

Paleomagnetism from multi-orogenic terranes is "not a simple game": Pyrenees' Paleozoic warning

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43 13 **Summary**
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47 14 Paleomagnetism is a versatile tool in the Earth sciences: it provides critical input to geological
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49 15 time scales and plate tectonic reconstructions. Despite its undeniable perks, paleomagnetism is
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51 16 not without complications. Remagnetizations overprinting the original magnetic signature of
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54 17 rocks are frequent, especially in orogens which tend to be the areas with better rock exposure.
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3 18 Unraveling the magnetic history of the rocks is a complicated task, especially in areas that
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5 19 underwent several orogenic pulses. In turn, constraining the timing of remagnetization represents
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7 20 an opportunity to solve post-magnetization structural and tectonic kinematics. Here, we evaluate
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9 21 the magnetization history of Silurian-Devonian carbonates from the Pyrenees (Spain). The
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11 22 Pyrenees are a multi-orogenic mountain belt where Silurian-Devonian rocks have seen the
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13 23 Variscan collision (late Paleozoic), the opening of the Atlantic-Biscay bay (early Cretaceous)
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15 24 and the Alpine orogeny (late Cretaceous to Miocene). Our results show widespread
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17 25 remagnetization(s) of the Silurian-Devonian series of the Pyrenees. The majority of the samples
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19 26 likely acquired their magnetization during waning episodes of the Variscan orogeny, i.e. late
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21 27 Carboniferous and early Permian times, although some apparently were subsequently
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23 28 remagnetized during the Alpine orogeny. The paleomagnetic results constrained that the Variscan
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25 29 orogeny was responsible for their main folding event, whereas the Alpine orogeny produced the
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27 30 thrusting and antiformal stacking of the Paleozoic rocks. In addition, we observed a general
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29 31 clockwise rotational pattern which could be related with the formation of the Cantabrian
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31 32 Orocline and/or rotations associated with the Alpine orogeny. The Silurian-Devonian carbonates
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33 33 are thus useful to understand the tectonic evolution of the Pyrenean mountain range after a
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35 34 systematic combination of paleomagnetism with structural and petrological observations. In
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37 35 contrast, the secondary character of magnetization and complications associated with the
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39 36 Variscan tectonics indicate that a reassessment of Siluro-Devonian poles from the Variscan
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41 37 elsewhere in Europe might be appropriate.
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49 38 **Keywords**

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53 39 Remagnetization, Pyrenees, Variscan, Alpine, Silurian-Devonian, orocline
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1. Introduction

The Earth's magnetic field has left a remnant signature in the geological record through eons. These magnetic signals in the rock archive have been crucial to almost any field of Earth Sciences, from the development of plate tectonics (e.g., Vine and Matthews, 1963), to the development of global time scales (e.g., Kuiper et al., 2008) or the origin and evolution of the core (e.g., Biggin et al., 2015). Paleomagnetism is still the only available technique that can quantify pre-Jurassic paleolatitudes (Domeier and Torsvik, 2019), intensities of the past magnetic field, or global reference times through reversals. The paleomagnetic imprint in rocks can last billions of years but may be also fragile. For example, remagnetizations that overprint or even delete the original magnetic signature are ubiquitous, especially in orogenic belts (e.g., Pueyo et al. 2007, 2016a; Van der Voo and Torsvik, 2012; Huang et al. 2017). The majority of the studies associated with the preservation and reacquisition of a magnetic remanence in rocks are relatively recent. Remagnetizations were initially recognized already during the 1960's and remarked its importance in the 1980's (McCabe et al., 1983; McCabe and Elmore, 1989). However, they have been studied in particular detail only from the first two decades of the 21st century onward (c.f. van der Voo and Torsvik, 2012), when paleomagnetists and rockmagnetists realized to the full that a plethora of chemical and physical processes are capable of resetting the magnetic signature in a rock (e.g., Jackson et al., 1993; Weil and van der Voo., 2002; Dekkers, 2012; Pastor-Galán et al., 2017; Aubourg et al., 2019; Huang et al., 2020). Many paleomagnetic studies were performed before our current understanding of remagnetization processes. However, for a given region, time frame, or lithology, those older paleomagnetic data may be the only data available. There is an evident need to review and critically reassess such paleomagnetic information: regional and even global geologic interpretations are still grounded in them.

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3 63 Remagnetization is often deemed a problem because it interferes with paleogeographic
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5 64 reconstructions that rely on the analysis of primary natural remanent magnetization (NRM), i.e. the
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7 65 age of the NRM is the same as the age of the sampled rock unit. However, despite the perceived loss
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9 66 of information, remagnetized rocks do represent valuable sources of geological information when it
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11 67 is possible to retrieve precisely the timing of the resetting of the original NRM acquisition, i.e. the
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13 68 remagnetization. Remagnetized rocks have been successfully used to unravel paleolatitudes of
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15 69 orogenic processes, orogenic kinematics, as geothermometers, to reconstruct inverted basins, ...
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17 70 (e.g., Dinarès-Turell and García-Senz, 2000, Huang et al., 2015; Villalaín et al., 2016; Auburg et al.,
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19 71 2019; Izquierdo-Llavall et al., 2020). Rocks with complex orogenic histories – the rule in many
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21 72 orogens – present a myriad of complications including the timing of their NRM acquisition. In such
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23 73 settings, paleomagnetism can be an excellent tool to understand multiphase orogenic systems if
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25 74 wisely used in concert with other geologic tools.

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31 75 In this paper, we reappraise the paleomagnetism from Silurian-Devonian limestones in the Pyrenean
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33 76 mountain belt, from which only three rather limited studies have been previously published (Tait et
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35 77 al., 2000; Gil-Peña et al., 2006; Izquierdo-Llavall et al., 2020). Our results, together with an enhanced
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37 78 geologic and paleomagnetic knowledge of the orogen, show that the sampled Silurian-Devonian
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39 79 carbonates were completely remagnetized during the late Carboniferous and Early Permian, when
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41 80 they experienced significant clockwise vertical axis rotations. These rocks were partially overprinted
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43 81 yet another time during the Cenozoic times, and experienced tilting and vertical axis rotations
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45 82 related to both the opening of the Bay of Biscay in the Late Mesozoic and the Cenozoic building of
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47 83 the Alpine chain in the Pyrenees. The magnetization history of the Pyrenees is a warning for
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49 84 paleomagnetists beautifully illustrated in Chris Scotese's anagram: PALEOMAGNETISM = NOT A
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51 85 SIMPLE GAME (Van der Voo, 1993).

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86 2. The Pyrenean mountain belt

87 The Pyrenees are a mountainous barrier that separates the Iberian Peninsula from the rest of
88 Eurasia. They are a prime example of the superposition of different tectonic events: (1) the
89 multiphase late Paleozoic Variscan orogeny; (2) the Jurassic and early Cretaceous major extension
90 during the opening of the Atlantic Ocean and the formation of the Bay of Biscay; (3) the opening of
91 a seaway including the exhumation of the mantle between the Neotethys and Atlantic Oceans along
92 the current Pyrenees; (4) the subduction of this previously opened seaway in the late Cretaceous; and
93 (5) the final collision between Iberia and Eurasia during the Cenozoic (Muñoz, 1992; 2019 and
94 references therein).

95 2.1 Paleozoic History: Variscan cycle

96 The tectonic evolution of the Paleozoic era was dominated by the progressive amalgamation of most
97 continents into Pangea (e.g., Domeier and Torsvik, 2014; Domeier, 2016), the latest supercontinent
98 (Pastor-Galán et al., 2019a). In western Europe the Pangean amalgamation history is recorded in the
99 Variscan orogen, which sutured the continents of Gondwana and Laurussia along with a variable
100 number of smaller plates that likely drifted away from Gondwana (e.g., Nance et al., 2010). On the
101 basis of paleomagnetic data, Iberia has been considered part of a ribbon continent (usually named
102 Armorica, Galatia, or Hun) that detached from Gondwana and drifted to the north or northwest in
103 the Late Silurian or Early Devonian (e.g., van der Voo, 1993; Tait et al., 2000; Tait, 1999; Stampfli et
104 al., 2013; Domeier and Torsvik, 2014). Other authors, however, place Iberia along the passive
105 margin of Gondwana throughout the Paleozoic based on the fossil record or the provenance of
106 detrital zircons (e.g., Robardet, 2003; Pastor-Galán et al., 2013a). Convergence leading up to the
107 Variscan orogen started ca. 420 Ma (e.g., Franke et al., 2017) and continued until the complete

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3 108 consumption of the Rheic ocean and other minor oceanic basins that existed between Gondwana
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5 109 and Laurussia at ca. 280 Ma; (e.g., Pastor-Galán, 2020). The final continent–continent collision was
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7 110 diachronic and became progressively younger westwards (in present-day coordinates) with Devonian
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9 111 continent–continent collision along the eastern boundary of the Variscan orogen, progressing to
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11 112 earliest Permian ages in the westernmost sector (e.g., Pastor-Galán et al., 2020 and references
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13 113 therein).

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17 114 Iberia has the largest exposure of the Variscan orogen in Europe, and an almost continuous cross
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19 115 section of the orogen (e.g., Azor et al., 2019). The majority of the Paleozoic outcrops in Iberia
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21 116 contain Gondwanan affinity rocks (e.g., Pastor-Galán et al., 2013a; Casas et al., 2019) and only a little
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23 117 sector of Southwest Iberia shows Laurussian affinity (e.g., Pérez-Cáceres et al., 2017).

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26 118 Geographically, the external zones of the Gondwana margin are nested to the north into the core of
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28 119 the Cantabrian Orocline (Fig. 1A), whereas the hinterland zones are to the west, center and
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30 120 northeast of Iberia (Fig. 1A; e.g., Azor et al., 2019). The stratigraphy of the Gondwanan autochthon
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32 121 consists of Neoproterozoic arc rocks (e.g., Fernández-Suárez et al., 2014), which evolved to a rift-to-
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34 122 drift sequence during the Cambrian to early Ordovician and then to an Ordovician to late Devonian
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36 123 passive margin basin sequence (e.g., Gutiérrez-Alonso et al., 2020). During the Carboniferous and
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38 124 early Permian, the rocks recorded up to 6 phases of deformation (e.g., Dias da Silva et al., 2020;
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40 125 Pastor-Galán et al., 2020 and references therein), metamorphism (e.g., Ribeiro et al., 2019) and
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42 126 synorogenic sedimentation processes that evolved to post-orogenic and intracontinental style basins
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44 127 during the Permian (e.g., Oliveira et al., 2019).

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50 128 The trend of the Variscan belt in Iberia follows a “S” shape; to the north and convex to the west is
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52 129 the Cantabrian Orocline, and to the center-south and convex to the east is the Central Iberian curve
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54 130 (e.g., Pastor-Galán et al., 2020). To the north, the Cantabrian Orocline displays a curvature of
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3 131 approximately 150°. In eastern Iberia the Cantabrian Orocline seems isoclinal, but this is likely the
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5 132 product of a retightening during the Alpine orogeny (e.g., Pastor-Galán et al., 2011; Leite Mendes et
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7 133 al., 2021; Fig. 1A). All kinematic data studied so far support a model in which the Cantabrian
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9 134 Orocline formed due to secondary vertical-axis rotation in a period of time later than 315 Ma and
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11 135 earlier than 290 Ma. Overall, the southern limb of the orocline rotated counterclockwise (CCW) and
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13 136 the northern limb clockwise (CW; e.g., Weil et al., 2013). Orocline formation postdates the main
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15 137 Variscan orogenic phases (e.g., Pastor-Galán et al., 2015a). The development of the Cantabrian
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17 138 Orocline implies the existence of a roughly linear orogenic belt during the early Variscan closure of
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19 139 the Rheic Ocean (with an approximately N–S orientation in present-day coordinates), which was
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21 140 subsequently bent in map-view into an orocline during the late stages of Pangea's amalgamation.
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23 141 This interpretation is grounded in extensive paleomagnetic, structural and geochronological studies
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25 142 (e.g., Weil et al., 2001; Pastor-Galán et al., 2014; Shaw et al., 2015; Gutiérrez-Alonso et al., 2015).
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27 143 The more southern Central Iberian curve has a similar magnitude, but opposite curvature compared
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29 144 to the Cantabrian Orocline (Fig. 1A), although its exact geometry is uncertain because this structure
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31 145 is largely covered by Mesozoic and Cenozoic basins (e.g., Pastor-Galán et al., 2020; Fig. 1A). The
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33 146 curvature is most recognizable at the boundary between the Galicia–Tras-os-Montes and Central
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35 147 Iberian zones (e.g., Aerden, 2004; Martínez Catalán, 2012). The most recent geometric and
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37 148 kinematic investigations suggest that the Central Iberian curve is not a structure formed by
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39 149 differential vertical-axis rotation as the Cantabrian Orocline, but one formed as a consequence of
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41 150 several complex, complementary and successive tectonic processes (e.g., Pastor-Galán et al., 2018).
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49 151 The Palaeozoic rocks of the Pyrenees form the backbone of the chain and crop out in two areas (the
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51 152 Axial Zone and the Basque Massif, to the east and west, respectively) that define an E-W elongated
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53 153 strip unconformably overlain by Mesozoic and Cenozoic rocks (Fig. 1A and B). These outcrops are
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55 154 geographically disconnected from neighboring Paleozoic outcrops of the Catalan Coastal Range and
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3 155 Balearic to the southeast, the Mouthoumet and Montagne Noire (southern French Central massifs)
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5 156 to the north, Corsica-Sardinia to the east and the Iberian Massif to the west and southwest. The pre-
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7 157 Permian rocks of the Pyrenees recorded a polyphase deformation during the Variscan orogeny with
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9 158 metamorphism that ranges from absent to high grade (e.g. Casas et al., 2019). So far, no relics of
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11 159 early Variscan deformation and/or subduction related high pressure metamorphism have been
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13 160 found. Most palaeogeographic reconstructions suggest that the Pyrenean Paleozoic outcrops (Fig. 1)
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15 161 may be equivalent to the northern branch of the Cantabrian Orocline (e.g. García-Sanseguendo, 2011;
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17 162 Pastor-Galán et al., 2020). Deformation, structural style and metamorphic grade show important
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19 163 differences along strike in the Pyrenees (Autran and García-Sanseguendo, 1996; Debon and Guitard,
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21 164 1996). The superposition of the later Mesozoic extension and subsequent Alpine orogeny markedly
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23 165 complicate an integral interpretation of the Variscan portions of the Pyrenees (see Casas et al., 2019).
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25 166 As a consequence, a comprehensive scheme integrating all the available data is lacking despite
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27 167 decades of geological research (e.g., de Sitter and Zwart, 1959; Kleinsmiede, 1960; Zwart, 1979;
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29 168 1986). In general terms, the Silurian, Devonian and Carboniferous successions show no to low-grade
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31 169 metamorphism and are composed of carbonates and shales (e.g., Casas et al., 2019). During the
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33 170 Carboniferous and Early Permian, the Pyrenees recorded an intense igneous activity including syn-
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35 171 to post-kinematic plutonism and volcanism (Fig. 1B; e.g., Gleizes et al., 1997, 2003; Denèle et al.,
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37 172 2011, 2014; Porquet et al., 2017), sometimes interpreted as subduction related (e.g., Pereira et al.,
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3 175 2.2 Mesozoic to present day evolution: Alpine cycle
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7 176 The final break-up of Pangea is marked with the opening of the Central and South Atlantic from late
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9 177 Triassic times onward (e.g., Müller et al., 2019). During the Jurassic, Iberia was attached to Europe
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11 178 and North America as another piece of Laurasia. In the Cretaceous, the breakup and spreading in
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13 179 the North Atlantic led to the separation of the Iberian microplate from Eurasia, North America and
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15 180 Africa (e.g., Vissers and Meijer, 2012). During the North Atlantic breakup, the Bay of Biscay
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17 181 opened, leading to approximately 35° of counterclockwise (CCW) rotation of Iberia (Van der Voo,
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19 182 1969; Neres et al., 2013) probably during the Aptian (Juárez et al., 1998; Gong et al., 2008). The
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21 183 opening of the Bay of Biscay to the west got recorded in the Pyrenees with the formation of a
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23 184 hyperextended margin with mantle exhumation during the Albian–Cenomanian (e.g. Lagabrielle et
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25 185 al., 2010). The rotation of Iberia, together with the increased convergence between Africa and
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27 186 Eurasia culminated in the collision between Iberia and Eurasia to form the Pyrenean range during
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29 187 Late Cretaceous-Miocene in the frame of the Alpine orogeny (e.g., Muñoz, 2019).
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35 188 Paleozoic rocks in the Spanish Pyrenees occur in three main Alpine thrust sheets (Muñoz 1992,
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37 189 2019; Barnolas et al., 2019) that provoked over 100 km of N-S shortening. So, the original position
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39 190 of the Palaeozoic rocks should be located northward from their present-day arrangement. The
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41 191 outcrops of Paleozoic rocks in the Axial Zone have witnessed more than 7000 meters of basement
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43 192 stacking (measured between the Balaitous peak and the San Vicente drill core; Fig. 1B; Lanaja, 1987).
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45 193 The geometry and kinematics of the thrust units affecting the Paleozoic rocks are not fully
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47 194 understood because of the complex superposition of deformation events and the unclear
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49 195 relationships with cover units where syntectonic sedimentation plays a key role to assign kinematic
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51 196 ages (Oliva-Urcia, 2018). Numerous structural studies (e.g., Muñoz et al., 1986, 2019; Muñoz, 1992;
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53 197 Puigdefábregas et al., 1992; Teixell, 1996, Millán et al., 2000; Martínez-Peña and Casas, 2003; Casas
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3 198 et al., 2003; Millán et al., 2006; Labaume et al., 2016; Labaume and Teixell, 2018) have identified a
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5 199 general Alpine piggy-back thrust sequence affecting the Paleozoic rocks (Fig. 2). Besides, numerous
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7 200 fission-track data on granites (Fitzgerald et al., 1999; Jolivet et al., 2007), on detrital rocks (Beamud
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9 201 et al., 2011; Bosch et al., 2016; Labaume et al., 2016) as well as $^{40}\text{Ar}/^{39}\text{Ar}$ and U/Pb dating of
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11 202 samples directly taken from fault planes (Abd Elmola et al., 2018) and calcite veins (Hoareau et al.,
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13 203 2021), see also recent reviews by Oliva-Urcia, 2018 and Calvet et al., 2020, and references therein),
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15 204 have improved the chronology of emplacement and exhumation of the basement units and their
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17 205 relationship with the cover ones. Basement thrusts partly reactivated previous Variscan, late-
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19 206 Variscan and/or Mesozoic structures. The Alpine structure defines an imbricate thrust system (Fig
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21 207 2) with a progressively increasing vertical overlap between the basement units from west (Fig. 2A) to
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23 208 east where thrusts define an antiformal stack (Fig. 2C). Basement units in the west include three
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25 209 thrusts in the central sector: Millares (Paleocene), Bielsa (Eocene) and Gavarnie (Eocene-Oligocene);
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27 210 and an undetermined number in the western sector (for example Lakora-Eaux Chaudes (Upper
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29 211 Cretaceous-Paleocene- even Eocene); Gavarnie (Eocene); Guara-Gedré (Eocene-Oligocene), Fiscal-
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31 212 Broto(Oligocene), and Guarga (Oligocene-Miocene, not indicated in Fig. 2). In the eastern sector
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33 213 main basement units are recognized, from the North to the South: Nogueras (emplaced in the
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35 214 Paleocene), Orri (Eocene), and Rialp (Oligocene). In the South Pyrenean Zone, all these basement
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37 215 units connect with an imbricate fold-and-thrust system with different Mesozoic and Cenozoic
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39 216 décollements (Cretaceous shales, Eocene marls and Upper/Middle Triassic evaporites); their
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41 217 geometry is controlled by salt-tectonics and Mesozoic inheritance (Millán et al., 2000; Huyghe et al.,
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43 218 2009; Labaume et al., 2016; Oliva-Urcia, 2018; Labaume and Teixell, 2018; Calvín et al., 2018;
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45 219 Santolaria et al., 2020; Muñoz et al., 2021).
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3 221 Thrusting was associated with important foreland flexure and foreland succession deposition both in
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5 222 the Northern Pyrenees (France; Biteau et al., 2006) and the Southern Pyrenees (Spain;
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7 223 Puigdefàbregas, 1975). Foreland deposits partly covered the Paleozoic units of the Axial Zone in the
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9 224 early orogenic stages and were subsequently exhumed and eroded (Beamud et al., 2011; Fillon and
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11 225 Van der Beek, 2012). The early, maximum burial conditions in the sampled portion of the Axial
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13 226 Zone are partly constrained by paleothermal studies in the overlying Meso-Cenozoic cover units
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15 227 (Izquierdo-Llavall et al., 2013; Labaume et al., 2016). They indicate Cenozoic-age, maximum
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17 228 temperatures of 160-190 °C in the Upper Cretaceous units of the western Axial Zone (Izquierdo-
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19 229 Llavall et al., 2013) that increase up to ~250 °C in the Eocene turbidites to the center of the Southern
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21 230 Pyrenean Zone. These values indicate that maximum temperatures in the underlying Paleozoic rocks
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23 231 were higher than 200-250 °C during the Cenozoic. They are in agreement with thermal modelling
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25 232 results of the Panticosa intrusion (Bosch et al., 2016) that indicate peak burial temperatures of ~300
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27 233 °C during the Oligocene. In the central Axial Zone, thermal models for the Paleozoic units of the
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29 234 Gavarnie and Orri units reveal peak temperatures below 300°C that were attained during the Early
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31 235 Paleogene (Waldner, 2019). Cenozoic burial favored the development of Alpine cleavage in the
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33 236 western Axial Zone (Choukroune and Séguret, 1972; Matte et al., 2000). Conversely, in the central
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35 237 Axial Zone Alpine cleavage developed only locally, the main cleavage being Variscan in age (Muñoz,
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37 238 1992).

38 39 40 41 42 43 44 45 239 2.3 Paleomagnetism in the Pyrenees

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48 240 Paleomagnetic investigations in the Pyrenees commenced with the pioneering studies of Van der
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50 241 Lingen (1960) and Schwarz (1963) in some Paleozoic rocks from the center of the Pyrenees. The
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52 242 available database has grown substantially during the following decades due to the excellent outcrop
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54 243 conditions (including world class stratigraphic sequences), the general exposure of synorogenic
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3 244 material throughout the chain allowing an accurate dating of deformation, the existence of well-
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5 245 exposed zones of lateral transference of deformation, etc. At present, the Pyrenean chain represents
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7 246 one of the most densely and homogeneously sampled paleomagnetic databases worldwide (Pueyo et
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9 247 al., 2017). Despite the quality and amount of paleomagnetic data in the Pyrenees, the Pyrenean
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11 248 Paleozoic rocks have remained largely unexplored. In the Axial Zone, very few data are known from
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13 249 sites older than Permian–Triassic red beds (Van Dongen, 1967; McClelland and McCaig, 1988, 1989;
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15 250 Keller et al., 1994; Tait et al., 2000; Gil-Peña et al., 2006; Izquierdo-Llavall et al., 2014, 2020; Ramón
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17 251 et al., 2016). And, to our knowledge, in the Pyrenees only three paleomagnetic studies collected and
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19 252 analyzed a limited number of sites dating from older than late Carboniferous (Stephanian) age: Tait
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21 253 et al. (2000), Gil-Peña et al. (2006), and Izquierdo-Llavall et al. (2020). The two latter studies found
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23 254 the rocks remagnetized in the Late Carboniferous and Paleogene, respectively. Previous studies in
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25 255 late Carboniferous and early Permian rocks (Izquierdo-Llavall et al., 2014) revealed shallow
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27 256 inclinations and clockwise rotations of $\sim 40^\circ$. Gil-Peña et al. (2006) showed analogous rotations
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29 257 ($\sim 50^\circ$ CW) for the Ordovician rocks that remagnetized during these times.

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35 258 The relatively good paleomagnetic control on undeformed areas in the vicinity of the Pyrenees (e.g.,
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37 259 Garcés et al., 2020; Oliva-Urcia and Pueyo, 2019) allows to define a reliable reference paleomagnetic
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39 260 direction and to understand the post-Variscan (late Permian to Eocene) magnetization and tectonic
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41 261 history during the Alpine orogeny in the Pyrenees. In the South Pyrenean Zone, significant rotations
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43 262 both CW and CCW ($40\text{--}60^\circ$) derived from proven primary paleomagnetic records from rocks of
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45 263 ages ranging from Permo-Triassic (e.g., Larrasoña et al., 2003) to Oligocene (e.g., Sussman et al.,
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47 264 2004) are found in the most external cover units in relation to lateral ramps in thrust sheets. These
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49 265 rotations are especially evident nearby the boundaries of the so-called South Pyrenan Central Unit
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51 266 (e.g., Sussman et al., 2004; Mochales et al., 2012, 2016; Muñoz et al., 2013; Rodríguez-Pintó et al.,
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53 267 2016) but also in the most external thrust units (Pueyo et al., 2021a, 2021b). Other moderate vertical
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3 268 axis rotations (15-25° CW and CCW) occur in the Permo-Mesozoic structural units immediately to
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5 269 the south of the Axial Zone: The Internal Sierras (Larra-Monte Perdido units; Oliva-Urcia and
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7 270 Pueyo, 2007a, 2007b; Oliva-Urcia et al., 2008; Izquierdo-Llavall et al., 2015), the Nogueras thrust
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9 271 unit (McClelland and McCaig, 1988, 1989; Dinarès et al., 1992; Oliva-Urcia et al., 2012; Izquierdo-
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11 272 Llavall et al., 2018) and in the eastern Cadi unit (Dinarès et al., 1992; Keller et al., 1994; Pueyo et al.,
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13 273 2016b). In the North Pyrenean Zone (France, Fig. 1B), paleomagnetic data are scarcer and evidence
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15 274 strong (over 70° CW in Aptian-Albian rocks, Oliva-Urcia et al., 2010; Rouvier et al., 2012) to null
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17 275 (Izquierdo-Llavall et al., 2020) vertical axis rotations in different areas.

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22 276 Early Cretaceous remagnetizations are only described, so far, in the out of the Axial Zone, in the
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24 277 Southwestern Pyrenees (Larrasoña et al., 2003) and in the Cotiella Massif (Garcés et al., 2016).
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26 278 These remagnetizations are relatively common in the deformed Cretaceous basins to the south of
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28 279 the Axial Zone. They are local and affect compartmentalized and highly subsident basins developed
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30 280 under high thermal gradient conditions (e.g., Lagabrielle et al., 2010) which played a key role to
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32 281 chemically remagnetize these rocks (e.g., Dinarès-Turell and García-Senz, 2000; Gong et al., 2008;
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34 282 2009). On top of that, Cenozoic remagnetizations have been described in the Meso-Cenozoic units
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36 283 just above the Axial Zone (Oliva et al., 2008; 2012; Izquierdo-llavall et al., 2015). These Cenozoic
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38 284 remagnetizations likely occurred due to the burial associated with the development of the Pyrenean
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40 285 orogenic wedge. The wedge generated important lithostatic and tectonic load in the internal units
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42 286 until the final collision, continentalization and exhumation of the Paleozoic rocks during Oligocene-
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44 287 Miocene times.

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50 288 Most of the paleomagnetic data to the south of the Axial Zone recorded, at least partially, Eocene
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52 289 secondary magnetizations (pre, syn and postfolding). Remanent magnetization in the Mesozoic units
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54 290 immediately to the North of the Axial Zone is in general terms post-folding and has been
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3 291 interpreted as a Cenozoic chemical (Oliva-Urcia et al., 2010) or thermal (Izquierdo-Llavall et al.,
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5 292 2020) remagnetization.
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8 9 293 **3. Sampling, methods and results**

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12 294 We drilled in 19 limestone sites from the Silurian or Devonian, one site of a late Carboniferous-early
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14 295 Permian granite (OG01, Panticosa intrusion) and one Permian dyke that intruded the surroundings
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16 296 of site OG12 (OG12dyke; Fig. 1B; Table 1) with a petrol-powered drill, in total 240 cores. We also
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18 297 collected 6 oriented hand samples (from the OG07 and OG08 sites, three samples each). Sites are
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20 298 distributed along-strike the southern and central Axial Zone from the Gallego valley in the west to
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22 299 the Valira valley in the east in eight different valleys (Figs. 1B; the kml file sample_locations.kml with
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24 300 exact locations is in the Supplementary material). Sites BN1 and OR15 were collected in the same
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26 301 area of sites published in Tait et al. (2000). Several sites allowed field tests: five site-scale fold-tests
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28 302 could be obtained (OG2; OG11; OG13; OG14; OG19); two tilt tests between sites within the same
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30 303 thrust unit (OG3-4; BN1-OR15), and two sites with a baked contact test (OG12 and OG17). We
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32 304 performed all analyses at Paleomagnetic Laboratory Fort Hoofddijk, Universiteit Utrecht, The
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34 305 Netherlands.
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41 306 Our sample collection comes from fresh, non metamorphic and weakly or non-internally deformed
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43 307 sites. Most limestone sites contained variable amounts of organic matter visible while drilling. A few
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45 308 of these sites show a spaced solution cleavage and evidence of recrystallization. We collected the
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47 309 bedding orientation and, when observable, that of the pressure-solution cleavage (Table 1). The
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49 310 sampled rocks were affected by both Variscan and Pyrenean orogenies. The large scale Pyrenean
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51 311 (Alpine) thrusting can be distinguished from the Variscan because its emplacement produced
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53 312 kilometric-scale folding of the basement that is well recorded by the Mesozoic units unconformably
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3 313 overlying the Paleozoic rocks. Basement folding resulted in dominantly southward and northward
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5 314 tilts in the southern and northern part of the Axial Zone, respectively, but not producing significant
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7 315 plunging axes ($<15^\circ$). To provide a detailed kinematic reconstruction separating the Variscan and
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9 316 Alpine events we inferred the approximate value of these Mesozoic-Cenozoic Alpine tilts from the
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11 317 surface or reconstructed orientation of the Mesozoic units that overlay the basement . To this end,
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13 318 we used geological maps and regional-scale cross sections where sampling sites were projected (Fig.
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15 319 2; Table 1).
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20 320 3.1 Methods

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23 321 Knowing when and how rocks magnetized is crucial towards an appropriate interpretation of
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25 322 paleomagnetic results, especially in terms of plate and structural kinematics. In this paper we
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27 323 combined rock magnetic, paleomagnetic and structural geology analyses to unravel the
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29 324 magnetization history of the rocks.
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31 325 Rock magnetism studies the magnetic properties of rocks and their magnetic minerals. The different
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33 326 magnetic properties of rocks, such as magnetic hysteresis, susceptibility and its anisotropy,
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35 327 magnetization vs. temperature (thermomagnetic analysis), can inform about the mineral(s) carrying
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37 328 the magnetic remanence and their crystal structure and grain size. This information is the most
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39 329 important to understand the geological processes involved in the magnetization of the rocks, and
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41 330 eventually also a key to unravel magnetization timings. In this research we have performed a series
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43 331 of rock magnetic analyses to fully characterize the magnetic mineralogy of the studied samples as a
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45 332 step towards understanding the magnetization process and timing. We measured 18 high-field
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47 333 thermomagnetic runs in an in-house-built horizontal translation-type Curie balance with a sensitivity
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49 334 of approximately $5 \cdot 10^{-9} \text{Am}^2$ (Mullender et al., 1993) and in an AGICO KLY-3 susceptibility bridge
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51 335 with a CS2 furnace attachment with nominal sensitivity ($5 \cdot 10^{-7}$ SI) and air forced into the tube. We
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3 336 also analyzed 20 magnetic hysteresis loops and one first order reversal curve (FORC) diagram. They
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5 337 were measured at room temperature with an alternating gradient force magnetometer (MicroMag
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7 338 Model 2900 with 2 Tesla magnet, Princeton Measurements Corporation, noise level $2 \times 10^{-9} \text{ Am}^2$).
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10 339 Finally, we obtained 88 isothermal remanent magnetization (IRM) acquisition curves from our
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12 340 Pyrenean limestone samples. Curves were obtained with the robotized magnetometer system of
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14 341 Utrecht University (Mullender et al., 2016).
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16 342 Our paleomagnetic analyses were focused towards determining the Natural Remanent Magnetization
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18 343 (NRM) of the rocks. NRM provides information about ancient latitudes (inclination of the magnetic
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20 344 remanence) and rotations (declinations with respect to the past north) so that we can constrain the
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22 345 magnetization timing. The NRM of the sample collection was investigated through alternating field
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24 346 (AF) demagnetization and thermal demagnetization. AF demagnetization was carried out using the
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26 347 aforementioned robotic 2G-SQUID magnetometer, through variable field increments (4–10 mT) up
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28 348 to 70–100 mT. In all limestone samples, where high-coercitivity, low-blocking temperature minerals
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30 349 (i.e. goethite, titano-hematite) were expected, heating step to 150 °C was performed previous to the
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32 350 AF demagnetization. At the same time this enhances the distinction between secondary and
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34 351 characteristic NRM components determined with AF demagnetization (van Velzen & Zijdeveld,
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36 352 1995). In samples demagnetized thermally, a stepwise thermal demagnetization was carried through
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38 353 10–100 °C increments up to complete demagnetization. Principal component analysis (Kirschvink,
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40 354 1980) was used to calculate magnetic component directions from orthogonal vector end-point
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42 355 demagnetization diagrams (Zijderveld, 1967) with the online open-source software
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44 356 Paleomagnetism.org (Koymans et al., 2016; 2020).
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46 357 Anisotropy of magnetic susceptibility (AMS) measures the induced magnetization in a rock when
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48 358 applying a magnetic field in different directions, defining an ellipsoid. The shape of the AMS
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50 359 ellipsoid depends on the crystallographic preferred orientation of the minerals; the shape, size, and
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3 360 preferred orientation of mineral grains; the occurrence of microfractures, its distribution and size...

