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Anisotropy induced by nonmagnetic impurities in *Cu*Mn spin-glass alloys

Peter M. Levy
Department of Physics, New York University, 4 Washington Place, New York, New York 10003

A. Fert
Laboratoire de Physique des Solides, Université de Paris-Sud, 91405 Orsay, France

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Recent measurements on *Cu*Mn spin-glass alloys have revealed that the anisotropy field maintaining the remanent magnetization in the direction of the initial applied field is strongly enhanced by the addition of nonmagnetic Au or Pt impurities. A similar enhancement of the anisotropy field of *Cu*Mn spin-glass alloys by nonmagnetic impurities is also indicated by ESR measurements. We show that these results can be accounted for by the existence of Dzyaloshinsky-Moriya-type interactions between the Mn spins arising from spin-orbit scattering of the conduction electrons by nonmagnetic impurities. We derive this interaction by using third-order perturbation theory on the conduction electrons and by retaining only the terms in the energy which are first order in the spin-orbit coupling of the nonmagnetic impurity. To calculate the macroscopic anisotropy energy from the anisotropic pair interaction, it is necessary to make some assumptions because of our incomplete knowledge of the spin-glass state. We are able to justify our assumptions *a posteriori* by comparing our predictions to the experimental results. In particular, we are able to explain the variation over two decades of the anisotropy energy with the type of nonmagnetic transition-metal impurity.

I. INTRODUCTION

Recent experiments by Préjean *et al.*[1] on *Cu*Mn doped with nonmagnetic impurities shine a new light on the crucial role played by anisotropic couplings between local moments in stabilizing the remanent magnetization of spin-glasses. From measurements of the hysteresis loops of *Cu*Mn spin-glass alloys, it has been found that the remanent magnetization induced by positive fields undergoes a steep reversal at a negative field H_r. Préjean *et al.* have found that $|H_r|$ is considerably enhanced by the addition of a few hundred ppm of Au or Pt nonmagnetic impurities. For example, $|H_r|$ is 170 Oe in pure *Cu*Mn 1 at.% and is increased by the addition of Au impurities at a linear rate of 6.2×10^3 Oe/at.% Au. This linear increase has been observed for concentrations of Au between 0.01 and 0.15 at.%. At higher concentrations, the width of the hysteresis loop continues to increase but there is no longer a sharp reversal of the magnetization and a reversal field H_r cannot be defined. Pt impurities are even more effective than Au and increase $|H_r|$ at a rate of 34×10^3 Oe/at.% Pt. By contrast the addition of Al has practically no effect. Thus Préjean *et al.* concluded that *the addition of nonmagnetic impurities with strong spin-orbit coupling (Au,Pt) sharply increases the anisotropy field which maintains the remanent magnetization in the direction of the initial applied field.*

Additional evidence for the selective effect of nonmagnetic impurities on the anisotropy field of spin-glasses is provided by electron-spin-resonance (ESR) measurements. In spin-glasses, a shift of the ESR is associated with the anisotropy field acting on the remanent part of the magnetization. This has been clearly shown by Monod and Berthier[2] in their low-frequency and low-field ESR studies of *Cu*Mn spin-glass alloys. These authors found that, in the field-cooled spin-glass state, the ESR frequency ω is described by the relation

$$\omega = \gamma(H_0 + H_A) \ ,$$

where H_0 is the applied field and H_A is the anisotropy field. The determination of H_A from the resonance shift is in agreement with that found by other methods in particular, NMR.[3] Some time ago Okuda and Date[4] studied the ESR of *Cu*Mn 2 at.% alloys containing Al, Zn, Ti, Ni, Fe, Co, or Pd impurities. They found that the ESR of these ternary alloys in the spin-glass state is shifted with respect to *Cu*Mn by an amount which varies linearly with the concentration of the added elements. The rate of increase of the shift varies from zero for Al impurities to 6.6×10^3 G/at.% for Co impurities. These results provide further evidence for the enhancement of the anisotropy field by nonmagnetic impurities (Ni and Co impurities in Cu do not carry a magnetic moment). As in the results of Préjean *et al.*,[1] the impurities with strong spin-orbit scattering (Co and the other transition elements) give much stronger effects than impurities with weak spin-orbit scattering (Zn,Al).

In a preceding article[5] referred to as FL we have shown that the enhancement of the anisotropy field in *Cu*Mn*T* ternary alloys can be accounted for by an additional term in the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction which is of the Dzyaloshinsky-Moriya (DM) type and is due to spin-orbit scattering of the conduction electrons by the nonmagnetic impurities. The existence of such interaction terms had been already predicted in low-symmetry crystals and in disordered magnetic systems.[6,7] In FL we derived a simple expression for the DM interaction induced by spin-orbit scattering from nonmagnetic transition-metal impurities as a function of some reliably known parameters and we compared our predictions with the experimental result. Here we present the details of the calculations of the DM interaction and the macroscopic anisotropy field arising from the DM interactions.

The salient feature of the results of Préjean *et al.*[1] and Okuda and Date[4] is the clear relation of the anisotropy energy of the ternary alloys $Cu\mathrm{Mn}_xT_y$ to the spin-orbit coupling scattering of the nonmagnetic impurity *T*. For platinum which produces large spin orbit scattering in *Cu*, the anisotropy energy as estimated by Préjean *et al.* is two orders of magnitude larger than for *Cu*Mn with no additional impurities.

We are able to explain this increase in anisotropy as follows. In the absence of additional impurities the dominant magnetic coupling between manganese magnetic moments is mediated by conduction electrons via the RKKY interaction.[8] It is rather isotropic, the residual anisotropy in *Cu*Mn is very small compared to that when nonmagnetic ternary impurities are introduced and is not of interest to us in this paper. The introduction of a nonmagnetic impurity perturbs the gas of conduction electrons. This leads to a reduction in the RKKY coupling due to the reduced mean free path of the conduction electrons. However, from the result of Préjean *et al.*[1] we see that this effect is not large because aluminum which is an effective scatterer does not appreciably change the properties of the *Cu*Mn alloys. In addition, when the nonmagnetic impurity has a sizable spin-orbit coupling Préjean *et al.* find a dramatic increase in the coercivity or anisotropy energy of the alloys without appreciable changes in either T_g the spin-glass freezing temperature, or μ_r the remanent magnetization. These impurities act in such a way as to induce locally a spin-orbit coupling in the conduction electrons. When Mn magnetic moments interact with these modified conduction-electron states the resultant magnetic coupling contains anisotropic terms[6,7] whose magnitudes are sufficiently large to explain the anisotropy energies observed by Préjean *et al.*[1]

There are two ways we can calculate the anisotropic coupling between Mn spins in *Cu* in the presence of nonmagnetic impurities with appreciable spin-orbit coupling. We can use the expression given originally by Ruderman and Kittel[8] and use in this second-order perturbation expression spin-orbit-coupled conduction-electron wave functions which include the phase shifts due to the impurities, i.e., spin-orbit-coupled distorted plane waves. The resulting coupling contains antisymmetric (Dzyaloshinsky-Moriya) and pseudodipolar interactions in addition to the conventional isotropic RKKY interaction. The DM term appears in the lowest order in the spin-orbit coupling λ, and we do not consider the higher-order pseudodipolar term in this paper. The difficulty with this approach is that we introduce perturbed wave functions (correct to first order in λ) into a second-order perturbation expression and we must assure ourselves that other contributions to the same order of λ do not enter. In Appendix B we show that this approach does in fact give us the correction coupling to lowest (first) order in the spin-orbit coupling.

The more satisfactory way (at least from a formal standpoint) to calculate the anisotropic coupling to first order in the spin-orbit coupling is to develop and evaluate an expression for the third-order perturbation of the ground-state energy of the gas of conduction electrons due to the nonmagnetic impurity and the Mn spins. We rapidly find the expression *trilinear* in the Mn spins and spin-orbit coupling parameter λ of the nonmagnetic impurity is of the antisymmetric DM type. While the existence of such a term has been previously shown[7] the formula derived was missing a crucial term due to double pole singularities in the perturbation expression. Furthermore, previously plane waves were used, while in our derivation we use distorted plane waves to include the effects of the impurity potential scattering on our coupling.

The crucial ingredient of the anisotropic coupling found in our calculation is that the interaction energy between spins depends on their orientation relative to the triangle formed by the manganese and impurity ions. This dependence is the origin of the anisotropy energy observed in the $Cu\mathrm{Mn}_xT_y$ alloys. An exact calculation of the macroscopic anisotropy energy from these anisotropic pair couplings is hindered by our lack of knowledge of the ground state of a spin-glass. We have made some plausible and justifiable assumptions to perform the averaging over the individual Mn pairs of spins to estimate the anisotropy energy that arises from our anisotropic pair couplings between Mn spins due to ternary nonmagnetic impurities.

In Appendix A we derive the third-order energy expression for an electron gas perturbed by one electron interactions. In the next section we use this expression to calculate the anisotropic coupling between Mn spins linear in the spin-orbit coupling of the nonmagnetic impurity *T*. In Sec. III we estimate the anisotropy energy. In the last section we introduce numerical values for the parameters entering our derived expression and compare our results to those found in the experiments of Préjean *et al.* and Okuda and Date.

II. ANISOTROPIC COUPLING BETWEEN Mn SPINS

To derive the anisotropic coupling between Mn spins in *Cu* induced by the addition of nonmagnetic impurities we consider the third-order energy correction to a gas of conduction electrons Eq. (A34),

$$E^{(3)}=\left[\frac{1}{8\pi^3}\right]^3\left[\frac{2m}{\hbar^2}\right]^2\int_0^{k_F}dk_1\left[\int_0^\infty dk_2\int_0^\infty dk_3\left[\frac{\mathcal{P}}{(k_1^2-k_2^2)}\frac{\mathcal{P}}{(k_1^2-k_3^2)}-\frac{\pi^2}{12k_1^2}\delta(k_2-k_1)\delta(k_3-k_2)\right]V(k_1k_2k_3)\right]\ , \tag{1}$$

where

$$V(k_1k_2k_3)=k_1^2k_2^2k_3^2\sum_{\sigma_1\sigma_2\sigma_3}\int d\Omega_{k_1}\int d\Omega_{k_2}\int d\Omega_{k_3}V(\vec{k}_1\sigma_1,\vec{k}_2\sigma_2)\,V(\vec{k}_2\sigma_2,\vec{k}_3\sigma_3)\,V(\vec{k}_3\sigma_3,\vec{k}_1\sigma_1)\ ,$$

due to the following perturbation potential,

$$V=-\Gamma\delta(\vec{r}-\vec{R}_a)\vec{s}\cdot\vec{S}_a-\Gamma\delta(\vec{r}-\vec{R}_b)\vec{s}\cdot\vec{S}_b+\lambda(r)\vec{s}\cdot\vec{l}\ . \tag{2}$$

The coupling constants Γ represent the interactions of the Mn spins S_a and S_b localized at $\vec{R}_a$ and $\vec{R}_b$ with the spins of conduction electrons and the last term presents the spin-orbit interaction on the site of a nonmagnetic transition-metal impurity at $\vec{R}=0$. When this perturbation is introduced in Eq. (1) there are 27 terms. However, there are only six terms of interest to us, i.e., those trilinear in the Mn spins S_a, S_b, and the spin-orbit coupling parameter λ.

On the site of the impurity, the spin-orbit coupling of a conduction electron is considerably enhanced because the admixture of the impurities' *d* states into the conduction band allows the conduction electrons to experience the strong spin-orbit forces of the *d* states. Therefore it is important to take into account this admixture when calculating the matrix elements entering Eq. (1). In the virtual bound state (vbs) model[9] the admixture of the atomic *d* states ϕ_{2m} with plane waves is written as

$$\phi_{\vec{k}}=\exp(i\vec{k}\cdot\vec{r})+\exp[i\eta_2(k)]\sin\eta_2(k)\frac{\langle d|V_0|k\rangle}{\Delta}\times\sum_{m=-2}^{2}Y_{2m}^*(\hat{k})\phi_{2m}(r)+\cdots\ , \tag{3a}$$

for electrons in the immediate region about the transition-metal impurity ($\vec{R}=0$). This is the wave function appropriate for the matrix elements of the perturbation $\lambda(r)\vec{s}\cdot\vec{l}$. For the matrix elements of the perturbations due to the Mn spins, which are situated at R_a, R_b about 10 Å from the transition-metal impurity, the appropriate wave function to use is

$$\phi_{\vec{k}}=\exp(i\vec{k}\cdot\vec{r})-4\pi\exp[i\eta_2(k)]\sin\eta_2(k)h_2^{(+)}(kr)\times\sum_{m=-2}^{2}Y_{2m}^*(\hat{k})Y_{2m}(\hat{r})\ . \tag{3b}$$

In these expressions Δ is the half-width of the virtual bound state, $h_2^{(+)}(kr)$ is an Hankel function of the first kind, and η_2 is the phase shift of the $l=2$ partial waves,

$$\eta_2(k)=\tan^{-1}\{\Delta/[E_r-E(k)]\}\ , \tag{4}$$

where E_r is the energy of the center of the vbs resonance. At the Fermi level the phase shift is related to the number of *d* electrons Z_d by the Friedel rule

$$\eta_2(E_F)=(\pi/10)Z_d\ , \tag{4a}$$

and the matrix element $\langle d|V_0|k\rangle$ is related to the density of states for one spin direction at the Fermi level $N(E_F)$ by the relation[9]

$$|\langle k|V_0|d\rangle|^2=4\Delta/N(E_F)\ . \tag{5}$$

As mentioned before there are six terms trilinear in the Mn spins and the spin-orbit coupling parameter. We will evaluate one term explicitly; the contributions from the remaining ones can be readily found by taking permutations of S_a, S_b, and λ in the expression we develop. By placing the perturbation Eq. (2) in the third-order energy correction Eq. (1), we find one trilinear term in S_a, S_b, and λ is

$$V(k_1k_2k_3)=k_1^2k_2^2k_3^2N^{-2}\int d\Omega_{k_1}\int d\Omega_{k_2}\int d\Omega_{k_3}\sum_{\sigma_1\sigma_2\sigma_3}\langle\vec{k}_1\sigma_1|\Gamma\delta(\vec{r}-\vec{R}_a)\vec{s}\cdot\vec{S}_a|\vec{k}_2\sigma_2\rangle\langle\vec{k}_2\sigma_2|\lambda(r)\vec{s}\cdot\vec{l}|\vec{k}_3\sigma_3\rangle\times\langle\vec{k}_3\sigma_3|\Gamma\delta(\vec{r}-\vec{R}_b)\vec{s}\cdot\vec{S}_b|\vec{k}_1\sigma_1\rangle\ , \tag{6}$$

where the factor N^{-2}, enters from the matrix elements of the δ functions evaluated by using Eq. (3b), and N is the number of lattice sites per unit volume in the crystal. To evaluate the spin-orbit interaction for the conduction electron when it is in the vicinity of the transition-metal impurity $R=0$ we use the wave function Eq. (3a). The plane-wave part of the function does not contribute much to the spin-orbit coupling because the radial portion of a plane wave is rather diffuse. The dominant part of the spin-orbit interaction comes from the atomic d states admixed in Eq. (3a) and we find

$$\begin{aligned}\langle \vec{k}_2\sigma_2|\lambda(r)\vec{l}\cdot\vec{s}|\vec{k}_3\sigma_3\rangle &= [4\lambda_d/\Delta N(E_F)]\exp\{i[\eta_2(k_3)-\eta_2(k_2)]\}\\ &\quad\times\sin\eta_2(k_2)\sin\eta_2(k_3)\sqrt{10}[Y^2(\hat{k}_2)\times Y^2(\hat{k}_3)]^1\cdot\langle\sigma_2|\vec{s}\,|\sigma_3\rangle\ ,\end{aligned}\tag{7}$$

