Theory of relaxation of magnetic clusters in a Stern-Gerlach setup

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We use a stochastic theory approach to discuss different scenarios such as ''locked moment'' and ''superparamagnetic'' behavior of magnetic clusters seen in Stern-Gerlach (SG) experiments. A dimensionless parameter $\lambda\tau_E$ (where λ is an internal spin relaxation rate and τ_E is the time spent by the cluster in the field gradient region) and the initial polarization of the cluster moment are shown to determine sensitively the position of the cluster beam on the SG screen. [S0163-1829(98)08517-8]

The study of magnetic clusters is a very active area of research in mesoscopic physics.[1–5] Apart from their potential importance in industrial applications, these clusters are interesting in their own right for a fundamental understanding of their magnetism in general. A useful experimental technique for studying magnetic properties of clusters is to employ them in a molecular beam in a Stern-Gerlach (SG) setup and measure deflections on a detector. A proper analysis of this experiment is expected to reveal important information about the cluster size, anisotropy energies, magnetic moment per atom, temperature dependence of magnetization, and, above all, thermal relaxation processes. In particular, it has been found that magnetic clusters show two limiting characteristics referred to as ''locked moment'' and ''superparamagnetic'' behaviors depending on the orientational relaxation of the moment as it traverses the SG apparatus.[3] In this paper we provide a unified picture of these different scenarios based on a stochastic theory approach and discuss our theoretical results in the light of experiments on transition-metal and rare-earth clusters.[3–5]

For the sake of definiteness we shall use the result for the deflection on the screen in the usual SG geometry (sketched schematically in Fig. 1) as a reference. If the beam consists of two-level atoms as in the historic experiment, the positive deflection, say, for the ''spin-up'' atoms, is given by[3]

$$d=\frac{\mu}{m}B'(0)\,\frac{LD}{v^2}\left(1+\frac{1}{2}\,\frac{L}{D}\right), \tag{1}$$

where μ is the atomic moment, m is the mass of the atom, $B'(0)$ is the gradient of the magnetic field [assumed to have a constant part B_0 which is much larger than $z_0B'(0)$, z_0 being the width of the beam], the distances L and D are as indicated in the figure, and v^2 is the mean-squared velocity of the atoms. The latter depends on the ''source temperature'' T_s through the relation $v^2=(k_BT_s)/m$, k_B being the Boltzmann constant. Considering that the beam traverses a length L in the magnetic field, we may already introduce the notion of the experimental time scale $\tau_E=L/v$. This will be used later as a yardstick for discussing relaxation effects which will be characterized by other time scales. Our objective is to give a *unified* theory which encompasses both transition-metal clusters, i.e., those of cobalt, nickel, or iron (believed to have ''small'' anisotropy energies), and rare-earth clusters, i.e., those of gadolinium (with ''large'' anisotropy energies). The central issue is, what is d (or more accurately its average), when the beam of clusters undergoes thermal relaxation during the traversal through the magnetic field?

The model which captures our proposed physical picture is the following: each cluster is viewed as a single-domain magnetic particle (consisting of up to hundreds of atoms) with the individual atomic moments locked up in a given direction, yielding a giant moment for the particle. Thus we ignore, for the present discussion, intraparticle[6] fluctuations. The moment of the particle is, however, not fixed in direction as it is expected to undergo thermal fluctuations (due to a spin-lattice interaction). The latter arise from a heat bath which is maintained at a temperature that has been referred to in the literature[3–5] as the vibrational temperature T_v. The effect of the thermal fluctuations is to relax the orientation of the magnetic moment of the particle, as in rotational Brownian motion.[7]

We should keep in mind that our Brownian particle (or, precisely, the moment) is, in fact, under the influence of a potential which is a combination of the anisotropy energy and the Zeeman energy. Assuming for the sake of simplicity that the anisotropy is uniaxial,[8] the potential may be written

$$\Phi(\theta)=N(K\,\sin^2\theta-B_0\mu_0\,\cos\,\theta), \tag{2}$$

where N is the number of atoms in the cluster, K and μ_0 are, respectively, the anisotropy energy and magnetic moment per atom, and θ is the angle between direction of the moment

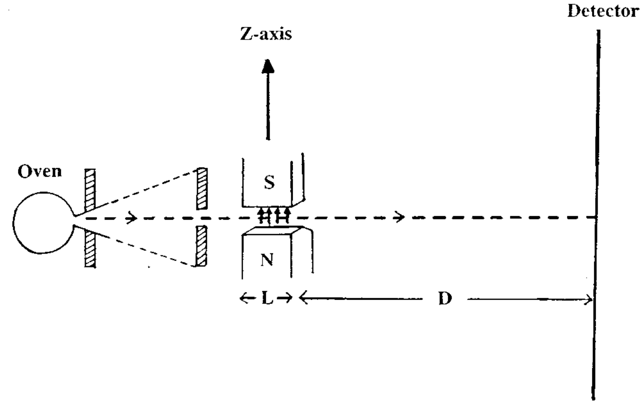

FIG. 1. A typical Stern-Gerlach setup.