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5 361 Frequently it is a good proxy for sedimentary and tectonic fabrics that are not visually obvious, but it

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7 362 is also a powerful method to investigate the effect of deformation on the NRM. We determined the

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9 363 composite fabric of the paramagnetic, diamagnetic and ferromagnetic grains by measuring the AMS

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11 364 of 148 samples from our collection with an AGICO MFK1-FA susceptometer (nominal sensitivity

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13 365 2×10^{-8} SI).

14 15 16 366 3.2 Rock magnetism results

17 18 19 20 367 3.2.1 Thermomagnetic analyses

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23 368 We place between 50–100 mg of powdered sample material from representative samples into quartz

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25 369 glass cup holders that hold the sample with quartz wool. We programmed stepwise thermomagnetic

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27 370 runs with intermittent cooling between successive heating steps. The heating and cooling segments

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29 371 were 150, 100, 250, 200, 300, 250, 400, 350, 520, 450, 620, 550, 700° C and finally back to 25° C,

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31 372 respectively. Heating and cooling rates were 10° C min⁻¹. Many samples show a paramagnetic

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33 373 contribution, sometimes uniquely (OG19; Supplementary File SF1), sometimes with a more or less

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35 374 noteworthy alteration reaction at about 400–450° C. This indicates the presence of non-magnetic

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37 375 sulfides (likely pyrite) that oxidize to magnetite during the thermomagnetic run (Fig. 3; OG14;

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39 376 Supplementary File SF1). Some samples show a small but sharp decay between 500 and 600° C,

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41 377 indicating the presence of magnetite (Fig. 3, OG13), others show the presence of pyrrhotite with a

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43 378 sudden increase at ~300°-320° C followed by a sharp decrease afterwards (OG8 in Fig. 3; OG06

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45 379 and OG07 in SF1), all samples showing pyrrhotite contained a less important, but observable,

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47 380 content of pyrite. In the susceptibility vs. temperature curve (Fig. 3, BN1), pyrrhotite is observable

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49 381 during cooling but it likely formed as a secondary mineral during heating.

382 3.2.2 Magnetic hysteresis

383 Representative samples with masses ranging from 20 to 50 mg were measured using a P1 phenolic
384 probe. Hysteresis loops were measured to determine the saturation magnetization (M_s), the
385 saturation remanent magnetization (M_{rs}), and the coercive force (B_c). These parameters were
386 determined after correcting for the paramagnetic contribution. The maximum applied field was 0.5
387 T. The field increment was 10 mT and the averaging time for each measurement was 0.15 s. We
388 found different loop shapes (Fig. 4, SF-2): (i) Loops that do not saturate at 0.5 T with a pseudo-
389 single-domain like shape which points to the presence of a relatively hard magnetic carrier likely
390 pyrrhotite (Fig. 4, OG08) and (ii) typical magnetite-like pseudo-single domain loops (Fig. 4, OG19).
391 We performed a first order reversal curve (FORC) diagram (Fig. 4, BN1-3) that shows a mixture
392 between superparamagnetic and single domain behaviour.

393 3.2.3 Isothermal Remanent Magnetization (IRM)

394 Before the actual IRM acquisition, samples were AF demagnetized with the static 3-axis AF protocol
395 with the final demagnetization axis parallel to the subsequent IRM acquisition field, a procedure that
396 generates IRM acquisition curves with a shape as close to a cumulative-lognormal distribution as
397 possible (Egli, 2004; Heslop et al., 2004). IRM acquisition curves consist of 61 IRM levels up to 700
398 mT. The shape of IRM curves is approximately a variably skewed cumulative log-Gaussian function
399 in which may contain more than one coercivity phase. IRM component analysis enables a semi-
400 quantitative evaluation of different coercivity components (magnetic minerals or particle sizes) to a
401 measured IRM acquisition curve. Every skewed log-normal curve is characterized by four
402 parameters: (1) The field ($B_{1/2}$) corresponding to the field at which half of the saturation isothermal
403 remanent magnetization (SIRM) is reached; (2) the magnitude of the phase (M_{ri}), which indicates the
404 contribution of the component to the bulk IRM acquisition curve; (3) the dispersion parameter

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3 405 (DP), expressing the width of the coercivity distribution of that mineral phase and corresponding to
4
5 406 one standard deviation of the log-normal function (Kruiver et al. 2001; Heslop et al. 2002); and (4)
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7 407 the skewness of the gaussian curve (Maxbauer et al., 2016). IRM curve unmixing was performed
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9 408 with IRM MaxUnmix package (Maxbauer et al., 2016). The interpretation of the coercivity
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11 409 components in terms of mineralogy and grain size is usually done in concert with thermomagnetic
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13 410 curves.

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17 411 Results from individual samples are characterized by two main IRM components: (a) a relatively soft
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19 412 component (C1 in Fig 5A) with $B_{1/2}$ between a minimum value of 23 and a maximum of 74 mT, but
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21 413 generally ~ 40 mT and dispersion parameter (DP) of ~ 0.33 and 0.38 (log units); and (b) a high
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23 414 coercivity component (C2) with a high $B_{1/2} > 200$ mT and DP ~ 0.5 (log units). Both components
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25 415 are present in all samples but in varying proportions (Fig. 5A) of the SIRM. We performed end-
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27 416 member modeling in all of the same lithology Silurian-Devonian samples (following the steps of
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29 417 Gong et al. (2009a) but without a 150°C preheating of the samples) to fully characterize the IRM set
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31 418 of samples. The program (Heslop and Dillon, 2007) to interpret the IRM acquisition curves uses the
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33 419 algorithm developed by Weltje (1997). End-member modeling assumes that the measured data can
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35 420 be represented by a linear mixture of a number of invariant constituent components, which are
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37 421 referred to as endmembers. The algorithm dictates that input IRM acquisition curves are monotonic;
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39 422 the curves were smoothed when appropriate to enforce them being monotonic. By least-squares
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41 423 minimization calculated normative compositions are optimized to the measured IRM acquisition
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43 424 curves, eliminating the need for prior knowledge of end-member properties (cf. Weltje, 1997). For
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45 425 further information about this technique in the framework of remagnetization see the review by
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47 426 Dekkers (2012). We found that a two end-member model shows an acceptable r^2 value of 0.6.
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49 427 Models with 3 to 9 end members show slightly better fits ($r^2 = 0.73$ to 0.88 respectively) although
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51 428 improvement is not deemed that significant (see discussion). The two end members are a soft (42
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3 429 mT component with a DP = 0.36) and a hard (a 200mT component and DP = 0.35) component,
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5 430 analogous to C1 and C2 in the individual IRM curves analyzed (Fig. 5C).
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8 9 431 3.3 Paleomagnetism results

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12 432 A minimum of five demagnetization steps was considered to characterize a remnant component. In
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14 433 specimens where directions were difficult to isolate, we used the approach of McFadden and
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16 434 McElhinny (1988) in combining great circles and linear best fits (set points). The Virtual
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18 435 Paleomagnetic Directions (VPD) software was also used (Ramón et al., 2017) at the site level
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21 436 (stacking routine, linearity spectrum analysis, and the virtual direction methods by Scheepers and
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23 437 Zijdeveld (1992); Schmidt (1982) and Pueyo (2000), respectively) to confirm the means derived
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25 438 from PCA analyses of individual specimens. Representative Zijdeveld diagrams are shown in Fig. 6.
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27 439 For the complete analyses, the reader can check the paleomagnetism.org files associated with this
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29 440 paper (check persistent identifier -PID- in the acknowledgements).
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33 441 Mean directions and uncertainties of each component were evaluated using Fisher's statistics (1953)
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35 442 of virtual geomagnetic poles (VGPs). We applied a fixed 45° cut-off to the VGP distributions of
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37 443 each site. In addition, we used the Deenen et al. (2011) criteria to evaluate the scatter of VGPs. As a
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39 444 general rule, if scatter is—mostly—due to paleosecular variation (PSV) of the geomagnetic field, the
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41 445 associated VGP distribution is roughly circular in shape. However, internal deformation, vertical axis
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43 446 rotation or inclination shallowing may add anisotropy to the scatter. In such cases, VGP
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45 447 distributions will show a certain degree of elongation or are otherwise not spherically uniform. Many
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47 448 samples show a NRM component with very low unblocking temperatures and low coercivities (100–
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49 449 180 °C or 10–12 mT). We consider this component as a viscous remanent magnetization (VRM),
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52 450 because of its similarity to the recent field (Fig. 7).
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3 451 After VRM removal, the samples show a single NRM component (Fig. 6), generally pointing to the
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5 452 origin, regardless of the mineral, magnetite (usually fully demagnetized at 40-60 mT and 500-580° C)
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7 453 and/or pyrrhotite (fully demagnetized at 330° C and little to barely demagnetized in AF). This
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9 454 characteristic remanent magnetization (ChRM) clusters well in all the sites (concentration parameter
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11 455 $k > 8$, but generally over 15; Table 2) with the exceptions of sites OG02 and OG08 (Figs. 8 and 9;
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13 456 Table 2). In addition, there are three sites with less than seven samples passing the 45° cut-off
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15 457 (OG05, OG06, OG14) and therefore their statistical parameters are not reliable. These five sites
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17 458 were excluded from further interpretation. The remaining 15 sites show quite variable ChRM
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19 459 declinations and inclinations which appear to be only comparable between sites within the same
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21 460 thrust unit (Figs. 2, 8 and 9, Table 2). To account for the different events of deformation we have
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23 461 used bedding corrections (Table 2) and fold tests (Fig. 10). In addition, we also performed
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25 462 inclination only statistics (Enkin and Watson, 1996; Arason and Levi, 2010) to eliminate clustering
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27 463 problems related to vertical axis rotations using both the bedding parameters and our inferred
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29 464 Alpine corrections (see top of section 3) (Table 3).

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35 465 In geographic coordinates the considered sites show clusterings that range from k (concentration
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37 466 parameter) ~ 8 (OG10 and 11) to $k \sim 188$ (OG19). Site average declinations range from 125° to
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39 467 297°, the majority of them in the south quadrants with sites OG15 and BN1-OR15 (combined sites
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41 468 separated by 100 m) being the only exceptions (Fig. 8 and 9; Table 2). Inclinations range from -50°
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43 469 to 50°. Bedding correction significantly changes the distribution of the site averages, but the
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45 470 scattering in declinations (from 111° to 289°) and inclinations (-65° to 56°) remains (Table 2), which
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47 471 means that magnetization timing is not the same for all samples and/or structural complications are
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49 472 larger than folding.
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3 473 All fold-tests whose samples passed the aforementioned quality criteria (Fig. 10; OG03-04; OG11;
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5 474 OG13 and OG19) were performed in folds with weakly plunging axes (Table 1; Fig. 11) with the
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7 475 exception of OG11, which in turn is the only one that is not negative (Fig. 10). OG11 shows a
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10 476 better clustering (τ_1) after tilt correction, however, the fold-axis in site OG11 is steeply plunging (the
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12 477 only case; Fig. 10). After back-tilting the plunging-axis (azimuth/plunge = 005/42), the fold test
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15 478 remains indeterminate, in this case with a greater clustering (τ_1) before tilt correction (Figs. 10
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17 479 (structural correction panel) and 11). The statistics of all sites that pass our quality criteria ($n \geq 7$ and
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19 480 $k > 8$) yield close to random distributions both considering all specimens ($k = 1.63$ and $K = 1.65$;
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21 481 Supplementary File SF3) and the mean of site averages ($k = 1.95$ and $K = 2.5$; Table 4). As expected
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23 482 from the negative within-site fold-tests, the concentration parameter does not change after bedding
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25 483 correction neither in all specimens together ($k = 1.51$ and $K = 1.59$; Supplementary File SF3) nor
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27 484 the mean of site averages ($k = 1.57$ and $K = 1.64$; Table 4).

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31 485 Inclination only statistics are independent to differential vertical axis rotations since declinations are
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33 486 not taken into account (e.g. Enkin and Watson, 1996). Inclination only statistics were performed on
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35 487 site averages to avoid weighting based on number of specimens (Table 4). The concentration
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37 488 parameter (k) equals to 0 in geographic coordinates and 2.17 in tilt corrected coordinates both
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39 489 figures representing very poor clusterings. k becomes close to 4 if we do not account for OG3 and
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41 490 OG4, which follow a different trajectory and may represent a different magnetization event. In
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43 491 contrast, when correcting the studied samples exclusively for our inferred Alpine tilt, the inclination
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45 492 only concentration parameter is ~ 4 , but becomes ~ 14 when excluding OG3 and OG4 (Table 4). An
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47 493 inclination only tilt test without OG3 and OG4 shows a best fit for a 110% correction, both using
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49 494 the Enkin and Watson, 1996 approach (with a maximum clustering around $k \sim 12$) and a stepwise
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51 495 untilting following Arason and Levi (2010) inclination only statistics with a maximum at $k \sim 15$ to
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3 496 (Fig. 12). OG3 and OG4 share a common true direction in geographic coordinates and their Alpine
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5 497 tilt correction does not change them too much (Fig. 13)
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8 498 3.4 Anisotropy of the Magnetic Susceptibility (AMS) results

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12 499 In pyrrhotite, the magnetic easy direction is confined to the basal crystallographic plane which
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14 500 implies an intrinsically strong anisotropy because of the 'hard' crystallographic c-axis (Schwarz and
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16 501 Vaughan, 1972; Schwarz, 1974). When pyrrhotite grows oriented in a preferred fabric (e.g. S1), the
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18 502 direction of the magnetic remanence can be biased towards the fabric plane (Fuller, 1963). The
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20 503 studied samples occasionally show pressure solution cleavage and a widespread presence of
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22 504 pyrrhotite as a partial or main carrier of the NRM. An inspection of the AMS fabrics can reveal
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24 505 whether pyrrhotite is oriented according to the S1 fabric.
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29 506 We measured the magnetic anisotropy in 148 samples from most sites to explore possible causes for
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31 507 the variety of ChRM directions found (Table 2). The degree of anisotropy (P) appears to be
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33 508 generally low (< 1.05 ; Fig. 14, Supplementary file SF4) although some individual samples showed up
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35 509 to 1.5. The samples' three principal ellipsoid axes (Kmax, Kint, and Kmin) follow the bedding (S0)
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37 510 in six sites (OG09, 15, 16, 17, 18, 19); in three samples the AMS ellipsoid corresponded to the
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39 511 foliation S1 (OG06, 10 and 13); and others showed a quasi-random pattern (OG03, 05, 11, 14) both
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41 512 in geographic and tectonic coordinates (Fig. 14). No observed AMS fabric (not Kmax, Kint, or
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43 513 Kmin axes) from the datasets studies coincides with the ChRM directions, implying that NRM and
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45 514 ChRM are not biased by rock fabric.
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515 4. Discussion

516 The paleomagnetic and rock magnetic results obtained from the Silurian-Devonian limestones in the
517 Pyrenees certify that the Paleozoic rocks from this mountain belt have been subject to at least one
518 widespread remagnetization event. Many samples contain a VRM that is similar to the recent
519 geoaixial dipole for recent times in the Pyrenees (Fig. 7). Apart from this VRM, all rocks, regardless
520 of their magnetic mineralogy, show a single stable component heading to the origin, with the
521 exception of samples not delivering results (Fig. 6 and 8). This component is not deviated toward
522 bedding/cleavage planes (as inferred from AMS patterns, Fig. 14) and displays negative fold tests
523 (Fig. 10; perhaps a syn-folding fold test in the case of site OG11).

524 4.1 Rock magnetism

525 Rock magnetic analyses show that both pyrrhotite and magnetite are the magnetic carriers in the
526 Silurian-Devonian limestones whereas magnetite is the carrier in the Panticosa late Carboniferous-
527 Permian granite (OG1) and sampled dyke (OG12dyke). All limestone sites contain variable amounts
528 of pyrrhotite and magnetite both in thermomagnetic curves (Fig. 3), in IRM acquisition curves and
529 during NRM demagnetization (Figs. 5 and 6). We applied the IRM end-member modeling technique
530 in an attempt to discriminate between different remagnetization events in the Pyrenees. The two
531 end-member model with a reasonably high r^2 value of 0.65 is our preferred model. Models with 3 to
532 9 end members evidently show slightly better fits ($r^2 = 0.73$ to 0.88 respectively). However, neither
533 the fit improves significantly, nor the shape of the end members shows more or less anticipated
534 IRM acquisition curves for any particular mineralogy (Fig. 5B). In addition, most of the additional
535 end members seem to represent the variable coercivity windows of magnetite (4 endmember
536 example in Fig. 5B: 3 endmembers (EM1-3) all represent magnetite and do not deliver meaningful

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3 537 results). All samples contain a significant amount of those additional endmembers (varying from
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5 538 10% to 60%) indicating that a variable grain-size or compositional magnetite is present in virtually
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7 539 every sample. The two end-member model further distinguishes a 42 mT component with a DP =
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9 540 0.36 (C1), which is typical for magnetite and a 200mT component and DP = 0.35 (C2), which we
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11 541 interpret as pyrrhotite (Fig. 5C). The two end members are in agreement with individual sample fits,
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13 542 but end-member IRM acquisition curves describe much better the IRM properties of each magnetic
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15 543 phase (Fig. 5). We interpret the soft component (C1) as magnetite varying from coarse to very fine
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17 544 grained (i.e. lower and higher coercivity respectively) as supported by hysteresis loops (Fig. 4 and
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19 545 SF2). It is reasonable that the high coercivity component (C2) reflects the observed pyrrhotite in the
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21 546 thermomagnetic curves as SD pyrrhotite has a rather high coercivity.
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26 547 The presence of variable amounts of pyrrhotite and magnetite in all Silurian-Devonian samples
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28 548 studied suggests that this is a common feature for the Paleozoic sedimentary and metasedimentary
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30 549 units of the mountain belt. Pyrrhotite is a stable mineral in low-grade metamorphic rocks under
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32 550 reducing conditions (e.g. Aubourg et al., 2012) such as the Pyrenean Silurian-Devonian limestones.
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34 551 Similar to the Pyrenees, pyrrhotite is the most common magnetic carrier in other limestone
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36 552 formations of the Iberian Variscides heavily affected by late Carboniferous magmatism (Pastor-
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38 553 Galán et al., 2015a; 2016; 2017; Fernández-Lozano et al., 2016).
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43 554 In general terms, the occurrence of pyrrhotite in limestones is a sign of their remagnetization.
44
45 555 Pyrrhotite is a frequent secondary mineral which is formed in limestones under anchimetamorphic
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47 556 and low-grade metamorphic conditions (Crouzet et al., 2001; Aubourg et al., 2019; Izquierdo-Llavall
48
49 557 et al., 2020) or in the presence of non oxidizing magmatic fluids (Pastor-Galán et al., 2016). The
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51 558 occurrence of pyrrhotite has been used as a geothermometer; increasing burial enhances the
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53 559 transformation of magnetite to pyrrhotite and the progressive replacement of magnetite-carried
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3 560 magnetizations by pyrrhotite-carried remagnetizations (e.g. Aubourg et al., 2019). In clay-rich rocks,
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5 561 magnetite and pyrrhotite coexist at burial temperatures <340 °C whereas at ~ 350 °C the
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7 562 concentration of magnetite decreases drastically and pyrrhotite becomes the dominant magnetic
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10 563 mineral (Aubourg et al., 2019). Izquierdo-Llavall et al. (2020) estimated the peak temperatures (~ 350
11
12 564 – 450 °C) in the North Pyrenean Zone (in the northernmost side of cross section 2c in Fig. 1)
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14 565 following the magnetite –pyrrhotite transformations. The temperature estimates for the south
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16 566 Pyrenees (Izquierdo-Llavall et al., 2013; Labaume et al., 2016) are much lower, in contrast to the
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18 567 North Pyrenean Zone. Taking that into account, we suggest that in our samples pyrrhotite was
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21 568 formed by a fluid induced chemical remagnetization during the latest stages of the Variscan orogeny.
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23 569 However, our westernmost sampled units (OG1-OG4) could surpass the Curie temperature of
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25 570 pyrrhotite (~ 320 ° C; e.g. Dekkers, 1989) during the Cenozoic burial, and possibly represent a
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28 571 Cenozoic TRM.

31 572 4.2 Paleomagnetism and timing of remagnetization

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35 573 We only consider sites for interpretation with at least 7 specimens passing the VGP's 45° cut-off
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37 574 criterion and with the concentration parameter $k > 8$. Sites OG02, 05, 06, 08, 14 and the dyke in
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39 575 OG12 do not pass these criteria and are not considered for further interpretation (Table 2). The rest
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41 576 of the sites show k values that range from barely above 8 in geographic coordinates (OG10 and 11),
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43 577 which could be expected also from sedimentary magnetizations, to over 40 (OG12, OG15, OG17-
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45 578 OG19) in geographic coordinates, which is unlikely in primary magnetizations of sediments (e.g.,
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47 579 Deenen et al., 2011). Site averages point generally to ESE-WSW with both positive and negative
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50 580 inclinations in geographic coordinates with the only exceptions of OG15 and BN1-OR15 which
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52 581 point WNW (Fig. 8 and 9; Table 2), something that does not change after bedding correction. With
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55 582 the exception of OG11, we have documented post-folding magnetizations. Importantly, the fabrics

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3 583 do not show consistency with NRM directions (particularly AMS fabric coincident with S1 does not
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5 584 occur) precluding any bias caused by a preferred orientation of pyrrhotite particles. Therefore, a post
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7 585 Variscan folding (i.e. late Carboniferous) is the oldest possible age for the magnetization since no
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10 586 earlier folding event has been described in the Pyrenees. The OG11 fold, which has a steep plunging
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12 587 axis (Fig. 11), yielded an inconclusive fold-test, but shows better clustering after tilt correction (Fig.
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14 588 10). Classical fold-tests assume horizontal axes and performing them in plunging axis' folds
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16 589 introduces spurious rotations and false positive/negative foldtests (e.g. Pueyo et al., 2016a). Pre-
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18 590 correcting the plunge of the fold axis may help to unravel the relative timing of magnetization, but in
19
20 591 turn the declination may not be reliable, introducing spurious rotations. In the case of OG11, pre-
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22 592 correcting the fold axis plunge produces another inconclusive result (Fig. 10). However, declinations
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24 593 in geographic, standard bedding correction, and Alpine tilt correction remain around 200° (Table 2
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26 594 and 3) and inclinations are in all cases relatively shallow (between -9 and 20), suggesting that OG11
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28 595 acquired its NRM during a reversed chron during a time when Iberia was at equatorial latitude:
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30 596 during late Carboniferous or Permian times (e.g. Pastor-Galán et al., 2018).

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35 597 When considering all site paleomagnetic directions, we found that declinations and inclinations are
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37 598 only compatible in geographic coordinates for those sites within the same tectonic unit (Figs. 1, 2, 8,
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39 599 9; Table 2). Such a directional pattern may be indicative of (a) different timing of NRM acquisition
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41 600 for each tectonic unit, (b) differential vertical axis rotations between units, (c) post-magnetization
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43 601 differential tilting between units or (d) a combination of the previous processes. To distinguish
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45 602 between these options inclination only statistics are appropriate (e.g. Enkin and Watson, 1996).
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47 603 Inclination only statistics do not consider declinations and therefore are independent of variations
48
49 604 due to differential vertical axis rotations. In order to evaluate potential timing of magnetization we
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51 605 performed statistical analyses in geographic coordinates, after bedding correction, and also after
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53 606 correction of the tilt related to the emplacement of Alpine basement thrusts (Figs. 2, 12, 13; Tables 3
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3 607 and 4). The concentration parameter of inclination data (k) is 0 in geographic coordinates (Table 4),
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5 608 which could mean that (i) sites magnetized at significantly different geological times when Iberia was
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7 609 at very different latitudes, and/or (ii) Alpine tilting postdates the magnetization and therefore it has a
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10 610 strong influence on the inclinations. Inclination only k is still too low (minimum $k \sim 8$ to consider
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12 611 an acceptable clustering) after bedding correction (~ 2) but also after Alpine tilt correction (~ 4).
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15 612 Two sites from the Gavarnie nappe (OG03 and OG04; Fig. 8 and 13, geographic coordinates) move
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17 613 in a very different direction both during bedding and Alpine tilt corrections. In contrast, OG01
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19 614 (Panticosa granite, late Carboniferous-Permian, Denèle et al., 2012), which is in the same Gavarnie
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21 615 nappe, correlates well with the rest of the sites in all thrust units after Alpine tilt correction but not
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23 616 with the neighboring OG03 and OG04. Thus, those sites might have acquired their magnetization at
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25 617 a significantly different time than the rest. After removing OG03 and OG04, the inclination only
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27 618 concentration parameter still indicates poor clustering after bedding correction ($k \sim 4$). However,
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29 619 when correcting only for the Alpine tilt k becomes 14.42 and a positive inclination only tilt test with
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31 620 a maximum in a 110% untilting (95% between 58 and 150; Fig. 12; Table 4). After the Alpine tilt
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33 621 correction, the mean inclination is $5^\circ \pm 11$ (Table 4) and all included sites show SW to SE
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35 622 declinations (Fig. 12). Despite the positive result, our Alpine tilt correction should be taken
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37 623 cautiously. We considered only regional tilt values, which were inferred from the average values of
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39 624 the overlying Mesozoic and thrust slopes in cross-sections (Fig. 2). Our estimated values took into
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41 625 account the kilometric-scale, thrust-related folding of the basement but can not consider the
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43 626 potential contribution of Alpine, outcrop-scale folding of the Paleozoic strata.
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50 627 We believe, however, that the inclination only k value of 14.42 together with the obtained shallow
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52 628 inclinations and southerly declinations are sufficiently convincing to argue for a common timing of
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54 629 NRM acquisition for the samples included in the tilt test (Fig. 12). The results imply a postfolding
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3 630 but a pre-Alpine tilt NRM. The shallow inclinations suggest that Iberia was located at equatorial
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5 631 latitudes and the southerly declinations suggest that this occurred during a reverse chron. We
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7 632 therefore suggest that all samples that passed the quality criteria, with the exception of sites OG03
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9 633 and OG04, magnetized during the latest Carboniferous to middle Permian times during the Kiaman
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11 634 reverse superchron, when Iberia was indeed located at equatorial latitudes (Weil et al., 2010). The
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13 635 late Carboniferous and early Permian times in the Pyrenees are characterized by widespread
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15 636 intrusions and volcanism (Panticosa granite, for example, OG01; Gleizes et al., 1998). We
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17 637 hypothesize that the remagnetization mechanism in the Pyrenees was triggered by fluids associated
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19 638 to the magmatic activity analogously to the remagnetizations observed in the Central Iberian Zone
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21 639 of west Iberia (e.g. Fernández-Lozano et al., 2016; Pastor-Galán et al., 2016; 2017).

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26 640 OG03 and OG04 show a negative fold test (Fig. 10) and a common true mean direction in
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28 641 geographic coordinates (Fig. 13). Their paleomagnetic direction is, however, significantly different
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30 642 from OG01 that is a late Carboniferous-early Permian site located in the same thrust sheet. OG03
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32 643 and OG04 show declinations to the south (both in geographic coordinates and after Alpine tilt
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34 644 correction) and upward inclinations of -43° and -48° (geographic coordinates) or -60° and -68° (after
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36 645 the restoration of the inferred Alpine tilt respectively). Both geographic and Alpine tilt corrected
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38 646 data indicate a remagnetization when Iberia was located at latitudes between 25° and 50° during a
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40 647 reverse chron. These results fit best to a remagnetization during late Cretaceous times, after the
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42 648 Cretaceous normal superchron (e.g. Izquierdo-Llavall et al., 2015 and references therein).

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45 649 Considering the steep $>55^\circ$ inclinations, we tentatively favor a late orogenic (Eocene) post Alpine
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47 650 tilting remagnetization. Such inclinations of $>55^\circ$ (after tilt correction) otherwise would only be
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49 651 possible after a remagnetization fairly long after the Alpine orogeny, when Iberia was located at
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51 652 similar latitudes as at present. We anticipate less remagnetization events long after the orogeny. Most
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53 653 structural units just above the basement units are remagnetized by tardi-orogenic burial
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3 654 remagnetizations (post-, syn- and pre- Alpine folding) that affected all kinds of rocks (limestones,
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5 655 calcarenites, redbeds...) (Dinarès et al., 1992; Dinarès, 1994; Keller et al., 1994; Oliva-Urcia and
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7 656 Pueyo, 2007b; Izquierdo-Llavall et al., 2015, Mujal et al., 2017 etc.) In addition, the burial
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10 657 temperature of OG03 and OG04 seem to have been sufficiently elevated to trigger a
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12 658 remagnetization after tilting during the unroofing of the Alpine orogeny.
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15 659 4.3 Tectonic significance

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19 660 The hypothesis of a detachment and northward drift of a peri-Gondwana microcontinent (Armorica
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21 661 s.l.) during the late Silurian or early Devonian (e.g. Torsvik et al., 2012; Stampfli et al., 2013;
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23 662 Domeier and Torsvik., 2014; Franke et al., 2017) is grounded largely on the basis of paleomagnetic
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25 663 data from the Silurian and Devonian rocks of the Pyrenees (Tait et al., 2000), Brittany (Tait et al.,
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27 664 1999) and Bohemian Massif (Tait et al., 1994). Our results show that pervasive remagnetizations
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30 665 have affected the Silurian-Devonian limestones of the Pyrenees during, at least, two episodes: late
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32 666 Carboniferous-Early Permian and at the end of the Alpine orogeny. Our site BN1-OR15 was
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35 667 collected in the vicinity of those from Tait et al. (2000) and shows the same direction in geographic
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37 668 coordinates (Fig. 9), but within-site clustering worsens after bedding correction (Table 2). After
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39 669 Alpine tilt correction, however, a very good fit results with the majority of our Silurian-Devonian
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42 670 collection. In addition, site BN1-OR15 contains pyrrhotite (Fig. 3), a feature common to all the
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44 671 other samples studied (Fig. 9) and a secondary mineral in (meta)sediments indicative of
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46 672 remagnetization (e.g. Pastor-Galán et al., 2017; Izquierdo-Llavall et al., 2020). We therefore conclude
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48 673 that the originally published data by Tait et al. (2000) also reflect a remagnetization. Since the other
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51 674 paleomagnetic results from the putative Armorican s.l. continent are similar to those from the
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53 675 Pyrenees and come from areas with intense Carboniferous deformation and enhanced thermal
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3 676 activity, we suspect that they could be remagnetized as well. We request using those paleolatitudes
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5 677 (Tait et al., 1994; 1999) with caution; their paleomagnetic veracity warrants to be reassessed.
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8 678 Despite the inherent loss of information due to the remagnetization, especially regarding the
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10 679 potential paleolatitudinal constraints, the Silurian-Devonian rocks of the Pyrenees do provide
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12 680 interesting insights. Based on the inclination data we interpret that the sampled rocks mostly
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14 681 remagnetized during late Carboniferous and Permian times when Iberia was located around the
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16 682 equator, thus previous to the Alpine orogeny. Our paleomagnetic results show a positive inclination
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18 683 only fold test when correcting the Alpine tilt inferred from the cross-sections but negative outcrop
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20 684 scale fold tests when using the bedding parameters. Thus, paleomagnetism in combination with
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22 685 detailed structural analysis, is a reliable tool to unravel the deformation style of multi-phase orogens
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24 686 like the Pyrenees. With our paleomagnetic data we now can separate the effects of Alpine and
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26 687 Variscan orogeny in the Silurian-Devonian carbonate series, something that classically is deemed
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28 688 challenging (e.g. Casas et al., 2019). With our data we can say that the Variscan orogeny was
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30 689 responsible for the main folding event observed in the Silurian and Devonian rocks since all
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32 690 remagnetizations are post-folding (Figs. 10 and 12). In contrast, our data supports that the tilting
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34 691 observed in the main Pyrenean thrusts postdates the late Carboniferous-Permian remagnetization.
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36 692 Consequently, those structures formed during the Alpine orogeny.
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43 693 After the Alpine tilt correction, we obtained a relatively good agreement in inclinations, but
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45 694 declinations are still really scattered from SE to SW. Alpine vertical axis rotations in the Pyrenees are
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47 695 frequent and very variable with magnitudes ranging from a few degrees to up to 80° both clockwise
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49 696 and counterclockwise (e.g. Sussman et al., 2004; Rodríguez et al., 2016). Although it is plausible that
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51 697 the basement also underwent significant vertical axis rotations, very little is known about the
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53 698 rotational activity of Pyrenean basement thrusts during the Alpine orogeny. The declinations
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3 699 observed in the Silurian-Devonian rocks of the Pyrenees are generally clockwise with respect to the
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5 700 Permian reference pole for stable Iberia (Weil et al., 2010; Oliva et al., 2012) ranging from a few
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7 701 degrees to ca. 90° (Fig. 1). We note that such results are in line with the paleomagnetic results from
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9 702 Carboniferous and Permian igneous rocks in the Pyrenees and Catalan coastal ranges (Edel et al.,
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11 703 2018). The Pyrenees lay in the northern branch of the Cantabrian Orocline, which rotated clockwise
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13 704 during the Late Carboniferous and Early Permian (e.g. Pastor-Galán et al., 2015b; Pastor-Galán,
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15 705 2020). Edel et al. (2018) interpreted their results as consistent with the rotations expected in the
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17 706 northern branch of the of the Cantabrian Orocline. Izquierdo-Llavall et al. (2014) also found similar
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19 707 data in late Carboniferous and Early Permian rocks of the Pyrenees and interpreted them as an
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21 708 Alpine rotation. Therefore, we would like to remain wary about their meaning. The variety of
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23 709 rotations found might be reflecting: (i) Differential timing of the remagnetization which occurred
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25 710 widely during the Cantabrian Orocline formation as observed in other areas of Iberia (e.g. Pastor-
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27 711 Galán et al., 2017; 2020); (ii) Vertical axis rotations associated with the Alpine orogeny (Izquierdo-
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29 712 Llavall et al., 2014); or (iii) a combination of both processes where the Alpine rotations may be
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31 713 opposite to and/or in the same sense as the late Carboniferous clockwise rotations.
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38 714 4.4 A blessing in disguise

