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Susceptibility resonance and magnetic viscosity

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It is suggested that the phenomenon of superparamagnetism in small ferromagnetic particles may manifest itself as a resonance in the magnetic susceptibility, measured at a frequency which corresponds to the average relaxation time τ of these particles. It is argued that this resonance has not been observed because it is difficult to know in advance at which frequency it should be looked for, and Néel's theory of this τ is inadequate for estimating the location of such a resonance. However, an appropriate analysis of the measurements of magnetic viscosity *in the same sample* can give a very good idea of the value of the resonance frequency and an estimation of its line width.

I. INTRODUCTION

Rather large ferromagnets are known to be subdivided into domains, with a complicated magnetization structure. But a sufficiently small ferromagnetic particle is a "single domain,"[1] which can always be considered (at least approximately) to be magnetized to saturation. When such a particle is magnetized in some direction, it remains that way, and its magnetization does not change its direction, because of an energy barrier. This barrier is due to the anisotropy energy, and as such is proportional to the particle's volume. Therefore, for a small volume the barrier can be small enough for thermal fluctuations to flip the direction of the magnetization *back and forth.* When the relaxation time, namely, the time it takes to flip the magnetization, is much shorter than the time it takes to do the experiment, the magnetization reverses many times while being measured. Therefore, the average of the magnetization vanishes in zero applied field, and on the whole, the particles behave as if they were huge paramagnetic *atoms:* There is no hysteresis, but there is saturation when the magnetizations of all the particles are aligned. The theoretical treatments of paramagnetism fit this case without any change, except for the *quantitative* difference that the spin number S is of the order of, say, 10^4, whereas the more conventional paramagnets have S of the order of 1. This transition from stable ferromagnetism to paramagnetism at a small particle size has been observed[2] in many systems. The phenomenon is known as superparamagnetism. Its existence was suggested theoretically by Néel[3] before any of these experiments were done.

There are also many experiments in the literature in which the relaxation time τ for the flipping of the magnetization is of the same order as the time it takes to do the measurement. However, the magnetization in these experiments is not allowed to flip *back and forth.* They all follow the original thought experiment of Néel[3] in which an ensemble of small particles is put in a very large field, then taken out of the field. The remanent magnetization is then observed to decay in time, as the thermal fluctuations keep changing the directions of the particles' magnetization. Because of this particular way of measurement, the phenomenon was given the name "magnetic aftereffect," or "magnetic viscosity."

It is also possible in principle to measure the particles of the magnetic-viscosity region in an *alternating* magnetic field. As will be discussed in the next section, such a measurement can result in a resonance similar to the one observed by Awschalom *et al.*[4] at very low temperatures. In searching for such a resonance it should help to know *a priori* where to look for it, but the theory cannot provide an adequate estimate, as will be shown in Sec. III. It is, therefore, suggested in Sec. IV to start the experiment by the conventional measurement of the magnetic viscosity in the same sample. If properly analyzed, this measurement can yield all the necessary information.

II. SUSCEPTIBILITY RESONANCE

Awschalom *et al.*[4] have recently observed resonance peaks in the magnetic susceptibility versus frequency plots of small, elongated particles of carbonyl iron. The peaks were fully developed at a temperature of about 70 mK, and disappeared above about 200 mK, so that there can be no doubt that they originate from a relaxation of the magnetization by a quantum tunneling mechanism, and not by thermal fluctuations. However, the mathematical form of magnetization versus time in quantum tunneling is identical[5] to that of magnetic viscosity controlled by thermal agitation. Therefore, the same peaks must also be observed for the latter case, in an appropriate range of temperature and frequency.

The important physical feature is that when a particle has a natural frequency ν for switching the direction of its magnetization, an alternating magnetic field applied at that frequency will be more effective in switching the magnetization back and forth than a field applied at other frequencies. A resonance peak should, therefore, appear at that frequency, or integral multiples of it, and there can be no doubt that this resonance is the source of the peaks observed by Awschalom *et al.*,[4] or that a similar peak can be found at higher temperatures, when the switching is due to thermal fluctuations.

When the sample contains many particles, with different natural frequencies, this resonance will obviously widen, and may even be completely smeared out, if the distribution of the relevant properties of the particles is too wide. Awschalom *et al.*[4] checked by scanning electron microscopy (SEM) that their particles had a very narrow size distribution, and the same must be required from any sample used in the magnetic viscosity region. However, such narrow distributions *have* been made and studied in that region. For example, the size distribution of the Fe particles of Yatsuya *et al.*[6] was so narrow that all their measurements of $M(H/T)$ superimposed into a single, pure Langevin function.

Besides this broadening of the resonance line due to the different relaxation times (or frequencies) for the different particles, there should also be a natural linewidth due to the statistical nature of the relaxation time in each of the particles. The latter would in principle be observed even if the measurement could be done on an isolated, single particle. The point is that the relaxation time of each particle is an average value for many cycles of the applied field. It may vary between one cycle and the next, because the magnetization vector may[7] perform a random walk around each of the energy minima before jumping to the other minimum.

