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## **Relaxation time of weakly interacting superparamagnets**

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**Abstract.** – The relaxation time of weakly interacting classical spins is calculated by introducing the averages of the local dipolar field, obtained by thermodynamic perturbation theory, in a rigorous expression for the single-spin thermoactivation rate in a weak but arbitrarily oriented field. At low temperatures the non-trivial dependence of the superparamagnetic blocking on the damping coefficient, numerically found by Berkov and Gorn, is reproduced by our model and interpreted in terms of the deviations from uniaxial anisotropy associated to the transversal component of the dipolar field acting on each spin.

Introduction. – The study of single-domain magnetic particles has been an active field of research since the pioneer work of Stoner and Wohlfarth [1], who studied the hysteretic rotation of the magnetisation over the magnetic-anisotropy energy barrier under the influence ofapplied fields, and N´eel [2] who predicted that at non-zero temperature the magnetisation can surmount the energy barrier as a result of thermal agitation. Important progress has been made since Brown [3] derived the Fokker-Planck equation for the probability distribution of spin orientations, starting from the stochastic Landau-Lifshitz equation, and calculated the relaxation time for uniaxial particles in a longitudinal field. Recent work on spins with non-axially symmetric potentials revealed  $[4]$  a large dependence of the relaxation time on the damping coefficient  $\lambda$  in the medium-to-weak damping regime ( $\lambda$  measures the relative importance of the precession and the damping in the dynamics). Experiments on individual nanoparticles  $[5]$  analysed with accurate asymptotes of the relaxation time  $[6]$ , gave damping coefficients in that regime:  $\lambda \approx 0.05{\text -}0.5$ .

Turning our attention from independent particles to systems of interacting particles, the complexity of the problem increases drastically, as it becomes a many-body problem with a long-ranged and reduced symmetry interaction mechanism—the dipolar interaction. The approach to study the relaxation time  $\tau$  has so far been based on how the energy barriers of the spins are modified by the interactions [7,8]. With the  $\tau$  so obtained, one can study the effects of the interaction on the superparamagnetic blocking (the maximum in the dynamical response at the temperature where  $\tau$  becomes of the order of the observation time). However, the barrierbased approach to determine  $\tau$  corresponds to assuming  $\lambda \to \infty$ , so that dynamical features such as the precession of the spins are disregarded, as incisively noted by Berkov and Gorn [9]. Indeed, numerical integration of the stochastic Landau-Lifshitz equation for weakly interacting systems revealed [9] non-trivial effects of finite damping on the superparamagnetic blocking, such as enhanced shifts of the temperature of the maximum of the dynamical response and non-monotonic behaviour of its height with the interaction strength.

In this article we apply thermodynamic perturbation theory (treating the anisotropy energy exactly and the dipolar energy perturbatively) to calculate the averages of the dipolar field produced at the position of a given spin. These averages are then introduced in a rigorous low-field expansion of the single-spin relaxation rate, which can be combined with a recently derived perturbative formula for the equilibrium susceptibility [10] to provide a model for the dynamical susceptibility of weakly interacting spins. In the low-temperature range, where the superparamagnetic blocking takes place, our model recreates the non-trivial damping dependence of the blocking found in ref. [9]. The analytical treatment permits us to ascribe the observed features to the deviations from uniaxial symmetry of the potential due to the transversal components of the dipolar field, deviations which render the relaxation time very sensitive to the damping [4].

