Macroscopic anisotropy in transition-metal spin-glass alloys

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The nature of the macroscopic anisotropy in a classical Ruderman-Kittel-Kasuya-Yosida (RKKY) spin-glass with weak Dzyaloshinskii-Moriya interactions is studied in a zero-temperature numerical simulation. For rigid rotations of the spin system from a metastable state, we find the expected unidirectional form for the anisotropy energy. Simulations of rotation and inversion of the magnetization by a magnetic field show nearly rigid rotations of the spin system if the anisotropy energy is much smaller than the RKKY energy. For stronger anisotropy, we find substantial deviations from rigidity, especially for large-angle rotations and inversions, resulting in a behavior characteristic of an anisotropy energy with both unidirectional and uniaxial terms. The simulations qualitatively reproduce several features observed in hysteresis and torque experiments.

I. INTRODUCTION

The existence of a macroscopic anisotropy in transition-metal spin-glass alloys (such as Cu-Mn and Ag-Mn) at low temperatures has been observed in a large number of experiments.[1-9] Fert and Levy[10,11] traced the microscopic origin of this anisotropy to a Dzyaloshinskii-Moriya (DM) interaction arising from spin-orbit scattering of the conduction electrons. This mechanism explained the dramatic enhancement[3,5] of the anisotropy energy produced by the addition of nonmagnetic impurities (such as Au and Pt) with strong spin-orbit coupling. Subsequently, several authors[12-14] developed a semiphenomenological description of the nature of the macroscopic anisotropy produced by the microscopic DM interactions. These authors argue from general symmetry considerations that when a weak DM interaction is present, the increase in energy due to a rotation of the spin system from a frozen metastable state with zero or small remanent magnetization should be linear in the trace of the SO(3) rotation matrix that describes, in an average sense, the changes in the noncollinear spin directions. Since the trace of the rotation matrix is equal to $(1+2\cos\theta)$, where θ is the rotation angle, the increase in energy is expected to have the form

$$\Delta E = K(1-\cos\theta)\,, \tag{1}$$

with the anisotropy constant K *independent* of the direction of the rotation axis. The macroscopic anisotropy is thus unidirectional in the sense that ΔE is a 2π-periodic function of θ. However, since the anisotropy energy depends only on the rotation angle and not on the rotation axis, no special directions are picked out.

This picture has been successful in explaining several interesting experimental results on Cu-Mn alloys. There are, however, some other experimental observations which are in conflict with this description. Results of electron-spin-resonance (ESR) experiments[6] on field-cooled samples of Cu-Mn agree with hydrodynamic calculations[13-15] based on this picture if the angular displacement of the remanent magnetization from the direction of the cooling field is small. However, the agreement deteriorates rapidly for rotation angles larger than 90°. Recent measurements[9] of the restoring torque during rotations of the remanent magnetization by an external field in concentrated Cu-Mn are consistent with a purely unidirectional anisotropy. Measurements[8] of the transverse ac susceptibility of Cu-Mn alloys in a uniform field perpendicular to the direction of the cooling field indicate a purely unidirectional anisotropy for modest ($<35°$) angular displacements of the remanent magnetization, whereas transverse ac susceptibility measurements[7] after the remanent magnetization is inverted by a magnetic field clearly indicate the presence of a uniaxial (quadratic in $\cos\theta$) component of the anisotropy. Finally, in order to explain the experimentally observed shape of hysteresis cycles[1-3] of Mn-based spin-glass alloys, it is necessary to include both unidirectional and uniaxial terms in the anisotropy energy. Thus, it appears that the anisotropy energy has the form of Eq. (1) for small deviations from the initial state, whereas for large-angle deviations and inversions, a uniaxial term has to be included. At present, there is no clear understanding of the origin of this uniaxial term. The theoretical picture described above is expected to be valid only if the changes in the spin orientations produced by an external perturbation can be described as a uniform rotation in the macroscopic sense. It has been suggested[6,8,9] that the experimentally observed deviations from this picture are due to nonrigid rotations of the spin system.

In this paper we report the results of a numerical study of the zero-temperature anisotropy properties of a classical Ruderman-Kittel-Kasuya-Yosida (RKKY) spin-glass with weak DM interactions. The system studied here corresponds to $CuMn_xT_y$, where T represents a nonmagnetic impurity that mediates the DM interaction, and x and y denote concentrations. We have used the Walker-Walstedt procedure[16] to locate metastable equilibrium configurations (EC's) of the spin system, and studied the nature of the anisotropy for rigid rotations from these EC's. We have also investigated how a particular EC changes under the action of a uniform magnetic field that tends to rotate or invert the magnetization. The main re-

sults of this study are summarized below.

(a) A rigid rotation of the spin system through angle θ from an EC increases the energy by an amount proportional to $1-\cos\theta$ for all θ. The constant of proportionality is roughly independent of the direction of the axis of rotation. These results are in agreement with the theoretical predictions mentioned above. However, the calculated value of the anisotropy constant K for T=Pt turns out to be more than an order of magnitude larger than the experimental value.