where λ_d represents the average of the spin-orbit coupling constant $\lambda(r)$ taken over the atomic radial d state wave function, and $[Y^2(\hat{k}_2)\times Y^2(\hat{k}_3)]^1$ is the vector product of second-rank spherical harmonics, i.e.,

$$[Y^2(\hat{k}_2)\times Y^2(\hat{k}_3)]^1_m=\sum_{m'm''}(2m',2m''|1m)\,Y^2_{m'}(\hat{k}_2)\,Y^2_{m''}(\hat{k}_3)\ ,\tag{8}$$

where $(2m',2m''|1m)$ is a Clebsch-Gordan coefficient. To evaluate the matrix element of the Mn spin interaction with the conduction electrons we use Eq. (3b) and find

$$\begin{aligned}\langle\vec{k}_1\sigma_1|\Gamma\delta(\vec{r}-\vec{R}_a)\vec{s}\cdot\vec{S}_a|\vec{k}_2\sigma_2\rangle &=\Gamma\{\exp(-i\vec{k}_1\cdot\vec{R}_a)-4\pi\exp[-i\eta_2(k_1)]\sin\eta_2(k_1)h_2^{(-)}(k_1R_a)Y^2(\hat{k}_1)\cdot Y^2(\hat{R}_a)\}\\ &\quad\times\{\exp(i\vec{k}_2\cdot\vec{R}_a)-4\pi\exp[i\eta_2(k_2)]\sin\eta_2(k_2)h_2^{(+)}(k_2R_a)Y^2(\hat{k}_2)\cdot Y^2(\hat{R}_a)\}\\ &\quad\times\langle\sigma_1|\vec{s}\cdot\vec{S}_a|\sigma_2\rangle\ ,\end{aligned}\tag{9}$$

where

$$Y^2(\hat{k})\cdot Y^2(\tilde{R})\equiv\sum_m(-)^mY^2_m(\hat{k})\,Y^2_{-m}(\hat{R})\tag{10}$$

is the scalar product of second-rank harmonics. The principal contribution to this matrix element comes from the undistorted plane wave; the distortions proportional to η_2 contribute very little. After placing Eqs. (7), (9), and one equivalent for the spin interaction at R_b, in Eq. (6) we must integrate over the solid angles of the k vectors and sum over the spin states of the conduction electrons. The integrations over Ω_k are done by using the following formulas

$$\begin{aligned}&\int d\Omega_k\,e^{i\vec{k}\cdot\vec{R}}Y^2_m(\hat{k})=-4\pi j_2(kr)\,Y^2_m(\hat{R})\ ,\\ &\int d\Omega_k[Y^2(\hat{k})\cdot Y^2(\hat{R})]\,Y^2_m(\hat{k})=Y^2_m(\hat{R})\ ,\\ &\int d\Omega_k[Y^2(\hat{k})\cdot Y^2(\hat{R}_a)][Y^2(\hat{k})\cdot Y^2(\hat{R}_b)]\\ &\qquad=Y^2(\hat{R}_a)\cdot Y^2(\hat{R}_b)\ ,\\ &\int d\Omega_k\exp(i\vec{k}\cdot\vec{R}_a)\,Y^2(\hat{k})\cdot Y^2(\hat{R}_b)\\ &\qquad=-4\pi j_2(kR_a)\,Y^2(\hat{R}_a)\cdot Y^2(\hat{R}_b)\ ,\end{aligned}\tag{11}$$

and

$$\int d\Omega_k\exp[i\vec{k}\cdot(\vec{R}_a-\vec{R}_b)]=4\pi\sin kR_{ab}/kR_{ab}\ ,$$

where $R_{ab}\equiv|\vec{R}_a-\vec{R}_b|$, and $j_2(kR)$ is a spherical Bessel function of rank 2. The sum over the spin states is

$$\begin{aligned}\sum_{\sigma_1\sigma_2\sigma_3}\langle\sigma_1|\vec{s}\cdot\vec{S}_a|\sigma_2\rangle\langle\sigma_2|\vec{s}|\sigma_3\rangle\langle\sigma_3|\vec{s}\cdot\vec{S}_b|\sigma_1\rangle &=\mathrm{Tr}_\sigma(\vec{s}\cdot\vec{S}_a)\vec{s}(\vec{s}\cdot\vec{S}_b)\\ &=-(i/4)(\vec{S}_a\times\vec{S}_b)\ .\end{aligned}\tag{12}$$

By using these formulas and performing the integrations and sums over states indicated in Eq. (6) we find

$$\begin{aligned}V(k_1k_2k_3)=\frac{4(4\pi)^3\lambda_d\Gamma^2}{\Delta N^2N(E_F)}k_1^2k_2^2k_3^2\,&(\sin k_1R_{ab}/k_1R_{ab}+\sin\eta_2(k_1)\{\exp[-i\eta_2(k_1)]h_2^{(-)}(k_1R_a)j_2(k_1R_b)\\ &+\exp[i\eta_2(k_1)]j_2(k_1R_a)h_2^{(+)}(k_1R_b)\}\\ &\times4\pi Y^2(\hat{R}_a)\cdot Y^2(\hat{R}_b)+\sin^2\eta_2(k_1)h_2^{(-)}(k_1R_a)h_2^{(+)}(k_1R_b)4\pi Y^2(\hat{R}_a)\cdot Y^2(\hat{R}_b))\\ &\times\sin\eta_2(k_2)\{\exp[-i\eta_2(k_2)]j_2(k_2R_a)+\sin\eta_2(k_2)h_2^{(+)}(k_2R_a)\}\sin\eta_2(k_3)\\ &\times\{\exp[i\eta_2(k_3)]j_2(k_3R_b)+\sin\eta_2(k_3)h_2^{(-)}(k_3R_b)\}(-i/4)\sqrt{10}[Y^2(\hat{R}_a)\times Y^2(\hat{R}_b)]^1\cdot\vec{S}_a\times\vec{S}_b\ .\end{aligned}\tag{13}$$

At the concentrations used in the various experiments the average distance between the transition-metal impurity and the Mn ions at R_a and R_b is about 10 Å. This is sufficiently large so that we can replace the Bessel and Hankel functions by their asymptotic expressions

$$h_2^{(\pm)}(kR) \to -\exp(\pm ikR)/kR$$

and

$$j_2(kR) \to -\sin kR/kR \ . \tag{14}$$

By placing these expressions in Eq. (13) we find the asymptotic limit of the terms in Eq. (13) in bold parentheses and curly brackets is

$$(\sin k_1R_{ab}/k_1R_{ab} + \sin\eta_2(k_1)\sin[k_1(R_a+R_b)+\eta_2(k_1)]/(k_1R_ak_1R_b)4\pi Y^2(\hat{R}_a)\cdot Y^2(\hat{R}_b))$$
$$\times\{\sin[k_2R_a+\eta_2(k_2)]/k_2R_a\}\{\sin[k_3R_b+\eta_2(k_3)]/k_3R_b\} \ . \tag{15}$$

In this limit of large R, $1/R_aR_b$ is much less than $1/R_{ab}$ and we will neglect the second term in the first curly brackets. We can rewrite the vector product between second rank harmonics as

$$\sqrt{10}i[Y^2(\hat{R}_a)\times Y^2(\hat{R}_b)]^1 = (15/4\pi)\hat{R}_a\cdot\hat{R}_b(\hat{R}_a\times\hat{R}_b) \ , \tag{16}$$

where $\hat{R} \equiv \vec{R}/|R|$ is a unit vector. With these simplifications we find that in the asymptotic limit Eq. (6) becomes

$$V(k_1k_2k_3) = -[15(4\pi)^2\lambda_d\Gamma^2/\Delta N^2N(E_F)]k_1^2k_2^2k_3^2(\sin k_1R_{ab}/k_1R_{ab})$$
$$\times\{\sin\eta_2(k_2)\sin[k_2R_a+\eta_2(k_2)]/k_2R_a\}\{\sin\eta_2(k_3)\sin[k_3R_b+\eta_2(k_3)]/k_3R_b\}$$
$$\times\hat{R}_a\cdot\hat{R}_b(\hat{R}_a\times\hat{R}_b)\cdot(\vec{S}_a\times\vec{S}_b) \ . \tag{17}$$

By placing this expression in Eq. (1) we find the contribution of Eq. (6) (one of six) to the anisotropic coupling between Mn spins is

$$E_1^{(3)} = -\frac{15}{32\pi^7}\frac{\lambda_d\Gamma^2}{\Delta N^2N(E_F)}\left[\frac{2m}{\hbar^2}\right]^2\frac{I_1}{R_aR_bR_{ab}}\hat{R}_a\cdot\hat{R}_b(\hat{R}_a\times\hat{R}_b)\cdot(\vec{S}_a\times\vec{S}_b) \ , \tag{18}$$

where

$$I_1 \equiv \int_0^{k_F} k_1\,dk_1\sin k_1R_{ab}\left(\mathcal{P}\int_0^\infty \sin\eta_2(k_2)\frac{\sin[k_2R_a+\eta_2(k_2)]}{(k_1^2-k_2^2)}k_2\,dk_2\,\mathcal{P}\int_0^\infty \sin\eta_2(k_3)\frac{\sin[k_3R_b+\eta_2(k_3)]}{(k_1^2-k_3^2)}k_3\,dk_3\right.$$
$$\left.-\frac{\pi^2}{12}\sin^2\eta_2(k_1)\sin[k_1R_a+\eta_2(k_1)]\sin[k_1R_b+\eta_2(k_1)]\right) \ . \tag{19}$$

To evaluate the Cauchy principal values of the integrals we perform contour integrations as shown in Appendix C and find for large R, see Eq. (C7),

$$\mathcal{P}\int_0^\infty \sin\eta_2(k')\frac{\sin[k'R+\eta_2(k')]}{k_1^2-k'^2}k'\,dk' = -\frac{\pi}{2}\sin\eta_2(k_1)\cos[k_1R+\eta_2(k_1)] \ .$$

By placing these integrals in Eq. (19) we find

$$I_1 = \frac{\pi^2}{4}\int_0^{k_F}\sin k_1R_{ab}\{\cos[k_1R_a+\eta_2(k_1)]\cos[k_1R_b+\eta_2(k_1)]$$
$$-\tfrac{1}{3}\sin[k_1R_a+\eta_2(k_1)]\sin[k_1R_b+\eta_2(k_1)]\}\sin^2\eta_2(k_1)k_1\,dk_1 \ . \tag{20}$$

In evaluating the other five trilinear contributions to Eq. (1), one can see by reviewing the steps from Eq. (6) to Eq. (18) that as we permute the three terms in S_a, S_b, and λ, the energies $E_2^{(3)}$ to $E_6^{(3)}$ differ from Eq. (18) only in the integrals I_n, and then only the first part of these integrals differ, see Eq. (19). After adding the six different contributions to Eq. (1), trilinear in S_a, S_b, and λ we find the energy of the form of Eq. (18) with an integral I of the form

$$
\begin{aligned}
I &= \sum_{i=1}^{6} I_i \\
&= \frac{\pi^2}{4}\int_0^{k_F} (2\sin kR_{ab}\cos[kR_a+\eta_2(k)]\cos[kR_b+\eta_2(k)] \\
&\qquad +2\cos kR_{ab}\{\sin[kR_a+\eta_2(k)]\cos[kR_b+\eta_2(k)]+\cos[kR_a+\eta_2(k)]\sin[kR_b+\eta_2(k)]\} \\
&\qquad -6(\tfrac{1}{3})\sin kR_{ab}\sin[kR_a+\eta_2(k)]\sin[kR_b+\eta_2(k)])\sin^2\eta_2(k)k\,dk \\
&= \frac{\pi^2}{2}\int_0^{k_F} \{\sin kR_{ab}\cos[k(R_a+R_b)+2\eta_2(k)]+\cos kR_{ab}\sin[k(R_a+R_b)+2\eta_2(k)]\}\sin^2\eta_2(k)\,k\,dk \\
I &= \frac{\pi^2}{2}\int_0^{k_F} \sin[k(R_a+R_b+R_{ab})+2\eta_2(k)]\sin^2\eta_2(k)k\,dk\ .
\end{aligned}
\tag{21}
$$

To evaluate this last integral we make the following observation. From the definitition Eq. (4), we note that for a vbs the phase shift η_2

$$
\sin^2\eta_2(k)=\frac{\Delta^2}{[E(k)-E_r]^2+\Delta^2}
\tag{22}
$$

is appreciable only for energies $E(k)$ in the immediate vicinity Δ (a half-width, i.e., about 0.25 eV) of the center of the vbs resonance, E_r. Furthermore over the interval when $\sin^2\eta_2(k)$ is nonzero, the phase η_2 rapidly goes from 0 to π while $k(R_a+R_b+R_{ab})$ is relatively constant for distances between impurities corresponding to concentrations greater than 1000 ppm. Therefore we can set k equal to k_f [which is close to k_r, i.e., $(2mE_r/\hbar)^{1/2}$]. (See Note added in proof.) By differentiating the phase shift, Eq. (4) with respect to the energy and using $E(k)=\hbar^2k^2/2m$, we find

$$
d\eta_2=(\hbar^2/m\Delta)\sin^2\eta_2\ kdk\ .
\tag{23}
$$

This allows us to replace the integral in k space, Eq. (21), to one over the phase shift η_2

$$
\begin{aligned}
I &\cong \frac{\pi^2}{2}\frac{m\Delta}{\hbar^2}\int_0^{\eta_2(E_F)} \sin[k_F(R_a+R_b+R_{ab})+2\eta_2]d\eta_2 \\
&= (\pi^2/2)(m\Delta/\hbar^2)\sin\eta_2(E_F)\sin[k_F(R_a+R_b+R_{ab})+\eta_2(E_F)]\ .
\end{aligned}
\tag{24}
$$

When this result is placed in the energy expression of the form of Eq. (18), we find the total of the six trilinear contributions to the third energy correction of the electron gas is

$$
\begin{aligned}
H_{\mathrm{DM}} \equiv \sum_{i=1}^{6} E_i^{(3)} &= -\frac{15}{64\pi^5}\frac{\lambda_d\Gamma^2}{N^2N(E_F)}\frac{1}{2}\left(\frac{2m}{\hbar^2}\right)^3 \sin\eta_2(E_F) \\
&\quad\times\frac{\sin[k_F(R_a+R_b+R_{ab})+\eta_2(E_F)]}{R_aR_bR_{ab}}\hat{R}_a\cdot\hat{R}_b(\hat{R}_a\times\hat{R}_b)\cdot(\vec{S}_a\times\vec{S}_b)\ .
\end{aligned}
\tag{25}
$$

This expression can be cast in its final form as

$$
H_{\mathrm{DM}}=-V_1\frac{\sin[k_F(R_a+R_b+R_{ab})+(\pi/10)Z_d]\hat{R}_a\cdot\hat{R}_b}{R_aR_bR_{ab}}(\hat{R}_a\times\hat{R}_b)\cdot(\vec{S}_a\times\vec{S}_b)\ ,
\tag{26}
$$

where

$$
V_1=\frac{135\pi}{32}\frac{\lambda_d\Gamma^2}{E_F^2k_F^3}\sin\left(\frac{\pi}{10}Z_d\right)\ .
$$

To arrive at this expression we used the relation, Eq. (4a) of the phase shift at the Fermi level to the number of d electrons on the transition-metal impurity; we assumed one electron per Cu atom, $N_e=N$ and the following definitions for a gas of free electrons

$$
E_F=\hbar^2k_F^2/2m\ ,
$$

$$
k_F^3=3\pi^2N_e\ ,
$$

and

$$
N(E_F)=3N_e/4E_F\ ,
$$

is the density of states for one direction of the spin at the Fermi level.