and the applied magnetic field. For simplicity, we assume the magnetic field to be along the direction of anisotropy; this assumption can, however, be relaxed easily. The equilibrium value of the magnetization is given by

$$M = N\mu_0 \langle \cos\,\theta \rangle_{\text{eq}}, \tag{3a}$$

where

$$\langle \cos\,\theta \rangle_{\text{eq}} = \frac{\int_0^{\pi} d\theta\,\sin\,\theta\,\cos\,\theta\,\exp(-\Phi(\theta)/k_B T_v)}{\int_0^{\pi} d\theta\,\sin\,\theta\,\exp(-\Phi(\theta)/k_B T_v)}. \tag{3b}$$

For very weak anisotropy $(K \approx 0)$, Eq. (3) can be approximated by

$$\langle \cos\,\theta \rangle_{\text{eq}} \approx \frac{k_B T_v}{N B_0 \mu_0} - \coth\left(\frac{N B_0 \mu_0}{k_B T_v}\right). \tag{4}$$

On the other hand, if the anisotropy is very large, the moment is ''locked'' at the orientation 0 or π, and hence

$$\langle \cos\,\theta \rangle_{\text{eq}} \approx \tanh\left(\frac{N B_0 \mu_0}{k_B T_v}\right). \tag{5}$$

Thus, in either of the limits, $\langle \cos\,\theta \rangle_{\text{eq}}$ is independent of K.

We turn our attention now to the main focus of our study, namely, the relaxational dynamics of the moment. Insofar as the dynamics is viewed to be described by rotational Brownian motion,[7] the underlying probability distribution $P(\theta,t)$ obeys a Fokker-Planck equation

$$\tau_0 \frac{\partial}{\partial t} P(\theta,t) = \frac{1}{\sin\,\theta} \frac{\partial}{\partial \theta}\left[\sin\,\theta\left(\frac{1}{k_B T_v}\frac{\partial \Phi}{\partial \theta} P(\theta,t) + \frac{\partial P(\theta,t)}{\partial \theta}\right)\right], \quad 0 \leq \theta \leq \pi, \tag{6}$$

where τ_0 is simply a parameter that sets the basic time scale of the process. Because the so-called drift term in the Fokker-Planck equation (6) is written in terms of the potential $\Phi(\theta)$, Eq. (6) is guaranteed to yield for $P(\theta,t)$ in the asymptotic (i.e., $t \to \infty$) limit, the equilibrium distribution function

$$P_{\text{eq}}(\theta) = \frac{\exp(-\Phi(\theta)/k_B T_v)}{\int_0^{\pi} d\theta\,\sin\,\theta\,\exp[-\Phi(\theta)/k_B T_v]}. \tag{7}$$

Evidently, it is this quantity which is used to evaluate Eq. (3b).

The Fokker-Planck equation is the starting point of our analysis. However, it is important to emphasize that this equation describes the stochastic dynamics of an ''internal'' variable such as the orientation of the magnetic moment. As such, it would have *no* influence on the barycentric motion of the cluster were it not for the inhomogeneous magnetic field present in the SG setup. Thus we need to tie up our previous discussions with the kinematics of the cluster center-of-mass motion along the z axis. The latter is naturally governed by the acceleration

$$a(t) = \frac{N\mu_0 B'(0)}{M} \cos\,\theta(t), \tag{8}$$

which is a stochastic process, because $\theta(t)$ is. Thus the relevant quantity to calculate is the mean velocity in the z direction, given by $\overline{\nu_z}(t) = b\int_0^t \langle \cos\,\theta(t') \rangle dt'$, where $b = N\mu_0 B'(0)/M$, M is the mass of the cluster, and we assume $\overline{\nu_z}(0) = 0$. The angular bracketed quantity is defined by $\langle \cos\,\theta(t) \rangle = \int_0^{\pi} d\theta \sin\,\theta \cos\,\theta\, P(\theta,t)$. Knowing $\overline{\nu_z}(t)$, the SG deflection can be evaluated as

$$d = \int_0^{\tau_E} \overline{\nu_z}(t)dt + \frac{D}{v}\,\overline{\nu_z}(\tau_E), \tag{9}$$

where τ_E is the experimental time scale defined in the paragraph below Eq. (1).

