Comment on “Thermal fluctuations of magnetic nanoparticles”, cond-mat/arXiv:1209.0298

J.-L. Déjardin, H. Kachkachi, and J.-M. Martinez
PROMES CNRS UPR8521, Université de Perpignan Via Domitia, 52 avenue Paul Alday, 66860 Perpignan, France

We comment on some misleading and biased statements appearing in the manuscript “Thermal fluctuations of magnetic nanoparticles”, cond-mat/arXiv:1209.0298, about the use of the damped Landau-Lifshitz equation in conjunction with the kinetic Langer theory for the calculation of the relaxation rate of magnetic nanoclusters. We provide simple scientific arguments, part of which is well known to the whole community, demonstrating that the authors overstate the issue and contradict a work they have co-published earlier.

Here we would like to draw the reader’s attention to the following scientifically misleading and biased statement on page 46 of the manuscript cond-mat/arXiv:1209.0298: “...... Unfortunately, some authors (see, e.g., Ref. 137 and 138) have ignored this property of the Landau-Lifshitz equation and, in consequence, have used this intrinsically under-damped equation in conjunction with the intrinsically IHD Langer formula for the calculation of the escape rate in all damping ranges. Thus the ensuing escape rate formulas [Refs. 137, 138] are misleading and not valid for experimental comparison both at low damping, where they coincide with the TST rate, and also in the IHD range, $\alpha \gtrsim 1$, where they predict nonphysical behavior of the rate, namely, a rate in excess of the TST one.” In the sequel, this review paper will be referred to as CK.

Let us now give a brief account of our scientific point of view concerning the issue of damping in the Landau-Lifshitz equation and its use in Refs. (1–3), Refs. (137, 138), together with Langer’s approach. This is, of course, known to the whole community working in this area, and we apologize for having to reiterate it once again [17].

It goes without saying that the damping issue is a subtle one and takes on special relevance in magnetism. The issue dates back to 1935 when Landau and Lifshitz published their seminal paper on magnetics, and has been since a strong point of debate and controversy through hundreds of publications and conference proceedings, especially after Gilbert proposed, in the 1955 MMM conference proceedings, a new form for the magnetization damping. Despite this long period of investigation, the issue has not been settled yet and the very origin of damping eludes any simple interpretation. The main reason is that damping is rooted in various kinds of correlation processes, both intrinsic and extrinsic, which cannot be captured by a few phenomenological parameters added to the equation of motion. It would be too long, if not impossible, to give a fair account of the divers approaches and interpretations of damping in magnetic systems. A brief account can be found in Ref. 4 [Landau-Lifshitz or Gilbert damping ? That is the question] and Ref. 5 [Origin of intrinsic Gilbert damping].

The equation of motion describing the magnetization dynamics with a phenomenological damping parameter can be represented as one of the following two well-known forms:

1. The Landau-Lifshitz equation (LLE)

$$\frac{d\vec{M}}{dt} = -\gamma \vec{M} \times \vec{H} - \lambda \frac{\gamma}{M} \vec{M} \times \left( \vec{M} \times \vec{H} \right),$$

with $\lambda$ being a (dimensionless) dissipation parameter and $\vec{H}$ the effective field.

2. The Landau-Lifshitz-Gilbert equation (LLGE)

$$\frac{d\vec{M}}{dt} = -\gamma \vec{M} \times \vec{H} - \frac{\alpha}{M} \vec{M} \times \frac{d\vec{M}}{dt},$$

where $\alpha$ is another (dimensionless) dissipation parameter.

Mathematically, the two equations (1) and (2) are equivalent. Indeed, substituting for $d\vec{M}/dt$ on the right-hand side of Eq. (2) the same right-hand side and working out the resulting double cross product $\vec{M} \times \left( \vec{M} \times \frac{d\vec{M}}{dt} \right)$, and using $\vec{M} \cdot \frac{d\vec{M}}{dt} = 0$ (because the module of $\vec{M}$ is constant), we obtain

$$\frac{d\vec{M}}{dt} = -\left( \frac{\gamma}{1 + \alpha^2} \right) \vec{M} \times \vec{H} - \left( \frac{\alpha}{1 + \alpha^2} \right) \frac{\gamma}{M} \vec{M} \times \left( \vec{M} \times \vec{H} \right),$$

which is just Eq. (1) upon making the following substitutions

$$\frac{\gamma}{1 + \alpha^2} \to \gamma, \quad \frac{\alpha}{1 + \alpha^2} \to \lambda$$

in the first and second terms, respectively. Note that this transformation depends on the normalization used in both equations [see e.g. http://en.wikipedia.org/wiki/Landau-Lifshitz-Gilbert_equation]. Another useful form of Eq. (3) consists in rewriting it in terms of the re-scaled time $\tau = t / (1 + \alpha^2)$, and thus also re-scaling the Néel free diffusion time $\tau_N$ [9].
Further discussion of the two equations and their comparison can be found in the textbooks [6,7]. It is worth mentioning the work in Ref. [8] where it is rigorously shown that the LLGE damping term is a mere re-scaling of time by a complex constant. Moreover, it can be easily shown [9] that the Fokker-Planck equations associated with the stochastic analogs of the two equations [1][2] are also identical.

From the experimental point of view, there is no clear cut proof as to which equation has to be used in general. In practice, based on many investigations, it has been agreed upon that for small damping, LLE and LLGE are almost the same and thereby the former is then assumed to be more suited to small damping regimes. Indeed, for small damping, the transformation in Eq. [4] boils down to identity. However, many workers obtain the LLE damping for low frequency, long wavelength dynamics [4]. For high damping one would expect a damping-dependent gyromagnetic ratio, but this effect has still to be confirmed by experiments.