(b) The evolution of an EC under the action of a magnetic field that tends to rotate or invert the magnetization is well described by a rigid rotation only if the anisotropy energy is much smaller than ($<0.1\%$) the RKKY energy. For larger anisotropies, the changes in the spin system show substantial deviations from a rigid rotation. The deviation from rigidity during a rotation of the magnetization increases with the rotation angle, and EC's corresponding to inversions of the magnetization show the largest amount of deviation from rigidity. The deviations from rigidity lead to a behavior characteristic of an anisotropy energy that has both unidirectional and uniaxial components. The qualitative behavior observed in the simulation of rotation and inversion processes is similar to the results of torque and hysteresis experiments.

Recently, Morgan-Pond[17] reported a numerical study similar to the one described in this paper. In Morgan-Pond's study, the emphasis is on a calculation of ESR frequencies in a field parallel to the remanent magnetization. This study thus probes the nature of the anisotropy for small deviations from the initial state, whereas in the present work we have concentrated on the behavior for large-angle deviations with a view towards understanding some of the results of hysteresis and torque experiments. Some of the general conclusions derived from our study were also obtained by Morgan-Pond.

This paper is organized as follows. Section II contains a definition of the model studied in this paper and a brief description of the numerical procedure employed. In Sec. III we present the results of our study of the nature of the anisotropy energy for rigid rotations. The simulations of rotation and inversion processes are described in Sec. IV. We present details of how the spin system behaves during (a) a rotation of the magnetization through 180°, (b) a subsequent rotation, through 180°, about an axis perpendicular to both the original direction of the magnetization and the axis of the first rotation, and (c) a hysteresis cycle in which a magnetic field applied in a direction opposite to the magnetization is first increased from zero to a value larger than that needed for a reversal of the magnetization, and then decreased back to zero. In Sec. V we summarize the main results and conclusions. Owing to the smallness of the sample size used in this simulation, some of the conclusions should be considered tentative.

II. MODEL AND NUMERICAL PROCEDURE

The model studied in this paper is appropriate for ternary spin-glass alloys $CuMn_xT_y$ where T represents a nonmagnetic impurity that mediates[18] the DM interaction through spin-orbit scattering. It consists of a system of classical Heisenberg spins interacting via isotropic RKKY and weak DM interactions. We consider a $L\times L\times L$ section of a fcc lattice with periodic boundary conditions. Spins are placed at $N_s=4xL^3$ randomly chosen sites and spin-orbit scatterers are placed at $N_i=4yL^3$ other sites, also chosen at random. The interaction Hamiltonian is a sum of two terms,

$$\mathscr{H}=\mathscr{H}_{\rm RKKY}+\mathscr{H}_{\rm DM}\ . \tag{2}$$

For the RKKY term we used the well-known form

$$\mathscr{H}_{\rm RKKY}=\sum_{i>j}\frac{A}{R_{ij}^3}\cos(2k_FR_{ij})\vec{\rm S}_i\cdot\vec{\rm S}_j\ , \tag{3}$$

where $\vec{\rm S}_i$ is the spin vector of unit length at site i and R_{ij} is the distance between sites i and j. For the lattice constant and the Fermi wave number k_F, we used values appropriate for Cu. Following Walker and Walstedt[16] we adopted a system of reduced units (r.u.) in which A/R_{ij}^3 is set equal to unity for i,j nearest neighbors. For the DM part we used the form derived by Fert and Levy,[10]

$$\mathscr{H}_{\rm DM}=-V\sum_{i>j}\sum_k\left[\sin\left[k_F(R_{ij}+R_{ik}+R_{jk})+\frac{\pi}{10}Z_d\right]\right]\frac{\vec{\rm R}_{ik}\cdot\vec{\rm R}_{jk}(\vec{\rm R}_{ik}\times\vec{\rm R}_{jk})\cdot(\vec{\rm S}_i\times\vec{\rm S}_j)}{R_{ik}^3R_{jk}^3R_{ij}}\ . \tag{4}$$

In Eq. (4), $\sum_k$ represents a sum over all sites occupied by the spin-orbit scattering impurity T, and Z_d is the number of d electrons in T. We took $Z_d=9.4$, a value appropriate for T=Pt, and treated V as an adjustable parameter. To avoid multiple interactions arising from periodic boundary condition, the interaction was set equal to zero if any one of the distances R_{ij}, R_{ik}, and R_{jk} was larger than $L/2$. In the presence of a uniform magnetic field $\vec{\rm h}$, a term $-\sum_i\vec{\rm h}\cdot\vec{\rm S}_i$ was added to the Hamiltonian.

