Reviewer: 1

Comments to the Author
The limitation to non interacting particles rule out too many materials of interest. For materials with hysteresis these results are only valid for minor loops attached to the major loop, There is no comparison with any other anisotropy or real measurements.

The limitation to noninteracting particles does rule out some systems of interest. However, this publication aims at a precisely delineated problem and proposes a methodology that can be easily extended to include magnetostatic interactions. The FORC methodology is based solely on minor loops attached to the major loop, so this second statement is puzzling. There is only comparison to uniaxial anisotropy as this is the only case that has been previously studied under the conditions we are concerned with, namely, noninteracting dispersions of randomly aligned fine particles.

Reviewer: 2

Comments to the Author
The manuscript by Grijalva et al. deals with a nontrivial topic, namely with the interpretation of FORC diagrams for nanoparticles with cubic crystal symmetry. The topic is basically suitable for IML, but I have several concerns about the paper and its scientific context.

(i) The first sentence of the abstract, namely "First order reversal curve (FORC) diagrams are routinely used as a material’s magnetic fingerprint", is a stretch. The method is cumbersome, and for any given phenomenon or new material, M(H) measurements have remained the most important routine method in magnetism. FORC diagrams are useful for some aspects of magnetism research, such as minor loops, but aside from this, FORC pattern are difficult to relate to structural and magnetic features of interest to a broader magnetism community. Exaggerating for simplicity, FORC patterns like a 'blue-bird research' in biology: blue birds are different from yellow or red birds, which is interesting to know, but does it really matter biologically?

FORC diagrams have become increasingly popular (Figs. 1, 2) as a method that provides a wealth of information beyond standard magnetic hysteresis M(H) measurements (Roberts et al. 2000, 2006, Muxworthy and Dunlop 2002, Pike et al. 2005, Dumas et al. 2007, Egli et al. 2010). The reviewer’s claim that the cited first statement is ‘a stretch’ denigrates the work of many researchers in this area. The reviewer is right to assert that ‘FORC patterns are difficult to relate to structural and magnetic features of interest’ as the method and its interpretation is still a hot area of research. This paper aims precisely at solving some of the problems faced by researchers using this technique.

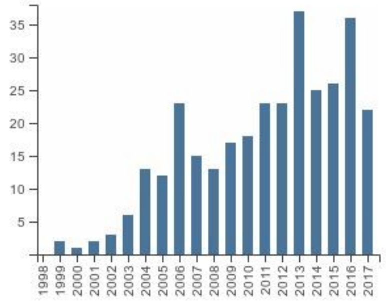

Figure 1. Number of papers on "first order reversal curve" or "forc diagram" per year. Source: Web of Science.

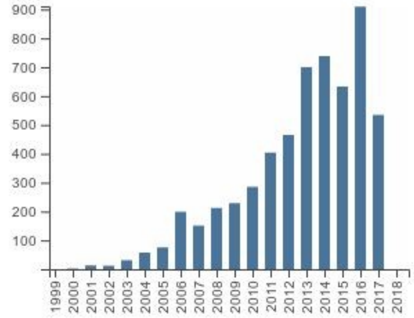

Figure 2. Number of citations of papers on "first order reversal curve" or "forc diagram" per year. Source: Web of Science.

Roberts, A.P., Pike, C.R. and Verosub, K.L., 2000. First-order reversal curve diagrams: A new tool for characterizing the magnetic properties of natural samples. *Journal of Geophysical Research: Solid Earth*, *105*(B12), pp.28461-28475.

Muxworthy, A.R. and Dunlop, D.J., 2002. First-order reversal curve (FORC) diagrams for pseudo-single-domain magnetites at high temperature. *Earth and Planetary Science Letters*, *203*(1), pp.369-382.

Pike, C.R., Ross, C.A., Scalettar, R.T. and Zimanyi, G., 2005. First-order reversal curve diagram analysis of a perpendicular nickel nanopillar array. *Physical Review B*, *71*(13), p.134407.

Dumas, R.K., Li, C.P., Roshchin, I.V., Schuller, I.K. and Liu, K., 2007. Magnetic fingerprints of sub-100 nm Fe dots. *Physical Review B*, *75*(13), p.134405.

Roberts, A.P., Liu, Q., Rowan, C.J., Chang, L., Carvallo, C., Torrent, J. and Horng, C.S., 2006. Characterization of hematite (α-Fe2O3), goethite (α-FeOOH), greigite (Fe3S4), and pyrrhotite (Fe7S8) using first-order reversal curve diagrams. *Journal of Geophysical Research: Solid Earth*, *111*(B12).

Egli, R., Chen, A.P., Winklhofer, M., Kodama, K.P. and Horng, C.S., 2010. Detection of noninteracting single domain particles using first-order reversal curve diagrams. *Geochemistry, Geophysics, Geosystems, 11*(1).

