From Disorder to Order: Inheritance of Magnetic Remanence in Tetrataenite Bearing Meteorites from Multi-Phase Micromagnetic Modeling

Response to Reviewers

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We are resubmitting the revised manuscript entitled “From Disorder to Order: Inheritance of Magnetic Remanence in Tetrataenite Bearing Meteorites from Multi-Phase Micromagnetic Modeling”. We would like to thank both referees for revising our manuscript and for their valuable comments. We have addressed our responses to their comments below and we believe that their criticisms were constructive and led to better readability and overall quality of the manuscript.

**Reviewer 1**

1. Line 16. "Iron meteorites are believed to be fragments of mantle-stripped planetary cores". Does this statement strictly apply to all iron meteorites? Are there not some irons thought to have originated in other ways (e.g. evidence that IABs formed as isolated pools within the parent body; Ruzicka A. (2014) Silicate-bearing iron meteorites and their implications for the evolution of asteroidal parent bodies. Chem. Erde Geochem. 74, 3-48.)

A: Thanks for pointing out. The text was updated.

2. Line 65. 40 nm sounds a little high to me. Our calculations had the SD threshold more in the 20-25 nm range. Looking at the SI for Devienne et al. 2023 I see a similar value of equidimensional particles. I think it would be helpful to be specific about the threshold size, i.e. when talking about a threshold, specify what shape of particle it refers to.

A: The sizes are expressed in equivalent spherical volume diameter (ESVD), so 40 nm sized particles have a volume equivalent to a sphere with a 40 nm diameter. I just noticed that it was not stated in the paper, so I just included it (please, see section 2.1, L. 110-111). In Devienne et al., 2023 we assessed the SD size range for taenite particles also as a function of elongation; we observed SD forming up to ~40 nm (e.g., see Fig. S1g in Devienne et al., 2023). I do agree, however, that this can be more clearly stated in the text. The text is updated.

3. Line 83. "Experimental evidence for nucleation and growth" One way to determine this is via group theoretical analysis of the group-subgroup relationship between the space groups of taenite and tetrataenite. I worked with Chris Howard, who is an expert in group theory. Using the ISOTROPY package, he determined that, according to group theory, the transition MUST be 1st order. Unfortunately, this was not published formally, so remains a pers comm. However, if correct, it means the approach used here is indeed the correct one, and the approach used by Einsle et al is not correct. You could include this as an indirect pers. comm. from me or perhaps better reach out directly to Chris ([chris.howard@newcastle.edu.au](mailto:chris.howard@newcastle.edu.au)) to ask about the proof.

A: Thanks for sharing this information. The nature of the 1st order A1 to L10 phase transition in FeNi alloy is also included in the references cited in the text: “The chemically ordered structure is the thermodynamically stable phase below a critical temperature TCR; in this structure each crystal lattice site has a different probability of being occupied by one of the two atom types. In ferromagnetic systems, the critical order–disorder temperature can vary rather widely and marks a thermodynamically first-order phase transition in the Ehrenfest sense” (Lewis et al., 2014). Therefore, I decided to keep this sentence unaltered.

4. Line 103. "Ellipsoidal shapes". The tomography indicates that in general shapes are far more complex. I think it would be good to recognize here that ellipsoids present a highly idealized picture of the reality of the cloudy zone, and in fact there is a wide range of size and shape. It is easy to cherry pick a few islands that have roughly ellipsoidal shapes from the tomography data, but the majority are more complex.

A: Noted and addressed. The text was updated. Thanks.

5. Fig. 1 Models of nucleation and growth. Did you consider nucleation at multiple points in the grain? What impact, if any, might that have?

