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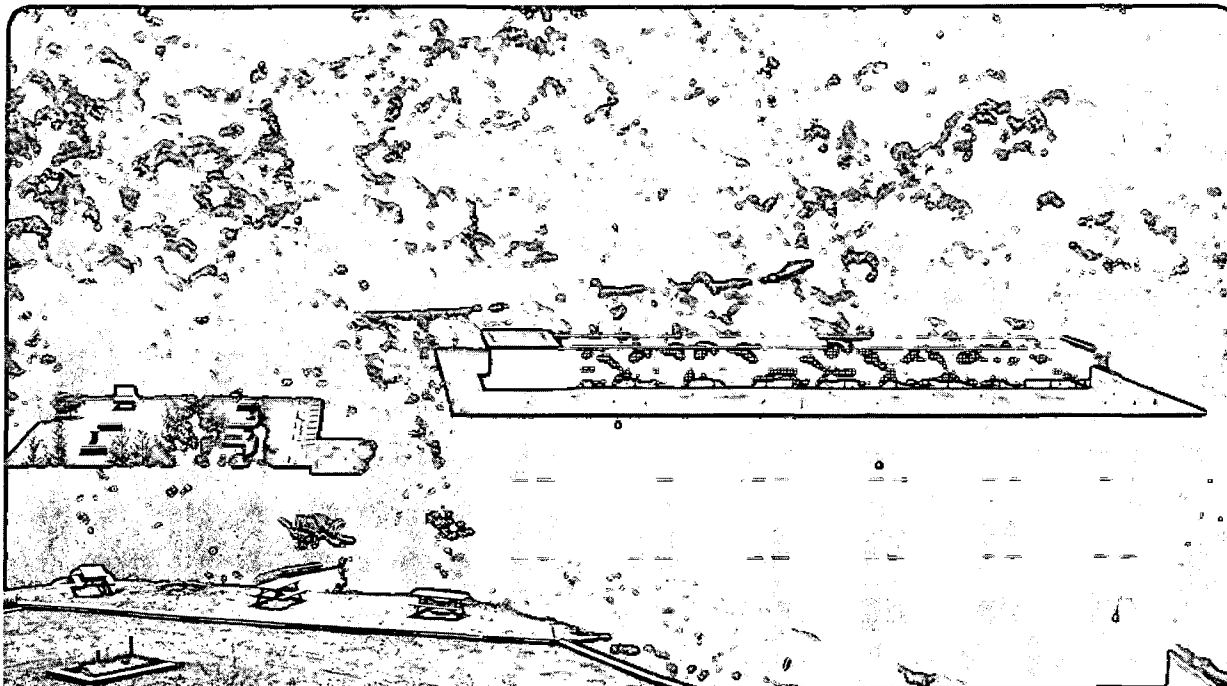
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A. Maiti and L.M. Falicov

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Stern-Gerlach Dynamics of Magnetic Clusters*

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Stern-Gerlach Dynamics of Magnetic Clusters

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Abstract

A classical theory of the deflection of a single-domain ferromagnetic cluster in a Stern-Gerlach experiment is presented. Two cases are discussed in detail; 1) *superparamagnetic relaxation*, in which the thermal fluctuation of the cluster spin occurs on a time scale much smaller than the transit time of the cluster through Stern-Gerlach apparatus; and (2) *anisotropy-induced relaxation*, in which the cluster is sufficiently isolated from the thermal bath during its transit through the apparatus so that its dynamics is governed by the interaction of the cluster spin with the anisotropic magnetic forces caused by the lattice and the shape of the cluster. Average spin-orientation and average spin-rocking during transit, as well as Stern-Gerlach intensity profiles are presented.

1. Introduction

It is well-known that for ferromagnetic materials there exists a critical size, usually a few nanometers in diameter, below which a particle possesses only a single magnetic domain¹. Such magnetic clusters, in which all the atomic spins are aligned parallel to each other, have recently become a subject of intensive experimental and theoretical study. Stern-Gerlach (SG) experiments on iron², cobalt^{3,4} and gadolinium⁵ clusters have revealed features in the deflection profile that are completely different from those observed for isolated atoms or molecules. Experiments on iron clusters have also been done using SQUID magnetometry and Mössbauer spectroscopy⁶. Theoretical models⁷⁻¹¹ have been built concurrently in order to interpret observed deflection patterns as functions of cluster-size, temperature and magnetic field strength. The dependence of the physical properties on cluster size involves the determination of the electronic structure and has been addressed elsewhere^{10,11}. The present contribution deals with the effects of temperature and magnetic field on the deflection pattern of a cluster of well defined, fixed size. All relevant experiments suggest that for very small clusters (of less than about 1000 atoms) the dynamics of the cluster spin is dominated by either of the following two kinds of behaviors: (1) *superparamagnetic relaxation*, in which the thermal fluctuation of the cluster spin occurs on a time-scale τ_{th} much smaller than τ_p , the time of transit of the cluster through the poles of the SG magnet, and (2) *anisotropy-induced relaxation*, in which the cluster is sufficiently isolated from the thermal bath during its transit through the SG apparatus, and its dynamics is governed by the interaction of the cluster spin with the anisotropic magnetic forces caused by the lattice and the shape of the cluster. The above two situations give rise to a variety of effects of temperature and magnetic field on the SG deflection profile. It is the purpose of this contribution to analyze, compare and contrast the effects of these different mechanisms of spin relaxation.

