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Theory of electron-spin resonance in spin-glasses with remanence and anisotropy

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A microscopic theory will be developed for the electron-spin resonances (ESR) in spin-glasses with remanence and anisotropy. The model consists of spin vectors randomly distributed and interacting through a dominant exchange interaction and a smaller anisotropic interaction of the Dzyaloshinski-Moriya type. The resulting ESR frequencies are in agreement with recent experimental findings on CuMn. The theory also yields a microscopic foundation for the anisotropy constant used in a recent phenomenological description.

I. INTRODUCTION

Spin-glasses are random dilute magnetic alloys in which the magnetic impurity concentration is of the order of a few percent. The classical example of this is the system of Mn ions dissolved in Cu. This system is metallic and the impurities primarily interact through a Rudermann-Kittel-Kasuya-Yosida (RKKY) interaction which is oscillating and of long range.

In recent experiments a vast amount of data on the low-temperature properties of spin-glasses has been accumulated.[1] Among the many interesting phenomena observed there are remanence-associated properties such as the hysteresis loops of the magnetization,[2] thermoremanent magnetization (TRM), isothermal remanent magnetization (IRM), and their extremely slow decay. Also electron-spin-resonance (ESR) measurements have been performed.[3] However, the result of the ESR depends critically on the magnetization of the sample and the previous ESR work refers to a superposition to the reversible magnetization of a partial remanent magnetization induced by an IRM process during the measurements. Only recently ESR experiments were performed with clear statements concerning the magnetization value of the samples. Monod and Berthier[4] cooled their system in such a way as to produce the saturated value of the thermoremanent magnetization. The ESR frequency was found to vary linearly with the external field, $\omega_+ \approx \alpha^{-1}\gamma H^{\rm ex} + \omega_a$, with a slope α^{-1} slightly smaller than 1. The quantity ω_a turned out to be inversely proportional to the total magnetization which was mainly determined by the remanent magnetization. Shortly afterwards new ESR measurements were carried out by Schultz *et al.*,[5] who adopted an opposite condition from Ref. 4. They cooled the spin-glass in a very small external field and used only small resonance fields in order not to produce a significant remanent magnetization by an IRM process. Their ESR frequency again represented a linear relationship of the form $\omega_+ \approx \gamma(\tilde{\alpha} H^{\rm ex} + H^i)$ but different values for the slope and the intercept from those reported in Ref. 4 were obtained. The slope with $\gamma H^{\rm ex}$ was far from unity and approximately 0.5. A second ESR mode was also reported which showed a linear relation of its frequency versus external field as well, but the slope was less than zero.

Halperin and Saslow[6] developed a hydrodynamical theory which explains the spin dynamics in spin-glasses. However, anisotropy and external fields were ignored in their treatment and systems with remanent magnetization were only briefly discussed. Therefore, Saslow[7] extended the hydrodynamical theory to include an external field as well as remanence and anisotropy. The results describe the observed behavior of the Monod and Berthier experiments. Since, however, Saslow's theory could not cover the experimental features of their low-remanence measurements, the authors of Ref. 5 invented a model free energy that incorporated magnetic remanence, anisotropy, and Zeeman energy. From their phenomenological free energy they derived equations of motion, the eigenfrequencies of which showed the expected behavior for systems with both small and large remanence.

The purpose here is to give a microscopic derivation for the ESR frequencies in spin-glass

systems with either small or large remanence. Note that the time scale involved in ESR experiments is extremely small compared to the time scale of the slow decay of magnetization mentioned above. This decay is believed to be caused by a slow time dependence of low-lying metastable states. Therefore, for an ESR experiment the metastable states can be treated as stable and the well-known theoretical concepts such as application of linear-response theory or linearization of the equation of motion can be applied.[8]

In the next section the model is introduced. In Sec. III we present the general formalism which is used to treat the problem of the ESR excitations in spin-glasses with anisotropy and remanence. It is based on the projection-operator method developed by Zwanzig and Mori. Section IV contains a general discussion of the ESR frequencies for both large and small remanence. The results will also be compared with those of the phenomenological theory of Schultz *et al.* (Sec. V). Here, a microscopic expression can be given for the anisotropy constant introduced by Schultz *et al.* Finally, in Sec. VI, the microscopic parameters appearing in the ESR frequency expressions will be evaluated by means of a Bethe-Peierls-Weiss—type mean-field theory for the case of low-remanent magnetization. The result for K shows a temperature and a concentration dependence which are in qualitative agreement with the experimental results.

II. THE MODEL

We shall consider a system of N-quantum spin operators $\vec{S}_i$, $i=1,\ldots,N$ in a metallic host. The spins are randomly distributed in a volume V. Their Hamiltonian is ($\hbar=1$)

$$\mathcal{H}=\mathcal{H}_H+\mathcal{H}_A-\gamma\vec{H}^{\rm ex}\sum_{i=1}^{N}\vec{S}_i \tag{2.1}$$

with

$$\mathcal{H}_H=-\frac{1}{2}\sum_{i\neq j}J_{ij}\vec{S}_i\cdot\vec{S}_j\ ,$$

$$\mathcal{H}_A=-\frac{1}{2}\sum_{i\neq j}\vec{K}_{ij}\cdot(\vec{S}_i\times\vec{S}_j)\ .$$

$\mathcal{H}_H$ is the isotropic RKKY interaction between the ionic spins, mediated by the conduction electrons, which is the dominant interaction in a metallic system (J_{ij} is the exchange integral). The term $\mathcal{H}_A$ describes an additional anisotropic perturbation between the spins. Here, we assume the anisotropy to be of the Dzyaloshinski-Moriya type[9] as was recently suggested by Fert and Levy.[10] They believe this interaction to be relevant in a metallic RKKY spin-glass like CuMn and explain it by spin-orbit scattering of conduction electrons at additional nonmagnetic transition-metal impurities, dissolved in the sample. Therefore, each interaction coefficient should be proportional to the spin-orbit coupling constant and should contain a sum over all nonmagnetic impurities. However, one should mention that the actual type of $\mathcal{H}_A$ is not essential for the final form of the resulting ESR frequencies. Any other interaction which is not invariant against rotations in spin space could contribute equally well to $\mathcal{H}_A$. In the explicit calculations of Sec. VI, however, we shall maintain the Dzyaloshinski-Moriya type for $\mathcal{H}_A$ and assume instead that each interaction coefficient $\vec{K}_{ij}$ contains two contributions: the Fert-Levy part and an additional contribution of unknown origin which is even present when no additional nonmagnetic impurities are dissolved in the sample.

To simplify the model for the explicit calculations in Sec. VI we shall not use the complicated position dependence of the interactions J_{ij} and K_{ij}. Instead we shall assume the interactions to be of infinite range and independent random variables distributed according to distribution functions $W(J_{ij})$ and $\overline{W}(\vec{K}_{ij})$ with vanishing first but nonvanishing second moments

$$\int dJ_{ij}\,W(J_{ij})J_{ij}^2=\frac{J^2}{N}\ ,$$

$$\int d^3K_{ij}\,\overline{W}(\vec{K}_{ij})K_{ij}^{\alpha}K_{ij}^{\beta}=\delta_{\alpha\beta}\frac{\kappa^2}{N} \tag{2.2}$$

($\alpha,\beta=x,y,z$). Here the normalization with N must be enforced to obtain an appropriate thermodynamic limit. The first moment of the distribution $\overline{W}(\vec{K}_{ij})$ is zero since $\vec{K}_{ij}$ is antisymmetric $\vec{K}_{ij}=-\vec{K}_{ji}$. A nonzero first moment of $W(J_{ij})$ would only lead to a transition between the spin-glass and ferromagnetic state and will be neglected here. The higher moments of $W(J_{ij})$ and $\overline{W}(\vec{K}_{ij})$ are assumed to be of higher order in $1/N$. Note that the second moment of the anisotropic interaction should contain two contributions, $\kappa^2=ac_I+f(c_{\rm Mn})$, according to its physical origin. The first contribution is proportional to the concentration of the nonmagnetic impurities and is caused by single-site contributions from the double sum in K_{ij} over the nonmagnetic impurities. The second term of κ^2 is a function of the magnetic ion concentration but is independent of c_I.

III. THE DYNAMICAL SUSCEPTIBILITY

In order to construct a dynamical theory one must be able to determine the macroscopic variables which will enter into the theory. The theoretical approaches of Saslow[7] and Schultz *et al.*[5] both used as dynamical variables the magnetization $\mathcal{M}$ and a second variable associated with the order parameter, which was interpreted in Ref. 7 as an average angle θ by which the spins are rotated from their equilibrium positions. For a spin-glass with a dominant Heisenberg interaction and smaller anisotropy and Zeeman energy the total magnetization is almost conserved and should therefore lead to a slow mode. Also, the variable $\vec{\theta}$ should lead to a slow mode because its k-dependent static susceptibility $\chi_\theta(\vec{k})$ diverges as $k\to 0$ in the case of a pure Heisenberg spin-glass. In a system with small anisotropy the susceptibility χ_θ no longer diverges but still is bounded by the inverse of the small anisotropy energy as is shown in Appendix A. Thus, both quantities $\mathcal{M}$ and θ should belong to the set of dynamical variables for the system (2.1), where we have large exchange and small anisotropic energy. However, we find it difficult to evaluate explicitly static quantities involving θ by starting from microscopic expressions. We shall consider instead the time derivative of the magnetization $\dot{\mathcal{M}}^- = iL\mathcal{M}^-$ as the second dynamical variable. This choice is equivalent to substituting χ_θ for its lower bound. Thus, one expects that $\dot{\mathcal{M}}^-$ should almost equally well lead to an adequate description of the spin-glass dynamics.