A: We did not consider multiple nucleation points in this work. However, I do believe that in general the main conclusions would still hold, i.e., that in SD taenite, inheritance is likely to occur, while SV states are likely to be modified to form MD states (i.e., no inheritance taking place). It’s important to note that for both nucleation scenarios we investigated, the results are virtually the same, indicating that the most important factors influencing the domain state modification through the phase transition are (1) the domain structure taenite precursor have before at the beginning of the phase transition (by means of the magnetic field it creates and that acts upon the nucleating phase) and (2) the orientation of tetrataenite’s MUA axis (which, in turn, is likely to be dictated by the field created by underlying precursor’s domain structure). As SD states produce a uniform magnetic field at the grain’s surface (see Fig. S2 in SI), the multiple nucleating sites would probably develop tetrataenite portions whose MUA axes align with the field — at the end we would have all regions with the same uniform state, thus creating a whole SD grain whose orientation is parallel to the precursor’s one (i.e., inheritance). For SV states it would probably be more complex, as vortex structures create non-uniform magnetic fields at the grain’s surface (Fig. S3 in SI), so different nucleating points would have different probabilities of alignment of tetrataenite’s MUA axis and, thus, could develop different domain structures according to the overall field at the nucleating site. I guess that, due to the very large magnetocrystalline anisotropy of tetrataenite, each individual region would develop a uniform domain structure aligned with the orientation of the MUA axis (dictated by the local field). In any case, it would still be true that, at the end of the ordering, the SV state in taenite is completely modified, such that any remanence possibly recorded previously would be reset. The transition from a MD to a SD led by magnetostatic interactions and/or external fields (as suggested by Einsle et al., 2018) would certainly be affected, though.

6. Line 122. MUA used here, but not defined until later in the manuscript.

A: Thanks for pointing out. It’s now defined in section 2.1 (L. 113).

7. Line 143. Threshold sizes for SD. Which dimension does this refer to? Long or short? I still feel slightly uneasy about referring to 50 nm as the threshold size for SD, as this refers to one arbitrarily chosen, elongated, grain geometry. Typically threshold sizes for SD in the literature are reported for equidimensional particles so they are more easily compared from material to material (e.g. 67 nm for magnetite is a well recognised number that refers to equidimensional particles).

A: I agree that referring to 50 nm as a threshold for taenite is indeed an overestimate. I have updated the text. Please also see my reply to comment #2.

8. Line 165 - relaxation times < age of solar system are 'unblocked'. Is this the right way to think about it? When considering unblocking, it is not the age of the solar system that is the key timescale, rather the time taken for the volume percentage of tetrataenite to increase to 100%? If the rate of ordering is faster than the relaxation time, then no unblocking would take place, as the transient state of partial order only exists for a short time? It may only take a very short time indeed (certainly a lot less than the age of the solar system) for an island to go from 0 to 100% ordered. Some discussion of what is the appropriate time scale is needed, and then if significantly shorter timescales are more important than some updating to the figures and conclusions might be necessary.

A: Thanks for pointing out. I think that the time-scale as the threshold for (un)blocking does not affect the main conclusions of the paper: that SD are preserved and SV are modified though chemical ordering. Moreover, all transition that are unblocked for relaxation times < age of solar system are also unblocked for relaxation times < 100 s. Therefore, we decided to keep Fig. 3 unaltered.

9. Fig. 2. Not immediately clear which dimension is 30 nm. Long or short? I don't think you state this explicitly anywhere.

A: You’re right. Please, see my replies to comments #2 and #7.

10. Line 168. Energy barriers. It is not clear what exactly the transition states are and what the barriers are referring to. If only a portion of the grain moment is unblocked, is there not a stable component remaining? Do we still call this unblocked? Need to be very clear with terms like 'unblocking' which most people associate with the complete thermal instability of the whole grain, and which might mean something different in this case as you have two phases. Is a grain that has only one part of it thermally unstable really 'unblocked'? To help this, it would be useful to have representative (e.g. 50% transition volume?) pictures of the 'before' and 'after' states corresponding to each of the relaxation times in Fig. 3. Without that it is very hard to picture what these energy barriers actually correspond to.

A: Thanks for pointing out. I created a figure (Fig. S1 in the SI) to illustrate the presentative ‘initial’ and ‘final’ LEM states adopted to calculate the energy barriers used to create the plots in Fig. 3.

11. Line 198 - experimental evidence suggesting c axis aligns with <100>. Actually, this is a fundamental consequence of the symmetry change involved in the phase transition. There is not really any flexibility or doubt here - the c axis of tetrataenite MUST be one of the pre-existing <100> directions of taenite.

A: Thanks for pointing out. The sentence was rewritten.