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41 715 The Pyrenees are a multi-orogenic mountain range whose kinematics is often complicated
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43 716 due to the superposition of different deformation events. Accumulation of geological processes,
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45 717 many of them involving relatively high temperatures and fluid percolation increases the chances of
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47 718 remagnetization for the rocks involved in the orogenies. In fact, we suspect that the majority of the
48
49 719 Silurian-Devonian carbonate series of the Pyrenees won't preserve a primary and syn-sedimentary
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51 720 magnetization. This makes the Pyrenees, despite the great outcrop quality and quantity a bad
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53 721 candidate to study pre-Variscan plate motions and kinematics. However, we found generally strong
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3 722 magnetizations containing with univocal ChRMs. Our paleomagnetic data, in combination with
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5 723 detailed structural observations, has proven the best way so far to unravel the complex tectonic
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7 724 evolution of the Axial Zone of the Pyrenees. We think that at least the Devonian carbonate rocks,
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10 725 but likely other Paleozoic series and igneous rocks are excellent targets to study: (1) the Variscan -
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12 726 Alpine structural relationships, (2) the Alpine rotational history of the basement thrusts; and perhaps
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14 727 (3) the late Variscan deformation events leading to the final amalgamation of Pangea.
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18 728 5. Conclusions and caution for paleomagnetists

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- 22 729 • The Silurian-Devonian carbonate series of the Pyrenees show varying amounts of pyrrhotite,
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24 730 a secondary magnetic mineral, and negative fold tests using bedding parameters, which
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26 731 indicate widespread remagnetization(s).
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- 29 732 • The majority of sites that passed the quality criteria ($n = 7$ and $k > 8$) show a positive
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31 733 inclination only fold test when correcting the Alpine tilt (with the exceptions of OG03 and
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33 734 OG04). The obtained inclinations are southerly and very shallow; they constrain the
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35 735 remagnetization to a reverse chron when Iberia was around the equator, only possible during
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37 736 late Carboniferous or early Permian times.
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- 40 737 • Sites OG03 and OG04 (Western Pyrenees, Gavarnie thrust sheet) were likely remagnetized
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42 738 after the main Alpine thrusting, during a pervasive burial remagnetization widely observed in
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44 739 the Internal Sierras and other along-strike equivalent units (Bóixols, Cadí).
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- 47 740 • Paleomagnetism from the Silurian and Devonian rocks suggest that the Variscan orogeny
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49 741 was responsible for their main folding event, whereas the Alpine orogeny produced their
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51 742 thrusting and antiformal stacking.
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3 743 • Our results also show general clockwise rotations which may be consistent with the northern
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5 744 branch of the Cantabrian Orocline. These rotations may as well represent Alpine vertical axis
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7 745 rotations or a combination of both.
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10 746 • Given the generally good paleomagnetic quality of the Devonian carbonates, they could be
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12 747 targeted to study the Alpine imprint on Paleozoic rocks and thus, unravelling the rotational
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14 748 history of basement thrusts.
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17 749 • The widespread remagnetizations found in the Paleozoic of the Pyrenees indicate that
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19 750 paleolatitudes inferred from Silurian and Devonian are very unlikely original and should be
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21 751 taken very cautiously. We urge a reassessment of Siluro-Devonian poles from the Variscan in
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23 752 Europe.
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26 753 • Paleomagnetism from multi-orogenic areas is NOT A SIMPLE GAME. However, the
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28 754 systematic combination of paleomagnetism with detailed structural observations, seems to
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30 755 be a foremost way to unravel complex tectonic evolutions.
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37 757 Acknowledgements

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41 758 This work was funded by postdoctoral (ISES) grant from NWO to DPG and the projects
42
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44
45 760 PID2019-104693GB-I00/CTA) from the Spanish Ministry of Science. We thank Mat Domeier
46
47 761 for providing help with the inclination only statistics. DPG thanks Edward Lodewijk Van Halen
48
49
50 762 for all times making him jump, we will keep on jumping.
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764 Data availability statement

765 All data is included in the paper and supplementary materials. In Addition, paleomagnetic data is
766 stored in paleomagnetism.org under the persisitent identifier (PID)
767 1871091757a6ef9d46ac59bb9f35c7a6387bc8fcece7bf0715eaba29164fbc7e and can be accessed
768 in the link <https://www.paleomagnetism.org/library/>.

770 Figure and Table captions

771 Figure 1: A) Simplified map of the Iberian Peninsula showing the main Paleozoic outcrops and
772 the areas affected by the Alpine orogeny (after Pastor-Galán et al., 2020). B) Geological map of
773 the Pyrenees (modified from Barnolas et al., 2008 according to Choukroune and Seguret, 1973)
774 showing our sampling locations (red dots) and the extensive paleomagnetic studies in the
775 Pyrenees focused mainly on Permo-Triassic rocks (blue dots). Lines show the trend of the cross
776 sections in figure 2.

777 Figure 2: Cross sections through the Pyrenees with projected positions of our sampling sites.

778 Figure 3: Selected magnetization vs. temperature curves (OG8, OG13, OG14) and susceptibility
779 vs. temperature (BN1). Note that magnetic and non magnetic sulfides are common. All
780 measurements performed are available in supplementary file SF1.

781 Figure 4: Selected slope corrected hysteresis loops and FORC diagram (plotted with FORCINEL
782 (Harrison and Feinberg, 2008, smoothing factor 13). All measurements performed are available
783 in supplementary file SF2.

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3 784 Figure 5: A) Unmixing of IRM acquisition curve for three samples showing different proportions
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5 785 of a ‘soft’ mineral that saturates below 75 mT (magnetite) and a ‘harder’ one that saturates over
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7 786 200 mT (pyrrhotite). B) Results from the endmember modeling showing our preferred two end-
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9 787 member solution (left) and the four end-member solution (right). C) Unmixing of the IRM
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11 788 synthetic acquisition curve from the two end members showing the same two magnetic
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13 789 mineralogies as in the forward modeling of different samples: magnetite to the left and pyrrhotite
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15 790 to the right.
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20 791 Figure 6: Examples of ‘Zijderveld’ (Zijderveld, 1967) vector-end point plots for selected
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22 792 samples. All samples plotted in geographic coordinates. Close-open circles represent declination
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24 793 and inclination projections respectively. Complete analyses are available in paleomagnetism.org
25
26 794 through the Persistent identifier PID given in the acknowledgements.
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30 795 Figure 7: Viscous remanent magnetization (VRM) from all samples is compatible with the
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32 796 present-day field (geographic coordinates). Red dots are those that fall outside of the 45° cut-off.
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34 797 Uncertainty envelope is in both cases VGP A95. The rather large scattering is likely due to the
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36 798 small number of demagnetization levels containing the VRM (3-4) and the possible migration of
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38 799 the VRM during transport, storage and analysis.
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43 800 Figure 8: Directional and VGP results in geographic coordinates of sites OG01 to OG11.
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45 801 Uncertainty envelope is in both cases VGP A95. Red dots are those that fall outside of the 45°
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47 802 cut-off. Sites OG02, 05 and 08 did not provide statistically meaningful results and were not
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49 803 interpreted.
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53 804 Figure 9: Directional and VGP results in geographic coordinates of sites OG12 to OG19 and
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55 805 BN1-OR15. Uncertainty envelope is in both cases VGP A95. Red dots are those that fall outside
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3 806 of the 45° cut-off. Sites OG02, 05 and 08 did not provide statistically meaningful results and
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5 807 were not interpreted.
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9 808 Figure 10: Within-site fold tests. All are negative but OG11, which is inconclusive.
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12 809 Figure 11: Pi diagrams for the studied folds. Only OG11 shows a steeply plunging axis.
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15 810 Figure 12: Results after the inferred Alpine tilt correction. Permian reference declination for
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17 811 Iberia is after Weil et al. (2010). The results show a positive inclination only tilt test following
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19 812 the methodology of Enkin and Watson (1996) (selected bootstraps in thin gray lines) and Arson
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21 813 and Levi (2010) approach (dashed line).
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25 814 Figure 13: OG03 and OG04 show a common true mean bootstrapped direction (after Tauxe,
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27 815 2010) in geographic (and Alpine tilt corrected) coordinates.
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31 816 Figure 14: Results from the anisotropy of the magnetic susceptibility analyses. Magnetic fabrics
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33 817 represent bedding or S1 cleavage. Magnetic fabric directions do not coincide with the
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35 818 paleomagnetic directions, which allows us to discard an internal deformation control of the
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37 819 paleomagnetic remanence.
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41 820 Table 1: Site Location and key structural data from each site.
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45 821 Table 2: Paleomagnetic results for all sites in geographic and tilt (bedding) corrected coordinates.
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48 822 Table 3: Paleomagnetic results after the Alpine tilt correction (tilt associated to the emplacement
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50 823 of the thrusts).
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53 824 Table 4: Concentration parameters of inclination only statistics from the mean values of the sites.
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3 825 Supplementary Files:
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6 826 SF1: All thermomagnetic curves analyzed.
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10 827 SF2: All Hysteresis loops analyzed.
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13 828 SF3: Synthesis of the paleomagnetic results (can be opened with paleomagnetism.org).
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16 829 SF4: KMZ file (Google Earth) with the sampled locations.
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20 830 SF5: Zip file containing all obtained raw results.
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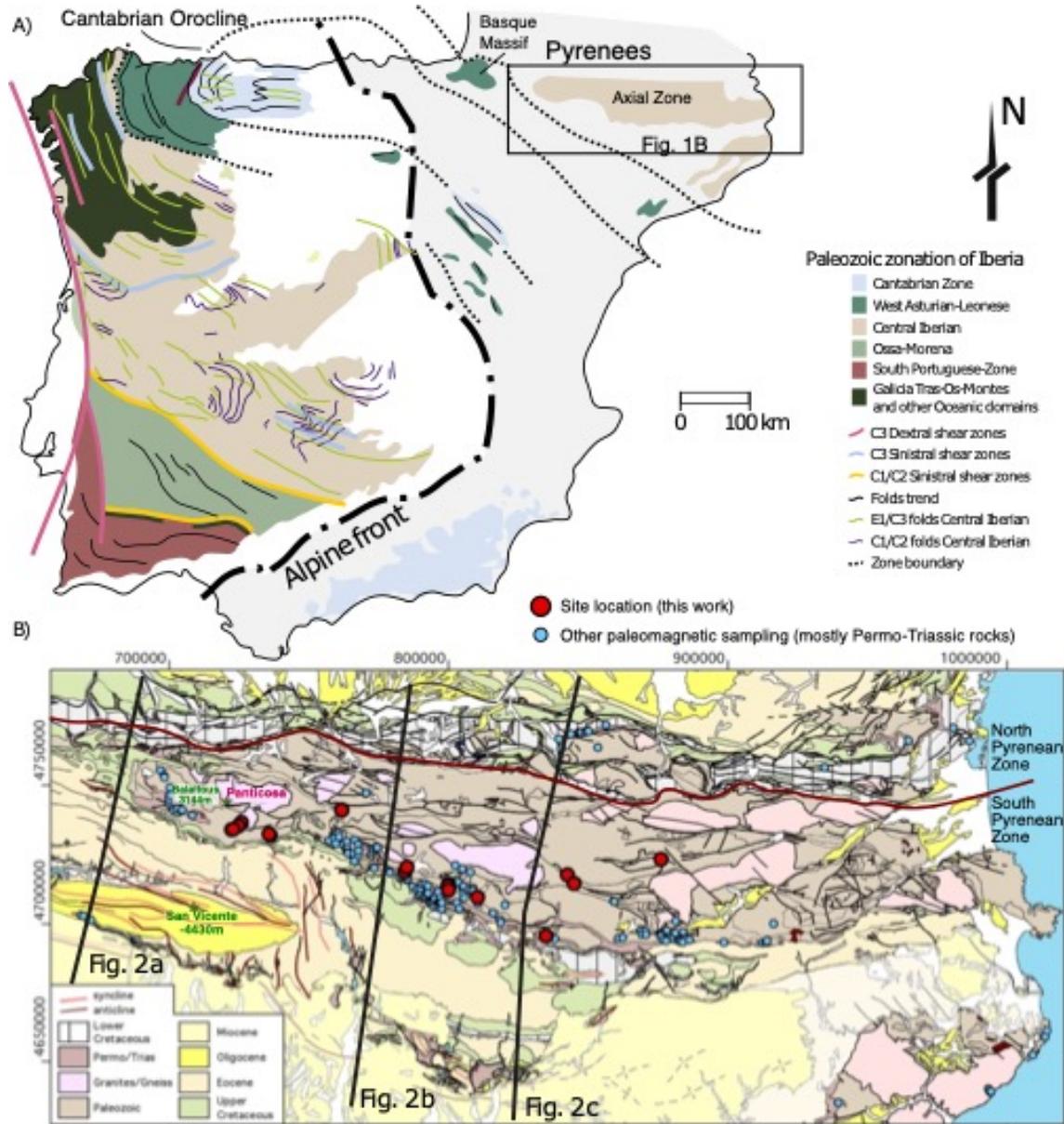
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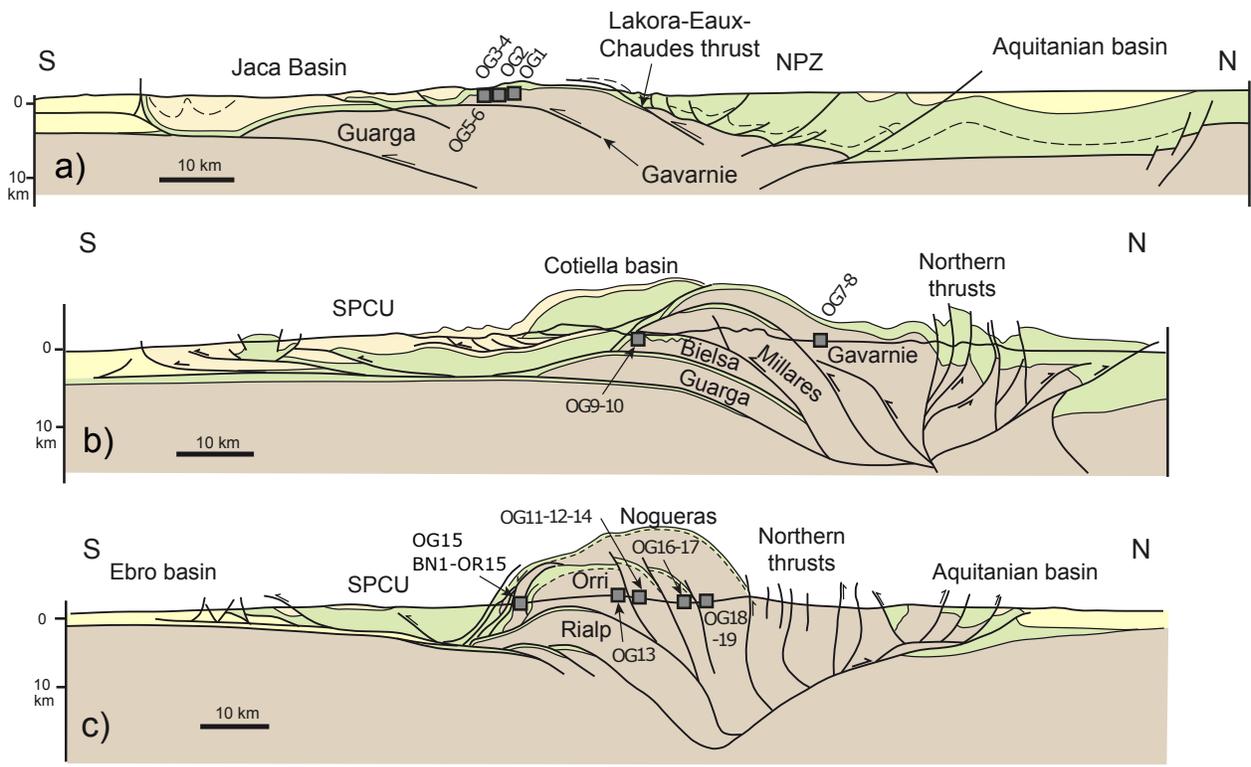
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1199 Figure 1: A) Simplified map of the Iberian Peninsula showing the main Paleozoic outcrops and
 1200 the areas affected by the Alpine orogeny (after Pastor-Galán et al., 2020). B) Geological map of
 1201 the Pyrenees (modified from Barnolas et al., 2008 according to Choukroune and Seguret, 1973)
 1202 showing our sampling locations (red dots) and the extensive paleomagnetic studies in the
 1203 Pyrenees focused mainly on Permo-Triassic rocks (blue dots). Lines show the trend of the cross
 1204 sections in figure 2.

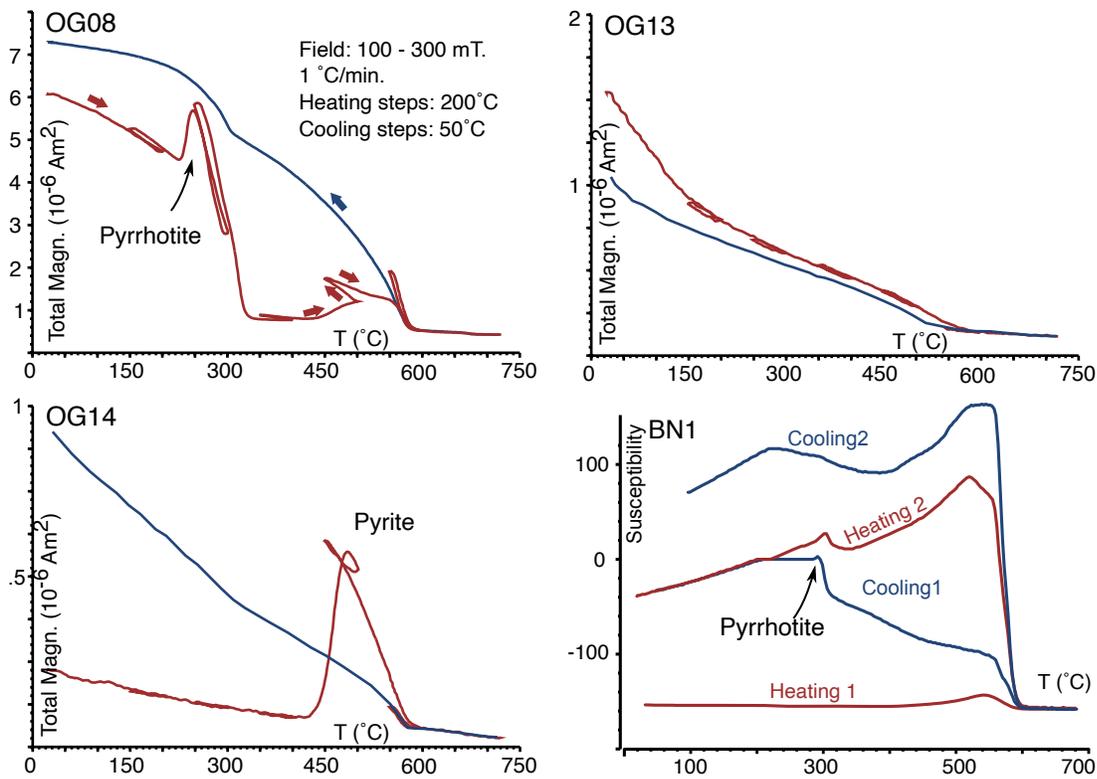
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Figure 2: Cross sections through the Pyrenees with projected positions of our sampling sites.

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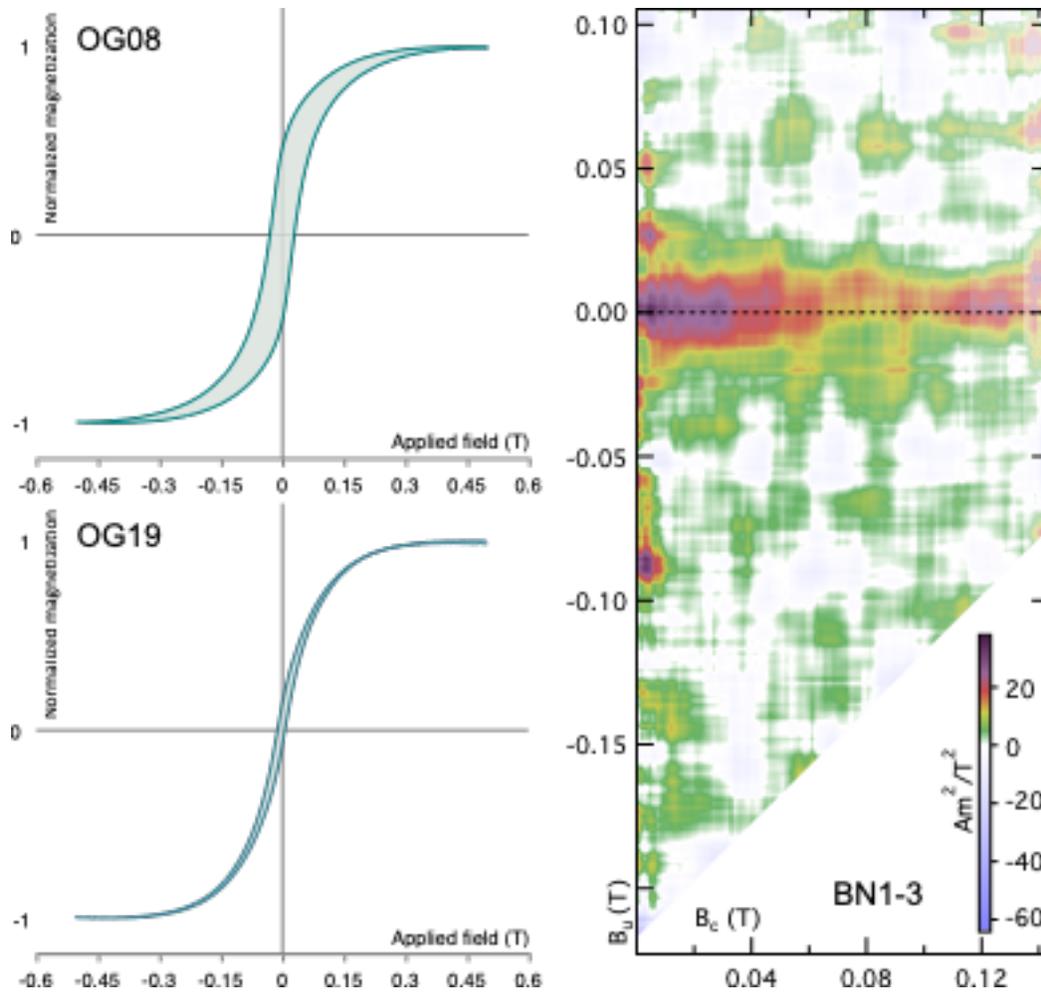
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1210 Figure 3: Selected magnetization vs. temperature curves (OG8, OG13, OG14) and susceptibility

1211 vs. temperature (BN1). Note that magnetic and non magnetic sulfides are common. All

1212 measurements performed are available in supplementary file SF1.

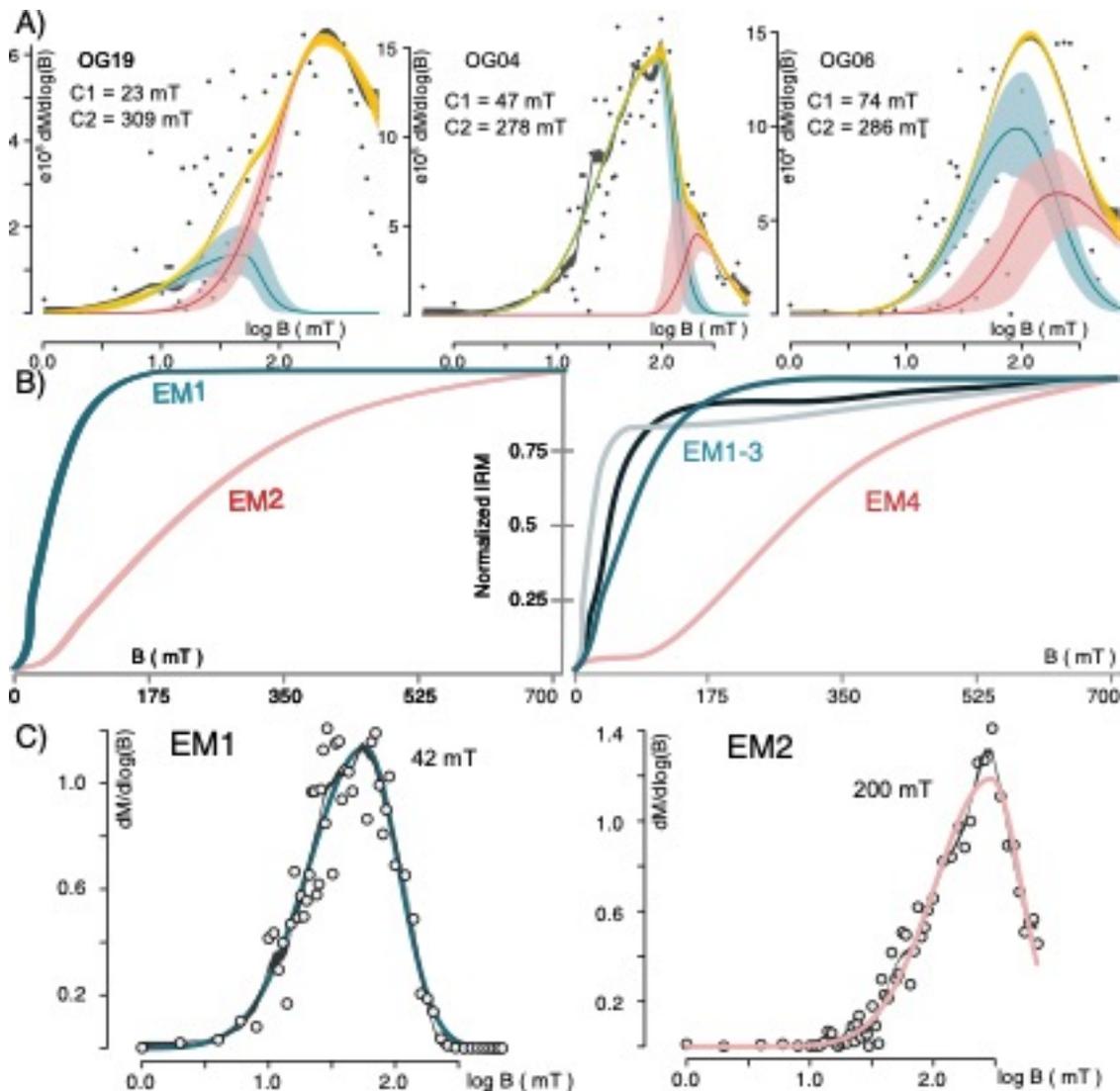
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1215 Figure 4: Selected slope corrected hysteresis loops and FORC diagram (plotted with FORCINEL
 1216 (Harrison and Feinberg, 2008, smoothing factor 13). All measurements performed are available
 1217 in supplementary file SF2.

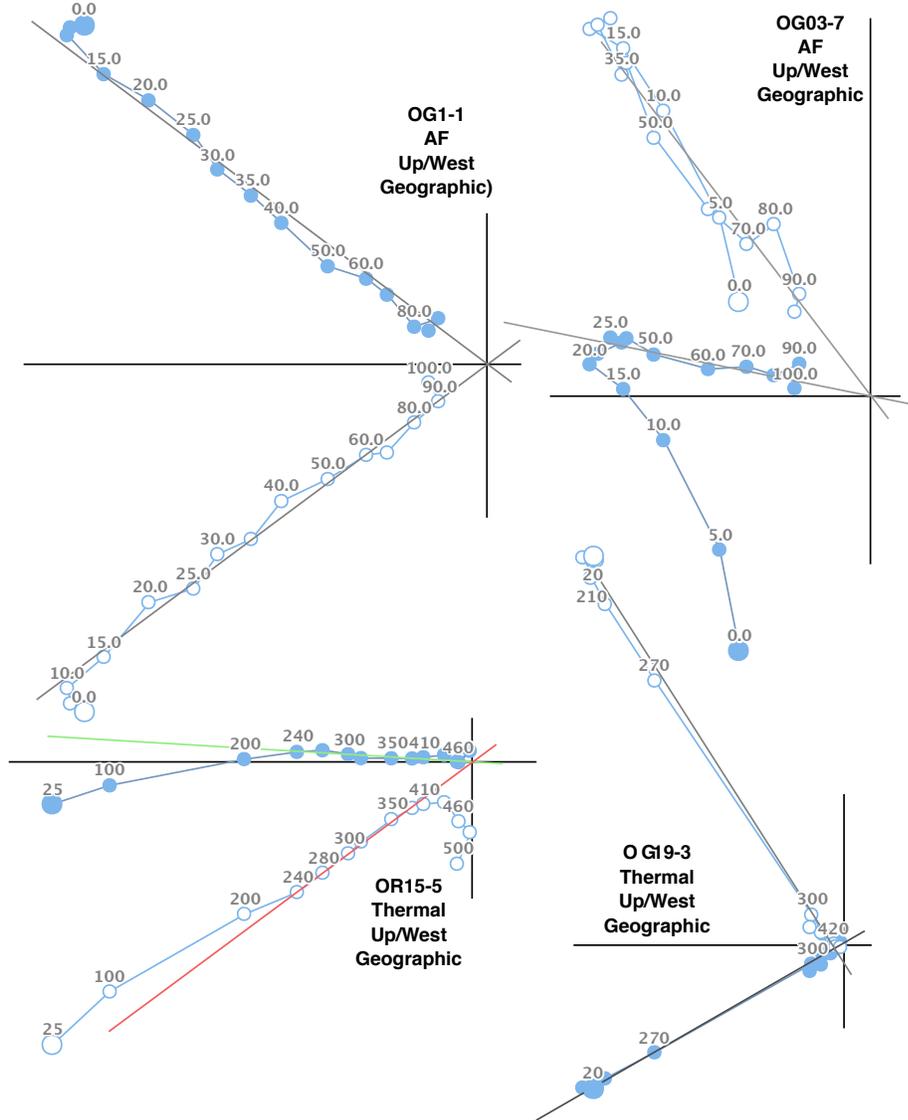
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1220 Figure 5: A) Unmixing of IRM acquisition curve for three samples showing different proportions
 1221 of a ‘soft’ mineral that saturates below 75 mT (magnetite) and a ‘harder’ one that saturates over
 1222 200 mT (pyrrhotite). B) Results from the endmember modeling showing our preferred two end-
 1223 member solution (left) and the four end-member solution (right). C) Unmixing of the IRM
 1224 synthetic acquisition curve from the two end members showing the same two magnetic
 1225 mineralogies as in the forward modeling of different samples: magnetite to the left and pyrrhotite
 1226 to the right.

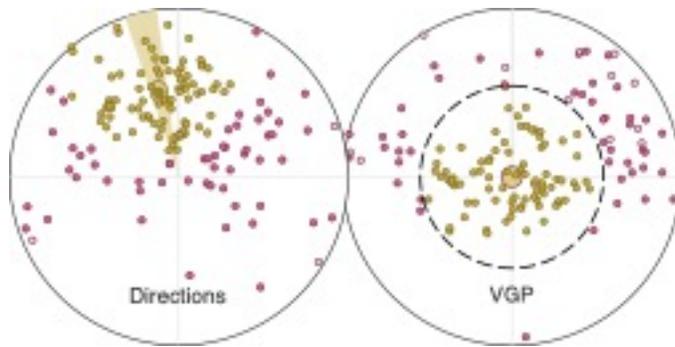
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1229 Figure 6: Examples of 'Zijderveld' (Zijderveld, 1967) vector-end point plots for selected
 1230 samples. All samples plotted in geographic coordinates. Close-open circles represent declination
 1231 and inclination projections respectively. Complete analyses are available in paleomagnetism.org
 1232 through the Persistent identifier PID given in the acknowledgements.