The numerical work in this study consisted of generating metastable EC's of the spin system. An EC is a local minimum of the energy surface in phase space. In an EC the spin $\vec{\rm S}_i$ at each site i is parallel to the local field $\vec{\rm h}_i$ at that site, defined by

$$h_i^\alpha\equiv-\frac{\partial\mathscr{H}}{\partial S_i^\alpha},\quad\alpha=x,y,z\ . \tag{5}$$

The EC's were generated by using the algorithm of Walker and Walstedt.[16] In this procedure the spins are updated one at a time. In the update of the spin at site i, one first calculates the instantaneous local field $\vec{\rm h}_i$, and then sets $\vec{\rm S}_i$ parallel to $\vec{\rm h}_i$. An iteration is defined as a block of N_s consecutive updates. We used both fixed and random sequences of updates. It is clear that each update decreases the energy (unless one is already at an EC), and the system is not allowed to go over any energy barrier during its relaxation. Thus this procedure is similar to a zero-temperature Monte Carlo simulation. Convergence to an

EC was determined from the rate of change of the total energy. As the system closely approaches an EC, the energy decreases very slowly and it becomes difficult to distinguish the energy change in one iteration from computer round-off errors. For better accuracy we calculated the energy after every 200 iterations, and stopped the iterations when the fractional charge in energy in the last 200 iterations was less that a convergence parameter δ, taken to be 10^{-6} in most of our calculations. Except in one situation mentioned in Sec. IV, decreasing δ to 10^{-7} did not produce any noticeable change in the EC. As a further check of convergence, we calculated for each EC the quantity

$$\Delta=\frac{1}{N_s}\sum_i\left[1-\frac{\vec{h}_i\cdot\vec{S}_i}{h_i}\right], \tag{6}$$

which is a measure of the average deviation of a spin from the direction of its local field. The value of Δ was less than 10^{-7} for all EC's obtained with $\delta=10^{-6}$.

III. ANISOTROPY FOR RIGID ROTATIONS

The results reported in this section were obtained from simulations of ten samples, each containing 161 spins and 32 spin-orbit scatterers, distributed in a 20^3 lattice with concentrations $x=0.5$ at.% and $y=0.1$ at.%. For the parameter V appearing in the DM interaction [Eq. (4)], we used the value expected[10] for $T=$Pt ($V/A=0.2$). Starting from a random configuration the spins were updated sequentially until convergence to an EC was obtained. Typically, several thousand iterations were needed for convergence. The energies of the EC's showed variations of $\sim$20% from sample to sample, with an average value of -0.06 r.u. per spin. This value is consistent with the results obtained in other simulations[16,19] of RKKY systems. Although no magnetic field was applied during the relaxation process, the EC's had a magnetization of order $1/\sqrt{N_s}$, arising from finite-size fluctuations.

After having located an EC, we calculated the energies of configurations obtained by rigid rotations of the spins from the EC. For each sample, we studied rotations about six difference axes: one parallel to the magnetization, two in the plane perpendicular to the magnetization, and the remaining three along the principal axes of the lattice. In all cases we found that the increase in energy per spin due to a rigid rotation through angle θ is well described by the form

$$\Delta E=K_r(1-\cos\theta) \tag{7}$$

for all values of θ. A typical set of ΔE-vs-$1-\cos\theta$ plots is shown in Fig. 1. For a particular EC the values of K_r obtained from rotations about different axes showed a variation of $\sim$15%. Sample-to-sample variations were also of the same order. We also calculated the energies of configurations obtained by applying a sequence of two rotations to an EC, the first one through 180° and the second one through angle θ about an axis perpendicular to the axis of the first rotation. It is easy to show[9] that the rotation angle associated with the rotation matrix that describes the net rotation of the spin system is equal to 180° for all values of θ. Thus, if the anisotropy energy depends only on the rotation angle, then the energy of the configuration obtained in this way should be independent of θ. As shown in Fig. 1, the calculated value of the energy was found to be nearly constant during the second rotation, with small variations arising from the fact that K_r is not completely independent of the direction of the rotation axis in a finite system. We also carried out similar studies of several smaller samples (60—100 spins) with larger concentrations of Pt. In all cases, we found good agreement with Eq. (7). The calculated values of K_r were consistent (within rather large error bars) with a linear increase of the anisotropy energy with the concentration of Pt. All the numerical results for rigid rotations are thus in good agreement with the theoretical prediction about the nature of the macroscopic anisotropy.

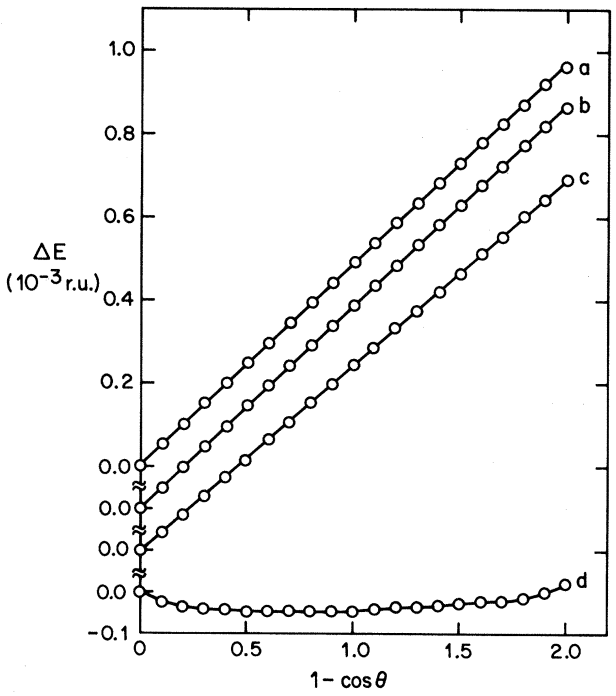

FIG. 1. Variation of energy with rotation angle for rigid rotations. (a) Rotation about the magnetization, (b) and (c) rotations about two mutually perpendicular axes perpendicular to the magnetization, and (d) rotation about the axis of (c) after a rotation of 180° about the axis of (b). r.u. represents reduced units.