Low-field relaxation rate of uncoupled spins. – In weak fields, we can handle the field dependence of the relaxation time  $\tau$  by expanding the relaxation rate  $\Gamma = 1/\tau$  in powers of the field components (for convenience we use the field in temperature units  $\vec{\xi} = m \vec{B}/k_{\rm B}T$ , where  $m$  is the magnetic moment). As the spins have inversion symmetry in the absence of the field, the relaxation rate will not change if the field is reversed (since  $\Gamma$  accounts for jumps over the energy barrier in both directions). Therefore,  $\Gamma$  should be an even function of  $\xi$ , and for spins with uniaxial anisotropy we have to third order

$$
\Gamma \simeq \Gamma_0 \Big( 1 + c_{\parallel} \xi_{\parallel}^2 + c_{\perp} \xi_{\perp}^2 \Big) , \qquad (1)
$$

where  $\Gamma_0$  is the zero-field relaxation rate, and  $\xi_{\parallel}$  and  $\xi_{\perp}$  are the longitudinal and transversal components of the field with respect to the anisotropy axis (the vanishing of the term  $\xi_{\parallel}\xi_{\perp}$ follows from the invariance of the relaxation rate upon field reflection through the barrier plane). The coefficients  $c_{\parallel}$  and  $c_{\perp}$  will be determined by choosing special configurations in which they are known (strictly longitudinal and transversal fields).

We shall restrict our attention to low temperatures where the superparamagnetic blocking takes place. Expanding the expression for  $\Gamma$  in the presence of a longitudinal field  $\xi_{\parallel}$  [3, 11], we find

$$
\Gamma(\xi_{\parallel},\xi_{\perp}=0)\simeq\Gamma_0\Big(1+\tfrac{1}{2}\xi_{\parallel}^2\Big)\;,\qquad\Gamma_0=\frac{1}{\tau_{\rm D}}\frac{2}{\sqrt{\pi}}\sigma^{3/2}e^{-\sigma}\;,
$$

where  $\tau_{\rm D}$  ( $\propto 1/\lambda$ ) is the relaxation time of isotropic spins,  $\sigma = A/k_BT$  is the anisotropy barrier in temperature units, and corrections of order  $1/\sigma$  are disregarded due to the low- $T$  assumption. Comparison with the general expansion  $(1)$  gives the longitudinal coefficient  $c_{\parallel} = 1/2$ . Note that in this case the damping parameter  $\lambda$  only enters through  $\tau_D$  and hence it only matters to establish a global time scale. In other words, the results for different  $\lambda$ presented in units of  $\tau_D$  show complete dynamical scaling, and in this sense the  $\lambda$ -dependence is said to be trivial.

There is no general expression for the relaxation time in the presence of a non-zero transversal field valid for all values of the relevant parameters. Nevertheless, Garanin *et al.* [4] have derived a low-temperature formula valid for weak transversal fields, which is perfectly suited for our purpose of determining  $c_{\perp}$ , namely

$$
\Gamma(\xi_{\parallel} = 0, \xi_{\perp}) \simeq \Gamma_0 \left[ 1 + \frac{1}{4} F(\alpha) \xi_{\perp}^2 \right], \qquad F(\alpha) = 1 + 2(2\alpha^2 e)^{1/(2\alpha^2)} \gamma \left( 1 + \frac{1}{2\alpha^2}, \frac{1}{2\alpha^2} \right).
$$

Here  $\alpha = \lambda \sigma^{1/2}$  and  $\gamma(a, z) = \int_0^z dt t^{a-1} e^{-t}$  is the incomplete gamma-function. Comparing with the expansion (1), one gets the transversal coefficient  $c_{\perp} = F/4$ .

On gathering these results we finally get the desired expression for the low-temperature relaxation rate in weak fields

$$
\Gamma \simeq \Gamma_0 \left[ 1 + \frac{1}{2} \xi_{\parallel}^2 + \frac{1}{4} F(\alpha) \xi_{\perp}^2 \right], \qquad (2)
$$

which constitutes a straightforward generalisation of the formula of Garanin  $et \ al.$  [4] to an arbitrary field orientation. The function  $F$  takes into account without further approximations the effects of the precession. F decreases towards 1 for strong damping (where  $c_{\parallel}$  and  $c_{\perp}$  are of the same order of magnitude), while F grows as  $1/\lambda$  for weak damping, where the relaxation time turns to be very sensitive to the damping (or to transversal fields).

