

Mode-coupling calculations of electron-spin resonance in long-range spin-glasses

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A full discussion of mode-coupling theories as applied to electron-spin resonance in long-range spin-glasses is presented. High-temperature linewidth and line-shift calculations, using the method of Kubo and Tomita, are presented and compared to results previously reported [P. M. Levy, C. Morgan-Pond, and R. Raghavan, *Phys. Rev. Lett.* **50**, 1160 (1983)], which used the Mori-Kawasaki formalism for all $T > T_g$. Justification for the decoupling procedure used and further work necessary for improved agreement close to the glass temperature T_g and in high resonance fields are discussed.

I. INTRODUCTION

Electron-spin resonance (ESR) experiments have been used for some time to investigate spin-glasses.¹ We present here a theoretical interpretation of some recent experiments on AgMn.² A brief account of the results has been reported earlier.³ We focus our interest here on AgMn (2.6 at. %) (Ref. 2), because the high-temperature susceptibility is Curie-like rather than Curie-Weiss-like, and results are available at different frequencies. The long-range model⁴ for spin-glasses predicts Curie-like behavior above the glass temperature T_g and is thus in accord with the results for AgMn (2.6 at. %). Results at different frequencies allow a more stringent test of the theories. The salient features of the data of Mozurkewich *et al.*² are the following. (1) As the temperature is lowered toward the glass temperature T_g , one finds an increase in the linewidth of the resonance over the expected linear (bottlenecked Korringa) behavior (see Fig. 1 of Ref. 2). (2) For temperatures close to T_g ($T/T_g \gtrsim 1.5$), the increase in linewidth becomes larger the smaller the measuring frequency, and it seems as if the linewidth would diverge at T_g for a zero-frequency resonance. (3) This divergence has been fitted to a power-law behavior $\Delta H = At^{-\gamma}$, where $t = T/T_g - 1$ and $\gamma \sim 1.5$, and is relatively independent of the concentration of Mn in the range studied. (4) A shift in the field for resonance or g shift occurs for temperatures below a crossover temperature t^* , where one finds the increase in linewidth depends on measuring frequency. (5) The temperature t^* decreases as the concentration of Mn ions increases. (6) Adding antimony impurities, which act as spin-orbit scatterers of the conduction electrons, increases the linewidths and shifts.

The increases in linewidth of the ESR resonance as T_g is approached is understood as follows.³ At high temperatures, $t \gg 1$, the linewidth comes from the relaxation of Mn ions to the lattice through the conduction electrons. This is described by a residual width of $\Delta H = a + bT$, with a bottlenecked Korringa rate b . Another contribu-

tion to the linewidth comes from the broadening of the resonance by anisotropic pair interactions between Mn ions. This is usually called dipolar broadening, but for transition-metal spin-glass alloys it is believed that the anisotropic Dzyaloshinsky-Moriya (DM) interaction rather than the dipole-dipole interaction dominates. At high temperatures these DM broadened resonances are narrowed by the isotropic exchange interaction between Mn ions and, as we will show, make a small contribution to the linewidth. However, as the temperature approaches T_g the Mn spins slow down and the exchange-narrowing mechanism becomes inoperative. Then the full DM broadened linewidth appears in the resonance.

Previous analyses of resonance in spin-glasses fall into two categories. Barnes⁴ has adapted the Bloch-Hasegawa theory for the ESR of magnetic alloys and extended it to spin-glasses in the *entire* paramagnetic regime. In the region close to the glass temperature he adopts Huber's theory⁵ of critical divergences of the spin-spin correlations for conventional antiferromagnets. In the second category of analyses attention is solely focused on the region close to T_g , and one describes the ESR properties in terms of the dynamical correlation functions of the spin-glass. This interpretation of the resonance behavior of spin-glasses was first made by Salamon and Herman¹ and more recently by Becker.⁶ Salamon and Herman decoupled their four-spin correlation functions and obtained some results which are at variance with extant data. Becker did not decouple his four-spin correlation functions but went on to the next-higher-spin correlation function. While the qualitative aspects of his solutions are in agreement with the experiment, unfortunately he is unable to obtain quantitative results.

In this paper we narrowly focus on explaining the increase in the linewidth and resonance shift in the region immediately above the glass temperature. We use the Mori-Kawasaki and memory-function formalisms as did Salamon and Becker. However, by adopting a different decoupling procedure from Salamon's, and by using the Dzyaloshinsky-Moriya interaction appropriate to spin-

glasses, we are able to show that the magnitude as well as the temperature dependences of the resonances linewidth and shift in AgMn can be understood on the basis of the slowing down of fluctuations in the spin system as it approaches the glass temperature.

In Sec. II, we introduce the Mori-Kawasaki and memory-function formalisms and discuss the approximations made in order to calculate resonance linewidths and shifts over the entire paramagnetic regime, and in particular close to T_g . In Sec. III, this formalism is applied to the calculation of the linewidth and shift due to the anisotropic DM interactions present in spin-glasses. A new decoupling procedure is used and justified, and we also use and justify the Kirkpatrick-Sherrington (KS) and Kinzel-Fischer (KF) formulas for the time-dependent spin-correlation functions for spin-glasses. To check the validity of our results in the high-temperature limit, we compare them in Sec. IV to the linewidth and shift calculated using the Kubo-Tomita method which is exact in this limit. In Sec. V we obtain quantitative estimates for our calculated widths and shifts and compare our results to the recent data on AgMn. We discuss the essential differences between resonance in spin-glasses and resonance in uniform systems, and point out what effects must be accounted for to obtain better agreement with experiment in the region very close to the glass temperature. We summarize our results in Sec. VI.

II. THE MEMORY-FUNCTION FORMALISM

In this section we review the memory-function formalism for the dynamics and decay of fluctuations in a statistical system and the simplified version of it that we use in our calculations.

Mori,⁷ following Kubo's⁸ earlier work, observed that the structure of linear operators in Hilbert space and of the Wigner-Weiskopf theory of line shape in quantum mechanics may be taken over to statistical mechanics by formally defining a space of dynamical variables or Heisenberg operators⁹ and defining the scalar product as

$$\chi_{AB} \equiv (A | B) \equiv \int_0^\beta d\lambda \text{Tr}[\rho A(i\lambda)B^*] - \beta \text{Tr}(\rho A)\text{Tr}(\rho B^*), \quad (2.1)$$

where $A(i\lambda) = e^{\lambda H} A e^{-\lambda H}$, β is the inverse temperature, $\beta = 1/k_B T$, ρ is the (equilibrium) density matrix, A and B are Heisenberg operators, and $*$ is used for the Hermitian adjoint. The Kubo correlation function χ_{AB} is an entry in the static isothermal susceptibility matrix.⁹ The operator analogous to the Hamiltonian H is iL , the Liouville operator which satisfies

$$A(t) = e^{iHt/\hbar} A(0) e^{-iHt/\hbar} \quad (2.1'a)$$

$$\equiv e^{-iL t} A(0) \quad (2.1'b)$$

so that

$$\frac{d}{dt} A(t) = iL A(t) \equiv \frac{1}{i\hbar} [A(t), H]. \quad (2.1'c)$$

In this notation

$$-iC_A(z) \equiv (A | \frac{1}{z-L} | A) \quad (2.2)$$

is related to the frequency-dependent susceptibility in linear-response theory as follows:

$$C_A(z) = - \frac{\chi(z) - \chi(z=0)}{z}. \quad (2.3)$$

The properties of $C(z)$, called the relaxation function, and $\chi(z)$ along with their time-dependent versions were discussed by Kubo.⁸ The imaginary part of the susceptibility gives the absorptive response and is related by the fluctuation-dissipation theorem to the correlation functions seen in scattering experiments.⁹

With these developments the analog of the Wigner-Weiskopf formulas¹⁰ may be immediately written⁷

$$iC_A(z) = - \frac{1}{z - w_0 + \Sigma(z)} \chi_A, \quad (2.4)$$

$$\chi_A = (A | A), \quad (2.4'a)$$

$$w_0 = -(A | L | A) \chi_A^{-1}, \quad (2.4'b)$$

$$\Sigma(z) = (A | LQ \frac{1}{z - QLQ} QL | A) \chi_A^{-1}, \quad (2.4'c)$$

where

$$1 - Q = |A) \chi_A^{-1} (A|. \quad (2.4'd)$$

Unlike the case in quantum mechanics the "states" are not orthonormal. Also, although we have written the formula for a single label A , more generally C , χ , ω , and Σ are matrices and the various multiplications, inverses, etc., must be considered as matrix operations.⁹ This is a necessary complication in the case of statistical dynamics. We also assume stationarity so that iL is Hermitian just as H is, i.e., $(A | iL | A) = (iL A | A)$.