The calculated value of the anisotropy constant, however, turned out to be much larger than the experimentally observed value. Averaging the results obtained for the ten 161-spin samples, we obtained $K_r=(4.5\pm0.3)\times10^{-4}$ r.u. per spin, which corresponds to $\sim2.7\times10^{-17}$ erg per spin if we use the value of the parameter A [Eq. (3)] given in Ref. 16 to convert reduced units to ergs.[20] In Ref. 11 the experimental value of the anisotropy constant per spin for 0.1 at.% Pt in Cu-Mn was estimated to be $\sim9.5\times10^{-19}$ erg. Thus the value of the anisotropy constant obtained from the simulation is about 30 times larger than the experimental value. There are several factors which indicate that the calculated anisotropy constant should be larger than the experimental value. First, when the spin system

is rotated from a metastable state by applying an external perturbation, the changes in the individual spin orientations do not correspond to a perfectly rigid rotation. In addition to the average rotation, the spins make readjustments to the changes in the local anisotropy field. These readjustments reduce the energy cost associated with the rotation, leading to an anisotropy constant that is smaller than the rigid-rotation value. The effects of these readjustments are studied in detail in the next section. Second, thermal relaxations which are always present in a real experiment but which are not allowed in our simulation also tend to reduce the value of the anisotropy constant. A decrease of the anisotropy constant with increasing temperature has been observed in experiments. Third, as pointed out by Morgan-Pond,[17] the anisotropy constant calculated from simulations of small systems contains a contribution coming from the fact that the DM energy of an isotropic (RKKY only) EC is, in general, nonzero in a small system. To estimate the size of this contribution, we calculated how the DM energy of the isotropic EC closest in phase space to an anisotropic EC changes under rigid rotations. We first located an isotropic EC by applying the relaxation algorithm to an anisotropic EC, with the DM interaction turned off. The resulting configuration was then rotated by a SO(3) matrix chosen to maximize the overlap of the rotated configuration with the anisotropic EC. The DM energy of the isotropic EC's obtained in this way was found to be negative in all cases. Rigid rotations increased this energy by amounts which were ~ 20—30% of the energy increases measured for the anisotropic EC. If this contribution is subtracted off, the calculated value of K_r would be reduced by $\sim 25\%$. Fourth, we note that the experimental value of the anisotropy constant quoted in Ref. 11 was obtained from hysteresis data. Anisotropy energies derived from the hysteresis loop are known to be somewhat smaller than those derived from experiments involving rotations of the magnetization. Finally, the expression for the DM interaction [Eq. (4)] used in this simulation did not include a correction term introduced in later work of Levy and Fert.[11] This term is expected to produce a small reduction in the calculated value of K_r. All these factors indicate that the value of the anisotropy constant obtained in our calculations should be larger than the experimental value. However, it is not clear whether these effects would be sufficient to account for the observed discrepancy of more than an order of magnitude.

The theoretical estimate of Levy and Fert[11] and the numerical work of Morgan-Pond[17] also showed a discrepancy of a similar magnitude between calculated and experimental values of the anisotropy constant. In Ref. 17, K_r was estimated from the relation

$$K_r = \tfrac{2}{3} E_{\mathrm{DM}} , \tag{8}$$

where E_{DM} represents the DM energy per spin in an anisotropic EC. In our calculations, the value of K_r obtained from this formula agreed to within 5% with the value obtained from the rigid-rotation data. However, we did not find agreement with the prediction made in Ref. 17 that E^0_{DM}, the DM energy of the isotropic EC corresponding to the anisotropic EC under consideration, should be half of E_{DM}. In our calculations, the ratio $E^0_{\mathrm{DM}}/E_{\mathrm{DM}}$ had values in the range 0.2—0.4, the smaller values being obtained for larger samples.

IV. SIMULATION OF ROTATION AND INVERSION

The results obtained in the preceding section clearly show that the macroscopic anisotropy in a RKKY spin glass with DM interactions is purely unidirectional for rigid rotations. Therefore, the origin of any departure from unidirectional behavior in this system must lie in nonrigid rotations of the spin system. We mentioned earlier that a rotation or reversal of the magnetization by an applied field does not correspond to a purely rigid rotation in the microscopic scale. Thus, it is important to understand under what circumstances the macroscopic behavior of the system is well decribed by a suitably averaged rigid rotation. Another important question is how the nature of the macroscopic anisotropy is affected by deviations from rigidity. With a view towards gaining some insight into these questions, we have simulated the response of the system to external magnetic fields which tend to rotate or invert the magnetization. The necessity of having to locate EC's for many different values and orientations of the magnetic field restricted our study to small samples. In these simulations, we used parameters different from those described in the preceding section. For samples with the parameters described in the preceding section, one would need unphysically large fields to produce a large-angle rotation or reversal of the magnetization. The simulations were done on ten samples divided into two groups. Each sample contained 81 spins and 20 spin-orbit scatterers distributed in a 10^3 fcc lattice, with concentrations $x=2$ at.% and $y=0.5$ at.%. The positions of the spins and the nonmagnetic impurities were the same in the two groups. The parameters V appearing in Eq. (4) was adjusted between $0.02A$ and $0.1A$ to give $K_r \simeq 6\times 10^{-5}$ r.u. per spin for the samples of the first group, and $K_r \simeq 3\times 10^{-4}$ r.u. per spin for those in the second group. The ratio of the DM energy (E_{DM}) to the RKKY energy (E_{RKKY}) was $\sim 1/2000$ in the first group and $\sim 1/400$ in the second group. The results obtained from the simulations are described below. Since the numerical procedure used to locate EC's did not allow transitions over energy barriers, the simulations correspond to situations in which the temperature and the time scale are such that thermally activated processes are effectively frozen out.