Let us compare the rigorous eq.  $(2)$  with the corresponding result of Mørup and co-workers (see, for instance, eq. (6) in ref. [8]), which for  $1/\sigma \ll 1$  can be written as

$$
\Gamma_{\text{Mørup}} \simeq \Gamma_0 \Big( 1 - \xi_{\parallel} + \frac{1}{2} \xi_{\parallel}^2 + \frac{1}{4} \xi_{\perp}^2 \Big) .
$$

This expression has the undesirable feature of a term linear in the field, which can be attributed to accounting for the escape from one of the potential wells only. Actually, if we average over both wells the linear term cancels out and the corrected formula equals the overdamped  $\lambda \to \infty$ limit of eq. (2) (then  $F \to 1$ ). This is natural, since in ref. [8] the effects of the field were only considered via barrier changes, so that no gyromagnetic effects were included, and their result can only be correct when the precession can be neglected ( $\lambda \to \infty$ ). Besides, as their linear term disappeared upon random anisotropy averaging, their results will correspond to the limits overdamped *plus* random anisotropy of those derived here from eq.  $(2)$ .

Averages of the dipolar field.  $-$  Let us consider a system of magnetoanisotropic spins coupled via the dipole-dipole interaction. The dipolar field at the position  $\vec{r}_i$  of the spin  $\vec{s}_i$ created by all other spins is given by

$$
\vec{B}_i = \frac{\mu_0 m}{4\pi a^3} \sum_j \mathbf{G}_{ij} \cdot \vec{s}_j , \qquad \mathbf{G}_{ij} = \frac{1}{r_{ij}^3} \left( 3 \,\vec{v}_{ij} \,\vec{v}_{ij} - \mathbf{I} \right) , \tag{3}
$$

where the term with  $j = i$  is omitted, m is the magnitude of the magnetic moment, and in the dipolar tensor  $\vec{r}_{ij} = \vec{r}_i - \vec{r}_j$  and  $\vec{v}_{ij} = \vec{r}_{ij}/r_{ij}$ . The action of a tensor dyadic  $\vec{A}\vec{B}$  on a vector  $\vec{C}$  is the usual one,  $(\vec{A}\vec{B}) \cdot \vec{C} \equiv \vec{A}(\vec{B} \cdot \vec{C})$ , and hence the tensor  $G_{ij}$ , when multiplied by  $\vec{s}_j$ , gives the field at the position of the *i*-th dipole created by  $\vec{s}_j$ . All lengths are measured in units of the characteristic length a, which is defined in such a way that  $a<sup>3</sup>$  is the mean volume occupied around each spin. In a simple cubic arrangement  $a$  is the lattice constant and for nanoparticles of volume V the volume concentration of particles is  $V/a<sup>3</sup>$ . For notational simplicity we are assuming that the parameters characterising the different spins are identical (it is immediate to generalise the expressions for different anisotropy constants, magnetic moments, volumes, etc.).

For spins with uniaxial anisotropy the total energy of the system can be written as

$$
-\beta E = \sigma \sum_{i} (\vec{s}_i \cdot \vec{n}_i)^2 + \xi_d \sum_{i>j} \vec{s}_i \cdot \mathbf{G}_{ij} \cdot \vec{s}_j , \qquad (4)
$$

where  $\beta = 1/k_B T$ ,  $\vec{n_i}$  is the unit vector along the anisotropy axis of the *i*-th spin,  $\sigma$  is the anisotropy barrier divided by the thermal energy, and

$$
\xi_{\rm d} = \frac{\mu_0}{4\pi a^3} \frac{m^2}{k_{\rm B}T} \,, \qquad h_{\rm d} = \frac{\xi_{\rm d}}{2\sigma} \,. \tag{5}
$$

The quantity  $h_d$  is a convenient temperature-independent measure of the interaction strength  $(\propto$  concentration) equal to the magnitude of the field, measured in units of the maximum anisotropy field  $B_K = 2A/m$ , produced at a given position by a spin located at a distance a.