Let us consider a sample which was cooled to low temperature from above the spin-glass regime in the presence of a magnetic field $\vec{H}_c$ along z. If $\vec{H}_c$ was not too small a remanent magnetization (TRM) remains after the field was turned off. We shall study the ESR frequencies in the spin-glass state for external fields $\vec{H}^{\rm ex}$ applied parallel to $\vec{H}_c$ for both large- and small-remanent magnetization. The linear response of the transverse magnetization due to a small oscillating transverse magnetic field $h_x\cos\omega t$ is given by the Kubo formula,[11]

$$\delta\langle\mathcal{M}^x(t)\rangle = \mathrm{Re}\left[\frac{\chi(\omega)+\chi(-\omega)}{2}e^{i\omega t}\right]h_x \ , \tag{3.1}$$

where

$$\chi(\omega)=\frac{i}{2}\int_0^\infty dt\, e^{+i(\omega+i\eta)t}\langle\langle[\mathcal{M}^+(t),\mathcal{M}^-]\rangle\rangle_{\rm av} \ , \qquad \eta\to 0^+ \tag{3.2}$$

($\mathcal{M}^\pm=\mathcal{M}^x\pm i\mathcal{M}^y=\gamma S^\pm$). Here $\chi(\omega)$ is the frequency-dependent dynamical susceptibility which contains all information concerning the positions and the linewidths of the electron-spin resonances. The infinitesimally small quantity η was introduced for the integral in (3.1) to converge. Note that Eq. (3.1) is based on the assumption of vanishing off-diagonal response between the x and y direction. This is approximately valid because the isotropic RKKY interaction is assumed to be the dominant energy in the system (compare Sec. VI).

To analyze the dynamical susceptibility we shall apply the memory-function formalism[12] and define the following scalar product:

$$(A\,|\,B)=\frac{1}{\beta}\int_0^\beta d\lambda\langle\langle A^+ e^{-\lambda\mathcal{H}} B\, e^{\lambda\mathcal{H}}\rangle\rangle_{\rm av} \ , \qquad \beta=1/kT \tag{3.3}$$

for any operator A and B. Here $\langle\ \rangle$ means the expectation value with the equilibrium density matrix and $\langle\ \rangle_{\rm av}$ indicates that an average has to be taken over all random interaction parameters. Using the relation $\langle\langle[A,B]\rangle\rangle_{\rm av}=\beta(A^+\,|\,LB)$, which connects commutator expressions with the scalar product (3.3) the transverse susceptibility $\chi(\omega)$ can be expressed as

$$\chi(\omega)=\beta\left[\mathcal{M}^-\left|\frac{L}{L-z}\mathcal{M}^-\right.\right]\Big/2 \ , \quad z=\omega+i\eta \tag{3.4}$$

where the Liouville operator L is a superoperator which acts on operators A as $LA=[\mathcal{H},A]$ ($\hbar=1$).

The projection-operator technique is based on the operator identity[13]

$$\frac{1}{L-z}P[PLP-z-\mathcal{M}(z)]+\frac{1}{QLQ-z}QLP=P \ ,$$

where

$$\mathcal{M}(z)=PLQ\frac{1}{QLQ-z}QLP \ . \tag{3.5}$$

In case P is chosen to be the projector into the subspace formed by the dynamical variables $\mathcal{M}^-$ and $\dot{\mathcal{M}}^-$ and $Q=1-P$ we obtain the exact representation of $\chi(\omega)$ (see Appendix B),

$$\chi(\omega)=\chi_\perp\left[1+\frac{\omega}{\omega_1-\omega-M_1(\omega)/(2\chi_\perp/\beta)}\right] \ ,$$

where

$$M_1(\omega)=\frac{\chi_2/\beta}{\omega_2-\omega-M_2(\omega)/(\chi_2/\beta)} \ . \tag{3.6}$$

Here the susceptibilities $\chi_\perp,\chi_2$ and the frequency terms ω_1,ω_2 are given by

$$\chi_\perp=\beta(\hat{\mathscr{M}}^- \mid \mathscr{M}^-)/2,\quad \chi_2=\beta(\hat{\dot{\mathscr{M}}}^- \mid Q_1\dot{\mathscr{M}}^-)\,,$$
$$\omega_1=\frac{(\hat{\mathscr{M}}^- \mid L\mathscr{M}^-)}{2\chi_\perp/\beta},\quad \omega_2=\frac{(\hat{\dot{\mathscr{M}}}^- \mid Q_1LQ_1\dot{\mathscr{M}}^-)}{\chi_2/\beta}\,, \tag{3.7}$$

and the self-energy or "memory function" by

$$M_2(\omega)=\left[\hat{\dot{\mathscr{M}}}^- \middle| Q_1LQ\frac{1}{QLQ-\omega-i\eta}QLQ_1\dot{\mathscr{M}}^-\right]. \tag{3.8}$$

Here, Q_1 is a projection operator into the subspace perpendicular to the magnetization operator $\mathscr{M}^-$, and a quantity[14] $\hat{A}$, defined by $\hat{A}=[L/(L+i\eta)]A$ means that part of the operator A which does not commute with the total Hamiltonian $\mathscr{H}$. This may be seen by taking matrix elements in a representation in which $\mathscr{H}$ is diagonal. $\hat{A}$ connects eigenstates of $\mathscr{H}$ having different energies. Note that the projector P, defined above, differs from the projector $P_1\equiv 1-Q_1$. P projects into the subspace formed by $\mathscr{M}^-$ and its time derivative whereas P_1 projects only into $\mathscr{M}^-$. Let us now discuss the different parameters $\chi_\perp,\omega_1,\chi_2,\omega_2$ defined in (3.7). The quantity $\chi_\perp$ is the transverse static susceptibility. [Strictly speaking, it is the isolated susceptibility[14] because of $\chi_\perp=\chi(\omega\to 0)$.] The frequency term ω_1 can be expressed in two ways. By using $L\mathscr{M}^-=L_A\mathscr{M}^-+\gamma H^{\rm ex}\mathscr{M}^-$ we find at first,

$$\omega_1=\gamma H^{\rm ex}+\frac{\Delta/2}{\chi_\perp},\quad \Delta=\beta(\{L_A\mathscr{M}^-\} \mid \mathscr{M}^-)\,. \tag{3.9}$$

Here $\{L_A\mathscr{M}^-\}$ is defined by $\{L_A\mathscr{M}^-\}=[L/(L+i\eta)]L_A\mathscr{M}^-$, $\gamma H^{\rm ex}$ is the Larmor frequency, and $\Delta/2\chi_\perp$ is a line-shift term proportional to the anisotropy energy. Or, we can use the relations $L\hat{\mathscr{M}}^-=L\mathscr{M}^-$ and $(A \mid LB)=\beta^{-1}\langle\langle[A^+B]\rangle\rangle_{\rm av}$ so that

$$\omega_1=\frac{1}{2\chi_\perp}\langle\langle[\mathscr{M}^+,\mathscr{M}^-]\rangle\rangle_{\rm av}=\frac{\gamma M_z}{\chi_\perp}\,, \tag{3.10}$$

where $M_z=\langle\langle\mathscr{M}^z\rangle\rangle_{\rm av}$ is the total magnetization. Therefore, the line-shift term Δ can be expressed by M_z and $\chi_\perp$, i.e.,

$$\Delta/2=\gamma M_z-\gamma H^{\rm ex}\chi_\perp\,. \tag{3.11}$$

Note that the total magnetization consists of a reversible and an irreversible part, $M_z=H^{\rm ex}\chi_{||}+M_0$, where M_0 is the remanent magnetization (in z direction) and $\chi_{||}$ the longitudinal susceptibility. The quantity χ_2 of (3.7) can be considered as a generalized static susceptibility for the variable $Q_1\dot{\mathscr{M}}^-$. Since $Q_1\dot{\mathscr{M}}^-=iQ_1L_A\mathscr{M}^-$ we have

$$\chi_2=\beta(\{L_A\mathscr{M}^-\} \mid Q_1L_A\mathscr{M}^-)=K-\frac{\Delta^2/2}{\chi_\perp}\,. \tag{3.12}$$

Note that χ_2 can also be expressed in terms of Δ and a new defined quantity

$$K=\beta(\{L_A\mathscr{M}^-\} \mid L_A\mathscr{M}^-)=\int_0^\beta d\lambda\langle\langle[\{\mathscr{H}_A,\mathscr{M}^-\}]^+e^{\lambda\mathscr{H}}[\mathscr{H}_A,\mathscr{M}^-]e^{\lambda\mathscr{H}}\rangle\rangle_{\rm av}\,, \tag{3.13}$$

where K lacks the projector Q_1 of (3.12). As it turns out K will take over the part of the anisotropy constant. Both quantities K and χ_2 are of second order in the anistropic interaction $\mathscr{H}_A$, where higher terms in K_{ij} should be negligible for a spin-glass with dominant Heisenberg interaction. Note that the quantity χ_2 is positive definite because it can be expressed by a norm of the scalar product (3.3). Therefore, the following inequality must be valid (Appendix C):

$$\chi_\perp K\gtrsim 2(\gamma M_z-\gamma H^{\rm ex}\chi_\perp)^2\,. \tag{3.14}$$

