Macroscopic anisotropy in Dzyaloshinsky-Moriya spin glasses

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We present a model of the macroscopic anisotropy in spin glasses, based on current ideas distinguishing the "frozen-in" and "adjustment" parts of the energy due to anisotropic interactions. Predictions of the model are tested by computer simulations of Ruderman-Kittel-Kasuya-Yosida spin glasses with Dzyaloshinsky-Moriya (DM) anisotropy. Results from the computer simulations are consistent with the identification of the macroscopic anisotropy measured from ESR or transverse susceptibility with a frozen-in "first-order" anisotropy. The model quantitatively predicts the measured "adjustment energy" due to the anisotropic interactions, using most of the assumptions used to calculate the "frozen-in" anisotropy which would be measured in a macroscopic sample. Calculations of the macroscopic anisotropy for experimental samples using two alternate forms of the microscopic DM interaction, which give anisotropies 20 to 150 times greater than the value derived from hysteresis experiments for CuMnPt depending on the parameters used, are presented in detail.

I. INTRODUCTION

Anisotropy in spin glasses, already observed in 1960 by Kouvel,[1] has been studied extensively in hysteresis loop,[2] transverse susceptibility,[3] ESR,[4,5] NMR,[6] magnetotransport,[7] and torque[8] experiments. Initial efforts to characterize the anisotropy as unidirectional or uniaxial have led to work using a specific form for the anisotropy in a macroscopic Hamiltonian,[5,9–11] and an anisotropy constant K, to predict the behavior expected for these experiments. For torque experiments and ESR measurements in which the remanent magnetization is kept within a small enough angle of the direction in which it was cooled ($<\pi/2$, and depending on the size of the anisotropy), the triad model[11] has had great success in explaining the results. Irreversibilities appear in hysteresis loop and other experiments in which the magnetization is reversed or greatly removed from the orientation in which it was cooled, indicating that relaxation over barriers has become important, and the models necessary for these situations become more complex.

The strong enhancement of anisotropy in dilute metallic spin glasses such as CuMn resulting from a low concentration of impurities with strong spin-orbit coupling has been explained by Dzyaloshinsky-Moriya (DM) interactions.[12] This anisotropy mechanism would result in a unidirectional macroscopic anisotropy if the spin configuration is rotated rigidly, and it is reasonable to assume that relaxation does not change the form of the anisotropy, but only reduces its magnitude. The first theoretical work done to relate this microscopic pair anisotropy to an observed, macroscopic anisotropy constant was the Fert-Levy estimate of an upper bound for K, K_{DM}, which was the value of K expected for rigid rotation with no relaxation, assuming that the anisotropy energy is due entirely to the spins adjusting to random isotropic and anisotropic fields, by rotating through a small angle toward the anisotropic fields, and away from being exactly parallel to the isotropic fields. The estimated K_{DM} for CuMn with a low concentration of Pt, a very strong spin-orbit scatterer, was about a factor of 30 greater than measured values of K, and an unknown proportion of this error was attributed to relaxation effects, i.e., the nonrigid character of the macroscopic rotation.

Further work[13,14] has clarified the origin of two different contributions, to lowest order in the DM interaction constant, to K_r, the macroscopic anisotropy expected under rigid rotation. These contributions are the anisotropy due to the second order adjustment energy K_{DM} and an anisotropy K_0 due to the freezing in of a "first-order" anisotropy—i.e., the preferential trapping of the spin glass at T_G in configurations with favorable anisotropy energies. (The adjustment and frozen-in contributions to the anisotropy seen under rigid rotation will be derived and defined in Sec. II). By analogy with other systems, the frozen-in anisotropy might be expected to be of the same order of magnitude as the adjustment anisotropy, thus justifying the use of K_{DM} as an estimate of the macroscopic anisotropy, but this must be justified by a calculation with explicit assumptions, and these assumptions should be verified by experiment or numerical simulations. This paper will present a discussion of the assumptions and approximations needed to estimate macroscopic anisotropies from microscopic parameters, and some numerical evidence for their validity.

In Sec. II, the macroscopic anisotropy is calculated from an expression for the frozen in first-order anisotropy (which is actually second order in the DM interaction constant, and comparable in magnitude to K_{DM}), and it is shown that the adjustment energy, which was used in the original Fert-Levy derivation, does not contribute to the macroscopic anisotropy, to lowest order in the anisotropy. However, both the frozen-in anisotropy and the adjustment energy contribute to the lowest order term in K_r. In Sec. III, these ideas are tested by investigating the effects of relaxation on realistic Ruderman-Kittel-Kasuya-Yosida

(RKKY) spin glasses with DM interactions by computer simulations, following the methods of Walker and Walstedt.[15] Analytic estimates of K_0 are calculated for two forms of the microscopic DM interaction, the validity of several approximations involved in these estimates are discussed, and comparisons with experiment are made in Sec. IV.

II. THEORY

For RKKY spin glasses with DM anisotropy, at temperatures well below the glass temperature T_G, any spin i is frozen (for short timescales) along the direction given by the sum of a large isotropic exchange field $\mathbf{H}_e^i$ and a small anisotropic field $\mathbf{H}_a^i$, given by

$$\mathbf{H}_e^i = \sum_{j=1}^{Z} J_{ij}\mathbf{S}_j \ , \tag{1a}$$

$$\mathbf{H}_a^i = -\sum_{j=1}^{Z} \mathbf{D}_{ij}\times\mathbf{S}_j \ , \tag{1b}$$

where the spins $\mathbf{S}_j$ are the Z interacting neighbors of spin i. For simplicity, we will assume the spins are classical spins of unit length in the following discussion. J_{ij} and $\mathbf{D}_{ij}$ are the RKKY and DM interaction constants, given in terms of the interaction parameters V_0, V_1, and Γ the Fermi wave vector k_F, the radii between spins i and j and between each spin j and each spin-orbit scatterer t, $\mathbf{R}_{ij}$ and $\mathbf{R}_{jt}$, and the phase angle ϕ, which depends on the number of electrons in the d shell of the spins Z_d:[12]

$$J_{ij} = V_0\cos(2k_F R_{ij})/R_{ij}^3 \ , \tag{2a}$$

$$\mathbf{D}_{ij} = \sum_t \frac{V_1\sin[(1+\gamma)\phi]\sin[k_F(R_{ij}+R_{it}+R_{jt})+\phi](\hat{R}_{it}\cdot\hat{R}_{jt})(\hat{R}_{it}\times\hat{R}_{jt})}{(1+\gamma)R_{ij}R_{it}R_{jt}\sin\phi} \ , \tag{2b}$$

$$\gamma = \Gamma(R_{ij}+R_{it}+R_{jt}) \ , \tag{2c}$$

$$\phi = \pi Z_d/10 \ . \tag{2d}$$

Unit vectors are written as $\hat{\mathbf{v}}$, where $\hat{\mathbf{v}}=\mathbf{v}/v$, and $v=|\mathbf{v}|$.

At temperatures well below T_G, and for anisotropies which are considerably weaker than the isotropic coupling, rotation of the spin system by a small angle involves only small deviations from a rigid rotation.[16] In this case, we may assume that each spin i is parallel to $\mathbf{H}_e^i+\mathbf{H}_a^i$. Since $|\mathbf{H}_e^i| \gg |\mathbf{H}_a^i|$, the local-spin configuration does not differ strongly from what it would be in the absence of anisotropy. Let us define the fields which would exist in the limit as the anisotropy goes to zero as

$$\mathbf{h}_e^i = \lim_{\{\mathbf{h}_a\}\to 0} \mathbf{H}_e^i = \sum_j J_{ij}\hat{\mathbf{h}}_e^j \ , \tag{1a'}$$

$$\mathbf{h}_a^i = -\sum_j \mathbf{D}_{ij}\times\hat{\mathbf{h}}_e^j \ . \tag{1b'}$$

The part of $\mathbf{h}_a^i$ which is perpendicular to $\mathbf{h}_e^i$ causes spin i to rotate slightly away from the direction of $\mathbf{h}_e^i$ by an amount $\Delta\mathbf{S}_i = \mathbf{h}_{a,\perp}^i/h_e^i$, for unit spins, to lowest order in h_a/h_e. The direction of $\mathbf{S}_i$ is then determined by $\mathbf{h}_{a,\perp}^i$ and $\mathbf{h}_e^i$. Since $\mathbf{h}_e^i$ rotates rigidly along with the spin system, any deviations from a rigid rotation result only from the $\{\mathbf{h}_{a,\perp}^i\}$, which do not rotate rigidly to follow the spin system, but are determined by the orientation of the lattice as well as that of the spin system.