12. Line 218. Probability of magnetization state proportional to dot product M.B. Why? Is this not more likely a Maxwell-Boltzmann distribution based on the energy difference between differently oriented nuclei? i.e. proportional to exp(M.B/kT)?

A: exp(M.B/kT) represents the probability of a magnetic moment M aligning with the external field B at a certain temperature T. This is fundamentally different from what we are interested here, i.e., to estimate a certain probability of tetrataenite to align its MUA axis along a certain field component. As the MUA must align with one of the pre-existing <100> directions of taenite (as you correctly pointed out), we are interested in estimating what is the probability that, at the right beginning of the phase transition, tetrataenite’s c-axis will lie along the <100> direction over which the local magnetic field created by the precursor’s domain structure is larger. exp(M.B/kT) would then represent the probability of certain volume of tetrataenite to align its magnetic moment with the magnetic field B created by the precursor’s domain state AFTER the MUA axis is set, thus relating to a different physical process. The works of Paulevé et al. (1968) and Néel et al., (1964) have shown that magnetic fields play an important role in dictating the orientation of the c-axis, but we still lack (as far as I’m concern) a model that quantitatively accounts for the influence of external fields on the orientation of tetrataenite’s c-axis. Our estimated probability distributions can be considered as ‘educated guesses’. However, due to the lack of additional evidence, we decided to remove the discussion of probability distributions from the manuscript.

13. Line 248. You need to make a very clear distinction between a) fine cloudy zone that is close to the tetrataenite rim, which is fine due to rapid cooling rates and b) fine cloudy zone that is far from the tetrataenite rim that is fine because the Ni content is less and so spinodal decomposition initiates at lower temperature where kinetics and diffusion distances are low. Even slowly cooled meteorites have a fine cloudy zone far from the rim. It might be useful to use the calculations of Maurel et al. 2109 to explore the likely distribution of SD particles sizes in CZs cooled at different rates that formed above 320 {degree sign}C.

A: Thanks for pointing out. Please see the updated text, sections 4.2 and 4.5.

14. Line 255 - a new mechanism? Not really new - this was essentially the model put forward by Bryson et al. (2015) prior to the realization that re-recording could occur at the TT ordering temperature. In that model, the time-resolved records were linked to the different times of cooling into the spinodal region for CZ as a function of distance from the rim. The assumption was that magnetic recording occurred at or below the spinodal temperature.

A: I agree. The text was updated.

15. Line 264 - Reference to Fig. S16b. Can't see this - figures in my supplemental only go as high as S5!

A: Thanks for pointing out. Noted and corrected.

16. Line 264 - discussion of pqaleointensity estimates. I would take all estimates of paleointensity with a very large pinch of salt, as the models all completely ignore interactions between cloudy zone islands, which will surely have a large impact on the quantitative link between paleofield and magnetisation state of the CZ, and the mechanism of remanence acquisition is not well constrained (certainly the classic idea of CRM whereby particles nucleate small and grow through a blocking volume does not apply, as the mechanism of island formation is spinodal decomposition, not nucleation and growth).

A: Regarding the remanence acquisition, we’ve included a more detailed discussion in sections 4.2 and 4.3 (please, see the main text). In summary, according to Devienne et al. (2023), the blocking volume for taenite varies from ~14 – 22 nm (ESVD) depending on grain elongation (see Fig. 4c and Table S4 in Devienne et al., 2023). We used the model of Maurel et al. (2019) to estimate the islands size in fast-cooled meteorites at 350, 320 and 210 oC (see Fig. 5 in the main text). We show that most IVAs and IVBs have an average island size < 14 nm at 350 oC (when they form via spinodal decomposition) and that upon subsequent grain coarsening, the islands reach taenite’s SD size range. Therefore, they are still likely to have formed smaller than their blocking volume, therefore are capable of recording grain growth/crystallization remanent magnetizations (CRMs) as put forward by Devienne et al. (2023). Therefore, we argue that the fact that the islands in CZs grow via spinodal decomposition does not rule out the possibility of CRM acquisition in fast cooled (IVA and IVB) meteorites and in the finer regions of slowly cooled meteorites (see section 4.5).