The formula we shall use is a perturbational approximation to (2.4). In fact this approximation was used earlier than the general formalism to discuss spin dynamics in ferromagnets.¹¹ We write

$$\chi_A \Sigma(z) = (A | L_1 Q \frac{1}{z - L_0} QL_1 | A) \quad (2.5)$$

which implies a separation of the Hamiltonian into two parts: one which commutes with A or results in a systematic conserved motion and another causing it to decay in time. In our case (see below) A will be the component of the total spin in a given direction (appropriate to an ESR experiment), H_0 the isotropic Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction, and H_1 the anisotropic Dzyaloshinsky-Moriya interaction. The Liouville operator L_0 is related to H_0 and L_1 to H_1 as L is to H in Eqs. (2.1'a) and (2.1'b).

Using the definitions of the Liouville operator and the Laplace transform, Eqs. (2.1'a) and (2.1'b), we see, ignoring the Q 's, that

$$\Sigma(z) = \chi_A^{-1} \int_0^\infty dt e^{izt} (\dot{A}(t) | \dot{A}(0)), \quad (2.6)$$

where the overdots denote time derivatives as usual. We

notice that $\Sigma(z)$ in Eq. (2.5) is also of the form of a diagonal matrix element of a modified resolvent. It is therefore obvious that $\Sigma(z)$ has a representation analogous to (2.4'c) resulting in a continued-fraction expansion of $C(z)$.¹²

We are now in a position to discuss the various approximations used in treating spin systems. The Hamiltonian of interest in our paper is

$$H = H_0 + H_A + H_Z, \quad (2.7)$$

where H_0 is the isotropic RKKY interaction, H_A the anisotropic Dzyaloshinsky-Moriya term, and H_Z the Zeeman term due to the static field for resonance in the ESR experiment. As explained in Sec. III, these may be considered to be the relevant terms in the temperature ranges we consider. The explicit forms for the terms above are written there, Eq. (3.1).

Whether we use the "exact" formula (2.4) or the approximate one (2.6) for the self-energy, the chief difficulty is in calculating $\langle A | (z - L)^{-1} | A \rangle$ or $\langle \dot{A}(t) | \dot{A}(0) \rangle$ (our interest is in the ESR line shape whereas we have set $A = S_{\text{tot}}^x = \sum S_j^x$). In order to calculate $\langle \dot{A}(t) | \dot{A}(0) \rangle$, we need to compute a four-spin time-correlation function in the presence of many-body interactions. In the past mode-mode coupling theories were invented to deal with this problem.¹¹ In uniform-spin systems, the four-spin correlations were decoupled in the random-phase approximations (RPA).

In attempting to adapt the mode-coupling approach to the present problem we encounter several problems. The equations of motion in a random system do not justify the RPA decoupling in momentum space. In a uniform system the RPA decoupling is justified as an approximation by invoking momentum conservation and the exactness of the RPA decoupling for the spherical model.

A possible decoupling using the space of the eigenvectors of the interaction matrix is suggested¹ by the infinite-range model for spin-glasses.¹³ The spectrum of H_0 is not that of the RKKY interaction matrix, owing to the many-body nature of the spin operators and the constraints they have to satisfy. However, in the spherical model where we dispense with the constraint $S_i^2 = S(S+1)$ for each i in favor of the single constraint $\sum S_i^2 = NS(S+1)$, the diagonalization of the interaction matrix does yield the spectrum of H_0 . It is easy to show that the decoupling becomes exact in this space.¹³ It is in fact this procedure we follow (see calculations in Sec. III). This is inexact for a Heisenberg-type model but is expected to be better the higher the dimension and the higher the temperature.

Even though we have potentially identified the appropriate space in which a decoupling may be performed, there are two essential complications related to the nature of the spin-glass transition and to configuration averaging in disordered systems.

Unlike magnetic transitions in uniform systems, the spin-glass transition even in the KS limit is not dominated by one mode. Rather all modes contribute with more or less equal significance near the transition.¹³ Thus in the decoupling procedure we cannot include only one mode in the two spin correlations resulting from the decoupling, as

was done in the RPA decoupling for uniform systems, and in earlier work on spin-glasses.

Salamon has previously implemented such a decoupling for the ESR problem.¹ However, as we show in Sec. III he neglected an $O(N)$ term in the decoupled correlations in favor of an $O(1)$ term. This error is related to the fact mentioned above; namely, that no one mode can be identified as the critical mode in a spin-glass transition. One of the central results of this paper is to show that Salamon's ideas properly implemented can lead to a quantitative understanding of metallic spin-glasses.

We also perform the configuration averaging of the DM interactions and the spin correlations separately. This is a good perturbative approximation in the ESR calculation. If we could not make the separation of the Hamiltonian H into $H_0 + H_A$ where H_A is small and entirely responsible for the decay, we could not make this approximation.

We are thus left with the evaluation of $\langle S_\lambda(t) | S_\lambda(0) \rangle$ where λ refers to an eigenvalue of the interaction matrix. We return to this point later, after first discussing the approaches of Becker and of Barnes in the context of the general formalism.

Instead of decoupling the four-spin correlations, Becker proceeds to write the next term in the continued fraction for $\Sigma(z)$, involving eight-spin correlations, which one may decouple in favor of lower-order correlations. He uses the structure of the resulting equations to predict the structure of the ESR line shape in spin-glasses. Our paper is concerned with a quantitative calculation, without adjustable parameters, and so we have not included "two-spin variables" in our basis. It is inevitable that near the spin-glass line of the phase diagram¹⁴ our description is inadequate. Nevertheless, it would be premature to attempt the more difficult calculations without first exploring the simpler consequences of the theory.

It is also important to distinguish between streaming (reversible) and dissipative (irreversible) dynamics. Although from a microscopic point of view one writes the equations of motion as arising from a Hamiltonian, this is sometimes not a useful point of view. One is interested in fluctuations of thermodynamic variables which are defined in terms of ensembles of possible configurations in the limit of large systems. Thus the relevant equations of motion are derived from free-energy functionals,¹⁵ since one always works with a reduced Hamiltonian that disregards many degrees of freedom. In this case the equations for slowly relaxing variables may be predominantly dissipative, predominantly streaming (Hamiltonian), or a mixture.

In uniform systems the choice between dissipative and streaming dynamics near a phase transition is more easily made. The existence of dynamic scaling, together with the different exponents arising from reversible and irreversible behavior for the relaxation of important variable, often results in an unambiguous choice from experimental data. Near the spin-glass transition, which may not even be a phase transition, we are not so fortunate. Barnes⁴ has discussed ESR in metallic spin-glasses from the dissipative point of view.