Using thermodynamic perturbation theory (ref. [12], § 32) to expand the Boltzmann distribution  $W = Z^{-1} \exp[-\beta E]$  in powers of  $\xi_d$ , one gets an expression of the form

$$
W = W_{a} [1 + \xi_{d} w_{1} + \cdots] . \tag{6}
$$

Here  $W_a = \prod_i Z_a^{-1} \exp[-\beta E_a(\vec{s}_i)]$  is the Boltzmann distribution of the non-interacting ensemble (in our case  $E_a$  includes the magnetic anisotropy) and  $w_1$  is linear in the dipolar energy (and hence quadratic in the spins). The calculation of the observables reduces to computing averages weighted by the non-interacting probability distribution  $\langle \cdots \rangle_{\rm a}$  =  $Z_{\rm a}^{-1} \int \frac{\mathrm{d}^2 \vec{s}}{2\pi} \exp[-\beta E_{\rm a}] (\cdots)$  of low grade powers of the spin variables. In this way, the results are exact in the magnetic anisotropy and only perturbational in the dipolar interaction.

The averages of terms linear and quadratic in  $\vec{s}$ , weighted by the non-interacting distribution, can be calculated by means of the following "algorithms" [10]:

$$
\langle (\vec{s} \cdot \vec{v}_1) \rangle_a = 0, \qquad \langle (\vec{s} \cdot \vec{v}_1)(\vec{s} \cdot \vec{v}_2) \rangle_a = \frac{1}{3} (1 - S_2) \ \vec{v}_1 \cdot \vec{v}_2 + S_2 (\vec{n} \cdot \vec{v}_1)(\vec{n} \cdot \vec{v}_2) , \qquad (7)
$$

where  $\vec{v}_1$  and  $\vec{v}_2$  are arbitrary constant vectors. The first average vanishes (actually any odd power) since the magnetic anisotropy has inversion symmetry  $[E_{a}(-\vec{s}_{i})] = E_{a}(\vec{s}_{i})$  and there is no external bias field. The quantity  $S_2$  is the average of the second-order Legendre polynomial  $S_2 = \langle \frac{1}{2} [3(\vec{s} \cdot \vec{n})^2 - 1] \rangle_a$ , and it can be written in terms of the one-spin partition function  $Z_a$ , as

$$
S_2(\sigma) = \frac{3}{2} \left( \frac{e^{\sigma}}{\sigma Z_a} - \frac{1}{2\sigma} \right) - \frac{1}{2} , \qquad Z_a = \sqrt{\pi/\sigma} \operatorname{erf}(i\sqrt{\sigma}) . \tag{8}
$$

In the equations for the relaxation rate the field enters squared (eq. (2)). Therefore, we do not calculate the statistical mechanical average of  $\vec{B}_i$  (eq. (3)) and plug it into Γ (which would be a sort of mean-field approach) but we average instead the combinations of field variables as they enter in the expression for  $\Gamma$ . Since  $B_{\perp}^2 = B^2 - B_{\parallel}^2$ , we can average the square of the field  $(\xi_i = m B_i / k_\text{B} T)$ 

$$
\left\langle \xi_i^2 \right\rangle = \xi_{\rm d}^2 \sum_{j,k} \left\langle \vec{s}_j \cdot \bm{G}_{ij} \cdot \bm{G}_{ik} \cdot \vec{s}_k \right\rangle \simeq \xi_{\rm d}^2 \sum_j \left\langle \vec{s}_j \cdot \bm{G}_{ij} \cdot \vec{s}_j \right\rangle_{\rm a},
$$

and the square of the projection along the local anisotropy axis

$$
\left\langle \xi_{i,\parallel}^2 \right\rangle = \xi_{\rm d}^2 \sum_{j,k} \left\langle (\vec{n}_i \cdot \mathbf{G}_{ij} \cdot \vec{s}_j)(\vec{n}_i \cdot \mathbf{G}_{ik} \cdot \vec{s}_k) \right\rangle \simeq \xi_{\rm d}^2 \sum_j \left\langle (\vec{n}_i \cdot \mathbf{G}_{ij} \cdot \vec{s}_j)^2 \right\rangle_{\rm a}.
$$

