Magnetic relaxation of Co nanoclusters in a bias magnetic field

F Luis[1,4], F Petroff[2,3] and J Bartolomé[1]

[1] Instituto de Ciencia de Materiales de Aragón, CSIC-Universidad de Zaragoza, 50009 Zaragoza, Spain
[2] Unité Mixte de Physique CNRS/THALES, Domaine de Corbeville, 91404 Orsay Cedex, France
[3] Université Paris-Sud, 91405 Orsay Cedex, France

E-mail: fluis@posta.unizar.es

Abstract

We report ac magnetic susceptibility data measured on two samples of Co nano-particles, with average diameters $\langle D \rangle \simeq 1.4$ and 3.1 nm, respectively. The temperatures T_{max} of the maxima of the real χ' and imaginary χ'' components of the susceptibility shift in opposite directions when an increasing bias magnetic field H is applied. We show that the increase of T_{max} of χ' with H is caused by the non-linear field dependence of the equilibrium magnetization and that only χ'' provides reliable information about the magnetic relaxation mechanisms in a bias field. Our data show that the magnetization reversal takes place via a classical thermally activated relaxation process. The influence that inter-cluster dipolar interactions have on the relaxation time depends on H. At zero field, the magnetic relaxation is slower than in the limit of no interactions. By contrast, as the magnetic moments of the particles become gradually polarized by H, the relaxation approaches the theoretical predictions for non-interacting particles.

1. Introduction

The temperature-dependent dynamic susceptibility of a sample of magnetic nanoparticles contains information on the potential energy barriers U that separate different minima in the energy landscape of their magnetic moments [1]. This relation provides a method to measure fundamental magnitudes, such as the magnetic anisotropy [2], which are also of primary interest for the applications of these materials. In principle, the susceptibility can also be employed to investigate how a bias magnetic field modifies the relaxation time τ. An important question

---

[4] Author to whom any correspondence should be addressed.

here is how to experimentally determine the blocking temperature T_B, i.e., the temperature at which the typical relaxation time τ equals the experimental time τ_e, when $H \neq 0$. Often [3–5], T_B is taken from the temperature T_{max} of the maximum of the dc susceptibility measured after cooling the sample in zero field (ZFC). However, this interpretation neglects non-linear effects in the equilibrium susceptibility [6, 7]. Furthermore, very small particles do not always behave classically. Important quantum effects are expected at sufficiently low temperatures [8, 9], that can also modify the field dependence of τ.

It is therefore important to test the validity of classical models for magnetic relaxation and susceptibility in well characterized systems for which relevant parameters, such as the distribution of particle sizes $g(D)$ and the size-dependent anisotropy, are known independently, as is the case with the Co spheres that we study. In this paper, the following issues are addressed:

(a) we study the relation between T_B and T_{max} obtained from ac susceptibility experiments;
(b) we check whether or not very small clusters of a few hundred Co atoms flip their spins via a quantum tunnelling mechanism as smaller single-molecule magnets do; and
(c) we investigate how the influence that dipolar interactions have on the magnetic relaxation mechanism is modified by a magnetic field.

The paper is organized as follows. First, we give details of the sample preparation and sample parameters and of the magnetic measurement techniques. In section 3 we discuss, from a qualitative point of view, the competition between different effects that the application of an external magnetic field has on the equilibrium and dynamical magnetic susceptibility of nanoparticles. Finally, in section 4, we extract some quantitative information on the influence of interparticle dipolar interactions, and how this influence is modified by the bias. In order to do so, we study the deviation of the experimental results from theoretical predictions that are valid for independent nanoparticles. We apply this strategy to data measured on two samples of nanoparticles having different average sizes, for which we expect the strength of interactions to be also markedly different. The paper ends with the conclusions.