2 . Model Lagrangian and equations of motion

The model consists of a spherical particle of moment of inertia I about any diameter, and a uniaxial magnetic anisotropy with a unique easy axis fixed to the cluster. Simulations with other familiar kinds of anisotropies, *e.g.* uniaxial anisotropy with an easy plane, or cubic anisotropy, yield qualitatively similar results and are not discussed here. The particle is assumed to be small enough (typically consisting of, $N \sim 100$ atoms) to sustain a single magnetic domain. Since all the atomic spins are aligned parallel to one another and $S^2 = \mathbf{S} \cdot \mathbf{S}$, the square of the total cluster spin \mathbf{S} is a constant of motion, the atomic spin-spin interaction in the Heisenberg model becomes a constant, which can be dropped out of the problem. One arrives at the familiar "single spin" model Lagrangian⁷⁻⁹:

$$L = E_K - (V_a + V_Z) \quad ; \quad (2.1)$$

where E_K is the cluster rotational kinetic energy, V_a is the energy caused by the coupling of the cluster magnetic moment to the magnetic anisotropic forces, and V_Z is the Zeeman energy, the coupling of the cluster magnetic moment to the magnetic field of the SG apparatus. The spacing of the quantized energy levels caused by the E_K term is much smaller than any experimental temperature and can be neglected. The problem then becomes one of a classical magnetic rigid rotator in the presence of a magnetic field and anisotropic magnetic forces. Thus the kinetic energy term becomes:

$$E_K = (1/2) I (\dot{\theta}^2 + \dot{\phi}^2 + \dot{\psi}^2 + 2 \dot{\phi} \dot{\psi} \cos\theta) \quad , \quad (2.2)$$

where θ, ϕ, ψ are Euler angles defining the orientation of the cluster with respect to the SG apparatus, *i.e.* the laboratory frame. Dotted quantities are time derivatives. Expressions for the other two terms in (2.1) are given by:

$$V_a = - K_0 (\mathbf{S} \cdot \hat{z})^2 / S^2 \quad ; \quad (2.3)$$

$$V_Z = \gamma S_z B \quad (2.4)$$

In (2.3) \hat{z}' is the unit vector in the direction of the positive z-axis of the reference frame fixed to the cluster body (henceforth referred to as the body frame) and defines the easy magnetic axis, while K_0 is the uniaxial-anisotropy constant. In (2.4) B , the magnetic field in the SG poles, is taken to be along the positive z-axis of the laboratory frame, and γ is the gyromagnetic ratio. The net variation of B within the SG poles is usually small compared to the average value. The magnetic field, therefore, is taken to be spatially constant. Because the values S of interest are of the order of 100 *i.e.*, large compared to a typical atomic spin and S^2 is a conserved quantity, it is more appropriate to consider a classical spin of constant magnitude S . The vector \mathbf{S} can thus be completely specified by its orientation angles α and β in the laboratory frame; α is the angle of inclination with the positive z-axis and β is the azimuthal angle measured anticlockwise from the positive x-axis. In the framework of this classical spin model, (2.3) and (2.4) take the forms:

$$V_a = - K_0 \{ \sin\alpha \sin\theta \sin(\phi-\beta) + \cos\alpha \cos\theta \}^2 \quad ; \quad (2.5)$$

$$V_Z = - \gamma S B \cos\alpha \quad (2.6)$$

Following Appendix A, it is straightforward to obtain the equations of motion corresponding to the dynamical variables $\theta, \phi, \psi, \alpha, \beta$:

$$I \dot{\phi} \sin^2\theta = S (C_1 - \cos\alpha - C_2 \cos\theta) \quad ; \quad (2.7)$$

$$I \dot{\psi} \sin^2\theta = S (C_2 - C_1 \cos\theta + \cos\alpha \cos\theta) \quad ; \quad (2.8)$$

$$I \ddot{\theta} = - I \dot{\phi} \dot{\psi} \sin\theta - \frac{\partial V_a}{\partial \theta} \quad ; \quad (2.9)$$

$$S \sin\alpha \dot{\alpha} = \frac{\partial V_a}{\partial \beta} \quad ; \quad (2.10)$$

$$S \sin \alpha \dot{\beta} = - \frac{\partial V_a}{\partial \beta} - \gamma S B \sin \alpha \quad ; \quad (2.11)$$

where C_1 and C_2 are related to conserved components of angular momentum (see Appendix A). It is useful to rewrite (2.7) - (2.11) in dimensionless forms in terms of the following quantities:

$$\tau = \frac{S}{I} t \quad ; \quad \Delta = \frac{2 K_0 I}{S^2} \quad ; \quad H = \frac{\gamma S B}{2 K_0}$$

Physically $\tau=1$ correspond to an interval of the order of a body rotation time of the cluster ($\sim 10^{-10}$ -- 10^{-11} sec) and the dimensionless magnetic field H represents the strength of the Zeeman term relative to the anisotropy constant K_0 . Another relevant energy scale is the temperature, T , that determines the value of τ_{th} , the thermal relaxation time of the cluster. Physically τ_{th} corresponds to the average time interval between successive "energy exchanges" of the cluster with the heat bath and shows an Arrhenius behavior¹²:

$$\tau_{th} = \tau_0 \exp(K_0 / k_B T) \quad , \quad (2.12)$$

where the constant τ_0 is derived from a complex expression which depends strongly on the gyromagnetic ratio γ and the friction in the path of the experimental cluster beam¹² (typically $\tau_0 \sim 10^{-10}$ -- 10^{-13} sec). A time-scale to be compared with τ_{th} is τ_p , the time of residence of the cluster inside the magnetic field of the SG poles (typically $\tau_p \sim 10^{-5}$ sec). When $k_B T \gg K_0$ one obtains the result $\tau_{th} \sim \tau_0 \ll \tau_p$. In this case the cluster thermally relaxes many times during its passage through the SG magnet. This is the case of *superparamagnetic relaxation* in which the cluster is essentially in thermal equilibrium throughout its transit in the field of the SG magnet. The other interesting case is $k_B T \ll K_0$ which implies $\tau_{th} \gg \tau_p$. This is the case of *anisotropy-induced relaxation* in which the cluster is effectively isolated from the thermal bath during its transit through the SG apparatus, and its motion is governed solely by the equations of motion (2.7) - (2.11).

3. Calculation and results

For any kind of relaxation process a quantity of calculational interest is the mean value of the z-component of cluster-spin $\langle \tilde{S}_z \rangle$ (to which the average deflection in the SG profile is proportional). The *single bracket* $\langle \rangle$ here corresponds to averaging over the initial conditions (angles and angular momenta when the cluster enters the SG field), while the *tilde* corresponds to a time average during the residence time τ_p in the SG apparatus. In the case of superparamagnetism τ_p is much larger than any other relevant time scale. In the extreme limit $\tau_p \rightarrow \infty$, any time-average is independent of the initial conditions, and therefore, $\langle \tilde{S}_z \rangle = \bar{S}_z$, which, assuming ergodicity, is equal to the canonical (thermodynamic) average $\langle\langle S_z \rangle\rangle$. Following results of Appendix B one arrives at the familiar Langevin equation for $\langle \tilde{S}_z \rangle = \langle\langle S_z \rangle\rangle$:

$$\langle \tilde{S}_z \rangle = S \left[\coth \left\{ \frac{2 K_0 H}{k_B T} \right\} - \frac{k_B T}{2 K_0 H} \right] \quad (3.1)$$

Figure 1 shows $[\langle \tilde{S}_z \rangle / S]$ versus H graphs for three different values of T [in units of (K_0 / k_B)]. The curves rise linearly from the origin and saturate to the upper limit $\langle \tilde{S}_z \rangle / S = 1$ for $H \gg 1$ (out of the graph's range). The remarkable property of (3.1) is that $\langle \tilde{S}_z \rangle$ is independent of the anisotropy constant K_0 as long as the condition $K_0 \ll k_B T$ is satisfied (see Appendix B for details). More specifically, K_0 does not have to vanish for $\langle \tilde{S}_z \rangle$ to satisfy the Langevin equation. Another quantity of interest is the average rocking of S_z during τ_p , given by:

$$\langle \sigma_S \rangle = \langle [S_z^2 - (\tilde{S}_z)^2]^{1/2} \rangle \quad (3.2)$$

For superparamagnetism $\langle \sigma_S \rangle$ is given by the canonical average (B.7) of Appendix B.

The important results for the case of superparamagnetic relaxation are summarized below:

- (1) $\langle \tilde{S}_z \rangle$ always increases with increasing H and decreasing T.
 - 2) For small magnetic fields ($\gamma S B \ll K_0$) one obtains $\langle \tilde{S}_z \rangle \propto H/T$;
for large fields ($\gamma S B \gg k_B T \gg K_0$) the results is
 $(1 - \langle \tilde{S}_z \rangle / S) \propto T/H$.
 - (3) The spin-rocking $\langle \sigma_S \rangle$ decreases monotonically with increasing H;
for large magnetic fields ($\gamma S B \gg k_B T \gg K_0$) one obtains
 $\langle \sigma_S \rangle \propto T/H^2$.
- It is also an always increasing function of the temperature.

The anisotropy-induced case is more difficult to treat because one needs to integrate the five coupled differential equations (2.7) - (2.11). The time-averaged quantities $\langle \tilde{S}_z \rangle$ and $\langle \sigma_S \rangle$ strongly depend on the initial conditions and it is necessary to average over eight continuous variables: the initial orientation angles θ, ϕ, ψ , the corresponding three angular momenta, and the spin angles α, β . Independence of the Lagrangian from ψ , and dependence on ϕ and β only in the form of $(\phi - \beta)$ effectively reduce the averaging procedure to a space of six variables; this is still computationally prohibitive. A major simplification is made by assuming that, before entering the SG field, the spin vector \mathbf{S} is locked into the magnetic easy axis of the body (since $K_0 \gg k_B T$), and that the rotational kinetic energy E_K is equal to the canonical average value $[(3/2) k_B T]$. This reduces the initial-condition averaging problem to one in a space of three variables. The time average is obtained by integrating the equations of motion (2.7) -(2.11). Integration is performed by using a modified form of the Runge-Kutta method; the constancy of the total energy given by (A.4) is checked after every 100 time steps. The total time of integration is 500 τ -units; longer runs do not change time-averaged results. Figure 2 shows results for $[\langle \tilde{S}_z \rangle / S]$ as a function of H for three different temperatures. Figure 3 shows the functional dependence of $\langle \sigma_S \rangle$ as a function of H at a given temperature. As can be easily seen, figure 2 is qualitatively very different from figure 1. The main features of figures 2 and 3 are summarized below:

- (1) The quantity $\langle \tilde{S}_z \rangle$ is not a monotonically increasing function of H . For small fields ($\gamma S B \ll k_B T \ll K_0$) it increases linearly in H , reaches a peak at $H \sim 0.5$ (i.e., $\gamma S B \sim K_0$), and starts decreasing, falling off as $(1/H)$ for $H \gg 1$.
- (2) For small fields ($H < 1$), the quantity $\langle \tilde{S}_z \rangle$ increases with decreasing T . However, the T -dependence is much weaker than in the superparamagnetic case, and there is no discernible T -dependence for $H > 1$.
- (3) The spin rocking $\langle \sigma_S \rangle$ decreases monotonically with increasing H , falling off as $(1/H)$ for $H \gg 1$.