During zero-field cooling, a spin glass freezing in a configuration α, specified by the $\{\hat{\mathbf{h}}_e\}$, will choose the orientation that minimizes the total energy, which is $(-\frac{1}{2})$ times the sum of the spins,

$$\mathbf{S}_i = \{1-[(\Delta S_i)^2/2]\}\hat{\mathbf{h}}_e^i + \Delta S_i\hat{\mathbf{h}}_{a,\perp}^i \ ,$$

dotted into their fields, $\mathbf{h}_e^i+\mathbf{h}_a^i$. We let this minimum energy orientation correspond to $\theta=0$, where θ is the angle of rotation of the spin system away from the minimum energy orientation. Both $\Delta\mathbf{S}_i$ and the fields $\mathbf{h}_e^i$ and $\mathbf{h}_a^i$ will vary as the spin system is rotated to a different orientation, θ. We assume that the $\{\hat{\mathbf{h}}_e\}$ are rotated rigidly—i.e., that the configuration α is rotated but the spin system does not hop into another low-energy configuration specified by a different set of isotropic fields $\{\hat{\mathbf{h}}'_e\}$. This assumption should be accurate for low-temperature experiments involving rotations by a small angle. This gives rise to a rigid rotation of the isotropic fields, $\mathbf{h}_e(0,\alpha)\to\mathbf{h}_e(\theta,\alpha)$. Note that this is not a rigid rotation of the whole spin system, but only a semirigid rotation. The new anisotropy fields $\{\mathbf{h}_a(\theta,\alpha)\}$ are not obtained by rigid rotation of the $\{\mathbf{h}_a(0,\alpha)\}$, as they depend on the orientation of both the lattice and the spin system. We assume that the individual spins in the rotated configuration are free to adjust individually to the new anisotropy fields: $\Delta\mathbf{S}_i(\theta,\alpha)=\mathbf{h}_a^i(\theta,\alpha)/h_e^i(\theta,\alpha)$.

Using the expression for the energy as $(-\frac{1}{2})$ times the sum of the dot products of the (unit-length) spins with their fields, we may express the energy to $O(h_a^2/h_e)$ for a particular configuration α and an orientation θ with respect to the zero-field cooled orientation as

$$E(\theta,\alpha) = -\tfrac{1}{2}\sum_{i,j}\left[[J_{ij}\hat{\mathbf{h}}_e^j(\theta,\alpha)-\mathbf{D}_{ij}\times\hat{\mathbf{h}}_e^j(\theta,\alpha)]\cdot\hat{\mathbf{h}}_e^i(\theta,\alpha)\left[1-\frac{|\Delta\mathbf{S}_i(\theta,\alpha)|^2}{2}\right]\left[1-\frac{|\Delta\mathbf{S}_j(\theta,\alpha)|^2}{2}\right]\right.$$
$$\left.-\mathbf{D}_{ij}\times\hat{\mathbf{h}}_e^j(\theta,\alpha)\cdot\Delta\mathbf{S}_i(\theta,\alpha)-\mathbf{D}_{ji}\times\hat{\mathbf{h}}_e^i(\theta,\alpha)\cdot\Delta\mathbf{S}_j(\theta,\alpha)\right] \tag{3a}$$

$$= -\tfrac{1}{2}\sum_{i,j}\{[J_{ij}\hat{\mathbf{h}}_e^j(\theta,\alpha) - \mathbf{D}_{ij}\times\hat{\mathbf{h}}_e^j(\theta,\alpha)]\cdot\hat{\mathbf{h}}_e^i(\theta,\alpha)[1-|\Delta\mathbf{S}_i(\theta,\alpha)|^2]$$

$$-2\mathbf{D}_{ij}\times\hat{\mathbf{h}}_e^j(\theta,\alpha)\cdot\Delta\mathbf{S}_i(\theta,\alpha)\} \tag{3b}$$

$$= -\tfrac{1}{2}\sum_i (h_e^i(\theta,\alpha)+h_{a,||}^i(\theta,\alpha)+\{[h_{a,\perp}^i(\theta,\alpha)]^2/h_e^i(\theta,\alpha)\}) , \tag{3c}$$

where $\mathbf{h}_{a,||}=(\mathbf{h}_a\cdot\hat{\mathbf{h}}_e)\hat{\mathbf{h}}_e$ and $\mathbf{h}_{a,\perp}=\mathbf{h}_a-\mathbf{h}_{a,||}$. Note that there are four contributions which enter the last term in Eq. (3c). The first two are due to the rotation of spin i toward $\mathbf{h}_{a,\perp}^i$ and away from $\mathbf{h}_e^i$, and the last two are due to changes in $\mathbf{h}_e^i$ and $\mathbf{h}_a^i$, resulting from all the other spins j rotating toward $\mathbf{h}_{a,\perp}^j$ and away from $\mathbf{h}_e^j$. The energy observed for a macroscopic sample rotated by an angle θ from the zero-field cooled orientation will be an average over configurations α, $E(\theta)=\langle E(\theta,\alpha)\rangle_\alpha$, where we write $\langle\ \rangle_\alpha$ for the average over all possible configurations, weighted with a Boltzmann probability distribution

$$P(\alpha)\sim\exp[-E(0,\alpha)/(k_BT_G)] . \tag{4}$$

Here we have assumed that the trapping in different configurations occurs close to T_G, and that the sample was cooled in zero field, so that the energy of each configuration during cooling would have been given by $E(0,\alpha)$.

We may define

$$E_e(\theta,\alpha)=-\tfrac{1}{2}\sum_i h_e^i(\theta,\alpha) ,$$
$$E_e(\theta)=\langle E_e(\theta,\alpha)\rangle_\alpha ; \tag{5a}$$

$$E_a^{(1)}(\theta,\alpha)=-\tfrac{1}{2}\sum_i h_{a,||}^i(\theta,\alpha) ,$$
$$E_a^{(1)}(\theta)=\langle E_a^{(1)}(\theta,\alpha)\rangle_\alpha ; \tag{5b}$$

$$E_a^{(2)}(\theta,\alpha)=-\tfrac{1}{2}\sum_i [h_{a,\perp}^i(\theta,\alpha)]^2/[h_e^i(\theta,\alpha)] ,$$
$$E_a^{(2)}(\theta)=\langle E_a^{(2)}(\theta,\alpha)\rangle_\alpha , \tag{5c}$$

so that

$$E(\theta)=E_e(\theta)+E_a^{(1)}(\theta)+E_a^{(2)}(\theta) .$$

We will call E_e the isotropic exchange energy, $E_a^{(1)}$ the first-order anisotropy energy, and $E_a^{(2)}$, which is due to relaxation of the spins toward $\mathbf{h}_{a,\perp}$ and away from $\mathbf{h}_e$, the second-order adjustment energy. The original Fert-Levy energy, which is related to the adjustment anisotropy by $E_{\rm DM}=3K_{\rm DM}/2$, was taken to be proportional to $E_a^{(2)}(0)$; the derivation of $K_{\rm DM}$ outlined below gives $E_{\rm DM}=2E_a^{(2)}(0)$.

For long-range interactions, such as RKKY and DM interactions, it is a reasonable first approximation to assume that the distribution of angles between interacting spins in the low-temperature state is random for the majority of spin pairs, and also that for small anisotropy the directions of spins i and j are uncorrelated with $\mathbf{D}_{ij}$, since the spin directions are predominantly determined by the random $\{J_{ij}\}$. These assumptions were previously used in calculating $E_{\rm DM}$.[11] There will be corrections to this picture due to the small correlation which develops between the spin pairs $\{\mathbf{S}_i,\mathbf{S}_j\}$ and the $\{\mathbf{D}_{ij}\}$ during the initial cooling. This frozen-in correlation leads to a nonzero value of $E_a^{(1)}$. There will also be corrections to the estimate of $E_a^{(2)}$ due to correlations between the spin pairs $\{\mathbf{S}_i,\mathbf{S}_j\}$ and the $\{\mathbf{D}_{ij}\}$, or equivalently, between the $\{\mathbf{h}_a\}$ and the $\{\mathbf{h}_e\}$. However, Sec. III will include evidence from computer simulations that the corrections to $E_a^{(2)}$ are small, and may be neglected.

We would expect corrections to $E_a^{(2)}$ due to preferential occupation of configurations with energetically favorable anisotropies to be higher order in h_a than the uncorrected value, by an extra factor of $[h_a/(k_BT_G)]\sim(h_a/h_e)$. The nonzero expectation value of $E_a^{(1)}$, due to preferential occupation of energetically favorable configurations, will also have an extra factor of (h_a/h_e), raising $E_a^{(1)}$ to be of $O(h_a^2/h_e)$. Let us keep terms of $O(h_a^2/h_e)$ and neglect terms of $O(h_a^3/h_e^2)$, such as the corrections to $E_a^{(2)}$. We note that if we neglect corrections to $E_a^{(2)}$, then $E_a^{(2)}(\theta)$ becomes independent of θ; in other words, $E_a^{(2)}$ does not contribute to the lowest order term in (h_a/h_e) of the macroscopic anisotropy. If we assume that the $\{J_{ij}\}$ and the $\{\mathbf{D}_{ij}\}$ are uncorrelated, which is a reasonable assumption for the DM interaction, which depends on the random placement of spin-orbit scatterers, $E_e(\theta)$ becomes independent of θ to $O(h_a^2/h_e)$, and does not contribute to the macroscopic anisotropy. This occurs because the reduction of the isotropic interactions due to the spins twisting away from the $\{\mathbf{h}_e^i\}$ and toward the $\{\mathbf{h}_{a,\perp}^i\}$ is the same to this order for different configurations: the same reason that $E_a^{(2)}$ does not contribute to the anisotropy in lowest order. The effect of E_e is then only to constrain the spins to freeze into a low-energy configuration which is locally similar to an isotropic equilibrium configuration, determined only by the isotropic interactions.