Finally, also the frequency term ω_2 can be rewritten. Using $Q_1L\mathscr{M}^-=Q_1L_A\mathscr{M}^-$ and (3.11) we find

$$\omega_2=\frac{1}{\chi_2/\beta}(\{L_A\mathscr{M}^-\} \mid Q_1LQ_1L_A\mathscr{M}^-)=\frac{\langle\langle[(L_A\mathscr{M}^-)^+,L_A\mathscr{M}^-]\rangle\rangle_{\rm av}}{\chi_2}-\frac{\Delta/2}{\chi_\perp}\left[2+\left[\gamma H^{\rm ex}+\frac{\Delta/2}{\chi_\perp}\right]\frac{\Delta}{\chi_2}\right]. \tag{3.15}$$

IV. THE ESR FREQUENCIES

The formal result (3.6) for the dynamical susceptibility $\chi(\omega)$ can easily be discussed. In case the self-energy $M_2(\omega)$ varies slowly with frequency, $\chi(\omega)$ describes two individual resonances,

$$\omega_{\pm}=\pm\frac{\omega_1+\omega_2}{2}+\left[\frac{\chi_2}{2\chi_\perp}+\left[\frac{\omega_1-\omega_2}{2}\right]^2\right]^{1/2}-i\frac{\mathrm{Im}M_2}{2\chi_2/\beta}\left[1\mp\frac{1}{2}\frac{\omega_1-\omega_2}{\{\chi_2/2\chi_\perp+[(\omega_1-\omega_2)/2]^2\}^{1/2}}\right], \tag{4.1}$$

where the first term determines the position and the last term the linewidth of the ESR excitations. Here, the real part of M_2 was neglected and the linewidth was assumed to be small compared to the resonance frequencies. Note the physical response of the transverse magnetization to an oscillating field perpendicular to H^{ex} is proportional to $\chi(\omega)+\chi(-\omega)$ [cf. (3.1)] and only two of the four poles of $\chi(\omega)+\chi(-\omega)$ are seen in an ESR experiment. As it turns out, Eq. (4.1) applies to a spin-glass system with small remanent magnetization. For the case of large remanence not $M_2(\omega)$ but the "first" self-energy $M_1(\omega)$, defined in (3.6) or (B5), can be considered as ω independent. Then, the dynamical susceptibility $\chi(\omega)$ describes only one ESR excitation,

$$\omega_+=\omega_1-i\frac{\mathrm{Im}M_1}{2\chi_\perp/\beta}, \tag{4.2}$$

where we have neglected the real part of M_1.

In the remaining part of this section we shall give a general discussion of the ESR excitations based on general considerations neglecting all linewidth effects. Let us consider the cases of large and small remanence separately.

A. Large remanence $[(\gamma M_0)/\chi_{||}\gg(K/2)/\gamma M_0]$

In Appendix C the inequality (3.14) is discussed for both large- and small-remanent magnetization. Solving for $\chi_\perp$ one obtains lower and upper bounds for the transverse susceptibility. In the case of small magnetic field, $\gamma H^{\mathrm{ex}}\ll(K/2)/\gamma M_z$, the lower bound is given by

$$\chi_\perp\geq\frac{(\gamma M_z)^2}{K/2}\left[1-\frac{\gamma M_z\gamma H^{\mathrm{ex}}}{K/2}+\cdots\right]. \tag{4.3}$$

For the case of large remanence $(\gamma M_0)/\chi_{||}\gg(K/2)/\gamma M_0$, which is now discussed, the transverse susceptibility is always very much larger than the longitudinal susceptibility $\chi_{||}$. Let us now assume the transverse susceptibility is of the order of its lower bound, i.e.,

$$\chi_\perp=\alpha\frac{(\gamma M_z)^2}{K/2}\left[1-\frac{\gamma M_z\gamma H^{\mathrm{ex}}}{K/2}\right],\quad \gamma H^{\mathrm{ex}}\ll\frac{K/2}{\gamma M_z} \tag{4.4}$$

where the constant α may be somewhat larger than 1, $\alpha\gtrsim 1$. Then we have

$$\omega_1=\frac{\gamma M_z}{\chi_\perp}=\frac{1}{\alpha}\left[\gamma H^{\mathrm{ex}}+\frac{K/2}{\gamma M_z}\right]. \tag{4.5}$$

The generalized susceptibility χ_2 would be zero in case $\chi_\perp$ fulfills its lower bound. So, $\chi_2=K-(\Delta^2/2)/\chi_\perp$ should be very small compared to K since $\chi_\perp$ has a value near its lower bound, and the second frequency term ω_2 should become very large because it is inversely proportional to χ_2. Thus, the first self-energy $M_1(\omega)$ is small and almost independent of ω, and only one resonance should be found in an ESR experiment,

$$\omega_+=\omega_1=\frac{1}{\alpha}\left[\gamma H^{\mathrm{ex}}+\frac{K/2}{\gamma M_z}\right],\quad \alpha\gtrsim 1. \tag{4.6}$$

This result means that the dynamical behavior of a spin-glass with large remanence is determined by the dynamics of the magnetization operator $\mathcal{M}^-$ alone. As already mentioned, an experimental ESR frequency described by (4.6), was indeed recently observed by Monod and Berthier in a CuMn spin-glass with large-remanent magnetization. The experiment showed a slope of the frequency with γH^{ex} which differed slightly, but significantly, from unity. Also a line shift was found which was inversely proportional to the total magnetization. The value for $K/\gamma M_0$, deduced from the experiment, is small compared to the experimental value for $(\gamma M_0)/\chi_{||}$. Therefore, the physical situation of the Monod and Berthier experiment should belong to the case of large remanence, considered here.

Note that the mathematical structure of (4.6) is very similar to that of wave-vector-dependent excitations in anisotropic Heisenberg ferromagnet in the ordered phase. In that case the dynamical variable is the Fourier transform $\mathcal{M}_k^-$ of the magnetization operator (k is the wave vector) and the anisotropy constant K reduces to $\rho_0 k^2$ (ρ_0 is the bare stiffness constant) since for small wave vectors the commutator of the Heisenberg Hamiltonian with $\mathcal{M}_k^-$ is proportionate to k ($[\mathcal{H}_H,\mathcal{M}_k^-]\sim k$). Thus, the k-dependent transverse susceptibility becomes $\chi_\perp(k)=(\gamma M_z)^2/\rho_0 k^2$, which shows the well-known divergence for small k, and (4.6) reduces to the expression

$$\omega(k)=\gamma H^{\mathrm{ex}}+[\rho_0/(2\gamma M_z)]k^2,$$

which represents the k-dependent excitations in a Heisenberg ferromagnet.[15]

B. Small remanence $[(\gamma M_0)/\chi_{||} \ll (K/2)/\gamma M_0]$

Let us now consider a sample which was prepared so that it shows no remanent magnetization, i.e., $M_0 \simeq 0$. In this case, the Schwartz inequality (3.14) yields no useful bounds (Appendix C). However, when only small Zeeman energies are used in the ESR experiment both the transverse and the longitudinal susceptibility should approximately be equal, $\chi_\perp \approx \chi_{||}$, because there is no preferred direction in the spin-glass state. Then, the quantity

$$\Delta/2 = \gamma M_0 + (\chi_\perp - \chi_{||})\gamma H^{\rm ex}$$

should become very small, so that the generalized susceptibility $\chi_2 = K - (\Delta^2/2)/\chi_\perp$ can be replaced by K and the frequency term $\omega_1 = \gamma H^{\rm ex} + (\Delta/2)/\chi_\perp$ by $\gamma H^{\rm ex}$, i.e.,

$$\chi_\perp = \chi_{||}\ , \quad \chi_2 = K\ , \tag{4.7}$$
$$\omega_1 = \gamma H^{\rm ex}\ , \quad \omega_2 = \frac{\langle\langle [(L_A \mathcal{M}^-)^+, L_A \mathcal{M}^-] \rangle\rangle_{\rm av}}{K}$$

Thus, the two ESR frequencies $\omega_\pm$ of (4.1) are given by

$$\omega_\pm = \pm \frac{\gamma H^{\rm ex} + \omega_2}{2} + \left[\frac{K/2}{\chi_\perp} + \left[\frac{\gamma H^{\rm ex} - \omega_2}{2} \right]^2 \right]^{1/2}\ , \tag{4.8}$$

where for small external fields the term $[(\gamma H^{\rm ex} - \omega_2)/2]^2$ of order $(\gamma H^{\rm ex})^2$ can be neglected against $(K/2)/\chi_\perp$ in the last term. This last approximation should be valid for the small external fields that are used in the ESR experiments of Ref. 5. The result (4.8) shows the expected experimental behavior if one neglects the frequency term ω_2 (no ω_2 term was employed in the phenomenological description of Ref. 5). Here ω_2 will be evaluated in Sec. VI. The result leads to a value of 0.6 for the slope of ω_+ with $\gamma H^{\rm ex}$ that is somewhat different from the experimental slope 0.5. However, the evaluation of ω_2 is rather crude and the result also depends on the actual type of the anisotropic interaction and its position dependence.