4. Discussion

In order to understand the qualitative differences between figures 1 and 2, it is necessary to analyze the SG deflection profile for each kind of relaxation. Such a profile displays the relative intensity of the cluster beam as a function of transverse deflection in the direction of the magnetic field. It is obtained by forming a histogram of the time-averaged quantity \tilde{S}_z , due to varying initial conditions. The quantity $\langle \tilde{S}_z \rangle$ is the average of \tilde{S}_z weighted by the relative intensities in the histogram. Figure 4 shows the SG deflection profile for the case of anisotropy-induced relaxation for three different magnetic fields at a given constant temperature. The main features of figure 4 are summarized below:

- (1) The distribution of \tilde{S}_z is not sharply peaked even for $H=0$. With increasing H the peak moves towards the right (positive \mathbf{B} direction), but at the same time the distribution widens (a large number of clusters deflect in the negative \mathbf{B} direction).
- (2) The average $\langle \tilde{S}_z \rangle$ of figure 2 increases linearly for $H \ll 1$. For $H > 0.5$, the peak still moves towards the right extreme $\tilde{S}_z / S = +1$. However, increase of relative intensity in the negative \mathbf{B} direction offsets positive effect of the peak's right-shift, with a net effect of a decrease in the average $\langle \tilde{S}_z \rangle$ with increasing H .

- (3) For $H \gg 1$ the profile is almost uniform with a small shift of weight from the extreme left ($\tilde{S}_z / S = -1$) to the extreme right ($\tilde{S}_z / S = +1$).

The reason for behavior (3) above (and also the related spin rocking behavior of figure 3) is that, for very large fields the Zeeman term in (2.1) dominates over the anisotropy term, making S_z an approximate constant of motion. The relatively weak anisotropy term causes a small fluctuation [proportional to $(1/H)$ in the first order of perturbation, as is shown in figure 3] around the initial value of S_z during the transit time through the SG magnet. Fluctuations of S_z towards the positive \mathbf{B} direction are favored energetically over fluctuations towards negative \mathbf{B} . The net effect is that the time-averaged \tilde{S}_z is right-shifted from the initial S_z by a constant amount for each value of S_z , resulting in the behavior shown in figure 4 for $H = 3.0$.

The calculation of histograms for the case of superparamagnetism requires additional subtlety, because the profile depends explicitly on the detailed mechanism of thermal relaxation. However, the following general features can be expected :

- (1) In the extreme limit of infinite τ_p , the distribution is a delta function peak at the value $\langle \tilde{S}_z \rangle$ given by (3.1). For finite τ_p , however, a finite width proportional to $\tau_p^{-1/2}$ is to be expected.
- (2) With an increase in H , dependence on the initial value of S_z is enhanced (as follows from the discussion of the anisotropy-induced relaxation case) and the distribution is expected to widen. Figure 1 of reference 2 illustrates exactly the behavior just described, although the interpretation of the relaxation mechanism is quite different in that article.

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Appendix A

This appendix sets the stage that leads to the equations of motion of Section 2. The time evolution of the Euler angles θ, ϕ, ψ follows from the respective Lagrange's equations:

$$\frac{d}{dt} \left(\frac{\partial L}{\partial \dot{q}} \right) - \frac{\partial L}{\partial q} = 0 \quad , \quad (\text{A.1})$$

where L is given by equations (2.1), (2.2), (2.5), (2.6); q represents the variables θ, ϕ, ψ , and \dot{q} describes the corresponding time-derivative. Equations of motion of the spin orientation angles α, β , on the other hand, follow the Bloch equations:

$$S \frac{d}{dt} (\cos \alpha) = \frac{\partial L}{\partial \beta} \quad ; \quad (\text{A.2})$$

$$S \frac{d\beta}{dt} = - \frac{\partial L}{\partial (\cos \alpha)} \quad . \quad (\text{A.3})$$

Equations (A.1) - (A.3) imply conservation of the total energy:

$$E_{tot} = E_K + V_a + V_Z \quad . \quad (\text{A.4})$$

Equation (A.1) is a second-order differential equation (in time); for the ϕ and ψ variables one obtains the following simple first integrals (first-order differential equations):

$$S \cos\alpha + I (\dot{\phi} + \dot{\psi} \cos\theta) = S C_1 \quad ; \quad (\text{A.5})$$

$$I (\dot{\phi} \cos\theta + \dot{\psi}) = S C_2 \quad ; \quad (\text{A.6})$$

where C_1 and C_2 are dimensionless constants. Equation (A.5) follows from the fact that ϕ and β always appear in the combination $(\phi - \beta)$ in the expression for Lagrangian L [see (2.5); this property also holds for cubic anisotropy], and therefore

$$\frac{\partial L}{\partial \phi} = - \frac{\partial L}{\partial \beta} = - S \frac{d}{dt} (\cos\alpha)$$

because of (A.2). Equation (A.6) follows from the fact that the Lagrangian L is independent of ψ (this result applies only to the case of uniaxial anisotropy).