The lowest-order term in the macroscopic anisotropy is then given by

$$E_a^{(1)}(\theta)=-\left\langle\tfrac{1}{2}\sum_i h_{a,||}^i(\theta,\alpha)\right\rangle_\alpha .$$

Since the distribution of $\{h_e\}$ is uncorrelated with $h_{a,||}^i(\theta,\alpha)$ to $O(h_{a,||}^i(\theta,\alpha))$, to lowest order $\exp[-E_e(0)/(k_BT_G)]$ factors out of the average and we are left with

$$E_a^{(1)}(\theta)=\frac{-\sum_\alpha\left[\left[\sum_i h_{a,\|}^i(\theta,\alpha)\right]\exp\{-[E_a^{(1)}(0,\alpha)+E_a^{(2)}(0,\alpha)]/(k_BT_G)\}\right]}{2\sum_\alpha\exp\{-[E_a^{(1)}(0,\alpha)+E_a^{(2)}(0,\alpha)]/(k_BT_G)\}}\ . \tag{6}$$

Let us note here that when we factor out the dependence on E_e, we must convert the sum from a sum over all configurations to a sum over configurations which are locally similar to a low-energy isotropic equilibrium configuration.

Let us expand the exponential, keeping only terms up to $O(h_a/(k_BT_G))$, and so dropping $E_a^{(2)}(0,\alpha)$, to get

$$E_a^{(1)}(\theta)=\frac{-\sum_\alpha\left\{\left[\sum_i h_{a,\|}^i(\theta,\alpha)\right]\left[1+\left[\sum_i h_{a,\|}^i(0,\alpha)\right]/(k_BT_G)\right]\right\}}{2\sum_\alpha\exp\{-[E_a^{(1)}(0,\alpha)+E_a^{(2)}(0,\alpha)]/(k_BT_G)\}}\ . \tag{7}$$

Assuming the distributions of $\{J_{ij}\}$ and $\{\mathbf{D}_{ij}\}$ are uncorrelated, we have for the sum over all configurations α which are locally similar to isotropic equilibrium configurations

$$\sum_\alpha\sum_i h_{a,\|}^i(\theta,\alpha)=0\ ,$$

to $O(h_a^2/h_e)$, which leaves only one term in the numerator of Eq. (7). At this point, we may also expand the exponentials in the denominator, taking the lowest-order term, which is 1, for each of the N_α configurations under consideration. This gives

$$\begin{aligned}E_a^{(1)}(\theta)&=-\sum_\alpha\left[\left[\sum_i h_{a,\|}^i(\theta,\alpha)\right]\left[\sum_i h_{a,\|}^i(0,\alpha)\right]\right]\Big/(2N_\alpha k_BT_G)\\&=-\sum_\alpha\left[\sum_i[h_{a,\|}^i(\theta,\alpha)h_{a,\|}^i(0,\alpha)]\right]\Big/(2N_\alpha k_BT_G)\\&=-\sum_\alpha\left[\sum_i\sum_j[(\mathbf{D}_{ij}\times\mathbf{S}_j)\cdot\hat{\mathbf{w}}\,'][(\mathbf{D}_{ij}\times\mathbf{S}_j)\cdot\hat{\mathbf{w}}]\right]\Big/(2N_\alpha k_BT_G)\ ,\end{aligned} \tag{8}$$

where $\hat{\mathbf{w}}=\hat{\mathbf{h}}_e^{\,i}(0,\alpha)=(\hat{\mathbf{w}}\cdot\hat{\boldsymbol{\theta}})\hat{\boldsymbol{\theta}}+[1-(\hat{\mathbf{w}}\cdot\hat{\boldsymbol{\theta}})^2]^{1/2}\hat{\mathbf{x}}$ and $\hat{\mathbf{w}}\,'=(\hat{\mathbf{w}}\cdot\hat{\boldsymbol{\theta}})\hat{\boldsymbol{\theta}}+[1-(\hat{\mathbf{w}}\cdot\hat{\boldsymbol{\theta}})^2]^{1/2}[\hat{\mathbf{x}}\cos\theta+\hat{\mathbf{y}}\sin\theta]$, where $\hat{\mathbf{x}}$ is chosen to be parallel to $\hat{\mathbf{w}}-(\hat{\mathbf{w}}\cdot\hat{\boldsymbol{\theta}})\hat{\boldsymbol{\theta}}$ and $\hat{\mathbf{y}}=\hat{\boldsymbol{\theta}}\times\hat{\mathbf{x}}$. Assuming a random distribution of angles [to lowest order in (h_a/h_e)] between the $\{\mathbf{D}_{ij}\}$, $\{\mathbf{S}_j\}$, and $\{\hat{\mathbf{h}}_e^{\,i}\}$, and using random averages $\langle(\hat{\mathbf{v}}\cdot\hat{\mathbf{u}})^2\rangle=\frac{1}{3}$ and $\langle\,|\hat{\mathbf{v}}\times\hat{\mathbf{u}}\,|^2\rangle=\frac{2}{3}$ in three dimensions, we have

$$\begin{aligned}E_a^{(1)}(\theta)&=-\sum_\alpha\left[\sum_i\sum_j\{(\mathbf{D}_{ij}\times\mathbf{S}_j)_x^2[1-(\hat{\boldsymbol{\theta}}\cdot\hat{\mathbf{w}})^2]\cos\theta+(\mathbf{D}_{ij}\times\mathbf{S}_j)_\theta^2(\hat{\boldsymbol{\theta}}\cdot\hat{\mathbf{w}})^2\}\right]\Big/(2N_\alpha k_BT_G)\\&=-\left[\sum_{(ij)}|\,\mathbf{D}_{ij}\,|^2/(k_BT_G)\right]\left[\left(\tfrac{4}{27}\right)\cos\theta+\left(\tfrac{2}{27}\right)\right]\ ,\end{aligned} \tag{9}$$

where v_x and v_θ are the x and θ components of any vector $\mathbf{v}$, and (ij) means the sum over pairs. [Note that one factor of $\frac{1}{2}$ has disappeared when we converted from the double sum over spins i and j to the sum over spin pairs (ij).]

Writing the energy in terms of an anisotropy constant, we have $E(\theta)=-K_0\cos\theta+\text{const}$, where

$$K_0=\sum_{(ij)}[4\,|\,\mathbf{D}_{ij}\,|^2/(27k_BT_G)]\ . \tag{10}$$

If the spin system were to be rotated completely rigidly, then each spin would not be allowed to readjust under the influence of the new anisotropy fields $\mathbf{h}_a(\theta,\alpha)$, but would be forced to remain rotated slightly away from the rotated $\mathbf{h}_e(\theta,\alpha)$ and toward the anisotropy field corresponding to a rigid rotation of the anisotropy fields of the unrotated system $\mathbf{h}_a(0,\alpha)$. Such a completely rigid rotation, which does not allow for any readjustment of individual spins to their new fields, will give rise to an energy $E_r(\theta,\alpha)$, which may be derived in the same way as the energy $E(\theta,\alpha)$ for a semirigid rotation was derived in Eqs. (3a)—(3c). In the derivation of $E(\theta,\alpha)$, individual spins were allowed to adjust to the anisotropic fields in the rotated position, but the spin system was not allowed to jump to a different low-energy equilibrium configuration characterized by a different set of isotropic fields $\{\hat{\mathbf{h}}_e\}$.

In deriving the energy as a function of configuration α and rotation angle θ for a completely rigid rotation, it is easiest to treat the rotation as a rotation of the $\{\mathbf{D}_{ij}\}$, which are fixed to the lattice, while keeping the spin system fixed. So we will write the energy in terms of the DM interaction constants obtained by rotating the lattice

through an angle $-\theta$, $\{\mathbf{D}_{ij}(-\theta)\}$, rather than the interaction constants $\{\mathbf{D}_{ij}\}=\{\mathbf{D}_{ij}(0)\}$ for the unrotated lattice, used in Eqs. (3a)—(3c).

Writing the energy as $(-\frac{1}{2})$ times the sum of the dot products of the (unit-length) spins with their fields, we have, for a completely rigid rotation,

$$E_r(\theta,\alpha)=-\tfrac{1}{2}\sum_{ij}\left[[J_{ij}\hat{\mathbf{h}}_e^j(0,\alpha)-\mathbf{D}_{ij}(-\theta)\times\hat{\mathbf{h}}_e^j(0,\alpha)]\cdot\hat{\mathbf{h}}_e^i(0,\alpha)\left[1-\frac{|\Delta\mathbf{S}_i(0,\alpha)|^2}{2}\right]\left[1-\frac{|\Delta\mathbf{S}_j(0,\alpha)|^2}{2}\right]\right.$$
$$\left.-\mathbf{D}_{ij}(-\theta)\times\hat{\mathbf{h}}_e^j(0,\alpha)\cdot\Delta\mathbf{S}_i(0,\alpha)-\mathbf{D}_{ji}(-\theta)\times\hat{\mathbf{h}}_e^i(0,\alpha)\cdot\Delta\mathbf{S}_j(0,\alpha)\right]$$
$$=-\tfrac{1}{2}\sum_{ij}\{[J_{ij}\hat{\mathbf{h}}_e^j(0,\alpha)-\mathbf{D}_{ij}(-\theta)\times\hat{\mathbf{h}}_e^j(0,\alpha)]\cdot\hat{\mathbf{h}}_e^i(0,\alpha)[1-|\Delta\mathbf{S}_i(0,\alpha)|^2]-2\mathbf{D}_{ij}(-\theta)\times\hat{\mathbf{h}}_e^j(0,\alpha)\cdot\Delta\mathbf{S}_i(0,\alpha)\}\ . \quad (11)$$