The exact expression for $\langle \cos\,\theta(t) \rangle$ requires the most general solution of Eq. (6) which is analytically intractable. We believe, however, that a range of cluster behavior can be described within the so-called transition state theory of Kramers which works very well within high barrier and weak thermal noise limit.[9,10] In that region, the solution of Eq. (6), with the initial condition $\theta = \theta_0$ at $t = 0$ can be approximately written as[11]

$$P(\theta,t) = P_{\text{eq}}(\theta) + [\delta(\cos\,\theta - \cos\,\theta_0) - P_{\text{eq}}(\theta)]e^{-\lambda t}, \tag{10}$$

where $P_{\text{eq}}(\theta)$ is given by Eq. (7). The rate parameter λ is the dominant eigenvalue in the Kramers regime which can be estimated from a variational treatment. The result is[7]

$$\lambda = \frac{\pi}{\tau_0}\left(\frac{2NK}{\pi k_B T_v}\right)^{3/2} \exp\left[-\frac{NK}{k_B T_v}\left(1 + \frac{B_0^2 \mu_0^2}{4K^2}\right)\right] \times \left\{\cosh\left(\frac{N B_0 \mu_0}{k_B T_v}\right) - \frac{B_0 \mu_0}{2K} \sinh\left(\frac{N B_0 \mu_0}{k_B T_v}\right)\right\}, \tag{11}$$

which is valid only when $B_0\mu_0/2K \leq 1$. In the absence of the Zeeman interaction, λ reduces to the well-known Néel formula[12]

$$\lambda(B_0 = 0) = \frac{\pi}{\tau_0}\left(\frac{2NK}{\pi k_B T_v}\right)^{3/2} \exp\left[-\frac{NK}{k_B T_v}\right]. \tag{12}$$

While employing Eq. (10) for evaluating d from Eq. (9), it is convenient to take an average over the initial cluster orientation θ_0, to be denoted as $\langle \cos\,\theta_0 \rangle$. For a polarized (unpolarized) beam $\langle \cos\,\theta_0 \rangle = 1(0)$. We then obtain

$$\begin{aligned} d = {} & \frac{bD}{v} \langle \cos\,\theta_0 \rangle \left\{\frac{1}{\lambda}(1 - e^{-\lambda \tau_E}) + \frac{v}{D\lambda}\left[\tau_E - \frac{1}{\lambda}(1 - e^{-\lambda \tau_E})\right]\right\} \\ & + b\langle \cos\,\theta \rangle_{\text{eq}} \left\{\frac{1}{2}\tau_E^2 + \frac{D}{v}\left(1 - \frac{v}{D\lambda}\right) \times \left[\tau_E - \frac{1}{\lambda}(1 - e^{-\lambda \tau_E})\right]\right\}. \end{aligned} \tag{13}$$

Equation (13) is our final result, which will form the basis of our further analysis of different regimes of relaxation. Before we carry out this analysis, it is important to check that in the *static* limit $(\lambda = 0)$, Eq. (13) does reduce to Eq. (1) for a polarized beam, as expected. We now discuss the two extreme limits of Eq. (13) in terms of the dimensionless parameter $\lambda\tau_E$ that characterizes the interplay of thermal relaxation time in relation to the experimental time scale.

(i) *Slow relaxation* ($\lambda\tau_E \ll 1$): In this limit the SG displacement d is given by

$$d \approx \frac{bD\tau_E}{v}\left[\langle\cos\,\theta_0\rangle\left(1+\frac{v\tau_E}{2D}\right)-\frac{1}{2}\lambda\tau_E\left(1+\frac{v\tau_E}{3D}\right)\right.$$
$$\left.\times(\langle\cos\,\theta_0\rangle-\langle\cos\,\theta\rangle_{\rm eq})\right]. \tag{14}$$

(ii) *Fast relaxation* ($\lambda\tau_E \gg 1$):

$$d \approx \frac{bD\tau_E}{v}\left[\left(1+\frac{v\tau_E}{2D}\right)\langle\cos\,\theta\rangle_{\rm eq}+\frac{1}{\lambda\tau_E}\left(1+\frac{v\tau_E}{D}\right)\right.$$
$$\left.\times(\langle\cos\,\theta_0\rangle-\langle\cos\,\theta\rangle_{\rm eq})\right]. \tag{15}$$

