Low-Temperature Features of Nano-Particle Dynamics

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March 23, 2022

Abstract

In view of better characterizing possible quantum effects in the dynamics of nanometric particles, we measure the effect on the relaxation of a slight heating cycle. The effect of the field amplitude is studied; its magnitude is chosen in order to induce the relaxation of large particles (∼7nm), even at very low temperatures (100mK). Below 1K, the results significantly depart from a simple thermal dynamics scenario.

Key words: nanoparticles, magnetic particles, magnetic relaxation, magnetization - quantum tunneling

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Submitted for the Proceedings of ICM97
The contribution of quantum tunneling events \[1\] to the reversal of the magnetic moment of nano-particles is commonly investigated by relaxation measurements \[2\]. However, the measured relaxation rate \(S\) (‘magnetic viscosity’, defined as the time-logarithmic derivative of the magnetization) is an intricate combination of the effective temperature \(T^\ast(T)\) (involved in the activated crossing of a barrier \(U\)) with the distribution of anisotropy barriers \(P(U)\). \(T^\ast(T)\) is the quantity of interest; it is equal to \(T\) for thermal dynamics, and should level off to some low-\(T\) plateau for quantum dynamics \[1\] (sketch in the inset of Fig.2). The knowledge of \(P(U)\) is highly uncertain for the smallest barriers which are explored at the lowest temperatures, where precisely one aims at extracting \(T^\ast(T)\).

Thermal dynamics is strongly accelerated by a temperature increase, whereas quantum processes should be much less affected; in this spirit, we have studied \[3\] the fraction of the relaxation rate at a given \(T\) which survives to a stage at a slightly higher temperature \(xT\). After the field change at \(T\) which triggers the relaxation \((t = 0)\), we heat the sample to \(xT\) during 200s \((1 \leq x \leq 3)\), and then measure the viscosity at \(T\) (see Fig.1). We normalize this viscosity to that measured without heating to \(xT\). In the obtained “Residual Memory Ratio” \((\text{RMR, [3]}), P(U)\) is (to first order) eliminated. More precisely, we have computed this ratio for some example distributions \(P(U)\) (which we consider temperature independent, as expected for independently relaxing particles). Fig.2 presents the results; remarkably, \(\text{RMR}(x)\) is nearly insensitive to extreme choices of \(P(U)\), while clearly picturing the thermal or quantum nature of the dynamics.

We have previously applied a similar measurement technique \[3\] to a sample of \(\gamma - Fe_2O_3\) particles diluted in a silica matrix with a \(4.3 \times 10^{-4}\) volume fraction \[4\]. The particle sizes correspond to a log-normal distribution of \(d_0 \sim 7nm\) and \(\sigma \sim 0.3\). The relaxations were induced by switching off a 62 Oe field in which the sample had been cooled from 6K. For \(0.06K \leq T \leq 1K\), the measured \(\text{RMR}(x)\) clearly departs from the thermal expectation (see details in \[3\]).

The main drawback of such measurements at fixed low field is that the relaxing objects at very low \(T\) are by far smaller than the typical size of \(7nm\), and cannot be clearly identified; one may wonder about the contribution of surface excitations in larger particles \[5\]. In order to measure quasi-macroscopic particles (close to the peak of the size distribution), we have performed measurements using higher fields. Due to the important flux drifts caused by this procedure, we study a less diluted sample of the same particles as before, with a \(3.4 \times 10^{-3}\) volume fraction; the dipolar fields \((H_{\text{dp}} = 4\pi M/V \sim \text{a few } Oe)\) remain small compared to the anisotropy field \(H_a \sim 1000Oe)\).

We prepare the sample at constant temperature in a negative field \(H_0 = -2000Oe\), in which almost all anisotropy barriers are overcome; then we sweep
this field to some value $H_1 (-100Oe < H_1 < 1500Oe)$. Small particles relax, but
the larger ones remain negatively oriented; we wait 1 hour, mainly for damping
the drift of the magnet. Then we increase the field from $H_1$ to $H_2$ (here we
keep $H_2 - H_1$ equal to $46Oe$). This last field increase triggers the relaxation of
a set of particles of the corresponding size; at several given temperatures $T$, we
have studied the relaxation rate $S(T, H_2)$ as a function of $H_2$. The results are
shown in Fig.3, where at each temperature a peak in the relaxation rate can be
seen, reflecting the distribution of the field-modulated barriers (see a discussion
of such $S(T, H)$ in [6]). At 100mK, we observe during our procedure a transitory
heating which is related to the large change in the sample magnetization. This
difficulty has already been noted in the very few existing experiments at high
field and very low $T$ [7], and related to avalanche processes. At 100 mK, the
effect is important for $H_2 > 500Oe$; the right-hand part of this curve is therefore
artificially depressed (Fig.3). At 300mK the effect is already much weaker [8].

Such a stray heating impedes a simple interpretation of viscosity measure-
ments, the distribution of objects which are about to relax being modified by the
initial temperature increase. However, if this initial state is reasonably repro-
ducible, it will not affect an RMR-type measurement, in which the initial distri-
bution is of very little influence (Fig.2). We have checked this point; our viscosity
values at 100mK remain reproducible within $\pm 20\%$, and within $\sim \pm 15 - 5\%$ at
higher temperatures. Besides, at 100 and 300mK, we have measured $RMR(x)$
for two different values of $H_2$; different sizes of particles are relaxing in each case,
but the $RMR(x)$ decrease with $x$ remains about the same. The $H_2$ choices at dif-
ferent temperatures (made for practical reasons [8]) are therefore of no significant
influence on our $RMR(x)$ discussion.

The results are displayed in Fig.4. From 1K and below, $RMR(x)$ clearly de-
parts from the simple thermal expectation, which is verified at 3K. As was the
case in our previous low-field results [3], no plateau of $RMR(x)$ is visible, sug-
gesting a more complex behavior than pictured for the quantum case in Fig.2.
If the $RMR$-anomalies are to be ascribed to quantum mechanics, then one must
think of a distribution of thermal-to-quantum crossover temperatures among the
particles. This may be due to a distribution of anisotropy energy densities; for
nanometric particles, the anisotropy arises from multiple origins (shape, sur-
face...). On the other hand, if the behavior is purely thermal, then the observed
anomalies imply that the very common description that we use here is not correct;
the $T$-independence of $U$ and $P(U)$ may be questioned, which means for instance
examining how far the particles are rigid macro-moments, or how independent of
each other they can be considered, even in highly diluted samples.
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Figure captions

Figure 1: Temperature and magnetic moment variations, recorded during a RMR procedure at 3K (dashed line: expectation for the quantum case [I]).

Figure 2: Calculated RMR(x), for very different P(U) choices, in the thermal (i) and quantum (ii) cases.

Figure 3: Measured viscosity as a function of the final field value H_2 (see procedure in the text), at different temperatures.

Figure 4: Measured RMR(x), at different temperatures (the negative values are compatible with zero within the error bars). The solid line is the expectation for a simple thermal dynamics scenario.
Fig. 1

$\Delta M \left(10^3 \, \text{e.m.u.}\right)$

$T = 3 \, \text{K}$

$x \cdot T \quad \left(x = 1.3\right)$

thermal

quantum

$\log_{10} \left( \text{time (s)} \right)$

Temperature (K)
Fig. 2
Fig. 3
Fig. 4

- T = 100 mK, $H_2 = 400$ Oe
- T = 300 mK, $H_2 = 500$ Oe
- T = 500 mK, $H_2 = 233$ Oe
- T = 1 K, $H_2 = 500$ Oe
- T = 3 K, $H_2 = 500$ Oe

- thermal dynamics