The field averages are considered to order  $\xi_d^2$  (so we just need eq. (6) to zero order;  $w_1$  only enters in the third-order corrections) and to get the last equalities we have used  $\langle \vec{s}_j \cdot \mathbf{T} \cdot \vec{s}_k \rangle_a = 0$ , if  $j \neq k$  (since then, when integrating over  $\vec{s}_j$ , we can use the first eq. (7) with  $T \cdot \vec{s}_k = \vec{v}_1$ ). Therefore, by means of the algorithms (7), we get

$$
\langle \xi_{i,\parallel}^2 \rangle = \frac{\xi_{\rm d}^2}{3} \sum_j \left[ \left( 1 - S_2 \right) \left( \vec{n}_i \cdot \boldsymbol{G}_{ij} \cdot \boldsymbol{G}_{ij} \cdot \vec{n}_i \right) + 3S_2 \left( \vec{n}_i \cdot \boldsymbol{G}_{ij} \cdot \vec{n}_j \right)^2 \right],\tag{9}
$$

$$
\langle \xi_{i,\perp}^2 \rangle = \frac{\xi_{\rm d}^2}{3} \sum_j \left[ 6r_{ij}^{-6} + 3S_2 r_{ij}^{-3} (\vec{n}_j \cdot \mathbf{G}_{ij} \cdot \vec{n}_j) - (1 - S_2) (\vec{n}_i \cdot \mathbf{G}_{ij} \cdot \vec{n}_i) - 3S_2 (\vec{n}_i \cdot \mathbf{G}_{ij} \cdot \vec{n}_j)^2 \right]. \tag{10}
$$

Note that the averaged fields depend on T via  $S_2(\sigma)$ , reflecting the fact that the field created at a given position can be different if, for example, the source spins are almost freely rotating (high T,  $S_2 \to 0$ ) or are almost parallel to their anisotropy axes (low T,  $S_2 \to 1$ ).

One may wonder about the validity of these field averages below the superparamagnetic blocking, where the spins are not in complete equilibrium. However, since at those temperatures the spins are still in quasi-equilibrium confined to one ofthe two wells, we can repeat the derivation of the algorithms (7) restricting the phase space for integration to one well. In this case, averages of the form  $\langle \vec{s} \cdot \vec{v}_1 \rangle_a$  do not vanish, and should be considered together with  $\langle (\vec{s} \cdot \vec{v_1})(\vec{s} \cdot \vec{v_2}) \rangle_a$ , which being even in  $\vec{s}$  is not modified. The extra terms associated with  $\langle \vec{s} \cdot \vec{v_1} \rangle_a$ , however, vanish if the overall state is demagnetised, and we recover eqs. (9), (10). This can be interpreted as iftwo nearby blocked spins, one in its upper well and the other in the lower, create a net field at the position of a third spin similar to that created by any of them when in equilibrium, since each of the spins approximately compensates for the restricted phase space of the other.

The general expressions for the longitudinal and transversal fields are notably simplified in some important situations. For a system with parallel anisotropy axes  $(e.g.,)$  in a single crystal of magnetic molecular clusters, or ferrofluids frozen in a strong magnetic field) we equate all the  $\vec{n}_j$  to  $\vec{n}$ . For a system with randomly distributed anisotropy axes we replace expressions involving  $f(\vec{n}_j)$  by integrals  $\int d^2\vec{n} f(\vec{n}) \equiv \overline{f}$ , and use  $\overline{(\vec{n} \cdot \vec{v}_1)(\vec{n} \cdot \vec{v}_2)} = \frac{1}{3}\vec{v}_1 \cdot \vec{v}_2$ . In both cases the final expressions involve some rapidly convergent sums over the lattice. Let us concentrate on the cases of "sufficiently isotropic" lattices, in the sense of fulfilling  $\sum (r_x)^k = \sum (r_y)^k = \sum (r_z)^k$ , e.g., cubic and completely random lattices (incidentally, the type of arrangements for which in the classical Lorentz cavity field calculation the contribution of the dipoles inside the "small" sphere vanishes). Similarly we consider large enough systems, so that all spins have approximately equivalent surroundings (then the index  $i$  on the different quantities can be dropped). Under these circumstances the "lattice sums" involved are

$$
\mathcal{R} = 2 \sum_{j} r_{ij}^{-6}, \qquad \mathcal{T} = \sum_{j} (\vec{n} \cdot \mathbf{G}_{ij} \cdot \vec{n})^2 , \qquad (11)
$$

where the terms with  $j = i$  are of course omitted.