Analysis similar to that above for the energy of a semirigid rotation, averaged over configurations α, $E(\theta)$, shows that the energy of a completely rigid rotation averaged over configurations may be expressed as

$$E_r(\theta)=E_{e,r}(\theta)+E_{a,r}^{(1)}(\theta)+E_{a,r}^{(2)}(\theta)\ .$$

The first term of Eq. (11) leads to similar results for completely rigid rotation as for semirigid rotation: $E_{e,r}(\theta)=E_e(\theta)$ and $E_{a,r}^{(1)}(\theta)=E_a^{(1)}(\theta)$. The second-order adjustment energy under rigid rotation $E_{a,r}^{(2)}(\theta)$ has two parts. The first part is due to the first term in Eq. (11). It is the constant contribution to the energy during the initial freezing due to the initial adjustment causing the spins to rotate slightly away from the isotropic fields. The second part is due to the second term in Eq. (11). This contribution to $E_{a,r}^{(2)}(\theta)$ depends on the correlation between the $\{\mathbf{h}_{a,\perp}(\theta,\alpha)\}$, after rotation of the $\{\mathbf{D}_{ij}\}$, and the original $\{\mathbf{h}_{a,\perp}(0,\alpha)\}$, toward which the spins have rotated slightly. We may write the adjustment energy as the sum of these two parts: $E_{a,r}^{(2)}(\theta)=X+Y$, where

$$X=\left\langle\sum_i |h_{a,\perp}^i(0,\alpha)|^2/[2h_e^i(0,\alpha)]\right\rangle_\alpha\ , \quad (11a)$$

$$Y=\left\langle-\sum_i\sum_j\left[\mathbf{D}_{ij}(-\theta)\times\hat{\mathbf{h}}_e^j(0,\alpha)\cdot\sum_k\{\mathbf{D}_{ik}(0)\times\hat{\mathbf{h}}_e^k(0,\alpha)-\hat{\mathbf{h}}_e^i(0,\alpha)[\mathbf{D}_{ik}(0)\times\hat{\mathbf{h}}_e^k(0,\alpha)]\cdot\hat{\mathbf{h}}_e^i(0,\alpha)\}/h_e^i(0,\alpha)\right]\right\rangle_\alpha\ . \quad (11b)$$

The first contribution to $E_{a,r}^{(2)}(\theta)$, x, may be calculated in the same way as was done above, in the calculation of the second order adjustment energy for a semirigid rotation, $E_a^{(2)}(\theta)$.

Let us define unit vectors $\hat{\mathbf{a}}_{ij}$ and $\hat{\mathbf{b}}_{ij}$ so that

$$\mathbf{D}_{ij}(-\theta)=D_{ij}(\sin\varphi_{ij}\cos\theta\hat{\mathbf{a}}_{ij}-\sin\varphi_{ij}\sin\theta\hat{\mathbf{b}}_{ij}+\cos\varphi_{ij}\hat{\theta})\ ,$$

where φ_{ij} is the angle between $\mathbf{D}_{ij}$ and $\hat{\theta}$. In calculating the second contribution to $E_{a,r}^{(2)}(\theta)$, Y, we note that to lowest order in (h_a/h_e), where the $\{\mathbf{D}_{ij}\}$ and the $\{\mathbf{h}_e\}$ are uncorrelated, averaging Eq. (11b) over a random distribution of $\{\mathbf{D}_{ij}\}$ leaves only the term proportional to D_{ij}^2, since the average of $D_{ij}D_{ik}$ over a random distribution is zero. This gives

$$Y=\left\langle-\sum_i\sum_j\{|\mathbf{D}_{ij}|^2(\sin\varphi_{ij}\cos\theta\hat{\mathbf{a}}_{ij}-\sin\varphi_{ij}\sin\theta\hat{\mathbf{b}}_{ij}+\cos\varphi_{ij}\hat{\theta})\right.$$
$$\left.\times\hat{\mathbf{h}}_e^j(0,\alpha)\cdot[(\sin\varphi_{ij}\hat{\mathbf{a}}_{ij}+\cos\varphi_{ij}\hat{\theta})\times\hat{\mathbf{h}}_e^j(0,\alpha)-\hat{\mathbf{h}}_e^i(0,\alpha)(\sin\varphi_{ij}\hat{\mathbf{a}}_{ij}+\cos\varphi_{ij}\hat{\theta})\times\hat{\mathbf{h}}_e^j(0,\alpha)\cdot\hat{\mathbf{h}}_e^i(0,\alpha)]/h_e^i(0,\alpha)\}\right\rangle_\alpha\ . \quad (11c)$$

If $\theta=0$ is the preferred orientation for the spin configuration to freeze into, as we have specified above, then the term proportional to $\sin\theta$ in Eq. (11c) must vanish, leaving only a constant term plus a term proportional to $\cos\theta$. Since $X=-E_a^{(2)}(0)$, we see that X is a constant independent of θ. So the configuration average of this second-order adjustment energy for rigid rotation may be written in terms of the Fert-Levy adjustment anisotropy as $E_{a,r}^{(2)}(\theta)=-K_{\mathrm{DM}}\cos\theta+\text{const}$, where

$$K_{\mathrm{DM}}=\left\langle\sum_i\sum_j(|\mathbf{D}_{ij}|^2[\sin\varphi_{ij}\hat{\mathbf{a}}_{ij}\times\hat{\mathbf{h}}_e^j(0,\alpha)]\cdot\{(\sin\varphi_{ij}\hat{\mathbf{a}}_{ij}+\cos\varphi_{ij}\hat{\theta})\times\hat{\mathbf{h}}_e^j(0,\alpha)\right.$$
$$\left.-\hat{\mathbf{h}}_e^i(0,\alpha)[\sin\varphi_{ij}\hat{\mathbf{a}}_{ij}+\cos\varphi_{ij}\hat{\theta})\times\hat{\mathbf{h}}_e^j(0,\alpha)]\cdot\hat{\mathbf{h}}_e^i(0,\alpha)\}/h_e^i(0,\alpha))\right\rangle_\alpha\ . \quad (12a)$$

Since θ and the $\{\mathbf{D}_{ij}\}$ are uncorrelated, and the $\{\mathbf{D}_{ij}\}$ and $\{\mathbf{h}_e\}$ are uncorrelated to lowest order in (h_a/h_e), the term proportional to $\sin\varphi_{ij}\cos\varphi_{ij}$ cancels out. $\langle\sin^2\varphi_{ij}\rangle=\frac{2}{3}$, and we use the same relation as above for the average cross product of uncorrelated unit vectors: $\langle|\hat{\mathbf{u}}\times\hat{\mathbf{v}}|^2\rangle=\frac{2}{3}$. This gives

$$K_{\rm DM}=\sum_{(ij)}(16|\mathbf{D}_{ij}|^2/27)\langle 1/h_e\rangle\ , \tag{12b}$$

where the sum has been converted into a sum over spin pairs (ij), instead of a double sum over spin i and spin j. This conversion gives rise to an additional factor of 2, as before. The total anisotropy constant for rigid rotation is $K_r=K_0+K_{\rm DM}$. We note that in calculating $E^{(2)}_{a,r}(\theta)$, $\langle 1/h_e\rangle$ should be taken over a random distribution of spins, and not over an actual distribution, including correlations, since h_e is used here as a measure of the resistance of the spin to rotating in the direction of $\mathbf{h}_{a,\perp}$. One should not include any effect that the spin has in polarizing its environment, since the spin should be equally effective in polarizing its environment in the rotated position. For this reason, any contribution to the exchange field defined by Eq. (1a′) due to correlations should not be included in the energy required to turn the spin. However, T_G may be estimated by taking an average of the fields over an actual equilibrium configuration, $\langle h_e\rangle_c \approx(3k_BT_G/s)=3k_BT_G$, for three-dimensional spins of unit length, since the glass temperature should occur when thermal fluctuations become comparable to the total energy, including correlations, of an equilibrium configuration. Here, we will use $\langle\ \rangle$ for averages over a random distribution of spins, and $\langle\ \rangle_c$ for averages over an actual equilibrium configuration, including correlations. According to Walker and Walstedt, $\langle 1/h_e\rangle_c \approx 2\langle 1/h_e\rangle/3$.[15] Using $\langle 1/h_e\rangle\approx\frac{3}{2}\langle 1/h_e\rangle_c \approx 3/\langle 2h_e\rangle_c \approx 1/(2k_BT_G)$, we get $K_r=K_{\rm DM}+K_0=3K_0$, or $K_0=0.33K_r$. In Ref. 16, $1/(3k_BT_G)$ was estimated as $\langle 1/h_e\rangle$, not $\langle 1/h_e\rangle_c$, giving $K_0=3K_{\rm DM}/4$, and $K_0=0.43K_r$, which was rounded up to $K_0=K_r/2$ by the argument that the temperature at which the freezing takes place is probably slightly lower than T_G, which would increase the magnitude of the frozen-in anisotropy. (Recent work by Sompolinsky, Kotliar, and Zippelius,[17] which assumed highly correlated isotropic and anisotropic interactions, suggests that the spin system is not as stiff as it seems to be from these arguments based on single spins in effective fields. Their calculation gives $K_0=0.2K_r$ for three dimensional spins.) In order to calculate K_0, we will estimate T_G using the distribution of correlated fields seen by Walker and Walstedt,[15]

$$1/(3k_BT_G)\approx\langle 1/h_e\rangle_c=2v_a/(\pi^2xV_0S)=\tfrac{2}{3}\langle 1/h_e\rangle\ , \tag{13}$$

where v_a is the volume per atom, and x is the fraction of atomic sites occupied by spins.