The effect of interactions in fine-grained CZs is discussed in section 4.3. An shown by Einsle et al. (2018) and Mansbach et al. (2022), interactions in coarse grained CZs (where the SV to 2D transformation occurs) are likely to play a role in creating stable SD states. In fine-grained CZs, however, where SD states are likely to be preserved though tetrataenite chemical ordering, we argue that interactions might play a minor role as the blocking takes place at very small sizes, when the interaction fields are weaker (due to the weaker magnetic moments of the grains) (Baker & Muxworthy, 2023).

17. Line 265 - discussion of blocking volumes etc. Maurel et al. 2019 demonstrate that islands form via spinodal decomposition at >>60% of their final size - a fundamental consequence of how spinodal decomposition works. The idea of islands nucleating small and growing significantly through a blocking volume is not appropriate to spinodal systems. I think the discussion here needs to be careful to make this point - there is a danger that misconceptions of how spinodal decomposition works propagate through the literature if we continue to make analogy to grain growth CRM.

A: Please refer to the previous comment.

18. Line 291. As noted above, the fine end of more slowly cooled meteorites formed at lower temperatures due to lower Ni content, so there might be a different mechanism operating. For quantitative XMCD studies, we tend to avoid analysis of the fine regions and focus deliberately on the coarser regions which are more likely to have been vortex state taenite that then transformed. According to our modelling, the generation of two-domain states and the subsequent response of those domain walls to the interaction fields between islands may be an integral part of the recording mechanism. For this reason, we tend to focus on regions we are confident started as vortex states initially. Your finding that 2D states are also generated by nucleation and growth is therefore significant.

A: Thanks for pointing out. The distinction between finer CZs (closer to tetrataenite rim) in fast-cooled meteorites and in slowly-cooled meteorites (further from tetrataenite rim) is presented better explained in the updated manuscript, sections 4.2 and 4.5. We used the model of Maurel et al. (2019) to estimate the extension of CZs of slowly-cooled meteorites where C the SV-to-2-domain transition occur and CRM inheritance are likely to occur, as well as the time differences between these distinct records (please, see section 4.5).

19. Line 294. See comment previously - this is wrong way to think about island formation in a system undergoing spinodal decomposition.

A: Please refer to my comments above.

20. Line 334. How many particles are needed to get a good paleomagnetic measurement. Most of these calculations are based on a lack of knowledge of the actual mechanism of remanence acquisition. I think I can confidently state that the mechanism operating in the coarse to medium sized CZ regions that are typically targeted in XPEEM studies bears no relation to the sorts of calculations performed by Berndt et al. to estimate numbers of particles needed, which are based on conventional TRM theory - the mechanism operating here is fundamentally different (i.e., transformation from SV-2D). I would be wary of making a definitive statement like this based on comparisons to any model calculation that uses a different mechanism. I agree that for rapidly cooled meteorites, where the taenite particles are SD, grew as taenite above 320 {degree sign}C, and acquired remanence via TRM and/or CRM mechanism {plus minus} interactions, will be hard/impossible to get a result from. However, the medium-coarse CZ is a different story. I think that needs to be made clear here.

A: I agree that the mechanism of remanence acquisition in the coarse to medium sized CZ is different from those depicted by Berndt et al. (2016). However, the equations derived by the authors are valid for both TRMs and CRMs; hence, based on my response to comment #16 (see section 4.3), we argue that they can still be used to evaluate the number of islands that have to be sampled to obtain a reliable paleointensity estimate from fast cooled meteorites.

21. Line 382. Again, if CZ formation is due to spinodal decompsition, then this picture of small grains growing to their blocking volume is incorrect. Spinodal decomposition is a different mechanism to nucleation and growth. It instead occurs via the growth in amplitude of a compositional fluctuation with a wavelength that is determined by the balance between gradient energy and diffusion length. This is partly why calculations based on conventional CRM theory may not be appropriate in this case.

A: Please, refer to my response to comment #16.

22. Line 385 - different regions of the CZ. How extensive would such a region be? It might be useful to take the results of Maurel et al. 2019, which predicts island sizes for different bulk composition (i.,e. different regions of the CZ) and for different cooling rates. It should be possible to predict the lateral extent of any region that fits these criteria (i.e. islands small enough to be SD forming at temperatures 400-320{degree sign}C). When deciding what is SD, however, you will have to decide what threshold size (or range of sizes) is appropriate. I'd suggest using 20-25 nm for equidimensional particles as a reference.