Our *initial* formulation of the equations of motion is

from the Hamiltonian dynamics, with the decoupling mentioned. The remaining step is to evaluate the two-spin correlations. The Hamiltonian dynamics that results from H_0 is complicated and perhaps incomplete for a description of the time correlations. We use a *dissipative* description for the above time dependence and use the results of Kirkpatrick and Sherrington and of Kinzel and Fischer.¹³ We then identify an arbitrary flipping rate in the KS dynamics with the strength of the RKKY exchange constants to make the resulting calculations free of unknown parameters. A partial justification of this is offered by our high-temperature calculation (Sec. IV). Again this procedure is somewhat arbitrary but we feel that the resulting quantitative agreement implies that metallic spin-glasses do indeed experience the precursor of an KS-type transition. Let us summarize our discussions and indicate the calculations that are performed explicitly in Sec. III.

If the oscillating field in the ESR experiment is in the x direction, we wish to calculate the dynamic response of $S_{k=0}^x$. From the general relation between the relaxation function and the dynamic susceptibility it is sufficient to calculate the former.

We use the Mori-Kawasaki formula Eq. (2.5) or (2.6), which is a perturbational approximation that involves neglecting the mixing of various times scales. In particular the formula is not valid at long times unless the relaxation rate is much smaller than the frequency of measurement.

To compute $\dot{S}_x(t)$ we use the Hamiltonian, neglecting dissipative dynamics. This can only be justified by the results, although it can be argued for by separation of the time scale of the spin and the other degrees of freedom.

To decouple the resulting four-spin correlation, we go into the eigenbasis of the RKKY interaction matrix. We have already discussed this as the analog of the RPA implemented for a random spin system. This involves neglecting the dynamics for the replica variables.

To evaluate the two-spin correlations we use the Kirkpatrick-Sherrington and Kinzel-Fischer results. Again, this involves neglecting the Poisson bracket or commutator constraints among the spin variables.

We have not included H_Z (the Zeeman term) in evaluating the dynamics of the spins. This is not valid near a spin-glass transition, for resonance in high applied fields.

We have discussed the meaning of these approximations in the preceding, and we do so again in Sec. III. The point of the various assumptions is that they allow an explicit calculation to be performed with no free parameters.

III. CALCULATION

The ESR resonance comes from the excitation of an essentially uniform, $k=0$, mode of the spin system. As

$$\begin{aligned} \langle \dot{S}_{\text{tot}}^x(t) | \dot{S}_{\text{tot}}^x(t) \rangle &= \frac{1}{3} \frac{\langle D^2 \rangle_c}{\hbar^2} \sum_{\substack{\alpha, \beta, \gamma, \\ \beta', \gamma'}} (\delta_{\alpha\gamma} \delta_{\beta\alpha} - \delta_{\alpha\alpha} \delta_{\beta\gamma}) (\delta_{\alpha\gamma} \delta_{\beta\alpha} - \delta_{\alpha\alpha} \delta_{\beta\gamma}) \left[\sum_{i,j} (S_i^\beta(t) S_j^\gamma(t) | S_i^\beta S_j^\gamma) - \sum_{i,j} (S_i^\beta(t) S_j^\gamma(t) | S_i^\beta S_j^\gamma) \right] \\ &= \frac{1}{3} \frac{\langle D^2 \rangle_c}{\hbar^2} \sum_{\alpha=y,z} \sum_{i,j} [(S_i^x(t) S_j^\alpha(t) | S_i^x S_j^\alpha) - (S_i^x(t) S_j^\alpha(t) | S_i^x S_j^\alpha)] . \end{aligned} \quad (3.6)$$

such the linewidth and shift of this resonance is calculated by using Eq. (2.6), and the following Hamiltonian:

$$\begin{aligned} -H &= \frac{1}{2} \sum_{i,j} J_{ij} \vec{S}_i \cdot \vec{S}_j + \frac{1}{2} \sum_{i,j} \vec{D}_{ij} \cdot \vec{S}_i \times \vec{S}_j \\ &+ g \mu_B h_{\text{ext}} \sum_i S_i^z . \end{aligned} \quad (3.1)$$

Here the J_{ij} are randomly distributed according to a Gaussian distribution with a zero mean, and the D_{ij} represent the DM coupling between Mn ions as given by Fert and Levy.¹⁶ The Mn spins lose their energy to the lattice mainly via the conduction electrons because the direct spin-lattice coupling for s -state ions is very small. This mechanism produces the bottlenecked Korringa linewidth seen at high temperatures, which we will not consider in this paper. The justification for the various approximations has been mentioned in Sec. II.

Here we focus on the broadening due to the DM interactions, and find

$$\begin{aligned} \dot{S}_{\text{tot}}^x &= (i/\hbar) [H_{\text{DM}}, \vec{S}_{\text{tot}}] \\ &= (1/\hbar) \sum_{i,j} (\vec{D}_{ij} \times \vec{S}_i) \times \vec{S}_j . \end{aligned} \quad (3.2)$$

By replacing this result in the expression for the Kubo-Mori inner product we find

$$\begin{aligned} \langle \dot{S}_{\text{tot}}^x(t) | \dot{S}_{\text{tot}}^x(t) \rangle &= \left[\frac{1}{\hbar^2} \right] \sum_{i,j,k,l} \sum_{\substack{\alpha, \beta, \gamma, \\ \alpha', \beta', \gamma'}} f^x(\alpha, \beta, \gamma) f^x(\alpha', \beta', \gamma') D_{ij}^\alpha D_{kl}^{\alpha'} \\ &\quad \times (S_i^\beta(t) S_j^\gamma(t) | S_i^\beta S_j^\gamma) , \end{aligned} \quad (3.3)$$

where $f^x(\alpha, \beta, \gamma)$ is defined by

$$[(\vec{D}_{ij} \times \vec{S}_i) \times \vec{S}_j]^x = \sum_{\alpha, \beta, \gamma} f^x(\alpha, \beta, \gamma) D_{ij}^\alpha S_i^\beta S_j^\gamma . \quad (3.4)$$

For transition-metal spin-glass alloys, such as AgMn , the anisotropic interactions are small compared to the isotropic and the D_{ij} 's are not correlated to the Mn spins. Therefore we immediately average over the product of DM pair couplings in Eq. (3.3) and find

$$\langle D_{ij}^\alpha D_{kl}^{\alpha'} \rangle_c = \frac{1}{3} \langle D^2 \rangle_c (\delta_{ik} \delta_{jl} - \delta_{il} \delta_{jk}) \delta_{\alpha\alpha'} , \quad (3.5)$$

where

$$\langle D^2 \rangle_c = (1/N) \sum_j \sum_\alpha \langle (D_{ij}^\alpha)^2 \rangle_c$$

and the angular brackets represent the configurational average over a *random* distribution of the DM couplings. This average will be performed in Sec. V. By using Eq. (3.5) in the relaxation function Eq. (3.3) we find

To make the calculation tractable we decouple the four-spin correlations. We do this in the basis which diagonalizes the RKKY interaction matrix:¹³

$$H_{\text{RKKY}} = -\frac{1}{2} \sum_{i,j} J_{ij} \vec{S}_i \cdot \vec{S}_j = -\frac{1}{2} \sum_{\lambda} J_{\lambda} \vec{S}_{\lambda} \cdot \vec{S}_{\lambda}, \quad (3.7)$$

$$J_{ij} = \sum_{\lambda} J_{\lambda} \langle i | \lambda \rangle \langle \lambda | j \rangle,$$

where

$$S_i^{\beta} = \sum_{\lambda} S_{\lambda}^{\beta} \langle \lambda | i \rangle.$$