The mathematical structure of the solution (4.8) for a spin-glass with small remanence is very similar to that of wave-vector-dependent excitations in a Heisenberg antiferromagnet in the ordered region. In both cases there is a second dynamical variable besides the magnetization operator that describes the dynamical behavior. In an antiferromagnet the second variable is the staggered magnetization. It corresponds to the time derivative of the magnetization operator $Q_1 \mathcal{M}^-$. For the antiferromagnet the frequency terms $\omega_{1,2}$ both reduce to $\gamma H^{\rm ex}$ and K again reduces to $\rho_0 k^2$. Thus, one gets the usual result for the k-dependent excitations in a Heisenberg antiferromagnet,

$$\omega_\pm(k) = \pm\gamma H^{\rm ex} + \left[\frac{\rho_0/2}{\chi_\perp} \right]^{1/2} |k|\ . \tag{4.9}$$

It is also instructive to introduce the eigenmodes of the system which are linear combinations of the two dynamical variables $\mathcal{M}^-$ and $\dot{\mathcal{M}}_\perp^- \equiv Q_1 \mathcal{M}^-$,

$$B_\pm = \left[\mathcal{M}^- + i \frac{2\chi_\perp}{\chi_2} (\omega_1 \mp \omega_\pm) \dot{\mathcal{M}}_\perp^- \right] \Big/ \sqrt{2}\ , \tag{4.10}$$

where $\omega_\pm$ are the complex eigenfrequencies (4.1). Using (4.10) we can express the dynamical susceptibility $\chi(\omega)$ by

$$\frac{\chi(\omega) - \chi_\perp}{\omega} = \left| \frac{\omega_1 + \omega_-}{\omega_+ + \omega_-} \right|^2 \frac{\chi_B^+}{\omega_+ - \omega} + \left| \frac{\omega_1 - \omega_+}{\omega_- + \omega_+} \right|^2 \frac{\chi_B^-}{(-\omega_-) - \omega}\ , \tag{4.11}$$

where χ_B^+, χ_B^- are the static susceptibilities of the eigenmodes $B_\pm$,

$$\chi_B^\pm = \chi_\perp \left[1 + \frac{2\chi_\perp}{\chi_2} |\omega_1 \mp \omega_\pm|^2 \right]\ . \tag{4.12}$$

The prefactors in (4.11) determine the coupling of $B_\pm$ to the magnetization operator $\mathcal{M}^-$. For small remanence the two excitations seen in an ESR experiment are given by the pole of the first term of the right-hand side (RHS) of (4.11) and by the pole of a contribution to $\chi(-\omega)$ that corresponds to the second term of (4.11). With increasing remanent magnetization the coupling strength of the second eigenmode in (4.11) decreases and vanishes for large remanence because of $\omega_+ \approx \omega_1$. Thus the large-remanence case of Sec. IV A is regained.

V. COMPARISON WITH THE PHENOMENOLOGICAL THEORY OF REF. 5

Our results shall be compared with those of Schultz *et al.*[5] We start by giving a dynamical

description equivalent to (3.7) in terms of two equations of motion for the variables $\mathcal{M}^-$ and $Q_1\dot{\mathcal{M}}^- \equiv \dot{\mathcal{M}}_\perp^-$,

$$\begin{aligned}\frac{d}{dt}\mathcal{M}^- &= i\omega_1\mathcal{M}^- + \dot{\mathcal{M}}_\perp^- ,\\ \frac{d}{dt}\dot{\mathcal{M}}_\perp^- &= -\frac{\chi_2}{2\chi_\perp}\mathcal{M}^- + i\omega_2\dot{\mathcal{M}}_\perp^- \\ &\quad - \int_0^t dt'\dot{\mathcal{M}}_\perp^-(t')\frac{M_2(t-t')}{\chi_2/\beta} + F(t) .\end{aligned} \tag{5.1a}$$

These equations can be obtained from Eq. (3.5) [with P and Q defined in (B7)] by multiplying from the right with $\mathcal{M}^-$ and $\dot{\mathcal{M}}_\perp^-$ and performing the inverse Laplace transformation. $M_2(t)$ is the inverse Laplace transform of $M_2(\omega)$ and $F(t)$ is the random force $F(t)=\exp(iQLQ)\dot{\mathcal{M}}_\perp^-$. The phenomenological equations of Schultz *et al.* did not include damping effects. Therefore, we shall neglect the third and fourth terms in the second equation and rewrite (5.1) in terms of the two new variables $\mathcal{M}^-$ and $iL_A\mathcal{M}^- \equiv \dot{\mathcal{M}}_A^-$, where $\dot{\mathcal{M}}_A^-$ is the part of the time derivative of $\mathcal{M}^-$ which is governed by the anisotropic energy $\mathcal{H}_A$. With $\dot{\mathcal{M}}_A^- = \dot{\mathcal{M}}_\perp^- + (\Delta/2\chi_\perp)\mathcal{M}^-$ we have

$$\begin{aligned}\frac{d}{dt}\mathcal{M}^- &= i\gamma H^{\mathrm{ex}}\mathcal{M}^- + \dot{\mathcal{M}}_A^- ,\\ \frac{d}{dt}\dot{\mathcal{M}}_A^- &= -\frac{\chi_2+\Delta(\gamma H^{\mathrm{ex}}-\omega_2)}{2\chi_\perp}\mathcal{M}^- \\ &\quad + i\left[\frac{\Delta/2}{\chi_\perp}+\omega_2\right]\dot{\mathcal{M}}_A^- .\end{aligned} \tag{5.1b}$$

The corresponding equations deducible from the paper of Schultz *et al.* are

$$\begin{aligned}\frac{d}{dt}\mathcal{M}^- &= i\gamma H^{\mathrm{ex}}\mathcal{M}^- + i\gamma K_s n_- ,\\ \frac{d}{dt}n_- &= i\frac{\gamma}{\chi_{s\perp}}\mathcal{M}^- - i\left[\frac{\gamma M_z}{\chi_{s\perp}} - \gamma H^{\mathrm{ex}}\right]n_- ,\end{aligned} \tag{5.2}$$

which lead to the ESR frequencies

$$\begin{aligned}\tilde{\omega}_\pm &= \pm\frac{\gamma H^{\mathrm{ex}} - \gamma M_0/K_s}{2} \\ &\quad + \left[\frac{(\gamma H^{\mathrm{ex}}+\gamma M_0/K_s)^2}{2} + \frac{K_s}{\chi_s}\right]^{1/2} .\end{aligned} \tag{5.3}$$

In (5.2) the second dynamical variable $\hat{n}$ is a direction operator associated with the order parameter. K_s is the phenomenological anisotropy constant and $\chi_{s\parallel}$ and $\chi_{s\perp}$ are the principal values of the susceptibility tensor which were set equal to each other, $\chi_{s\parallel}=\chi_{s\perp}=\chi_s$. Now we can compare (5.1b) with (5.2). From the first equations we find that $\dot{\mathcal{M}}_A^-$ can be identified with $i\gamma K_s n_-$. The second equations give the following relations between the various quantities of both approaches:

$$\frac{\gamma^2 K_s}{\chi_s} \hat{=} \frac{\chi_2+\Delta(\gamma H^{\mathrm{ex}}-\omega_2)}{2\chi_\perp} , \tag{5.4a}$$

$$\gamma H^{\mathrm{ex}} - \frac{\gamma M_z}{\chi_s} \hat{=} \frac{\Delta}{2\chi_\perp} + \omega_2 , \tag{5.4b}$$

which must be valid if both dynamical descriptions are equivalent. Let us discuss the cases of small and large remanence separately.

For *small remanence* $[\gamma M_0/\chi_\parallel \ll (K/2)/\gamma M_0]$ the RHS of (5.4a) reduces to $K/(2\chi_\parallel)$ because the quantity Δ is small and also the frequency term ω_2 is small. Thus, if we identify χ_s with $\chi_\parallel$ $(=\chi_\perp)$ the phenomenological anisotropy constant K_s of Schultz *et al.* is determined by the microscopic quantity K,

$$\gamma^2 K_s = \frac{K}{2} = \frac{\beta}{2}(\{L_A\mathcal{M}^-\}\,|\,L_A\mathcal{M}^-) . \tag{5.5}$$

The microscopic expression for K will be evaluated in the next section.

In the case of *large remanence* $[\gamma M_0/\chi_\parallel \gg (K/2)/\gamma M_0]$ a comparison between both sides of (5.4) is more complicated. To ensure a finite value for ω_2 we assume the following form for the transverse susceptibility:

$$\chi_\perp = \chi_\parallel + \frac{(\gamma M_z)^2}{K/2+\gamma M_z\gamma H^{\mathrm{ex}}} , \tag{5.6}$$

which can be deduced from the phenomenological free energy of Ref. 5. Note that (5.6) also agrees with both the large- and low-remanence results (4.4) and (4.7). For small magnetic fields $\gamma H^{\mathrm{ex}} \ll (K/2)/\gamma M_z$ one now finds $\chi_2 = [K^2/(\gamma M_z)^2]\chi_\parallel/2$ and

$$\omega_2 = \frac{\langle\langle[[\mathcal{H}_A,\mathcal{M}^-]^+,[\mathcal{H}_A,\mathcal{M}^-]]\rangle\rangle_{\mathrm{av}}}{\chi_{2\mathrm{av}}} - \frac{\gamma M_0}{\chi_\parallel} , \tag{5.7}$$

where $\Delta/\chi_\perp = K/\gamma M_0$ was used. If the first term in (5.7) can be neglected, i.e., $\omega_2 \approx -\gamma M_0/\chi_\parallel$, the RHS of (5.4a) again yields $(K/2)/\chi_\parallel$ as in the low-remanence case. Thus the relation (5.5) between the phenomenological anisotropy constant K_s and the microscopic expression K is also derived for the large-remanence case. Note that the two approximations used here, i.e., the assumed

form (5.6) for $\chi_\perp$, and the approximation $\omega_2 \approx -\gamma M_0/\chi_{||}$, were not used in the derivation of the ESR frequency (4.6) for the large-remanence case. They are, however, implicitly contained in the theory of Ref. 5.