A. Rotations

For each sample, we first located an EC in zero field,[21] and then calculated the values of K_r for rigid rotations about different axes. We then applied a magnetic field $\vec{h}$ parallel to the magnetization $\vec{M}^0_r$, and located the new EC. The value of h was taken to be somewhat larger than K_r/M^0_r. The reversible susceptibility χ was calculated from the increase in the magnetization. The values of χ obtained in this way were found to be consistent with other calculations.[16] The field was then rotated by 180° in steps of 30° about the axis perpendicular to $\vec{M}^0_r$ for which the value of K_r was the smallest. We call this axis the x axis, and $\vec{M}^0_r$ defines the z axis. At each step the new EC

was located using the EC obtained at the previous step as the starting point. We used a random sequence of spin updates in the relaxation. Test runs with different random sequences were found to converge to the same EC. For the samples of the first group, we also studied the behavior during a subsequent rotation of the magnetic field about an axis (the y axis) perpendicular to both $\vec{M}_r^0$ and the axis of the first rotation. Before describing the results, we briefly discuss the behavior expected for rigid rotations and a purely unidirectional anisotropy. During the first rotation, the equilibrium magnetization per spin, $\vec{M}$, is expected[14] to be given by the relation

$$\vec{M}=\chi\vec{h}+\overleftrightarrow{R}\cdot\vec{M}_r^0 , \tag{9}$$

where the matrix $\overleftrightarrow{R}$ describes a rotation about the x axis by an angle θ which is obtained by balancing the anisotropy torque with the torque due to the magnetic field,

$$K\sin\theta=hM_r^0\sin(\theta_h-\theta) . \tag{10}$$

Here θ_h represents the angle between $\vec{M}_r^0$ and $\vec{h}$, and K is the macroscopic anisotropy constant per spin. The behavior expected during the second rotation has been analyzed by Fert and Hippert.[9] If the system rotates about the y axis from the state obtained by a π rotation of the initial state about the x axis, then there should be no anisotropy torque and $\vec{M}$ should remain parallel to $\vec{h}$. However, it is easy to show that a rotation about $\vec{M}$ would reduce the anisotropy energy of the state obtained in this way. Since such a rotation does not cost any magnetic energy, the system can lower the total energy by rotating about $\vec{M}$, ultimately reaching a state that corresponds to a rotation of the initial state about the y axis. In a recent experiment on concentrated samples of Cu-Mn, Fert and Hippert[9] found that the torque is essentially zero for small-angle rotations ($<10°$), whereas for larger angles they found a torque corresponds to a rotation of the original state about the y axis.

The results of our numerical experiments on the samples of the first group are in good agreement with these predictions. In Fig. 2 we have shown the variation of θ_M (the angle between $\vec{M}$ and $\vec{M}_r^0$), with θ_h during a rotation of one of the samples about the x axis. The magnetic field used to rotate the magnetization was 1.2×10^{-3} r.u., which corresponds to ~4 kG. The solid line represents the best fit to Eqs. (9) and (10) using the "experimental" values of χ (equal to 12.8 r.u./spin) and M_r^0 (equal to 0.054), and treating K as an adjustable parameter. It is clear that a purely unidirectional anisotropy fits the data very well. The value of the anisotropy constant obtained from the best fit ($K=5.4\times10^{-5}$ r.u./spin) was $\sim15\%$ smaller than the value obtained from a rigid rotation about the same axis. As mentioned in the preceding section, small readjustments of the spins to the changes in the local anisotropy field are expected to produce such a reduction in the value of the anisotropy constant. Similar behavior was observed in all other samples in this group.