By using the above transformation and the orthonormality of the eigenvectors

$$\sum_i \langle \lambda | i \rangle \langle i | \lambda' \rangle = \delta_{\lambda\lambda'}$$

we find

$$\sum_{i,j} \langle S_i^{\beta}(t) S_j^{\gamma}(t) | S_i^{\beta} S_j^{\gamma} \rangle = \sum_{\lambda,\lambda'} \langle S_{\lambda}^{\beta}(t) S_{\lambda'}^{\gamma}(t) | S_{\lambda}^{\beta} S_{\lambda'}^{\gamma} \rangle. \quad (3.8)$$

It is at this stage that we decouple the four-spin products in Eq. (3.6). The basic idea is that variables which are weakly correlated can be separately evaluated. If the modes λ are eigenmodes of the interaction *Hamiltonian* then the expectation values entering Eq. (3.8) for different modes are evaluated independent of one another. Even where the λ 's are not eigenmodes we can make the assumption the different modes are weakly correlated and find that for thermal averages, denoted by $\langle \rangle$,

$$\begin{aligned} & \sum_{\lambda,\lambda'} \langle S_{\lambda}^{\alpha}(t) S_{\lambda'}^{\alpha}(t) S_{\lambda}^{\alpha} S_{\lambda'}^{\alpha} \rangle \\ & \cong \left[\sum_{\lambda} \langle S_{\lambda}^{\alpha}(t) S_{\lambda}^{\alpha} \rangle \right]^2 \\ & + \sum_{\lambda} (\langle S_{\lambda}^{\alpha}(t) S_{\lambda}^{\alpha}(t) S_{\lambda}^{\alpha} S_{\lambda}^{\alpha} \rangle - \langle S_{\lambda}^{\alpha}(t) S_{\lambda}^{\alpha} \rangle^2), \end{aligned} \quad (3.9a)$$

and

$$\sum_{\lambda,\lambda'} \langle S_{\lambda}^{\alpha}(t) S_{\lambda'}^{\alpha}(t) S_{\lambda}^{\alpha} S_{\lambda'}^{\alpha} \rangle \cong \sum_{\lambda} \langle S_{\lambda}^{\alpha}(t) S_{\lambda}^{\alpha}(t) S_{\lambda}^{\alpha} S_{\lambda}^{\alpha} \rangle. \quad (3.9b)$$

where $\alpha = y, z$ and we used the fact that

$$(S_{\lambda}^x(t) | S_{\lambda}^x) = (S_{\lambda}^y(t) | S_{\lambda}^y) = (S_{\lambda}^z(t) | S_{\lambda}^z) \quad (3.9c)$$

for a random system with cubic symmetry. We may decouple the Kubo-Mori inner product in a way analogous to the decoupling of the simple averages in Eqs. (3.9) by making the assumption that

$$\langle S_{\lambda}^{\alpha}(t) S_{\lambda'}^{\alpha}(t) | S_{\lambda}^{\alpha} S_{\lambda'}^{\alpha} \rangle \approx k_B T \left[\sum_{\lambda} \langle S_{\lambda}^{\alpha}(t) | S_{\lambda}^{\alpha} \rangle \right]^2. \quad (3.10)$$

By replacing the sums over λ in Eq. (3.9) by integrals and by recognizing that for spin-glasses *all modes* contribute¹³ we find that single sums over λ in Eqs. (3.9) and

(3.10) are of order $1/N$ compared to the first terms (double sum over λ, λ') and we will neglect them. By placing our approximation for the four-spin Kubo-Mori inner product in Eq. (3.6) we find the self-energy due to the DM interaction Eq. (2.6) is given as

$$\begin{aligned} \Gamma(\omega, T) &= \frac{2}{3} \frac{\langle D^2 \rangle_c}{\hbar^2} \frac{k_B T}{(S_{\text{tot}}^x | S_{\text{tot}}^x)} \\ &\times \int_0^{\infty} e^{-i\omega t} \left[\sum_{\lambda} \langle S_{\lambda}^x(t) | S_{\lambda}^x \rangle \right]^2 dt \end{aligned} \quad (3.11)$$

using condition (3.9c). For a homogeneous system where q space is used the above condition is valid only when one assumes isotropic pair interactions. The spin susceptibility in the denominator of (3.11) can be written above T_g in the λ basis as¹³

$$(S_{\text{tot}}^x | S_{\text{tot}}^x) = \sum_{\lambda} (S_{\lambda}^x | S_{\lambda}^x). \quad (3.12)$$

The noteworthy point in the expression for the self-energy Eq. (3.11) is that we find the term

$$\int_0^{\infty} \left[\sum_{\lambda} \chi_{\lambda}(t) \right]^2 e^{-i\omega t} dt, \quad (3.11'a)$$

while in the equivalent expression for homogeneous systems one finds⁵

$$\sum_k A_k \int_0^{\infty} [\chi_k(t)]^2 e^{-i\omega t} dt. \quad (3.11'b)$$

To complete our calculation we will assume the time-dependent spin correlations in Eq. (3.11) are governed by a Langevin equation with a flipping rate for the exchange field, $\Delta \approx k_B T_g \equiv \theta_g$. This assumption is physically reasonable, and similar to the assumption made by Anderson and Weiss¹⁷ for exchange narrowing in ferromagnets. The approximation is better at high temperatures, see Sec. IV, but in the final analysis our approximations can only be justified by the results. The correlation functions we use should become incorrect for T close enough to T_g in that mean-field theory breaks down. The region where corrections to mean field are important may extend up to $t \sim \frac{1}{4}$ in AgMn (2.6 at. %), based on a recent estimate by Morgan-Pond.¹⁸

The Langevin equation for the time dependence of the spins is

$$\frac{d}{dt} S_i^{\alpha}(t) = -\Delta [S_i^{\alpha}(t) - \gamma^{-1} \chi_0 h_i^{\alpha}(t)], \quad (3.13)$$

where χ_0 is the single-ion susceptibility, γ the gyromagnetic ratio for a spin, h_i is the field on the i th spin due to the others and Δ is the flipping rate. In Sec. IV we will determine its value by comparing our results using Eq. (3.13) to the linewidth and shift found by the method of Kubo and Tomita. From the work of Anderson and Weiss we can identify the Δ^{-1} as the characteristic time it takes the exchange field from other Mn spins to change,

$$\Delta \approx g \mu_B H_e \approx \frac{S(S+1)}{3\hbar} \tilde{J}, \quad (3.14)$$

where for a spin-glass the effective coupling \tilde{J} is

$$\tilde{J} \equiv \left[\sum_j J_{ij}^2 \right]^{1/2}. \quad (3.15) \quad \left[1 + \frac{1}{\Delta} \frac{d}{dt} + \beta^2 (\tilde{J}')^2 \right] (S_i^\alpha(t) | S_j^\alpha) = \beta \sum_k J'_{ik} (S_k^\alpha(t) | S_j^\alpha), \quad (3.19)$$

If we define the glass temperature as

$$k_B T_g = \theta_g = \frac{1}{3} S(S+1) \tilde{J} \quad (3.16)$$

we find that the flipping rate is

$$\Delta \approx \theta_g / \hbar. \quad (3.17)$$

When we make the additional assumption of neglecting the relaxation of the energy fluctuations in the time scale of interest, i.e., when we do not keep the energy in our basic set of dynamical variables (which is consonant with our decoupling procedure) we find

$$\frac{1}{\Delta} \frac{d}{dt} (S_i^\alpha(t) | S_j^\alpha) = -(S_i^\alpha(t) | S_j^\alpha) + \gamma^{-1} \chi_0 (h_i^\alpha(t) | S_j^\alpha). \quad (3.18)$$