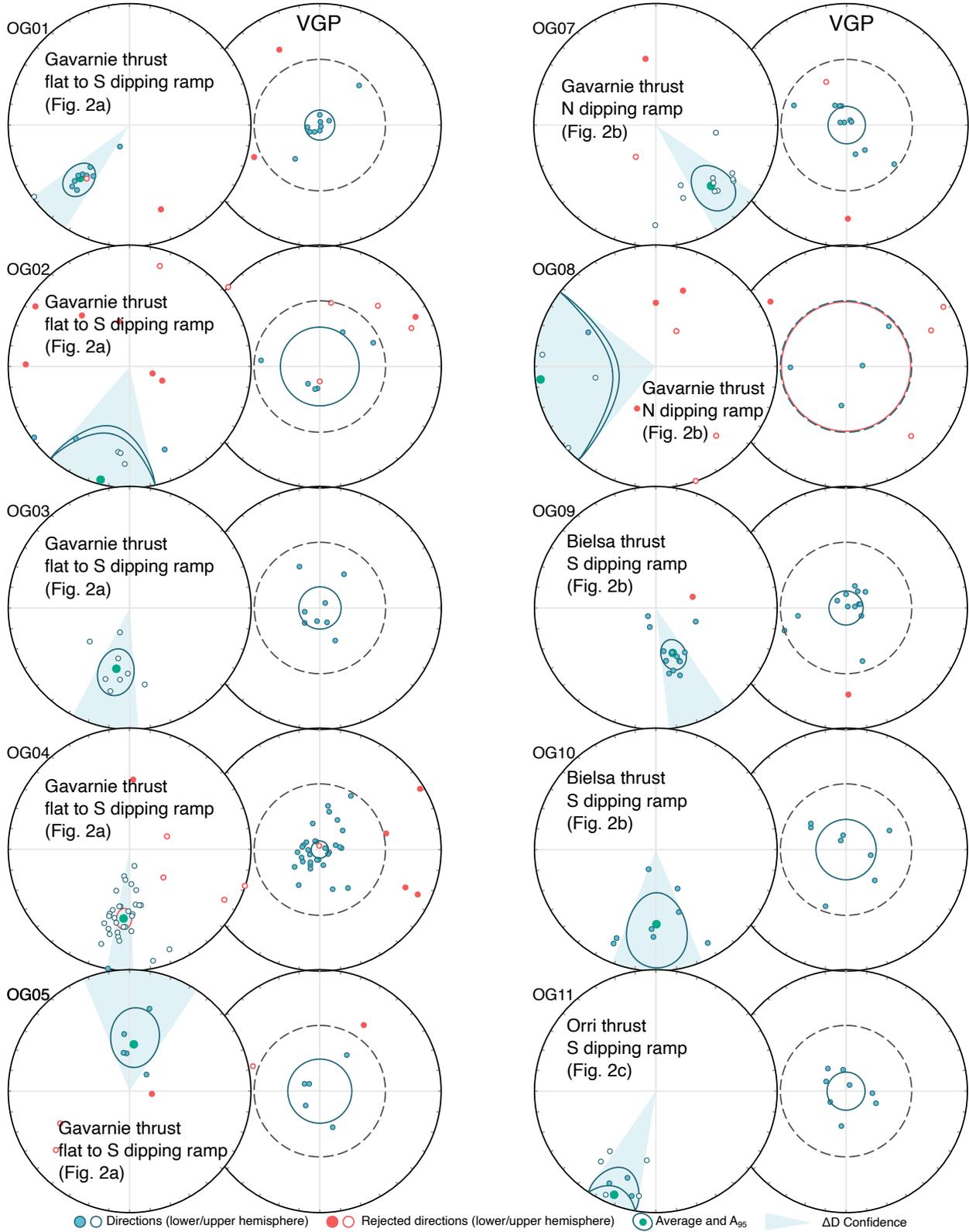
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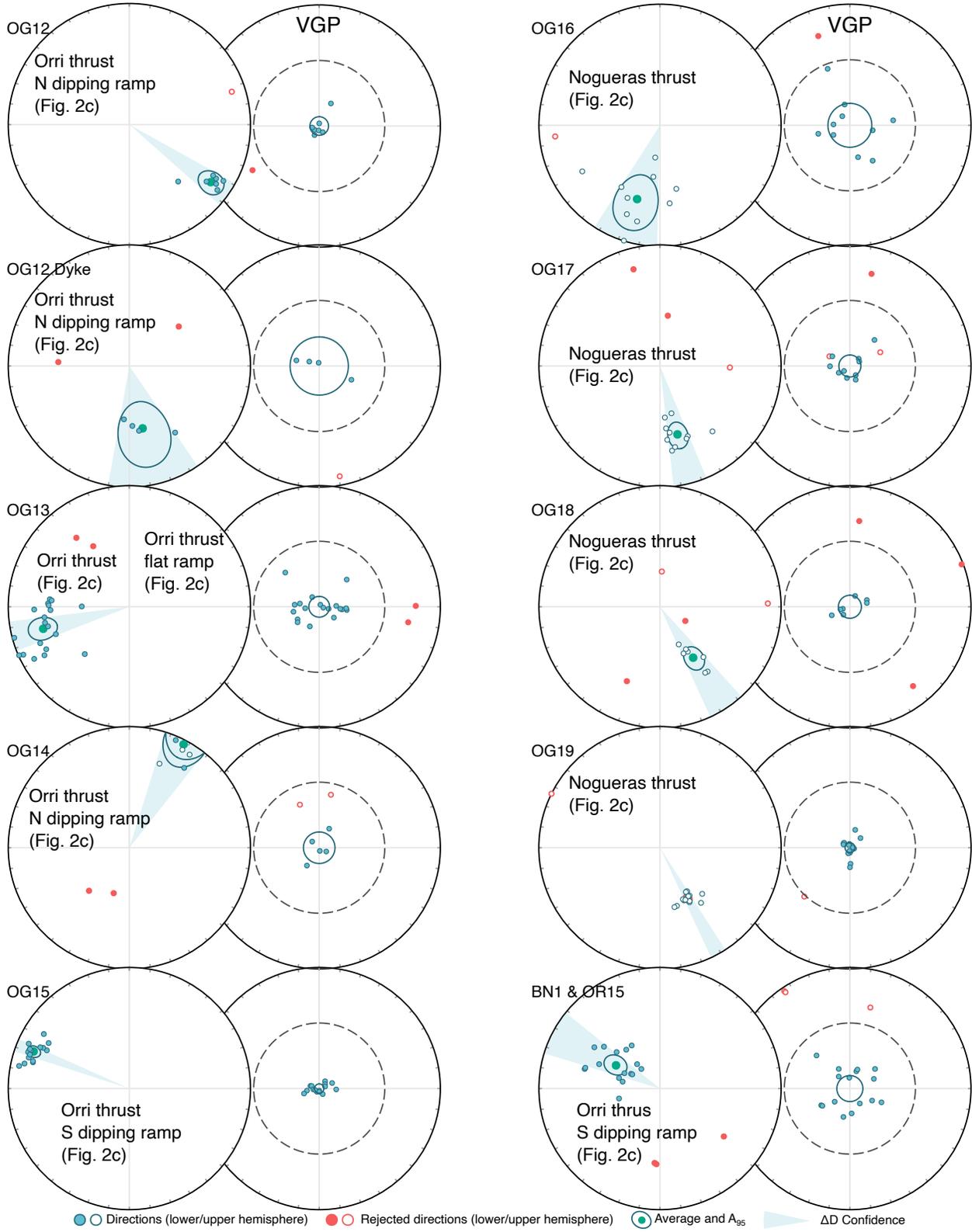
1235 Figure 7: Viscous remanent magnetization (VRM) from all samples is compatible with the
1236 present-day field (geographic coordinates). Red dots are those that fall outside of the 45° cut-off.
1237 Uncertainty envelope is in both cases VGP A95. The rather large scattering is likely due to the
1238 small number of demagnetization levels containing the VRM (3-4) and the possible migration of
1239 the VRM during transport, storage and analysis.

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3 1242 Figure 8: Directional and VGP results in geographic coordinates of sites OG01 to OG11.
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5 1243 Uncertainty envelope is in both cases VGP A95. Red dots are those that fall outside of the 45°
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7 1244 cut-off. Sites OG02, 05 and 08 did not provide statistically meaningful results and were not
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9 1245 interpreted.
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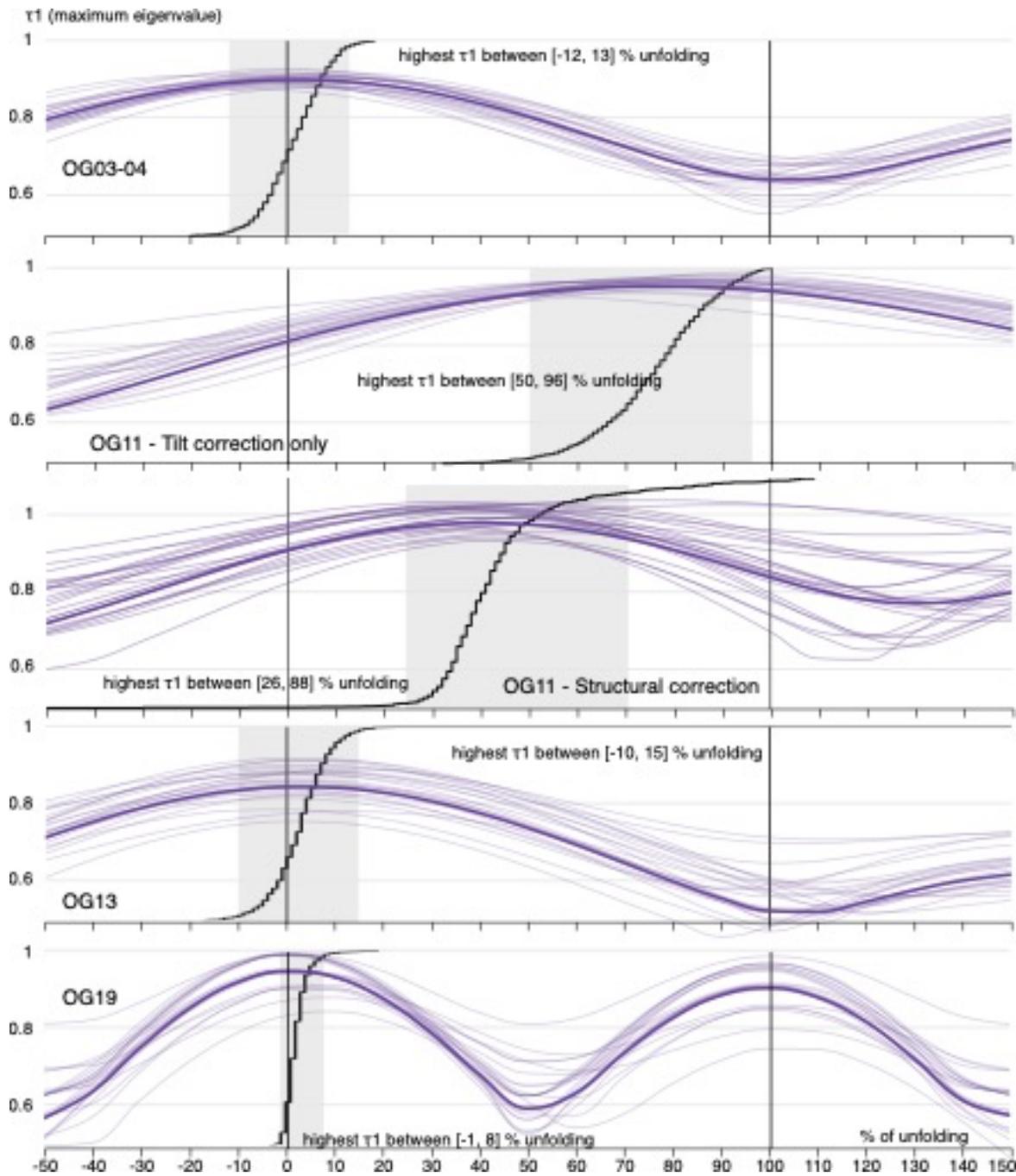


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5 1249 BN1-OR15. Uncertainty envelope is in both cases VGP A95. Red dots are those that fall outside
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7 1250 of the 45° cut-off. Sites OG02, 05 and 08 did not provide statistically meaningful results and
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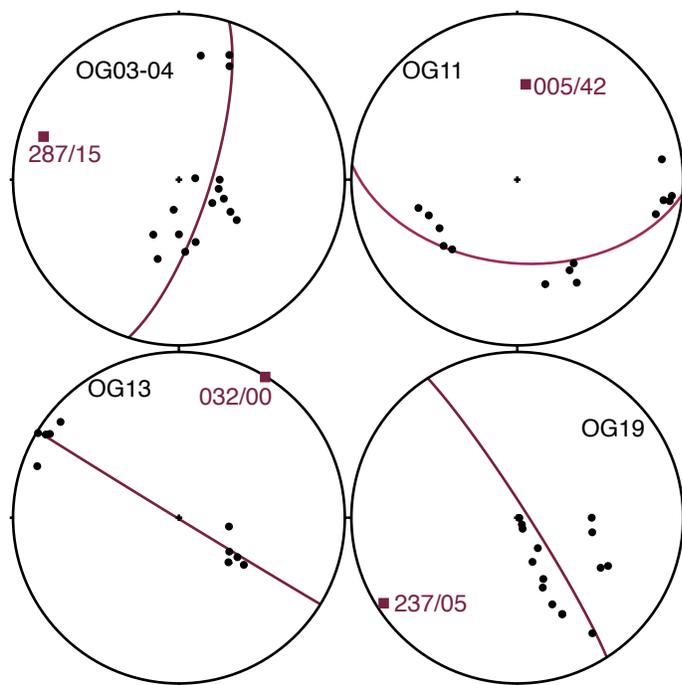
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1254 Figure 10: Within-site fold tests. All are negative but OG11, which is inconclusive.

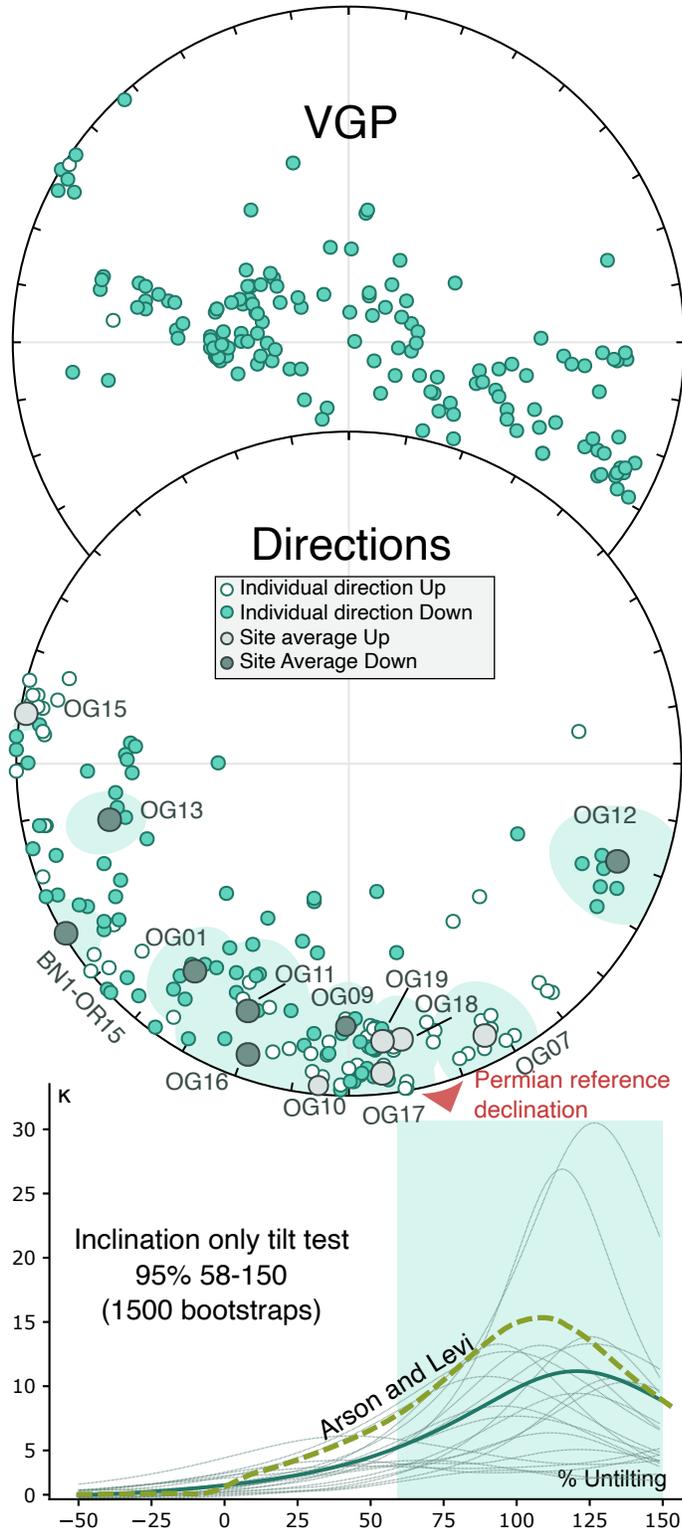
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1257 Figure 11: Pi diagrams for the studied folds. Only OG11 shows a steeply plunging axis.

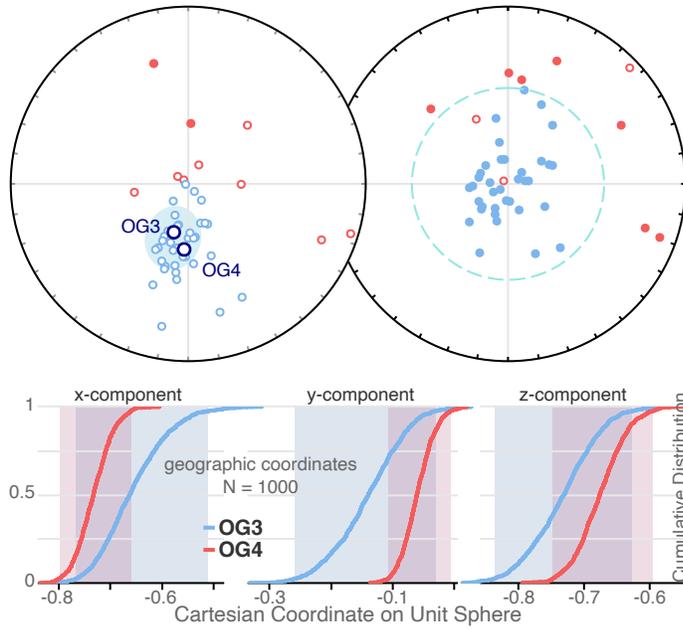
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1260 Figure 12: Results after the inferred Alpine tilt correction. Permian reference declination for
 1261 Iberia is after Weil et al. (2010). The results show a positive inclination only tilt test following

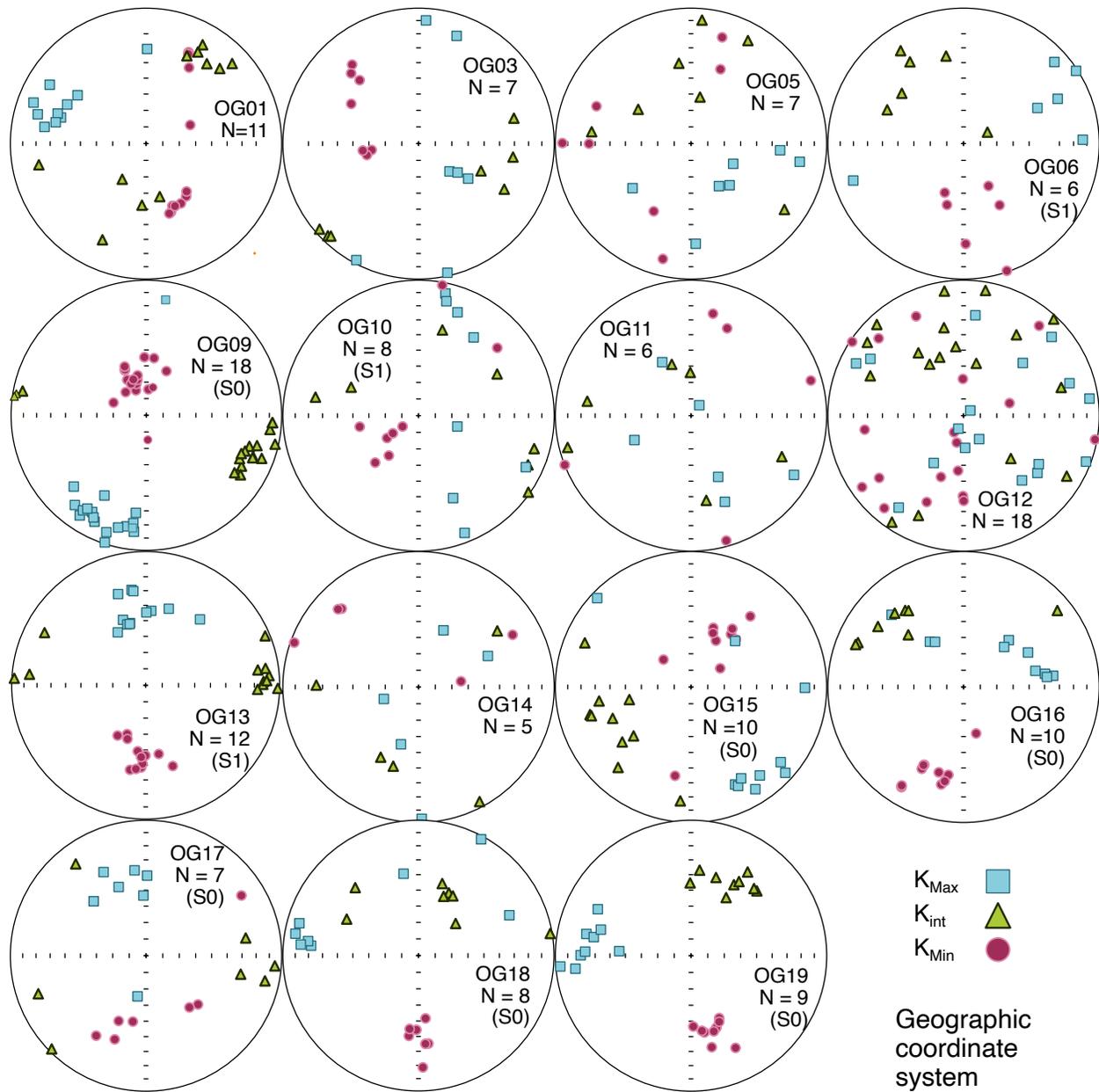
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1266 Figure 13: OG03 and OG04 show a common true mean bootstrapped direction (after Tauxe,
1267 2010) in geographic (and Alpine tilt corrected) coordinates.

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1270 Figure 14: Results from the anisotropy of the magnetic susceptibility analyses. Magnetic fabrics
 1271 represent bedding or S1 cleavage. Magnetic fabric directions do not coincide with the
 1272 paleomagnetic directions, which allows us to discard an internal deformation control of the
 1273 paleomagnetic remanence.

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1275 Table 1: Site Location and key structural data from each site.

Structure	S0 (Bedding)		Fold Axis		S1		Inferred Alpine tilt		Location	
	Dip Direction	Dip	Trend	Plunge	Dip direction	Dip	Dip Direction	Dip	Latitude	Longitude
OG01	-	-	-	-	-	-	185	20	42.7473	-0.24497
OG02*	Fold		191	14	-	-	185	20	42.7361	-0.25841
OG03	199	66	287	15	-	-	185	20	42.7249	-0.28900
OG04	Fold				20	60	185	20	42.7259	-0.28300
OG05*	299	17	-	-	-	-	180	20	42.7023	-0.12022
OG06*	358	62	-	-	256	78	180	20	42.7064	-0.12464
OG07	36	42	-	-	35	55	25	45	42.7771	0.19758
OG08*	10	36	-	-	-	-	25	45	42.7759	0.19858
OG09	89	13	-	-	-	-	205	45	42.5673	0.46444
OG10	17	31	-	-	-	-	205	45	42.5824	0.47447
OG11	Fold		5	42	-	-	25	30	42.5222	0.65244
OG12	40	39	-	-	-	-	25	30	42.5167	0.65100
OG13	Fold		32	0	-	-	25	0	42.5028	0.65228
OG14*	Fold		292	19	-	-	25	30	42.4752	0.77733
OG15	234	48	-	-	-	-	205	90	42.3380	1.06883
OG16	16	51	-	-	-	-	25	45	42.5339	1.17550
OG17	17	50	-	-	-	-	25	45	42.5048	1.20275
OG18	359	51	-	-	254	67	25	45	42.5689	1.59106
OG19	Fold		237	5	250	60	25	45	42.5682	1.58550
BN1 & OR15	185	25	-	-	-	-	205	90	42.3220	1.10466

*Unreliable paleomagnetic result

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1279 Table 2: Paleomagnetic results for all sites in geographic and tilt (bedding) corrected coordinates.

Geographic	N	Ns	Cutoff	S	Dec	Inc	R	k	a95	K	A95	A95min	A95max	ΔDx	ΔIx	λ
OG01	10	12	45	16.7 1	222.7	40.19	9.48	17.27	11.96	24.19	10.02	4.78	19.22	10.89	13.79	22.9
OG02*	6	13	45	30.3 8	194.24	-4.41	4.97	4.86	33.88	7.34	26.5	5.86	26.52	26.52	52.76	-2.21
OG03	8	8	45	20.2 3	191.98	-47.6	7.66	20.67	12.47	16.32	14.13	5.22	22.12	16.16	16.7	-28.7
OG04	32	37	45	18.0 8	184.57	-42.53	30.18	17.02	6.34	20.49	5.75	3	9.24	6.33	7.56	-24.64
OG05*	5	8	45	22.1 6	5.71	58.01	4.78	17.93	18.57	13.61	21.51	6.3	29.75	28.01	19.81	38.68
OG06*	2	3	45	24.0 6	248.92	-20.54	1.88	8.48	100.98	11.42	82.51	9.09	52.99	NaN	149.7 8	-10.61
OG07	11	13	45	21.7 7	137.77	-32.97	10.37	15.83	11.84	14.29	12.51	4.6	18.1	13.16	19.46	-17.97
OG08*	4	10	45	35.8 5	263.68	-5.38	3.11	3.36	59.54	5.28	44.25	6.89	34.24	44.31	87.91	-2.7
OG09	13	14	45	21.8 7	159.34	57.48	12.55	26.68	8.18	14.19	11.4	4.3	16.29	14.55	10.64	38.1
OG10	8	8	45	28.2 6	179.3	38.56	7.17	8.42	20.28	8.45	20.23	5.22	22.12	21.86	28.68	21.73
OG11	8	8	45	18.2 6	201.72	-9.18	7.18	8.58	20.06	19.89	12.73	5.22	22.12	12.77	24.98	-4.62
OG12	7	8	45	8.33	125.38	18.71	6.89	53.04	8.36	95.15	6.22	5.51	24.07	6.31	11.48	9.61
OG12-Dyke	4	6	45	16.9 9	167.95	46.44	3.9	28.77	17.42	23.01	19.57	6.89	34.24	22.23	23.72	27.73
OG13	19	21	45	16.5 7	255.81	27.7	18.01	18.2	8.09	24.22	6.96	3.7	12.83	7.2	11.67	14.71
OG14*	5	7	45	11.1 1	27.96	-4.07	4.79	19.12	17.95	53.41	10.56	6.3	29.75	10.57	21.05	-2.04
OG15	15	15	45	6.56 23.9 6	291.06	16.61	14.87	107.2 5	3.71	152.91	3.1	4.06	14.89	3.14	5.82	8.48
OG16	10	11	45	12.2 8	197.11	-36.31	9.14	10.48	15.66	11.72	14.72	4.78	19.22	15.71	21.7	-20.18
OG17	10	13	45	10.2 9	165.55	-41.26	9.79	41.99	7.54	43.86	7.38	4.78	19.22	8.06	9.94	-23.69
OG18	7	11	45	188.0 2	146.65	-48.47	6.88	49.71	8.64	62.26	7.71	5.51	24.07	8.86	8.94	-29.45
OG19	15	16	45	6.98	151.24	-50.69	14.93	2	2.8	135.26	3.3	4.06	14.89	3.87	3.64	-31.41
BN1 & OR15	16	19	45	18.7 6	297.88	56.61	15.43	26.13	7.35	18.89	8.71	3.96	14.3	10.96	8.32	37.18
Tilt corrected																
OG01	10	12	45	16.7 1	222.7	40.19	9.48	17.27	11.96	24.19	10.02	4.78	19.22	10.89	13.79	22.9
OG02*	5	13	45	28.8 4	290.83	0.19	4.02	4.07	43.34	8.09	28.63	6.3	29.75	28.63	57.26	0.1
OG03	8	8	45	24.8 1	30.57	-65.75	7.66	20.67	12.47	10.9	17.56	5.22	22.12	26.79	13.23	-47.98
OG04	32	37	45	18.2 7	179.62	-19.66	29.27	11.35	7.89	19.99	5.83	3	9.24	5.92	10.66	-10.13
OG05*	6	8	45	27.9 1	19.38	45.79	5.54	10.9	21.23	8.67	24.12	5.86	26.52	27.35	29.65	27.2
OG06*	2	3	45	16.5 9	230.08	-10.25	1.88	8.48	100.98	23.93	53.52	9.09	52.99	53.83	104.4 9	-5.17
OG07	11	13	45	16.8	156.89	-16.86	10.37	15.83	11.84	23.56	9.61	4.6	18.1	9.72	18	-8.62
OG08*	3	10	45	26.6 9	247.31	-2.42	2.63	5.46	58.88	9.34	42.85	7.73	41.04	42.86	85.58	-1.21
OG09	13	14	45	17.6 1	164.17	45.05	12.55	26.68	8.18	21.64	9.12	4.3	16.29	10.21	11.39	26.6
OG10	6	8	45	30.8 9	169.06	56.53	5.52	10.48	21.7	7.09	27.03	5.86	26.52	34.74	25.85	37.1
OG11	8	8	45	11.7 2	200.07	-4.48	7.75	27.93	10.66	48.06	8.07	5.22	22.12	8.08	16.07	-2.24
OG12	8	8	45	16.1 6	111.8	5.44	7.43	12.25	16.48	25.83	11.11	5.22	22.12	11.12	22.06	2.72
OG12-Dyke	4	6	45	19.2 19.6 6	127.4	56.64	3.85	20.52	20.78	18.07	22.22	6.89	34.24	28.35	21.19	37.21
OG13	11	21	45	183.15	41.74	10.46	18.38	10.94	17.25	11.31	4.6	18.1	12.4	15.1	24.04	
OG14*	5	7	45	21.6	46.47	-55.84	4.79	19.12	17.95	14.34	20.91	6.3	29.75	26.32	20.34	-36.39
OG15	15	15	45	5.25 20.1 8	289.1	-11.31	14.87	107.2 5	3.71	238.05	2.48	4.06	14.89	2.5	4.82	-5.71
OG16	10	11	45	16.5 7	15.07	-14.68	9.14	10.48	15.66	16.44	12.28	4.78	19.22	12.38	23.37	-7.46
OG17	11	13	45	17.9 9	171.02	0.43	10.41	16.87	11.45	24.55	9.4	4.6	18.1	9.4	18.8	0.21
OG18	8	11	45	217.2 2	159.26	-8.83	7.4	11.66	16.93	20.94	12.39	5.22	22.12	12.43	24.35	-4.44
OG19	9	16	45	4.48	155.85	10.2	8.96	2	3.5	327.87	2.85	4.98	20.54	2.86	5.56	5.14
BN1 & OR15	14	19	45	21.4 6	257.17	56	13.43	22.91	8.49	14.47	10.82	4.18	15.55	13.51	10.49	36.55

* Less than n = 7 was not considered

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6 1282 Table 3: Paleomagnetic results after the Alpine tilt correction (tilt associated to the emplacement
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8 1283 of the thrusts).