This natural linewidth is not easy to estimate, and one should always carry in mind that it may in principle suppress the whole resonance in extreme cases. There is, however, one indication that this problem is not very important in most practical cases. The relaxation times obtained from the theory of Brown,[7] which takes this random walk into account, are rather close to the ones obtained from Néel's theory,[3] which assumes just one jump, at least for[8] an energy barrier that is much larger than k_BT. For *uniaxial* anisotropy, the difference between these two theories is actually quite small[9] even when this barrier is comparable to k_BT, so that the linewidth cannot possibly be so large as to make the resonance unobservable.

For a cubic anisotropy, the possibilities of a random walk in all sorts of directions are more intricate and more complicated than for a uniaxial symmetry, and it makes a large difference. Except for the case of a very large energy barrier, the relaxation time computed[10] from Brown's differential equation for the random walk in a cubic anisotropy was considerably different[11] from that in Eq. (1), obtained from the simple approach of Néel. Moreover, Krop *et al.*[12] measured the relaxation times for different sizes of some cubic particles, at different temperatures, and demonstrated that the difference between these theories was *measurable*. In their experiments, the numerical solution of Brown's differential equation fitted the experimental data, while the Néel equation did not. However, even for this case the theoretical difference does not seem to be so large that it can smear out the resonance in the susceptibility altogether. Therefore, one should try to find this resonance even in cubic materials, but it makes more sense, of course, to try first to look for it in a *uniaxial* material.

A broadening due to the effect of a random walk will naturally be larger at higher frequencies. Therefore, higher harmonics, at integral multiples of the fundamental resonance frequency, should exist in principle but may be too wide to be observed. Awschalom *et al.*[4] did not see peaks at the higher harmonics, but it is possible that these were just too wide in their particular sample, and it should be interesting to look for such peaks in other samples. It should also be interesting to try the more conventional experiment of magnetic viscosity at the same temperature and with the same sample of Awschalom *et al.*[4], and see if it leads to the same relaxation time as the resonance frequency, which the present discussion implies.

Interaction between the particles may also smear out this resonance, and indeed Awschalom *et al.*[4] report that their resonance was suppressed when the interparticle distance was reduced. This result means that one should be careful in choosing the sample, but there have also been magnetic viscosity measurements on particles that were very well separated. For example, the particles of Yatsuya *et al.*[6] were dispersed in oil so that there was hardly any interaction between them. Some of the older measurements[2] used precipitation out of dilute alloys, and these "particles" are certainly sufficiently well separated to avoid interaction. Noninteracting, spherical particles have also been prepared[13] by trapping them in pores.

Susceptibility resonance thus seems feasible and detectable in at least some of the samples on which magnetic viscosity measurements can be found in the literature. But in searching for such a resonance it should always be helpful to know at which frequency to look for it, in a given sample. An obvious first approach is to estimate this frequency theoretically, as discussed in the next section.

III. THEORETICAL ESTIMATES

The theory of Néel[3] estimated the relaxation time τ of an isolated particle by using the (classical) probability of jumping over an energy barrier. It led to the famous relation

$$\frac{1}{\tau} = f_0\, e^{-\alpha}, \qquad \alpha = \frac{KV}{k_BT}, \tag{1}$$

where K is the anisotropy constant, V is the particle's volume, k_B is Boltzmann's constant, and T is the absolute temperature. The pre-exponential factor f_0 is a constant, which Néel[3] estimated to be of the order of 10^9 s^{-1}. In more recent years it has become more customary to take this constant as 10^{10} s^{-1}, or larger, which probably results from the use of the bulk anisotropy constant in Eq. (1), while small particles have actually a different value of K. Equation (1) is for a uniaxial anisotropy, but can also be adapted, with a slight modification,[2] to the case of a cubic anisotropy.

Brown[7] argued that this oversimplified theory of Néel considered only a single jump from one energy minimum to the other, and did not allow the magnetization vector to spend some of the time in between these two energy minima, before jumping. He also noticed that Néel had ignored the probability of a jump back to the original minimum. In his first attempt to take these effects roughly into account[7], Brown came up with the same relation as in Eq. (1) here, only with

$$f_0 = \frac{2K\gamma_0}{M_s}\sqrt{\frac{\alpha}{\pi}}, \tag{2}$$

for a *uniaxial* anisotropy, where γ_0 is the gyromagnetic ratio and M_s is the saturation magnetization.