2. Experimental details

The Co clusters were prepared by sputter deposition of Co on a smooth alumina surface [10–12]. Oxidized Si was used as a substrate. The amount of Co that is deposited per layer is given by the thickness t_{Co} that the deposits would have if they were continuous. The average cluster diameter $\langle D \rangle$ increases with t_{Co}. A given sample is usually made by piling up a number N of Co/Al$_2$O$_3$ layers [12]. The clusters exhibit a well defined in-plane local order with constant inter-particle separations. The centre to centre distance inside each layer $\Lambda_\parallel \simeq D + 2$ nm. Multilayers show also a periodical vertical organization which resembles that of a close-packed hexagonal structure. For the present study, we have chosen a sample with $N = 40$ layers and $t_{Co} = 0.3$ nm that we hereafter label by I, and another with $N = 30$ layers and $t_{Co} = 0.7$ nm (sample II). Samples I and II have $\langle D \rangle = 1.4$ and 3.1 nm, respectively. The distributions $g(D)$ in these samples are approximately Gaussian and have widths $\sigma_D = 0.3$ and 0.2, respectively. The samples contain a fraction of paramagnetic atoms. Their concentration x_{para} was determined comparing the mass of Co which forms clusters, as observed in the TEM images, to the total amount of Co that was deposited. It was also estimated from magnetic measurements [2], with comparable results. For samples I and II, $x_{para} = 0.22$ and 0.25, respectively. The susceptibility of these paramagnetic spins is completely negligible with respect to that of the clusters in the field range of interest to the present work [2].

The ac susceptibility was measured by applying an oscillating magnetic field of small amplitude (4.5 Oe) to the sample and using the ac detection option of a commercial
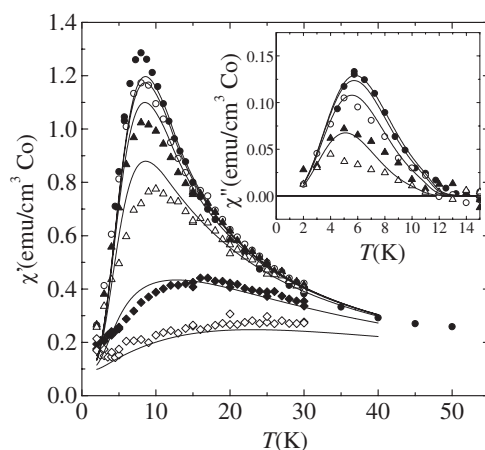
Figure 1. Real (main plot) and imaginary (inset) components of the ac susceptibility of sample I ($\langle D \rangle = 1.4$ nm) for $\omega/2\pi = 1$ Hz: $\bullet$, $\circ$, $\blacktriangle$, $\triangle$, $\blacklozenge$, and $\lozenge$ are for $H = 0$, 50, 100, 200, 500, and 1000 Oe, respectively. The full curves are theoretical predictions for non-interacting clusters (see the text).

SQUID magnetometer. The frequency $\omega/2\pi$ was varied between 1 and 90 Hz. The samples had a diamagnetic signal arising from the silicon substrate. This contribution was estimated by measuring a bare substrate of the same batch and found to be linear in field and independent of temperature. It was subtracted from all experimental data.

3. Ac susceptibility in a dc field: dynamical versus equilibrium effects

We show in figure 1 the ac susceptibility of sample I as a function of temperature. At zero field, the real χ' and the imaginary χ'' components of the susceptibility show frequency-dependent maxima that can be associated with the superparamagnetic blocking of the magnetic moments. It takes place when the relaxation time τ becomes of the order of $\tau_e = 1/\omega$. As H increases, the susceptibility decreases at all temperatures. This is expected, because the field polarizes the magnetic moments of the clusters (the slope of the magnetization curve is smaller at larger fields). A remarkable additional feature is that the maxima of χ' and χ'' shift in opposite directions. We denote the temperatures of these maxima as $T_{\max}(\chi')$ and $T_{\max}(\chi'')$. We observe that, while $T_{\max}(\chi'')$ decreases, the peak of χ' moves towards higher temperatures as H increases.