VI. EVALUATION OF PARAMETERS

In this section the various parameters which enter the ESR frequencies (4.1) shall be evaluated by starting from the microscopic expressions (3.7). Because of the complexity of the system a Bethe-Peierls-Weiss-type mean-field theory shall be employed which, however, cannot explain the appearance of a remanent magnetization due to an applied field during the cooling process. A more sophisticated method could make use of the concept of spin clusters, which was recently utilized by Ma[16] in explaining the remanence in spin-glasses as well as other low-temperature phenomena. Therefore, we shall henceforth restrict ourselves to the zero-remanent magnetization case.

Our model (2.1) with random-exchange and anisotropic interactions J_{ij} and K_{ij}, both of infinite range, represents a generalization of the Sherrington-Kirkpatrick spin-glass model.[17] The evaluation method used here can avoid the $n \to 0$ replica trick of Ref. 17. In a modified form it was applied to the Sherrington-Kirkpatrick model by Thouless *et al.*,[18] who obtained a result that remained physical to $T=0$ in contrast to the original treatment.[17] Here, however, we shall use a simpler treatment by Plefka, who could rederive the Sherrington-Kirkpatrick solution with the nonphysical $T=0$ behavior.[19] This approach uses the concept of an internal-field distribution. First we replace the Hamiltonian (2.1) by an effective Hamiltonian

$$\mathscr{H}_{N+1} = -\sum_{i=1}^{N} \vec{H}_i \cdot \vec{S}_i - \vec{S}_0 \left[\vec{H}_{ex} + \sum_{i=1}^{N} (J_{0i}\vec{S}_i + \vec{S}_i \times \vec{K}_{0i}) \right] . \tag{6.1}$$

The first term is the mean-field Hamiltonian of N spins with internal fields $\vec{H}_i$. These fields are distributed with a site-independent yet unknown function $P(\vec{H}_i)$. The second term represents the exact interaction of an additional spin $\vec{S}_0$, added at position 0, with the N original spins. By construction, $\vec{H}_i$, J_{0i}, and $\vec{K}_{0i}$ $(i=1,\ldots,N)$ are independent random variables with distribution functions $P(\vec{H}_i)$, $W(J_{0i})$, and $\overline{W}(\vec{K}_{0i})$, respectively.

The distribution function $P(\vec{H})$ can be defined by (Appendix D)

$$P(\vec{H}) = \left\langle \delta \left[\vec{H} - \vec{H}^{ex} - \sum_{i=1}^{N} (J_{0i}\langle \vec{S}_i \rangle_0 + \langle \vec{S}_i \rangle_0 \times \vec{K}_{0i}) \right] \right\rangle_{av} , \tag{6.2}$$

where $\langle\ \rangle_{av}$ means again the average over the random variables and $\langle\ \rangle_0$ is the thermal average taken with the single-particle Hamiltonian

$$\mathscr{H}_0 = -\sum_{i=1}^{N} \vec{H}_i \vec{S}_i - \vec{S}_0 \left[\vec{H}_{ex} + \sum_{i=1}^{N} (J_{0i}\langle \vec{S}_i \rangle_0 + \langle \vec{S}_i \rangle_0 \times \vec{K}_{0i}) \right] , \tag{6.3}$$

which is obtained from (6.1) by replacing $\vec{S}_i$ by its expectation value $\langle \vec{S}_i \rangle_0$. For details of the averaging we refer to Appendix D. In the limit of vanishing external field H^{ex} we obtain

$$P(\vec{H}) = (2\pi)^{-3/2} \frac{1}{[(J^2+2\kappa^2)q/3]^{3/2}} \exp\left[-\frac{H^2}{2(J^2+2\kappa^2)q/3} \right] , \tag{6.4}$$

where the order parameter q is

$$q = \int d^3H \langle S_i^z \rangle_0^2 P(\vec{H}) .$$

$P(\vec{H})$ has a Gaussian shape with linewidth $[(J^2+2\kappa^2)q/3]^{1/2}$ and possesses spherical symmetry $P(\vec{H}) = P(|\vec{H}|)$ even though anisotropic interaction between the spins is present. For finite fields $H^{ex} \neq 0$, $P(\vec{H})$ reduces to cylindrical symmetry.

Equation (6.4) and $q = \int d^3H \langle S_i^z \rangle_0^2 P(H)$ represent a self-consistent equation for q leading immediately to the spin-glass transition temperature for the onset of a nonzero q solution

$$T_f = [S(S+1)/3](J^2+2\kappa^2)^{1/2} .$$

Since the anisotropy energy is small compared to the exchange energy, the spin-glass temperature should depend little on the amount of nonmagnetic impurities added to the sample (e.g., Ni added to CuMn) but might be observable in experiment if the ratio κ/J can be made large enough. The magnetization M_z is evaluated in Appendix D. It cannot explain remanence because it has only a reversible part depending on the external field. However, from the expression for M_z the longitudinal and the transverse susceptibility can be obtained by differentiating with respect to a small external field in the z or in the x direction. For simplicity we restrict ourselves to the limit $H^{\rm ex}=0$, in which both susceptibilities become equal, $\chi_{||}=\chi_{\perp}\equiv\chi$,

$$\chi=N\gamma^2\beta\int d^3H\,P(H)(S_0^\alpha\,|\,S_0^\alpha-\langle S_0^\alpha\rangle_0)_0\,,\qquad \alpha=x,y,z\,. \tag{6.5}$$

Here the quantity $(A\,|\,B)_0$ is defined similarly to (3.3) but using the mean-field Hamiltonian $\mathscr{H}_0$ instead of $\mathscr{H}_{N+1}$,

$$(A\,|\,B)_0=\frac{1}{\beta}\int_0^B d\lambda\langle A^+B(i\lambda)\rangle_0\,,\qquad B(i\lambda)=e^{-\lambda\mathscr{H}_0}Be^{\lambda\mathscr{H}_0}\,, \tag{6.6}$$

and without an average taken over the random variables. The time dependence of $B(i\lambda)$ is best evaluated by transforming the spins S^α into a new frame of reference (x',y',z') with a new z' axis parallel to the internal-field direction $\hat{H}$. From the final result for χ, given in Appendix E, the values at T_f and $T=0$ can easily be deduced,

$$\chi(T=0)=\frac{N\gamma^2}{3}2S\int d^3H\frac{P(H)}{H}=\frac{4}{\sqrt{6\pi}}N\gamma^2\frac{1}{(J^2+2\kappa^2)^{1/2}}\,,$$
$$\chi(T=T_f)=\frac{N\gamma^2}{3}\frac{S(S+1)}{T_f}=\frac{N\gamma^2}{(J^2+2\kappa^2)^{1/2}}\,. \tag{6.7}$$

Note that at the transition temperature the linewidth of the internal-field distribution goes to zero and $P(H)H^2$ degenerates to a δ function: $P(H)=\delta(H)/(H^2 4\pi)$. The result shows that the $T=0$ value of χ is slightly smaller than that at T_f: $\chi(0)/\chi(T_f)\approx 0.92$.

Next, we evaluate the anisotropy constant K, given by (3.13). In a first approximation we replace $\mathscr{H}_{N+1}$ by $\mathscr{H}_0$, since K is already of second order in the anisotropy energy K_{ij}. Next, the quantity $\{L_A\mathscr{M}^-\}$ representing that part of $[\mathscr{H}_A,\mathscr{M}^-]$ that does not commute with the total Hamiltonian is approximated by $\{L_A\mathscr{M}^-\}=L_A\mathscr{M}^--\langle L_A\mathscr{M}^-\rangle_0$. Evaluating $[\mathscr{H}_A,\mathscr{M}^-]$ and taking the average over the random variables we finally obtain

$$K=\frac{4\kappa^2\gamma^2N}{9}\left[\frac{q}{4}\int_0^\beta d\lambda\int d^3H_0P(H_0)\int d^3H_1P(H_1)\left[\langle S_0^zS_0^z(i\lambda)\rangle_0+\langle S_0^yS_0^y(i\lambda)\rangle_0\right]\langle S_1^xS_1^x(i\lambda)\rangle_0-\beta q^2\right]. \tag{6.8}$$

The λ integration leads to a rather lengthy expression (Appendix E). Therefore, we only quote the result. For high temperatures the anisotropy constant decreases as T^{-1},

$$K(T)=4\kappa^2\gamma^2N\left[\frac{S(S+1)}{3}\right]^2\beta\,,\qquad \beta=\frac{1}{kT}\,. \tag{6.9}$$

Thus the value at the spin-glass transition temperature is

$$K(T=T_f)+4\kappa^2\gamma^2N\frac{S(S+1)}{3}\frac{1}{(J^2+2\kappa^2)^{1/2}}\,, \tag{6.10}$$

whereas at $T=0$ we have

$$K(T=0)=4\kappa^2\gamma^2N\frac{4S}{\sqrt{6\pi}}\frac{\alpha+2S/3}{(J^2+2\kappa^2)^{1/2}}\,. \tag{6.11}$$