Simulations of rotations about the y axis after a π rotation about the x axis showed behavior similar to that observed in the torque experiment of Fert and Hippert[9] on concentrated Cu-Mn samples. In Fig. 3 we have shown the results for the same sample as in Fig. 2. Here, θ_M and θ_h are measured from the $-\vec{M}_r^0$ direction. For $\theta_h\leq60°$, the magnetization remained nearly parallel to $\vec{h}$. For larger angles θ_M jumped to a value appropriate for a rotation of the initial state about the y axis. This changeover can be seen more clearly in the inset where we have shown the variation of the angle θ_R associated with the SO(3) rotation matrix $\overleftrightarrow{R}$ that maximizes the projection,

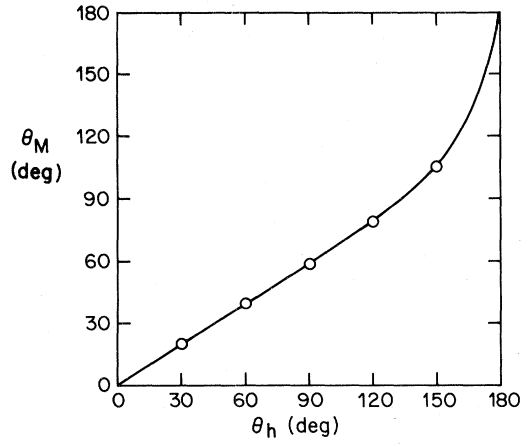

FIG. 2. Variation of θ_M, the angle of the total magnetization with θ_h, the angle of the applied magnetic field during a rotation about the x axis. The sample belongs to the first group (weak anisotropy). The solid line is a fit to Eqs. (9) and (10).

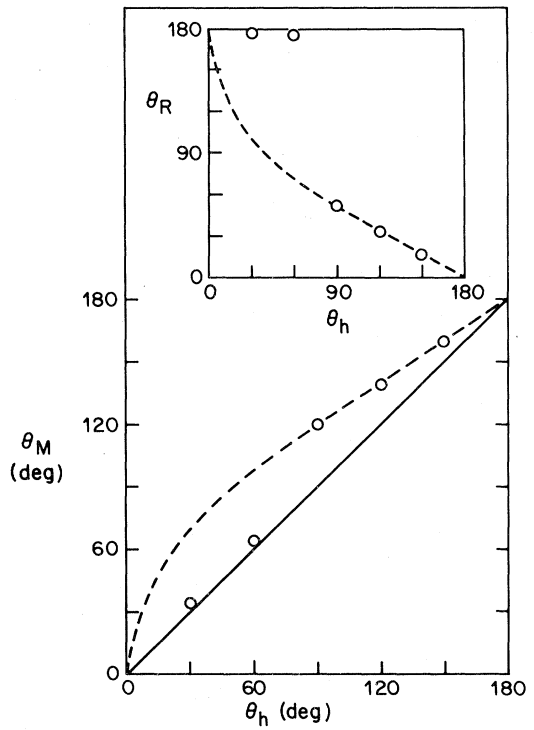

FIG. 3. Variation of θ_M with θ_h during a rotation about the y axis after a rotation of 180° about the x axis. The angles θ_M and θ_h are measured from the $-\hat{z}$ direction. The sample is the same as in Fig. 2. The solid line corresponds to $\theta_M=\theta_h$. The inset shows the variation of θ_R (see text) with θ_h. The dotted lines represent the expected behavior for a rotation of the initial state about the y axis.

$$P=\frac{1}{N_s}\sum_i \vec{S}_i'\cdot(\vec{R}\cdot\vec{S}_i)\,, \tag{11}$$

where $\{\vec{S}_i\}$ and $\{\vec{S}_i'\}$ represent the initial and new EC's, respectively. The angle θ_R remained nearly constant at $\sim 180°$ for $\theta_h \leq 60°$, and then changed over to a value corresponding to a rotation of the initial state about the y axis. When θ_h was increased from 60° to 90°, the system first rotated about the y axis until $\vec{M}$ was nearly parallel to $\vec{h}$, then rotated by $\sim 180°$ about $\vec{h}$, and finally rotated about the y axis to reach the equilibrium configuration. This process was very slow, requiring more than 30 000 iterations for convergence. The stabilization of the π-rotated state against a rotation about $\vec{M}$ for small values of θ_h was partly due to small readjustments of the spins to the new anisotropy fields. Since the first rotation was made about the axis with the smallest K_r, small variations of the anisotropy constant with the direction of the rotation axis also tended to stabilize the π-rotated state. This finite-size effect is probably the reason why the value of θ_h at which the π-rotated state became unstable was somewhat larger than the experimentally observed value. The changes in the spin orientations during both the first and the second rotation were well described by rigid rotations, with values of $P_{\max}$, the maximum value of the projection defined in Eq. (11), close to 0.99 in all cases.

The results for the samples in the second group showed some deviations from purely unidirectional behavior. In Fig. 4 we have shown the results for a sample that differed from the one of Figs. 2 and 3 only in the value of the DM interaction parameter V. The magnetic field used to rotate the magnetization was 5×10^{-3} r.u. (~ 17 kG). The solid line shows a best fit to Eqs. (9) and (10). The purely unidirectional form still provides a fairly good description of the data. However, the quality of the fit is clearly worse than that in Fig. 2. In particular, the value of K obtained from a fit to the data for $\theta_h \leq 90°$ was found to be larger than that obtained from the data for larger values of θ_h in all of the samples of this group. This indicates an effective θ_h-dependent anisotropy constant that decreases with increasing θ_h. This behavior would be consistent with the presence of a small uniaxial component in the anisotropy energy. Comparison of the new EC's with the initial one showed increased derivations from rigid rotations. The values of $P_{\max}$ varied from ~ 0.99 for $\theta_h = 30°$ to ~ 0.95 for $\theta_h = 180°$, indicating an increase in derivations from rigidity for larger rotation angles. A spin-by-spin comparison of the rotated state with the initial state showed that the observed deviations from rigidity were not produced by one or more small clusters rotating differently from the rest of the system. The values of K obtained from fits to the rotation data were found to be ~ 30—$50\,\%$ smaller than the rigid-rotation values.