We argue next that these apparently contradictory behaviours can be qualitatively explained within a classical model of magnetic relaxation. In figure 2, we show data of sample II measured using two different H values and three frequencies. At low bias ($H = 50$ Oe), the maxima of both χ' and χ'' move towards higher temperatures as ω increases. These maxima therefore mark the freezing of the magnetic moments, i.e. $T_{\max} \simeq T_B$ as happens for $H = 0$. The situation becomes different for higher bias ($H = 300$ Oe, see figure 2, bottom panel): $T_{\max}(\chi')$ becomes almost independent of ω whereas the frequency-dependent part of the curve shifts towards lower temperatures, in the same way as the maximum of χ'' does. These results clearly show that the broad χ' peak merely reflects the temperature dependence of the *equilibrium* susceptibility and it is not related to the superparamagnetic blocking. A similar effect has been observed for the ZFC dc susceptibility [6, 7].
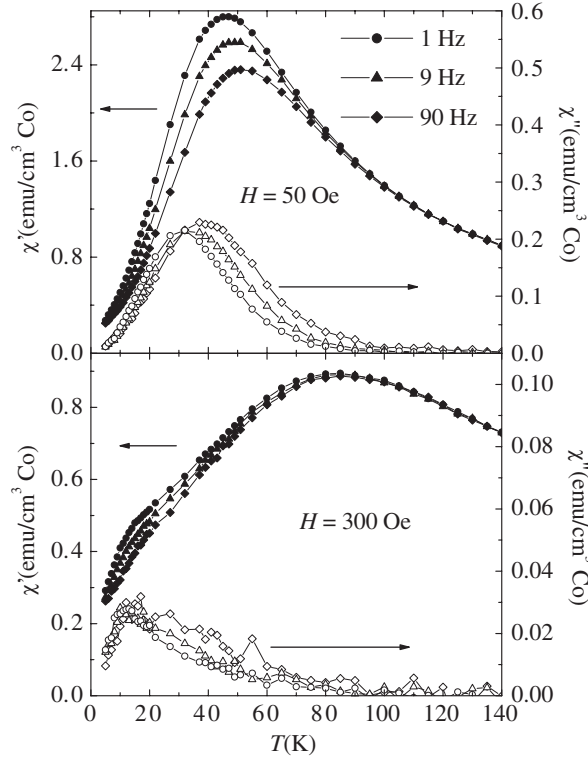
Figure 2. Ac susceptibility of sample II measured for two different bias fields and three different frequencies of the ac magnetic field. Solid and open symbols correspond to χ' and χ'' data, respectively.

The equilibrium susceptibility χ_T of each cluster can be obtained by performing the derivative of the equilibrium magnetization with respect to the field. Differentiating expression (3.38) of [13], which is valid for the high barrier limit (that is, for $\sigma \equiv U(H = 0)/k_B T \gg 1$), gives

$$\chi_T = \chi_{\sigma \to \infty} \left\{ 1 - \frac{1}{\sigma} \left[1 + \frac{x(1 - \cosh 2x)}{\sinh 2x} \right] \right\} + \frac{M_{sb}}{H_c(0)} \sin^2 \psi \qquad (1)$$

where $x = \mu H \cos \psi / k_B T$, μ is the magnetic moment of a cluster, ψ is the angle between the magnetic field and the anisotropy axis, $\chi_{\sigma \to \infty} = M_{sb} \mu \cos^2 \psi / k_B T \cosh^2 x$, and $M_{sb} = 6\mu/\pi D^3$. $\chi_{\sigma \to \infty}$ is the susceptibility of Ising spins (infinite anisotropy) and the other terms are corrections for finite anisotropy. For $H \neq 0$, χ_T has a maximum at a temperature $T_{eq} \simeq 2\mu H/k_B$, related to the saturation of the magnetic moments by H. When T_{eq} becomes larger than T_B the susceptibility peak reflects mainly the behaviour of χ_T. Under these conditions, $T_{max} \simeq T_{eq}$.