Physically C_1 corresponds to the z -component of the total angular momentum $\mathbf{J} = \mathbf{S} + I \boldsymbol{\Omega}$ (where $\boldsymbol{\Omega}$ is the angular-velocity vector of the rotating cluster) and C_2 corresponds to the conserved ψ -angular momentum.

Appendix B

This appendix derives the result that the canonical partition function Z , given by integration of $\exp(-E_{tot}/k_B T)$ over the phase-space of all angles and angular momenta, can be factored into a product of three components Z_K , Z_a and Z_Z to be defined below. It is convenient to define the differentials $d\phi_x$, $d\phi_y$, $d\phi_z$, which are the cartesian components (in the laboratory frame) of an arbitrary infinitesimal rotation of the body, and are given by

$$\begin{aligned}
d\phi_x &= d\theta \cos\phi + d\psi \sin\theta \sin\phi & ; \\
d\phi_y &= d\theta \sin\phi - d\psi \sin\theta \cos\phi & ; \\
d\phi_z &= d\phi + d\psi \cos\theta & .
\end{aligned}$$

The Jacobian of the transformation is given by

$$\frac{\partial(\phi_x, \phi_y, \phi_z)}{\partial(\theta, \phi, \psi)} = \sin\theta \quad (B.1)$$

It is also convenient to rewrite the rotational kinetic energy E_K in terms of cartesian components of the rotational angular momentum (in the laboratory frame) :

$$E_K = (1 / 2 I) (L_x^2 + L_y^2 + L_z^2) \quad (B.2)$$

where $L_x = I \dot{\phi}_x$ is the angular-momentum component along the positive x -axis of the laboratory frame, and similarly for y and z . The canonical partition function Z is given by:

$$\begin{aligned}
Z &= \int dL_x dL_y dL_z d\phi_x d\phi_y d\phi_z \sin\alpha d\alpha d\beta \exp(-E_{tot}/k_B T) \\
&= Z_K Z_V \quad (B.3)
\end{aligned}$$

where

$$Z_K = \int dL_x dL_y dL_z \exp(-E_K/k_B T) = (2 \pi I k_B T)^{3/2}$$

and

$$\begin{aligned}
Z_V &= \int d\phi_x d\phi d\phi_z \sin\alpha d\alpha d\beta \exp[-\{ V_\alpha(\theta, \alpha, \phi-\beta) + 2 K_0 H \cos\alpha \} / k_B T] \\
&= 2\pi \int d\phi_x d\phi_y d\phi_z \sin\alpha d\alpha \exp[-\{ V_\alpha(\theta, \alpha, \phi) + 2 K_0 H \cos\alpha \} / k_B T]
\end{aligned}$$

after integrating over β .

In order to decouple the integrations over θ and ϕ from that over α one needs to make a transformation from the laboratory frame (unprimed) in which the z -axis is in the direction of \mathbf{B} to the body frame (primed coordinates) in which the z' -axis is along \mathbf{S} . There is a proper orthogonal transformation matrix \mathbf{A} that transforms the unprimed into the primed system:

$$(d\phi_{x'}, d\phi_{y'}, d\phi_{z'}) = (d\phi_x, d\phi_y, d\phi_z) \cdot \mathbf{A}$$

where matrix multiplication is implied on the right hand side. Noting that the function $V_a(\theta, \alpha, \phi)$ transforms into $[-K_0 \cos^2\theta']$ in the primed system, and the Jacobian of transformation is:

$$\frac{\partial(\phi_{x'}, \phi_{y'}, \phi_{z'})}{\partial(\phi_x, \phi_y, \phi_z)} = \det \mathbf{A} = 1$$

one obtains

$$Z_V = Z_\alpha Z_Z \quad , \quad (\text{B.4})$$

where

$$\begin{aligned} Z_\alpha &= \int d\phi_{x'} d\phi_{y'} d\phi_{z'} \exp(K_0 \cos^2\theta' / k_B T) \\ &= \int \sin\theta' d\theta' d\phi' d\psi' \exp(K_0 \cos^2\theta' / k_B T) \end{aligned}$$

and

$$Z_Z = 2\pi \int \sin\alpha d\alpha \exp(2 K_0 H \cos\alpha / k_B T)$$

Finally

$$Z = Z_K Z_\alpha Z_Z \quad . \quad (\text{B.5})$$

It is important to note that all the magnetic-field dependence of Z is in the Z_Z factor. With the use of the double bracket notation $\langle\langle \rangle\rangle$ to denote canonical averaging one obtains:

$$\langle\langle S_z \rangle\rangle / S = \langle\langle \cos\alpha \rangle\rangle = \frac{\partial}{\partial H} \ln Z = \frac{\partial}{\partial H} \ln Z_Z, \quad (\text{B.6})$$

which yields the familiar Langevin equation. One also obtains the following formula for σ_S^2 :

$$\begin{aligned} \sigma_S^2 &= \langle\langle \cos^2\alpha \rangle\rangle - \langle\langle \cos\alpha \rangle\rangle^2 \\ &= \frac{\partial^2}{\partial H^2} \ln Z = \frac{\partial^2}{\partial H^2} \ln Z_Z \end{aligned} \quad (\text{B.7})$$

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Figure Captions

- Figure 1. The average normalized z-component of the cluster spin $\langle \tilde{S}_z \rangle / S$ as a function of the dimensionless magnetic field H (defined in the text) at three different temperatures for the case of superparamagnetism. Temperatures are in units of (K_0 / k_B) .
- Figure 2. The average normalized z-component of the cluster spin $\langle \tilde{S}_z \rangle / S$ as a function of the dimensionless magnetic field H (defined in the text) at three different temperatures for the case of anisotropy-induced relaxation. Temperatures are in units of (K_0 / k_B) .
- Figure 3. The average spin rocking $\langle \sigma_S \rangle$ as a function of H for the case of anisotropy-induced relaxation. The temperature is $k_B T = 0.07 K_0$.
- Figure 4. Stern-Gerlach intensity profile for three different values of H and a constant temperature $k_B T = 0.07 K_0$.