Now, the work [3] (Ref. 137 in CK) uses the LLE for obtaining the attempt frequency that enters the prefactor of Langer’s relaxation rate. Had we used the Landau-Lifshitz-Gilbert damping instead we would have obtained expressions that can be recovered by making the substitution [4]. A concrete example illustrating this procedure is provided by the work published in Ref. [10]. In this reference, Appendix B summarizes the analytical expressions obtained in Ref. [1] (1st paper in Ref. 137 in CK), for a system of two exchange-coupled magnetic moments using Langer’s approach. Then, these analytical expressions were compared in Figs. 2 and 3 of Ref. [10] to the results of the fully independent numerical method of matrix continued fractions, with a fairly good agreement.

Last but not least, let us mention a few points about this review that deserve special attention from the reader.

- It is curious how the authors’ select their references when they write “...some authors (see, e.g., Ref. 137 and 138) have ignored this property of the Landau-Lifshitz equation”; this is a rather biased and non objective manner in reviewing the literature, at variance with what a reader expects from a review article. Indeed, one of the well-known specialist in this area, Dmitry Garanin, and who is acknowledged by the authors for his “direct or indirect” contribution to this review, has published fundamental and well-known contributions with strong impact on the developments in this area of physics. The authors seem to ignore the fact that all of Garanin’s papers exclusively use the Landau-Lifshitz equation. The reason is, of course, scientifically motivated and is as explained above. In the work [11] a kind of phase diagram was obtained for uniaxial anisotropy with precise crossovers between various damping regimes.

- The authors claim that the analytical expressions published in Refs. 137, 138 are misleading and “not valid for experimental comparison”. It is indeed very important to show some care for comparison between theory and experiments. However, this manuscript which is a big review that covers at least two decades fails to provide a single comparison of experiments with any of the authors’ own theoretical work that goes beyond the Néel-Brown model. The only two figures 8 and 22 that show a comparison between experiments and Néel-Brown model, are borrowed from the literature. A successful comparison between the Néel-Brown model and experimental measurements on single magnetic nanoparticles was achieved many years ago by W. Wernsdorfer et al. in the seminal work [12–14].

- The 2nd article in Ref. 137 in CK (which is Ref. 2) was published as a review article in the special edition of the Journal of Molecular Liquids that was edited and prefaced by the first author of the review CK. This article summarized the main steps of Langer’s calculation of the relaxation rate [15,16] and clearly started the validity of the approach with respect to damping.

It is regretful that this big review does not provide a wider and more objective view of the work available in the literature on the dynamics of magnetic nanoclusters, for the benefit of a new comer to the field. It is also unfortunate that the authors have not provided a discussion of the huge amount of experimental work that shows the state-of-the-art understanding of the real situation about these systems.

[1] H. Kachkachi, Eur. Phys. Lett. 62, 650 (2003).
[2] H. Kachkachi, J. Mol. Liquids 114, 113 (2004).
[3] A. F. Franco, J.M. Martinez, J. L. Déjardin, H. Kachkachi, Phys. Rev. B 84, 134423 (2011).
[4] W. M. Saslow, J. Appl. Phys. 105, 07D315 (2009).
[5] M. C. Hickey and J. S. Moodera, Phys. Rev. Lett. 102, 137601 (2009).
[6] A.G. Gurevich and G.A. Melkov, Magnetization oscillations and waves (CSC Press, Florida, 1996).
[7] J. Stöer and H. C. Siegmann, Magnetism: from fundamentals to nanoscale dynamics (Springer, Berlin, 2006).
[8] M. Lakshmanan and K. Nakamura, Phys. Rev. Lett. 53, 2497 (1984).
[9] J.L. Garcia-Palacios, Adv. Chem. Phys. 112, 1 (2000).
[10] S. Titov, H. Kachkachi, Yu. Kalmykov, W.T. Coffey, Phys. Rev. B 72, 134425 (2005).
[11] D. A. Garanin, E. Kennedy, D. S. F. Crothers, and W. T. Coffey, Phys. Rev. E 60, 6499 (1999).
[12] W. Wernsdorfer, E. Boner Orozco, K. Hasselbach, A. Benoit, B. Barbara, N. Demoncy, A. Loiseau, H. Pascard, D. Mailly, Phys. Rev. Lett. 78, 1791 (1997).
[13] W. Wernsdorfer, K. Hasselbach, A. Benoit, B. Barbara, B. Doudin, J. Meier, J.-Ph. Ansermet, and D. Mailly, J. Mag. Mag. Mat. 55, 11552 (1997).
[14] M. Jamet, W. Wernsdorfer, C. Thirion, D. Mailly, V. Dupuis, P. Mélinon, and A. Pérez, Phys. Rev. Lett. 86, 4676 (2001).
[15] J.S. Langer, Phys. Rev. Lett. 21, 973 (1968).
[16] J.S. Langer, Ann. Phys. (N.Y.) 54, 258 (1969).
[17] We deem it our duty to inform the reader that the second author has already sent to Phys. Rev. B a comment on the article [3], Ref. 138 in CK. After we sent our reply and after one more round in the referral process of Phys. Rev. B, we decided that the various comments and replies be sent to a third referee. To the best of our knowledge, Kalmykov’s comment has not appeared in Phys. Rev. B.