This expression, Eq. (26) is the sought after anisotropic coupling of the Dzyaloshinsky-Moriya (DM) form between Mn spins located at sites A and B. As we can see the magnitude of the coupling depends on the orientation of the spins relative to the axes $(\vec{R}_a \times \vec{R}_b)$; for this reason it is called anisotropic. In addition the coupling depends on the relative orientation of one spin to the other; this of itself does not produce anisotropy energy. To this order in the spin-orbit coupling λ there is another coupling term which is proportional to $R_a^{-2}R_b^{-2}$. As this term contains one more power of $1/R$ than Eq. (26) and R is large (order of 10 Å), we have neglected its contributions; i.e., Eq. (26) is the leading term in $1/R$ (see Note added in proof). If one goes to higher powers in λ, i.e., fourth- or higher-order corrections to the energy of the electron gas, other anisotropic couplings between the Mn spins arise, e.g., pseudodipolar couplings. To recapitulate, the dominant anisotropic coupling between Mn spins in *Cu*Mn ($1\sim2$ at.% Mn) with transition-metal impurities is of the Dzyaloshinsky-Moriya form given by H_{DM} Eq. (26). For spin-glasses with higher concentrations of Mn or T and for impurities with very large spin-orbit coupling parameters (λ/E_F), other couplings enter.

To obtain an estimate of the strength of this DM interaction let us compare it to the leading term in $1/R$ of the RKKY interaction. For one electron per atom, this is given by

$$H_{\text{RKKY}} = V_0 \frac{\cos(2k_F R_{ab})}{R_{ab}^3} \vec{S}_a \cdot \vec{S}_b \ , \tag{27}$$

with

$$V_0 = \frac{9\pi\Gamma^2}{32E_F k_F^3} \ . \tag{28}$$

The ratio of V_1, Eq. (26), to V_0 is a simple function of the reliably known parameters λ_d, E_F, and Z_d,

$$\frac{V_1}{V_0} = 15\frac{\lambda_d}{E_F}\sin\left[\frac{\pi}{10}Z_d\right] \ . \tag{29}$$

In Table I we list calculated values of V_1/V_0 for various *Cu*Mn$_x T_y$ alloys. When λ_d is large V_1 is surprisingly large, e.g., $V_1/V_0 \sim \frac{1}{5}$ for $T=$Pt. In the next section we use the anisotropic coupling Eq. (26) to calculate the anisotropy energy due to the addition of transition-metal impurities in *Cu*Mn alloys.

Note added in proof

When one takes into account the variation of $k(R_a+R_b+R_{ab})$ across the vbs resonance[9a] in calculating the integral Eq. (24) one finds

$$I \cong (\pi^2/2)(m\Delta/\hbar^2)\frac{1}{1+\gamma}\sin[(1+\gamma)\eta_2(E_F)]$$
$$\times\sin[k_F(R_a+R_b+R_{ab})+(1+\gamma)\eta_2(E_F)] \ , \tag{24a}$$

where

$$\gamma = (\Delta/2\pi E_F)k_F(R_a+R_b+R_{ab}) \ .$$

We can neglect $\gamma\eta_2(E_F)$ compared to $k_F(R_a+R_b+R_{ab})$ in the argument of the sine function and find the final form of the DM coupling is similar to Eq. (26), only the parameter V_1 is modified as follows:

$$V_1 \cong \frac{135\pi}{32}\frac{\lambda_d\Gamma^2}{E_F^2 k_F^3}\frac{\sin[(1+\gamma)(\pi/10)Z_d]}{1+\gamma} \ . \tag{26a}$$

For *Cu*Mn spin-glass alloys with $\Delta=0.25$ eV, $E_F=7$ eV, and $k_F=1.36$ Å^{-1} we find

$$\gamma = 0.0077(R_a+R_b+R_{ab}) \ , \tag{24b}$$

where the distances R_i are given in units of angstroms.

As seen in Sec. III the distances R_i are related to the concentrations of the manganese x, and other impurities y. Provided x and y are greater than 1000 ppm, the correction γ is small. For example, for $x=0.01$ and $y=0.001$ and for a triangle of average length one finds for *Cu* $\gamma \cong 0.24$. Therefore, if we limit ourselves to spin-glass alloys for which x and y are greater than 1000 ppm we can neglect the correction factor γ, as we do in this article, in calculating the coupling between the near lying neighbors. However, for very weak concentrations of impurities y, and in the asymptotic limit as $R\to\infty$, the R_i's increase and the correction factor is important. In this limit the DM coupling goes as

$$H_{\text{DM}}|_{R\to\infty} \sim \frac{1}{R_a R_b R_{ab}(R_a+R_b+R_{ab})} \ . \tag{26b}$$

Thus, the coupling energy when summed over a crystal is finite. If one neglected the correction term γ for the coupling at large R, see Eq. (26), one would find the erroneous result that the coupling energy diverges.

In summary, the form of the DM coupling Eq. (26) has been derived on the assumptions (1) that R is sufficiently large so that one can replace the spherical Bessel and Hankel functions by their asymptotic forms, see Eq. (14), and (2) that R is small enough so that the correction factor γ, see Eq. (24a), can be neglected. These conditions are not overly restrictive and cover the *Cu*Mn$_x T_y$ spin-glass alloys studied with $x<0.1$ and $y>0.001$.

III. MACROSCOPIC ANISOTROPY ENERGY

We consider a spin-glass ternary alloy such as the *Cu*Mn$_x T_y$ alloys investigated by Préjean *et al.*[1] and Okuda and Date.[4] The spins of the Mn impurities interact via the RKKY and DM interactions described above. We write the interactions of a spin $\vec{S}$ with its neighboring spins in the following forms

$$H^{(i)}_{\rm RKKY} = \vec{S}_i \cdot \sum_j J_{ij}\vec{S}_j \equiv \vec{S}_i \cdot \vec{h}_e^{(i)} \ , \tag{30}$$

$$H^{(i)}_{\rm DM} = \sum_{ij} \vec{D}_{ij} \cdot (\vec{S}_i \times \vec{S}_j) \equiv \vec{S}_i \cdot \sum_j (\vec{S}_j \times \vec{D}_{ij}) \equiv \vec{S}_i \cdot \vec{h}_a^{(i)} \ , \tag{31}$$

where $\vec{h}_e^{(i)}$ and $\vec{h}_a^{(i)}$ are effective exchange and anisotropy fields, respectively. The total interaction energy of the spin $\vec{S}_i$ is written as

$$H_{\rm int} = \vec{S}_i \cdot (\vec{h}_e^{(i)} + \vec{h}_a^{(i)}) \ . \tag{32}$$

The crucial difference between $\vec{h}_e$ and $\vec{h}_a$ is that, when the spin system rotates as a whole, $\vec{h}_e$ follows the rotations while $\vec{h}_a$ does not. With only RKKY isotropic interactions, the spin system rotates freely. In contrast, a rotation of the spin system changes the angle between the spins and their anisotropy field $\vec{h}_a^{(i)}$, therefore the DM interactions $\vec{h}_a^{(i)} \cdot \vec{S}_i$ [see Eq. (31)] introduce anisotropy forces. We shall now determine the macroscopic anisotropy energy induced by these DM interactions.

Turning on the anisotropy field $\vec{h}_a$ in addition to the much stronger exchange field $\vec{h}_e$ results in small reorientations of the spins by angles $d\theta_i$ which are of the order of magnitude of h_a/h_e. The maximum energy gain (per magnetic impurity) arising from the orientations is of the order of magnitude of $Sh_a d\theta$, i.e., Sh_a^2/h_e. As the anisotropy energy is only that part of the above stabilization energy which depends on the orientation of the remanent magnetization with respect to the initial field, its maximum possible value per Mn ion corresponds to the mean value of the quantity Sh_a^2/h_e over the sample. As we will now show the consequences of assuming that the anisotropy energy of $Cu\mathrm{Mn}_x T_y$ alloys is proportional to $\langle Sh_a^2/h_e \rangle$ are (i) a contribution to the anisotropy energy proportional to y from the impurity T simply adds to the intrinsic energy of CuMn, (ii) the magnitude of the contribution scales with the *square* of the quantity $\lambda_d \sin[(\pi/10)Z_d]$. These predictions are borne out by the experimental data, which, *a posteriori*, justifies our assumptions.

1. Additivity of the anisotropy energy induced by nonmagnetic impurities and the intrinsic anisotropy energy of "pure" CuMn

Let us suppose that the anisotropy field acting on a magnetic Mn impurity arises from two independent mechanisms

$$\vec{h}_a = \vec{h}_a^{(1)} + \vec{h}_a^{(2)} \ . \tag{33}$$

By independent we mean that there is a random relative orientation of $h_a^{(1)}$ with respect to $h_a^{(2)}$ over the sample. On a particular Mn site, we have

$$\frac{h_a^2}{h_e} = \frac{(h_a^{(1)})^2}{h_e} + \frac{(h_a^{(2)})^2}{h_e} + 2\frac{h_a^{(1)}h_a^{(2)}}{h_e}\cos\theta \ , \tag{34}$$

where θ is the angle between $\vec{h}_a^{(1)}$ and $\vec{h}_a^{(2)}$. By taking into account the random orientation of $\vec{h}_a^{(1)}$ with respect to $\vec{h}_a^{(2)}$, i.e., $\langle\cos\theta\rangle = 0$ we obtain for the mean values over the sample:

$$\left\langle \frac{h_a^2}{h_e} \right\rangle = \left\langle \frac{(h_a^{(1)})^2}{h_e} \right\rangle + \left\langle \frac{(h_a^{(2)})^2}{h_e} \right\rangle \ . \tag{35}$$

As our working assumption is that the anisotropy energy E_a is proportional to $\langle h_a^2/h_e \rangle$, we immediately find from Eq. (35) that the anisotropy energies arising from the two mechanisms are additive.

This additivity has been borne out by the experiments on $Cu\mathrm{Mn}_x T_y$ alloys described above.[1,4] It turns out that the contribution to the anisotropy energy from the nonmagnetic impurities T simply adds to the intrinsic anisotropy energy of the bare CuMn, i.e.,

$$E_a(x,y) = E_a(x,0) + \alpha y \ . \tag{36}$$

The intrinsic anisotropy energy $E_a(x,0)$ of the bare CuMn is probably due either to the dipolar interactions between the Mn moments or to (weak) DM interactions, Eq. (26), associated with Mn triangles. The nonmagnetic impurities T induce an additional anisotropy due to the DM interactions associated with Mn–T–Mn triangles. The observed additivity simply implies that the intrinsic and induced anisotropy fields are not correlated and that Eq. (35) is valid. If the anisotropy energy were not proportional to h_a^2 but, for example, to h_a, the additivity of the anisotropy energies would not be predicted.

2. Dependence of the macroscopic anisotropy energy on concentration

We want to calculate the magnitude of the anisotropy field h_a acting on a given impurity $\mathrm{Mn}^{(0)}$ (at $R=0$) in $Cu\mathrm{Mn}_x T_y$ alloys with $x \gg y$. To do this we have to add up the contributions from all the triangles formed by this impurity $\mathrm{Mn}^{(0)}$ with other Mn and T impurities; the smallest triangles make the strongest contributions. We begin by adding up contributions from all the triangles formed by $\mathrm{Mn}^{(0)}$ with a particular impurity T at distance R from $\mathrm{Mn}^{(0)}$ and any of the numerous Mn impurities which, for $x \gg y$, are inside the sphere of radius R centered on $\mathrm{Mn}^{(0)}$. From Eq. (26) with

$$R_{ab} \equiv |\vec{R}(\mathrm{Mn}^{(0)}) - \vec{R}(\mathrm{Mn})| = r \ ,$$

$$R_a \equiv |\vec{R}(\mathrm{Mn}^{(0)}) - \vec{R}(T)| = R \ ,$$

and

$$R_b \equiv |\vec{R}(\mathrm{Mn}) - \vec{R}(T)| \simeq R \ ,$$

the order of magnitude of the anisotropy field due to a particular triangle is

$$\frac{V_1 S}{rR^2} \ .$$

The number of Mn atoms in the volume between r and $r+dr$ can be written as

$$dn = 4\pi r^2 \rho(x,r)\, dr \ ,$$

where $\rho(x,r)$ is the density of the Mn atoms at distance r from $\mathrm{Mn}^{(0)}$. As the anisotropy fields associated with these Mn atoms are randomly oriented, we have to add up the squares of the anisotropy fields of the individual triangles. Thus we obtain the following contribution from the shell dr

$$\begin{aligned} dh_a^2 &\simeq 4\pi \left(\frac{V_1 S}{rR^2} \right)^2 r^2 \rho(x,r)\, dr \\ &\simeq 4\pi \frac{V_1^2 S^2}{R^4} \rho(x,r)\, dr \ . \end{aligned} \tag{37}$$

It is interesting to note that the contribution of the shell $(r, r+dr)$ does not decrease with r, at least for $r < R$. Thus all the shells up to $r = R$ contribute with equal weight. In contrast, for $r > R$, it can be easily shown in a similar way that the contribution of a shell dr decreases as r^{-2}. Therefore the contribution from the region $r > R$ does not significantly change our final result. In this way we can estimate the contribution from a given impurity T at distance R as

$$h_a(R)^2 \simeq \frac{4\pi V_1^2 S^2}{R^4} \int_0^R \rho(x,r)\, dr \ . \tag{38}$$

As there are many Mn atoms in the sphere $r < R$, we can represent the density as a continuous distribution and write the integral as

$$\int_0^R \rho(x,r)\, dr = \frac{x}{v_a} R \ , \tag{39}$$

where v_a is the atomic volume of the host (Cu). Thus we obtain

$$h_a(R)^2 \simeq 4\pi V_1^2 S^2 \frac{x}{v_a} \frac{1}{R^3} \ . \tag{40}$$

Note that, after taking into account the triangles formed with all the Mn atoms, the contribution of a given atom T at distance R from the central Mn atom is proportional to R^{-3}. The main contribution to the square of the anisotropy field at a Mn atom comes from the nearest neighbor T at distance R_0. For a given concentration y of impurities T, the mean value of R_0^{-3} is

$$\left\langle \frac{1}{R_0^3} \right\rangle \simeq \frac{y}{v_a} \ . \tag{41}$$

By placing Eq. (41) in Eq. (40), we find the following mean value of the square of the anisotropy field

$$\langle h_a^2 \rangle \simeq 4\pi V_1^2 S^2 \frac{xy}{v_a^2} \ . \tag{42}$$

From Eq. (27), the mean value of the isotropic exchange field is

$$\langle h_e \rangle \simeq V_0 S \left\langle \frac{1}{r_0^3} \right\rangle \simeq V_0 S \frac{x}{v_a} \ . \tag{43}$$

Finally, as shown in the beginning of this section the macroscopic anisotropy energy $E_a(x,y)$ per Mn atom is expected to be proportional to the mean value of Sh_a^2/h_e. From Eqs. (42) and (43) we predict,

$$E_a(x,y) \simeq 4\pi \left(\frac{V_1}{V_0} \right)^2 V_0 S^2 \frac{y}{v_a} \ . \tag{44}$$

Therefore the macroscopic anisotropy energy of $Cu\mathrm{Mn}_x T_y$ spin-glass alloys per Mn atom is proportional to the concentration of the impurities T and *independent* of the concentration of the Mn impurities.