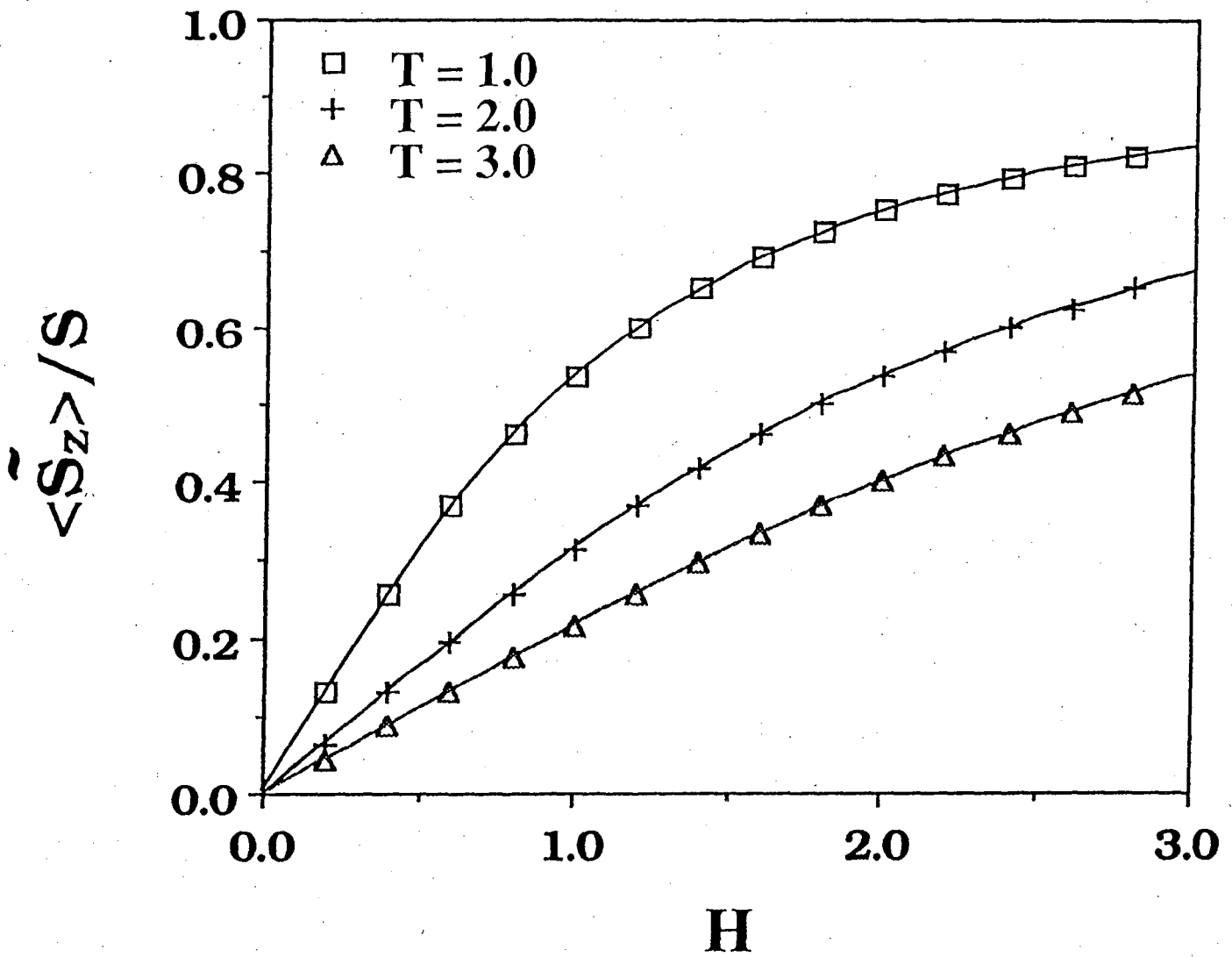


Figure 1

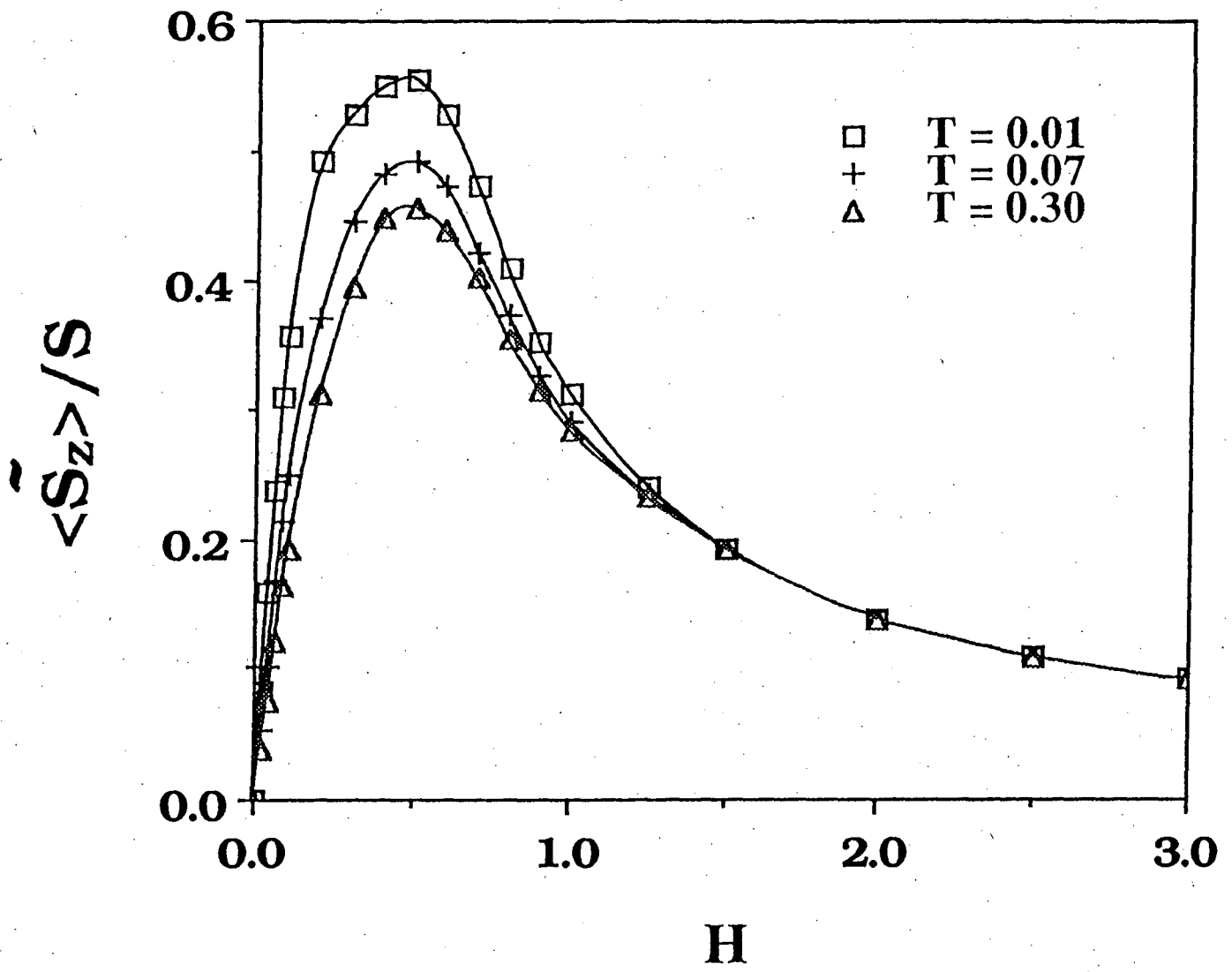