Therefore, only χ'' provides reliable information on the mechanism of magnetic relaxation in a bias field. In particular, the observed decrease of $T_{max}(\chi'')$ shows that the activation barrier U for the magnetization reversal becomes smaller as H increases. This is precisely the behaviour expected for classical relaxation [14–20] for which U can be accurately approximated [17, 18] by

$$U(H, \psi) = U(H = 0) \left[1 - H/H_c(\psi) \right]^{\eta(\psi)} . \qquad (2)$$

Here $H_c(\psi) = H_c(0) \left[\sin(\psi)^{2/3} + \cos(\psi)^{2/3} \right]^{-3/2}$, $H_c(0) = 2U(H = 0)/\mu$ is the anisotropy field, and the exponent $\eta(\psi) = 0.86 + 1.14 H_c(\psi)/H_c(0)$. The variation of U with H would have been very different if quantum tunnelling had taken place. Indeed, the field $H \cos(\psi)$ along the anisotropy axis splits the initially degenerate levels, and dramatically suppresses the probability of quantum tunnelling. This effect causes an increase of U and therefore of $T_{max}(\chi'')$ [9], just the opposite to the classical and also to the observed behaviours. It then follows from our data that the magnetization reversal takes place via thermal activation over the *classical* energy barrier.

In order to discuss the effect of interactions, which we shall do in the following section, it is useful to first know how T_{max} should vary with H in the limit of no interactions. In this limit, the susceptibility and therefore also $T_{max}(\chi')$ and $T_{max}(\chi'')$ can be calculated since the anisotropy and $g(D)$ are known for both samples [2]. We employed equation (1) to calculate χ_T and estimated the adiabatic susceptibility as $\chi_S \simeq M_{sb}/H_c(0) \sin^2 \psi$, independent of H. When $T \ll T_B$, this reversible response, which for randomly oriented anisotropy axes equals $2M_{sb}/3H_c(0)$, should be the only non-vanishing contribution to χ'. Indeed, the low-T experimental limit of χ' varies only weakly with H (see figure 1).

Finally, it is worth discussing in some detail the calculation of $\tau(H)$. The case of a longitudinal field ($\psi = 0$) was solved by Brown [14] for the limit of $\sigma \gg 1$, which is of applicability to our experimental conditions because $U/k_B T_B$ is of order 25 or more[5]. However, when the magnetic field is not parallel to the anisotropy axis, the problem loses its axial symmetry and it becomes very difficult to solve analytically [18, 21–23]. In addition, the relaxation time depends then in a nontrivial form on the damping parameter λ, describing the interaction of the cluster spin with the surrounding medium. Expressions for $\tau(H)$ have been obtained for low and high damping limits [24, 25] and even for the crossover regions between these limits [21, 22]. However, these expressions are difficult to implement for our experimental case, since the clusters' easy axes are oriented at random and we have to deal with a distribution of particle sizes. Therefore, in order to keep it sufficiently simple for our purposes, we decided to use equation (2) for U, which provides a good approximation of the numerically exact result [18]. We also introduced the effect that the longitudinal magnetic field has on the attempt time τ_0 using the same expression as found by Brown. The expression for τ that we used in our calculations then reads

$$\frac{1}{\tau} = \frac{1}{2\tau_0}(1 - h_\parallel^2) \exp\left(\frac{U}{k_B T}\right) \left[(1 - h_\parallel) + (1 + h_\parallel) \exp\left(\frac{-\xi}{k_B T}\right) \right], \quad (3)$$

where $h_\parallel = H \cos\psi/H_c(\psi = 0)$, and $\xi = 2\mu H \cos\psi$. We have set $U(H = 0) = K_s D^2$ with $K_s = 0.18$ and 0.31 erg cm^{-2} for samples I and II, respectively [2]. The values $\tau_0 = 2 \times 10^{-12}$ s for sample I and $\tau_0 = 10^{-13}$ s for sample II were determined from the dependence of $T_B(\chi'')$ on ω at zero field.