Experimentalists characterize the magnetic anisotropy induced by nonmagnetic impurities by the anisotropy per unit *volume*, i.e., for xN Mn atoms,

$$\mathcal{E}_a(x,y) = xNE_a \sim xy \tag{44a}$$

or by the macroscopic anisotropy field

$$H_A \equiv \frac{E_a}{\mu_r} \ , \tag{45}$$

where μ_r is the remanent magnetization per Mn atom. Well below the freezing temperature and under similar freezing conditions, μ_r is expected to be independent of the concentration x of the Mn atoms. From Eq. (44), this leads to

$$H_A(x,y) \sim y \ . \tag{46}$$

The proportionality of $\mathcal{E}_a(x,y)$ and $H_A(x,y)$ to y is observed in the experiments on $Cu\mathrm{Mn}_x T_y$ alloys described in Sec. I. Both the reversal field of the remanent magnetization and the ESR line shift increase linearly with y.[1,4] The dependence on x had not been thoroughly tested in those experiments. However, recent ESR results on $Cu\mathrm{Mn}_x\mathrm{Ni}_y$ alloys have just confirmed that the contribution of Ni impurities to $\mathcal{E}_a$ varies as xy.[10]

Finally, let us consider the contribution to $\mathcal{E}_a$ from DM interactions associated with triangles of Mn impurities, i.e., DM interactions between two Mn arising from weak spin-orbit scattering by a third Mn. By placing $y = x$ in Eq. (44a) we find the contribution to $\mathcal{E}_a$ from such interactions is expected to be proportional to x^2. Thus we find for the total anisotropy energy the following dependence on x and y

$$\mathcal{E}_a = \alpha x^2 + \beta xy \ . \tag{47}$$

This is in agreement with experimental results in

*Cu*Mn$_x$ and *Cu*Mn$_x$Ni$_y$ alloys where it was found that $\beta >> \alpha$.[10] The term proportional to x^2 suggests that the intrinsic anisotropy of *Cu*Mn$_x$ spin-glass alloys could be also due to DM interaction.

IV. NUMERICAL CALCULATIONS AND COMPARISON WITH EXPERIMENTAL DATA

In a previous paper[5] (FL) we calculated the macroscopic anisotropy energy for only *Cu*Mn$_x$$T_x$ alloys. In such alloys with equal concentrations of Mn and T, the mean distance between nearest-neighbor impurities is the same for Mn—Mn pairs and Mn—T pairs

$$R_a \simeq R_b \simeq R_{ab} \sim r_0 = (v_a/x)^{1/3}$$

so that, from Eqs. (26) and (27), the following mean magnitudes of the exchange field, anisotropy field, and macroscopic anisotropy energy per Mn atom can be predicted

$$h_e \simeq V_0 S \frac{x}{v_a} \ , \tag{48a}$$

$$h_a \simeq V_1 S \frac{x}{v_a} \ , \tag{48b}$$

$$E_a(x,x) \sim \frac{S h_e^2}{h_a} \sim \left[\frac{V_1}{V_0}\right]^2 V_0 S^2 \frac{x}{v_a} \ . \tag{49}$$

In FL we extrapolated the variation of the experimental anisotropy energy found in *Cu*Mn$_x$$T_y$ alloys (with $y << x$) to $y = x$. Then we compared the extrapolated value with the prediction of Eq. (49). Here we will compare more directly theory and experiments because, in the previous section, we derived the expected anisotropy energy *Cu*Mn$_x$$T_y$ alloys with $y << x$, which corresponds to the experimental conditions. From Eq. (44), we expect the anisotropy energy per Mn atom is

$$E_a(x,y) \simeq 4\pi \left[\frac{V_1}{V_0}\right]^2 V_0 S^2 \frac{y}{v_a} \ . \tag{50}$$

Alternatively this corresponds to the following rate of increase of E_a with y

$$\left[\frac{dE_a}{dy}\right]_{\text{calc}} \simeq 4\pi \left[\frac{V_1}{V_0}\right]^2 \frac{V_0 S^2}{v_a} \ . \tag{51}$$

By using Eq. (29) we find,[11]

$$\left[\frac{dE_a}{dy}\right]_{\text{calc}} \simeq 4\pi \left[15 \frac{\lambda_d}{E_F} \sin\left[\frac{\pi}{10} Z_d\right]\right]^2 \frac{V_0 S^2}{v_a} \ . \tag{52}$$

We notice that except for a factor of 4π the same expression is obtained for dE_a/dy for $y << x$ from Eq. (51) and for $E_a^{\text{calc}}(x,x)/x$ from Eq. (49).

In comparing the calculated values of dE_a/dy, Eq. (52) to the experimental rates of increase of the anisotropy energy with y, we want primarily to test the predicted dependence of the anisotropy energy on the type of nonmagnetic impurity [mainly through λ_d and Z_d in Eq. (52)]. In Table I and Fig. 1 we present the experimental rates of increase of the anisotropy energy with y together with the values of dE_a/dy calculated for Eq. (52). The experimental rates come from

TABLE I. Calculated and experimental values of the rate of increase of the macroscopic anisotropy energy of *Cu*Mn$_x$$T_y$ alloys with y. The table also gives the values of the parameters Z_d and λ_d which have been used in the calculations (the values of λ_d are atomic ones, see Ref. 12) and the calculated values of V_1/V_0. The calculations were performed by using Eqs. (29) and (52) with the listed values of Z_d and λ_d, $S = 1.88$, $V_0 = 7.5 \times 10^{-37}$ erg cm^3 (see Ref. 13), $E_F = 7$ eV, and $v_A = 1.178 \times 10^{-23}$ cm^3. The experimental values were derived from Refs. 1 and 4 as explained in Appendix D.

				Rate of increase with y of the anisotropy energy per Mn		
T	Z_d	λ_d (eV)	$\frac{V_1}{V_0}$	$\left[\frac{dE_a}{dy}\right]_{\text{calc}}$ (10^{-16} erg)	$\left[\frac{dE_a}{dy}\right]_{\text{expt}}$ (10^{-16} erg)	$\frac{\left[\frac{dE_a}{dy}\right]_{\text{calc}}}{\left[\frac{dE_a}{dy}\right]_{\text{expt}}}$
Ti	2	0.014	0.0176	8.79	0.136	65
Fe	6	0.049	0.100	282	3.1	91
Co	7	0.065	0.113	359	4.3	83
Ni	9.4	0.076	0.0305	26.3	2.1	12.5
Pd	9.4	0.177	0.0711	143	1.75	82
Pt	9.4	0.51	0.205	1186	9.5	124

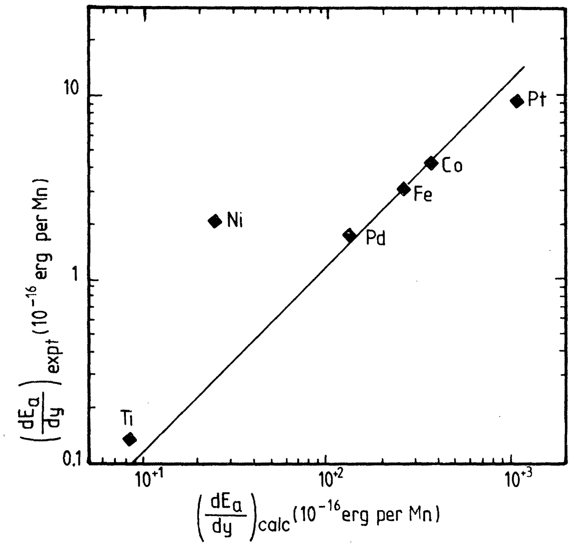

FIG. 1. Experimental vs calculated values of the rate of increase of the macroscopic anisotropy energy with y in $Cu\mathrm{Mn}_xT_y$ alloys. The straight line corresponds to $(dE_a/dy)_{\mathrm{exp}}=(1/85)(dE_a/dy)_{\mathrm{calc}}$ and is a guide to the eye. The numerical values are listed in Table I. The calculation is described in the text and in the caption of Table I. The derivation of the experimental values is explained in Appendix D.

the experiments of Refs. 1 and 4. The analysis of the experimental results is explained in Appendix D. The calculations have been performed by using atomic values of λ_d,[12] values of Z_d indicated in Table I and experimental values of V_0 and S found by Smith[13] and given in the caption of Table I. It can be seen that, with the exception of Ni, *the calculation reproduces very well over two decades the variation of the anisotropy energy with the type of the nonmagnetic impurity T.* This firmly supports our interpretation that the anisotropy energy induced by nonmagnetic impurities is due to DM interactions arising from spin-orbit scattering and, in addition, our assumption that E_a varies as the *square* of the macroscopic anisotropy field.

The predicted numerical values of dE_a/dy are about 80 times larger than the experimental ones. Such a discrepancy is not surprising because we have calculated the maximum possible energy which corresponds to the total stabilization energy and not the part dependent on the orientation of the remanent magnetization. In addition, if one does not assume the optimum configuration that produces the maximum energy gain arising from the reorientation of the spins due to the anisotropy field, but takes an average over all configurations, and if we take account of the increase of the RKKY energy when calculating the stabilization energy our estimate of the anisotropy energy is reduced by a factor of 3. Second, by taking into account the prefactors in the DM interaction [see Eq. (26)], which were ignored in estimating the contributions to the anisotropy field [see Eq. (37)], and by averaging the anisotropy field $|h_a|$ over all orientations of the spins, Caroline Morgan-Pond finds the result, Eq. (40), is reduced by a factor of 6. In any case our results show that the DM interactions are easily strong enough to account for the experimental data.

V. CONCLUSION

We have presented the calculation of the Dzyaloshinsky-Moriya interactions between impurity spins arising from the spin-orbit scattering of conduction electrons by nonmagnetic transition-metal impurities. These interactions which can be remarkably large account for the magnitude of the anisotropy fields measured in $Cu\mathrm{Mn}T$ spin-glass alloys and for their dependence on the nonmagnetic element T.

We are also able to comment on the origin of the anisotropy fields in *binary* spin-glass alloys. In pure CuMn the anisotropy is very small (corresponding to $E_a \sim 0.06\times10^{-18}$ per Mn for 1 at.%, see Ref. 1). It could be due either to weak DM interactions arising from the spin-orbit scattering of neighboring Mn atoms of Mn pairs or to dipolar coupling. In AuFe spin-glass alloys the anisotropy is much larger, corresponding to $E_a \simeq 20\times10^{-18}$ erg per Fe in AuFe 3.7 at.%.[14] The difference with respect to CuMn is that Fe impurities in Au carry a small orbital angular momentum, as has been demonstrated by the existence of skew scattering contributions to the Hall effect.[15] In a future publication we show that the existence of orbital angular momentum results in single-ion anisotropy and pseudodipolar interactions between Fe impurities. The strong anisotropy fields in AuFe spin-glass alloys could be due to these interactions. Alternatively they could also be due to Dzyaloshinsky-Moriya interactions associated with triangles of Fe impurities, i.e., DM interactions between a pair of Fe spins arising from spin-orbit scattering by a third Fe impurity. This latter spin-orbit scattering could be due either to the spin-orbit coupling of the $3d$ electrons or to the spin-orbit coupled band structure of the host (Au). It remains a challenge for us to reliably determine the hierarchy of anisotropic interactions in alloys containing transition-metal magnetic impurities.

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APPENDIX A: DERIVATION OF INTERACTION ENERGY TO THIRD ORDER

At the low temperatures at which most work on spin-glasses is done only the ground state or $T=0$ K state of the electron gas needs to be considered. The third-order correction to the ground-state energy of a gas of conduction electrons due to a perturbation V is[16]

$$E^{(3)}=\left\langle V\frac{1-P_0}{E_0-H_0}V\frac{1-P_0}{E_0-H_0}V\right\rangle - \langle V\rangle\left\langle V\frac{1-P_0}{(E_0-H_0)^2}V\right\rangle \ . \qquad \text{(A1)}$$

The angular brackets denote expectation values taken over the ground state of the system; H_0 is the unperturbed Hamiltonian for the system which is primarily the kinetic energy of the electrons and $(1-P_0)$ is a projection operator which *removes* the ground state when one sums over the intermediate states.

The perturbations we consider involve only one conduction electron, and are written in second quantized form as

$$V=\sum_{\vec{k},\vec{k}'} V_{\vec{k},\vec{k}'}C^{\dagger}_{\vec{k}}C_{\vec{k}'} \ . \qquad \text{(A2)}$$

We do not explicitly show the spin indices here but assume them implicitly. As shown in Sec. II when we include them we must sum over $\sigma,\sigma'=\pm\frac{1}{2}$ in Eq. (A2). We also represent the energy denominator as

$$\frac{1}{E_0-H_0}=-\int_0^\infty d\lambda \exp[(E_0-H_0)\lambda] \ . \qquad \text{(A3)}$$

By using these expressions in Eq. (A1) and the relation for fermion operators

$$C_1^{\dagger}C_2e^{-H_0\lambda}=e^{-H_0\lambda}e^{\hbar\omega_{12}\lambda}C_1^{\dagger}C_2 \ , \qquad \text{(A4)}$$

where $\hbar\omega_{12}=E_1-E_2$, we find

$$E^{(3)}=\int_0^\infty d\lambda\int_0^\infty d\lambda' \sum_{1\cdots 6}(1-\delta_{12})(1-\delta_{56})\exp(\hbar\omega_{12}\lambda-\hbar\omega_{56}\lambda')V_{12}V_{34}V_{56}\langle 0|C_1^{\dagger}C_2C_3^{\dagger}C_4C_5^{\dagger}C_6|0\rangle$$
$$-\sum_3 V_{33}\langle 0|C_3^{\dagger}C_3|0\rangle\int_0^\infty d\lambda\sum_{1,2,5,6}(1-\delta_{12})[\exp(\hbar\omega_{12})^2\lambda]V_{12}V_{56}\langle 0|C_1^{\dagger}C_2C_5^{\dagger}C_6|0\rangle \ , \qquad \text{(A5)}$$

where the ground-state many-body wave function $|0\rangle$ represents the Fermi sea of conduction electrons,

$$|0\rangle=\left(\prod_{k\leq k_F}C_k^{\dagger}\right)|\text{vac}\rangle \ . \qquad \text{(A6)}$$

The nontrivial combinations of creation and annihilation operators entering $E^{(3)}$ are

$$\langle 0|C_1^{\dagger}C_2C_2^{\dagger}C_3C_3^{\dagger}C_1|0\rangle=n_1(1-n_2)(1-n_3) \ , \qquad \text{(A7a)}$$

$$\langle 0|C_1^{\dagger}C_2C_3^{\dagger}C_1C_2^{\dagger}C_3|0\rangle=n_1(1-n_2)(\delta_{13}-n_3) \ , \qquad \text{(A7b)}$$

$$\langle 0|C_1^{\dagger}C_2C_3^{\dagger}C_3C_2^{\dagger}C_1|0\rangle=n_1(1-n_2)(n_3+\delta_{23}-\delta_{13}) \ , \qquad \text{(A7c)}$$

and

$$-\langle 0|C_3^{\dagger}C_3|0\rangle\langle 0|C_1^{\dagger}C_2C_2^{\dagger}C_1|0\rangle=-n_1(1-n_2)n_3 \ , \qquad \text{(A7d)}$$

where n_α is the Fermi distribution function which reduces to a step function ($n_\alpha=1$ if $k\leq k_F$ and 0 otherwise) in the limit $T=0$ K. There is a fair cancellation of terms. It should be noted that the term proportional to δ_{23} in the third process of Eq. (A7c) should not be counted as we consider this diagram in the first process, Eq. (A7a), when $2=3$. By placing the nonzero expectation values Eqs. (A7) in Eq. (A5) and after some cancellations we find

$$E^{(3)}=\sum_{1,2,3}(1-\delta_{12})(1-\delta_{13})\times\frac{n_1(1-n_2)(1-n_3)}{\hbar\omega_{12}\hbar\omega_{13}}V_{12}V_{23}V_{31}-\sum_{1,2,3}(1-\delta_{12})(1-\delta_{23})\times\frac{n_1(1-n_2)n_3}{\hbar\omega_{12}\hbar\omega_{32}}V_{12}V_{23}V_{31} \ . \qquad \text{(A8)}$$