Figure 2

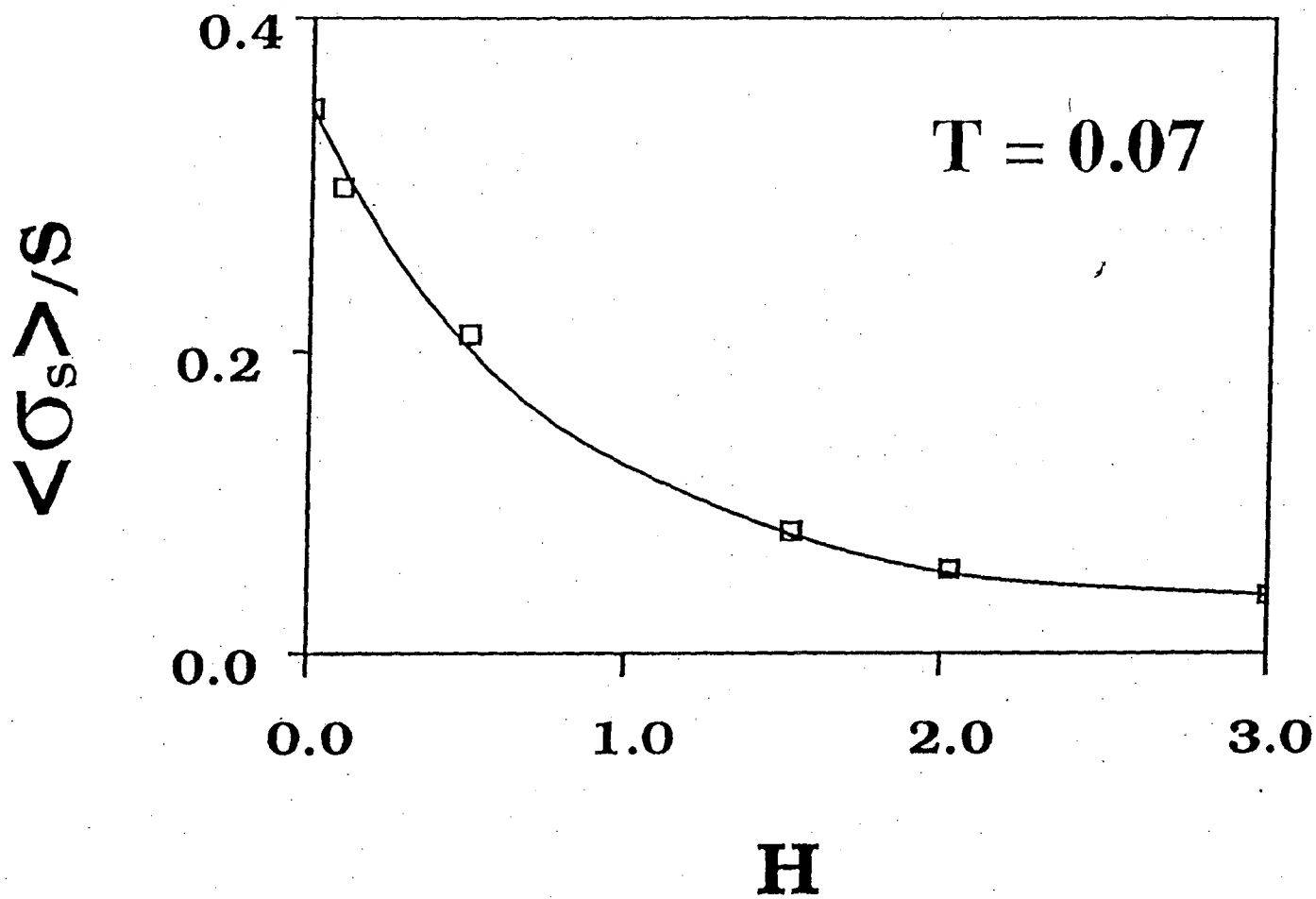


Figure 3

Relative Intensity (arb. units)