A: Thanks for pointing out. We used the model of Maurel et al. (2019) to estimate the extension of CZs of slowly-cooled meteorites where the SV-to-2-domain transition occur and CRM inheritance are likely to occur, as well as the time differences between these distinct records (please, see section 4.5).

**Reviewer 2**

1. Throughout the paper, there are various places where the English can make the key points more difficult to comprehend. For instance, in line 92, the first clause should read "Within the taenite SV size range..." (this persists throughout most of the paper). I was not able to understand the sentences on lines 105-108 and lines 168-170. If possible, a thorough proofread should pick up most comments like these, which should hopefully serve to improve the readability of the paper.

A: This sentence is essentially based a previous work where we showed that taenite forms SD for sizes between ~14 – 34 nm and non-SD – i.e., single vortex (SV) – states for sizes between ~40 nm up to 90 nm. Both SD and SV states are shown to be highly stable, meaning that they can preserve paleomagnetic records over geological (i.e., billion-year) timescales. The text was updated to improve its readability.

2. Some sentences are light on specific information, which can make them difficult to understand completely or easier to misinterpret. For instance, the sentence on lines 61-64 uses phrases such as "nm-sized taenite" and "magnetization states that are stable over billion-year timescales". Given a key outcome of this study concerns island size and domain state, it would be good if specific size ranges could be presented and specific domain states could be stated.

A: Please, see my comment above. The text was updated to improve its readability.

2.1 This would help to increase clarity and understanding. Another example is discussing the time discrepancy of 10^5 years: the paper refers to this discrepancy throughout, but doesn't say regularly if this is earlier or later. Going through the paper to look for examples of unclear or easily misinterpretable language would help the understandability of the paper.

A: This was cited in the abstract (please, see L. 29-32 in the first version; L. 26-30 in the updated version: “We show that in cases where tetrataenite inherits the domain states of its precursor taenite, the origin of the remanence can be up to ~105 years older than in cases where tetrataenite resets the precursor SV magnetization.”. I understand, however, that due to the importance of such statement, if should be put more clearly in the text. Please see section 4.2 in the updated manuscript.

3. Line 34: remove "and asteroids"

A: Noted and corrected.

4. Lines 123 and 136: Acronyms are used before they are defined.

A: Thanks for pointing out. Noted and corrected.

5. Line 178: replace 'dislocated' with 'displaced'.

A: Noted and corrected.

6. Line 327: replace 'reflecting' with 'resulting'.

A: Noted and corrected.

Major comments

7. One key finding of the paper is that CZs with small islands (i.e., those in quickly cooled meteorites) recorded their magnetization ~10^5 years earlier than previously thought. The authors state that this corresponds to "very different stages" of planetary evolution. High-precision Al-Mg dating of meteorites has a typical lower limit on its uncertainty of ~1.5x10^5 years. As such, the change of 10^5 years presented in this study is within the error of all previously reported radiometric dates. If this time period corresponds to very different behaviors (as suggested by this paper) then all radiometric dating to date doesn't capture any of these changes, suggesting that we are unable to understand early solar system chronology at all. Moreover, this comment specifically referred to quickly cooled iron meteorites, like the IVA iron meteorites. A paper that models the solidification of their parent core (Neufeld et al., 2019) predicts this process takes ~6.7-20 Myr. As such, the timing change presented in this paper is small compared to the overall solidification of this core (corresponding to <~1.5% of the total solidification time). The paper states that "...small planetesimals usually have short-lived thermomagnetic activity (~10^5 - 10^6 years)", but doesn't cite a paper to demonstrate an example of where ~10^5 years corresponds to widely different behaviors. Bearing all of these in mind, I believe the proposed consequences of a change of ~10^5 years in recording time should not be emphasized as heavily as it is throughout the paper.