We believe that, despite the shortcomings mentioned above, equation (3) provides an adequate description of the field-dependent relaxation time of noninteracting nanoparticles. The reason is that the relaxation time, thus also $T_{max}(\chi'')$, is dominated by the field dependence of the activation energy U, whereas the influence of the pre-factor is much smaller. In fact, the measured frequency dependence of $T_{max}(\chi'')$ reveals that τ_0 is nearly independent of H, within the experimental uncertainties (i.e. its order of magnitude does not change with H). It should also be mentioned that different expressions of τ corresponding to the different damping regimes approach the axial limit at sufficiently low fields [21] and that, in our experiments, $H/H_c(0)$ is always smaller than 0.1.

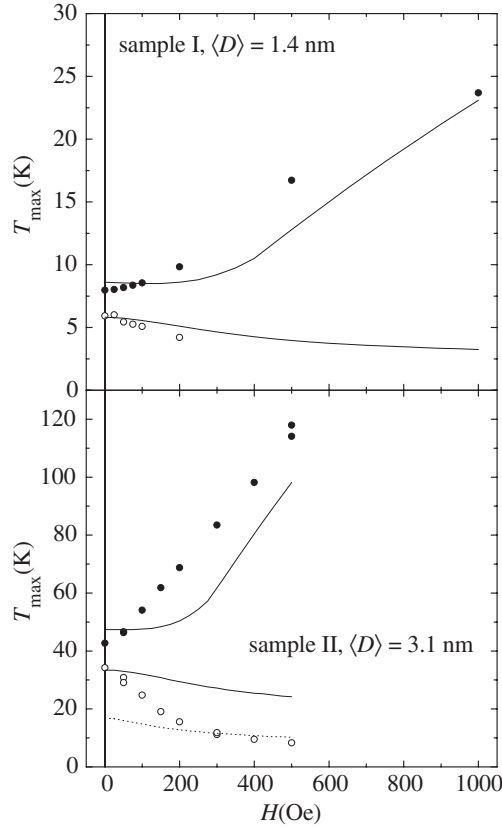[5] $U(H = 0) \simeq 87.1$ and 687 K for the average diameters of samples I and II, respectively.

Figure 3. Field dependence of $T_{\max}(\chi')$ (●) and of $T_{\max}(\chi'')$ (○) for the two samples investigated. The solid curves are extracted from calculations of the susceptibility performed with the model described in the text for non-interacting magnetic particles. The dotted curve in the lower panel was calculated by the same method, but fixing $U(H = 0)$ to half the value found for sample II at zero field.

4. Competition between dipolar interactions and the applied magnetic field

As shown in figures 1 and 3, the model of noninteracting particles describes qualitatively the overall trends: increase of $T_{\max}(\chi')$ and decrease of $T_{\max}(\chi'')$ as H increases. However, some important quantitative differences remain: the observed decrease of χ' and χ'' is larger than predicted, and also $T_{\max}(\chi'')$ decreases more rapidly than in the calculations. The discrepancy is more important for sample II, which contains the largest clusters. The deviation of the experimental $T_{\max}(\chi'')$ from the theoretical predictions is a direct indication that equation (3) overestimates τ at finite H. We have checked by varying K_s or $\langle D \rangle$ that it is impossible to simultaneously account for $T_{\max}(\chi'')$ measured at zero and finite bias.

Let us then discuss the effect of inter-particle dipolar interactions. It is important to mention here that interactions should be much stronger in the case of sample II. Nearest neighbours are located in adjacent layers at a distance $r_\perp \sim 0.7\Lambda_\parallel$ [12, 28]. The interaction energy is then of order $E_{\mathrm{dip}} = m^2/r_\perp^3$, where m is the particle's magnetic moment. For nanoparticles of average size in sample II ($\langle D \rangle = 3.1$ nm), this gives $E_{\mathrm{dip}} \sim 80$ K approximately, that is, about 12% of the total activation energy U. By contrast, for sample I we estimate $r_\perp \sim 2.4$ nm, thus

$E_{\text{dip}} = 2$ K only. This amounts to about 2% of the anisotropy energy. Therefore, these two samples provide a test case to investigate comparatively the effect of interactions.

