Theory of electron-spin-resonance linewidth and line-shift effects in spin-glasses with anisotropy and zero remanent magnetization

Klaus W. Becker*
Department of Physics, University of California, San Diego, La Jolla, California 92093 and Institute for Pure and Applied Physical Sciences, University of California, San Diego, La Jolla, California 92093

(Received 13 July 1981; revised manuscript received 16 February 1982)

The dramatic increase in the width of the electron-spin-resonance line in Ruderman-Kittel-Kasuya-Yosida spin-glasses at temperatures above the transition temperature T_f, recently regarded as evidence in favor of a phase transition, is explained as an effect of the small measuring frequency. The present theory also gives an explanation for the line-shift behavior as a function of the measuring frequency and the temperature.

I. INTRODUCTION

Only recently considerable progress has been made in understanding electron-spin-resonance (ESR) measurements in Ruderman-Kittel-Kasuya-Yosida (RKKY) spin-glasses such as CuMn. In the earlier work[1-4] a large shift of the resonance field was observed in the spin-glass regime and at first regarded as a "special kind of antiferromagnetic resonance." However, as it turned out, it is essential to refer to the magnetization properties of the spin-glass state because the ESR results for large-remanent magnetization differ completely from those at small or zero remanence.[5,6] In the former case only one ESR frequency was found whereas two resonances were seen in the latter case with completely different values for slopes and intercepts in the frequency-field relationship. In order to interpret the experimental results, Schultz *et al.*[6] invented a phenomenological free energy incorporating magnetic remanence, anisotropy energy, and Zeeman energy. In the preceding paper,[7] hereafter referred to as I, a microscopic derivation was given for the ESR excitations in RKKY spin-glass systems with remanence and anisotropy. 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 more similar to a Heisenberg antiferromagnet. The anisotropy constant, introduced in Ref. 6, was shown to be caused by anisotropic interactions between the spins. However, in this theoretical approach all linewidth effects were considered as small. This approximation, certainly valid for low temperatures, might be violated for temperatures at or above T_f. Here the anisotropy constant which determines the ESR positions can become very small so that linewidth effects can no longer be neglected.

On the other side, for the same temperature regime, ESR measurements were reported that showed a dramatic increase in the linewidth.[8-12] This was considered to be evidence in favor of a phase transition in spin-glass systems and also explained in this way by an exchange-narrowing effect.[8-10] However, caution should be exercised in analyzing spin-glass ESR in terms of critical dynamics, since the linewidth increase for higher measuring frequencies is far less pronounced.[12] Indeed it will be shown that the dramatic linewidth behavior can be understood as an effect of the small measuring frequency without any need to refer to critical properties of phase transitions.

II. DYNAMICAL SUSCEPTIBILITY

As in I we consider a spin-glass system consisting of N quantum spins randomly distributed in a metallic host,

$$\mathscr{H}=\mathscr{H}_H+\mathscr{H}_A-\gamma\vec{H}^{\text{ex}}\cdot\sum_{i=1}^{N}\vec{S}_i\ , \tag{1}$$

where $\mathscr{H}_H$ is the dominant RKKY exchange interaction mediated by the conduction electrons, $\mathscr{H}_A$ is a smaller anisotropic interaction between the spins, and the last term is the Zeeman energy. To discuss the dynamical behavior, one has to investigate the dynamical susceptibility $\chi(\omega)$ since all information concerning the widths and positions of the ESR lines is contained in $\chi(\omega)$. Based on the projection-operator technique an exact transformation of $\chi(\omega)$ can be obtained (cf. I),