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11 ALPINE TILT CORRECTION

	Dec	Inc
13 OG01	215.64	23.55
14 OG03	197.28	-67.34
15 OG04	184.57	-60.42
16 OG07	153.46	-8.93
17 OG09	145.47	21.97
18 OG10	180.36	-3.3
19 OG11	185.14	20.77
20 OG12	110.54	15.53
21 OG13	255.81	27.7
22 OG15	278.36	-3.78
23 OG16	198.58	8.37
24 OG17	173.76	-6.78
25 OG18	168.95	-16.46
26 OG19	172.82	-16.4
27 BN1 & OR15	238.36	1.58

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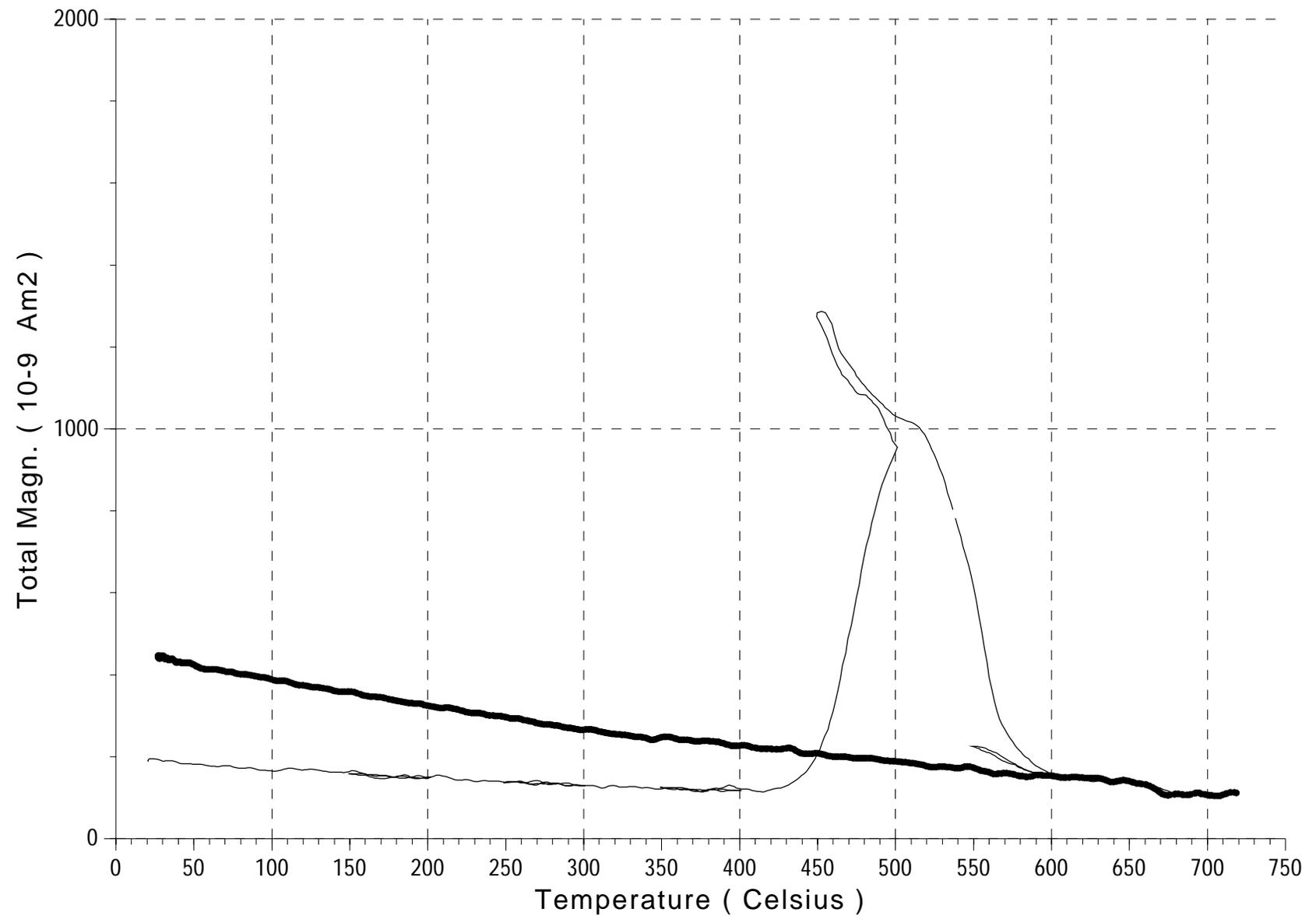
1286 Table 4: Concentration parameters of inclination only statistics from the mean values of the sites.

Inclination only statistics				
Geographic	k	Inclination		α_{95}
Site means avg.		0	0	90
Bedding corrected				
Site means avg.		2.17	18.25	38.91
Site means avg. but OG03 & 04		3.93	22.67	25.44
Alpine tilt corrected				
Site means avg.		4.19	-3.4	21.25
Site means avg. but OG03 & 04		14.42	5.08	11.3

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OG2----TOTAL

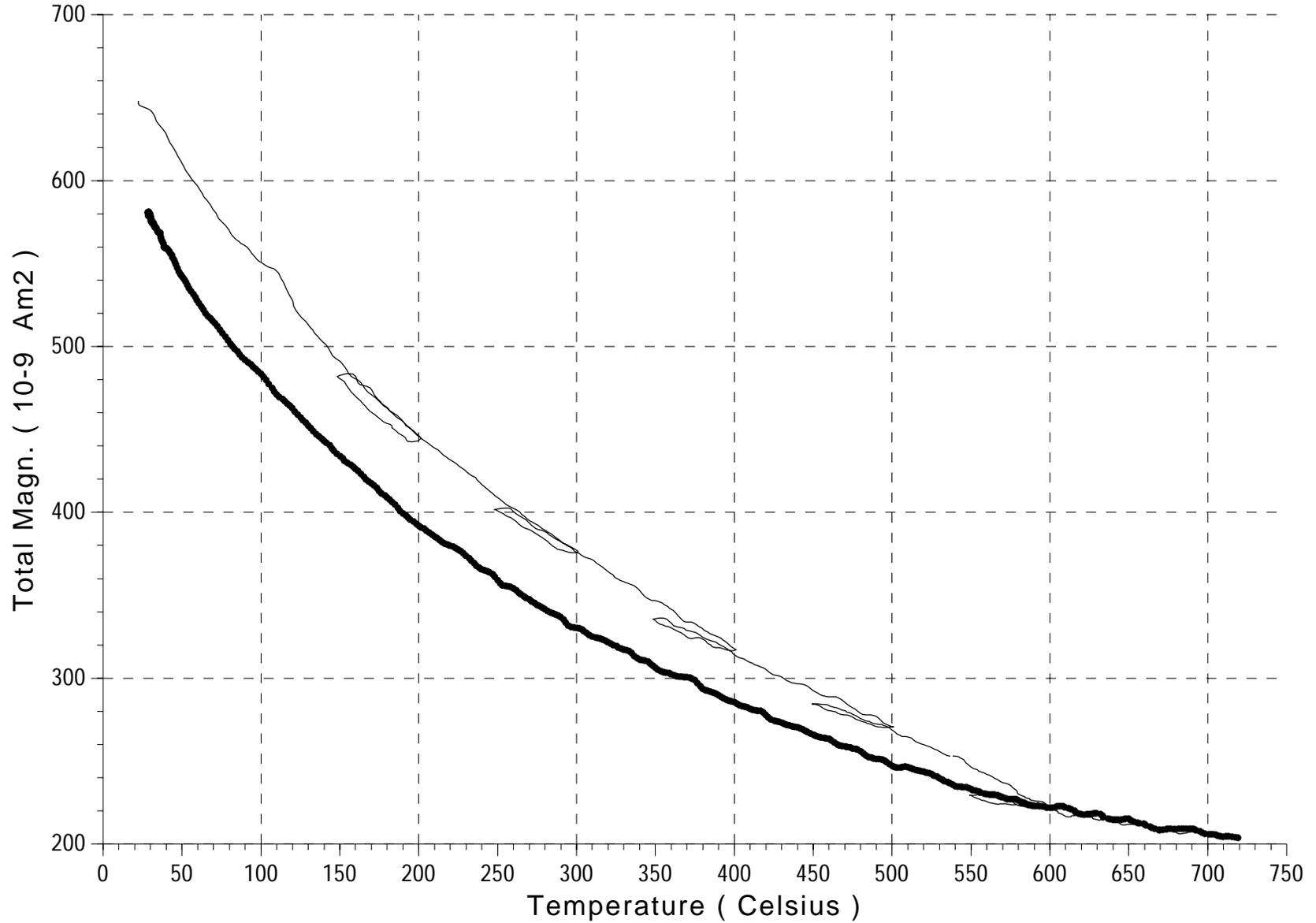
Field: 100 - 300 mT. 10/ 10 T/min. Weight: .06682. Max. temp.: 200



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OG3----TOTAL

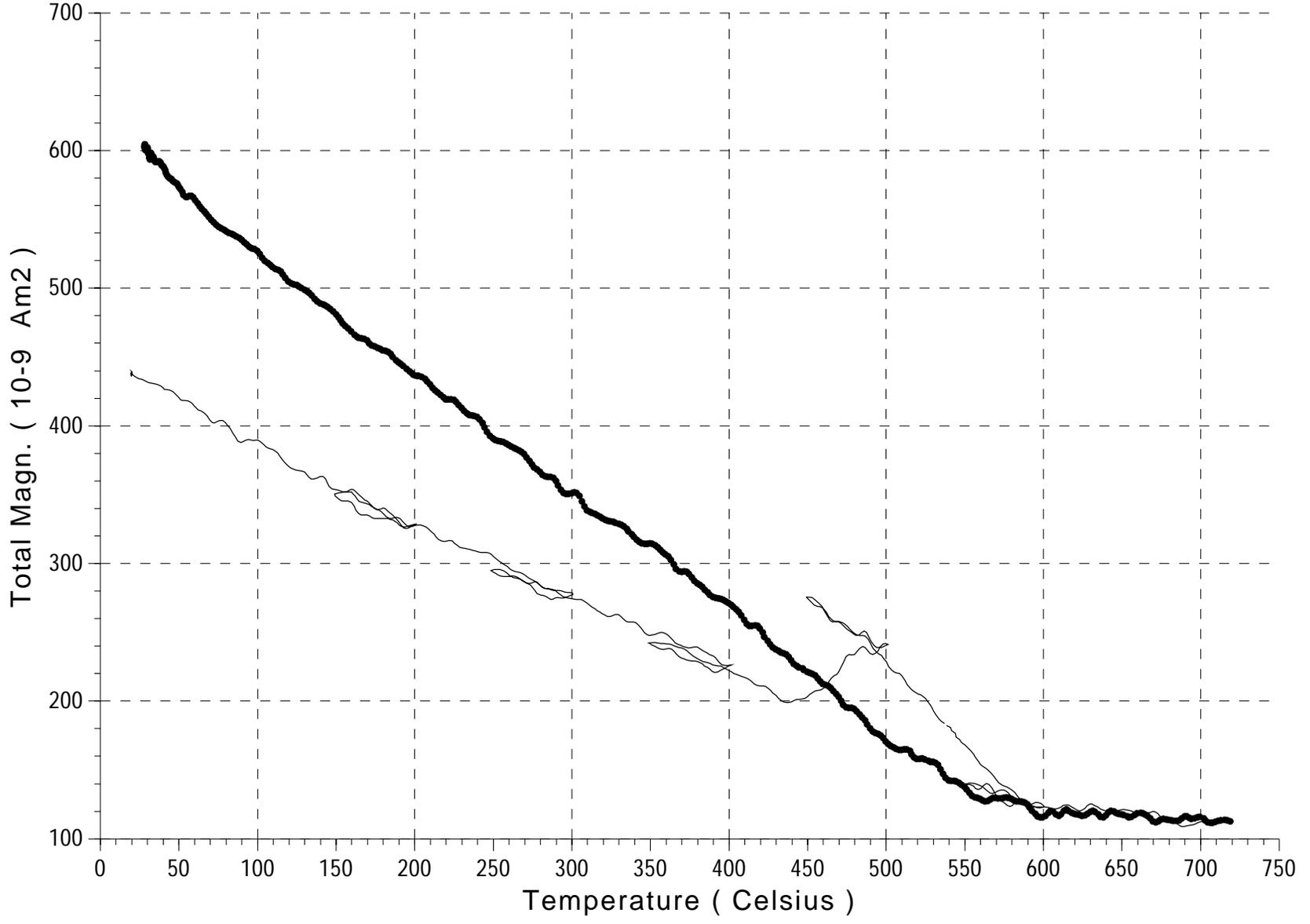
Field: 100 - 300 mT. 10/ 10 T/min. Weight: .07077. Max. temp.: 200



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OG4----TOTAL

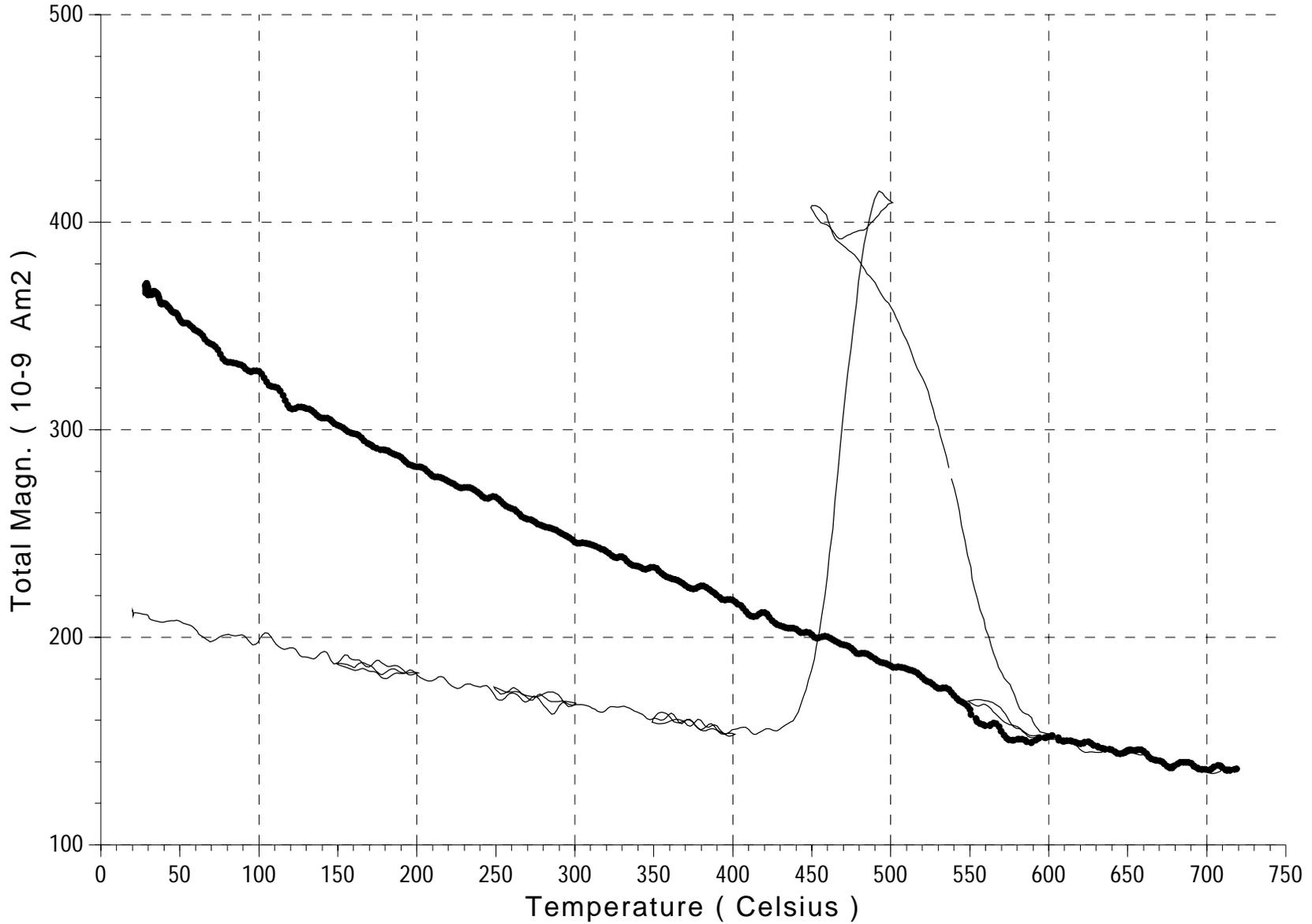
Field: 100 - 300 mT. 10/ 10 T/min. Weight: .06625. Max. temp.: 200



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OG5----TOTAL

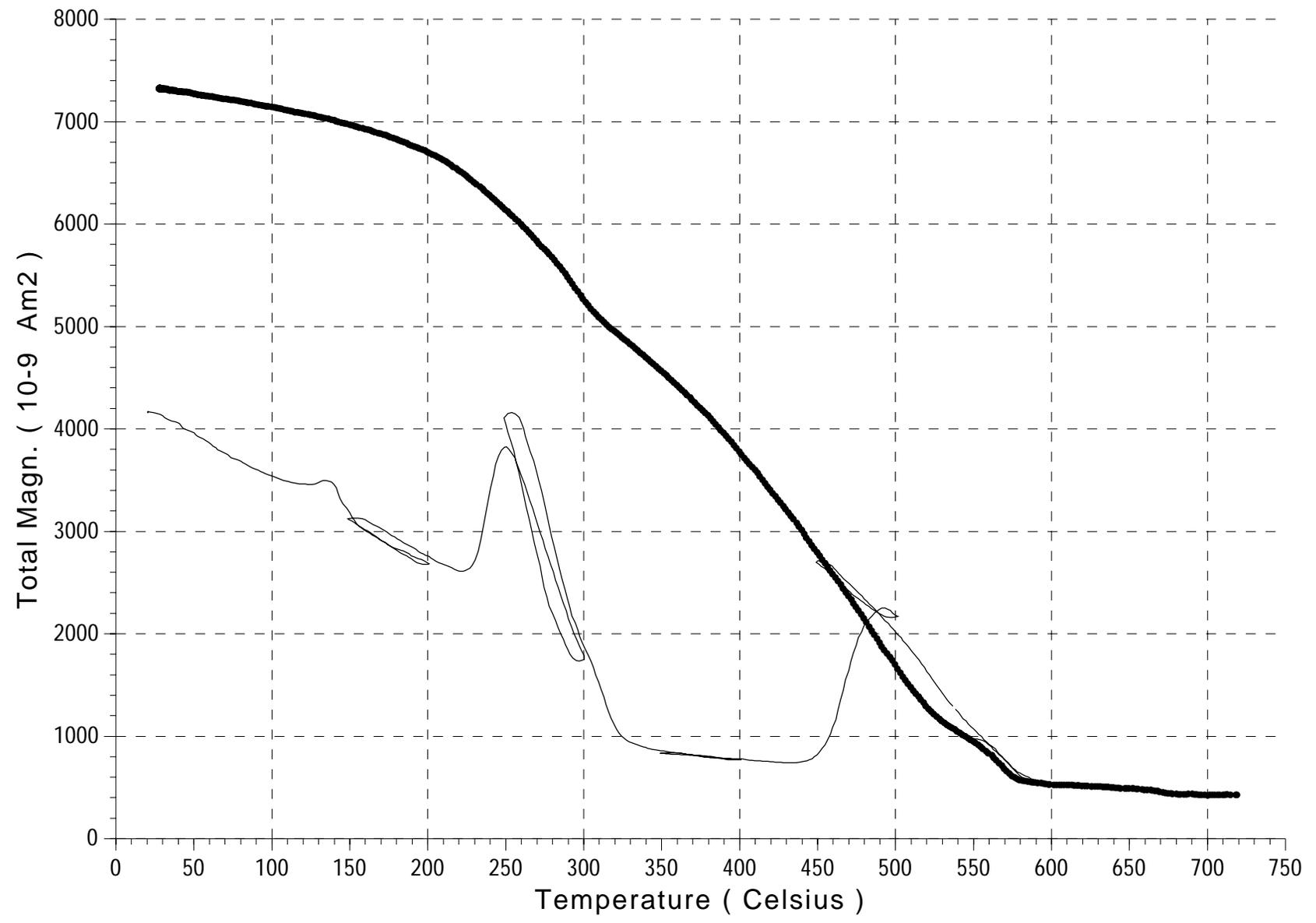
Field: 100 - 300 mT. 10/ 10 T/min. Weight: .0882. Max. temp.: 200



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OG6----TOTAL

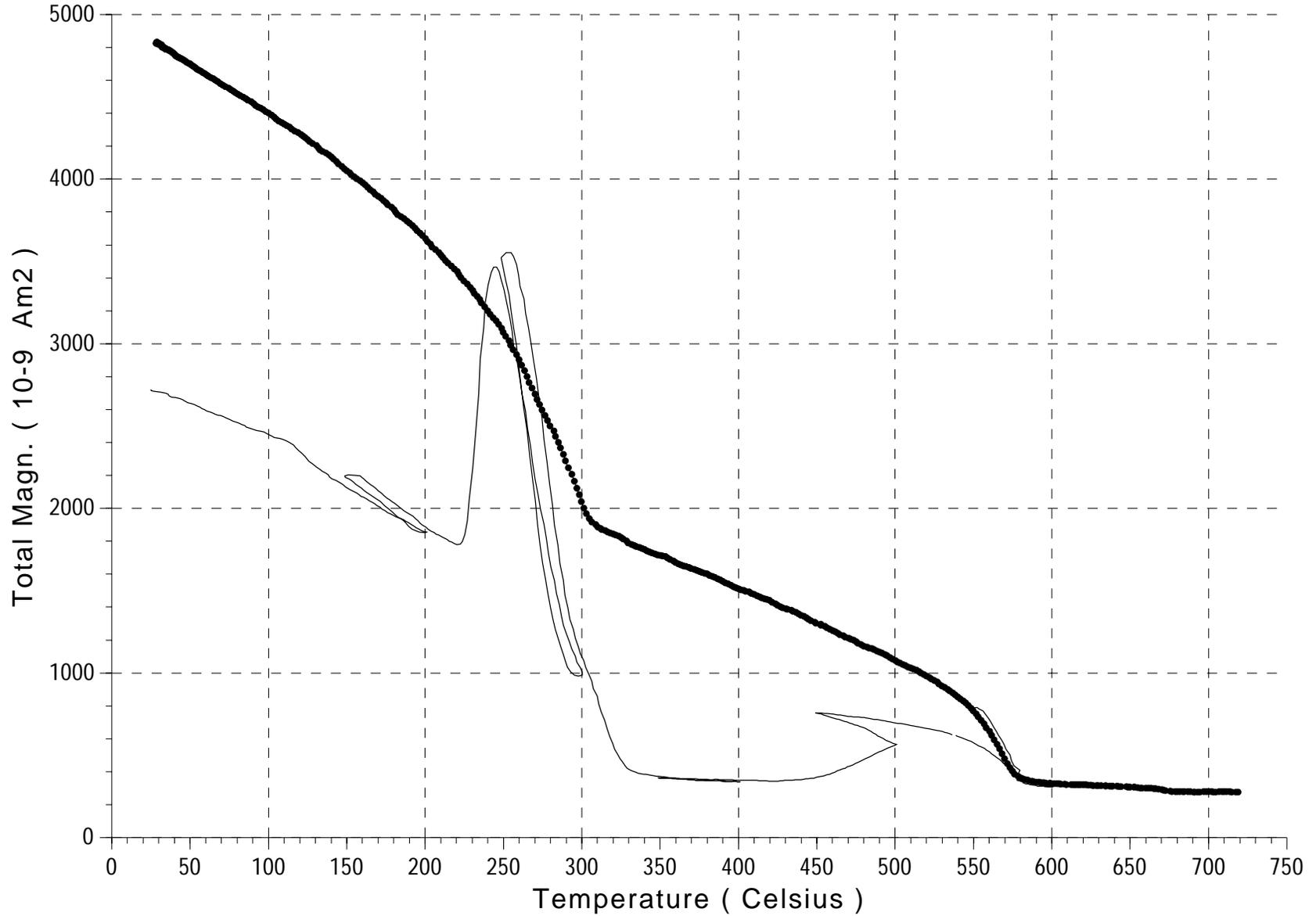
Field: 100 - 300 mT. 10/ 10 T/min. Weight: .09919. Max. temp.: 200



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OG7----TOTAL

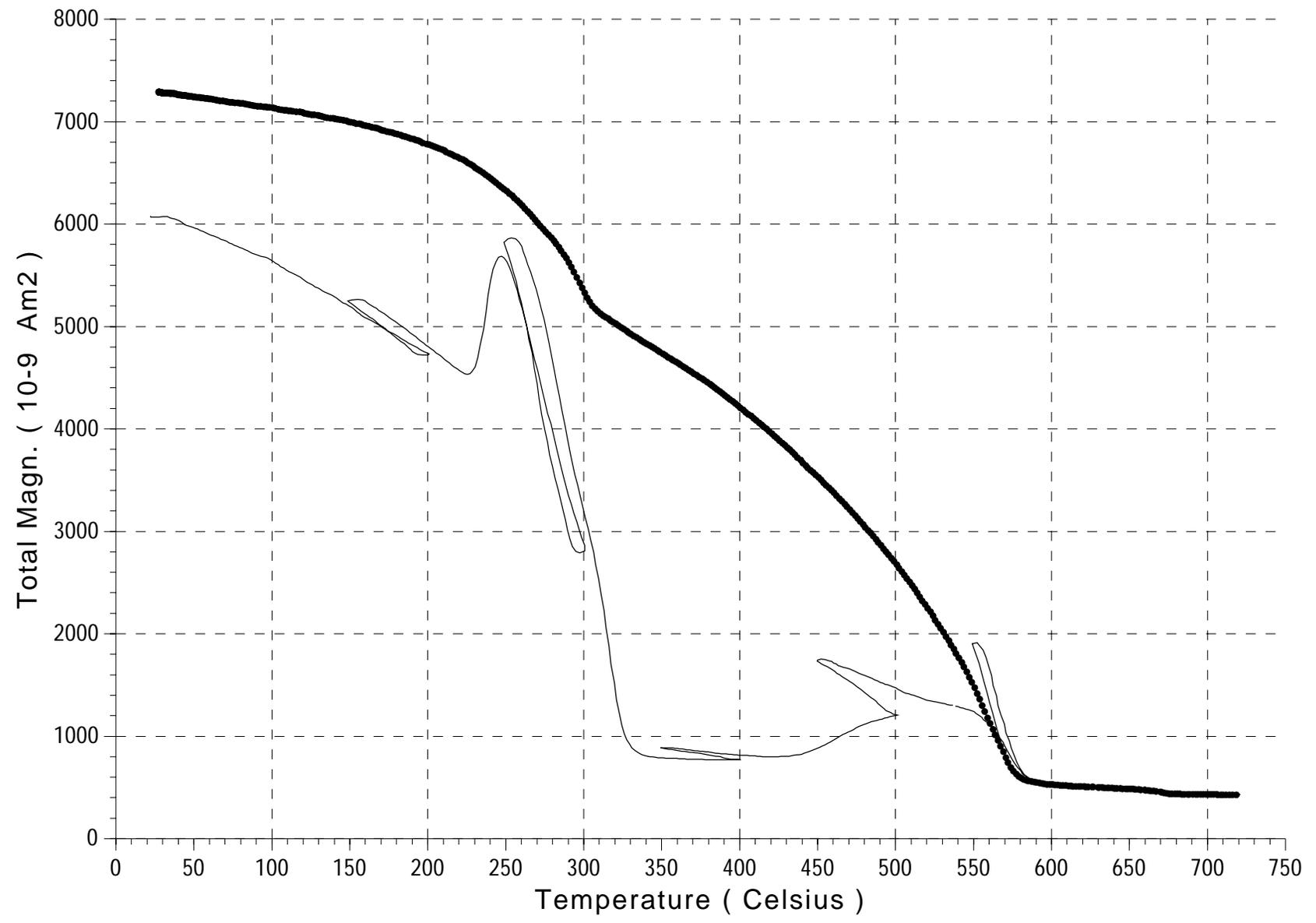
Field: 100 - 300 mT. 10/ 10 T/min. Weight: .0755. Max. temp.: 200



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OG8----TOTAL

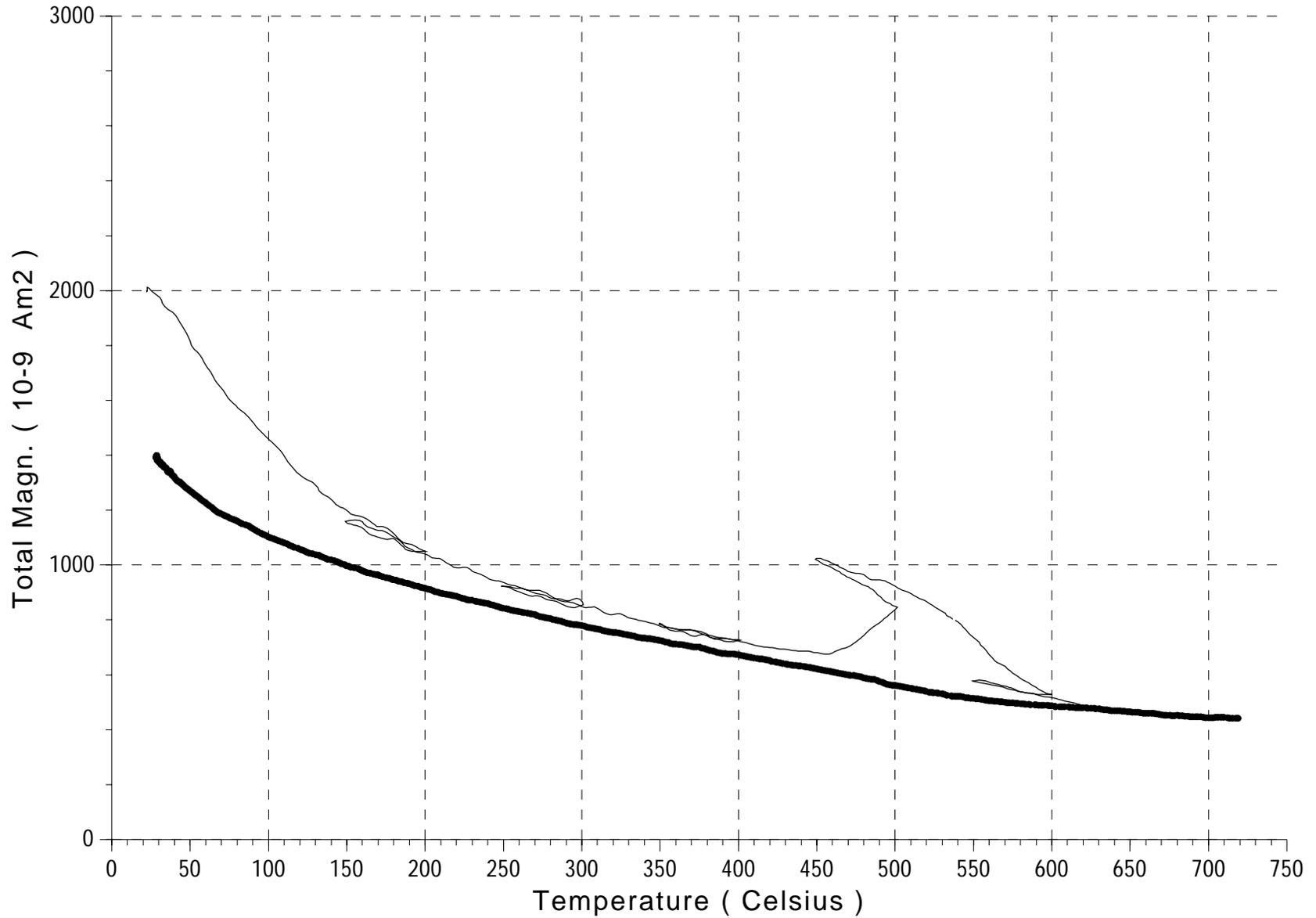
Field: 100 - 300 mT. 10/ 10 T/min. Weight: .06738. Max. temp.: 200



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OG9----TOTAL

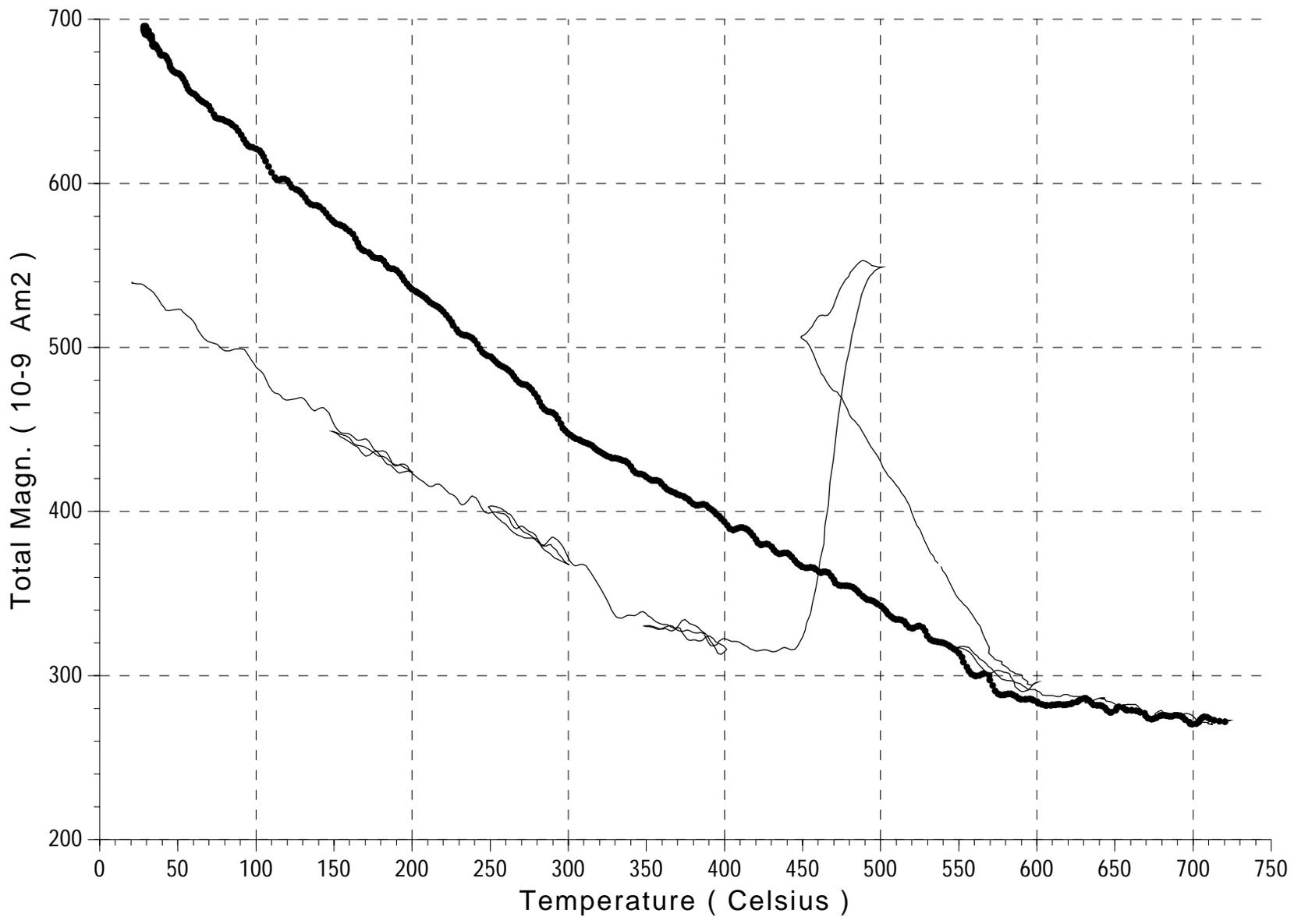
Field: 100 - 300 mT. 10/ 10 T/min. Weight: .06554. Max. temp.: 200