In addition to the activation energy U_0 arising from the anisotropy, we then have to consider additional contributions U_{int} caused by dipolar interactions. It was found that $U_{\text{int}} > 0$ at zero field [2, 28]. Therefore $U(H = 0)$ and thus also $H_{\text{c}}(\psi = 0)$ are larger than for non-interacting particles. As shown in figure 3 (dotted curve in the lower panel), $T_{\text{max}}(\chi'')$ data measured for sample II above 200 Oe can be reproduced reasonably well by the model if we replace $U(H = 0, D = \langle D \rangle) = 687$ K, which applies near zero field, by $U(H = 0) \approx 345$ K. It is then tempting to ascribe the difference between these two activation energies to the effect of interactions. We obtain in this way $U_{\text{int}} \approx 342$ K for $D = \langle D \rangle$, i.e., about one-half of the total U, in rather good agreement with previous independent estimates [2, 28]. The magnetic field apparently induces a cross-over from a regime controlled by a large U_{int} to a regime in which interactions play only a minor role. A similar decrease of the role played by interactions has also been found for colloids of larger ($\langle D \rangle \simeq 5.3$ nm) and strongly interacting FeC nanoparticles [29].

Modelling magnetic relaxation in the presence of dipolar interactions is a complex many-body problem. Therefore, we content ourselves here with a qualitative discussion of interaction effects on the basis of a simple model [27, 28], which accounts for the variation of T_{B} with N at zero field. The susceptibility is dominated near T_{B} by the response of the largest particles of a sample[6]. When the magnetic moment of one of these particles flips, the surrounding magnetic moments rapidly [28] reorient themselves in order to minimize the energy of dipolar interaction with the central spin. As a result of 'carrying' the neighbouring spins, $U(H = 0)$ of the central spin becomes higher than in the non-interacting case, as observed. A magnetic field polarizes the neighbouring magnetic moments. It is then expected that the influence of interactions on T_{B} becomes relatively less important as H increases because the field decouples the largest particles from their neighbours. The cross-over found experimentally (see figure 3) is in agreement with this qualitative picture. In addition, this interpretation is supported by the fact that the typical value of the dipolar fields ≈ 300 Oe is of the same order of the bias field at which the cross-over is observed.

We shall illustrate these ideas with the following simple example (see figure 4) for which the effect of a magnetic field can be studied quantitatively. A central particle ($D = 3.55$ nm) with magnetic moment μ_1 has two smaller neighbours ($D = \langle D \rangle = 3.1$ nm) with magnetic moments $\mu_2 = \mu_3$ at a distance $r = 4.5$ nm. The anisotropy axes z of the three particles are parallel and we treat them as Ising spins. Replacing μ_{2z} and μ_{3z} by their thermal averages, the energy of μ_1 is

$$E_1 = -U_0 \cos \theta_1^2 - \mu_1 H \cos \theta_1 + E_{\text{dip}} \cos \theta_1 \tanh\left(\frac{-E_{\text{dip}} \cos \theta_1 + H \mu_2}{k_{\text{B}} T}\right)$$
$$- 2 E_{\text{dip}} \cos \theta_1 \tanh\left(\frac{2 E_{\text{dip}} \cos \theta_1 + H \mu_2}{k_{\text{B}} T}\right), \tag{4}$$

where $E_{\text{dip}} \equiv \mu_1 \mu_2 / r^3$, $\cos \theta_1 \equiv \mu_{1z} / \mu_1$, and $U_0 = 450$ K (sample II). For $H \mu_2 \lesssim E_{\text{dip}}$, the orientations of μ_2 and μ_3 are governed by the dipolar interaction with μ_1. In this regime U decreases with H as predicted by equation (2) but with $U(H = 0) = U_0 + 3 E_{\text{dip}}$ (dotted curve in figure 4). However, when $H \mu_2 \approx E_{\text{dip}}$ the increasing magnetic field gradually polarizes the 'slave' particles (2) and (3). This causes a crossover in the U versus H curve to a new regime. At even higher H, U again follows equation (2) but this time with $U(H = 0) \simeq U_0 - E_{\text{dip}}$ (dashed