A: Thanks for pointing out. Paleomagnetism in meteorites, in particular that performed with the CZ, opens a window of opportunity events and time windows that are, as you correctly pointed out, inaccessible with other dating methods. For example, by investigating different regions of the CZ in Esquel and Imilac pallasite, Bryson et al. (2015) was capable of reconstructing a ~10-20 Myr dynamo history of the pallasites’ parent body; each subregion in the CZ of these meteorites corresponds to a ~1-2 Myr records of dynamo activity (i.e., very close to the Al-Mg dating you cited). In another work, Bryson et al. (2017) reconstructed a time-resolved record spanning ~200 kyr from the CZ of the IVA Steinbach meteorite which included a field reversion over a minimum period of ~30 kyr. This, therefore, suggests that ~105-years time windows can encompass distinct and important events of the dynamo history of a planetesimal (i.e., excursions and reversals), and provide a wealth of information about planetary differentiation and dynamo evolution that are inaccessible using other dating methods.

Moreover, it was recently shown that the mechanism leading dynamo generation in IVA parent body may be different from that proposed Neufeld et al. (2019) (i.e., through inwards solidification), and may have probably involved the formation of a “rubble-piled” inner core, which in turn extracted heat from the surrounding molten metal and led to solidification and light element expulsion, ultimately powering a compositionally-driven dynamo (Zhang & Bercovici, PNAS, 2023). Notably, this work predicts a way shorter dynamo activity in the IVAs parent body, spanning from ~1 to ~3 Myr. Therefore, time periods spanning ~105 years, as indicated to occurs due to CRM inheritance in fast cooled meteorites (see section 4.2 and 4.3) can correspond to ~10% to > 50% of the dynamo history predicted to operate in the IVAs’ parent body. We argue, therefore, that taking inheritance of SD states though tetrataenite ordering into account when assessing paleomagnetic records in the CZ of fast-cooled IVAs is an important finding, as it indicates that such magnetic signature measured from SD tetrataenite-containing CZs reflects up to ~105 years older records, thus represent a different stage (possibly the onset) of the IVAs’ parent body dynamo field.

8. In section 4.3, the authors consider the impact of their results on paleointensity estimates. Namely, they adopt an island size of 14 nm to reanalyze the results of Bryson et al., (2017). However, the results of Bryson et al., (2017) have already be reanalyzed by Maurel et al (2019), who found that the specific growth mechanism of spinodal decomposition means that islands do not simply grow through a blocking volume, and in fact can form with a size close to their present day size. I recommend that the impact of the findings of this study on paleointensitiy are considered alongside the island growth history and the reanalyzed paleointensities presented by Maurel et al (2019).

A: Thanks for your suggestion. We included a more detailed discussion about the nature of the paleomagnetic signature in fast-cooled IVA meteorites (i.e., a phase-transition-inherited CRM) in section 4.2. We also included a discussion about how our findings affect the paleointensity estimated firstly obtained by Bryson et al. (2017) and lately revisited by Maurel et al. (2019) for the IVA Steinbach.

9. In the paragraph from line 289-307, the authors consider the possibility that a time-resolved remanence can be recorded across the CZ. They argue that small islands in the fine CZ (further from the tetrataenite rim) inherit their remanence from pre-existing taenite islands, while large islands (closer to the tetrataenite rim) recorded a new remanence on ordering. As such, the remanence in the fine CZ would predate that in the coarse CZ in this model. However, the fine CZ forms from regions of taenite with lower bulk Ni concentrations, and so forms at lower temperatures (and later times). At some distance across the CZ (i.e., at a critical bulk Ni concentration), this will occur once the material has cooled below 320 C, such that the islands will form as tetrataenite rather than forming initially as taenite and then ordering to form tetrataenite. As such, the fine CZ cannot inherit a remanence from existing taenite in this case because it formed directly as tetrataenite. It may be possible that an intermediate distance across the CZ formed following the history assumed by the authors (i.e., islands form as taenite at >320 C and are small enough to be SD), but, again, the actual growth mechanism of spinodal decomposition (i.e., the results of Maurel et al., 2019) need to be considered here because these small islands do not need to have existed. I suggest that this proposed recording history across the CZ is considered and presented carefully, and that a discussion of the consequences of the smallest islands having formed as tetrataenite (rather than forming as taenite and ordering to become tetrataenite) and the impact of this on remanence acquisition is included.

A: Thanks also for pointing this out. We used the model of Maurel et al. (2019) to estimate the extension of CZs of slowly-cooled meteorites where the CRM inheritance is likely to occur (please, see section 4.5).