By using the mean-field approximation with the reaction-field correction term¹³ to write the field $h_i^\alpha(t)$ in terms of exchange fields coming from other sites and recognizing that for $T > T_g$ the susceptibility χ_0 is proportional to $S(S+1)/3k_B T$ we find (with $\beta \equiv 1/k_B T$)

where $\tilde{J}' = \frac{1}{3} S(S+1) \tilde{J} \equiv \theta_g$, and $J'_{ik} = \frac{1}{3} S(S+1) J_{ik}$. This equation is identical to the linearized random kinetic equation for the decay of spin correlations above T_g derived by KS (Ref. 13) for a spin- $\frac{1}{2}$ Ising system. We conclude that within the approximations made the relaxation function $(S_\lambda^\alpha(t) | S_\lambda^\alpha)$ for a Heisenberg spin-glass, representing the time decay of spin correlations, is given by the equivalent function found by KS,¹³

$$(S_\lambda^\alpha(t) | S_\lambda^\alpha) = \frac{1}{3} \beta S(S+1) \frac{\exp[-\Delta(1+\beta^2\theta_g^2-\lambda)t]}{(1+\beta^2\theta_g^2-\lambda)}, \quad (3.20)$$

where on the right-hand side $\lambda = \beta J'_\lambda = \frac{1}{3} \beta S(S+1) J_\lambda$, and we set \tilde{J}' equal to θ_g .

To determine the linewidth and shift [Eq. (3.11)] with this time-dependent function we must evaluate the following integrals:

$$\int_0^\infty e^{-i\omega t} \left[\sum_\lambda (S_\lambda^x(t) | S_\lambda^x) \right]^2 dt = \frac{1}{9} N^2 \beta^2 [S(S+1)]^2 \int_{-\infty}^\infty \frac{\rho(\lambda) d\lambda}{1+\beta^2\theta_g^2-\lambda} \int_{-\infty}^\infty \frac{\rho(\lambda') d\lambda'}{1+\beta^2\theta_g^2-\lambda'} \int_0^\infty \exp\{-\Delta[2(1+\beta^2\theta_g^2)-\lambda-\lambda'+i\omega/\Delta]t\} dt. \quad (3.21)$$

When we use the semicircular density of states¹³ appropriate to a Gaussian distribution of random pair interactions J_{ij} , we find for the susceptibility in the denominator of Eq. (3.11)

$$(S_{\text{tot}}^x | S_{\text{tot}}^x) = \frac{1}{3} N \beta S(S+1). \quad (3.22)$$

By using this density of states, integrating over time in Eq. (3.21) and by placing the result Eq. (3.22) in Eq. (3.11) we find for the linewidth and shift

$$\Gamma(\omega, T) = A_0 I(\omega, T), \quad (3.23)$$

where the amplitude is given as

$$A_0 = \frac{2N \langle D^2 \rangle_c S(S+1)}{9\hbar^2 \Delta} \quad (3.24)$$

and the frequency and temperature dependence is given by

$$I(\omega, T) = \left[\frac{2}{\pi} \right]^2 \frac{1}{(2\beta\theta_g)^3} \int_{-1}^1 \int_{-1}^1 dx dy \frac{(1-x^2)^{1/2} (1-y^2)^{1/2}}{(a-x)(a-y)} \left[\frac{2a-x-y}{(2a-x-y)^2 + \tilde{\omega}^2} - \frac{i\tilde{\omega}}{(2a-x-y)^2 + \tilde{\omega}^2} \right], \quad (3.25)$$

where

$$a = \frac{1 + (\beta\theta_g)^2}{2\beta\theta_g},$$

and

$$\tilde{\omega} = \frac{\omega}{\Delta} \frac{1}{2\beta\theta_g}.$$

The real part of Γ determines the linewidth, and the imaginary term the second-order (in the anisotropy) line shift. The first-order shift coming from the presence of anisotropic interactions vanishes for a spin-glass, see Eq. (5.2).

The integrals in Eq. (3.25) may be written formally as a combination of rational functions and elliptic functions of complex parameters. At high temperatures and for $\omega/\Delta \ll 1$

$$I(\omega, T = \infty) = \frac{1}{2} \left[1 - \frac{i\omega}{2\Delta} \right] \quad (3.26)$$

so that there is both a temperature-independent linewidth (which would appear in phenomenological equations as part of the residual linewidth) and a shift that is linear in frequency and leads to a positive g shift. Also for zero field, i.e., at zero resonance frequency,

$$I(\omega=0, T) = \frac{1}{(\beta\theta_g)^2} \left\{ \frac{2a}{\pi\beta\theta_g} \left[K \left[\frac{1}{a} \right] - E \left[\frac{1}{a} \right] \right] - 1 \right\}, \quad (3.27)$$

where

$$K \left[\frac{1}{a} \right] - E \left[\frac{1}{a} \right] = \frac{1}{a} \int_0^{\pi/2} d\phi \frac{\sin^2\phi}{(a^2 - \sin^2\phi)^{1/2}},$$

and $K(1/a)$ and $E(1/a)$ are elliptic functions. Finally, at the glass temperature $T = T_g$, we find the linewidth diverges as the frequency goes to zero as

$$I(\omega \rightarrow 0, T_g) = -2\pi \ln \omega + \text{const} \quad (3.28)$$

while the line shift (imaginary part) remains finite for all frequencies.

In concluding this section, we may point out that it would be interesting to have an experimental determination of the time-dependent spin-correlation functions (via neutron-diffraction), similar to what has been published for Cu-Mn.¹⁹ This would serve as a useful check on the validity of the assumed spin-correlation functions.

IV. HIGH-TEMPERATURE CALCULATION

In order to check our estimate of the flipping rate for the exchange field, $\Delta \sim k_B T_g / \hbar$, we may calculate the high-temperature linewidth using the method of Kubo and Tomita.²⁰ This method uses a different form for the time correlations, not the Mori-Kawasaki form, so the ratio of the line shift to the linewidth differs from our ratio. This difference is not a constant factor, but depends on the value chosen for the flipping rate. In this section we will calculate the value of Δ needed to give the same linewidth in the high-temperature limit as is given by the Kubo-Tomita method, and the value of Δ needed to give the same line shift. We expect a choice of Δ in between these two limits to give the best parametrization fitting both linewidth and line shift in the high-temperature limit.

We use Kubo and Tomita's expressions for the exchange-narrowed linewidth $\Delta\omega$, and line shift δ , in the limit of high temperature and low applied field. These are

$$\Delta\omega = \sum_{\gamma=0,+,-} \sigma_\gamma^2 \tau'_\gamma, \quad (4.1)$$

$$\delta = \sum_{\gamma=0,+,-} (-\sigma_\gamma^2 \tau''_\gamma), \quad (4.2)$$

where

$$\sigma_\gamma^2 = \frac{\langle |[M_-, H_\gamma]|^2 \rangle}{\hbar^2 \langle |M_-|^2 \rangle}, \quad (4.3)$$

$$\tau'_\gamma = \left[\frac{\pi}{2} \right]^{1/2} \left[\frac{1}{\omega_{e\gamma}} \right], \quad (4.4a)$$

$$\tau''_\gamma = \frac{-f(\gamma)\omega_Z}{\omega_{e\gamma}^2}, \quad (4.4b)$$

$$f(0)=0, \quad f(+)=1, \quad f(-)=-1, \quad (4.4c)$$

$$\omega_Z = \frac{g\mu_B H_Z}{\hbar}, \quad (4.5)$$

and

$$\omega_{e\gamma}^2 = \frac{\langle |[H_{\text{ex}}, [M_-, H_\gamma]]|^2 \rangle}{\hbar^2 \langle |[M_-, H_\gamma]|^2 \rangle}. \quad (4.6)$$