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

where $\chi_\perp$ is the static transverse susceptibility (strictly speaking it is the isolated susceptibility) and $M_1(\omega)$ is an ω-dependent self-energy, defined in Eq. (B5) of I, which describes all relaxation mechanisms of the magnetization $\mathscr{M}^-$. Furthermore, $M_1(\omega)$ can be expressed by

$$M_1(\omega)=\frac{\chi_2/\beta}{\omega_2-\omega-M_2(\omega)/(\chi_2/\beta)}, \tag{2b}$$

according to I. The new quantity $M_2(\omega)$ now describes the relaxation of the variable $\dot{\mathscr{M}}^-$ (time derivative of $\mathscr{M}^-$) into all nonmacrovariables (i.e., all operators perpendicular to $\mathscr{M}^-$ and $\dot{\mathscr{M}}^-$) and is responsible for linewidth effects. Finally, ω_1,ω_2 are frequency terms and χ_2 is a generalized susceptibility,

$$\begin{aligned}\omega_1&=\frac{\gamma M_z}{\chi_\perp},\quad \chi_2=K-\frac{\Delta^2/2}{\chi_\perp},\\ \omega_2&=\frac{\langle\langle[[\mathscr{H}_A,\mathscr{M}^-]^+,[\mathscr{H}_A,\mathscr{M}^-]]\rangle\rangle_{av}}{\chi_2}\\ &\quad-\frac{\Delta}{2\chi_\perp}\left[2+\left[\gamma H^{ex}+\frac{\Delta/2}{\chi_\perp}\right]\frac{\Delta}{\chi_2}\right].\end{aligned} \tag{3}$$

Here the quantity M_z is the total magnetization $M_z=M_0+\chi_{\parallel}H^{ex}$ ($\chi_{\parallel}$ is the longitudinal susceptibility and M_0 is the remanent magnetization), and K is the anisotropy constant defined by ($\mathscr{M}^-$ is the magnetization operator)

$$K=\int_0^\beta d\lambda\langle\langle[\mathscr{H}_A,\mathscr{M}^-]^+e^{-\lambda\mathscr{H}}[\mathscr{H}_A,\mathscr{M}^-]e^{\lambda\mathscr{H}}\rangle\rangle_{av}, \tag{4}$$

where $\langle\ \rangle_{av}$ indicates the average over the random distribution of the magnetic ions. Finally, the quantity Δ means

$$\frac{\Delta}{2}=\gamma M_z-\gamma H^{ex}\chi_\perp. \tag{5}$$

The above equations (2)–(5) describe the spin dynamics in a spin-glass with anisotropy and either large- or small-remanent magnetization. For the dynamical behavior in the spin-glass regime at or above T_f, however, we can assume the remanent magnetization to be small, i.e., $M_0\approx 0$, in agreement with the experimental findings.[13] For this case of zero remanence and not too large values of the applied resonance field the longitudinal and the transverse static susceptibility are approximately equal because of the lack of a preferred direction in the spin-glass state, i.e., $\chi_\perp\approx\chi_{\parallel}$. Thus we have $\Delta\approx 0$ and the above quantities reduce to

$$\chi_2\approx K,\quad \omega_1\approx\gamma H^{ex},\quad \omega_2\approx 0, \tag{6}$$

where we have neglected the commutator term of ω_2 in agreement with experiment (cf. I). In the following, we shall neglect the real part of $M_2(\omega)$ and assume that the imaginary part is almost frequency independent at least in the interesting frequency regime of the ESR lines, i.e., $M_2(\omega)\approx iM_2$ (Markov approximation). Thus (2) becomes

$$\chi(\omega)=\chi_\perp\left[1+\frac{\omega}{\gamma H^{ex}-\omega-(K/\chi_\perp)/[-\omega-iM_2/(K/\beta)]}\right] \tag{7}$$

or by separating the real and the imaginary part in the denominator of (7),

$$\begin{aligned}\chi(\omega)=\chi_\perp\{1+\omega/[\gamma H^{ex}-\omega(1-(K/\chi_\perp)/\{\omega^2+[M_2/(K/\beta)]^2\})\\ -i(K/\chi_\perp)[M_2/(K/\beta)]/\{\omega^2+[M^2/(K/\beta)]^2\}]^{-1}\}.\end{aligned} \tag{8}$$