---

[6] The largest contribution to χ is given by particles with volume approximately equal to $\int V^2 g(D) \, dD / \int V g(D) \, dD$. Using the size distribution of our samples, this expression gives a typical diameter $D \simeq 1.81$ nm for sample I and $D \simeq 3.55$ nm for sample II. Most particles (about 85% in sample I and 77% in sample II) are smaller than these.
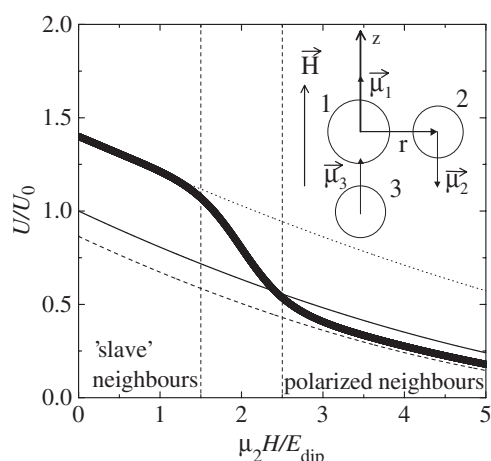
Figure 4. Field dependence of the activation barrier calculated under the approximations described in the text for the system shown in the inset. Thin full curves stand for the isolated cluster; thick curves stand for the interacting case. The dotted and dashed lines are calculated with equation (2) using $U(H = 0) = U_0 + 3E_{dip}$ and $U(H = 0) = U_0 - E_{dip}$, respectively. The parameters are $D_1 = 3.55$ nm, $D_2 = 3.1$ nm, $r = 4.5$ nm, and $U_0 = 450$ K, as appropriate for sample II.

line in figure 4). This behaviour is in qualitative agreement with what we observed for T_{max} (see figure 3). Therefore, we suggest that the 'freezing' of the fluctuations of superparamagnetic spins by the bias field may be responsible for the gradual decrease of the effect of interactions.

5. Conclusions

Summarizing, we have shown that the ac susceptibility provides an appropriate tool to investigate separately equilibrium and non-equilibrium phenomena. In particular, direct and reliable information on the mechanism of magnetic relaxation can be extracted from the imaginary part of the susceptibility. Our data show that magnetic relaxation of very small Co clusters proceeds classically. We have also observed that dipolar interactions cannot be neglected, and that they slow down the relaxation process at zero field. Nevertheless, in external magnetic fields stronger than the typical internal dipolar fields the magnetic relaxation approaches the relaxation of non-interacting particles.

Acknowledgments

References

[1] Shliomis M I and Stepanov V I 1994 *Adv. Chem. Phys.* **87** 1
[2] Torres J M *et al* 2002 *J. Magn. Magn. Mater.* **242–245** 575
     Luis F *et al* 2002 *Phys. Rev.* B **65** 094409
[3] Luo W, Nagel S R, Rosenbaum T F and Rosensweig R E 1991 *Phys. Rev. Lett.* **67** 2721
[4] El-Hilo M, O'Grady K and Chantrell R W 1992 *J. Magn. Magn. Mater.* **114** 307
[5] Tejada J, Zhang X X, del Barco E, Hernández J M and Chudnovsky E M 1997 *Phys. Rev. Lett.* **79** 1754