III. COMPUTER SIMULATIONS

Computer simulations of realistic RKKY spin glasses with DM anisotropy, following the methods of Walker and Walstedt,[15] have been used to investigate the total energy due to anisotropic interactions, which is related to K_r, and the effects of relaxation on the actual macroscopic anisotropy constant as measured by transverse susceptibility K, or by ESR, K', for low temperature equilibrium configurations. Samples of 96 and 204 spins, randomly substituted with a concentration of about 0.9% in an fcc lattice with the lattice constant of Cu, were used to simulate dilute CuMn. Spin-orbit scatterers were randomly substituted with concentrations of about 0.006—0.008 %. The parameters used for the RKKY interaction, V_0 and the Fermi wave vector k_F, were held fixed at values appropriate for CuMn. The parameters used for the DM interaction, V_1, Γ, and ϕ, were varied, and the extra oscillating factor in the DM interactions, $\sin[(1+\gamma)\phi]/\sin\phi$, was set to 1 in some cases, in order to vary the size of the anisotropy. Due to the periodic boundary conditions, to avoid multiple interactions, the range for the interactions had to be cut off for distances R_{ij}, R_{it}, or R_{jt} greater than half of the sample length. The magnitude, or effective range, of the anisotropy was also varied by sometimes allowing each spin pair to interact with only one spin-orbit scatterer, chosen randomly. The ratio of the anisotropic to the isotropic energy of the equilibrium configurations ranged from 0.0% to 1.25%. The equilibrium configurations were initially obtained in zero external field, but nevertheless have nonzero magnetization $\mathbf{M}$ before application of any measuring field, due to finite-size fluctuations.

We should remark here that small samples of 100 and even 200 spins have only a small number of isotropic equilibrium configurations,[15] and for small anisotropy, they freeze into a low-energy configuration closely corresponding to one or another of these isotropic configurations, depending on the initial configuration and the convergence algorithm. In order to see the true macroscopic frozen-in anisotropy, it would be necessary to take a sufficiently large sample to divide into many macroscopic regions, each with very many isotropic equilibrium configurations, and then cool the sample slowly enough through the glass temperature to allow time for preferential freezing into the configurations with the most favorable anisotropy energy. When small samples such as those studied here have frozen into one of their few available equilibrium configurations, which we may call configuration α, the frozen-in anisotropy $K_0(\alpha)$, which arises from the $\{\mathbf{h}_{a,||}\}$, and the adjustment anisotropy $K_{\rm DM}(\alpha)$, which arises from the $\{\mathbf{h}_{a,\perp}\}$, are simply due to the properties of configuration α, and may fluctuate strongly. Here we define $K_0(\alpha)=\frac{2}{3}E^{(1)}_a(0,\alpha)$, analogous to the macroscopic frozen-in anisotropy constant which is an average over all the macroscopic configurations: $K_0=\langle K_0(\alpha)\rangle_\alpha$. In these small samples, to lowest order in (h_a/h_e), the $\{\mathbf{h}_{a,||}\}$ are the same for α and its corresponding isotropic configuration, so $K_0(\alpha)$ was calculated from the $\{\mathbf{h}_{a,||}\}$ of the isotropic configuration.

Diagonalizing the matrix for the linearized equations of motion about the equilibrium configuration α, for each case studied, gave the frequencies and eigenvectors of the dynamic excitation modes, and diagonalizing the matrix of the quadratic form for the change in energy due to small displacements from equilibrium gave the static polarization modes needed to calculate the transverse and

longitudinal susceptibilities. As reported previously,[16] three dynamic resonance modes which are well described by the triad model[11] were observed for spin glasses with small-to-moderate anisotropy. The change in mode frequency as a function of measuring field was correctly predicted by the triad model, and a comparison of the magnetization and anisotropy constants obtained from a fit to the three identified ESR modes, $M'(\alpha)$ and $K'(\alpha)$, with the remanent magnetization calculated directly, $M(\alpha)$, and the anisotropy constant calculated from the transverse and longitudinal susceptibilities, $K(\alpha)$, showed good agreement.

The projection of these modes on uniform rotation modes, which is 1 for isotropic spin glasses, decreased as the anisotropy increased and relaxation became more important. However, when the ratio of anisotropic to isotropic energy was sufficiently increased so that these modes were less than 80% pure rotation, $K'(\alpha)$ and $K(\alpha)$ began to agree more and more poorly. In these cases, the part of the DM energy due to adjustment, $E_{\rm adj}(\alpha) = (H_{\rm DM})_\alpha - (H_{\rm DM})_{{\rm iso}(\alpha)}$, was 60–85% of the total DM energy $(H_{\rm DM})_\alpha$, where $(H_{\rm DM})$ is the expectation value of the DM Hamiltonian with a nonzero anisotropic interaction constant in the anisotropic equilibrium configuration α, or the corresponding isotropic equilibrium configuration iso(α), obtained by setting the DM interaction constant equal to zero and letting the configuration α relax, and then rotating the isotropic configuration to minimize $(H_{\rm DM})_{{\rm iso}(\alpha)}$.

For these large anisotropies, higher-order terms and statistical fluctuations in the adjustment energy begin to make $K'(\alpha)$ and $K(\alpha)$ deviate, independently, from being equal to the value expected for the configuration α due to the first-order anisotropy,

$$K_0(\alpha)=\tfrac{1}{3}\sum_i h^i_{a,\|}(0,\alpha)\ . \tag{14}$$

In this case, there is a nonzero contribution to the macroscopic anisotropy due to the relaxation effects which cause the sample to break up into domains instead of rotating as a whole. Therefore, calculations based on phenomenological models which assume an average, macroscopic angle of rotation (such as the triad model) no longer lead to an unambiguous determination of one anisotropy parameter, independent of whether it was determined from dynamic mode frequencies or static susceptibilities. In a macroscopic sample, the expectation value for these fluctuations in the adjustment energy should be zero to lowest order in h_a/h_e, while the expectation value for the frozen-in "first-order" anisotropy constant, $K_0 = \langle K_0(\alpha)\rangle_\alpha$, should be nonzero to the same order. [In Ref. 16, the distinction between quantities referring to one specific equilibrium configuration of a small sample, such as $K_0(\alpha)$, and quantities referring to the average over configurations of a macroscopic sample, such as $K_0=\langle K_0(\alpha)\rangle_\alpha$, was not made as clear as would have been desirable. Here an attempt has been made to write all parameters which refer to a single configuration rather than the average over configurations of a macroscopic sample as explicit functions of the configuration α: $K_0(\alpha)$, $M(\alpha)$, etc.]

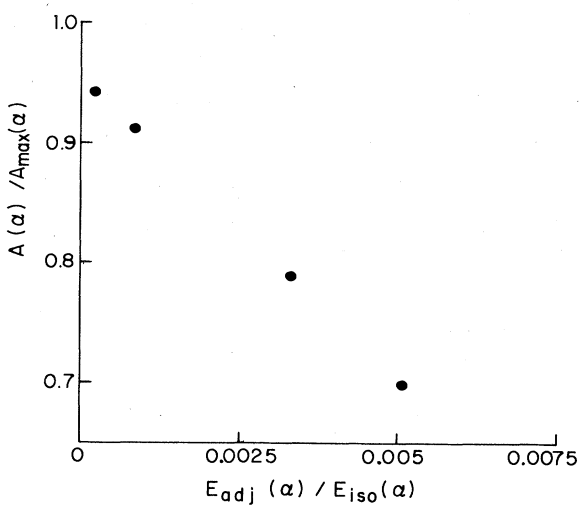

FIG. 1. Plot of the projection of the three identified modes onto rigid rotations $A(\alpha)/A_{\rm max}(\alpha)$, against the ratio of the anisotropic adjustment energy to the isotropic energy $E_{\rm adj}(\alpha)/E_{\rm iso}(\alpha)$, for one sample, varying the anisotropy constant V_1.

Figure 1 shows the decrease in the projections of the three triad modes on uniform rotations as h_a/h_e increases, increasing the fraction of the DM energy due to adjustment. The average angle of rotation for the eigenvector ϵ of configuration α, about axis $\hat{\mathbf{b}}$, $\hat{\mathbf{b}}=\hat{\mathbf{x}}$, $\hat{\mathbf{y}}$, or $\hat{\mathbf{z}}$, is defined as

$$\delta\theta_b(\alpha,\epsilon)=\sum_{i=1}^{N}\mathbf{S}_i^0(\alpha)\times d\mathbf{S}_i(\alpha,\epsilon)\cdot\hat{\mathbf{b}}/N\ , \tag{15}$$

where $d\mathbf{S}_i(\alpha,\epsilon)$ is the deviation from equilibrium of spin i for eigenvector ϵ, and $\mathbf{S}_i^0(\alpha)$ is the equilibrium position of spin i in the configuration α. Choosing $\hat{\mathbf{z}}\|\mathbf{M}$, we may define a longitudinal rotation angle $\delta\theta_L(\alpha,\epsilon)=\delta\theta_z(\alpha,\epsilon)$, and a transverse rotation angle

$$\delta\theta_T(\alpha,\epsilon)=[\,|\,\delta\theta_x(\alpha,\epsilon)\,|^2+|\,\delta\theta_y(\alpha,\epsilon)\,|^2]^{1/2}\ .$$

In all samples with small to moderate anisotropy, the mode with the largest $\delta\theta_L$ had a frequency in zero field between the frequencies of the two modes with largest $\delta\theta_T$, and these three modes were identified with the longitudinal and transverse modes of the triad model: ϵ_L, ϵ_+, and ϵ_-.[11] Adding up the average angles of rotation for the three identified modes, we define

$$A(\alpha)=\delta\theta_L(\alpha,\epsilon_L)+\delta\theta_T(\alpha,\epsilon_+)+\delta\theta_T(\alpha,\epsilon_-)\ .$$

We may normalize $A(\alpha)$ by dividing by $A_{\rm max}(\alpha)$, the value we would expect for a sample of the same size, assuming a uniform distribution of spins, if the three modes were composed of pure rotations about the different axes. $A(\alpha)/A_{\rm max}(\alpha)$, which measures the projections of the ESR modes on uniform rotations, is plotted in Fig. 1 against the ratio of the adjustment part of the DM energy to the isotropic energy,

$$E_{\rm adj}(\alpha)/E_{\rm iso}(\alpha)=[(H_{\rm DM})_\alpha-(H_{\rm DM})_{{\rm iso}(\alpha)}]/(H_{\rm RKKY})_{{\rm iso}(\alpha)}\ ,$$

for one particular configuration of one particular sample,

where only V_1 was varied to vary the magnitude of the anisotropy.