In a later, more elaborate study[8] Brown derived a differential equation to describe the *Brownian motion* of the direction of magnetization vector during its random walk from one energy minimum to the other. The smallest eigenvalue λ of this differential equation, is related to the relaxation time τ through the equation

$$\lambda = \frac{V}{k_B T\tau\eta}\left(\frac{1}{\gamma_0^2} + \eta^2 M_s^2\right), \tag{3}$$

where all the symbols have been defined in the foregoing, except for the dissipation constant η. Brown mentioned that the latter parameter should in principle be obtained from the experimental linewidth of the ferromagnetic resonance of the material, but took it for simplicity to be the value which minimizes λ of Eq. (3), namely,

$$\eta = \gamma_0^{-1} M_s^{-1}. \tag{4}$$

Bessais *et al.*[14] objected to this value, on several grounds which will be discussed in the following.

Brown did not solve that equation, but he did realize that it should lead to Eq. (2) in the limit of a very large energy barrier, namely, for $\alpha \gg 1$. In terms of λ, this asymptotic formula is

$$\lambda = 4\pi^{-1/2}\alpha^{3/2}e^{-\alpha}. \tag{5}$$

Numerical solution of Brown's differential equation for a uniaxial material in zero applied field[9] showed this relation to be quite a good approximation even down to $\alpha = 1$. For smaller values of α, results were given[9] in terms of a power series in α, with the coefficients specified explicitly up to that of α^7, so that $\lambda(\alpha)$ could be conveniently evaluated for any value of α.

More recently, Bessais *et al.*[14] repeated the numerical computation for the uniaxial case, by what they claim to be an easier method. They found that their numerical values could be approximated, to a very high accuracy, by the empirical relation

$$\lambda = 2\left(1 + \frac{\alpha}{4}\right)^{5/2} e^{-\alpha}. \tag{6}$$

This relation does not quite fit the power-series expression[9] for small α, neither does it fit the asymptotic expression of Brown, Eq. (5), for very large α. Bessais *et al.*[14] consider the latter misfit as an advantage, because their expression in Eq. (6) leads to a better temperature dependence of the dissipation rate than Brown's relation in Eq. (5). However, unlike their relation in Eq. (6), which is an arbitrary curve fitting, Eq. (5) is based on an analytic proof for the asymptotic form of the solution of Brown's differential equation. There is no way a numerical solution of that equation can lead, asymptotically, to any other functional form. If a different temperature dependence seems necessary for any reason, it can only be achieved by correcting the differential equation, or by modifying the temperature dependence of the constants of that equation. No other way is possible in principle, and a numerical similarity may be only fortuitous. At least the closeness of the empirical formula Eq. (6) to the numerical computation results of Bessais *et al.*[14] does not seem very conclusive to this particular author. The ratio of the two asymptotic expressions is

$$\rho = \frac{\lambda_{\text{of Eq. (5)}}}{\lambda_{\text{of Eq. (6)}}} = \frac{2\alpha^{3/2}}{\sqrt{\pi}\,(1+\alpha/4)^{5/2}}. \tag{7}$$

The value of this ratio is, for example, 1.56 for $\alpha = 10$ or 0.6 for $\alpha = 50$. Such a ratio may be adequate for most applications (where a factor of 2 is usually good enough), but may not suffice for some cases.

Nevertheless, the idea of Bessais *et al.*[14] to have a single empirical expression that can fit both the large and the small α regions, is a very sound one. Presenting all the numerical results in one equation is certainly nicer and more elegant than the way used in Ref. 9, and can be very helpful for the evaluation of λ in practical cases. It is, therefore, suggested to replace Eq. (6) by

$$\lambda = 2\left(\frac{2 + 9\alpha/5 + (4/\pi)^{1/3}\alpha^2}{2+\alpha}\right)^{3/2} e^{-\alpha}. \tag{8}$$

This equation tends to the *correct* power of $\alpha^{3/2}$ in the limit $\alpha \gg 1$, even with the *correct* numerical coefficient. Also, the power-series expansion, for small α, of Eq. (8) starts with

$$\lambda = 2\left(1 - \frac{2\alpha}{5}\right), \tag{9}$$

which is the same beginning as in Ref. 9. If this expansion is extended to the next term, the coefficient of $(2\alpha/5)^2$ in the present approximation is 2.96, which is much larger than the value of 0.343 of the exact[9] series expansion, but such a difference does not contribute very much to the actual values of λ. For the intermediate value $\alpha = 1$ the rigorous computation[9] gives $\lambda = 1.306$, for which $\lambda = 1.528$ from Eq. (8) is a sufficiently good approximation. Equation (8) should thus be a convenient approximation for all practical cases of a uniaxial anisotropy. It should be particularly useful for computations of the magnetization processes in particles, as, e.g., in Ref. 15. So far, such computations, which are otherwise long and tedious, just insert the crude Néel approximation of Eq. (1) when it comes to evaluating the switching probability. Therefore, these computations can significantly benefit from the use of Eq. (8).