The brackets $[]$ stand for commutators, and $\langle \rangle$ stand for configuration averages over lattice variables and expectation values for spin operators. ω_Z is the Zeeman frequency for the applied field H_Z and the above expressions for $\Delta\omega$ and δ are correct only for $\omega_Z \ll \omega_{e\gamma}$, for any γ such that $\sigma_\gamma^2 \neq 0$. H_{ex} is the exchange Hamiltonian that does the time-averaging of the anisotropy:

$$H_{\text{ex}} = \frac{1}{2} \sum_{i,j} J_{ij} \vec{S}_i \cdot \vec{S}_j, \quad (4.7)$$

and the anisotropy is given by

$$\begin{aligned} H_{\text{DM}} &= \frac{1}{2} \sum_{i,j} \vec{D}_{ij} \cdot \vec{S}_i \times \vec{S}_j \\ &= \sum_{\gamma=0,+,-} H_\gamma, \end{aligned} \quad (4.8)$$

where

$$H_0 = \frac{1}{2} \sum_{i,j} D_{ij}^z (S_i^x S_j^y - S_i^y S_j^x), \quad (4.9)$$

$$\begin{aligned} H_+ &= \frac{1}{4} \sum_{i,j} (D_{ij}^x - iD_{ij}^y) [(S_i^y S_j^z - S_i^z S_j^y) \\ &\quad + i(S_i^z S_j^x - S_i^x S_j^z)], \end{aligned} \quad (4.10)$$

$$\begin{aligned} H_- &= \frac{1}{4} \sum_{i,j} (D_{ij}^x + iD_{ij}^y) [(S_i^y S_j^z - S_i^z S_j^y) \\ &\quad - (S_i^z S_j^x - S_i^x S_j^z)]. \end{aligned} \quad (4.11)$$

M_- is given by

$$M_- = \frac{1}{2} (M_x - iM_y) = \frac{1}{2} g\mu_B \sum_j (S_j^x - iS_j^y). \quad (4.12)$$

Since $[M_-, H_-] = 0$ and $\sigma_-^2 = 0$, the sums will include only the terms $\gamma = 0, +$. For the $\gamma = 0$ term, we have

$$\begin{aligned} [M_-, H_0] &= \frac{g\mu_B}{4} \sum_{i,j} D_{ij}^z [i(S_i^x S_j^z - S_i^z S_j^x) \\ &\quad + (S_i^z S_j^y - S_i^y S_j^z)]. \end{aligned} \quad (4.13)$$

In the high-temperature limit, the expectation values of the spins are independent of the D_{ij} , and averages over different components may be taken separately, using $\langle (S_i^\alpha)^2 \rangle = 0$ and $\langle (S_i^\alpha)^2 \rangle = S(S+1)/3$. We may calculate $\langle |[M_-, H_0]|^2 \rangle$ by squaring Eq. (4.13) and taking the ex-

pectation values of the spins and the configuration averages of the D 's independently, noting that $\langle D_{ij}^\alpha D_{kl}^\beta \rangle_c \langle S_i^\alpha S_j^\beta S_l^\gamma S_l^\delta \rangle = 0$ unless either $i = k$ and $j = l$ or $i = l$ and $j = k$. This gives an additional factor of 2, and the sum over i, j, k , and l reduces to a sum over i and j :

$$\langle |[M_-, H_0]|^2 \rangle = \frac{g^2 \mu_B^2}{18} \sum_{i,j} |D_{ij}^z|^2 S^2 (S+1)^2. \quad (4.14)$$

We expect that D_{ij} will not be preferentially orientated along any axis in a random sample, so we may define an average anisotropy $\langle D^2 \rangle_c$, such that

$$NZ \langle D^2 \rangle_c \equiv 3 \sum_{i,j} (D_{ij}^\alpha)^2, \quad (4.15)$$

for $\alpha = x, y$, or z , where N is the total number of spins, and Z is the number of interacting neighbors per spin. Using $\langle D^2 \rangle_c$ we have

$$\langle |[M_-, H_0]|^2 \rangle = \frac{g^2 \mu_B^2}{54} NZ \langle D^2 \rangle_c S^2 (S+1)^2. \quad (4.16)$$

In a similar way, we may derive the following results:

$$\langle |M_-|^2 \rangle = \frac{1}{6} Ng^2 \mu_B^2 S(S+1), \quad (4.17)$$

$$\begin{aligned} \langle |[H_{ex}, [M_-, H_0]]|^2 \rangle \\ = \frac{2}{81} g^2 \mu_B^2 NZ^2 \langle D^2 \rangle_c \langle J^2 \rangle_c S^3 (S+1)^3, \end{aligned} \quad (4.18)$$

where we have defined

$$NZ \langle J^2 \rangle_c \equiv \sum_{i,j} J_{ij}^2. \quad (4.19)$$

We have kept only the lowest-order terms in $1/Z$ in Eq. (4.18), ignoring corrections of $O(NZ)$.

For $\gamma = +$, we have

$$\langle |[M_-, H_+]|^2 \rangle = \frac{g^2 \mu_B^2}{54} NZ \langle D^2 \rangle_c S^2 (S+1)^2, \quad (4.20)$$

$$\begin{aligned} \langle |[H_{ex}, [M_-, H_+]]|^2 \rangle \\ = \frac{2}{81} g^2 \mu_B^2 \langle D^2 \rangle_c NZ^2 \\ \times \langle J^2 \rangle_c S^3 (S+1)^3. \end{aligned} \quad (4.21)$$

Substituting these values into Eq. (4.3) and (4.4), we get

$$\sigma_0^2 = \sigma_+^2 = \frac{Z \langle D^2 \rangle_c S(S+1)}{9\hbar^2}, \quad (4.22)$$

$$\tau_0' = \tau_+' = \left[\frac{3\pi}{8} \right]^{1/2} \frac{\hbar}{Z^{1/2} \langle J^2 \rangle_c^{1/2} S^{1/2} (S+1)^{1/2}}, \quad (4.23)$$

$$\tau_0'' = 0, \quad (4.24a)$$

$$\tau_+' = \frac{3\hbar^2 \omega_Z}{4Z \langle J^2 \rangle_c S(S+1)}. \quad (4.24b)$$

We may simplify Eqs. (4.23) and (4.24) by making the approximation

$$k_B T_g = \frac{\langle J^2 \rangle_c^{1/2} S(S+1) Z^{1/2}}{3}. \quad (4.25)$$

By using Eqs. (4.1) and (4.2) for linewidth and line shift we find

$$\Delta\omega = \left[\frac{\pi}{6} \right]^{1/2} \frac{S^{3/2} (S+1)^{3/2} Z \langle D^2 \rangle_c}{9\hbar k_B T_g}, \quad (4.26)$$

$$\delta = \frac{Z \langle D^2 \rangle_c S^2 (S+1)^2 \omega_Z}{108 (k_B T_g)^2}. \quad (4.27)$$

We note that a positive value of δ corresponds to an increase in the resonance frequency for a particular applied field, to this line shift is in the same direction as the line shift we have calculated using the Mori-Kawasaki formalism. For the Mori-Kawasaki results, we have taken $Z = N$. If we assume $S = 2$ for AgMn, and set the Kubo-Tomita high-temperature linewidth equal to the Mori-Kawasaki value $A_0/2$ [Eq. (3.26)], we will need $\hbar\Delta = k_B T_g / \pi^{1/2} \cong 0.56 k_B T_g$.