By interchanging indices in the second term we are able to rewrite the third-order correction to the ground-state energy as

$$E^{(3)}=\sum_{\substack{1,2,3\\1\neq 2\\1\neq 3}}\frac{V_{12}V_{23}V_{31}}{\hbar\omega_{12}\hbar\omega_{13}}\times[n_1(1-n_2)(1-n_3)-(1-n_1)n_2n_3] \ . \qquad \text{(A9)}$$

As the energy levels of the conduction electrons form a quasicontinuous spectrum the sums will be replaced by integrals. The restrictions on the sums are accounted for by using the Cauchy principal value of the integrals. As the energy eigenvalues of the unperturbed Hamiltonian are those of the kinetic energy we write Eq. (A9) as

$$E^{(3)}=\left(\frac{1}{8\pi^3}\right)^3\left(\frac{2m}{\hbar^2}\right)^2\int d^3k_1\,\mathcal{P}\int d^3k_2\,\mathcal{P}\int d^3k_3\frac{V(\vec{k}_1,\vec{k}_2)\,V(\vec{k}_2,\vec{k}_3)\,V(\vec{k}_3,\vec{k}_1)}{(k_1^2-k_2^2)(k_1^2-k_3^2)}$$

$$\times\{n(k_1)[1-n(k_2)][1-n(k_3)]-[1-n(k_1)]n(k_2)n(k_3)\}\ ,\qquad\text{(A10)}$$

where $\mathcal{P}$ means the Cauchy principal value of the integral, and the unperturbed energy of the electrons is written as $\hbar^2k^2/2m$. As the direction of the k vectors only enter the matrix elements of the perturbation we can write the energy as a product of radial integrals

$$E^{(3)}=\left(\frac{1}{8\pi^3}\right)^3\left(\frac{2m}{\hbar^2}\right)^2\int_0^\infty dk_1\,\mathcal{P}\int_0^\infty dk_2\,\mathcal{P}\int_0^\infty dk_2\frac{V(k_1k_2k_3)}{(k_1^2-k_2^2)(k_1^2-k_3^2)}$$

$$\times\{n(k_1)[1-n(k_2)][1-n(k_3)]-[1-n(k_1)]n(k_2)n(k_3)\}\ ,\qquad\text{(A11)}$$

where

$$V(k_1k_2k_3)\equiv k_1^2k_2^2k_3^2\int d\Omega_{k_1}\int d\Omega_{k_2}\int d\Omega_{k_3}V(\vec{k}_1,\vec{k}_2)\,V(\vec{k}_2,\vec{k}_3)\,V(\vec{k}_3,\vec{k}_1)$$

and

$$\int d\Omega_k\equiv\int_0^\pi\sin\theta_k\,d\theta_k\int_0^{2\pi}d\phi_k\ .$$

The function $V(k_1k_2k_3)$ possesses the important property that it is invariant under cyclic permutations of the indices 1, 2, and 3, i.e.,

$$V(k_1k_2k_3)=V(k_2k_3k_1)=V(k_3k_1k_2)\ .$$

We assume that $V(k_1k_2k_3)$ does not vanish at $k_1=k_2$ or $k_1=k_3$ and that it is sufficiently regular so as not to give any pathological behavior.

Usual practice is to convert the integral

$$I=\int_0^\infty dk_1\,\mathcal{P}\int_0^\infty dk_2\,\mathcal{P}\int_0^\infty dk_3\frac{V(k_1k_2k_3)}{(k_1^2-k_2^2)(k_1^2-k_3^2)}\{n(k_1)[1-n(k_2)][1-n(k_3)]-[1-n(k_1)]n(k_2)n(k_3)\}\ ,\qquad\text{(A12)}$$

into the form where only $n(k_1)$ appears in the integrand since the resulting integral is often easier to evaluate. However, care must be exercised when we have the possibility of double-pole singularities as is the case above. We will show the above integral can be reduced to

$$I=\int_0^\infty dk_1\,\mathcal{P}\int_0^\infty dk_2\,\mathcal{P}\int_0^\infty dk_3\frac{V(k_1k_2k_3)n(k_1)}{(k_1^2-k_2^2)(k_1^2-k_3^2)}-\frac{\pi^2}{3}\int_0^\infty dk_1\frac{V(k_1k_1k_1)n^2(k_1)[3-2n(k_1)]}{4k_1^2}\ .\qquad\text{(A13)}$$

For the ground-state problem $[n(k)=1,\ k\leq k_F;\ n(k)=0,\ k>k_F]$ we then have

$$I=\int_0^{k_F}dk_1\,\mathcal{P}\int_0^\infty dk_2\,\mathcal{P}\int_0^\infty dk_3\frac{V(k_1k_2k_3)}{(k_1^2-k_2^2)(k_1^2-k_3^2)}$$

$$-\frac{\pi^2}{12}\int_0^{k_F}dk_1\frac{V(k_1k_1k_1)}{k_1^2}\ .\qquad\text{(A14)}$$

We begin by writing the integral Eq. (A12) in the form

$$I=\lim_{\epsilon\to 0}I(\epsilon)\ ,\qquad\text{(A15)}$$

where

$$I(\epsilon)=\underset{\substack{|k_1-k_2|>\epsilon\\|k_1-k_3|>\epsilon}}{\iiint_0^\infty}dk\frac{F(k_1k_2k_3)}{(k_1^2-k_2^2)(k_1^2-k_3^2)}\qquad\text{(A16)}$$

and

$$F(k_1k_2k_3)=V(k_1k_2k_3)\{n(k_1)[1-n(k_2)][1-n(k_3)]-[1-n(k_1)]n(k_2)n(k_3)\}\ . \tag{A17}$$

Next we split $I(\epsilon)$ into two parts

$$I(\epsilon)=I'(\epsilon)-R_1(\epsilon)\ , \tag{A18}$$

where $I'(\epsilon)$ is the integral Eq. (A16) further restricted to the region $|k_2-k_3|>\epsilon$

$$I'(\epsilon)=\int\int_0^\infty\int_{\substack{|k_1-k_2|>\epsilon\\|k_1-k_3|>\epsilon\\|k_2-k_3|>\epsilon}} dk\,\frac{F(k_1k_2k_3)}{(k_1^2-k_2^2)(k_1^2-k_3^2)}\ , \tag{A19}$$

and $R_1(\epsilon)$ is the remainder

$$R_1(\epsilon)=-\int\int_0^\infty\int_{\substack{|k_1-k_2|>\epsilon\\|k_1-k_3|>\epsilon\\|k_2-k_3|<\epsilon}} dk\,\frac{F(k_1k_2k_3)}{(k_1^2-k_2^2)(k_1^2-k_3^2)}\ . \tag{A20}$$

Now the domain of integration for $I'(\epsilon)$ is symmetric under cyclic permutations of the k_i's and we write the integrand F in Eq. (A19) in two parts

$$I'(\epsilon)=\int\int_0^\infty\int_{|k_1-k_j|>\epsilon} dk\,\frac{V(k_1k_2k_3)n(k_1)}{(k_1^2-k_2^2)(k_1^2-k_3^2)}-\int\int_0^\infty\int_{|k_i-k_j|>\epsilon} dk\,\frac{\bar{F}(k_1k_2k_3)}{(k_1^2-k_2^2)(k_1^2-k_3^2)}\ , \tag{A21}$$

where

$$\begin{aligned}\bar{F}(k_1k_2k_3)&=-F+Vn(k_1)\\&=V(k_1k_2k_3)\\&\quad\times\{-2n(k_1)n(k_2)n(k_3)+n(k_1)n(k_2)\\&\quad+n(k_2)n(k_3)+n(k_3)n(k_1)\}\ .\end{aligned}$$

Now $\bar{F}$ is also symmetric under cyclic permutations of k_i's. Therefore the second integral of Eq. (A21) can be shown to vanish by adding the three identical terms generated by cyclic permutation of k_i's. For the remaining first term of Eq. (A21) we write

$$I'(\epsilon)=\int\int_0^\infty\int_{\substack{|k_1-k_2|>\epsilon\\|k_1-k_3|>\epsilon}} dk\,\frac{V(k_1k_2k_3)n(k_1)}{(k_1^2-k_2^2)(k_1^2-k_3^2)}-\int\int_0^\infty\int_{\substack{|k_1-k_2|>\epsilon\\|k_1-k_3|>\epsilon\\|k_2-k_3|<\epsilon}} dk\,\frac{V(k_1k_2k_3)n(k_1)}{(k_1^2-k_2^2)(k_1^2-k_3^2)}\ . \tag{A22}$$

Thus from Eqs. (A18), (A20), and (A22) we find

$$I(\epsilon)=\int\int_0^\infty\int_{\substack{|k_1-k_2|>\epsilon\\|k_1-k_3|>\epsilon}} dk\,\frac{V(k_1k_2k_3)n(k_1)}{(k_1^2-k_2^2)(k_1^2-k_3^2)}-R(\epsilon)\ , \tag{A23}$$

where

$$R(\epsilon)=\int\int_0^\infty\int_{\substack{|k_1-k_2|>\epsilon\\|k_1-k_3|>\epsilon\\|k_2-k_3|<\epsilon}} dk\,\frac{\bar{F}(k_1k_2k_3)}{(k_1^2-k_2^2)(k_1^2-k_3^2)}\ , \tag{A24}$$

and $\bar{F}$ is given by Eq. (A21). The first term of Eq. (A23) reduces in the limit $\epsilon\to 0$ to the first term of Eq. (A13), and our task has been reduced to showing that

$$\lim_{\epsilon\to 0}R(\epsilon)=\frac{\pi^2}{3}\int_0^\infty dk_1\frac{\bar{F}(k_1k_1k_1)}{4k_1^2}\ . \tag{A25}$$

First we give an heuristic argument for Eq. (A25). The domain of integration in Eq. (A24) is, for a given k_1, over a thin strip of width $\sim O(\epsilon)$ in the (k_2,k_3) plane as shown in Fig. 2. Therefore, except for the double pole region $k_2\approx k_3\approx k_1$ the integral should vanish as $\epsilon\to 0$ and any nonzero contribution should come from the region where k_2,k_3 are close to k_1. Thus we may write

$$\begin{aligned}R(\epsilon)&=\int_0^\infty dk_1\int\int_S dk_2\,dk_3\frac{\bar{F}(k_1k_2k_3)}{(k_1+k_2)(k_1+k_3)}\\&\quad\times\frac{1}{(k_1-k_2)(k_1-k_3)}\\&\approx\int_0^\infty dk_1\frac{\bar{F}(k_1k_1k_1)}{4k_1^2}\int\int_S\frac{dk_2dk_3}{(k_1-k_2)(k_1-k_3)}\ ,\end{aligned} \tag{A26}$$

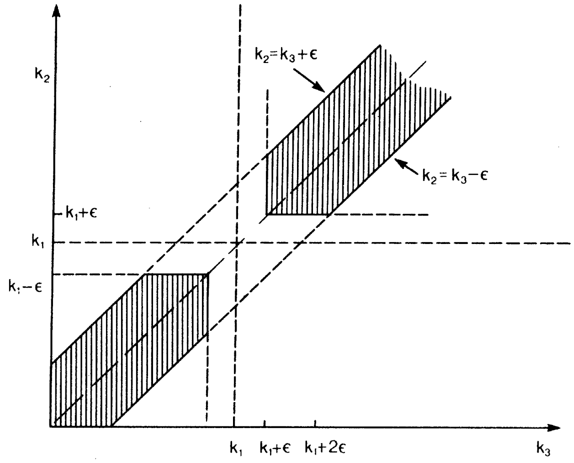

FIG. 2. Domain of integration S for the integrals in Eq. (A26).

where we assume that the regular parts of the integrand do not change appreciably over the region where most of the contribution to $R(\epsilon)$ comes from, and S denotes the domain of integration shown in Fig. 2. Now we consider the integration

$$\iint_S \frac{dk_2 dk_3}{(k_1-k_2)(k_1-k_3)} \equiv a(\epsilon) \quad . \tag{A27}$$

Let us define

$$k_2 \equiv k_1+\epsilon y, \quad k_3 \equiv k_1+\epsilon z \quad ,$$
$$dk_2=\epsilon dy, \text{ and } dk_3=\epsilon dz \quad . \tag{A28}$$

Then we write Eq. (A27) as

$$a(\epsilon)=\iint\limits_{\substack{-k_1/\epsilon \\ |y|>1 \\ |z|>1 \\ |y-z|<1}}^{\infty} \frac{dydz}{yz} \quad , \tag{A29}$$

which in the limit $\epsilon \rightarrow 0$ becomes

$$\lim_{\epsilon\rightarrow 0} a(\epsilon)=\iint\limits_{\substack{-\infty \\ |y|>1 \\ |z|>1 \\ |y-z|<1}}^{\infty} \frac{dydz}{yz} \quad . \tag{A30}$$

Now, as seen from Fig. 3 this integral has the following limits of integration

$$\iint\limits_{\substack{-\infty \\ |y|>1 \\ |z|>1 \\ |y-z|<1}}^{\infty} \frac{dydz}{yz}=\int_{-\infty}^{-1} dy \int_{y-1}^{\min(-1,y+1)} dz\frac{1}{yz}$$
$$+\int_1^{\infty} dy \int_{\max(1,y-1)}^{y+1} dz\frac{1}{yz} \quad . \tag{A31}$$

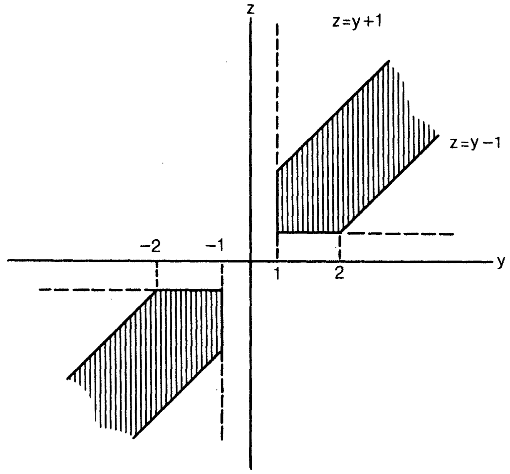

FIG. 3. Domain of integration for integrals in Eq. (A31).

The first term can be shown to be identical to the second by changing the variables $y \rightarrow -y'$, $z \rightarrow -z'$. As seen from Fig. 3

$$\int_1^{\infty} dy \int_{\max(1,y-1)}^{y+1} dz\frac{1}{yz}$$
$$=\int_1^2 dy\,\frac{1}{y}(\ln z)_1^{y+1}+\int_2^{\infty} dy\,\frac{1}{y}(\ln z)_{y-1}^{y+1}$$
$$=\int_1^2 dy\,\frac{\ln(y+1)}{y}+\int_2^{\infty} dy\,\frac{\ln(y+1)-\ln(y-1)}{y}$$
$$=\int_1^{\infty} dy\,\frac{\ln[(y+1)/(y-1)]}{y}+\int_1^2 dy\,\frac{\ln(y-1)}{y} \quad ,$$

and by making the substitution $y=t^{-1}$ in the first integral and $y=t'+1$ in the second we find

$$=\int_0^1 dt\,\frac{\ln[(1+t)/(1-t)]}{t}+\int_0^1 dt'\,\frac{\ln t'}{t'+1} \quad ,$$

which by partial integration of the second integral yields

$$=\int_0^1 dt\,\frac{\ln(1+t)}{t}-\int_0^1 dt\,\frac{\ln(1-t)}{t}$$
$$+[\ln t'\ln(t'+1)]_0^1-\int_0^1 dt'\,\frac{\ln(t'+1)}{t'}$$
$$=-\int_0^1 dt\,\frac{\ln(1-t)}{t} \quad .$$

This integral is evaluated by writing the integrand as a power series in t, and

$$\int_1^{\infty} dy \int_{\max(1,y-1)}^{y+1} dz\frac{1}{yz}=\int_0^1 dt\,\frac{1}{t}\sum_{n=1}^{\infty}\frac{t^n}{n}$$
$$=\sum_{n=1}^{\infty}\frac{1}{n}\int_0^1 t^{n-1}\,dt$$
$$=\sum_{n=1}^{\infty}\frac{1}{n^2}=\frac{\pi^2}{6} \quad . \tag{A32}$$

As $a(\epsilon)$ Eq. (A30) is twice this value, see Eq. (A31), $a(\epsilon)$ reduces to $\pi^2/3$ as $\epsilon \rightarrow 0$ and $R(\epsilon)$ Eq. (A26) gives the desired result Eq. (A25).