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OG10----TOTAL

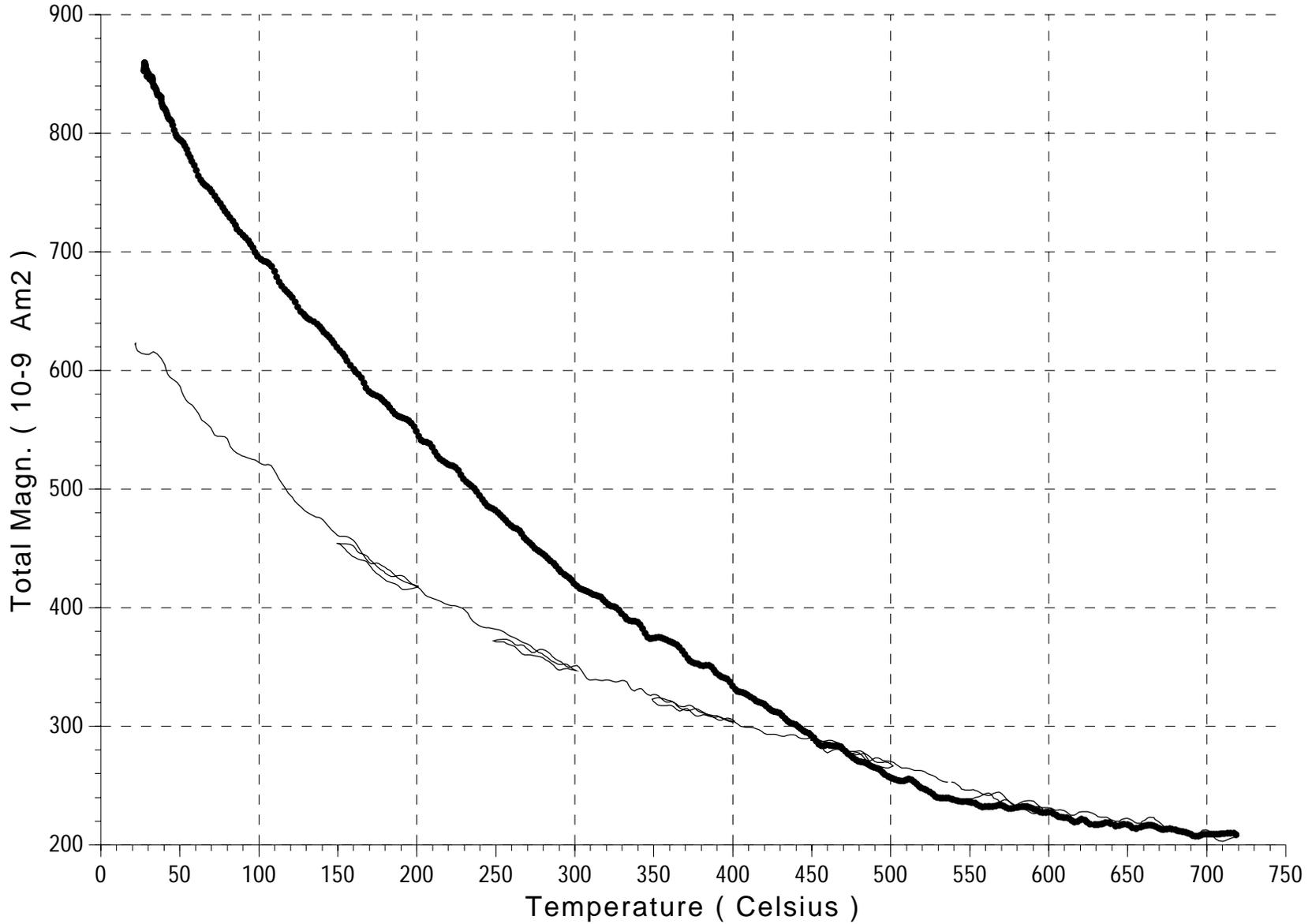
Field: 100 - 300 mT. 10/ 10 T/min. Weight: .0882. Max. temp.: 200



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OG11----TOTAL

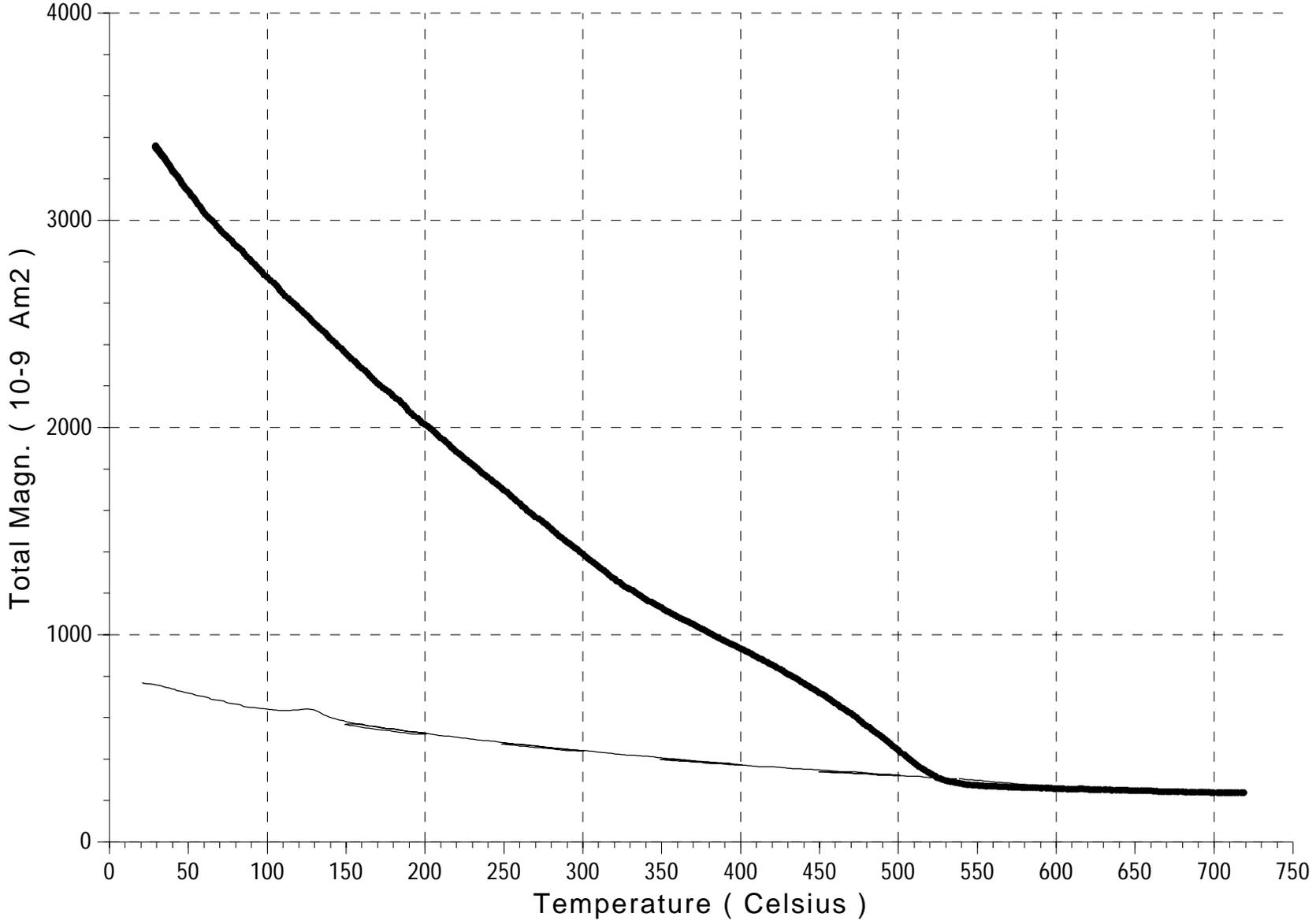
Field: 100 - 300 mT. 10/ 10 T/min. Weight: .0642. Max. temp.: 200



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OG12dyke----TOTAL

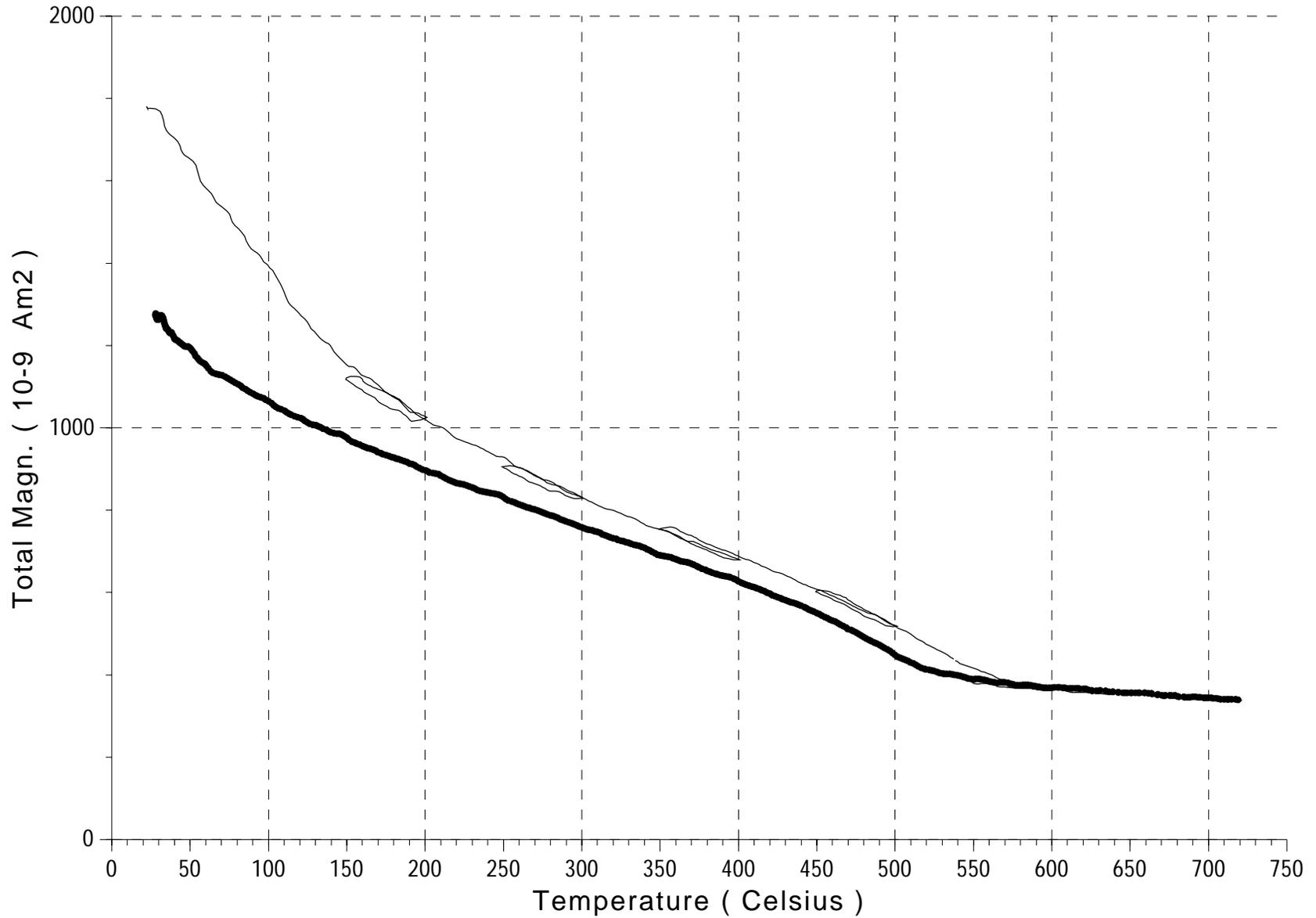
Field: 100 - 300 mT. 10/ 10 T/min. Weight: .04737. Max. temp.: 200



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OG13----TOTAL

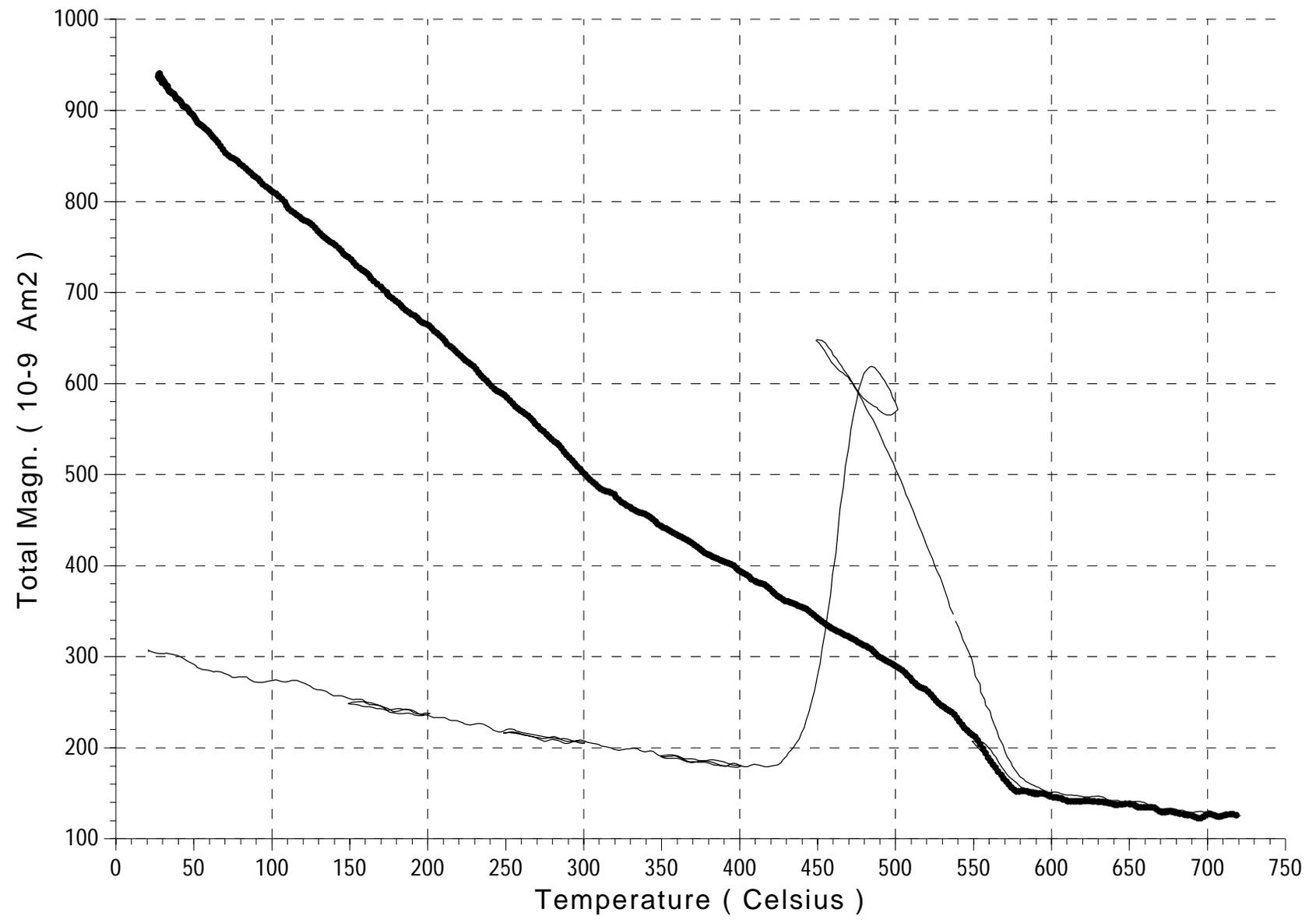
Field: 100 - 300 mT. 10/ 10 T/min. Weight: .06606. Max. temp.: 200



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OG14----TOTAL

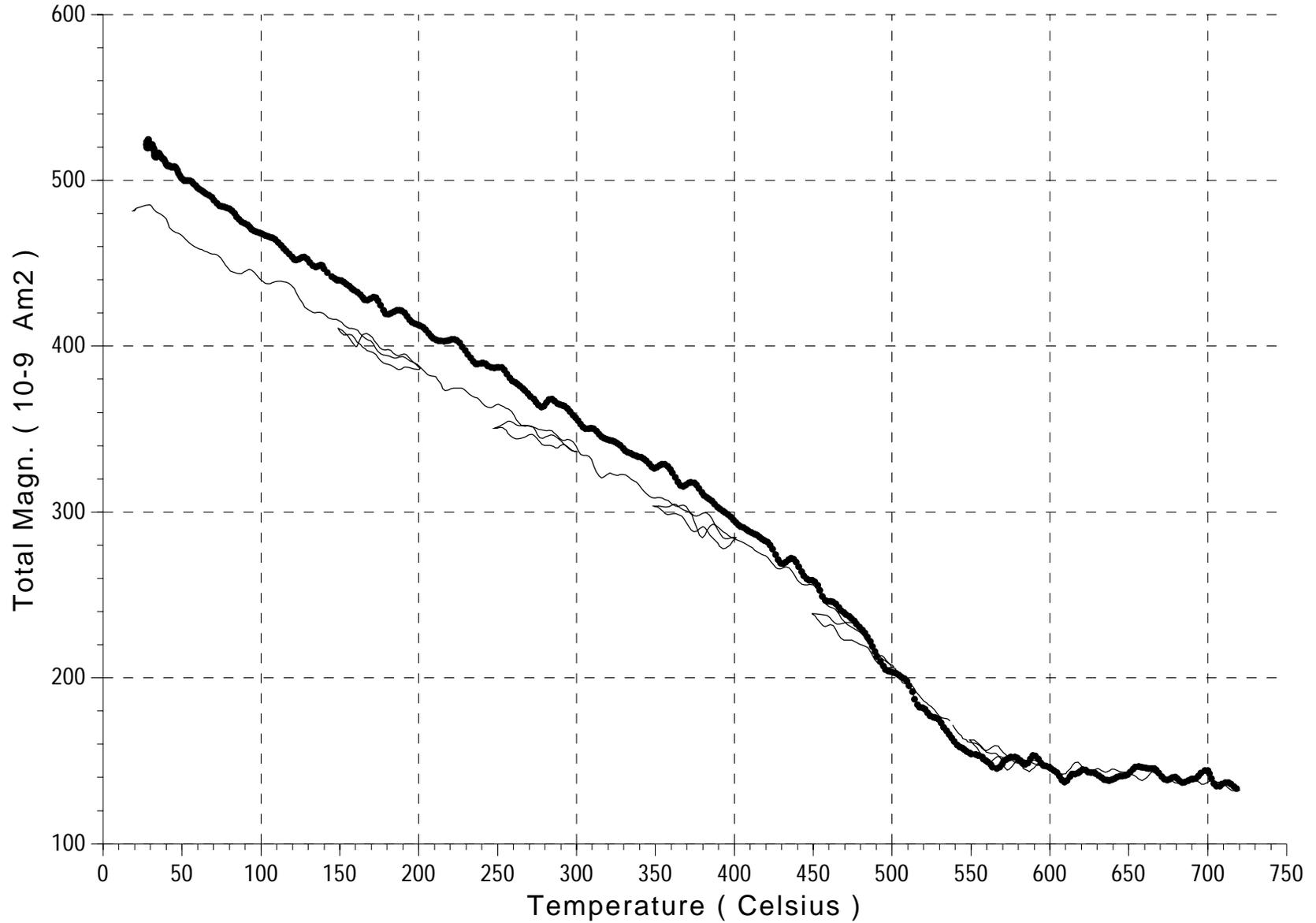
Field: 100 - 300 mT. 10/ 10 T/min. Weight: .06951. Max. temp.: 200



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OG15----TOTAL

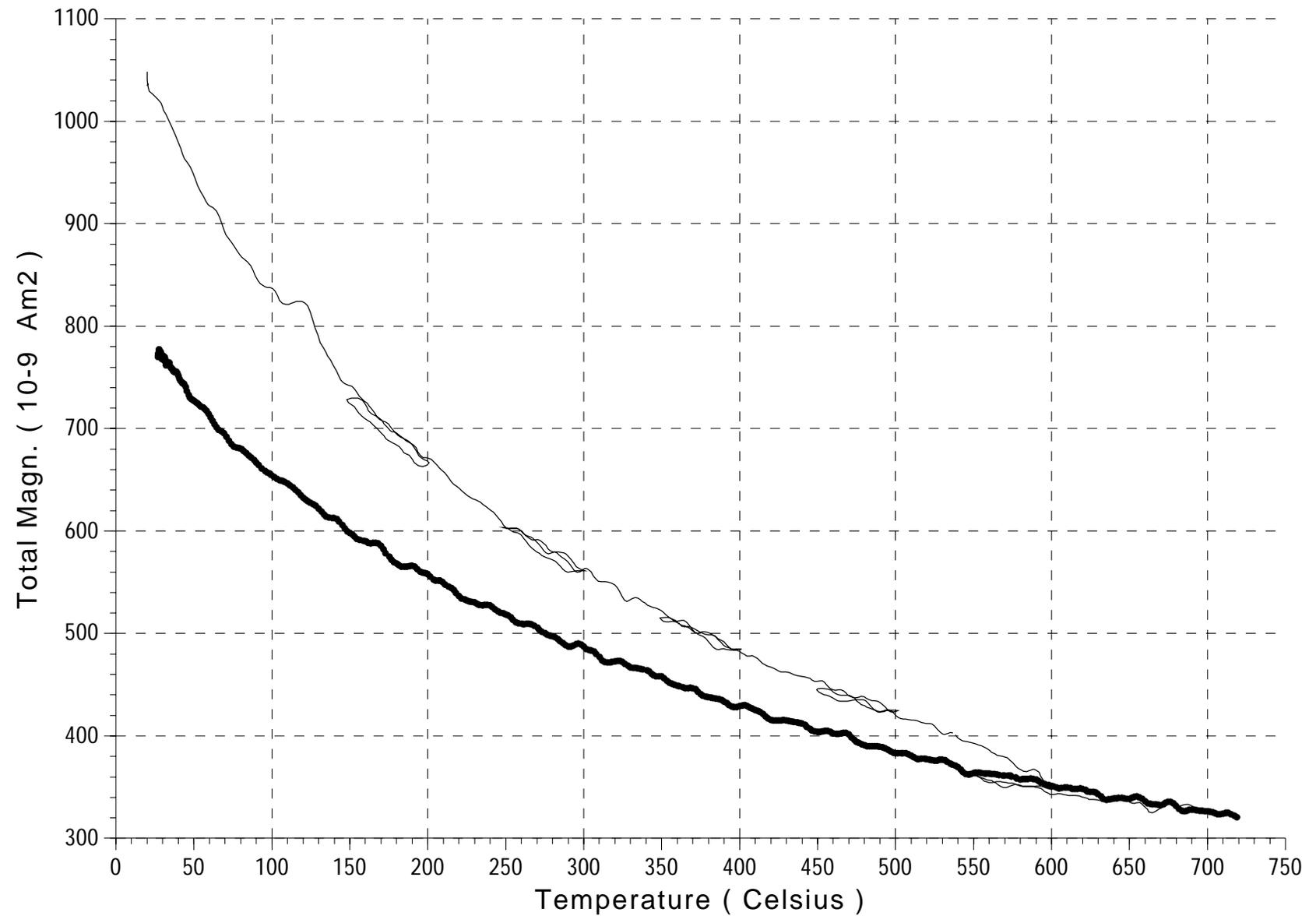
Field: 100 - 300 mT. 10/ 10 T/min. Weight: .03646. Max. temp.: 200



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OG16----TOTAL

Field: 100 - 300 mT. 10/ 10 T/min. Weight: .05961. Max. temp.: 200



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Og17----TOTAL

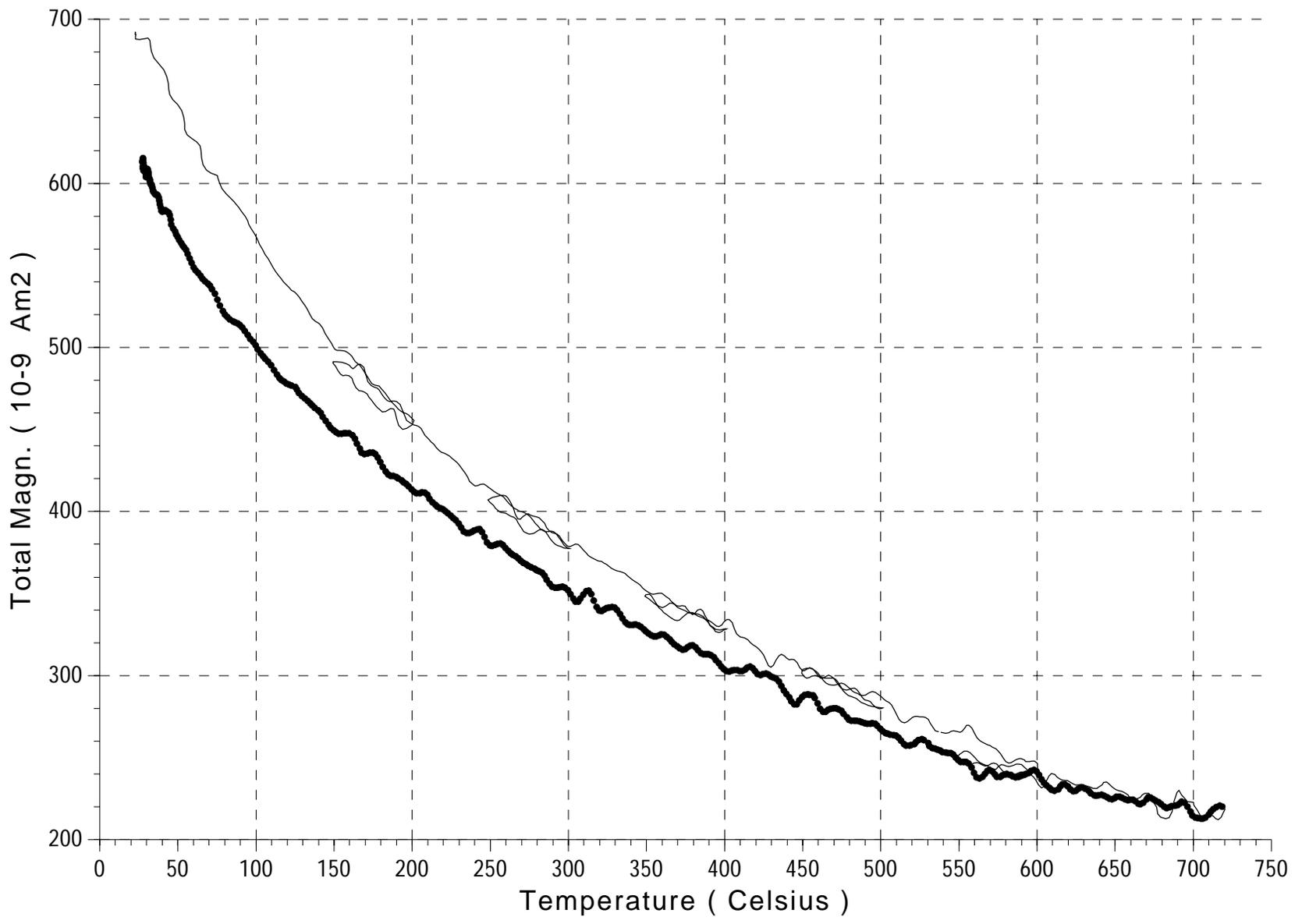
Field: 100 - 300 mT. 10/ 10 T/min. Weight: .02144. Max. temp.: 200



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OG18----TOTAL

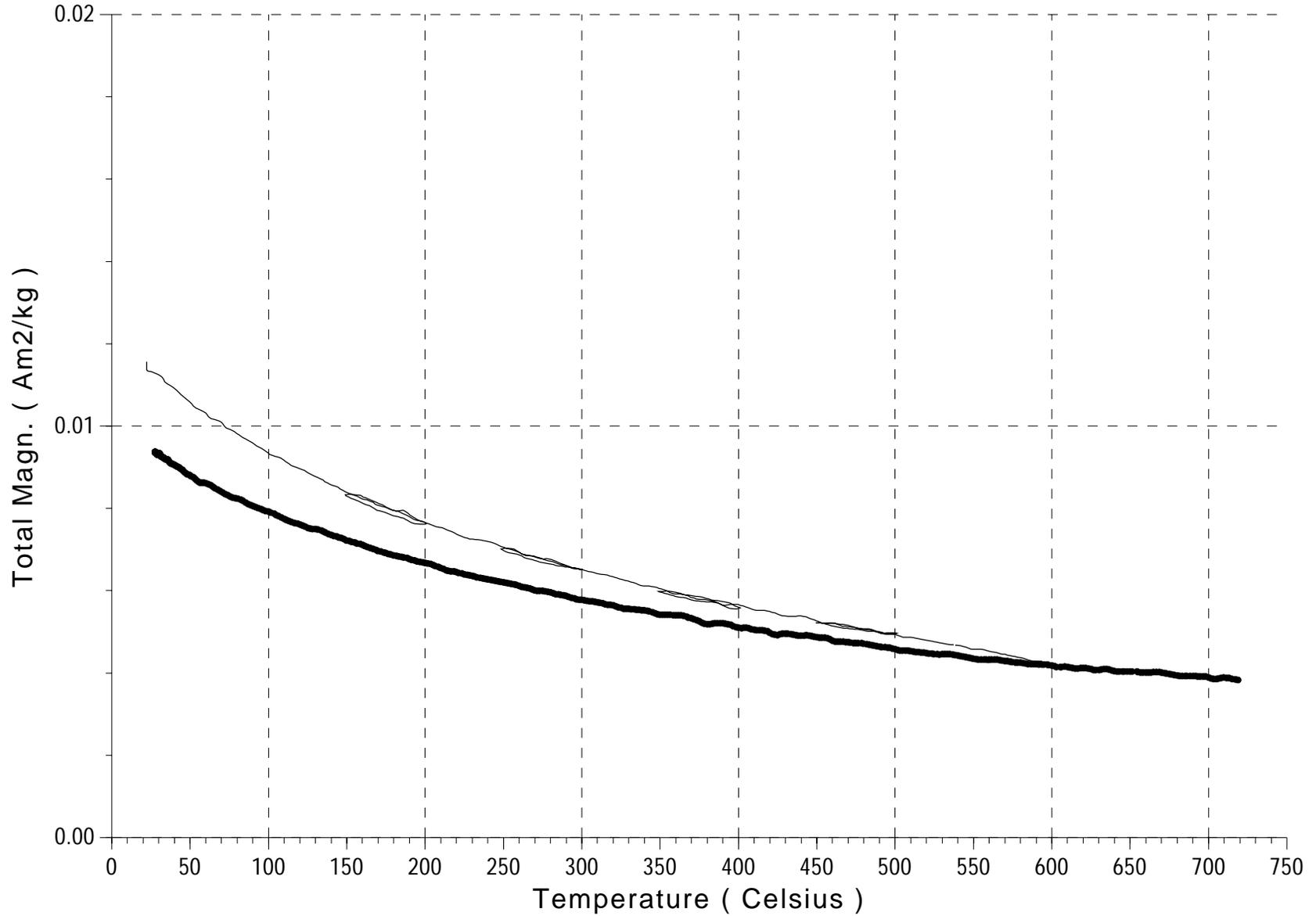
Field: 100 - 300 mT. 10/ 10 T/min. Weight: .07244. Max. temp.: 200