In the limit of a strongly exchange-narrowed line, where $\langle D^2 \rangle_c \ll \langle J^2 \rangle_c$, at high temperature the Zeeman frequency ω_Z is approximately the resonant frequency ω , because the line shift is small. In this case, if we set the Kubo-Tomita high-temperature line shift equal to the Mori-Kawasaki value $A_0 \omega / 4\Delta$, we will need $\hbar\Delta = k_B T_g / \sqrt{2}$. The general expressions for general S are $\hbar\Delta = \{6/[S(S+1)\pi]\}^{1/2} k_B T_g$ for equal linewidths, and $\hbar\Delta = \{12/[S(S+1)]\}^{1/2} k_B T_g$ for equal line shifts. As we stated in the beginning of this section, a value of $\hbar\Delta$ intermediate between these two values should give the best fit to both linewidth and shift in the high-temperature limit.

V. DISCUSSION OF RESULTS AND COMPARISON WITH EXPERIMENT

From the general result Eq. (3.25), the solutions in various limits Eqs. (3.26)–(3.28), and Sec. IV we obtain the following description of the ESR behavior for Mn spins in a metallic host such as silver. Mori and Kawasaki assumed a time dependence

$$\begin{aligned} \frac{d}{dt} S_{\text{tot}}^x(t) &= [i\omega - \Gamma(\omega, T)] S_{\text{tot}}^x(t) \\ &= (i\{\omega - \text{Im}[\Gamma(\omega, T)]\} \\ &\quad - \text{Re}[\Gamma(\omega, T)]) S_{\text{tot}}^x(t). \end{aligned}$$

From this equation one can see that $\text{Im}\Gamma$ is the line shift and $\text{Re}\Gamma$ is the linewidth. As

$$\text{Im}[\Gamma(\omega, T)] \sim \omega,$$

one finds

$$\omega' = \omega \left[1 + \frac{A_0}{4\Delta} \right]$$

or

$$g_{\text{eff}} = g \left[1 + \frac{A_0}{4\Delta} \right].$$

In other words, at high temperatures, the part of the self-energy coming from anisotropic pair interactions contributes a temperature independent linewidth and a positive

g shift, which we have calculated:

$$\Delta H = \frac{1}{2} A_0, \quad g_{\text{eff}} = g \left[1 + \frac{A_0}{4\Delta} \right]. \quad (5.1)$$

These contributions are in addition to the "normal" linewidth and g shift coming from the local-moment-conduction-electron-lattice mechanisms described by the Block-Hasegawa equations which we have not considered.

As the characteristic temperature T_g is approached, the contribution from the anisotropic pair interactions increases. The increase in the linewidth appears sooner than the shift in the resonance as the temperature approaches T_g . At temperatures close to T_g , i.e., $T/T_g \sim 1.3$ the width becomes frequency dependent and it diverges in the limit as $\omega \rightarrow 0$ at $T = T_g$, or as $T \rightarrow T_g$ at $\omega = 0$. The shift of the resonance beyond the high-temperature value given in Eq. (3.26) appears at temperature close to T_g . For low frequencies $\omega \rightarrow 0$ the shift is zero except for temperatures very close to T_g , where it rapidly increases. At higher frequencies the shift appears at higher temperatures; its increase is more gradual and at T_g it reaches a value *smaller* than that for *lower* frequencies. In contrast to the width, which diverges at T_g , the shift remains *finite* at the glass temperature.

The results for $\text{Re}I(\omega, T)$ and $\text{Im}I(\omega, T)$, see Eq. (3.25), were plotted in Ref. 3 as the linewidth and shift, respectively, for two different frequencies, $\Omega \equiv \omega/\Delta = 5 \times 10^{-3}$ and 5×10^{-2} , corresponding to $\omega = 1$ and 9.3 GHz for AgMn with 2.6 at. % Mn, where we made the identification $\Delta \cong \theta_g$. The curves reproduce the *qualitative* behavior of the linewidth and shift observed² in AgMn remarkably well, given the severity of our assumptions. However, our results for the frequency dependence of the linewidth and the line shift showed only trends and not the shape of the experimental curves, since we have not correctly considered higher-frequency effects. We could better reproduce the experimental data at 9.3 GHz by using values of $\Omega = \omega/\Delta = 1$, which corresponds to either 200 GHz for $\Delta = \theta_g$, or $\Delta = 0.05\theta_g$ for $\omega = 9.3$ GHz. Neither of these seems physically correct and we are inclined to conclude that our result, Eq. (3.25), is incorrect at temperatures close to T_g and at high frequencies. There are several possible reasons for this. First, we took a mean-field approach and used the KS correlation function. As estimated by Morgan-Pond,¹⁸ corrections to mean-field theory which we have neglected may enter for $T \sim 1.25T_g$. Second, we have neglected the two spin (replica) variables in our solution and they may become important as T approaches T_g . Third, our naive scheme of decoupling the spin operators in λ space, see Eq. (3.9), breaks down when mixing of states with different λ by the Zeeman term becomes important. Finally, we should include the Zeeman term in our Eq. (3.13) for the time dependence of the spins. However as soon as we do this an extra term enters Eq. (3.19) and when we write the function $\langle S_i^\alpha(t) | S_j^\alpha \rangle$ in terms of $\langle S_{\lambda'}^\alpha(t) | S_{\lambda}^\alpha \rangle$ it does not decouple, i.e., terms with $\lambda' \neq \lambda$ exist.

There are two noteworthy differences between the resonance properties of a spin-glass and that of a homogene-

ous system where the ordered state can be identified with one mode, e.g., the $k=0$ mode of a ferromagnet. First, since in an ESR experiment on a spin-glass one does not couple directly to an order parameter, we do not expect the excess linewidth $I(\omega, T) - I(\omega, \infty)$ to have a simple power-law behavior in \tilde{t} ($\tilde{t} = T/T_g - 1$) as $\tilde{t} \rightarrow 0$. The linewidth, as given by Eq. (3.11), is represented by a double sum over *all* modes λ ; see Eq. (3.11'a). For homogeneous systems, see Eq. (3.11'b), only one mode goes "critical" and the exponent of the susceptibility ($S_k^\alpha | S_k^\alpha$) together with the dependences of the diffusion constant and dipole sums on the wave vector in the critical region determine the power-law behavior with temperature of the linewidth. In a spin-glass, one cannot single out one mode and we do not predict a power-law behavior of the linewidth as $\tilde{t} \rightarrow 0$. Nevertheless, when we plot our results for the linewidth on a log-log plot we could approximate them for $\frac{3}{4} < \tilde{t} < 4$ by a straight line with $\gamma \cong 1.4$. This is close to the observed exponent, see Ref. 2, however the observed linewidth has a power-law behavior for temperatures *closer* to T_g , i.e., down to $\tilde{t} \sim \frac{1}{3}$.

The second difference in the ESR behavior of spin-glass is that the line shift first enters in second order in the anisotropic interaction H_{DM} , whereas for homogeneous systems an anisotropic pair interaction could contribute to the shift in first order of H_{aniso} . For homogeneous systems, anisotropic pair interactions can produce an anisotropy of the susceptibility and therefore produce a shift to first order in H_{aniso} . However, for random systems the first-order contribution of H_{aniso} vanishes if we average over the random anisotropy vectors D_{ij} independently of the spin averaging, as we have done in our calculation. The first moment of the frequency spectrum, as given by Mori and Kawasaki,¹¹ is

$$\omega_0 = \chi^{-1} \int_0^\infty dt \langle \dot{S}(t) | S(0) \rangle. \quad (5.2)$$

We readily see that when we average over the D_{ij} we obtain $\omega_0 = 0$ because the configurational average over the D_{ij} 's vanishes.