(ii) The authors claim a "novel approach for numerically calculating the FORC diagram of a uniform noninteracting dispersion" of particles (p. 2) but later admit that similar cubic systems have been investigated in the past (p. 4). More generally, FORC curves are additive as far as the magnetization is concerned, so the only thing one basically needs is the FORC diagrams of single Fe- and Ni-type nanoparticles as a function of field and measurement direction. The respective two elements have K1 > 0, with <001> easy axes, and K1 < 0, <111> easy axes, and the behavior of the particles is somewhat affected by K2. The present paper also investigates <110> easy axes, but K2 is not independent in cubic materials but strongly coupled to K1.

The reviewer is missing the point, explicit in the paper, that previous studies use specific grain arrangements, i.e., their computed FORC diagrams lack generality and by definition cannot isolate the effect of magnetostatic interaction between the particles. The reviewer's statement that the FORC curves are additive is correct, in the noninteracting case. This is precisely one of the facts we explicitly use in the paper to compute our FORC diagrams. However, the nonlinear nature of the problem means that it is difficult to extrapolate results from one material to another. We have chosen to model magnetic minerals that are essential to rock magnetics and included a 'dummy' material that has <110> easy axes so we cover all easy axes possibilities. Magnetite, iron and greigite have negligible K2 at room temperature, so we find his last statement somewhat puzzling.

(iii) It would be useful to compare the present FORC diagrams with those of aligned cubic particles with K1 > 0 or K1 < 0. The authors mention some research in this direction (p. 2), but no explicit comparison has been made. For example, Sect. III, which is not bad to start with, would greatly benefit from such a comparison.

The focus of this paper is noninteracting particles. This is not just for simplicity, rather, identification of a noninteracting single-domain phase is essential for rock magnetic studies, e.g., for determination of reliable palaeointensities. In the noninteracting case, if the particles were aligned, the FORC diagram will be identical to the FORC diagram of a single, isolated particle. We concede that a more explicit comparison could be made and we have amended our manuscript to reflect this.

(iv) The single-domain character of a particle (p. 2) is irrelevant to the applicability of the Stoner-Wohlfarth model (uniform rotation). The former is an equilibrium property, whereas the latter is a nonequilibrium (or hysteretic) feature. In practice, many nanoparticles are single-domain but undergo nonuniform magnetization reversal. This limits the experimental relevance of the present paper.

This is not the case. Both characteristics, single-domain and coherent rotation are essential for the Stoner-Wohlfarth model. The reviewer's claim that many single-domain nanoparticles undergo nonuniform magnetisation reversal is partially correct. This behaviour is only expected to occur for particles only slightly smaller, up to 4 nm smaller, than the single-domain to single-vortex threshold. This effect has been modelled by Valdez-Grijalva et al. 2017 by a systematic study of the energy barriers between local energy minima and found evidence that there exists a range of fine particles which are stable single-domain (in laboratory timescales) and undergo coherent rotations. Identification of this type of particles is essential por palaeomagnetic studies.

Valdez-Grijalva, M. A., Nagy, L., Muxworthy, A. R., Williams, W., Fabian, K., 2017. The micromagnetic structure and palaeomagnetic recording fidelity of sub-micron greigite (Fe3S4). *Submitted to Earth and Planetary Science Letters.*

(v) Explaining micromagnetism in terms of a minimization of a Gibbs free energy (p. 3) is wrong. This minimization would yield an equilibrium state, but hysteresis and FORC diagrams involve nonequilibrium spin configurations. Micromagnetic calculations do no minimize the free energy but trace metastable energy minima as a function of field, which is a very different thing.

Micromagnetic algorithms are in fact based on minimisation of the Gibbs free-energy functional; this is a widely accepted method. Our algorithm obtains local energy minima (metastable states) as a function of the external field, which is what accounts for the hysteretic behaviour.

(vi) How can the FORC distribution of noninteracting cubic single-domain particles be "strongly sensitive to the mineralogy", as stated in the conclusions?

This is evidenced by the different FORC patterns obtained for each mineral.

(vii) As most FORC manuscripts, the present one is extremely difficult to read for non-specialists. This includes some intellectual in-breeding, such as crediting the discovery of coercivity to Mayergoyz 1986. The abstract is rather vague scientifically but full of acronyms (FORC, SD, MCA). It should be the other way around!

This paper does not aim to give the most detailed introduction to the FORC methodology. References in the paper like Roberts et al. 2000 do. Nowhere in our paper do we attribute the discovery of coercivity to Mayergoyz 1986 but only the use of the second order mixed derivative that defines the FORC distribution. The use of acronyms in the abstract is to avoid repetition and help the readability of it. Also, these acronyms are of standard use.

In summary, this paper is not of a letter-type for a variety of reasons. A carefully improved version of the manuscript would probably be suitable as a regular article in IEEE Transactions on Magnetics, but this is not the question here.

Small changes have been made we feel improve the manuscript. We accept the reviewer's reccomendation of submitting to IEEE Transactions on Magnetics, instead.