Here α is a c number of value ~ 0.17. The results (6.10) and (6.11) show that K is of the order κ^2/J. The ratio $K(0)$ to $K(T_f)$ depends on the spin quantum number S. For $S=\frac{1}{2}$ the zero-temperature value of K is slightly smaller than the value at T_f. However, for larger values of S,$K(0)$ becomes larger than $K(T_f)$ for instance, by a factor $\frac{3}{2}$ for $S=\frac{5}{2}$ (spin quantum number of CuMn). Thus for CuMn the calculated anisotropy constant shows an overall decrease with increasing temperature: at first by a factor $\frac{3}{2}$ when T goes from $T=0$ to T_f and as T^{-1} for temperatures $T>T_f$ [note: The theoretical value of K is non-negative because it can be expressed by the norm of the

scalar product (3.3)]. The experimental result for K, deduced from the experiments,[4] shows a qualitatively similar T behavior. The experimental K also decreases with increasing T but linearly with $-T$,

$$K(T)=K(0)[1-\beta(T/T_f')]$$

(β is a c number), and has, therefore, no cusp at T_f as might be expected from the theoretical result. [Note that K was measured in a field of ~ 3 kG where $T_f' \approx 0.9 T_f$ ($H=0$), whereas the theoretical result is only valid for zero magnetic field.] Moreover, it is worthwhile to mention that the experimental prefactor β was found to be the same for different concentrations of Mn ions. Again, this behavior can be understood from the theoretical result for K (neglecting the difference between T_f and T_f'). From (E5) one finds that the ratio $K(T)/K(0)$ only depends on the quantity $\beta(J^2+2\kappa^2)^{1/2}$, i.e., T_f/T, but is independent of the concentration of the magnetic or nonmagnetic impurities dissolved in the sample. The experiments also gave a dependence of the zero-temperature value of K on the concentration of the magnetic and nonmagnetic impurities (c_{Mn} and c_I),

$$\frac{K_{\mathrm{exp}}(T=0)}{V}=c_{\mathrm{Mn}}(ac_I+bc_{\mathrm{Mn}}) \tag{6.12}$$

(V is the volume) with $a \gg b$. From the theoretical point of view the two terms in parentheses should be attributed to the two different physical contributions to κ^2 discussed in Sec. II, whereas the common factor c_{Mn} is the number of magnetic ions N divided by V.

Finally, the frequency term ω_2 has to be evaluated starting from expression (3.15),

$$\omega_2=\frac{\langle\langle[(L_A\mathcal{M}^-)^+,L_A\mathcal{M}^-]\rangle\rangle_{\mathrm{av}}}{\chi_2} \tag{6.13}$$

where we have already used $\Delta \simeq 0$ for the low-remanence case. By evaluating the commutator in (6.13) and taking the average over the random variables, we find

$$\omega_2=\frac{2\kappa^2\gamma^2 N}{K}\langle\langle S^z\rangle_0\rangle_{\mathrm{av}}\langle\langle(S_z)^2\rangle_0\rangle_{\mathrm{av}}\,, \tag{6.14}$$

which is proportional to the magnetization

$$\gamma\langle\langle S^z\rangle_0\rangle_{\mathrm{av}}=\gamma\int d^3H\,P(H)\langle S^z\rangle_0$$

of the sample. The factor

$$\langle\langle(S^z)^2\rangle\rangle_{\mathrm{av}}=\int d^3H\,P(H)\langle(S^z)^2\rangle\,,$$

which is best evaluated by transforming $(S^z)^2$ to the new (x',y',z') frame of reference, reduces to $S^2/3$ at low temperatures $\beta J \gg 1$. Thus the frequency term ω_2 is found to be proportional to γH^{ex},

$$\omega_2=\frac{2\kappa^2\gamma^2S^2\chi(0)}{3K(0)}\gamma H^{\mathrm{ex}}$$

$$=\frac{S}{6(\alpha+2S/3)}\gamma H^{\mathrm{ex}} \tag{6.15}$$

($\beta J \gg 1$) with a slope always less than $\frac{1}{4}$. For $S=\frac{5}{2}$ (CuMn) we have $\omega_2 \simeq 0.23\gamma H^{\mathrm{ex}}$. Thus, the expression (4.8) for the ESR frequency ω_+ in a sample with zero remanence yields a slope with γH^{ex} of ~ 0.61, somewhat in opposition to the experiments which gave a slope of ~ 0.5. The difference between theory and experiment should be attributed to the rather crude theoretical method used here in the evaluation of the various parameters appearing in (6.14).

From the experiments one can deduce a value of ~ 3 kG for the frequency ω_+ at zero external field $\omega_+(H^{\mathrm{ex}}=0)=\sqrt{K/\chi}$. With (6.11) and (6.7), this leads to a value of ~ 0.9 kG for the anisotropic interaction κ ($S=\frac{5}{2}$). For the exchange interaction J one finds a value of the order of 10^2 kG by assuming a realistic value for

$$T_f=[S(S+1)/3](J^2+2\kappa^2)^{1/2}\,.$$

Thus the anisotropic interaction is indeed very small compared to the exchange interaction as was assumed from the beginning.

VII. CONCLUSION

The purpose of the present investigation was to derive microscopic expressions for the ESR excitations in RKKY spin-glass systems with remanence and anisotropy. Based on the projection-operator formalism general features of the ESR frequencies for small and large remanence were discussed without evaluating the various parameters involved. The mathematical structure of the dynamical equations in the large-remanence case turned out to be similar to that of an isotropic Heisenberg ferromagnet in the ordered phase, whereas in the low-remanence case it was closer to an Heisenberg antiferromagnet. The theory presented gives a microscopic foundation for a recent phenomenological description of the spin dynamics in spin-glasses. Our method yielded a microscopic expression for the anisotropy constant used in that approach. This constant and other parameters, con-

tained in the final result for the ESR frequencies, were evaluated by means of a Bethe-Peierls-Weiss—type approach for the case of vanishing remanent magnetization. The theoretical findings were compared with the experiments. The influence of the self-energy on linewidth effects was neglected in the present theory. However, the self-energy is believed to be important for relatively high temperatures $T \gtrsim T_f$ within the spin-glass regime, when the anisotropy constant K and therefore the excitation frequencies become rather small. Linewidth and line-shift effects in this regime will be discussed in the following paper.

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APPENDIX A

The variable $\theta(r)$ describes the average angle by which the spins in the spin-glass system are rotated from equilibrium[6] and is defined by

$$\vec{\theta}(\vec{r})=\frac{1}{2\gamma}\sum_{i\in R}\langle\vec{S}_i\rangle\times\vec{S}_i\ , \tag{A1}$$

where the region R contains a large number of spins centered around point r. $\theta(r)$ and the magnetization operator $\mathcal{M}(r)$ obey the commutation relation

$$\int d\vec{r}\,[\mathcal{M}^\alpha(\vec{r}),\theta^\alpha(\vec{r}\,')]=\frac{(-i)}{2}\left[\sum_\gamma t_\gamma(\vec{r}\,')-t_\alpha(\vec{r}\,')\right] \tag{A2}$$

with $t_\alpha(\vec{r})=\sum_{i\in R}\langle S_i^\alpha\rangle S_i^\alpha$. Taking the thermal averages of (A2) and averaging over the random variables gives the Sherrington-Kirkpatrick order parameter $q_\alpha=\int dr\langle\langle t_\alpha(\vec{r})\rangle\rangle_{\rm av}$,

$$q_\alpha=i\int d\vec{r}\int d\vec{r}\,'\langle\langle[\mathcal{M}^\alpha(\vec{r}),\theta^\alpha(\vec{r}\,')]\rangle\rangle_{\rm av}\ . \tag{A3}$$

For a pure Heisenberg spin-glass q_α is independent of α. The RHS of (A3) can be expressed in terms of the scalar product (3.3) due to $\langle\langle[A^+,B]\rangle\rangle_{\rm av}=\beta(A\mid LB)$. So, we can use the Schwartz inequality $(A\mid A)(B\mid B)\geq|(A\mid B)|^2$, where A and B are the Fourier transforms of θ^α and $\dot{\mathcal{M}}^\alpha=iL\mathcal{M}^\alpha$ and obtain an upper bound for the wave-vector-dependent static susceptibility $\chi_\theta(k)=\beta(\theta_k^\alpha\mid\theta_k^\alpha)$ of the variable θ^α,

$$\chi_\theta(k)\gtrsim\frac{q^2}{\beta(\dot{\mathcal{M}}_k^\alpha\mid\dot{\mathcal{M}}_k^\alpha)}\ . \tag{A4}$$

For a pure Heisenberg spin-glass the total magnetization $\vec{\mathcal{M}}_{k=0}$ is a constant of motion, so that we have $\dot{\mathcal{M}}_k^\alpha\approx ikj^\alpha$ for small wave vectors. Thus $\chi_\theta(k)$ diverges at least as k^{-2}. If the divergence is the minimal one, i.e., $\chi_\theta(k)=q^2/(\rho_0k^2)$, the stiffness constant ρ_0 is bounded by

$$\rho_0(k)\lesssim\lim_{k\to0}\beta(\hat{j}_\alpha\mid j_\alpha)\ , \tag{A5}$$

$j_\alpha=\hat{j}_\alpha$, as already quoted by Halperin and Saslow.[6] In case the system includes anisotropic interactions the susceptibility $\chi_\theta(k)$ no longer diverges for $k\to0$ but is limited by the anisotropy energy

$$\chi_\theta(k\to0)\gtrsim\frac{q^2}{\beta([\{\mathcal{H}_A,\mathcal{M}^\alpha\}]\mid[\mathcal{H}_A,\mathcal{M}^\alpha])}\ . \tag{A6}$$

For the transverse direction the denominator represents the anisotropy constant, defined in (5.5).