It is thus possible to find the theoretical value of λ for a given α, either from Eq. (8) if that approximation is adequate, or from the numerical integration if a higher accuracy is needed. However, obtaining the value of τ from such an evaluation of λ is not straightforward, because the physical parameters in Eq. (3) are usually known to an insufficient accuracy. The volume V of the particle, which appears in the definition of α in Eq. (1), is particularly difficult to estimate, even when the particles are very regular, which is not always the case. Thus, e.g., Awschalom *et al.*[4] mention that their ellipsoidal particles had "a very narrow size distribution, but accurate determination of the smallest dimensions are limited to $\sim 10\%$."

The difficulty is that 10% of the radius can make a very large uncertainty in τ, at least in some regions. To demonstrate it, the following values have been computed, using for simplicity Eq. (1) with $f_0 = 10^9$ s^{-1}, and using the known anisotropy constant for bulk cobalt, with $T =$ 300 K: For a sphere of Co with a radius $R = 36$ Å, the relaxation time is $\tau = 0.1$ s, and for $R = 44$ Å it is $\tau = 6 \times 10^5$ s. Obviously, the theoretical estimate is completely useless in this range of particle size of cobalt, and a similar range exits for *any* material.

For ellipsoids the uncertainty may be even larger than it is for spheres, because only two of the three dimensions can be seen in electron micrographs, and because of the contribution of the shape anisotropy to K. Certain tricks of tilting the sample[16] can give some information of the third dimension, but it is not adequate. And for particles like those studied in Ref. 16 neither the size nor the shape anisotropy can be defined to any satisfactory approximation. Moreover, even for spherical particles without any shape anisotropy, the value of K is often very poorly known, especially for small particles.

On top of all these problems, Bessais *et al.*[14] have shown that the value of η deduced from some experimental linewidths of the ferromagnetic resonance can differ by orders of magnitude from the value of η in Eq. (4). They also argued that the temperature dependence of this parameter should be different from that of the saturation magnetization in Brown's simplified relation of Eq. (4). This extra complication makes it even more difficult to have a reliable theoretical value for τ in all but some very simple cases, at least until the study of this subject is much more advanced than it is now.

IV. MAGNETIC VISCOSITY

A. Logarithmic function

Since the theory has been shown in the previous section to be highly unreliable for finding the relaxation time τ of a given system, one must rely on a direct measurement of this τ. The most natural way for such a measurement is already contained in the original thought experiment of Néel[3] in which an ensemble of small particles is put in a very large field, then taken out of the field, and the remanent magnetization is observed to decay in time. Néel[3] argued that most decays are usually exponential to a first order, so that if t is the time, one should observe

$$M_r(t) = M_r(0)e^{-t/\tau}. \tag{10}$$

Fitting experimental data of remanent magnetization versus time should, thus, yield the value of the relaxation time τ of the particular system.

The relation in Eq. (10) has been widely used in *other* fields of physics, but *not* for analyzing experimental data of the time decay of magnetization. It is normally argued that because of particle-size distribution in the measured sample, there must be a distribution in τ, and the time dependence cannot possibly be a simple exponential. In analyzing other experiments, the use of an exponential time dependence is justified[17] by saying that τ is the average over a certain range. But for the decay of the remanent magnetization, Eq. (10) is not even tried in cases in which the size distribution is so narrow that the use of that equation may have been appropriate. Instead, it has become customary to analyze the experimental data in terms of

$$M_r(t) = C - S \ln(t/t_0), \tag{11}$$

where C, S, and t_0 are constants. Actually, most workers omit t_0 by absorbing it into C, which makes the whole relation dimensionally wrong, because the logarithm is defined only for a dimensionless number.

The functional form of Eq. (11) originates from an old theory[18] based on rough approximations, and never meant to be more than a quick, order-of-magnitude kind of an estimate. It assumes that the distribution function of the energy barriers is a *constant,* with two sharp cutoffs at energies E_1 and E_2, and that the measurement is done at a time which is much larger than $\tau(E_1)$, and much smaller than $\tau(E_2)$. Obviously, such assumptions can only be a crude approximation to any real physical system, but somehow this form of Eq. (11) was adopted by almost everybody. It even reached an extreme case[19] when the whole $M_r(t)$ curve was not measured. The resulting conclusions about the time decay of M_r are based on the remanent magnetization as measured at only two points: 100 s and 1000 s after switching off the magnetic field.

Even in less extreme cases, measurements of the $M_r(t)$ curve are only too often done over one decade of the time, t, and for one decade there is no advantage over the use of Eq. (11), because Eq. (10) may[20] fit just as well. In order to demonstrate this point, Fig. 1 plots the logarithmic and exponential functions, from which it can be seen that they are not significantly different, in this range of the argument. There is no way to decide that a set of experimental data, with just a little noise, fits one of these curves and not the other, and the only difference is that fitting the data to Eq. (10) determines the value of the (average) τ, while this important information is lost when Eq. (11) is used.