To be more precise, we have from Eqs. (A24), (A30), and from the transformation Eq (A28),

$$R(\epsilon)=\int_0^{\infty} dk_1 \int\limits_{\substack{-k_1/\epsilon \\ |y|>1 \\ |z|>1 \\ |y-z|<1}}^{\infty} dy \int_{-k_1/\epsilon}^{\infty} dz\,\frac{\bar{F}(k_1,k_1+\epsilon y,k_1+\epsilon z)}{(2k_1+\epsilon y)(2k_1+\epsilon z)}\frac{1}{yz}=\int_0^{\infty} dk_1 \iint\limits_{\substack{-\infty \\ |y|>1 \\ |z|>1 \\ |y-z|<1}}^{\infty} dy\,dz\,\frac{\bar{\bar{F}}(\epsilon y,\epsilon z,k_1)}{yz}+R'(\epsilon) \quad , \tag{A33}$$

where we have extended the lower limits of integration to $-\infty$, and $R'(\epsilon)$ is the correction term. Here we have written

$$\bar{\bar{F}}(y,z,k_1) \equiv \frac{\bar{F}(k_1,k_1+y,k_1+z)}{(2k_1+y)(2k_1+z)} \quad .$$

By using an argument similar to the one given below Eq. (A25) one convinces oneself that $R'(\epsilon)$ is of the order of ϵ. We then have

$$\lim_{\epsilon\to 0} R(\epsilon)=\lim_{\epsilon\to 0}\int_0^\infty dk_1 \iint_{\substack{-\infty\\ |y|>1\\ |z|>1\\ |y-z|<1}}^{\infty} dy\, dz\, \frac{\bar{\bar{F}}(\epsilon y,\epsilon z,k_1)}{yz} .$$

Thus if one assumes here that $\bar{\bar{F}}(\epsilon y,\epsilon z,k_1)$ uniformly converges to $F(0,0,k_1)$ as $\epsilon\to 0$ in the domain of integration (see Figs. 2 and 3), then we can interchange the limit as $\epsilon\to 0$ and the integration. This gives us

$$\lim_{\epsilon\to 0} R(\epsilon)=\int_0^\infty dk_1 \iint_{\substack{-\infty\\ |y|>1\\ |z|>1\\ |y-z|<1}}^{\infty} dy\, dz\, \frac{\bar{\bar{F}}(0,0,k_1)}{yz}$$

$$=\frac{\pi^2}{3}\int_0^\infty dk_1\, \bar{\bar{F}}(0,0,k_1) ,$$

which is just the sought after result Eq. (A25). For most of our applications, the uniform convergence criterion is satisfied for any reasonable $V(k_1k_2k_3)$.

In summary we have shown that the integral entering the third-order energy correction, Eq. (A12) can be written in the desired form Eq. (A14). Thus we can write the third-order energy correction as

$$E^{(3)}=\left(\frac{1}{8\pi^3}\right)^3\left(\frac{2m}{\hbar^2}\right)^2 \times\int_0^{k_F} dk_1\left[\int_0^\infty dk_2\int_0^\infty dk_3\left(\frac{\mathcal{P}}{(k_1^2-k_2^2)}\frac{\mathcal{P}}{(k_1^2-k_3^2)}-\frac{\pi^2}{12k_1^2}\delta(k_2-k_1)\delta(k_3-k_2)\right)V(k_1k_2k_3)\right] . \tag{A34}$$

Equivalently we can write this as integrals over d^3k,

$$E^{(3)}=\left(\frac{1}{8\pi^3}\right)^3\int_{k_1\le k_F} d^3k_1\left[\int d^3k_2\int d^3k_3\left(\frac{\mathcal{P}}{(E_1-E_2)}\frac{\mathcal{P}}{(E_1-E_3)}-\frac{\pi^2}{3}\delta(E_2-E_1)\delta(E_3-E_2)\right) \times V(\vec{\mathrm{k}}_1,\vec{\mathrm{k}}_2)V(\vec{\mathrm{k}}_2,\vec{\mathrm{k}}_3)V(\vec{\mathrm{k}}_3,\vec{\mathrm{k}}_1)\right] , \tag{A35}$$

by using the definition of the Dirac δ function

$$\delta(k)=(\hbar^2/2m)2k\,\delta(E)$$

and by using the definition of $V(k_1k_2k_3)$ [see Eq. (A11)].

APPENDIX B: ALTERNATE DERIVATION OF COUPLING

An alternate way of deriving the anisotropic coupling between Mn spins due to the presence of a transition-metal impurity is to take into account the change in the conduction-electron wave functions due to the potential and spin-orbit scattering by the impurities d levels in the second-order correction to the ground-state energy. The second-order correction to an electron gas is[16]

$$E^{(2)}=\left\langle V\frac{1-P_0}{E_0-H_0}V\right\rangle . \tag{B1}$$

By using techniques analogous to those used in Appendix A, we find

$$E^{(2)}=\sum_{\vec{\mathrm{k}}\sigma} f(k)\left\langle\vec{\mathrm{k}}\sigma\right|V\frac{(1-|\vec{\mathrm{k}}\sigma\rangle\langle\vec{\mathrm{k}}\sigma|)}{E_k-H_0}V\left|\vec{\mathrm{k}}\sigma\right\rangle=\left(\frac{1}{8\pi^3}\right)^2\int_{k\le k_F} d^3k\,\mathcal{P}\int d^3k'\sum_{\sigma\sigma'}\frac{V(\vec{\mathrm{k}}\sigma,\vec{\mathrm{k}}'\sigma')V(\vec{\mathrm{k}}'\sigma',\vec{\mathrm{k}}\sigma)}{E_{k\sigma}-E_{k'\sigma'}} . \tag{B2}$$

This expression when evaluated by using the conduction-electron-Mn spin interactions at sites A and B leads directly to the RKKY coupling.

We can replace the integrations over $\vec{\mathrm{k}}$ space by those over energy as follows

$$\left(\frac{1}{8\pi^3}\right)d^3k=\frac{1}{4\pi}\rho(\epsilon,\hat{k})\,d\epsilon\, d\Omega_k . \tag{B3}$$

For spherical bands the density of states $\rho(\epsilon,\hat{k})$ does not depend on $\hat{k}$ and following Coqblin and Schrieffer[17] we can write

$$\frac{1}{4\pi}\int d\Omega_k\,|\vec{\mathrm{k}}\sigma\rangle\langle\vec{\mathrm{k}}\sigma|\equiv\sum_{lm}|klm\,\sigma\rangle\langle klm\,\sigma|$$

$$=\sum_{lm}|\epsilon lm\,\sigma\rangle\langle\epsilon lm\,\sigma| , \tag{B4}$$

where $\epsilon=\hbar^2k^2/2m$. With these definitions the second-order correction can be written as integrals over the energy

$$E^{(2)}=\int_0^{E_F}\rho(\epsilon)\,d\epsilon\,\mathcal{P}\int_0^{\infty}\rho\frac{(\epsilon')d\epsilon'}{\epsilon-\epsilon'}\times\sum_{\substack{lm\sigma\\ l'm'\sigma'}}V(\epsilon lm\sigma,\epsilon' l'm'\sigma')\times V(\epsilon' l'm'\sigma',\epsilon lm\sigma)\ . \tag{B5}$$

When we take into account the spin-orbit coupling of the transition-metal impurity in the zeroth-order Hamiltonian we must use spin-orbit coupled basis states, i.e., $|kljm_j\rangle$ states defined as follows

$$|kljm_j\rangle=\sum_{m\sigma}\langle lm\tfrac{1}{2}\sigma|l\tfrac{1}{2}jm_j\rangle\,|klm\sigma\rangle\ , \tag{B6}$$

where $j=l\pm\frac{1}{2}$. In these basis states the second-order correction to the ground-state energy is

$$E^{(2)}=\sum_{jj'}\int_0^{E_F}\rho^j(\epsilon)\,d\epsilon\times\mathcal{P}\int_0^{\infty}\rho^{j'}\frac{(\epsilon')\,d\epsilon'}{\epsilon-\epsilon'}\times\sum_{m_jm_j'}V(\epsilon jm_j,\epsilon' j'm_j')\,V(\epsilon' j'm_j',\epsilon jm_j)\ . \tag{B7}$$

For our problem the effects of the spin-orbit coupling are contained in the phase shifts $\eta_j(k)$ of the conduction-electron states. The density of states for the conduction electrons in the presence of the potential and spin-orbit scattering of the impurity is[9]

$$\rho^j(\epsilon)=\rho^0(\epsilon)+\rho^j_{\mathrm{vbs}}(\epsilon)\ , \tag{B8}$$

where $\rho^0(\epsilon)$ is the density of states for the conduction electrons along and $\rho^j_{\mathrm{vbs}}(\epsilon)$ represents the Lorentzian hump due to the perturbing influence of the spin-orbit coupled impurity. As

$$\int\rho^j_{\mathrm{vbs}}(\epsilon)\,d\epsilon\sim O(1)\ ,$$

while

$$\int\rho^0(\epsilon)\,d\epsilon\sim O(N)\ ,$$

we can safely neglect $\rho^j_{\mathrm{vbs}}(\epsilon)$ and use the free-electron density of states $\rho^0(\epsilon)$ in Eq. (B7). Therefore we find

$$E^{(2)}=\int_0^{E_F}\rho^0(\epsilon)\,d\epsilon\,\mathcal{P}\int_0^{\infty}\frac{\rho^0(\epsilon')d\epsilon'}{\epsilon-\epsilon'}\times\sum_{\substack{jm_j\\ j'm_j'}}V(\epsilon jm_j,\epsilon' j'm_j')\times V(\epsilon' j'm_j',\epsilon jm_j)\ . \tag{B9}$$

We conclude that the effects of the spin-orbit coupling of the impurity are accounted for in the second-order correction to the ground-state energy by using distorted wave function with j dependent phase shifts.

Before we proceed to do this we will show that the terms in Eq. (B9) linear in λ are identical to the third-order energy expression Eq. (A35). Towards this end we revert to a discrete sum notation and to distorted wave basis functions $\phi_{k\sigma}(\lambda)$ by using the identity

$$\sum_{kljm_j}|kljm_j\rangle\langle kljm_j|=\sum_{klm\sigma}|klm\sigma\rangle\langle klm\sigma|\ .$$

In this way we find the second-order energy is written as

$$E^{(2)}=\sum_{\substack{\vec k\sigma\\ \vec k'\sigma'\\ k'\neq k}}f(k)\frac{{}_\lambda\langle\vec k\sigma|V|\vec k'\sigma'\rangle_\lambda\,{}_\lambda\langle\vec k'\sigma'|V|\vec k\sigma\rangle_\lambda}{E_k-E_{k'}}\ . \tag{B9$'$}$$

From first-order perturbation theory the wave function correct to $O(\lambda)$ is

$$|k\rangle_\lambda=|k\rangle+\sum_{k''}\frac{V_\lambda(k'',k)}{E_k-E_{k''}}|k''\rangle\ , \tag{B10}$$

where the unperturbed wave functions $|k\rangle$ contain the spin variable, i.e., $|k\rangle\equiv|\vec k,\sigma\rangle$ and

$$\langle k''|V_\lambda|k\rangle\equiv V_\lambda(k'',k)\ .$$

By placing this expansion for the wave function in the second-order energy Eq. (B9′) we find

$$E^{(2)} = \sum_{\substack{k,k' \\ k'\neq k}} f(k)\frac{V(k,k')V(k',k)}{E_k - E_{k'}}$$

$$+ \sum_{\substack{k,k',k'' \\ k'\neq k \\ k''\neq k}} f(k)\left\{\frac{V(k,k')V(k',k'')V_\lambda(k'',k) + V_\lambda(k,k'')V(k'',k')V(k',k)}{(E_k - E_{k'})(E_k - E_{k''})}\right.$$

$$\left. + \frac{V(k,k')V_\lambda(k',k'')V(k'',k) + V(k,k'')V_\lambda(k'',k')V(k',k)}{(E_k - E_{k'})(E_{k'} - E_{k''})}\right\} + O(\lambda^2) \ . \qquad \text{(B11)}$$

The first term yields the conventional RKKY isotropic spin coupling. It remains to show that the second term is identical to Eq. (A35). While the denominator of the first term in the large parentheses is of the correct form, i.e., the energy difference between ground and excited states, the denominator of the second must be recast. By a change of variables the second term can be written as

$$\sum_{\substack{kk'k'' \\ k'\neq k \\ k''\neq k}} f(k)V(k,k')V_\lambda(k',k'')V(k'',k)\left[\frac{1}{(E_k - E_{k'})(E_{k'} - E_{k''})} - \frac{1}{(E_k - E_{k''})(E_{k'} - E_{k''})}\right] \ . \qquad \text{(B12)}$$

When the sums are written as integrals the restrictions $k'\neq k$, $k''\neq k$ lead to Cauchy principal values of the integrals over d^3k' and d^3k'', i.e., Eq. (B12) is written as

$$\left(\frac{1}{8\pi^3}\right)^3 \int_{k\leq k_F} d^3k\,\mathcal{P}\int d^3k'\,\mathcal{P}\int d^3k''\frac{1}{E_{k'} - E_{k''}}\left[\frac{1}{E_k - E_{k'}} - \frac{1}{E_k - E_{k''}}\right] V(kk')V_\lambda(k'k'')V(k''k) \ . \qquad \text{(B13)}$$

As shown by Fano[18] the principal values of this combination of energy denominators can be rewritten as

$$\frac{\mathcal{P}}{E_{k'} - E_{k''}}\left[\frac{\mathcal{P}}{E_k - E_{k'}} - \frac{\mathcal{P}}{E_k - E_{k''}}\right] = \frac{\mathcal{P}}{E_k - E_{k'}}\,\frac{\mathcal{P}}{E_k - E_{k''}} - \pi^2\delta(E_{k'} - E_k)\delta(E_{k''} - E_{k'}) \ . \qquad \text{(B14)}$$

By placing this result in Eq. (B13) and writing the remaining sum in Eq. (B11) as integrals we find the part of $E^{(2)}$ linear in λ is

$$E^{(2)}(\lambda) = \left(\frac{1}{8\pi^3}\right)^3 \int_{k\leq k_F} d^3k \int d^3k' \int d^3k'' \left\{\mathcal{P}\mathcal{P}\frac{1}{(E_k - E_{k'})(E_k - E_{k''})}[V(k,k')V(k',k'')V_\lambda(k'',k)\right.$$

$$+ V_\lambda(k,k')V(k',k'')V(k'',k)$$

$$+ V(k,k')V_\lambda(k',k'')V(k'',k)]$$

$$\left. - \pi^2\delta(E_{k'} - E_k)\delta(E_{k''} - E_{k'})V(k,k')V_\lambda(k,k'')V(k'',k)\right\} \ . \qquad \text{(B15)}$$

In the present application

$$V = -\Gamma\delta(\vec{r} - \vec{R}_a)\vec{s}\cdot\vec{S}_a - \Gamma\delta(\vec{r} - \vec{R}_b)\vec{s}\cdot\vec{S}_b = V_A + V_B \ , \qquad \text{(B16)}$$

and we will denote the six permutation of the matrix elements of V_A, V_B, and V_λ by

$$P_{\alpha\beta\gamma} V_\alpha(k,k')\, V_\beta(k'k'')\, V_\gamma(k''k) \ .$$

By recognizing that the term in the integrand with the δ function is the same for all six permutations we can simplify Eq. (B15) by introducing a factor of $\frac{1}{3}$ and writing the energy as

$$E^{(2)}(\lambda)=\left[\frac{1}{8\pi^3}\right]^3 \int_{k\leq k_F} d^3k \int d^3k' \int d^3k'' \left[\left[\frac{\mathcal{P}}{E_k-E_{k'}}\,\frac{\mathcal{P}}{E_k-E_{k''}}-\frac{\pi^2}{3}\delta(E_{k'}-E_k)\,\delta(E_{k''}-E_{k'})\right] \right.$$

$$\left. \times P_{\alpha\beta\gamma} \sum_{\sigma\sigma'\sigma''} V_\alpha(\vec{k}\sigma,\vec{k}'\sigma')\, V_\beta(\vec{k}'\sigma',\vec{k}''\sigma'')\, V_\gamma(\vec{k}''\sigma'',\vec{k}\sigma)\right] \ , \tag{B17}$$

where we have explicitly inserted vectors over labels for wave functions and included the spin variables. This coupling is identical to that derived from third-order perturbation theory when the potential Eq. (2) is inserted into Eq. (A35) and only the trilinear terms in S_a, S_b, and λ are kept. Thus we have shown in principle that if one takes the second-order energy expression, Eq. (B2), replaces the wave functions $|k\sigma\rangle$ by spin-orbit coupled functions $|k\sigma\rangle_\lambda$ and linearizes the result so as to keep only terms to first order in λ, one arrives at the same trilinear coupling as that found using third-order perturbation theory. We now explicitly show that this is the case.