For a system with aligned anisotropy axes the averaged fields are given in terms of the lattice sums (11) by the compact expressions

$$
\left\langle \xi_{\parallel}^{2} \right\rangle = \frac{\xi_{\rm d}^{2}}{3} \left[ (1 - S_{2}) \mathcal{R} + 3S_{2} \mathcal{T} \right], \qquad \left\langle \xi_{\perp}^{2} \right\rangle = \frac{\xi_{\rm d}^{2}}{3} \left[ (2 + S_{2}) \mathcal{R} - 3S_{2} \mathcal{T} \right], \tag{12}
$$

while for randomly distributed anisotropy axes they read

$$
\overline{\langle \xi_{\parallel}^2 \rangle} = \frac{\xi_{\rm d}^2}{3} \mathcal{R}, \qquad \overline{\langle \xi_{\perp}^2 \rangle} = \frac{\xi_{\rm d}^2}{3} 2 \mathcal{R} \ . \tag{13}
$$

Superparamagnetic blocking.  $-$  The dependence of the features of the superparamagnetic blocking on the interaction strength  $h_d = \xi_d/2\sigma$  has been a subject of some controversy. Two main approaches  $[7, 8]$  addressed this problem on the basis of the modifications of the energy barriers by the interactions. However, this type ofapproach overlooks the fact that not only the energy landscape is important, but also how the spin evolves in it, depending on whether the spin is precessing almost freely (weak damping) or strongly damped. Indeed, Berkov and Gorn [9] have shown with rigorous Langevin dynamics simulations that for weak interactions the position of the blocking temperature  $T_b$  (where  $\chi''$  reaches its maximum) of strongly damped spins is hardly affected by the interaction strength, whereas for weak damping  $T<sub>b</sub>$  significantly



Fig. 1 – Imaginary component of the dynamical susceptibility vs. temperature (the real component is shown in the inset) for a spherical sample and spins placed in a simple cubic lattice. The anisotropy axes are all parallel and the response probed along their common direction. The dipolar interaction strength  $h_d = \xi_d/2\sigma$  is  $h_d = 0$  (thick lines), 0.004, 0.008, 0.012, and 0.016 from (a) top to bottom and (b) right to left. The frequency is  $\omega \tau_{\rm D}/\sigma = 2\pi \times 0.003$ .

decreases with  $h_d$  and the peak height behaves non-monotonically. The apparent discrepancy with some experimental results, in which  $T<sub>b</sub>$  increases with  $h<sub>d</sub>$ , was attributed in ref. [9] to the different behaviour of systems with weak anisotropy (or moderate-to-high interactions), where the energy barriers are mostly due to the interactions and hence grow with  $h_d$ .

In order to assess the features of the superparamagnetic blocking emerging from our model, we make use of a rigorous perturbative expansion of the equilibrium susceptibility  $\chi_{eq}$  in powers of  $\xi_d$  (to second order, see ref. [10]). This expression, together with the relaxation rate  $Γ$  obtained when the averaged fields  $(12)$ ,  $(13)$  are introduced in eq.  $(2)$ , can be combined in a simple Debye-type formula

$$
\chi = \chi_{\text{eq}} \frac{\Gamma}{\Gamma + i \omega} \ .
$$

Naturally, at low enough temperatures [10] the results will become invalid by the very nature of the approximations involved, although this does not affect the characteristics of the superparamagnetic blocking for weak interactions.