It should be particularly emphasized that there is nothing significant in the values of A and S in Fig. 1, which are only curve-fitting parameters, without any physical meaning. If 500 points are used, the least-squares fitting of the two curves in Fig. 1 leads to $A = 0.363\,13$ and $S = 0.325\,82$, so that any theory that involves Eq. (11) and the value of S can at most be a rough approximation to some physical situation, which is what can be expected anyway from the derivation[18] of Eq. (11).

It should also be emphasized that the time t in Fig. 1 is scaled by the factor τ, which is completely arbitrary in this kind of a plot, and can only be defined for a specific physical system. The figure may be describing a time

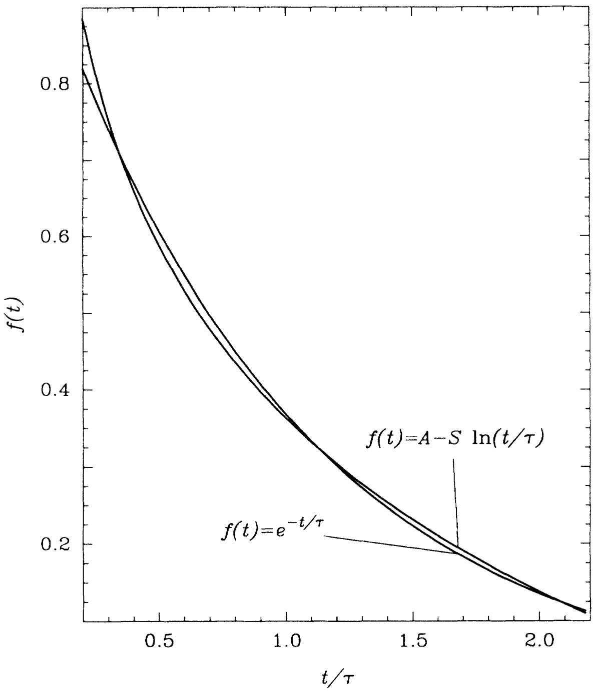

FIG. 1. Comparing the logarithmic decay, Eq. (11) with the exponential decay, Eq. (10). The former is plotted with $A = 0.362\,77$ and $S = 0.324\,46$, obtained from least-squares fitting of 100 equidistanced points on the curves.

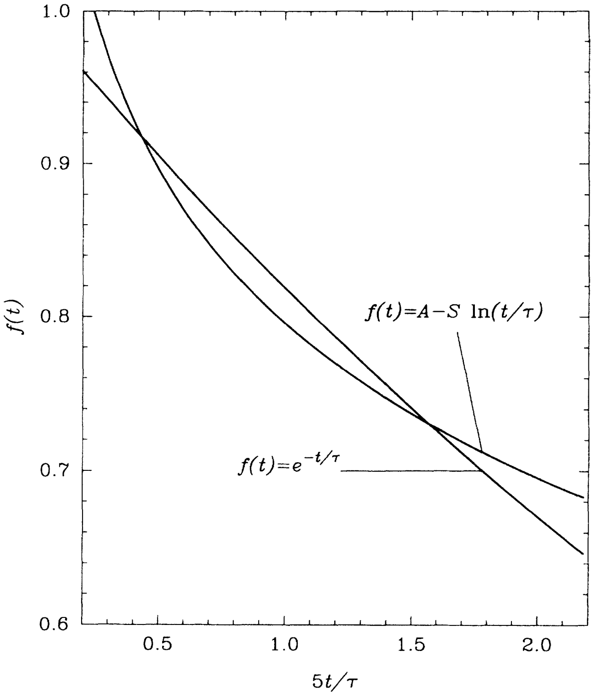

FIG. 2. Same as Fig. 1, for a factor of 5 increase in the argument of the functions. The logarithmic decay, Eq. (11), is plotted with $A = 0.563\,09$ and $S = 0.144\,59$, obtained from least-squares fitting of 100 equidistanced points on the curves.

dependence of the remanent magnetization between 0.2 and 2.2 μs, or that many years, or centuries, or any other units of time. When the time is measured in seconds, the value of 1000 s is sometimes defined[19,21] to be a "long time" after the removal of the magnetic field. But there is nothing special about the second as a unit, and the use of other units of time will make the numerical value of the same time much larger or much smaller than its numerical value in seconds.

Actually, the systems reported in Ref. 21 were shown in Ref. 20 to have a time scale around 10^4 s or larger, so that in these experiments, the time of 10^3 s is a very *short* time. The same must also be true for all published experiments, because a "long time" after removing the sample from the applied field means that the *exponent* has changed by at least an order of magnitude during that time, so that the magnetization has changed by *many* orders of magnitude. It is not easy to design an experimental system that has a sufficient dynamic range to measure many decades of magnetization, so that all the published experimental data must have been taken a very *short* time after removing the magnetic field, and it does not matter if the data were taken after hours or after days.