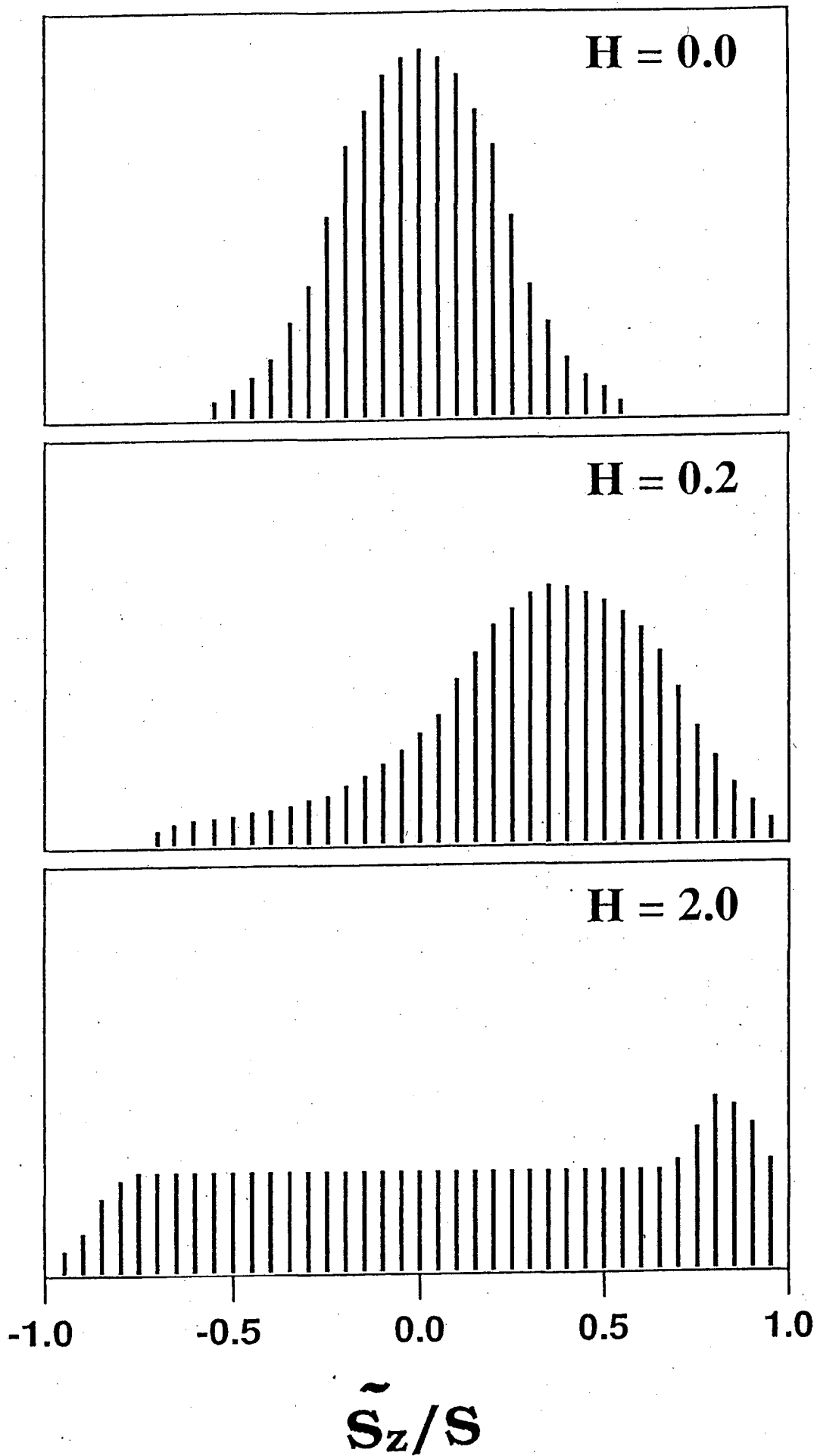


Figure 4

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