Finally, we calculate the amplitude A_0 , Eq. (3.23), by estimating $\langle D^2 \rangle_c$, see Eq. (3.5). In spin-glasses involving S -state ions as Mn the major contribution to the anisotropy comes from the spin-orbit coupling of the conduction electrons *induced* by a Mn ion or by a ternary impurity, such as antimony in the experiments of Mozurkewich *et al.* As shown by Fert and Levy,¹⁶ the anisotropic pair coupling between two Mn spins induced by the spin-orbit scattering of the conduction electrons by a third ion is of the Dyzaloshinsky-Moriya form with the coupling constant

$$\vec{D}_{ij} = \frac{V_1 \sin[k_F(R_i + R_j + R_{ij}) + \phi] \hat{R}_i \cdot \hat{R}_j (\hat{R}_i \times \hat{R}_j)}{R_i R_j R_{ij} [1 + \alpha(R_i + R_j + R_{ij})]}, \quad (5.3)$$

where

$$V_1 = 15 \frac{\lambda_d}{E_F} \sin \left[\frac{\pi}{5} \left(\frac{5}{2} - S \right) \right] V_0, \quad (5.4)$$

$$\alpha = (\Delta/2\pi E_F) k_F, \quad (5.5)$$

V_0 is the RKKY interaction constant, and we used, for the phase shift η_2 of Mn, $\eta_2 = (\pi/5)$, $(\frac{5}{2} - S)$. The position vectors \vec{R} are referred to the ternary site as the ori-

$$\begin{aligned} N\langle D^2 \rangle_c &= V_1^2 \langle \sin^2[k_F(r+R + |\vec{r}-\vec{R}|) + \phi] \rangle_c \\ &\times \frac{C_{\text{Mn}}C_{\text{imp}}}{v_A^2} \int_0^\infty dR 4\pi R^2 \int_0^\infty dr \int_0^\pi d\theta \sin\theta \frac{2\pi r^2 \cos^2\theta \sin^2\theta}{r^2 R^2 (r^2 + R^2 - 2rR \cos\theta) [1 + \alpha(r+R + |\vec{r}-\vec{R}|)]^2} \\ &= 8\pi^2 V_1^2 \frac{C_{\text{Mn}}C_{\text{imp}}}{v_A^2} \int_0^\infty dr \frac{1}{R} \int_0^\infty ds \int_{-1}^1 \frac{x^2(1-x^2)}{(1+s^2-2sx) \left[1 + \alpha \left(\sum R\right)\right]^2}, \end{aligned} \quad (5.6)$$

where we have used $\sum R = r+R + |\vec{r}-\vec{R}|$, $s = r/R$, choosing always r to be the smaller of r and R , and multiplying the whole integral by 2 to include both the case where the Mn is closer to the origin and the case where the impurity is closer. Also, $\langle \sin^2[k_F(\sum R) + \phi] \rangle \cong \frac{1}{2}$, v_A denotes atomic volume of a host ion, i.e., for a fcc lattice $v_A = a^3/4$, and $C_{\text{Mn}}, C_{\text{imp}}$ are the concentrations of Mn and impurity ions. The radial integral over $[1 + \alpha(\sum R)]^{-2}$ is approximated as

$$\begin{aligned} \int_{r_0}^\infty dR \frac{1}{R} \frac{1}{\left[1 + \alpha \left(\sum R\right)\right]^2} & \\ &\cong \int_{r_0}^{R_c} dR \frac{1}{R} + \int_{R_c}^\infty dR \frac{1}{(3\alpha R)^2 R} \\ &= \ln \frac{R_c}{r_0} + \frac{1}{2} \frac{1}{(3\alpha R_c)^2} \\ &= \frac{1}{2} + \ln(R_c/r_0), \end{aligned} \quad (5.7)$$

where r_0 is the nearest-neighbor distance on the host lattice, and where R_c is defined as that value R for which

$$\alpha \sum R \cong \alpha(r+R + |\vec{r}-\vec{R}|) \cong 3\alpha R.$$

is comparable to the other term in the denominator, i.e.,

$$3\alpha R_c = 1. \quad (5.8)$$

The remaining integral is written as

$$2 \int_0^\infty ds \int_{-1}^1 dx \frac{x^2(1-x^2)}{1+s^2-2sx} \cong \frac{8}{15}. \quad (5.9)$$

By placing these results in Eq. (5.6) we find

$$\begin{aligned} N\langle D^2 \rangle_c &= \frac{32\pi^2}{15V_A^2} \left[\ln \frac{R_c}{r_0} + \frac{1}{2} \right] \\ &\times [(V_1^{\text{Mn}})^2 x^2 + (V_1^T)^2 xy], \end{aligned} \quad (5.10)$$

where x is the concentration of Mn and y is the concentration of the ternary impurity, if any.

We use $\lambda_d = 0.03$ eV as the spin-orbit coupling strength, $E_F = 5.5$ eV as the Fermi energy, $V_0/v_A = 2.32 \times 10^6 \text{G} \mu_B$ per Mn,¹⁶ and estimate $S = 2$ for the Mn spin value. In Eq. (5.10), we use $r_0 = 2.89$ Å, $x = 0.026$, $y = 0$, and $R_c = 38.7$ Å, estimated using the values $k_F = 1.19$ Å⁻¹ as the Fermi wave vector, and $\Delta = 0.25$ eV as the width of

gin. By using this coupling constant and assuming a random distribution of Mn and impurity ions, we can write the configurational average of D_{ij}^2 , Eq. (3.5), as

the virtual bound state. This gives us a final value of

$$N\langle D^2 \rangle_c = 1.51 \times 10^8 (\text{G} \mu_B)^2. \quad (5.11)$$

By using $T_g = 10.3$ K for 2.6 at. % Mn in Ag, $S = 2$, and identifying the flipping rate Δ as $\theta_g = 1.53 \times 10^5 \text{G} \mu_B / \hbar$ we find A_0 , Eq. (3.24), is given by

$$A_0 = 1320 \text{G} \mu_B / \hbar = 660 \text{G} \quad (5.12)$$

in units of field, using $g = 2$.

As discussed in Ref. 3, this value represents an upper limit on A_0 , since we have used $S = 2$. From considerations of the screening of Mn 3d electrons, we expect $S_{\text{Mn}} > 2$ and we see from Eqs. (5.4) and (5.10) a rapid reduction of A_0 . Since $A_0^{\text{expt}} = 312$ G, we find that the DM interaction is amply able to explain the observed magnitude of the low-frequency linewidth.

As a function of ternary impurity concentration we predict on the basis of Eq. (5.10) that the linewidth and shift scale linearly with the concentration of a ternary impurity y , provided the concentration is not so large as to alter T_g . This is in agreement with the data on AgMn with antimony.² Although from simplest considerations of the RKKY interaction one expects T_g to increase linearly with the concentration of Mn, experiments have shown, and Walker and Walstedt have given the rationale²¹ for, a less rapid increase. Causes similar to those which produce this deviation from linearity will change our dependence of $\langle D^2 \rangle_c$ on x . Therefore it is not surprising that the experimental variation of A_0 with Mn concentration found by Mozurkewich *et al.* is also sub-linear.²

VI. SUMMARY

In conclusion, we have attempted a microscopic theory of the ESR resonance in AgMn and have found quantitative agreement, at low frequencies, for the linewidth. The line shift, the high-frequency linewidth as well as the constant high-temperature value found, see Eq. (3.26), all await further developments to receive satisfactory explanation. In particular, the mechanism explored here must be integrated with other mechanisms indigenous to spin-glasses. It is inevitable that near the spin-glass temperature our description is inadequate, i.e., we cannot simply decouple considering only single-spin variables. Nevertheless, before attempting more difficult and at this time intractable calculations, it was worthwhile exploring the simpler consequences of the theory presented here.

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