APPENDIX B

We want to derive Eq. (3.6). First, we use the identity

$$\frac{L}{L-\omega-i\eta}=\frac{L}{L-i\eta}+\omega\frac{L}{L-i\eta}\frac{1}{L-\omega-i\eta} \tag{B1}$$

and rewrite (3.4) as

$$\chi(\omega)=\chi_\perp+\omega\beta\left[\hat{\mathcal{M}}^-\left|\frac{1}{L-\omega-i\eta}\mathcal{M}^-\right.\right]\Big/2\ , \tag{B2}$$

where $\chi_\perp$ and $\hat{\mathcal{M}}^-$ are defined in (3.7). Next, we apply the operator identity (3.5) choosing at first a projector P_1 that projects on the magnetization operator,

$$P_1=\frac{|\mathcal{M}^-)(\hat{\mathcal{M}}^-|}{2\chi_\perp/\beta},\quad Q_1=1-P_1\ . \tag{B3}$$

By multiplying Eq. (3.5) from the left with

$(\hat{\mathscr{M}}^-|=(\hat{\mathscr{M}}^-|P_1$ and from the right with $|\mathscr{M}^-)=P_1|\mathscr{M}^-)$ we obtain [with $(\hat{\mathscr{M}}^-|Q_1=0$]

$$\chi(\omega)=\chi_\perp\left[1+\frac{\omega}{\omega_1-\omega-M_1(\omega)/(2\chi_\perp/\beta)}\right]. \tag{B4}$$

Here ω_1 is defined in (3.7) and the "first" self-energy $M_1(\omega)$ by

$$M_1(\omega)=\left[\hat{\mathscr{M}}^-\left|Q_1\frac{1}{Q_1LQ_1-\omega-i\eta}Q_1\dot{\mathscr{M}}^-\right.\right]. \tag{B5}$$

Note that $M_1(\omega)$ has the same mathematical structure as the original expression for $[\chi(\omega)-\chi_\perp]/\omega$. Thus, we can apply the identity (3.5) again with

$$P_2=\frac{|Q_1\dot{\mathscr{M}}^-)(\dot{\mathscr{M}}^-|Q_1}{\chi_2/\beta}, \tag{B6}$$

where χ_2 is defined in (3.7) and arrive at the final result (3.6)–(3.8), where

$$Q=(1-P_1)(1-P_2)=1-P_1-P_2\equiv 1-P \tag{B7}$$

projects on the operator space perpendicular to $\mathscr{M}^-$ and $\dot{\mathscr{M}}^-$.

APPENDIX C

To derive mathematical bounds for the susceptibility $\chi_\perp$ we employ the Schwartz inequality $|(\hat{A}|\hat{B})|^2\leq(\hat{A}|\hat{A})(\hat{B}|\hat{B})$ with $\hat{A}=\{L_A\mathscr{M}^-\}$ and $\hat{B}=\hat{\mathscr{M}}^-$, where the scalar product is defined in (3.3). Using the relation $(\hat{A}|\hat{B})=(\hat{A}|B)$ (for any A,B) the inequality reads

$$|(\{L_A\mathscr{M}^-\}|\mathscr{M}^-)|^2 \lesssim(\{L_A\mathscr{M}^-\}|L_A\mathscr{M}^-)(\hat{\mathscr{M}}^-|\mathscr{M}^-) \tag{C1}$$

or

$$(\gamma H^{\rm ex}2\chi_\perp-2\gamma M_z)^2\lesssim K2\chi_\perp \tag{C2}$$

using (3.13), (3.11), and (3.7). K and $\chi_\perp$ are positive definite quantities. (C2) can easily be solved for $\chi_\perp$ leading to a lower and an upper bound for the transverse susceptibility,

$$\gamma H^{\rm ex}2\chi_\perp\gtrless\left[2\gamma M_z+\frac{K}{2\gamma H^{\rm ex}}\right]-\frac{K}{2\gamma H^{\rm ex}}\left[1+\frac{4\gamma M_z}{K/(2\gamma H^{\rm ex})}\right]^{1/2}$$

and (C3)

$$\gamma H^{\rm ex}2\chi_\perp\lesssim\left[2\gamma M_z+\frac{K}{2\gamma H^{\rm ex}}\right]+\frac{K}{2\gamma H^{\rm ex}}\left[1+\frac{4\gamma M_z}{K/(2\gamma H^{\rm ex})}\right]^{1/2}.$$

The bounds depend on the total magnetization $M_z\approx M_0+H^{\rm ex}\chi_{||}$, the anisotropy constant K, and the applied field $H^{\rm ex}$. For low magnetic field $\gamma H^{\rm ex}\ll K/(2\gamma M_z)$, the lower bound is

$$\chi_\perp|_{\rm lb}=\frac{(\gamma M_z)^2}{K/2}\left[1-2\frac{\gamma M_z\gamma H^{\rm ex}}{K}+\cdots\right], \tag{C4}$$

which is discussed below (4.5). It is appropriate to discuss the cases of large- and small-remanent magnetization separately.

For large remanence, $\gamma M_0/\chi_{||}\gg K/\gamma M_0$, both bounds are decreasing functions of the external field [Fig. 1(a)]. Equations (C3) should be useful in this case. For small remanence, however, the lower bound is an increasing function and the upper bound a decreasing function of $\gamma H^{\rm ex}$. Then Eqs. (C3) can be used only for large external fields [compare Fig. 1(b)].

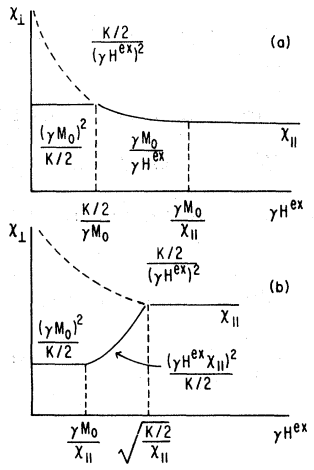

FIG. 1. Qualitative behavior for the lower and upper bound of the transverse susceptibility $\chi_\perp$ for (a) large remanence, $\gamma M_0/\chi_{||}\gg K/\gamma M_0$, and (b) small remanence, $\gamma M_0/\chi_{||}\ll K/\gamma M_0$. The leading contributions are indicated in the different external field regions.

APPENDIX D

For the Hamiltonian $\mathscr{H}_{N+1}$, defined in (6.1), the expectation values $\langle S_i \rangle_{N+1}$, $i=1,\ldots,N$ and $\langle S_0 \rangle_{N+1}$ can be evaluated. After applying $J_{0i}, \vec{K}_{0i} \sim N^{-1/2}$ we obtain [$B_s(x)$ is the Brillouin function]

$$\begin{aligned} &\langle \vec{S}_i \rangle_{N+1} = \langle \vec{S}_i \rangle_0 = \hat{u}_i S B_s(\beta S H_i) , \\ &\langle \vec{S}_0 \rangle_{N+1} = \hat{u}_0 S B_s(S\beta H_0) , \\ &\vec{H}_0 \equiv \vec{H}^{ex} + \sum (J_{0i}\langle S_i \rangle_0 + \langle \vec{S}_i \rangle_0 \times \vec{K}_{0i}) , \end{aligned} \tag{D1}$$

where $\langle\ \rangle_0$ is the expectation value defined below (6.2). $\hat{u}_i = \vec{H}_i/H_i$ and $\hat{u}_0 = \vec{H}_0/H_0$ are unit vectors parallel to the internal fields $\vec{H}_i$ and $\vec{H}_0$ at site i and 0, respectively. $\vec{H}_0$ is the usual molecular-field expression. Note that there is no contribution to the internal field at site i due to the presence of the added spin at site 0. Such a contribution was necessary for the Thouless, Anderson, and Palmer solution of the Sherrington-Kirkpatrick model to remain physical to $T=0$. Using

$$1 = \int d^3H\, \delta(\vec{H} - \vec{H}_0)$$

in (D1) and averaging over the random variables leads to

$$\langle \langle \vec{S}_0 \rangle_{N+1} \rangle_{av} = \int d^3H\, \hat{u} S B_s(S\beta H) P_0(H) , \tag{D2}$$

$\hat{u} = \vec{H}/H$, where the internal-field distribution $P_0(\vec{H})$ at site 0 is given by

$$P_0(\vec{H}) = \left\langle \delta \left[\vec{H} - \vec{H}^{ex} - \sum_{i=1}^{N} (J_{0i}\langle \vec{S}_i \rangle_0 + \langle \vec{S}_i \rangle_0 \times \vec{K}_{0i}) \right] \right\rangle_{av}$$

$$= \frac{1}{(2\pi)^3} \int d^3k\, e^{i\vec{k}(\vec{H} - \vec{H}^{ex})} \langle \{ \exp[-i\vec{k}(J_{0i}\langle \vec{S}_i \rangle_0 + \langle \vec{S}_i \rangle_0 \times \vec{K}_{0i})] \}^N \rangle_{av} . \tag{D3}$$