$A(\alpha)/A_{\max}(\alpha)$ decreases monotonically as relaxation becomes more important, and the adjustment part of the DM energy increases. But there is some variation in the slope of the curves for different samples, because of statistical fluctuations in the amount of second order relaxation from the isotropic equilibrium configuration seen in the unrotated position and the amount of relaxation seen in rotated positions. There is some variation in curvature, because of statistical fluctuations in higher-order terms. Deviations from a uniform distribution of spins, due to statistical fluctuations, have reduced the value of $A(\alpha)/A_{\max}(\alpha)$ extrapolated to the isotropic case to be less than 1. [This does not mean that the isotropic sample has a projection on uniform rotation modes of less than 1, only that the $A(\alpha)$ for the isotropic configuration, which has a slightly nonuniform distribution of spins due to statistical fluctuations, is a little less than the $A_{\max}(\alpha)$.]

The average of the measured anisotropy constants, $\tilde{K}(\alpha)=[K(\alpha)+K'(\alpha)]/2$, is plotted for 12 96-spin samples and 2 204-spin samples in Fig. 2, against $K_0(\alpha)$, where $K_0(\alpha)$ is taken to be the anisotropy corresponding to a rigid rotation of the isotropic configuration, iso(α). A line of unit slope is included for comparison. The points for small and moderate anisotropies show reasonably good agreement between $\tilde{K}(\alpha)$ and $K_0(\alpha)$. The two points for 204-spin samples are beginning to show a larger $\tilde{K}(\alpha)$ than the $K_0(\alpha)$ calculated from the $\{\mathbf{h}_{a,\|}\}$ of the isotropic configuration obtained by setting the anisotropy equal to zero and letting configuration α relax. This may be because these larger samples have isotropic equilibrium configurations spaced as closely as 0.0003 (reduced units)

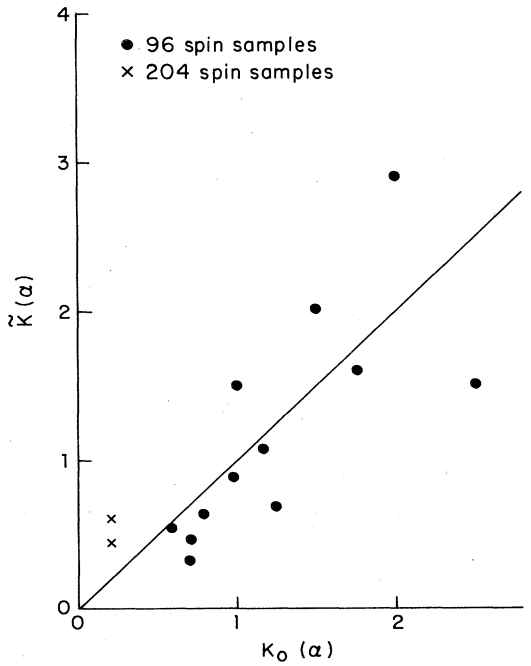

FIG. 2. Plot of the measured anisotropy constant $\tilde{K}(\alpha)$ vs the calculated "frozen-in" anisotropy constant $K_0(\alpha)$ [reduced units (r.u.)]. Line with unit slope is included for comparison.

in energy, while $(H_{\rm DM})_\alpha \approx 0.03$ (reduced units). Therefore, the anisotropy may already be mixing in enough of the other isotropic equilibrium configurations to increase the $\{\mathbf{h}_{\alpha,\|}\}$ in the anisotropic configuration α over those seen in iso(α). This effect is not as large as would be expected in a truly macroscopic sample, since the projection of α onto iso(α) is still $\sim 98\%$, and the projections of α onto the other isotropic configurations are not greatly changed from the projections of iso(α) onto these configurations. The points for the small samples with large anisotropies are beginning to deviate markedly from the line. As discussed above, this is because fluctuations in the relaxation and higher-order terms begin to dominate, for single configurations in finite samples with large anisotropies, over the frozen-in anisotropy which would be the only contribution to the anisotropy constant observed in macroscopic samples, to lowest order in the ratio of anisotropic to isotropic interaction strength.

Dasgupta and Yao have recently published a study[18] of the macroscopic anisotropy observed for large-angle deviations from the orientation of cooling. They also see unidirectional anisotropy well parametrized by one value of $K(\alpha)$ for samples with small anisotropies. For stronger anisotropies, where relaxation becomes more important and higher-order terms and statistical fluctuations in the adjustment energy should begin to dominate over $K_0(\alpha)$, they find complicated behavior which reproduces qualitatively some of the features of torque and hysteresis experiments. As observed experimentally, they find larger deviations from a simple unidirectional model when the deviation from the orientation of cooling is increased.

In our simulations, $K_0(\alpha)$ and $K_{\rm DM}(\alpha)$ scale as $(\sum_{ij} |\mathbf{D}_{ij}|^2)^{1/2}$ and $\sum_{ij} |\mathbf{D}_{ij}|^2$, respectively; i.e., $K_0(\alpha)$ is dominated in these samples by $N^{1/2}$ fluctuations, and $K_{\rm DM}(\alpha)$ goes as predicted for macroscopic samples in Sec. II. This behavior for $K_0(\alpha)$ occurs because $K_0(\alpha)$ arises from the sum of the (parallel) anisotropic fields for the isotropic configuration [denoted iso (α) in subscripts]: $K_0(\alpha)=\frac{1}{3}(\sum h^i_{\alpha,\|})_{\rm iso(\alpha)}$. For a small sample of N spins, $(\sum h^i_{\alpha,\|})_{\rm iso(\alpha)} \sim N^{1/2}$, due purely to statistical fluctuations. This is analogous to the freezing in of magnetic moments $M \sim N^{1/2}$ for small samples, even in the absence of an applied field $\mathbf{H}$, which would cause preferential freezing into configurations with a nonzero moment $\mathbf{M}||\mathbf{H}$. As h_a/h_e is increased, and for larger samples which have more low-energy equilibrium configurations, preferential freezing of the spin glass into configurations with favorable anisotropy energies becomes more important relative to the $K_0(\alpha) \sim N^{1/2}$ seen for a single low-energy configuration, due to statistical fluctuations. For the larger samples in this study,

$$K_0(\alpha)/K_r(\alpha)=K_0(\alpha)/[K_0(\alpha)+K_{\rm DM}(\alpha)] \approx 0.2\text{—}0.3 \ .$$

If we assume that due to the mixing in of different isotropic equilibrium configurations to configuration α, the true value of $K_0(\alpha)$ is not equal to the value which we have used, taken from the isotropic configuration iso(α), but rather equal to $\tilde{K}(\alpha)$, then we would have

$$K_0(\alpha)/K_r(\alpha)=K_0(\alpha)/[K_0(\alpha)+K_{\rm DM}(\alpha)] \approx 0.4\text{—}0.6$$

for the 204-spin samples. For the 96-spin samples with small anisotropies, the contributions to $K_r(\alpha)$ due to adjustment and to preferential freezing into favorable configurations were considerably less important than the $K_0(\alpha)\sim N^{1/2}$ due to statistical fluctuations, so that ratios of $K_0(\alpha)/K_r(\alpha)$ were seen up to 0.9. Even if we were to assume that the 204-spin samples are large enough to show macroscopic behavior, there is still sufficient fluctuation in these results that it is not possible to say whether K_0 is more nearly $0.33K_r$, as is given by Eqs. (4)—(6), or $0.2K_r$, as is given in Ref. 17.

Since the $K_0(\alpha)$'s we observe are dominated by $N^{1/2}$ fluctuations, we cannot predict the macroscopic value of K_0 directly from the simulations. In addition, the values of $K_{\rm DM}(\alpha)$ and $K_0(\alpha)$ derived from the simulations must be corrected to remove the cutoff introduced for $\sum_{ij}|\mathbf{D}_{ij}|^2$ because of the finite size of the samples. We defer to Sec. IV the discussion of the full estimate of $\sum_{ij}|\mathbf{D}_{ij}|^2$ with no cutoff, and the resulting estimate of K_0, using Eqs. (10) and (13), for a macroscopic sample of CuMnPt.