[6] Hanson M, Johansson C and Mørup S 1995 *J. Phys.: Condens. Matter* **7** 9263
    Hanson M, Johansson C and Mørup S 1998 *Phys. Rev. Lett.* **81** 735
[7] Kachkachi H, Coffey W T, Crothers D S F, Ezzir A, Kennedy E C, Noguès M and Tronc E 2000 *J. Phys.: Condens. Matter* **12** 3077
[8] Friedman J R, Sarachik M P, Tejada J and Ziolo R 1996 *Phys. Rev. Lett.* **76** 3830
    Hernández J M, Zhang X X, Luis F, Bartolomé J, Tejada J and Ziolo R 1996 *Europhys. Lett.* **35** 301
    Thomas L, Lionti F, Ballou R, Gatteschi D, Sessoli R and Barbara B 1996 *Nature* **383** 145
[9] Luis F, Bartolomé J and Fernández J F 1998 *Phys. Rev.* B **57** 505
    Garanin D A and Chudnovsky E M 1997 *Phys. Rev.* B **56** 11102
    Leuenberger M N and Loss D 1999 *Europhys. Lett.* **46** 692
[10] Maurice J L, Briático J, Carrey J, Petroff F, Schelp L F and Vaurès A 1999 *Phil. Mag.* A **79** 2921
[11] Briático J, Maurice J L, Carrey J, Imhoff D, Petroff F and Vaurès A 1999 *Eur. Phys. J.* D **9** 517
[12] Babonneau D, Petroff F, Maurice J L, Fettar F and Vaurès A 2000 *Appl. Phys. Lett.* **76** 2892
[13] García-Palacios J L 2000 *Adv. Chem. Phys.* **112** 1
[14] Brown W F Jr 1963 *Phys. Rev.* **130** 1677
[15] Aharoni A 1969 *Phys. Rev.* **177** 793
[16] Victora R H 1989 *Phys. Rev. Lett.* **63** 457
[17] Pfeiffer H 1990 *Phys. Status Solidi* a **122** 377
[18] Coffey W T, Crothers D S F, Kalmykov Y P and Waldron J T 1995 *Phys. Rev.* B **51** 15947
    Coffey W T, Crothers D S F, Dormann J L, Geoghegan L J, Kalmykov Y P, Waldron J T and Wickstead A W 1995 *Phys. Rev.* B **52** 15951
[19] Garanin D A 1996 *Phys. Rev.* E **54** 3250
[20] Coffey W T, Crothers D S F, Dormann J L, Kalmykov Y P, Kennedy E C and Wernsdorfer W 1998 *Phys. Rev. Lett.* **80** 5655
[21] Garanin D A, Kennedy E C, Crothers D S F and Coffey W T 1999 *Phys. Rev.* E **60** 6499
[22] Déjardin P M, Crothers D S F, Coffey W T and McCarthy D J 2001 *Phys. Rev.* E **63** 021102
[23] Coffey W T, Crothers D S F, Kalmykov Yu P and Titov S V 2001 *Phys. Rev.* B **64** 012411
[24] Brown W F 1979 *IEEE Trans. Magn.* **15** 1197
[25] Geoghegan L J, Coffey W T and Mulligan B 1997 *Adv. Chem. Phys.* **100** 475
[26] Shtrikman S and Wohlfarth E P 1981 *Phys. Lett.* A **85** 457
    Hansen M F and Mørup S 1998 *J. Magn. Magn. Mater.* **184** 262
    Dormann J L, Fiorani D and Tronc E 1999 *J. Magn. Magn. Mater.* **202** 251
[27] Jonsson T, Nordblad P and Svedlindh P 1998 *Phys. Rev.* B **57** 497
[28] Luis F, Petroff F, Torres J M, García L M, Bartolomé J, Carrey J and Vaurès A 2002 *Phys. Rev. Lett.* **88** 217205
    See also the comment by Hansen M F and Mørup S 2003 *Phys. Rev. Lett.* **90** 059705
    and the author's reply, Luis F, Petroff F, Torres J M, García L M, Bartolomé J, Carrey J and Vaurès A 2003 *Phys. Rev. Lett.* **90** 059706
[29] Jönsson P E, Felton S, Svedlindh P, Norblad P and Hansen M F 2001 *Phys. Rev.* B **64** 212402