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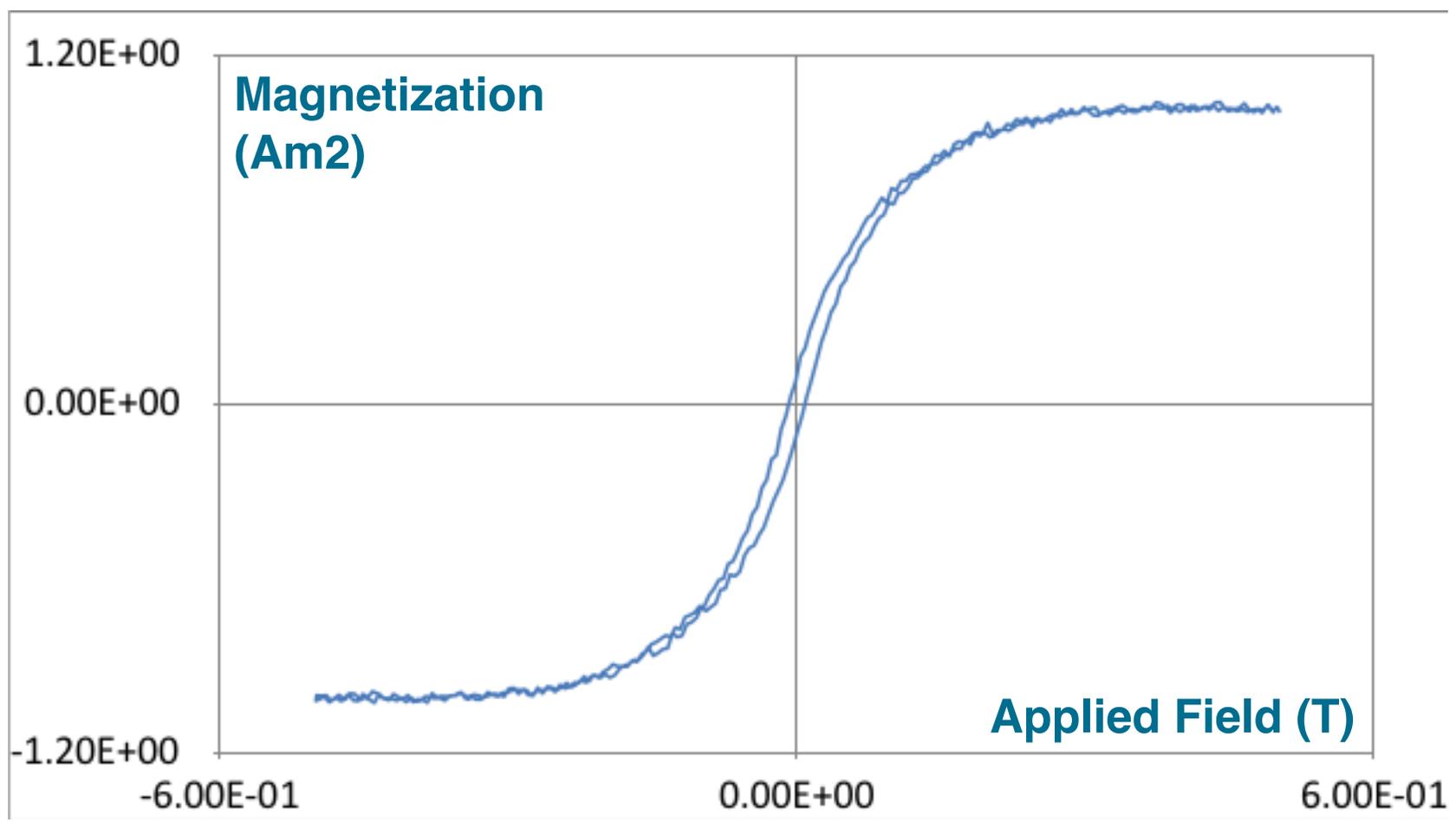
OG19---please work-TOTAL

Field: 100 - 300 mT. 10/ 10 T/min. Weight: .06479. Max. temp.: 200

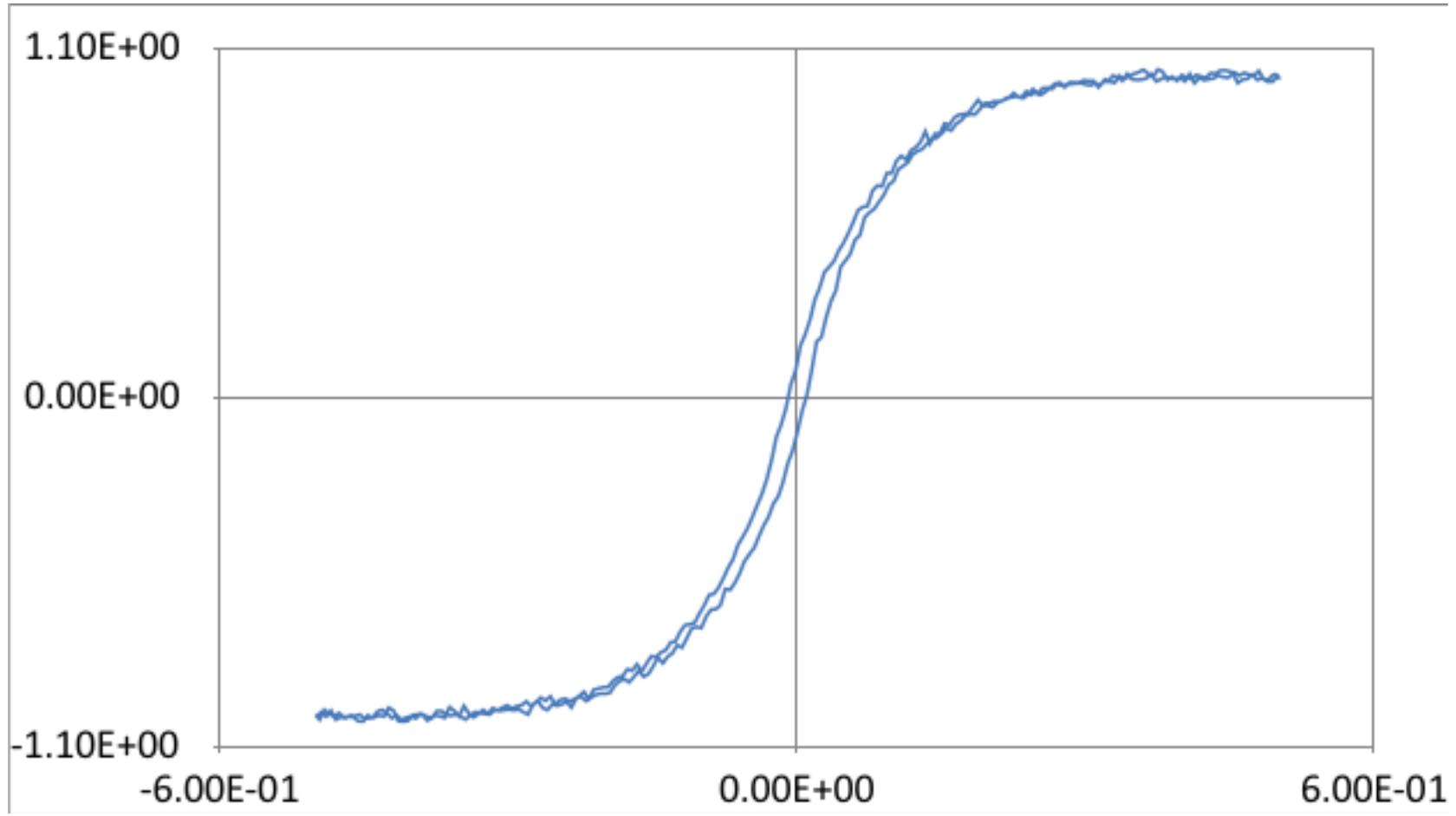


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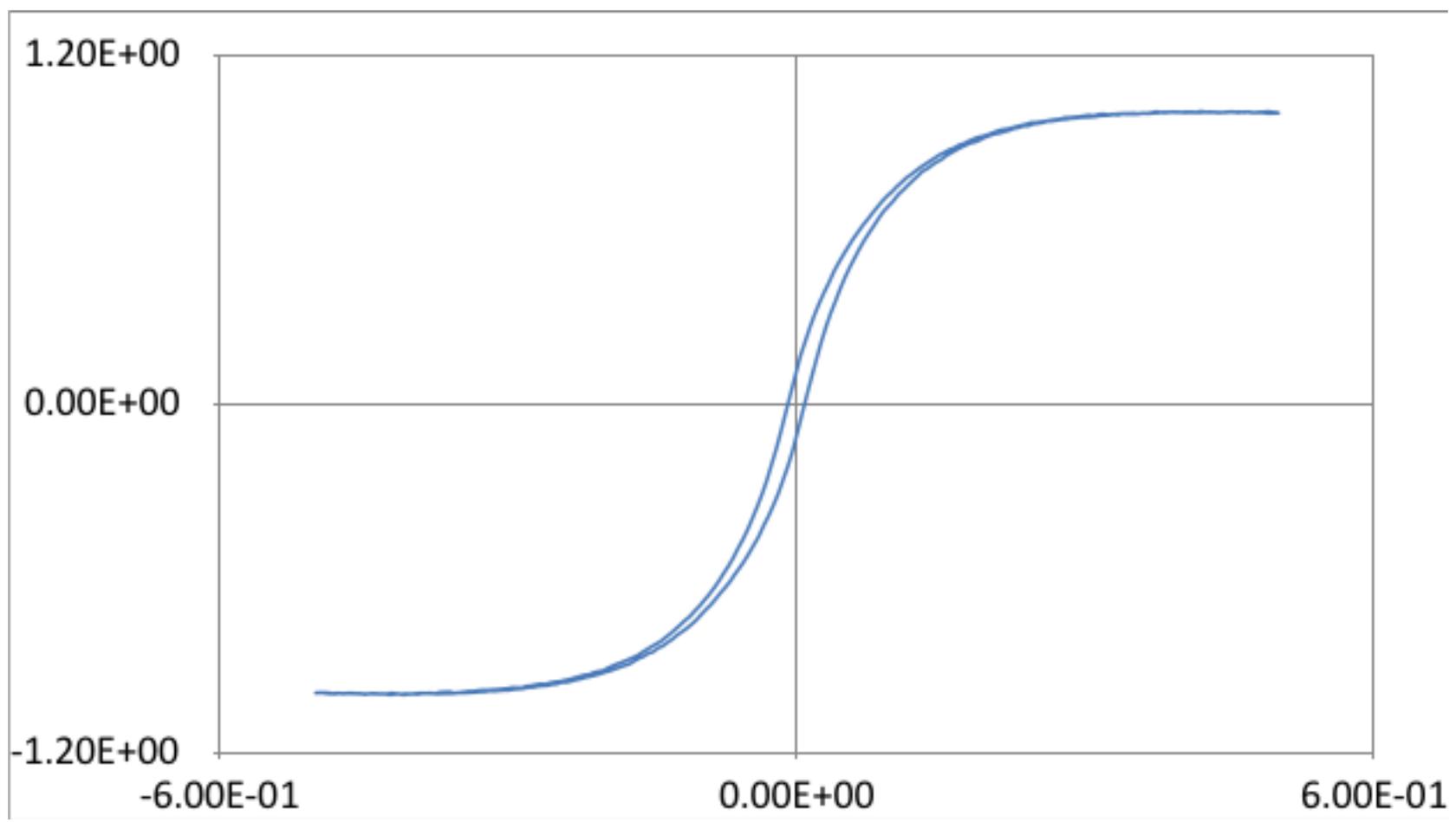


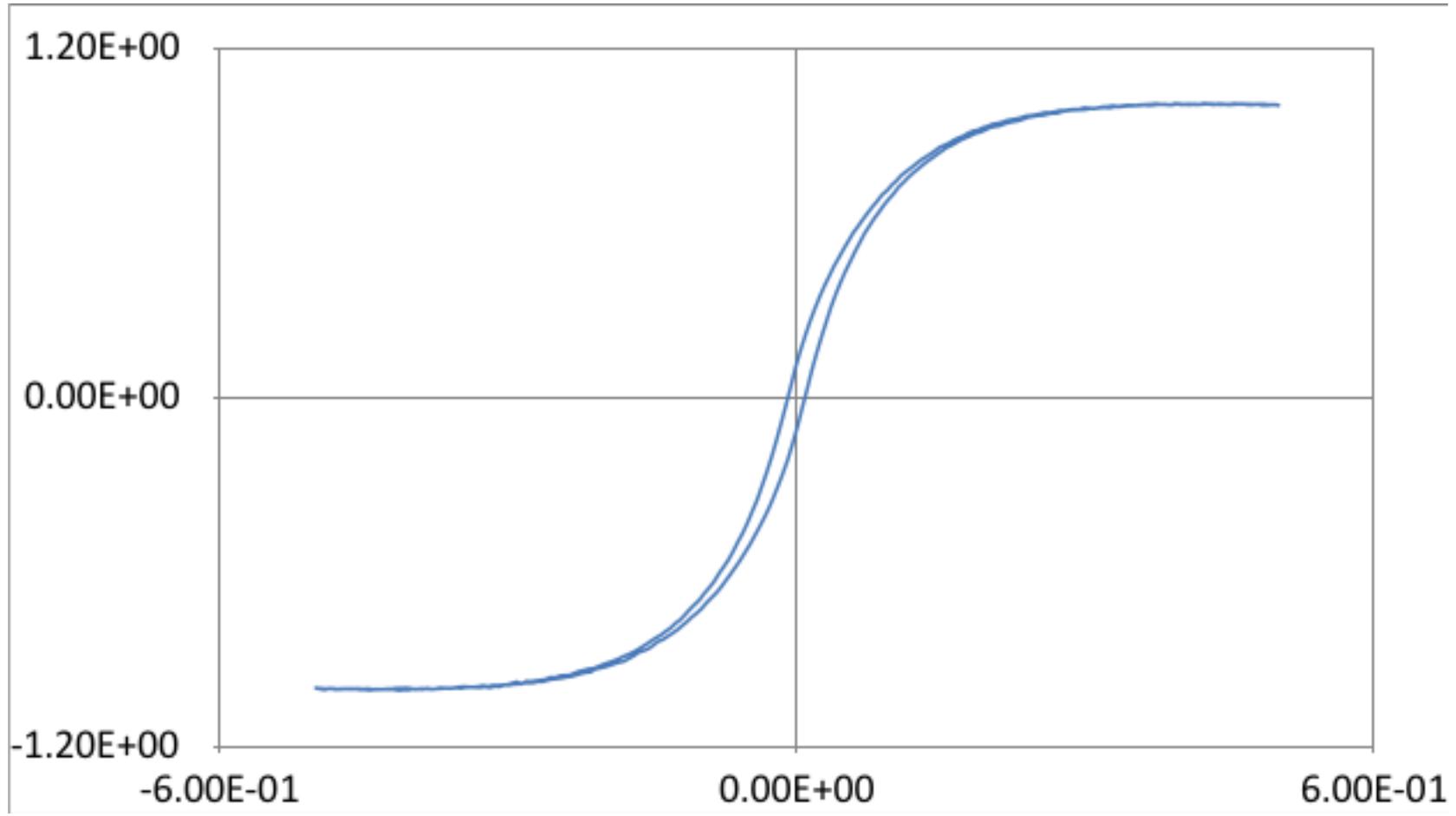
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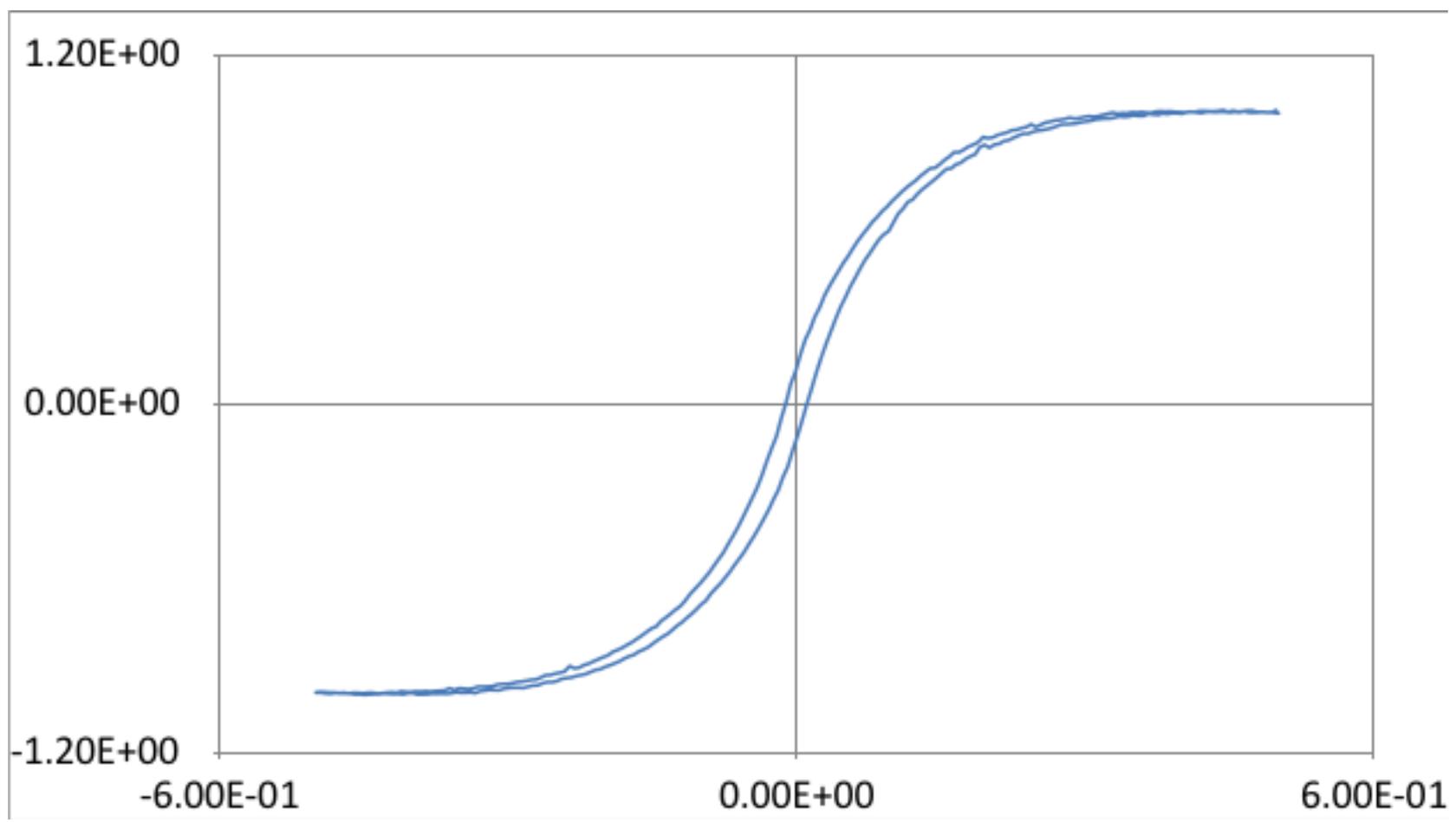
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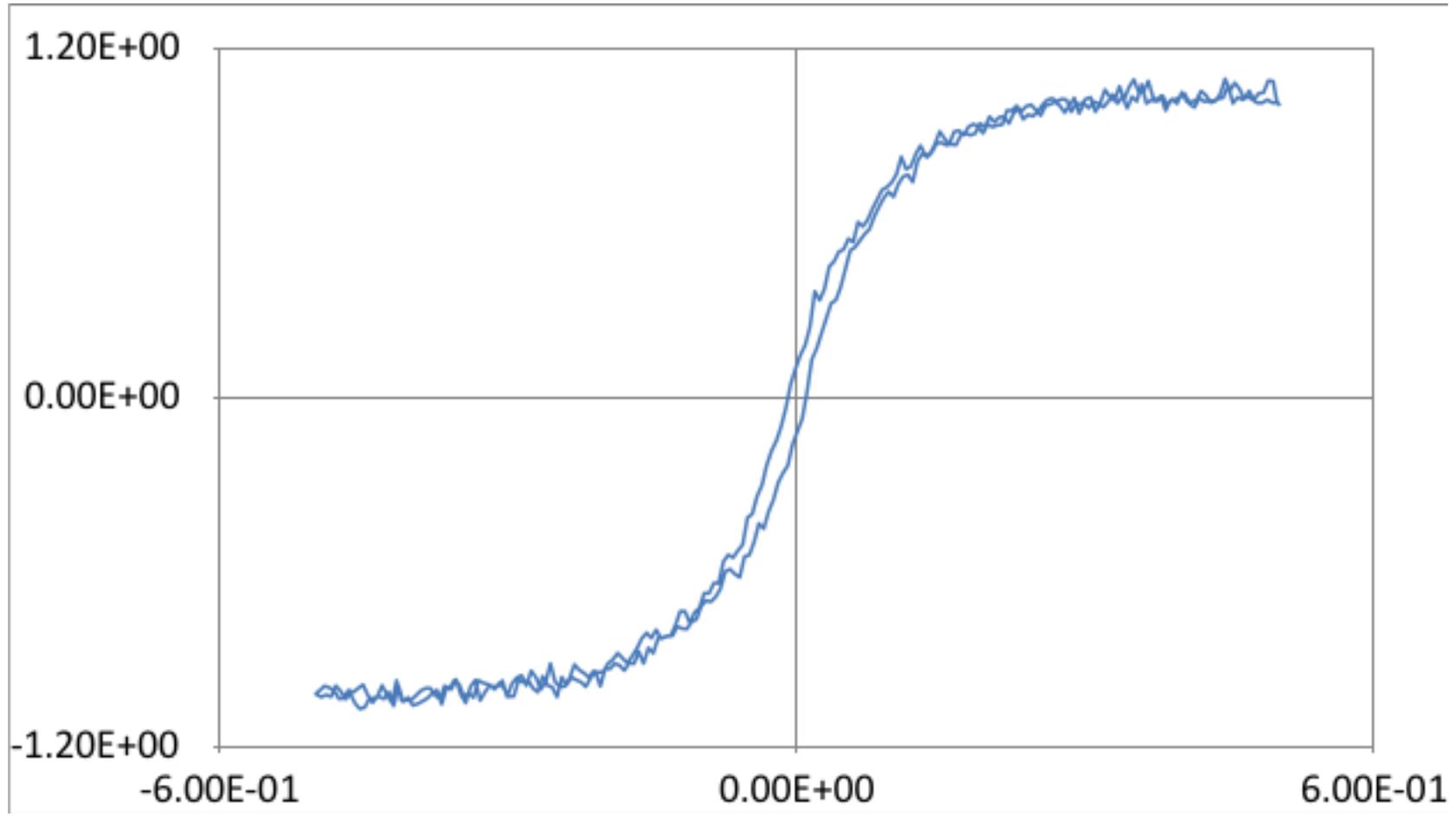




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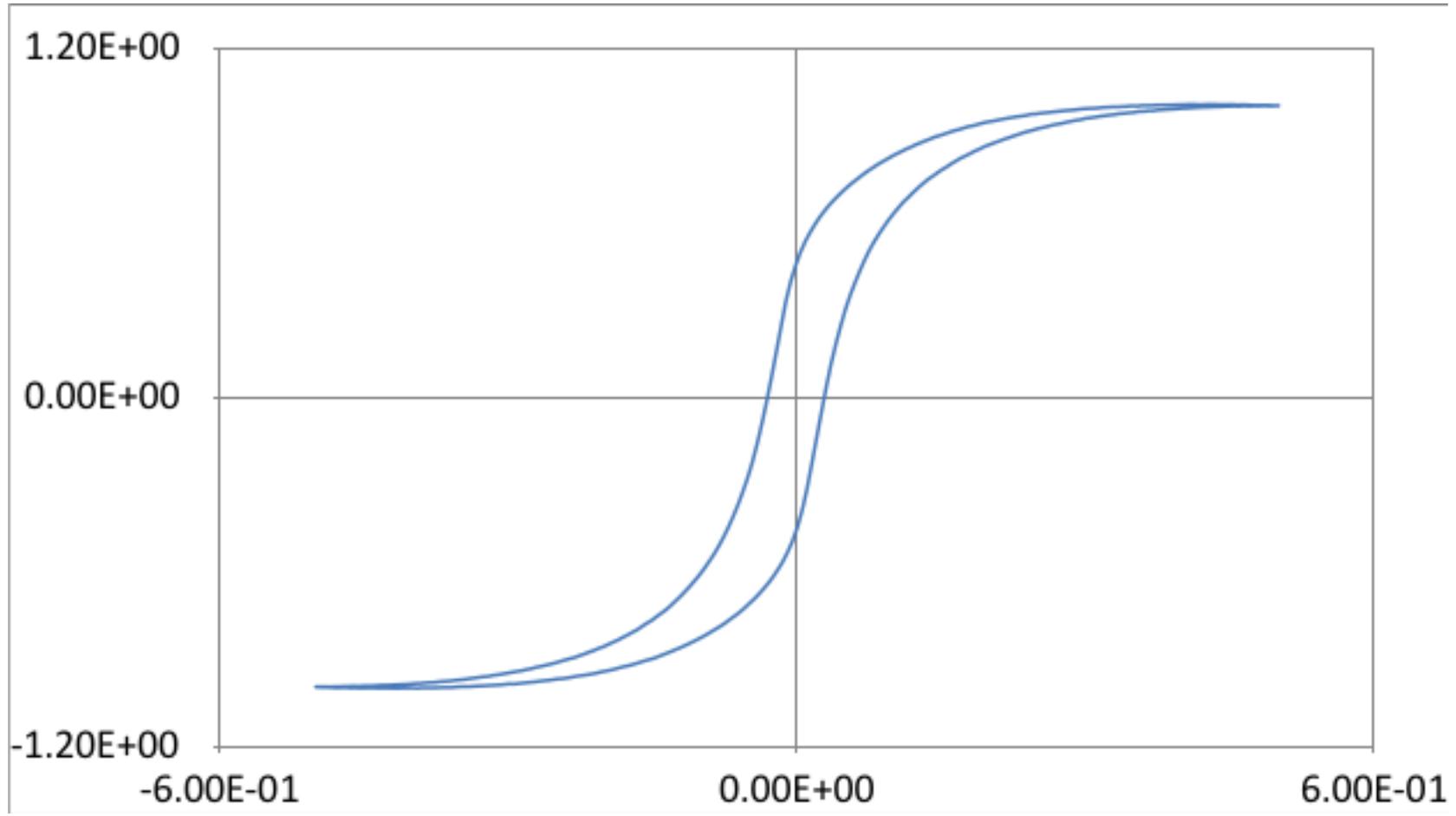
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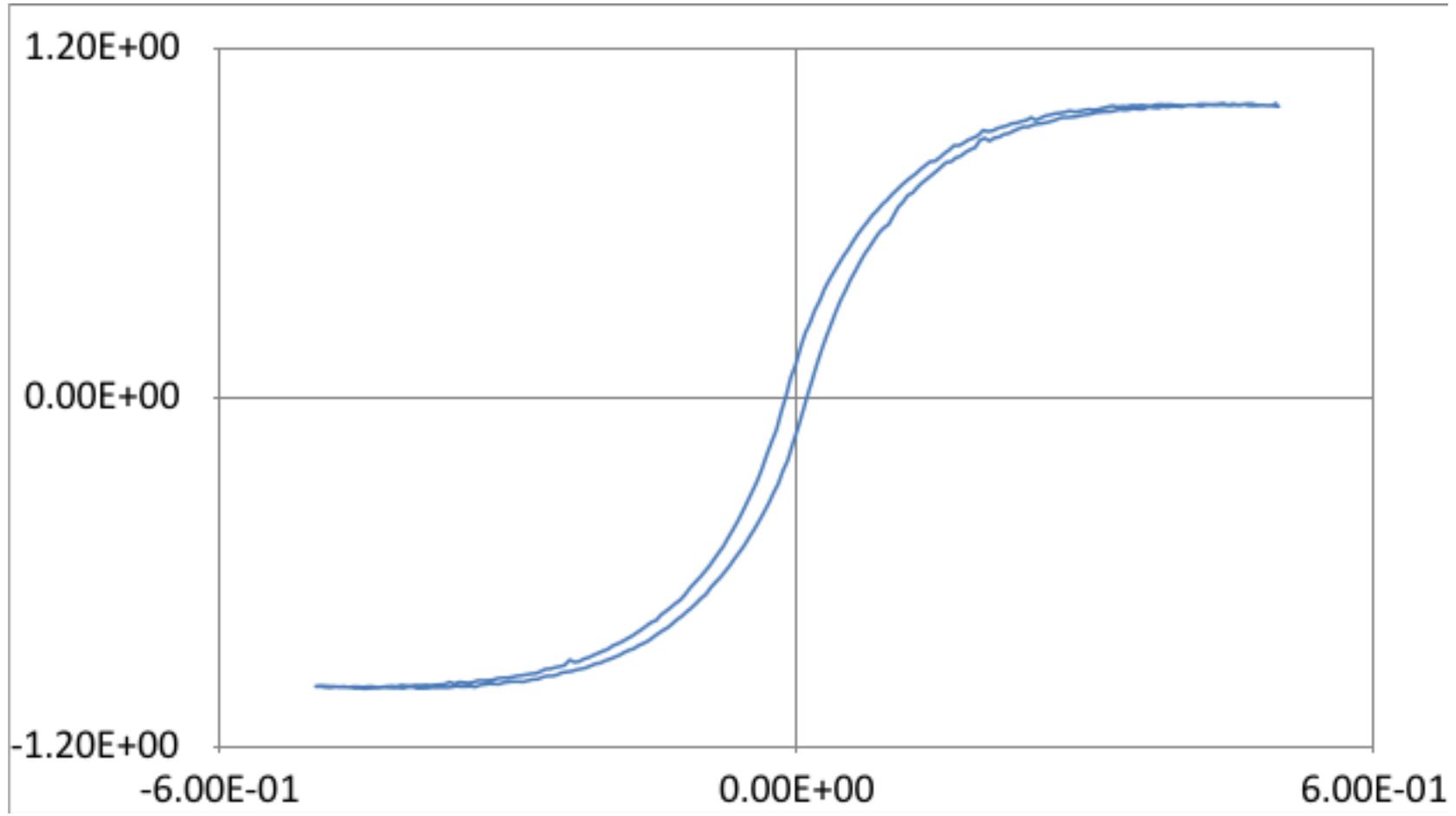




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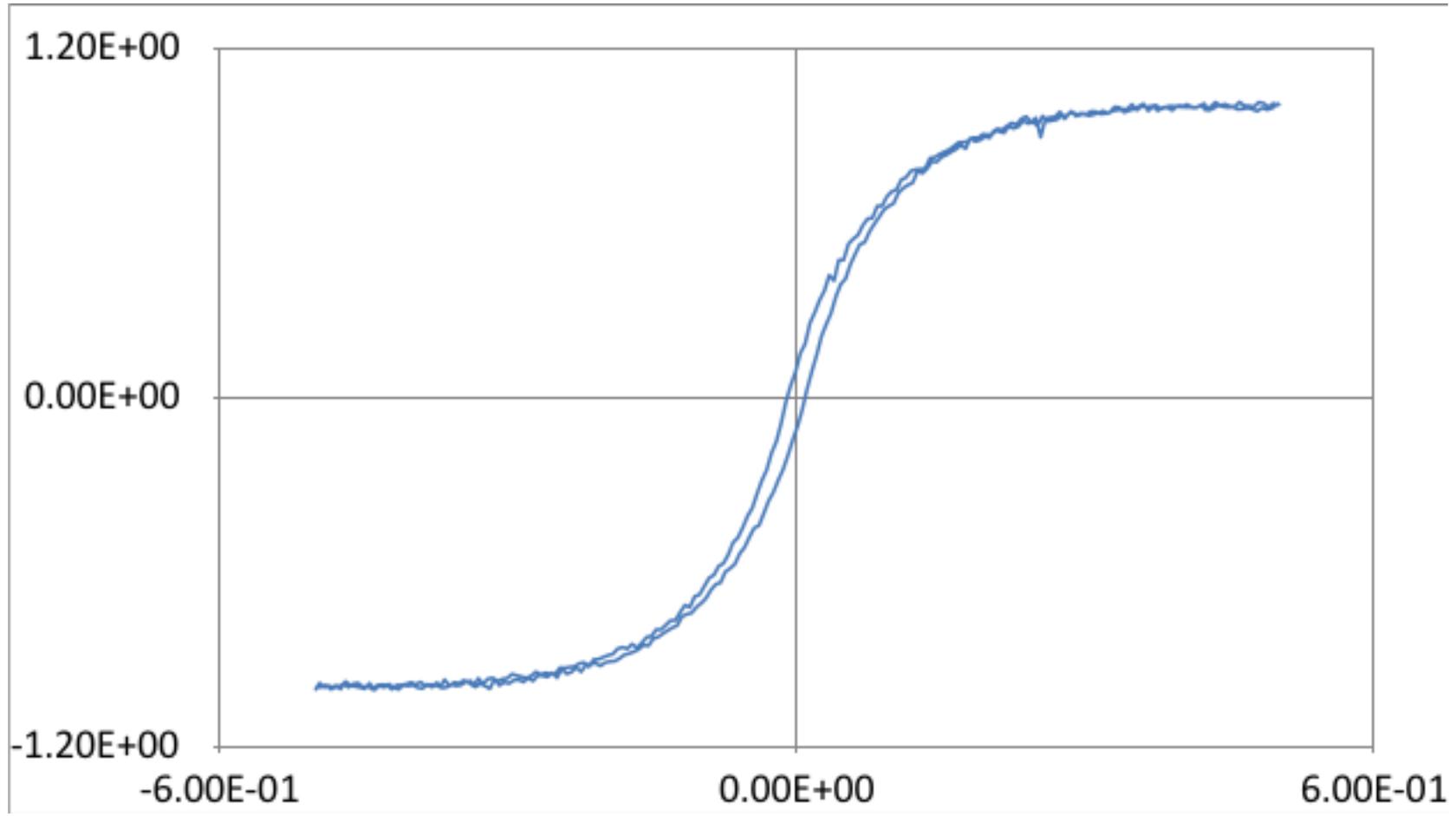
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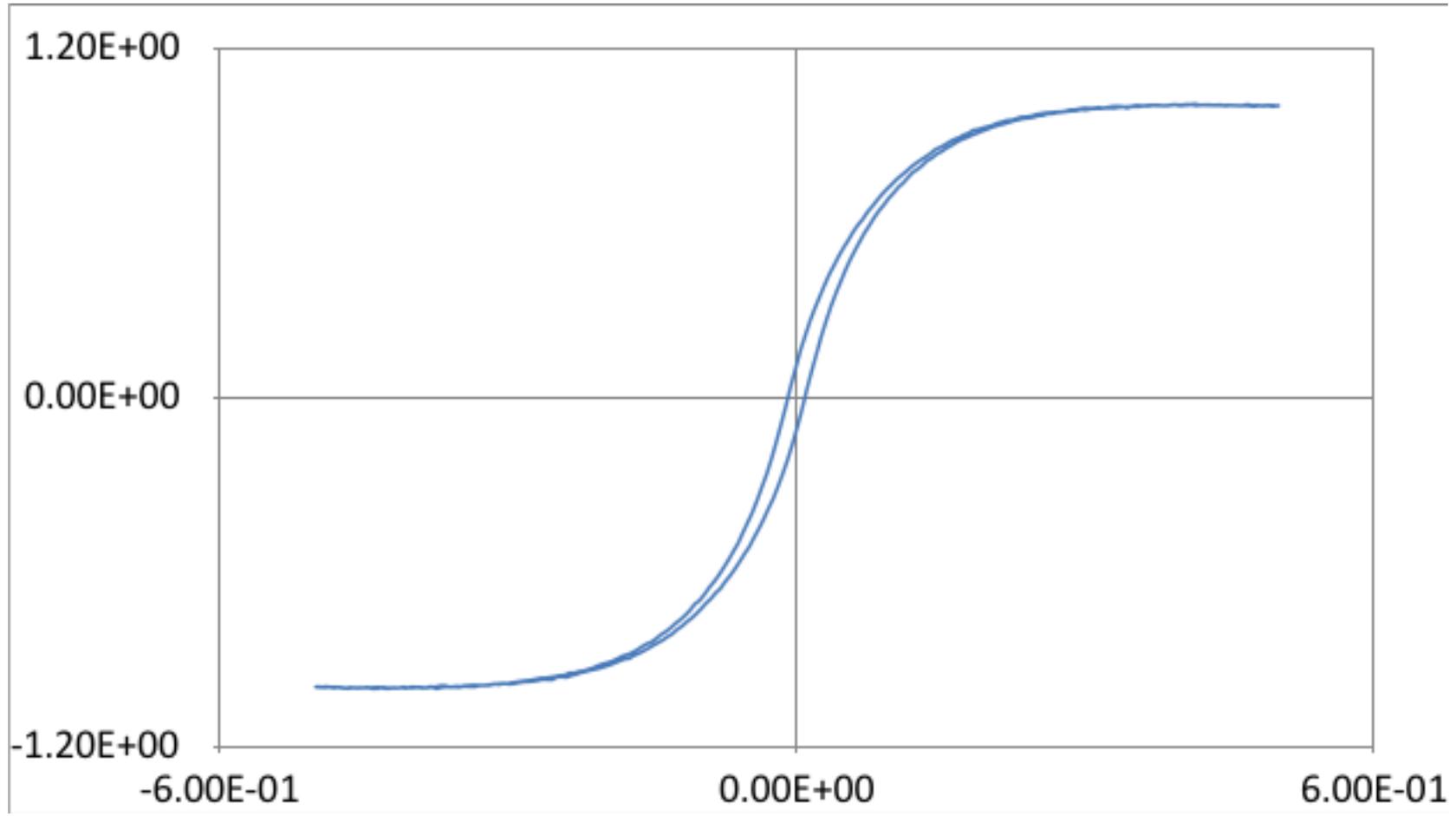