Several comments may now be made on the basis of the above results. (a) As the relaxation picks up from the extreme static limit ($\lambda=0$), there is a discernible shift in d, and the direction of the shift with respect to $z=0$, the center of the screen, depends on the sign of $q=(\langle\cos\,\theta_0\rangle-\langle\cos\,\theta\rangle_{\rm eq}$. (b) In the extreme rapid relaxation limit, the deflection is proportional to the average magnetization of the cluster [cf. Eq. (15)]. This is expected because the magnetic moment tumbles over so frequently during the traversal of the cluster through the gradient field that the magnetization equilibrates to the vibration temperature. This regime is known to be characterized by superparamagnetic relaxation.[4,5] Direction of the shift for finite $\lambda\tau_E$ also depends on the sign of q. (c) The fast relaxation limit is particularly relevant for transition-metal clusters for which the anisotropy is low. In addition, for a partially polarized beam, if k_BT_v is much larger than the Zeeman energy, then $\langle\cos\,\theta\rangle_{\rm eq}$ is vanishingly small [cf. Eq. (7)] and can be neglected compared to $1/\lambda\tau_E\langle\cos\,\theta_0\rangle$. In that case [cf. Eq. (15)],

$$\lim_{\lambda\to{\rm large}} d \approx b\,\frac{D}{v\lambda}\,\langle\cos\,\theta_0\rangle, \tag{16}$$

where D/L is also suitably large. Hence the deflection is now inversely proportional to v or to $\sqrt{T_s}$, T_s being the source temperature. In contrast, in the static limit (perhaps appropriate to the rare-earth clusters in which the moments are ''locked''), the deflection is inversely proportional to v^2 or to T_s. This may be used to experimentally discern the low- to high-anisotropy behavior.

In Fig. 2 we plot as a function of $\alpha=\lambda\tau_E$ (which is a measure the ratio of the time the cluster spends inside the gradient magnet to the relaxation time) the dimensionless quantity

$$\bar{d}=\frac{d}{b\tau_E^2}\,\frac{L}{D}=\frac{d}{d_0},$$

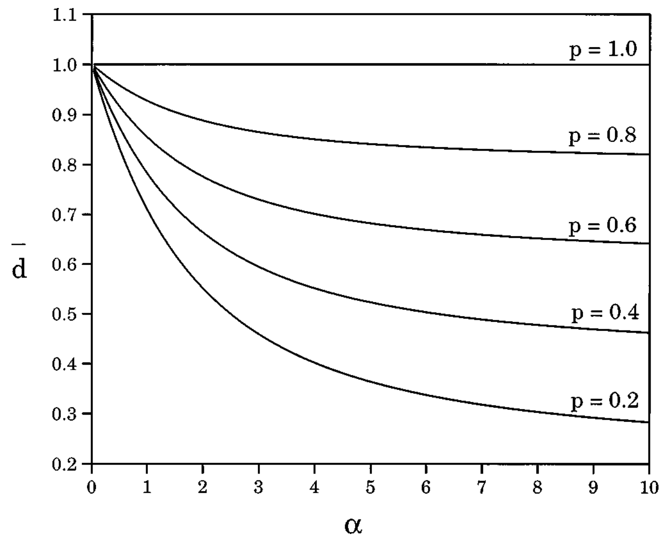

FIG. 2. Stern-Gerlach deflection d (measured in units of d_0 defined in the text) for a polarized beam, i.e., $\langle\cos\,\theta_0\rangle=1$, as a function of $\alpha=\lambda\tau_E$, where τ_E is the time the cluster spends inside the magnetic field gradient and λ is a characteristic spin orientational relaxation rate. The different curves correspond to different values of the thermal average moment $p=\langle\cos\,\theta\rangle_{\rm eq}$.

where $d_0=\mu_0B'(0)LD/mv^2$, for different values of $p=\langle\cos\,\theta\rangle_{\rm eq}$ and $\langle\cos\,\theta_0\rangle=1$. Typical experimental values are $B_0=1.034$ T, $B'(0)=310.1$ T/m, $D=1.183$ m, $L=0.25$ m, and $T_{\rm vib}$ in the range 100–300 K. In Fig. 2, the small-α limit corresponds to the ''locked'' moment regime, whereas the large-α limit corresponds to the ''superparamagnetic'' regime. If $p\approx 1$, then the displacement is quite insensitive to the value of α. On the other hand, the rate of decrease of $\bar{d}$ with α is extremely rapid for smaller values of the average moment p. The characteristic value of α that separates the slow and fast relaxation regimes is about 3 for smaller values of p.

A few challenging theoretical problems remains open. (1) The Fokker-Planck equation (9) has been solved here within the transition state theory of Kramers.[9] A more general solution requires a more sophisticated treatment.[13] (2) Within the present model one can also look at the distribution in d (line shape) to compare with experiment. (3) The present model assumes that the magnetization vector of the cluster is a hindered rotor, i.e., inertial effects have been ignored, as in the overdamped limit of the Brownian motion. There may be cases with magnetic clusters in which a complete phase space dynamics will be necessary involving ''spin-rotation'' coupling. This aspect may shed light on the issue of rotational temperature mentioned in the literature.[2] (4) Finally, at ultralow temperatures and with large anisotropy, the magnetic moment of the cluster may quantum mechanically tunnel from one energy minimum to another.[14] To what extent is such mesoscopic quantum tunneling important for SG experiments is an open issue.

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