Nevertheless, the equivalence of the logarithmic and the exponential functions does depend on the particular range of the *argument* of these functions, which is also related to the time scaling, only in an indirect way. To demonstrate this point, the best fit of the same two functions is plotted in Fig. 2, for a *larger* argument, and it can be seen that the agreement is much worse than in Fig. 1. Inasmuch as a different argument can be absorbed into a different value of τ, the curves can always be brought into the better agreement of Fig. 1. However, in a real physical set of data, the value of $M_r(0)$ in Eq. (10) is essentially determined by the beginning of the set, and τ is determined by the slope. Equation (10) needs a third parameter in order to bring it to the range of Fig. 1, which is, thus, not always possible. From a physical point of view, a simple exponential would not fit the data if there is some distribution of relaxation times. Therefore, the extra parameter is obviously the width of the distribution function, which cannot be determined by the special form of Eq. (11), even if a superficial look at the data seems to make them fit to this equation. Moreover, even if the fit to Eq. (10) is as bad as in the example of Fig. 2, there is more physical information in plotting the data in this way than in a better fit to Eq. (11). It gives at least some approximate idea of what the average relaxation time is, which is better than no information at all.

B. Logarithmic scale

One way of adding the third parameter is to use[22]

$$M_r(t) = M_0 + M_1 e^{-t/\tau_1} + M_2 e^{-t/\tau_2}, \tag{12}$$

for fitting the data. Details of the fit were not given, only that the fitting to *one* exponential was[22] "significantly poorer." The range of the measuring time was not specified either, but could not have been much more than one decade. Charap[23] has already remarked that "experiments of this sort will benefit from extension to longer times." In principle, two exponential functions cannot be a very good approximation to a whole distribution of exponentials, but the fit must be better than that of one exponential, although it is not actually necessary to have all the parameters of Eq. (12). Only *one* extra parameter is needed in Eq. (10) over one decade of the time.

The recent data of Liu *et al.*[24] are chosen to illustrate this point. The data they give in their Fig. 1 fit quite nicely a straight line on a logarithmic scale, starting at $t \approx 0.45$ min. They will, therefore, be simulated here by 46 equidistant values in t, starting at $t = 0.45$ min, for which

$$M(t) = 10.667 - 3.366 \ln t. \tag{13}$$

Forty-five of these forty-six points were fitted by least squares to Eq. (10), leaving the point at $t = 0.45$ min out of the fitting procedure, which yielded the parameters $M_r(0) = 13.1880 \times 10^{-3}$ T, and $\tau = 4.8757$ min. The points taken from Eq. (13), and those fitted to Eq. (10), are plotted in Fig. 3 as the dashed and the solid line, respectively. The same points were also fitted to

$$M(t) = M_0 + M_1 e^{-t/\tau}, \tag{14}$$

for which the best fit to the "data" was for $M_0 = 4.720\,78 \times 10^{-3}$ T, $M_1 = 10.3881 \times 10^{-3}$ T, and $\tau = 1.892\,54$ min. In this fitting, all 46 points were used, from $t = 0.45$, to $t = 5.05$ min. The values fitted in this way to Eq. (14) are plotted as circles in Fig. 3 and it can be seen that the agreement between these points and the straight, solid line is very good indeed.

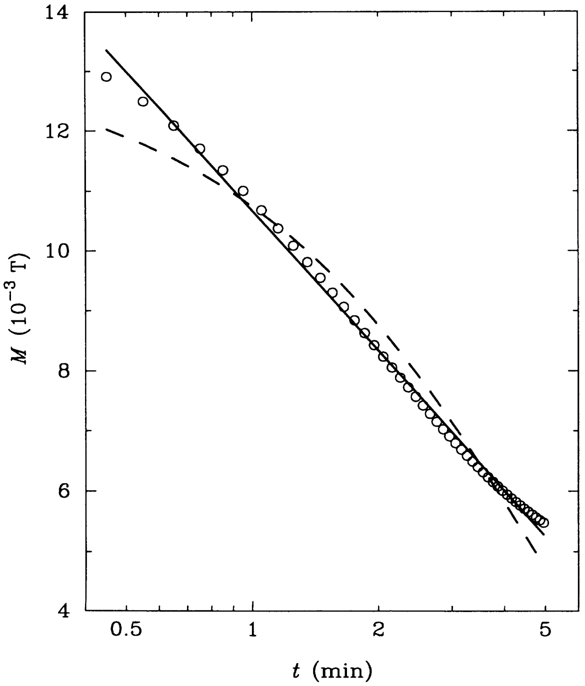

FIG. 3. The data of Ref. 24 plotted in different ways. The solid line is a plot of Eq. (13), the dashed curve is the fitting of points on the solid line to Eq. (10), and the circles are a fitting to Eq. (14).