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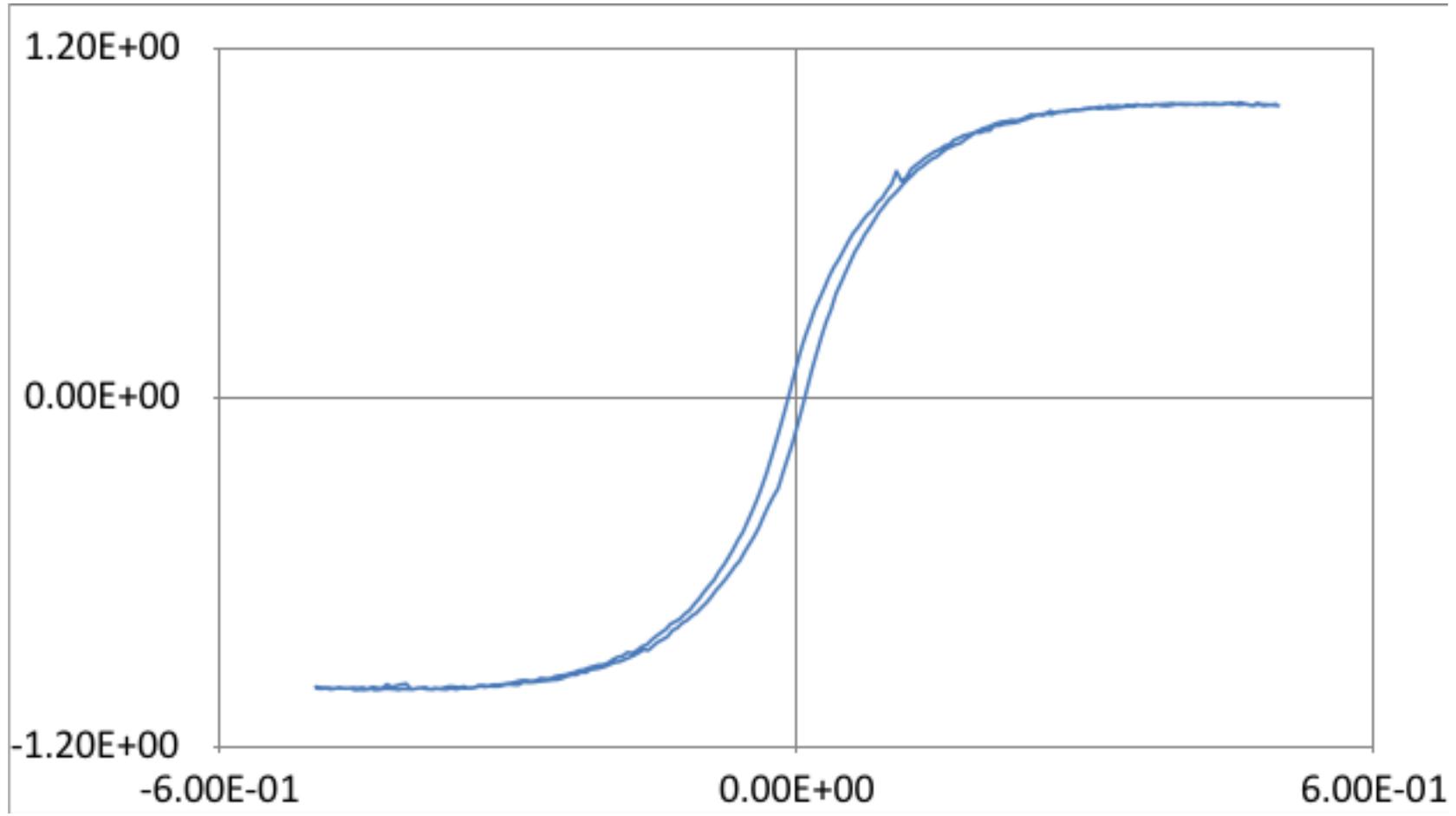
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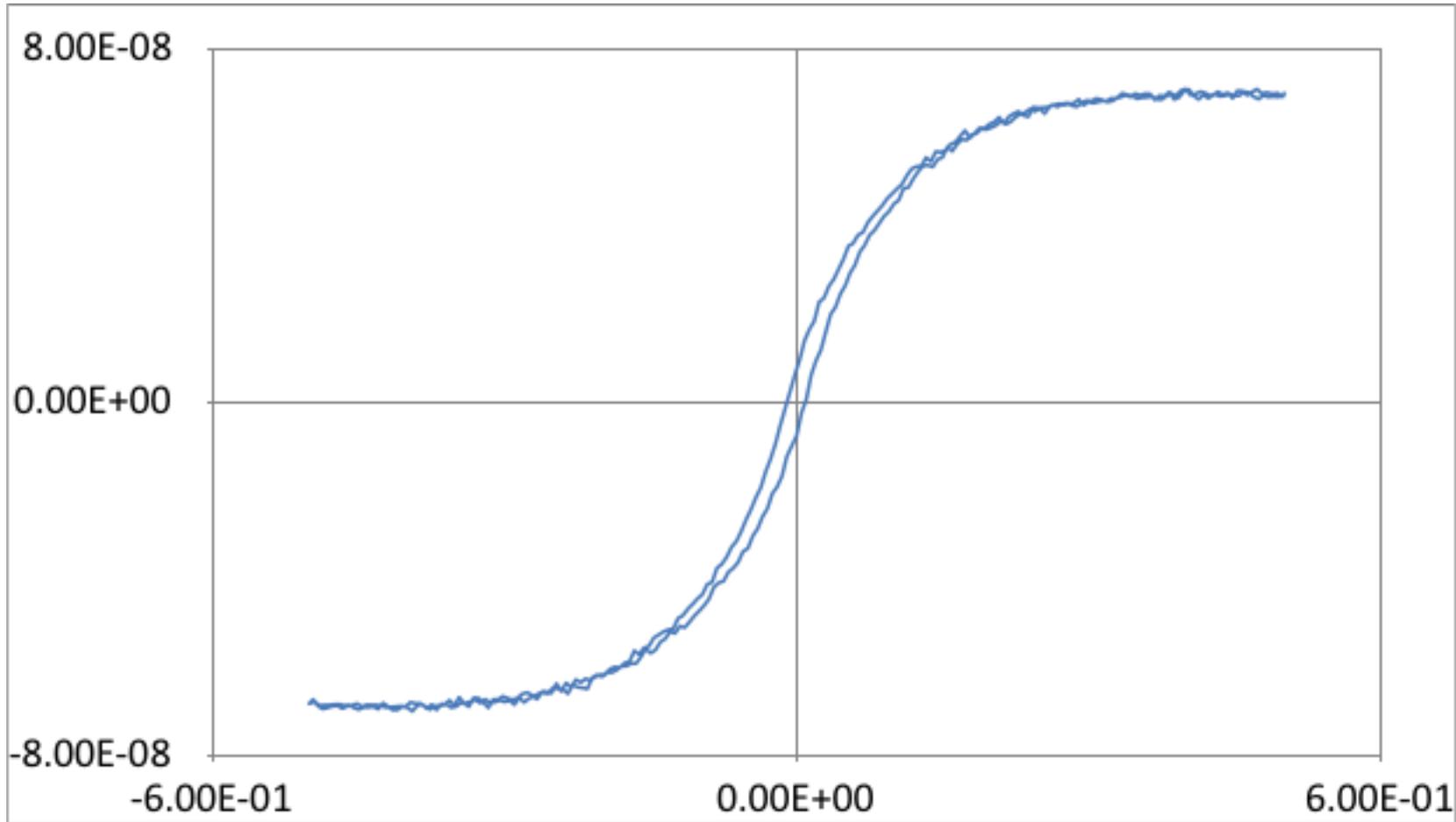




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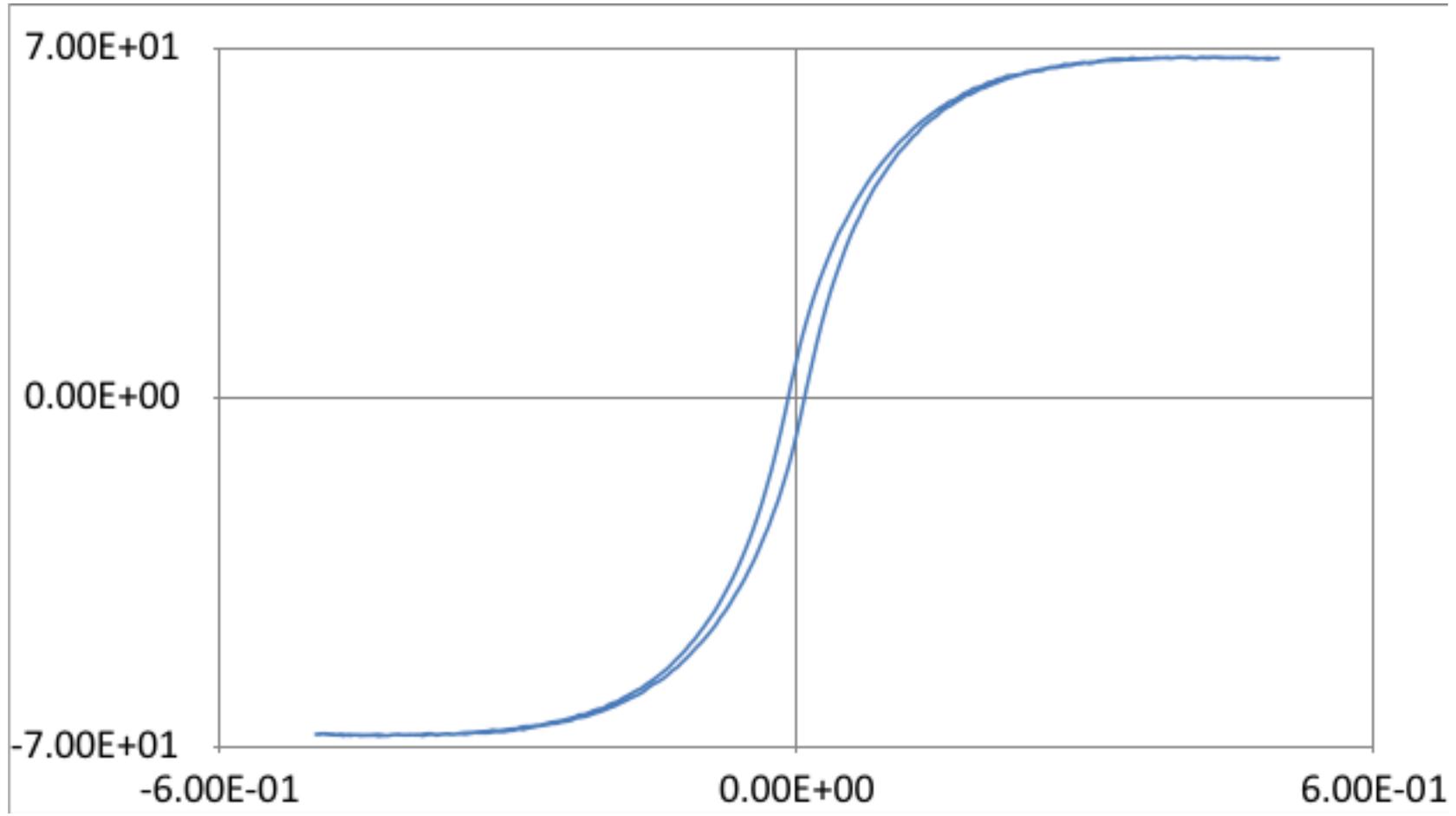
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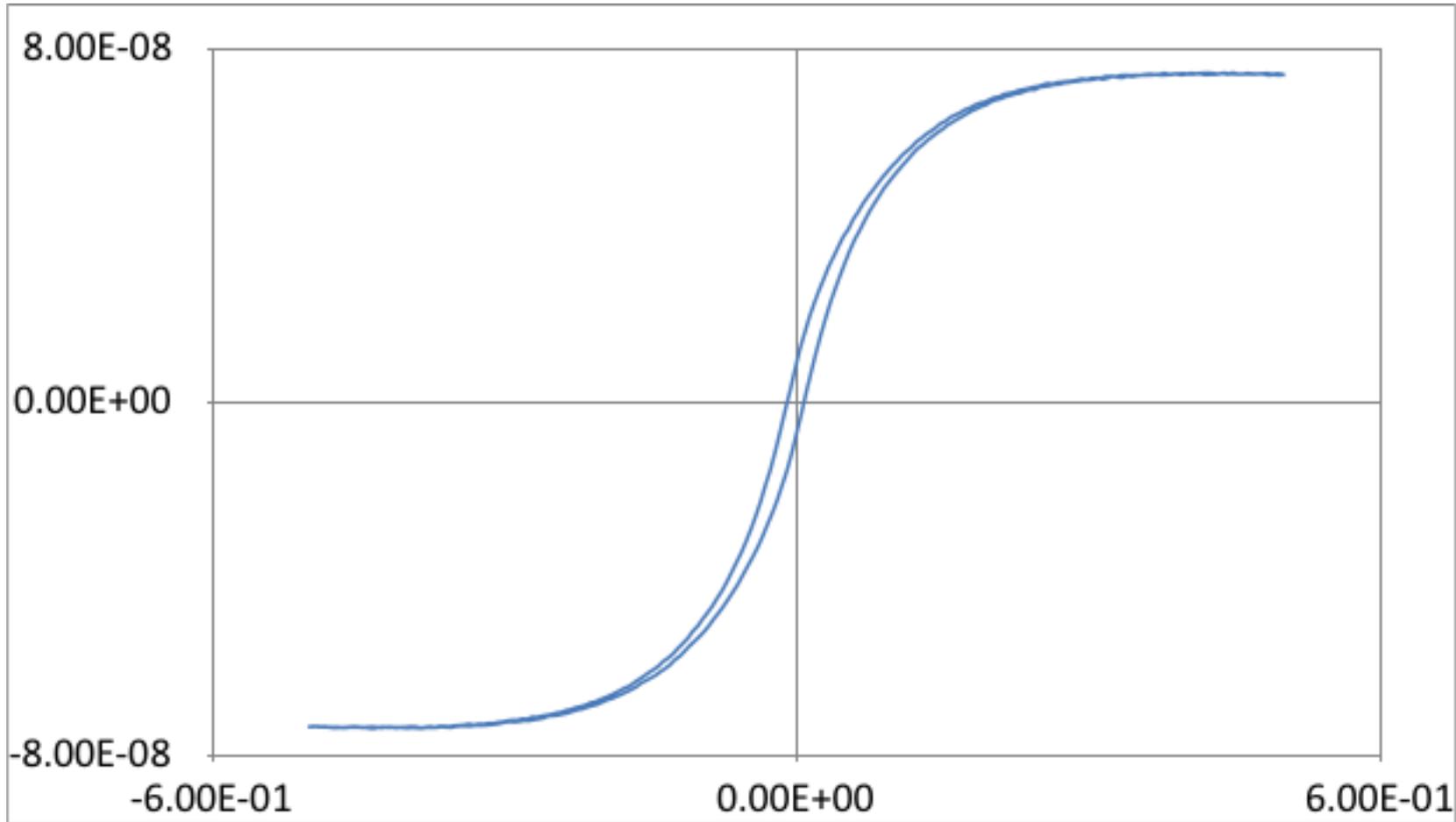




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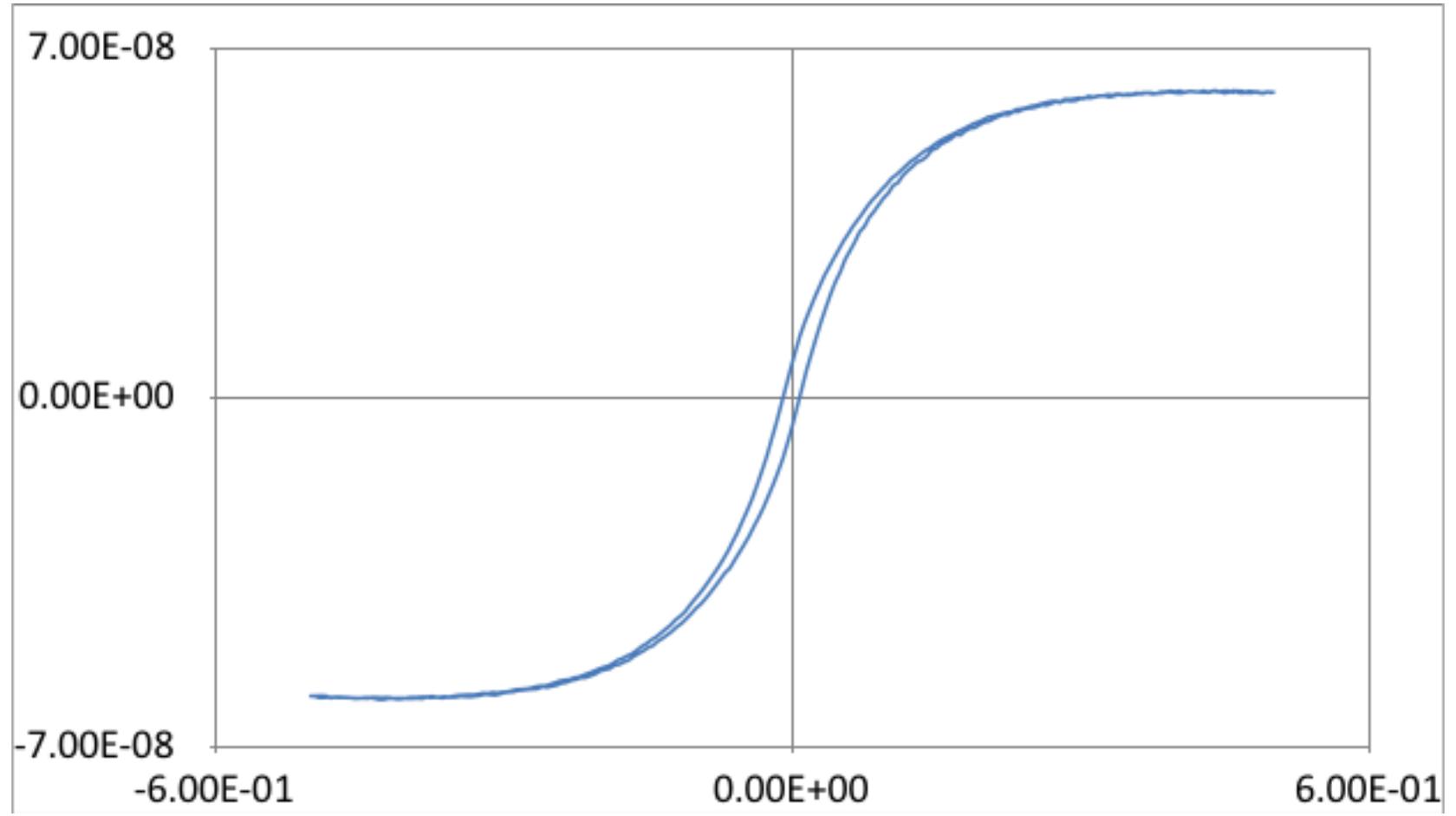
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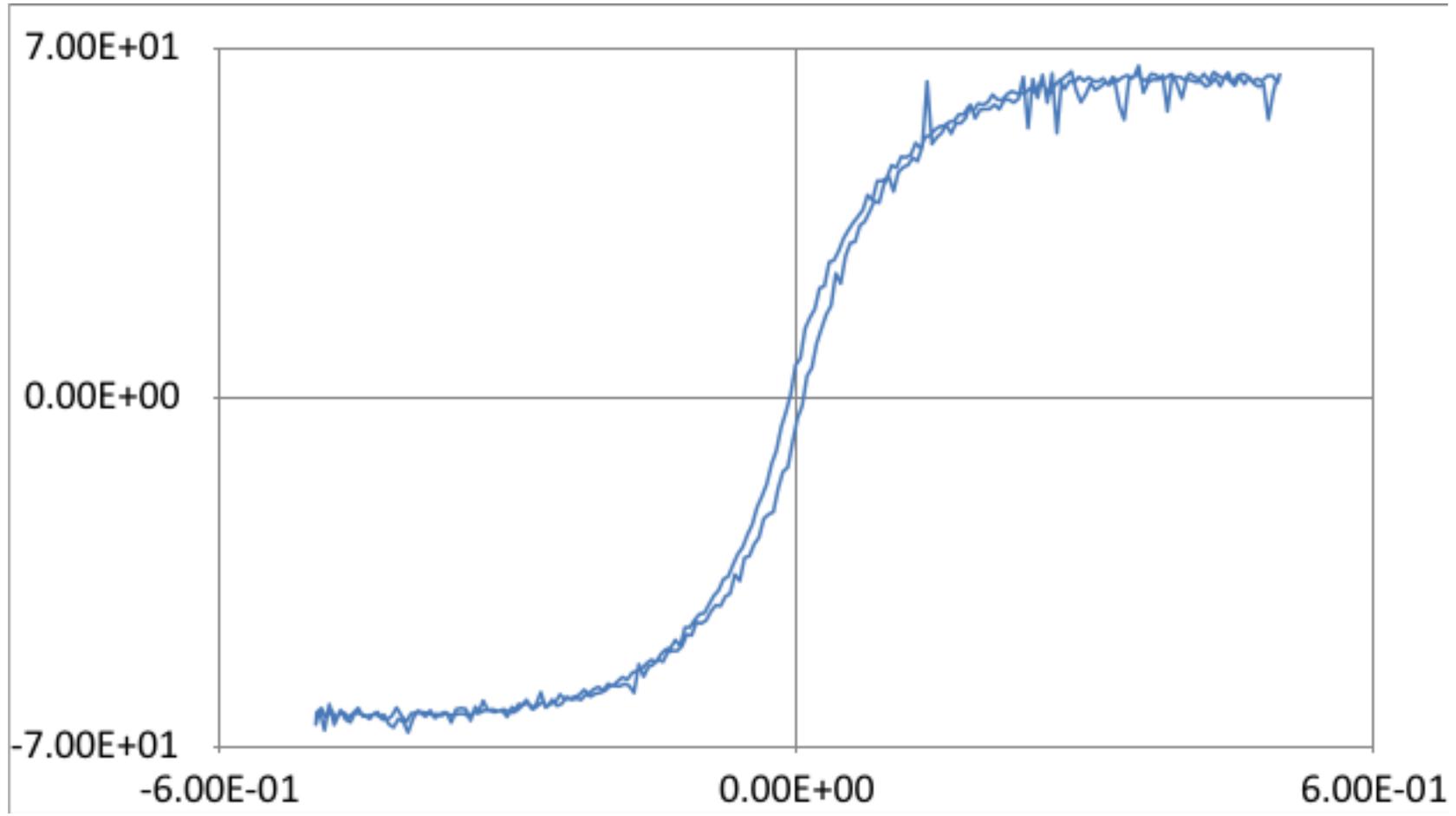




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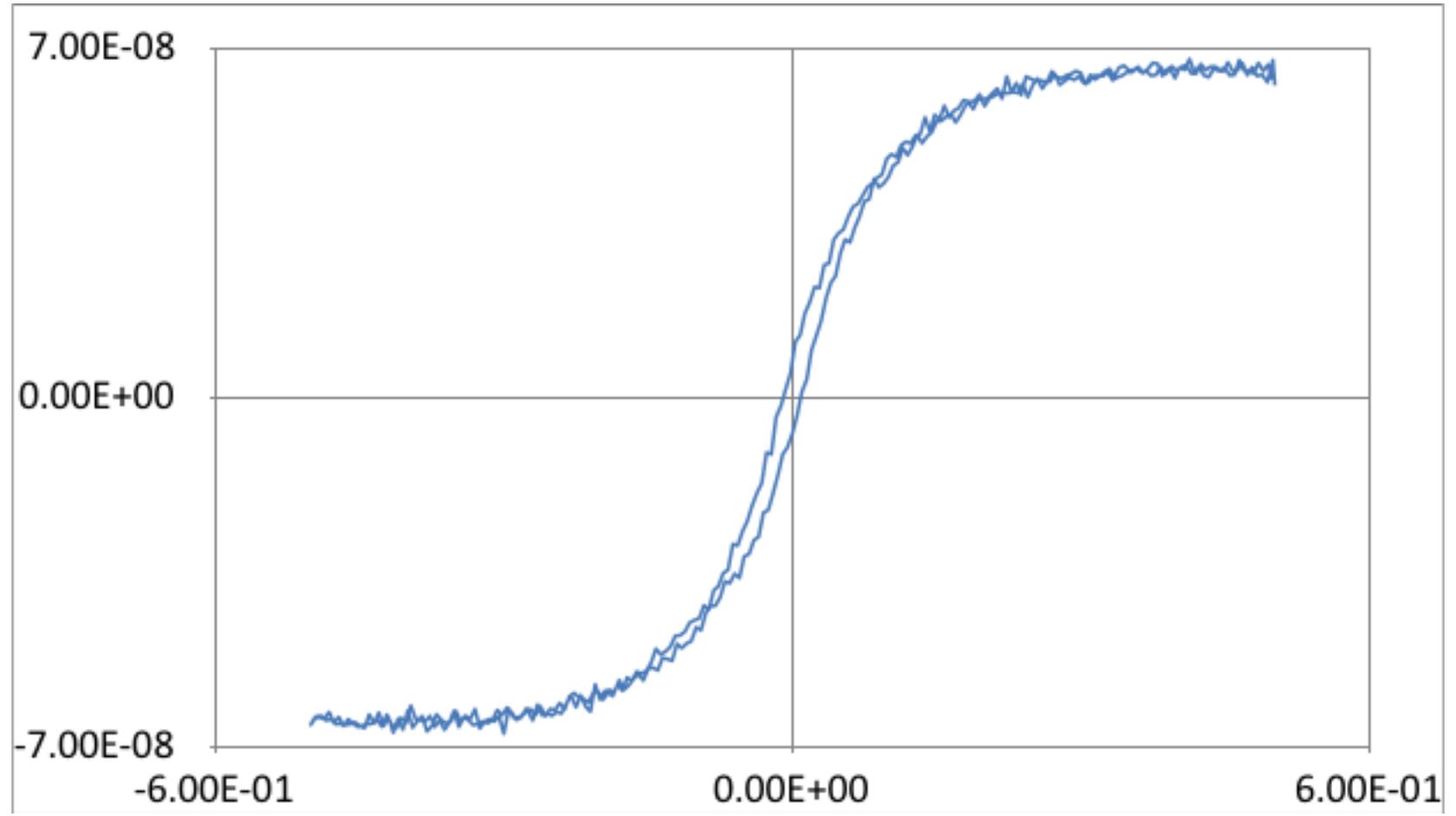
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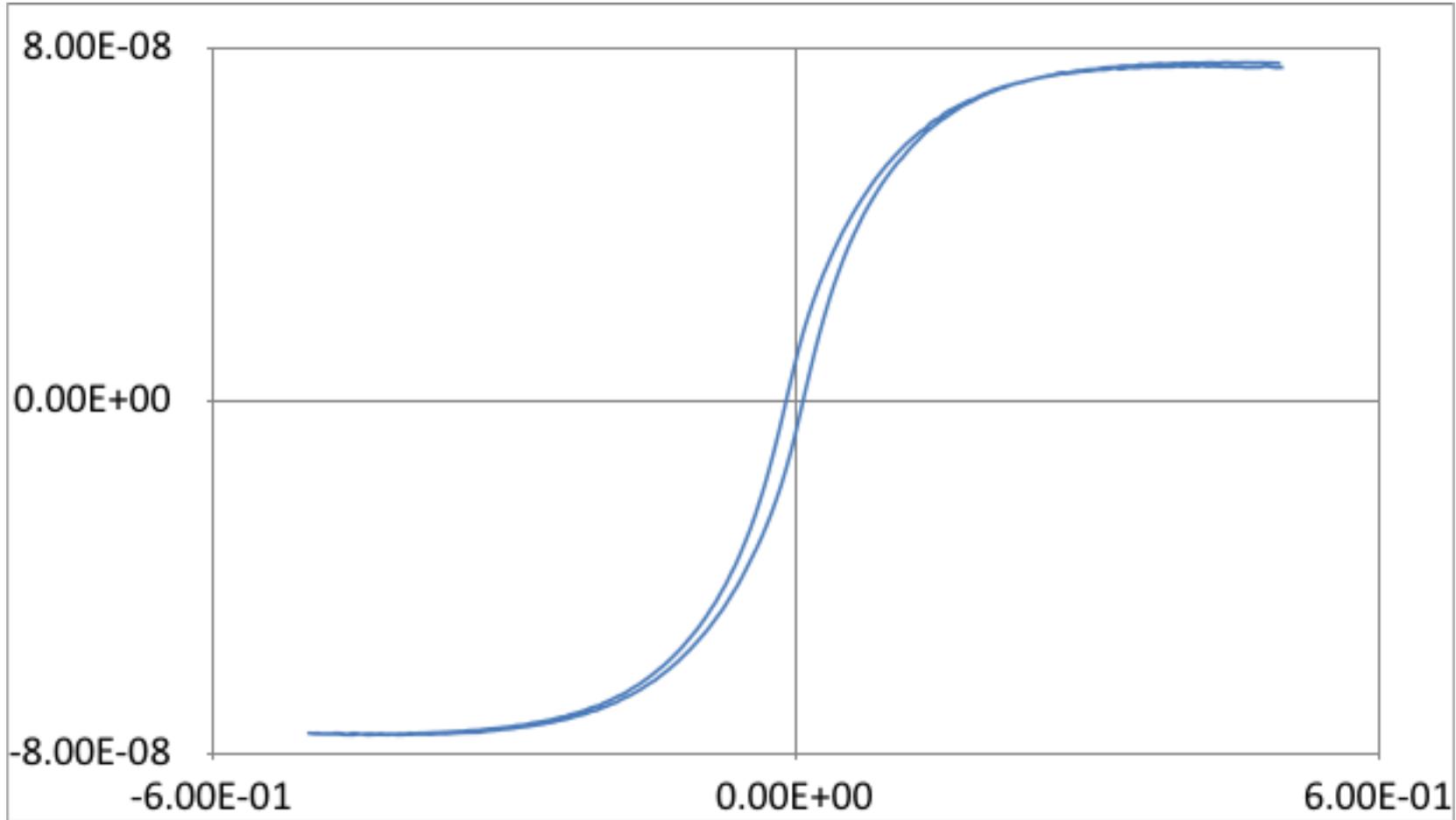




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