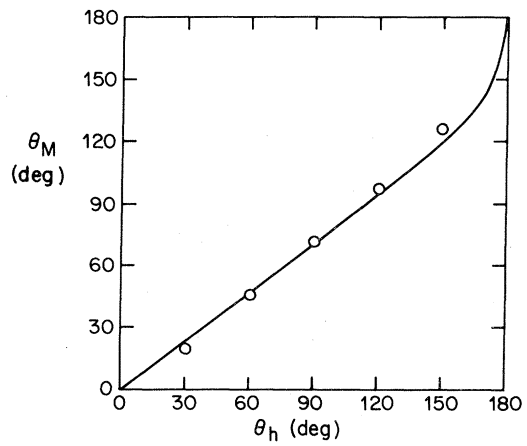

FIG. 4. Variation of θ_M with θ_h during a rotation of a sample of the second group (stronger anisotropy) about the x axis. The solid line represents the best fit to Eqs. (9) and (10).

B. Inversion and hysteresis

In these simulations, a magnetic field applied in a direction opposite to $\vec{M}_r^0$ was increased from zero in small steps until the magnetization turned over. The field was increased for a few more steps and then gradually decreased to zero. (In all samples the magnetization turned back before the field was reduced to zero.) At each step, the new EC was located using the EC at the previous step as the starting point. In carrying out this procedure we encountered a numerical difficulty. The calculated value (h_1) of $|h_z|$ at which the magnetization turned over, and also the value (h_2) at which it subsequently turned back, were found to depend on the choice of the convergence parameter δ that determined when to stop the iterations. In particular, a change of δ from 10^{-6} to 10^{-7} produced a decrease in the calculated value of h_1 and an increase in the value of h_2. The reason behind this problem is not difficult to understand. As an EC becomes unstable as a result of increasing (or decreasing) $|h_z|$ beyond a critical value h_c, the rate at which the energy decreases under iterations is an increasing function of $|h_c - |h_z||$. In the numerical procedure used here a state is considered to be stable if the rate at which the energy decreases under iterations is less than the convergence parameter δ. Thus it is not surprising that a state that appears stable in a calculation with a particular value of δ may turn out to be unstable in a calculation with a smaller value of δ. This is precisely the behavior we found. The numerical difficulty is made worse by the fact that for $\theta_h = 180°$, the energy varies very slowly with θ_M for θ_M close to zero. Monitoring the rate of change of θ_M during iterations improved the accuracy, but did not eliminate the problem. In simulations of the samples of the first group, the numerical uncertainties (estimated from the dependence on δ) in the values of h_1 and h_2 were comparable to the difference between h_1 and h_2. For this reason we cannot say anything definite about the hysteresis behavior of these samples. All samples of this group showed sharp one-step reversals of the magnetization. The EC's after inversion were related to the initial ones through nearly rigid rotations, with $P_{\max} > 0.98$. This result indicates that if a hysteresis loop exists at all, it will be very narrow. Sharp hysteresis cycles with small width have been observed in low-temperature experiments[2] on pure Cu-Mn in which the anisotropy energy is known to be very small. However, we are not aware of any experimental evidence indicating that an in-

version of the magnetization corresponds to a nearly rigid rotation. Some of the experimentally observed properties of the inverted state have been ascribed[9] to a breaking up of the system into domains which rotate by 180° about different axes. We find it difficult to reconcile this description with the sharp, one-step (or few-step) reversals of the magnetization seen in some hysteresis experiments.[2] However, due to the smallness of sample size we cannot draw any conclusion about the validity of this picture from the results of this simulation.

The numerical problems were less severe in the simulations of the samples of the second group, and we found well-defined hysteresis loops of varying sharpness. The results (obtained with $\delta=10^{-7}$) for the sample of Fig. 4 are shown in Fig. 5. The numerical uncertainty in determining h_1 and h_2 was less than 10% of the width of the hysteresis cycle. In order to describe the shape of this hysteresis loop it would be necessary to include an uniaxial term in the anisotropy energy. Similar hysteresis loops have been observed[3] for Cu-Mn samples containing small amounts of spin-orbit-scattering impurities, and also for pure Cu-Mn at relatively high temperatures. The inverted state had values of $P_{\max}$ between 0.7 and 0.9, indicating larger deviations from rigidity than the π-rotated state. The field needed to invert $\vec{M}_r^0$ was always smaller than the value expected from the analysis of the rotation data. This is consistent with experimental observations. A study of the spatial variation of the local relative rotation matrix [defined as the SO(3) matrix that maximizes a projection similar to the one defined in Eq. (11), but with the sum restricted to a small group of neighboring spins] did not show any evidence of different clusters rotating by 180° about different axes. Both the angle and the direction cosines of the axis of the local rotation matrix showed broad distributions, indicating a complicated rearrangement of the spins during an inversion.