Before proceeding to the estimate of K_0 for a macroscopic sample, let us consider the results of the simulations for $K_{\rm DM}(\alpha)$. As we have used the assumption that the probability distributions for $\mathbf{S}_i$, $\mathbf{S}_j$, and $\mathbf{D}_{ij}$ are only weakly correlated in deriving Eq. (10), which we will use to estimate K_0, it would be a good idea to check the validity of this assumption in our estimate of the anisotropy due to adjustment, $K_{\rm DM}$. Figure 3 shows the Fert-Levy anisotropy constant per spin, $\bar{K}_{\rm DM}(\alpha)=K_{\rm DM}(\alpha)/N$, calculated from Eq. (12b), which assumes uncorrelated probability distributions, to lowest order, for $\mathbf{S}_i$, $\mathbf{S}_j$, and $\mathbf{D}_{ij}$. This gives

$$\bar{K}_{\rm DM}(\alpha)=16\langle 1/h_e\rangle\left[\sum_{(ij)}|\mathbf{D}_{ij}|^2\right]/(27N)\ . \tag{16}$$

$\bar{K}_{\rm DM}(\alpha)$ is plotted against measured anisotropy constant per spin under rigid rotation due to adjustment,

$$\bar{K}_{\rm adj}(\alpha)=2\bar{E}_{\rm adj}(\alpha)/3$$
$$=2[(H_{\rm DM})_\alpha-(H_{\rm DM})_{{\rm iso}(\alpha)}]/(3N)\ ,$$

where N is the total number of spins. A line with unit slope is included for comparison. The agreement between $\bar{K}_{\rm DM}(\alpha)$ and $\bar{K}_{\rm adj}(\alpha)$ is very good.

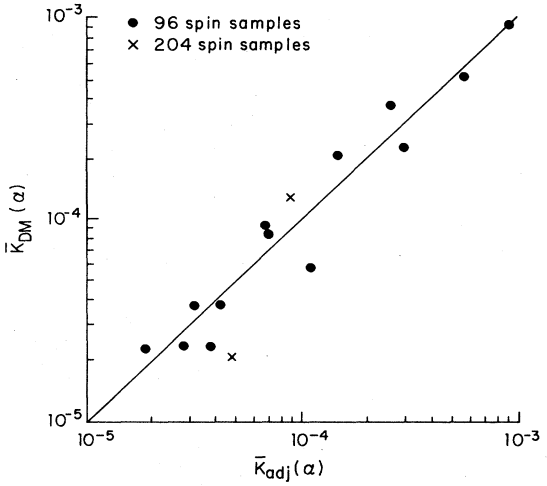

FIG. 3. Calculated anisotropy constant per spin due to adjustment, for a completely rigid rotation $\bar{K}_{\rm DM}(\alpha)$, plotted against the measured anisotropy constant per spin due to adjustment, for a completely rigid rotation $\bar{K}_{\rm adj}(\alpha)$ (r.u.). Line with unit slope is included for comparison.

IV. CALCULATION OF K_0

In order to estimate K_0 for a macroscopic sample, using Eq. (10) we will need an estimate of $\sum_{ij}|\mathbf{D}_{ij}|^2$. Using Equation (2b) we have, for each spin orbit scatterer t,

$$\sum_{(ij)}|\mathbf{D}_{ij,t}|^2=\sum_{(ij)}\frac{\{V_1\sin[(1+\gamma)\phi]\sin[k_F(R_{ij}+R_{it}+R_{jt})+\phi](\hat{\mathbf{R}}_{it}\cdot\hat{\mathbf{R}}_{jt})(\hat{\mathbf{R}}_{it}\times\hat{\mathbf{R}}_{jt})\}^2}{[(1+\gamma)R_{ij}R_{it}R_{jt}\sin\phi]^2}\ . \tag{17a}$$

Since k_Fa_0 is large, for a_0 the lattice constant and k_F the Fermi wave vector of copper, we may approximate $\sin^2[k_F(R_{ij}+R_{it}+R_{jt})+\phi]\approx\frac{1}{2}$, assuming that the argument fluctuates rapidly enough to be uncorrelated with the other factors.

For the analytic estimate, we may replace each sum over Mn sites by the concentration of Mn, $c_{\rm Mn}$, times an integral over volume, and the sum over spin-orbit scatterers (in this case, Pt) by $c_{\rm Pt}$ times another integral over volume. Doing first the sum over (ij), let us take $\mathbf{R}$ as the position of the more distant Mn relative to scatterer t, and $\mathbf{r}$ as the position of the closer Mn. In this case, $2R<(R_{ij}+R_{it}+R_{jt})\leq 4R$, so let us approximate $(R_{ij}+R_{it}+R_{jt})$ by aR, where a is a parameter between 2 and 4, so that $\gamma=\Gamma aR$. The integrals over all the angular variables except $x=\cos\theta$, where θ is the angle between $\mathbf{R}$ and $\mathbf{r}$, may be done immediately, leaving us with

$$\sum_{(ij)}|\mathbf{D}_{ij,t}|^2=4\pi^2V_1^2c_{\rm Mn}^2\int_0^\infty dR\int_0^R dr\int_{-1}^1 dx\frac{\sin^2[(1+\Gamma aR)\phi]x^2(1-x^2)}{(1+\Gamma aR)^2\sin^2\phi(R^2+r^2-2Rrx)}\ . \tag{17b}$$

Let us change to the dimensionless variables $s=r/R$, so that this becomes

$$\sum_{(ij)}|\mathbf{D}_{ij,t}|^2=\frac{4\pi^2V_1^2c_{\rm Mn}^2}{\sin^2\phi}\int_0^\infty dR\frac{\sin^2[(1+\Gamma aR)\phi]}{R(1+\Gamma aR)^2}\int_0^1 ds\int_{-1}^1 dx\frac{x^2(1-x^2)}{(1+s^2-2sx)}\ . \tag{18}$$

The integral over s gives

$$\int_0^1 ds\left[\frac{1}{1+s^2-2s\cos\theta}\right] = \frac{1}{\sin\theta}\left[\arctan\left[\frac{s-\cos\theta}{\sin\theta}\right]\right]_0^1 = \frac{1}{\sin\theta}\left[\arctan\left[\frac{\sin(\theta/2)}{\cos(\theta/2)}\right]-[\theta-(\pi/2)]\right] = \frac{\pi-\theta}{2\sin\theta} .$$

Substituting this into the integral over θ, we have

$$\int_0^\pi d\theta \sin^2\theta\cos^2\theta(\pi-\theta)/2 = (\pi/4)\int_0^\pi d\theta\sin^2\theta\cos^2\theta = \pi^2/32 . \quad (19)$$

This leaves us with an integral over R. We notice at this point that the sum will tend to diverge if we let $R\rightarrow 0$, and that physically, R must not be less than the nearest-neighbor distance on the lattice, $\tau=2.55$ Å for a Cu lattice. In order to check the approximation of replacing the sum over near neighbors by an integral over the continuum, we let the upper limit on R be a variable cutoff ν. (The result we will want to use to calculate K_0 will be the limit of $\sum_{ij}|\mathbf{D}_{ij,t}|^2$ as $\nu\rightarrow\infty$.) So we need to calculate

$$I(a,\nu)=\int_\tau^\nu dR\frac{\sin^2[(1+\Gamma aR)\phi]}{R(1+\Gamma aR)^2} . \quad (20)$$

Writing this out in partial fractions, we get

$$I(a,\nu)=I_R(a,\nu)-\Gamma aI_1(a,\nu)-\Gamma aI_2(a,\nu) , \quad (21)$$

$$I_R(a,\nu)=\int_\tau^\nu dR\frac{\sin^2[(1+\Gamma aR)\phi]}{R} , \quad (21a)$$

$$I_1(a,\nu)=\int_\tau^\nu dR\frac{\sin^2[(1+\Gamma aR)\phi]}{(1+\Gamma aR)} , \quad (21b)$$

$$I_2(a,\nu)=\int_\tau^\nu dR\frac{\sin^2[(1+\Gamma aR)\phi]}{(1+\Gamma aR)^2} \quad (21c)$$

These may be expressed in terms of sine and cosine integrals:

$$I_R(a,\nu)=\{[\ln(\phi\Gamma aR)/2]-[\cos(2\phi)/2]\mathrm{ci}(2\phi\Gamma aR)+[\sin(2\phi/2]\mathrm{si}(2\phi\Gamma aR\}_\tau^\nu , \quad (22a)$$

$$I_1(a,\nu)=([\ln(1+\Gamma aR)/(2\Gamma a)]-\{\mathrm{Ci}[2\phi(1+\Gamma aR)]/(2\Gamma a)\})_\tau^\nu , \quad (22b)$$

$$I_2(a,\nu)=\left[-\left[\frac{\sin^2[(1+\Gamma aR)\phi]}{\Gamma a(1+\Gamma aR)}\right]+[\phi/(\Gamma a)]\mathrm{si}[2\phi(1+\Gamma aR)]\right]_\tau^\nu . \quad (22c)$$

Putting these together, we have

$$I(a,\nu)=\left[[\ln(R)/2]-[\cos(2\phi)/2]\mathrm{ci}(2\phi\Gamma aR)+[\sin(2\phi)/2]\mathrm{si}(2\phi\Gamma aR)-[\ln(1+\Gamma aR)/2]+\{\mathrm{Ci}[2\phi(1+\Gamma aR)]/2\}+\left[\frac{\sin^2[(1+\Gamma aR)\phi]}{(1+\Gamma aR)}\right]-\phi\,\mathrm{si}[2\phi(1+\Gamma aR)]\right]_\tau^\nu . \quad (23)$$