Part of the agreement is actually psychological. Most observers have a tendency to fit points to a straight line, which may also be the reason for the popularity of using Eq. (11). To begin with, there is no reason whatsoever to plot any *one-decade* data on a logarithmic scale, but once it is done, it is natural to look for a fit of the type of Eq. (11). When shown the plot in Fig. 3, everybody seems to agree that the solid line is a very good approximation for the "experimental" points. However, when the *same* figure is reversed, and the "theoretical" Eq. (14) is plotted as a line, with Eq. (11) as points, the fit does not seem so good to most of a small sample asked about it, probably because the human eye is not accustomed to fit data to a curved line.

The plot of Fig. 3 thus shows that all the published one-decade data, reported to fit the logarithmic function, would fit Eq. (14) equally well, and a whole lot of other functions too. It is even possible to give some physical meaning to the parameter M_0 in Eq. (14) by claiming that measurements do not start at $t = 0$, but at some unknown beginning. After all, the form of Eq. (11) cannot possibly be extended to the measurement at time $t = 0$, so that those who report the experiments may not have noted the starting time, and in a case like Ref. 22 it is even implied by the mere use of Eq. (12). Moreover, in the case of a superconducting magnet, switching off the field is a lengthy procedure, and some of the magnetization decays *during* the removal of the field, and before the measurement actually starts. This shift in the zero of the time has been noted by Beck,[25] and was used[26] to explain the maximum in thermoremanence magnetization, which used to be considered as due to some special mechanism, existing only in the so-called spin glasses. Moreover, the explanation[26] of that maximum in terms of the start of measuring time has recently been supported by the same maximum being observed[27] in *particles*, and not only in a spin glass.

However, in spite of the possible physical significance of M_0 in Eq. (14) the above-mentioned fitting requires nearly $\frac{1}{3}$ of the magnetization to decay before starting the count of time, which does not seem reasonable. Therefore, the agreement between the curves in Fig. 3, nice as it looks, is most probably fortuitous. All it can really show is that one decade of time is far from being adequate for revealing the structure of the physical system involved. More generally speaking, an agreement between a theory and the experimental data is always necessary, but it is never a sufficient condition for a meaningful theory.

C. Bessel function

A more satisfactory way is to start with a distribution of the *relaxation times* and generalize Eq. (10) to

$$M_r(t) = M_r(0) \int_0^\infty P(\tau) e^{-t/\tau} d\tau, \tag{15}$$

for some distribution function P. Since there is not much difference between all the different possible bell-shaped functions, it was suggested[20] to use the one for which the integration in Eq. (15) can be carried out analytically, thus avoiding the computational errors of numerical integration. That function is known as the Γ distribution function,

$$P(\tau) = \frac{1}{\tau_0 \Gamma(p)} \left(\frac{\tau}{\tau_0}\right)^{p-1} e^{-\tau/\tau_0}. \qquad (16)$$

It contains two adjustable parameters: The mean $p\tau_0$ and the variance $p\tau_0^2$, and these parameters contain all the physical information that is necessary to characterize an ensemble of particles.

Using this distribution function in Eq. (15), the observed time dependence should be[20]

$$\frac{M_r(t)}{M_r(0)} = \frac{2}{\Gamma(p)} \left(\frac{t}{\tau_0}\right)^{p/2} K_p \left[2\left(\frac{t}{\tau_0}\right)^{1/2}\right], \qquad (17)$$

where K_p is the modified Bessel function of the third kind. Some experimental data of Chamberlin *et al.*,[21] which had originally been fitted to stretched exponentials were analyzed[20] in this way, and were found to be in excellent agreement with Eq. (17). However, this agreement did not encourage others to use this analysis for their data, and even the most recent experiments[28] use Eq. (11). El-Hilo *et al.*[27] quote the analysis of Ref. 20, but report their data in terms of Eq. (11). It is very strange that a relation which is so unphysical and so inconvenient to use or to interpret became so popular.

Charap[23] used the *same* distribution function as in Eq. (16), for $f = 1/\tau$, but instead of Eq. (15) he used it for a distribution of energy barriers. He related these entities via the Néel relation in Eq. (1), and after introducing an approximation which is valid for $t \gg \tau_0$, he ended up with some power law. The trick is somewhat unnecessary when there is an analytic solution, but the result is not very different, and Charap also found[23] very good agreement with some experiments.

It may thus be concluded that for a reliable estimate of the location of the resonance discussed in Sec. II, one has to measure the magnetic viscosity *in the same sample* over several decades of time. An analysis of the data according to Eq. (17) will then give the average τ, which is the value of the resonance frequency. It also gives the width of the distribution function in τ, which is a lower bound to the resonance linewidth. As mentioned in Sec. II, there is an additional width, which exists even for a single particle, so that there is no simple relation between the resonance linewidth and the width of the τ distribution. However, it is at least qualitatively clear that it is not worth looking for such a resonance unless the τ distribution is rather narrow. This information is automatically obtained from fitting the data to Eq. (17).