In a recent simulation of a Heisenberg spin-glass with random nearest-neighbor exchange and DM interactions, Soukoulis *et al.*[22] found sharp magnetization reversals only when the exchange interaction had a large ferromagnetic average. From this observation, they concluded that a tendency towards ferromagnetism is necessary for maintaining rigidity of the spin system during rotations and inversion of magnetization. In all the samples studied in our simulations, the average value of the exchange interaction showed a slight tendency towards antiferromagnetism. The results obtained in the present work (especially the results for the first group of samples) are thus in contradiction with the conclusion of Soukoulis *et al.* We believe that this disagreement is primarily due to a large difference in the values of the anisotropy parameters used in these two calculations. In Ref. 22 the DM energy was comparable to the isotropic exchange energy, whereas in our calculation the ratio of these two energies was of order 10^{-3}. In view of the fact that we found substantial deviations from rigidity when the ratio of DM to RKKY energy was $\sim 2.5\times 10^{-3}$, it is not surprising that the calculation of Ref. 22 did not show any rigid behavior unless a ferromagnetic tendency was imposed. It is interesting to note here that there exist experimental results which show a decrease in the sharpness of the hysteresis loop with increasing anisotropy. Prejean *et al.*[3] found sharp reversals of the magnetization in $CuMn_{1\ at.\%}Pt_y$ for $y \leq 100$ ppm, whereas for $y=300$ ppm, the reversal of the magnetization was found to be smooth. Another important difference between the systems studied in Ref. 22 and in the present work comes from the range of the exchange interaction. The long-range nature of the RKKY interaction used in our simulation correlates spins over long distances and thus enhances the rigidity of the spin system.

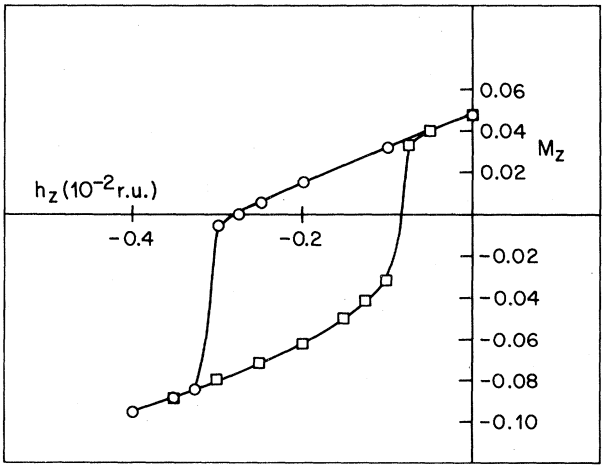

FIG. 5. Hysteresis cycle for the sample of Fig. 4. The circles and the squares, represent, respectively, the variation of the magnetization during an increase of $|h_z|$ from zero and a subsequent decrease of $|h_z|$ to zero. r.u. represents reduced units.

V. SUMMARY

To summarize the main results obtained in this study we have verified that the macroscopic anisotropy in a classical RKKY spin-glass with weak DM interactions has the theoretically predicted unidirectional form for rigid rotations. However, the estimated value of the anisotropy constant for rigid rotations is ~ 30 times larger than the experimental value. From simulations of the response of the spin system to a magnetic field that tends to rotate or invert the magnetization we have found that rotations and inversions of samples with weak anisotropy ($E_{DM}/E_{RKKY}\sim 5\times 10^{-4}$) are well described by rigid rotations and a purely unidirectional anisotropy. In samples with stronger anisotropy ($E_{DM}/E_{RKKY}\sim 2.5\times 10^{-3}$) we find substantial deviations from rigidity in large-angle rotations and inversions. The deviations from rigidity cause noticeable departures from purely unidirectional behavior. The observed behavior is consistent with an anisotropy energy that has both unidirectional and uniaxial components. Thus, we have shown that deviations from rigidity may produce some uniaxiality in a system in which the anisotropy for rigid rotations is purely unidirectional. It is not necessary to invoke the presence of new microscopic interactions (such as dipolar interaction) to explain the presence of an uniaxial term in the anisotropy energy. This work thus confirms previous suggestions[8,9] that the uniaxiality seen in torque and hysteresis experiments on Cu-Mn is due to nonrigid rotations. We also find that the amount of nonrigidity depends on the degree of deviation

from the initial state and also on the manner in which this deviation is produced. This may explain why different experiments lead to different conclusions about the nature of anisotropy in Cu-Mn. The trends found in our simulations are consistent with the experimental results. The observed increase in the deviation from rigidity with increasing anisotropy implies that a description in which the spin-glass state is represented by a single orthonormal triad[13–15] will break down for large anisotropy. The same conclusion was reached in the numerical study of Morgan-Pond.[17]

In this study we have left out important contributions to nonrigidity coming from thermally activated processes. Because of this reason and the smallness of sample size, a quantitative comparison of the results of these simulations with experiments would not be appropriate. However, we expect the qualitative picture that emerges from this study to remain valid in more realistic situations.

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