Substituting these results into Eq. (18), we get

$$\sum_{(ij)}|\mathbf{D}_{ij,t}|^2=\frac{\pi^4V_1^2c_{\mathrm{Mn}}^2}{8\sin^2\phi}I(a,\nu) , \quad (24)$$

where the limit $\nu\rightarrow\infty$ is taken for the full sum out to all spins. Finite sums of $\sum|\mathbf{D}_{ij,t}|^2$, out to all neighbors within seven unit cells or less, calculated in this way and using $\phi=2.9531$, the value appropriate for Pt scatterers,[12] are strongly dependent on the value of a used, but a sum out to $9999\approx\infty$ unit cells gives the same value to within 0.2% over the whole range, $2\leq a\leq 4$. Figure 4 shows finite sums of $\sum|\mathbf{D}_{ij,t}|^2$, out to all neighbors within n unit cells, calculated from Eqs. (23) and (24) and using $\phi=2.9531$, plotted as a function of n for $a=2$, 3, and 4, and $n\leq 7$, together with the results of exact summations of $|\mathbf{D}_{ij,t}|^2$ over lattice sites out to n unit cells, using $\phi=2.9531$. Values for the finite sums calculated from Eqs. (23) and (24), which were derived assuming a continuum of possible spin sites, would be equal to the values for an exact summation of $\sum|\mathbf{D}_{ij,t}|^2$ over lattice sites for a value of a somewhere between 2 and 3. This is taken as evidence that the continuum approximation is reasonable, and can be used for the total sum out to infinite distance, where the results do not depend on the value used for a, and an exact summation is not possible.

Because Eq. (18) has a fluctuating factor $\sin^2[(1+\Gamma aR)\phi]$ in the numerator and a constant factor $\sin^2\phi$ in the denominator, $\sum|\mathbf{D}_{ij,t}|^2$ peaks strongly when $\phi\approx\pi$, i.e., when $Z_d\approx 10$, and is very dependent on Z_d in this region. Figure 5 shows a plot of $\sum_{ij}|\mathbf{D}_{ij,t}|^2$ as a function of ϕ, including all spins i,j out to 9999 unit cells, using $a=3$, in arbitrary units. The point marked by an x is the value assumed for Pt spin-orbit scatterers; other points are marked by dots.

In order to get the full sum of $|\mathbf{D}_{ij}|^2$ over all spin pairs and spin-orbit scatterers, we simply multiply the sum per spin orbit scatterer t by the concentration of Pt spin-orbit scatterers, c_{Pt}, and the total volume of the sample V. The macroscopic anisotropy will be given by Eq. (10), and the anisotropy per spin will be this value divided by $(c_{\mathrm{Mn}}V)$. Using Eqs. (10) and (13), the value of $\sum_{ij}|\mathbf{D}_{ij}|^2$ obtained for $\phi=2.9531$ and $V_1/V_0=0.2$, $v_A=(3.61\times10^{-8}\ \mathrm{cm})^3/4$, $S=2.5$, and $SV_0=1.02\times10^{-36}$ erg cm^3,[15] we estimate the macroscopic anisotropy per spin for Pt in CuMn with about 1% Mn as $\bar{K}_0=K_0/N=y(1.5\times10^{-13}$ erg), where y is the atomic concentration of Pt.[19] This may be compared with the experimental value, $\bar{K}_0=y(9.5\times10^{-16}$ erg).[2]

There is currently some question whether the oscillating

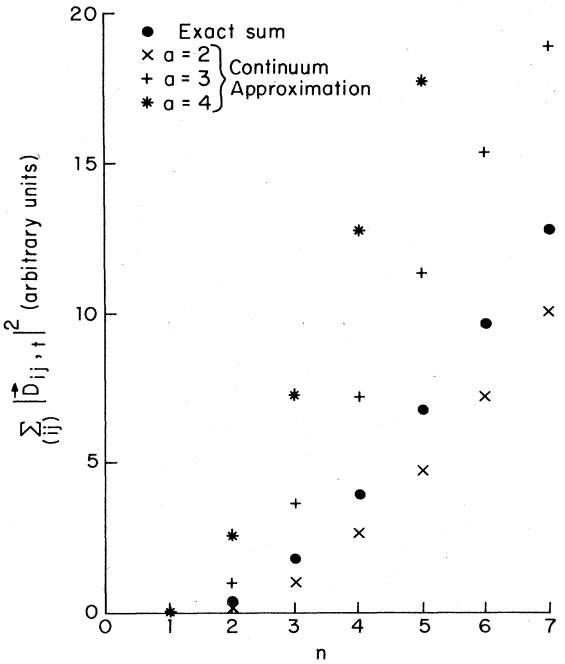

FIG. 4. Finite sums of the DM interaction strength over spins i and j, for spin orbit scatterer t, $\sum_{(ij)} |\mathbf{D}_{ij,t}|^2$, including spins out to n unit cells, plotted as a function of n, for the DM phase factor $\phi=2.9531$ [value for Pt (Ref. 12)].

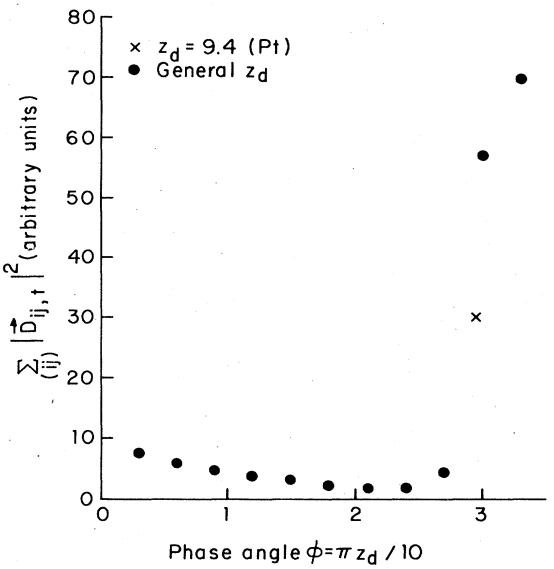

FIG. 5. Sum over spins i and j of the DM interaction strength due to spin-orbit scatterer t, $\sum_{(ij)} |\mathbf{D}_{ij,t}|^2$, as a function of the DM phase factor ϕ, using Eqs. (23) and (24).

factor $\sin^2[(1+\Gamma aR)\phi]$ is spurious, arising from an artificial cutoff in the microscopic calculation.[20] Since this factor strongly affects the results, as shown in Fig. 5, let us look at the magnitude of the anisotropy expected if the factor $\{\sin^2[(1+\Gamma aR)\phi]/\sin^2\phi\}$ is replaced by 1. In this case, the factor of $(1/\sin^2\phi)$ is removed from Eq. (24), and Eqs. (21a)—(21c) are replaced by Eqs. (25a)—(25c):

$$I(a,\nu)=I_R(a,\nu)-\Gamma aI_1(a,\nu)-\Gamma aI_2(a,\nu)\ , \tag{25}$$

$$I_R(a,\nu)=\int_\tau^\nu dR\frac{1}{R}\ , \tag{25a}$$

$$I_1(a,\nu)=\int_\tau^\nu dR\frac{1}{(1+\Gamma aR)}\ , \tag{25b}$$

$$I_2(a,\nu)=\int_\tau^\nu dR\frac{1}{(1+\Gamma aR)^2}\ . \tag{25c}$$

Solving the integrals gives

$$I(a,\nu)=\left[\ln R-[\ln(1+\Gamma aR)]+\left[\frac{1}{(1+\Gamma aR)}\right]\right]_\tau^\nu . \tag{26}$$

Using the new value for $I(a,\nu)$ as $\nu\to\infty$ given by Eq. (26), and removing the factor of $(1/\sin^2\phi)$ in Eq. (24), we get $\bar{K}_0=y(1.8\times10^{-14}$ erg), which is a factor of about 20 greater than the experimental value. If we had estimated $1/(3k_BT_G)=\langle 1/h_e\rangle$ instead of $\langle 1/h_e\rangle_c$, we would have gotten a result 30 times greater than the experimental value.

Clearly, estimates of the macroscopic anisotropy per spin vary strongly with the microscopic form of the interaction used to estimate $\sum|\mathbf{D}_{ij}|^2$. Both the latest published expression and that expression with a questionable oscillating factor removed yield macroscopic anisotropies much larger than the experimental value.

V. CONCLUSION

Current ideas distinguishing the "frozen-in" and the "adjustment" parts of the energy due to anisotropic interactions[13,14] are summarized, and the results of computer simulations of spin glasses with DM anisotropy are reported. These results are consistent with the identification of the macroscopic anisotropy measured from ESR or transverse susceptibility with a frozen-in first-order anisotropy. The measured adjustment energy due to anisotropic interactions is quantitatively predicted, using most of the assumptions used to estimate the frozen-in anisotropy which would be measured in a macroscopic sample. An expression for the macroscopic anisotropy for experimental samples is evaluated using two alternate forms of the microscopic DM interaction. This results in an estimate which ranges from 20 to 150 times greater than the experimental value for CuMnPt, depending on the parameters used.

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[19]The conversion from the reduced units used in Figs. 2 and 3 to ergs is 1 r.u. $=(2)^{3/2}(3.61\ \text{Å})^{-3}V_0S^2=1.533\times10^{-13}$ erg.

[20]S. Goldberg (private communication and unpublished).