To represent the potential and spin-orbit scattering effects of the transition-metal impurity on the conduction electrons we introduce phase shifts which now depend on the number j. For the Mn ions at distances $R_a \sim R_b$ relatively far from the impurity Eq. (3b) is the proper form of the wave function. When we make the spherical wave expansion of the plane wave (about the origin $R=0$) and couple the individual angular momenta to the spin we write the distorted wave function as

$$\phi_{\vec{k}\sigma}=4\pi \sum_{jl} i^l[j_l(kr)+\delta_{l2}e^{i\eta_j}\sin\eta_j h_2^{(+)}(kr)] \sum_{mm_j} Y^*_{lm}(\hat{k})\,\langle jm_j|lm\sigma\rangle\,|jm_j\rangle \tag{B18}$$

$$=-\frac{4\pi}{kr}\sum_j e^{i\eta_j}\sin(kr+\eta_j)\sum_{mm_j} Y^*_{2m}(\hat{k})\,\langle jm_j|m\sigma\rangle\,|jm_j\rangle+\cdots \quad (l\neq 2) \ . \tag{B19}$$

The dominant phase shifts produced by the impurity are for $l=2$, and the spin-orbit coupling information is carried by the phase shifts η_j, where

$$\eta_j(k)=\tan^{-1}[\Delta/(E_{rj}-E_k)] \ , \tag{B20}$$

for $j=l\pm\frac{1}{2}=\frac{5}{2},\frac{3}{2}$. Therefore we are only interested in the $l=2$ part of the wave function Eq. (B18), as the other terms do not contain the spin-orbit parameter λ and do not produce an anisotropic coupling between the Mn spins.

To obtain the leading term in $1/R$ and linear in λ of the anisotropic pair coupling, we need consider only one of the four states in Eq. (B9′) of the form of Eq. (B19). For the others it is sufficient to use plane wave states. When one performs the angular integrations over k space implied in the sums it becomes apparent that only the $l=2$ part of the plane wave contributes to the coupling. This term with $l=2$ is just that given by Eq. (B19) when one sets $\eta_j=0$. As we must consider the spin-orbit coupled states in all four positions in Eq. (B9′) and as we eventually linearize our results, it is expedient to consider two of the four states of the form of Eq. (B19). In this manner we simultaneously pick up contributions from two of the four ways of introducing the spin-orbit coupled state in Eq. (B9′). With the above in mind we can write the second-order energy expression which produces the leading contribution in $1/R$ and linear in λ as

$$E^{(2)}=\left[\frac{1}{8\pi^3}\right]^2\left[\frac{\Gamma}{N}\right]^2\int_{k\leq k_F} d^3k\,\mathcal{P}\int\frac{d^3k'}{E_k-E_{k'}}\sum_{\sigma\sigma'}\{\exp[i\vec{k}'\cdot(\vec{R}_b-\vec{R}_a)]\,\langle\sigma|\vec{S}_a\cdot\vec{s}|\vec{k}'\sigma'\rangle_\lambda\,{}_\lambda\langle\vec{k}'\sigma'|\vec{S}_b\cdot\vec{s}|\sigma\rangle$$

$$+\exp[-i\vec{k}'\cdot(\vec{R}_b-\vec{R}_a)]\,{}_\lambda\langle\vec{k}\sigma|\vec{S}_a\cdot\vec{s}|\sigma'\rangle\,\langle\sigma'|\vec{S}_b\cdot\vec{s}|\vec{k}\sigma\rangle_\lambda+A\rightleftarrows B\} \ . \tag{B21}$$

We use plane waves $e^{i\vec{k}\cdot\vec{r}}$ for two states and Eq. (B19) for the other two. By inserting the states Eq. (B19) and performing the angular integrations in k space we find for the first term of Eq. (B21)

$$E_1^{(2)}=\left[\frac{1}{8\pi^3}\right]^2\left[\frac{\Gamma}{N}\right]^2\frac{(4\pi)^3}{R_aR_bR_{ab}}\int_0^{k_F}k\,dk\,\sin kR_{ab}\,\mathcal{P}\int_0^{\infty}\frac{dk'}{E_k-E_{k'}}\sum_j\sin(k'R_a+\eta_j)\sin(k'R_b+\eta_j)$$

$$\times\sum_{m_j}\langle jm_j(R_b)|(\vec{S}_b\cdot\vec{s})(\vec{S}_a\cdot\vec{s})|jm_j(R_a)\rangle\ ,\quad\text{(B22)}$$

where

$$|jm_j(R)\rangle\equiv\sum_{m\sigma}\langle m\sigma|jm_j\rangle Y_{2m}(R)|\sigma\rangle\ .$$

To arrive at this result we used the relations

$$\int d\Omega_{k'}\,Y_{2m}^*(k')Y_{2m'}(k')=\delta_{mm'}\ ,$$

$$\int d\Omega_k\exp[i\vec{k}\cdot(\vec{R}_b-\vec{R}_a)]=4\pi\frac{\sin kR_{ab}}{kR_{ab}}\ ,$$

and

$$\sum_{m'\sigma'}\langle jm_j|m'\sigma'\rangle\langle m'\sigma'|j'm_j'\rangle=\delta_{jj'}\delta_{m_jm_j'}\ .$$

We evaluate the matrix elements of the conduction-electron spin operator by using the identity

$$(\vec{S}_b\cdot\vec{s})(\vec{S}_a\cdot\vec{s})=\tfrac{1}{4}\vec{S}_a\cdot\vec{S}_b+(i/2)\vec{s}\cdot(\vec{S}_b\times\vec{S}_a)\ .$$

By keeping only the anisotropic term of interest to us (the isotropic term makes an additional contribution of the RKKY coupling but has a different range dependence) and by recoupling angular momenta with the aid of $6j$ symbols we find for general l

$$\sum_{m_j}\langle jm_j(R_b)|\vec{s}|jm_j(R_a)\rangle=\frac{(-1)^l(2j+1)\langle\vec{l}\cdot\vec{s}\rangle_j}{2\sqrt{6}[l(l+1/2)(l+1)]^{1/2}}$$

$$\times[Y^l(\hat{R}_a)\times Y^l(\hat{R}_b)]^1\ ,$$

where

$$\langle\vec{l}\cdot\vec{s}\rangle_j=\tfrac{1}{2}[j(j+1)-l(l+1)-s(s+1)]\ .$$

For our case $l=2$ and by using Eq. (16) we find this becomes

$$\sum_{j_j}\langle jm_j(R_b)|\vec{s}|jm_j(R_a)\rangle$$

$$=(1/16\pi)(2j+1)\langle\vec{l}\cdot\vec{s}\rangle_j\hat{R}_a\cdot\hat{R}_b(\hat{R}_a\times\hat{R}_b)\ .$$

$$\text{(B23)}$$

By placing this result in Eq. (B22) we find the contribution of the first term of Eq. (B21) to the *anisotropic* coupling of Mn spins is

$$E_1^{(2)}=-\frac{(\Gamma/N)^2(2m/\hbar^2)}{16\pi^4R_aR_bR_{ab}}$$

$$\times\left[\sum_j(2j+1)\langle\vec{l}\cdot\vec{s}\rangle_jI_1(j)\right]$$

$$\times\hat{R}_a\cdot\hat{R}_b(\hat{R}_a\times\hat{R}_b)\cdot(\vec{S}_a\times\vec{S}_b)\ ,\quad\text{(B24)}$$

where

$$I_1(j)\equiv\int_0^{k_F}\sin kR_{ab}\,k\,dk\,\mathcal{P}\int_0^{\infty}\frac{dk'}{k^2-k'^2}\sin(k'R_a+\eta_j)$$

$$\times\sin(k'R_b+\eta_j)\ .\quad\text{(B25)}$$

The principal value integral is solved by first rewriting

$$\sin(k'R_a+\eta_j)\sin(k'R_b+\eta_j)$$

$$=-\tfrac{1}{2}\{\cos[k'(R_a+R_b)+2\eta_j]-\cos k'(R_a-R_b)\}\ .$$

$$\text{(B26)}$$

As the second term does not involve η_j it does not contribute to the coupling Eq. (24) because

$$\sum_j(2j+1)\langle\vec{l}\cdot\vec{S}\rangle_j=0\ .$$

The remaining integral is readily solved by using the contour integration techniques used in Appendix C and we find

$$\mathcal{P}\int_0^{\infty}\frac{dk'}{k^2-k'^2}(-1/2)\cos[k'(R_a+R_b)+2\eta_j(k')]$$

$$=-(\pi/4k)\sin[k(R_a+R_b)+2\eta_j(k)]\ .$$

The final integral for $I_1(j)$ becomes

$$I_1(j)=-\frac{\pi}{4}\int_0^{k_F}\sin kR_{ab}$$

$$\times\sin[k(R_a+R_b)+2\eta_j(k)]dk\ .$$

$$\text{(B27)}$$

When we repeat the steps from Eqs. (B22) through (B27) for the second term in the second-order energy Eq. (B21) we find a contribution of the same form as

Eq. (B24) but with the integrals

$$I_2(j) \equiv \int_0^{k_F} \sin(kR_a+\eta_j)\sin(kR_b+\eta_j)\,dk \times \mathcal{P}\int_0^{\infty} \frac{k'dk'}{k^2-k'^2}\sin k'R_{ab} \ . \tag{B28}$$

The principal value of the second integral is readily ascertained and is

$$\mathcal{P}\int_0^{\infty} \frac{\sin(k'R_{ab})k'dk'}{k^2-k'^2} = -\frac{\pi}{2}\cos kR_{ab} \ .$$

By placing this in $I_2(j)$ and by using Eq. (B26) we find the nonvanishing contribution of $I_2(j)$ Eq. (B28) to the anisotropic coupling part of $E^{(2)}$ is

$$I_2(j) = \frac{\pi}{4}\int_0^{k_F} \cos kR_{ab} \times \cos[k(R_a+R_b)+2\eta_j(k)]\,dk \ . \tag{B29}$$

By adding these two contributions and the additional two that come from exchanging indices A and B [see Eq. (B21)], we find the total anisotropic coupling of the form of Eq. (B24) with

$$\begin{aligned} I(j) &= 2[I_1(j)+I_2(j)] \\ &= \frac{\pi}{2}\int_0^{k_F} \cos[k(R_a+R_b+R_{ab})+2\eta_j(k)]\,dk \ . \end{aligned} \tag{B30}$$

To evaluate this integral we first linearize it with respect to the spin-orbit coupling by expanding the phase shift η_j about the one without spin-orbit coupling η_l

$$\cos(\alpha+2\eta_j) \cong \cos(\alpha+2\eta_l) - 2\sin(\alpha+2\eta_l)(\eta_j-\eta_l) \ . \tag{B31}$$

From the definition of the phase shift,[9] Eq. (B20), we find

$$\eta_j - \eta_l = -(1/\Delta)\sin^2\eta_l\ \lambda_d\langle \vec{1}\cdot\vec{s}\rangle_j + O(\lambda_d^2) \ . \tag{B32}$$

After linearizing and neglecting the term independent of λ in Eq. (B31) we find $I(j)$, Eq. (B30), becomes

$$I(j) = \frac{\pi\lambda_d\langle \vec{1}\cdot\vec{s}\rangle}{\Delta}\int_0^{k_F} \sin[k(R_a+R_b+R_{ab})+2\eta_2(k)] \times \sin^2\eta_2(k)\,dk \ . \tag{B33}$$

This integral is solved by using the same approximations as those used to solve the integral Eq. (21). By replacing the integral over k space by one over the phase shift, see Eq. (23), we find

$$\begin{aligned} I(j) &\cong \pi\lambda_d\langle \vec{1}\cdot\vec{s}\rangle_j(m/\hbar^2)(1/k_F)\int_0^{\eta_2(E_F)} \sin[k_F(R_a+R_b+R_{ab})+2\eta_2]\,d\eta_2 \\ &= \pi\lambda_d\langle \vec{1}\cdot\vec{s}\rangle_j(m/\hbar^2)(1/k_F)\sin\eta_2(E_F)\sin[k_F(R_a+R_b+R_{ab})+\eta_2(E_F)] \ . \end{aligned} \tag{B34}$$

When this result is placed in the second-order energy expression we find the anisotropic coupling linear in λ is

$$H_{\mathrm{DM}} = \sum_{i=1}^{4} E_i^{(2)} = -\frac{(\Gamma/N)^2\lambda_d}{32\pi^3 k_F}\left(\frac{2m}{\hbar^2}\right)^2\left[\sum_j (2j+1)\langle \vec{1}\cdot\vec{s}\rangle_j^2\right] \times \frac{\sin\eta_2(E_F)\sin[k_F(R_a+R_b+R_{ab})+\eta_2(E_F)]}{R_aR_bR_{ab}}\hat{R}_a\cdot\hat{R}_b(\hat{R}_a\times\hat{R}_b)\cdot(\vec{S}_a\times\vec{S}_b) \ . \tag{B35}$$

By evaluating the sum over j for $l=2$, we find

$$\sum_j (2j+1)\langle \vec{1}\cdot\vec{s}\rangle_j^2 = 15$$

so that after using the same relations used for the coefficient in Eq. (26) the numerical coefficient of the anisotropic coupling Eq. (B35) is

$$\frac{15\lambda_d\Gamma^2}{32\pi^3N^2k_F}\left(\frac{2m}{\hbar^2}\right)^2 = \frac{135\pi}{32}\frac{\lambda_d\Gamma^2}{E_F^2k_F^3} \ .$$

Thus we see that this anisotropic coupling which is trilinear in λ_d, S_a, and S_b is identical to Eq. (26).