Since the published experimental data are not detailed enough to be used for a proper analysis of the remanent magnetization versus time, an example for this fitting will be given for the same fictitious material used in the previous paragraph. The data of Liu *et al.*[24] are taken again as if they were given by the straight line of Eq. (13). On this line, 55 equidistant values in t were chosen, starting at $t = 0.45$ min and ending at $t = 5.85$ min. These points were fitted by least squares to Eq. (17), which yielded the parameters $M_r(0) = 28.2348 \times 10^{-3}$ T, $p = 0.2303$, and $\tau_0 = 20.221$ min. The fit is very good indeed, as can be seen in Fig. 4.

From these values, the average τ is $p\tau_0 = 4.6569$ min, and it is interesting to note that it is not very different from the value $\tau = 4.8757$ min, obtained in the foregoing from the fitting to the simple exponential of Eq. (10), even though the fit there was not good. It illustrates the point that a bad fitting to the exponential function contains more physical information than a good fitting to the logarithmic function.

The cycle for flipping the magnetization back and forth is $2p\tau_0$, which is 9.31 min, in this case. Therefore, a susceptibility resonance should be expected for the sample of Liu *et al.*[24] at a frequency of 1/9.31 per *minute*, which is 1.8×10^{-3} Hz, and is obviously too low for practical observation. Moreover, the resonance linewidth, which is at least $p\tau_0^2$, is much too large, and this particular sample cannot be recommended for this experiment. A measurement of the resonance is feasible only for a large p and a small τ_0.

More practical cases must be worked out by the experimentalists who have the original data, especially those data that extend over several decades of time. Such an analysis of the original data of Chamberlin on 2.6% Ag:Mn+0.46% Sb, at the temperture $T/T_g = 0.961$, led[20] to $p = 0.070$ and $\tau_0 = 9.66 \times 10^4$ s. At that temperature the resonance of that sample should, therefore, also be very wide, and at an extremely low frequency, which is not surprising for a spin glass.

Finally, it may be worth noting that the application

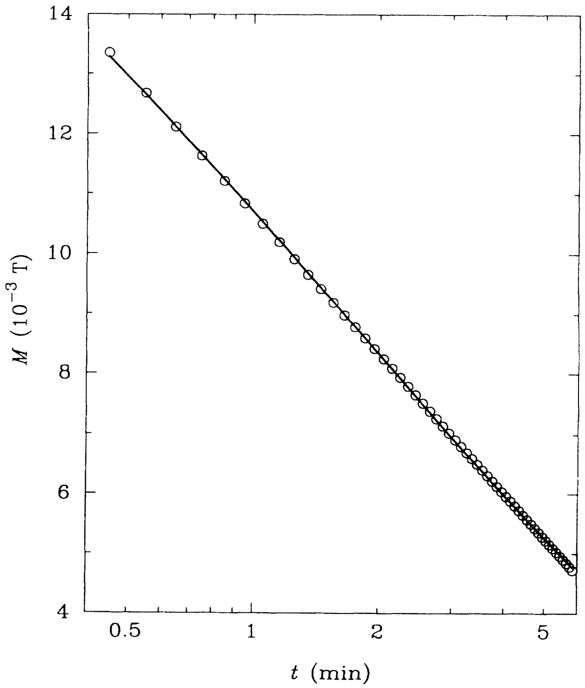

FIG. 4. The data of Ref. 24, approximated by Eq. (13), are plotted as circles. The solid line is a fitting of these "data" points to Eq. (17).

of Eq. (17) is not necessarily limited to the systems of small particles discussed here, or to the spin glasses to which it originally[20] referred. It should certainly be valid for magnetic viscosity measurements[29] of the *increase* in the magnetization with time, after applying the field, because this study is essentially the same as the study of the magnetization decay discussed in the foregoing. But it may also apply to completely different physical systems, which have only a mathematical similarity to the present study. For example, the magnetic relaxation in superconductors has been fitted[30] to $\ln[\ln(t/t_0)]$, or to a power of the logarithm. The fit is very good, but these functions just conceal the physical issues, and it may be better to try the function in Eq. (17). The same applies to a recent study of the magnetization decay in a nonzero magnetic field,[31,32], which led either to a[31] power law, $[\ln(t)]^n$, or to a[33] power-series expansion in $\ln(t/t_0)$. Both approaches conserve the unphysical and unnecessary singularities at both large and small t.

Similarly, for the decay of the magnetization in a bulk sample, subdivided into *domains*, the analysis[34] is in terms of the same Eq. (15) here, only with a different $P(\tau)$. They[34] claim their $P(\tau)$ to be based on a theory, but the physical processes are rather poorly known, and all theories involve some approximations. It is not impossible that a different distribution in τ is just as valid, and that the use of Eq. (17) may prove a more practical approach. After all, it is not absolutely necessary for any analysis to be based on a sound theory which starts from basic principles, and the approximation involved in replacing the correct $P(\tau)$ by any other distribution may not be worse than the errors involved in the numerical integration of the correct formula. It is at least worth trying the $P(\tau)$ for which the integration can be carried out analytically.

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