$\langle\ \rangle_{av}$ indicates the average over the independent random variables $\vec{H}_i$, J_{0i}, and $\vec{K}_{0i}$ ($i=1,\ldots,N$),

$$\langle A \rangle_{av} = \int \cdots \int A \prod_{i=1}^{N} P(\vec{H}_i) d^3H_i W(J_{0i}) dJ_{0i} \overline{W}(\vec{K}_{0i}) d^3K_{0i} . \tag{D4}$$

Since all spins are equivalent the internal field distributions must be the same at all sites, i.e., $P_0(\vec{H}) = P(\vec{H})$. Thus, Eq. (D3) represents a self-consistent integral equation for $P(\vec{H})$. It can be solved by expanding the second exponent in (D3) and using (2.2). In the large-N limit we find

$$P(H) = \frac{1}{(2\pi)^3} \int d^3k\, e^{i\vec{k}(\vec{H} - \vec{H}^{ex})} \exp\left(-\tfrac{1}{2} \{ J^2 q_z + 2\kappa^2 q_\perp) k_z^2 + [J^2 q_\perp + \kappa^2 (q_z + q_\perp)](k_x^2 + k_y^2) \} \right) , \tag{D5}$$

where we have introduced two order parameters q_z and $q_\perp$,

$$q_z = \int \langle S_i^z \rangle_0^2 P(H_i) d^3H_i , \quad q_\perp = \int \langle S_i^x \rangle_0^2 P(H_i) d^3H_i , \tag{D6}$$

which, however, become equal for vanishing external fields. The integration over k leads to the final result,

$$P(H) = \frac{1}{(2\pi)^{3/2}} \frac{1}{(J^2 q_z + 2\kappa^2 q_\perp)^{1/2}} \exp[-(H_z - H^{ex})^2 / 2(J^2 q_z + 2\kappa^2 q_\perp)]$$

$$\times \frac{1}{[J^2 q_\perp + \kappa^2 (q_\perp + q_z)]} \exp\{ -(H_x^2 + H_y^2) / 2[J^2 q_\perp + \kappa^2 (q_\perp + q_z)] \} . \tag{D7}$$

The magnetization is given by $\vec{M} = N \cdot \langle \langle \vec{S}_0 \rangle_{N+1} \rangle_{av}$, where (D3) has to be entered into the RHS. The result for $\vec{M}$ cannot describe the remanent magnetization, as mentioned in Sec. V.

APPENDIX E

The time-dependence (6.6) of a single spin,

$$S^\alpha(i\lambda) = e^{-\lambda\mathscr{H}_0} S^\alpha e^{\lambda\mathscr{H}_0} = e^{\lambda\gamma\vec{H}\cdot\vec{S}} S^\alpha e^{-\lambda\gamma\vec{H}\cdot\vec{S}} , \quad \alpha = x,y,z \tag{E1}$$

where the single-particle Hamiltonian $\mathscr{H}_0$ is given by (6.3), shall be evaluated by introducing a new frame of reference with the new z' axis parallel to the internal field $\vec{H}$. The new y' axis is chosen to be perpendicular to the plane formed by the z' axis and the old z axis. Then the transformation reads

$$S^x=\frac{H_x}{H}S^{z'}-\frac{H_xH_z}{H^2}\frac{1}{w}S^{x'}+\frac{H_y}{H}\frac{1}{w}S^{y'},$$
$$S^y=\frac{H_y}{H}S^{z'}-\frac{H_yH_z}{H^2}\frac{1}{w}S^{x'}-\frac{H_x}{H}\frac{1}{w}S^{y'}, \quad \text{(E2)}$$
$$S^z=\frac{H_z}{H}S^{z'}+wS^{x'},$$

where $w=[1-(H_z/H)^2]^{1/2}$. H_α, $\alpha=x,y,z$ are the components of $\vec{H}$ in the original x,y,z frame of reference and $H=|\vec{H}|$. The evaluation of the time dependence in the new x',y',z' frame is trivial and we immediately give the results for the correlation functions $\langle S^\alpha S^\beta(i\lambda)\rangle_0$ averaged over the internal fields. Here $\langle\ \rangle_0$ means again the average with $\mathscr{H}_0$. We find

$$\int d^3H\,P(H)\langle S^\alpha S^\beta(i\lambda)\rangle_0=\delta_{\alpha\beta}c_\alpha(i\lambda) \quad \text{(E3)}$$

with

$$c_z(i\lambda)=\int d^3H\,P(H)\left[\frac{H_z^2}{H^2}\phi_{||}+\left[1-\frac{H_z^2}{H^2}\right]\phi_\perp(i\lambda)\right],$$
$$c_x(i\lambda)=c_y(i\lambda)=\tfrac{1}{2}\int d^3H\,P(H)\left[\frac{H_x^2+H_y^2}{H^2}\phi_{||}+\left[1+\frac{H_z^2}{H^2}\right]\phi_\perp(i\lambda)\right],$$

where $\phi_{||}$ and $\phi_\perp$ are defined by

$$\phi_{||}=\langle(S_0^{z'})^2\rangle=S\frac{\partial}{\partial\beta H}B_s(S\beta H)+[SB_s(S\beta H)]^2,$$
$$\phi_\perp(i\lambda)=\frac{SB_s(S\beta H)}{2}\left[\frac{e^{-\lambda H}}{1-e^{-\beta H}}+\frac{e^{\lambda H}}{e^{\beta H}-1}\right], \quad \text{(E4)}$$

the susceptibility χ immediately follows from (E4) by integrating over λ,

$$\chi=\frac{N\gamma^2}{3}\int d^3H\,P(H)\left[\beta\phi_{||}+2\frac{SB_s(S\beta H)}{H}\right]-\frac{N\gamma^2}{3}\beta q,$$

and the anisotropy constant K is evaluated by inserting (E3) into (6.8) and integrating over λ,

$$K=\frac{4\kappa^2\gamma^2N}{9}\left[\beta\left[\int d^3H\,P(H)\phi_{||}\right]^2-\beta q^2+4\int d^3H_0P(H_0)\phi_{||}(H_0)\int d^3H_1P(H_1)SB_s(S\beta H_1)\frac{1}{H_1}\right.$$
$$+2\int d^3H_0P(H_0)SB_s(S\beta H_0)\int d^3H_1P(H_1)SB_s(S\beta H_1)$$
$$\times\left[\frac{1-e^{-\beta(H_0+H_1)}}{(1-e^{-\beta H_0})(1-e^{-\beta H_1})}\frac{1}{H_0+H_1}\right.$$
$$\left.\left.+\frac{1-e^{-\beta(H_0-H_1)}}{(1-e^{-\beta H_0})(e^{\beta H_1}-1)}\frac{1}{H_0-H_1}\right]\right]. \quad \text{(E5)}$$

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[1]See articles in the Proceedings of the International Conference on Low Temperature Physics, LT 15, Grenoble, 1978 [J. Phys. (Paris) Colloq. C 6, (1978)].
[2]P. Monod, J. J. Préjean, and B. Tissier, J. Appl. Phys. 50, 7324 (1979).

[3]J. Owens, M. E. Browne, V. Arp, and A. F. Kip, J. Phys. Chem. Solids 2, 85 (1957); K. Okuda and M. Date, J. Phys. Soc. Jpn. 27, 839 (1969).
[4]P. Monod and Y. Berthier, J. Magn. Magn. Mater. 15-18, 149 (1980).
[5]S. Schultz, E. M. Gullikson, D. R. Fredkin, and M. Tovar, Phys. Rev. Lett. 45, 1508 (1980).
[6]B. I. Halperin and W. M. Saslow, Phys. Rev. B 16, 2154 (1977).
[7]W. M. Saslow, Phys. Rev. B 22, 1174 (1980).
[8]U. Krey, Wiss. Z. Hochsch. Verkehrswes., Fviedrich List' Dres. 3, Suppl., 49 (1981); A. J. Bray and M. A. Moore, Phys. Rev. Lett. 47, 120 (1981).
[9]For instance, T. Moriya, in *Magnetism I,* edited by G. T. Rado and H. Suhl (Academic, New York, 1963).
[10]A. Fert and P. M. Levy, Phys. Rev. Lett. 44, 1538 (1980); P. M. Levy and A. Fert, Phys. Rev. B 23, 4667 (1981).
[11]R. Kubo and K. Tomita, J. Phys. Soc. Jpn. 9, 888 (1954).
[12]H. Mori, Prog. Theor. Phys. (Kyoto) 34, 399 (1965).
[13]See, e.g., A. Messiah, *Quantum Mechanics* (North-Holland, Amsterdam, 1963), Vol. II, p. 994.
[14]R. M. Wilcox, Phys. Rev. 174, 624 (1968); G. Sauermann, Z. Phys. 231, 161 (1970).
[15]D. Forster, *Hydrodynamics Fluctuations, Broken Symmetry and Correlation Functions* (Benjamin, New York, 1975).
[16]S. Ma, Phys. Rev. B 22, 4484 (1980).
[17]D. Sherrington and S. Kirkpatrick, Phys. Rev. Lett. 35, 1792 (1973).
[18]D. J. Thouless, P. W. Anderson, and R. G. Palmer, Philos. Mag. 35, 593 (1977).
[19]T. Plefka, J. Phys. F 6, L327 (1976).