In summary we have shown that one can find the anisotropic coupling between Mn spins linear in λ by using the same second-order energy expression as that used by RKKY. The perturbations introduced by the transition-metal impurities are taken into account by replacing the plane wave states by distorted waves whose phase shifts are proportional to the spin-orbit coupling of the impurity. When the resulting expression for the coupling is expanded in powers of λ, the first-order term gives the same anisotropic coupling found from third-order perturbation theory—as it should!

APPENDIX C: EVALUATION OF INTEGRAL

In evaluating the anisotropic coupling between Mn spins it is necessary to evaluate the integral in Eq. (19) of the form,

$$I=\mathcal{P}\int_0^\infty \frac{kdk}{k'^2-k^2}\sin\eta_2(k)\sin[kR+\eta_2(k)]$$

$$=\mathcal{P}\int_0^\infty \frac{kdk}{k^2-k'^2}\left(\frac{1}{2i}\frac{\delta e^{ikR}}{k^2-k_r^2+i\delta}+\text{c.c.}\right) , \qquad \text{(C1)}$$

where $\delta\equiv 2m\Delta/\hbar^2$ and $k_r^2=2mE_r/\hbar^2$. To arrive at this form we used the definition of the phase shift η_2, Eq. (4) to write

$$\sin\eta_2(k)\cos\eta_2(k)=\frac{\Delta(E_r-E)}{(E-E_r)^2+\Delta^2}$$

$$=\frac{\delta(k_r^2-k^2)}{(k^2-k_r^2)^2+\delta^2} , \qquad \text{(C2)}$$

and

$$\sin^2\eta_2(k)=\frac{\Delta^2}{(E-E_r)^2+\Delta^2}=\frac{\delta^2}{(k^2-k_r^2)^2+\delta^2} .$$

To determine the positions of the poles in the integrand Eq. (C1) we write

$$\frac{k}{(k^2-k'^2)(k^2-k_r^2+i\delta)}=\frac{1}{2[k'^2-k_r^2+i\delta]}\left(\frac{1}{k-k'}+\frac{1}{k+k'}-\frac{1}{k-k_i}-\frac{1}{k+k_i}\right) ,$$

where

$$k_i\equiv(k_r^2-i\delta)^{1/2}=k_r(1-i\delta/k_r^2)^{1/2} .$$

Therefore the integral to evaluate, Eq. (C1) is written as

$$I=\frac{1}{2(k'^2-k_i^2)}\left(\frac{\delta}{2i}\right)$$

$$\times\left(\mathcal{P}\int_0^\infty\frac{e^{ikR}dk}{k-k'}+\mathcal{P}\int_0^\infty\frac{e^{ikR}dk}{k+k'}\right.$$

$$\left.-\mathcal{P}\int_0^\infty\frac{e^{ikR}dk}{k-k_i}-\mathcal{P}\int_0^\infty\frac{e^{ikR}dk}{k-k_i}\right)+\text{c.c.} . \qquad \text{(C3)}$$

To evaluate these integrals we use the contour of integration shown in Fig. 4. There are no poles inside our contour and we find for the first integral

$$0=\mathcal{P}\int_0^\infty\frac{e^{ikR}dk}{k-k'}-\pi i e^{ik'R}+\int_{i\infty}^0\frac{e^{izR}}{z-k'}dz , \qquad \text{(C4)}$$

where the second term is the contribution from the integration in the immediate vicinity of the pole at k' and z represents the dummy variable of integration along the imaginary axis, see Fig. 4. By making the transformation $z=it/R$, we find

$$\mathcal{P}\int_0^\infty\frac{e^{ikR}dk}{k-k'}=\pi i e^{ik'R}-\frac{i}{k'R}\int_0^\infty\frac{e^{-t}dt}{1-(i/k'R)t}$$

$$=\pi i e^{ik'R}-\frac{i}{k'R}\sum_{n=0}^\infty n!\left(\frac{i}{k'R}\right)^n .$$

By a similar procedure we find for the second integral in Eq. (C3)

$$\mathcal{P}\int_0^\infty\frac{e^{ikR}dk}{k+k'}=\frac{i}{k'R}\sum_{n=0}^\infty n!\left(\frac{-i}{k'R}\right)^n .$$

By adding these integrals and the analogous results for the third and fourth integrals in Eq. (C3), i.e., with k' replaced by k_i, we find the four integrals in Eq. (C3) are equal to

$$\left(\mathcal{P}\int_0^\infty\frac{e^{ikR}dk}{k-k'}+\cdots\right)$$

$$=\pi i e^{ik'R}+2\left(\frac{1}{k'R}\right)^2\sum_{n=0}^\infty(2n+1)!\frac{(-1)^n}{(k'R)^{2n}}$$

$$-2\left(\frac{1}{k_iR}\right)^2\sum_{n=0}^\infty(2n+1)!\frac{(-1)^n}{(k_iR)^{2n}} .$$

As the pole at k_i is not on the contour of integration we do not pick up a contribution from integrating about this pole, i.e., we do not have a contribution

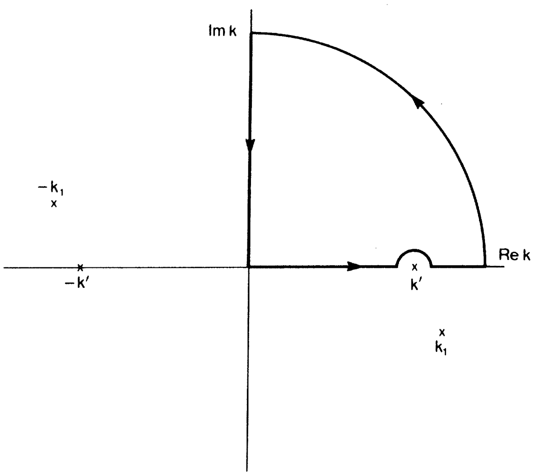

FIG. 4. Poles of the integrand in Eq. (C1) and the contour C used to evaluate the integrals in Eq. (C3).

$\pi i e^{ik_iR}$. By placing this result in Eq. (C3) we find our integral is

$$I=\frac{\delta}{2(k'^2-k_r^2+i\delta)}\left[\frac{\pi}{2}e^{ik'R}+\frac{1}{i}\left(\frac{1}{k'R}\right)^2\sum_{n=0}^{\infty}(2n+1)!\frac{(-)^n}{(k'R)^{2n}}-\frac{1}{i}\left(\frac{1}{k_iR}\right)^2\sum_{n=0}^{\infty}(2n+1)!\frac{(-)^n}{(k_iR)^{2n}}\right]+\text{c.c.}\ . \tag{C5}$$

By taking the indicated complex conjugate of the first term and by using the definitions of k_i and Eqs. (C2) we find

$$\begin{aligned}I=&-\sin\eta_2(k')\cos\eta_2(k')\Bigg[\frac{\pi}{2}\cos k'R-\operatorname{Im}\Bigg(\frac{1}{(k_iR)^2}-\frac{6}{(k_iR)^4}+\cdots\Bigg)\Bigg]\Bigg]\\&+\sin^2\eta_2(k')\Bigg[\frac{\pi}{2}\sin k'R-\Bigg(\frac{1}{(k'R)^2}-\frac{6}{(k'R)^4}+\cdots\Bigg]+\operatorname{Re}\Bigg[\frac{1}{(k_iR)^2}-\frac{6}{(k_iR)^4}+\cdots\Bigg]\Bigg]\ ,\end{aligned} \tag{C6}$$

where Re and Im refer to the real and imaginary parts of the terms in brackets. Therefore to lowest order in $1/k'R$ we find the integral is

$$I=-\frac{\pi}{2}\sin\eta_2(k')\cos[k'R+\eta_2(k')]\ . \tag{C7}$$

APPENDIX D: DETERMINATION OF ANISOTROPY ENERGY FROM EXPERIMENTAL DATA

In this Appendix we show how we extract from the experimental data the anisotropy energy of the $Cu\mathrm{Mn}_xT_y$ spin-glass alloys given in Table I and Fig. 1. For $T=$Ti, Fe, Co, Ni, and Pd the experimental data come from electron spin resonance (ESR) experiments.[4] For $T=$Pt, they come from magnetic measurements.[1]

The relation between the shift of the ESR in the spin-glass state and the macroscopic anisotropy field acting on the remanent magnetization (magnetostatic model) has been clearly shown by Monod and Berthier.[2] These authors have found that, in the low-frequency and low-field limit, the ESR frequency of $Cu\mathrm{Mn}_x$ alloys in the field-cooled spin-glass state is well described by the relation

$$\frac{\omega}{\gamma}=H_0+H_A\ . \tag{D1}$$

Here H_0 is the applied field and H_A can be identified as the macroscopic anisotropic field which acts on the remanent magnetization (H_A in the notation of Monod and Berthier[2]; $H_A=H_d+H_{ax}$ in the notation of Alloul and Hippert[3]). The values of H_A determined from the resonance shift, i.e., $H_A=\omega/\gamma-H_0$ turn out to be in approximate agreement with those determined from experiments of NMR or transverse susceptibility but always larger than those derived from the reversal field of the remanent magnetization. For example, in CuMn 1.35 at.% at $T \ll T_g$ and for the maximum remanent magnetization, the following values of H_A have been obtained: $H_A\simeq 445$ G from ESR,[2] $H_A\simeq 375$ G from NMR.[3,19] In contrast, the analysis of the hysteresis loop[1] of a CuMn 1 at.% alloys leads to only $H_A\simeq 170$ G, as will be explained below.

Let us now consider the ESR measurements on $Cu\mathrm{Mn}_xT_y$ alloys; $x=2$ at.%, $y<2$ at.%. Okuda and Date[4] found that, in the spin-glass state, there is a shift of the resonance which is proportional to y. This shift can be used to determine the anisotropy field H_A and the macroscopic anisotropy energy but, as the measurements were not made under the low frequency and low field conditions as the measurements of Monod and Berthier,[2] the relation between the resonance shift and the anisotropy field does not have the simple form of Eq. (D1). It is necessary to take into account the significant magnetization χH_0 which supplements the remanent magnetization μ_r. According to Hurdequint *et al.*,[19] this leads to replacing Eq. (D1) by the following relation

$$\frac{\omega}{\gamma}=H_0+\frac{\mu_r}{\mu_r+\chi H_0}H_A\ . \tag{D2}$$

By using Eq. (D2), we can relate the macroscopic anisotropy energy to the resonance shift in the following way:

$$E_a\equiv\mu_rH_A=\left(\frac{\omega}{\gamma}-H_0\right)(\mu_r+\chi H_0)\ , \tag{D3}$$

With this relation the rate of increase of the anisotropy energy with concentration of nonmagnetic impurities y is related to the rate of increase of the resonance shift by

$$\left(\frac{dE_a}{dy}\right)_{\text{expt}}=(\mu_r+\chi H_0)\frac{d}{dy}\left(\frac{\omega}{\gamma}-H_0\right)\ . \tag{D4}$$

The values of $(dE_a/dy)_{\text{expt}}$ in Table I and Fig. 1 have been derived (i) by taking the experimental values of $(d/dy)(\omega/\gamma-H_0)$ at 4.2 K given in Table II of Ref. 4 for several types of nonmagnetic element T, and (ii) by estimating the factor $(\mu_r+\chi H_0)$ to be approximately $0.07\mu_B$ ($\pm 0.025\mu_B$) per Mn atom for the experimental conditions given in Ref. 2. The factor $(\mu_r+\chi H_0)$ cannot be accurately derived because the experimental conditions of Ref. 2 are not precisely known. Thus relative values of the anisotropy energies induced by various types of nonmagnetic impurities are more reliably known than their absolute values. This is one of the reasons why in Sec. III we attached more importance to the variation of the anisotropy energy with the type of nonmagnetic impurity than to the absolute values.

For $T = \mathrm{Pt}$ an experimental value of dE_a/dy can be derived from the work of Préjean, Joliclerc, and Monod[1] who studied the hysteresis loop of $Cu\mathrm{Mn}_x\mathrm{Pt}_y$ alloys (with $x = 0.01$) at 1.45 K after cooling in 28 kG. For $y << x$, they observed a steep reversal of the remanent magnetization at a negative field H_r proportional to y. When the field is increased back to positive values, there is an opposite reversal of the remanent magnetization at about zero field. The absolute value of the negative reversal field $|H_r|$ amounts to about 170 G in "pure" CuMn 1 at.% and increases with y at the rate of 34×10^3 G per at.% Pt. When these results are analyzed in the magnetostatic model of an anisotropy field,[3,19,20] the half-width of the hysteresis loop $(\sim \frac{1}{2}|H_r|)$ has to be identified with the uniaxial anisotropy field H_{ax}, and the displacement of the loop $(\sim \frac{1}{2}|H_r|)$ is identified with the displacement field H_d. This leads us to conclude that

$$H_A \equiv H_d + H_{ax} \simeq |H_r| \qquad \text{(D5)}$$

and

$$\left(\frac{dE_a}{dy}\right)_{\mathrm{expt}} \equiv \frac{d}{dy}(\mu_r H_A) = \frac{d|H_r|}{dy}\mu_r \ . \qquad \text{(D6)}$$

By using the experimental data of Préjean *et al.*[1] on $Cu\mathrm{Mn}_x\mathrm{Pt}_y$ alloys, i.e., $d|H_r|/dy = 34 \times 10^5$ G and $\mu_r \simeq 0.03\mu_B$ per Mn atom, we have calculated the value of $(dE_a/dy)_{\mathrm{expt}}$ for $T = \mathrm{Pt}$ which appears in Table I and Fig. 1.

As we mentioned above, the anisotropy energies derived from the hysteresis loop generally turn out to be smaller than those derived from ESR or NMR experiments. This could explain way the experimental point for Pt in Fig. 1 lies somewhat below the expected line. ESR and NMR measurements on $Cu\mathrm{Mn}_x\mathrm{Pt}_y$ alloys probably yield larger values of dE_a/dy which would be in better agreement with the variation predicted by our theory.

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[11] Equation (29) is relevant when T is a *nonmagnetic* transition-metal impurity, i.e., for Pt, Pd, Ni, Co, and Ni impurities in CuMn. In contrast, although Fe impurities are nonmagnetic when diluted in pure Cu, most Fe impurities in $Cu\mathrm{Mn}_x\mathrm{Fe}_y$ alloys probably develop a magnetic moment induced by their interactions with Mn moments. For magnetic Fe impurities, our single vbs model should be replaced by a model with spin up and spin down vbs. It can be easily shown that the spin-orbit splitting of these vbs still gives rise to spin-orbit scattering and that, in the expression for V_1/V_0, Eq. (29), $\sin[(\pi/10)Z_d]$ has simply to be replaced by $(1/2)\{\sin[(\pi/5)Z_{d+}] + \sin[(\pi/5)Z_{d-}]\}$, where Z_{d+} and Z_{d-} are the number of spin up and spin down $3d$ electrons which can be derived from the total number of $3d$ electrons and the value of the impurity spin. We calculated V_1/V_0 for $Cu\mathrm{Mn}_x\mathrm{Fe}_y$ alloys for both cases of magnetic and nonmagnetic Fe impurities and found only slightly different values. Consequently, to estimate the anisotropy energy of $Cu\mathrm{Mn}_x\mathrm{Fe}_y$ alloys, it is not necessary to make an assumption on the proportion of magnetic to nonmagnetic Fe impurities. The values of V_1/V_0 and $(dE_a/dy)_{\mathrm{calc}}$ given in Table I have been calculated for nonmagnetic Fe impurities, i.e., by using Eqs. (29) and (52).

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