The dynamical response for a large spherical sample with parallel anisotropy axes and simple cubic lattice structure is shown in fig. 1 (the lattice sums required are  $\mathcal{R} = 16.8$  and  $\mathcal{T} = 13.4$ ). In the overdamped case,  $T<sub>b</sub>$  is not noticeably affected by the dipolar interaction and the height of the susceptibility peak decreases monotonically with  $h_d$ . This corresponds to the effect of a slight decrease of  $T<sub>b</sub>$  found in ref. [8] for random anisotropy, which is observable only at very high frequencies (e.g., Mössbauer). For weak damping, however, fig. 1 shows that the blocking temperature significantly decreases as the interaction strength increases and, in addition, the peak height of  $\chi''$  initially rises for small values of  $h_d$  and then decreases for larger values. The same behaviour can be seen in  $\chi'$  with the appropriate choice of parameters.

The features shown in fig. 1 are in complete agreement with the results obtained by Berkov and Gorn [9]. The analytical treatment employed here, however, allows us to readily trace back the origin of the results obtained and to interpret them in terms of the different  $h_d$ -dependences of Γ and  $\chi_{\text{eq}}$ . For overdamped systems the coefficients  $c_{\parallel} = 1/2$  and  $c_{\perp} = F/4 \simeq 1/4$  in the expression (2) for the relaxation rate are of order unity and lead to a slight increase of  $\Gamma$  (decrease of  $T_{\rm b}$ ) with  $h_{\rm d}$ , while the entire curve is lowered by the reduction of  $\chi_{\rm eq}$  with  $h_{\rm d}$ . For weak damping, however,  $F \propto 1/\lambda$  is large and makes Γ very sensitive to the interaction strength, moving the peak quickly towards low T as soon as  $h_d$  departs from zero. Then, as the coefficients in  $\chi_{\text{eq}}$  are not as large as  $c_{\perp}$ , the initial decrease of the equilibrium susceptibility can be smaller than the increase associated to the quick shift of the blocking to low T, where  $\chi_{eq}$  is higher (roughly  $\propto 1/T$ ), producing the rise of the peak. If  $h_d$  is further increased the decrease of  $\chi_{\text{eq}}$  starts to be competitive and the peaks are reduced as they shift to lower temperatures.

The different behaviours are related to the presence of transversal components of the local fields, which create a saddle point in the uniaxial potential barrier of the spins, turning the relaxation rate sensitive to  $\lambda$  [4]. We can picture the underlying physical mechanism as follows [13]. Consider one spin that after a "favourable" sequence of fluctuations, reaches a point close to the top of the barrier but does not surmount it. In the subsequent spiralling down back to the bottom of the potential well, a strongly damped spin descends almost straightly, whereas a weakly damped spin executes several rotations ( $\sim 1/\lambda$ ) about the anisotropy axis. This allows the latter spin to pass close to the saddle area, where it will have additional opportunities, not available for the damped spin, to cross the barrier, enhancing the relaxation rate. As we see, this mechanism duly combines the characteristics of the potential and the dynamical evolution ofthe spins in the potential. Note finally that the transversal components are non-zero even for parallel anisotropy axes  $(eq. (12))$ , so there is no need, as in the case of non-interacting particles, to appeal to oblique fields, applied  $[4]$  or probing  $[13]$ , to find a large sensitivity to the damping in interacting systems.

Summary and conclusions. – We have proposed a model for the relaxation time of weakly interacting superparamagnets. The single-spin relaxation time at low fields of Garanin  $et$   $al$ . is generalised for an arbitrary directed field, and the components of the local field calculated by thermodynamical perturbation theory. The non-monotonic behaviour of the height of the dynamical susceptibility peak with the interaction strength and the enhanced lowering of the blocking temperature for weak damping, discovered numerically by Berkov and Gorn, are captured by our model. These features are interpreted in terms of the different sensitivity, depending on the damping strength, of the relaxation rate to transversal fields, which for interacting spins are provided by the dipolar interaction.

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