Note $\chi(\omega)$ is not only a function of the measuring frequency ω but also a function of the applied field H^{ex} (and of the temperature T as well). Indeed, Eq. (8) just describes a resonance behavior for the dynamical susceptibility as a function of the applied field (for fixed value of ω) assuming the different quantities K, $\chi_\perp$, and M_2 are independent of H^{ex}. Though these quantities do certainly depend on H^{ex}, as is, for instance, well known for the static susceptibility $\chi_\perp$, as an approximation we shall henceforth neglect this dependence. We believe that for spin-glasses such as CuMn this approximation does not change the general characteristics of $\chi(H^{ex},\omega_{fixed})$ but might change quantitative results. Thus, the resonance behavior of the dynamical susceptibility as a function of the external field is given by

$$\chi(H^{ex},\omega_{fixed})=\chi_\perp\left[1+\frac{\omega/\gamma}{(H^{ex}-H_r)-i\Delta H}\right], \tag{9}$$

where H_r is the resonance field and ΔH the linewidth,

$$H_r = \frac{\omega}{\gamma}\left[1 - \frac{K/\chi_\perp}{\omega^2 + [M_2/(K/\beta)]^2}\right], \tag{10}$$

$$\Delta H = \frac{K}{\gamma\chi_\perp}\frac{M_2/(K/\beta)}{\omega^2 + [M_2/(K/\beta)]^2}. \tag{11}$$

The resonance field H_r and the linewidth ΔH both depend on the measuring frequency and on the temperature. For the temperature behavior, we refer to the temperature dependence of the anisotropy constant. Here the experiments[6] have shown that K decreases linearly from its zero-temperature value with increasing temperature as $-T$, i.e.,

$$K(T) = K(0)(1 - 0.67T/T_f')$$

and is zero for higher temperatures $T > 1.5T_f'$. (The theoretical investigation in I gave a less decreasing function for $T > T_f$ as T^{-1}; however, an infinite-range model and an evaluation method equivalent to the replica method was used.) For the following discussion, we shall assume a temperature dependence for $M_2/(K/\beta)$ which is mainly determined by the T dependence of K alone (that is, $M_2\beta$ is assumed not to depend very much on T). Thus, the quantity $M_2/(K/\beta)$ can be considered large (i.e., $[M_2/(K/\beta)]^2 \gg \omega^2$) for sufficiently high temperatures and small (i.e., $[M_2/(K/\beta)]^2 \ll \omega^2$) for the lower-temperature regime. The transition between both regimes depends on the value of the measuring frequency ω.

III. LINE-SHIFT EFFECTS

Owing to the assumed temperature dependence of $M_2/(K/\beta)$ we expect characteristic differences in the experimental behavior of the line shift and the linewidth for the two temperature regimes $[M_2/(K/\beta)]^2 \gg \omega^2$ and $[M_2/(K/\beta)]^2 \ll \omega^2$. Of course, the temperatures where the changes occur depend on the value of the frequency. At first let us consider the line shift $\delta H \equiv \omega/\gamma - H_r$ which is defined as the difference between the Larmor frequency and the actual resonance field,

$$\delta H = \frac{K}{\gamma\chi_\perp}\frac{\omega}{\omega^2 + [M_2/(K/\beta)]^2}. \tag{12}$$

In the low-temperature regime, where $[M_2/(K/\beta)]^2 \ll \omega^2$ is valid, δH reduces to

$$\delta H = \frac{K}{\gamma\chi_\perp}\frac{1}{\omega}. \tag{13}$$

This equation was used in Ref. 6 to extract the anisotropy constant and its T dependence from experiment. However, relation (13) is only valid when $[M_2/(K/\beta)]^2$ can be neglected compared to ω^2. Deviations are expected to be seen for higher temperatures when $[M_2/(K/\beta)]^2 \gtrsim \omega^2$ or for smaller values of the measuring frequency when T is fixed. Let us define the quantity $\tilde{K} = \gamma\delta H\chi_\perp\omega$, which can be extracted from the experimental data. Using (12) we find

$$\tilde{K} = \frac{\omega^2}{\omega^2 + [M_2/(K/\beta)]^2}K. \tag{14}$$

Therefore, for fixed temperature $\tilde{K}$ is equal to K only for sufficiently high frequencies. However, for smaller frequencies deviations from K should be seen in $\tilde{K}$ (Fig. 1). This behavior was indeed found in experiment.[14] As a function of the measuring frequency the quantity $\tilde{K}/\omega^2$ should behave like a Lorentzian curve with a linewidth $M_2/(K/\beta)$ that decreases with decreasing temperature.

A similar check of the theory can be made by measuring the lineshift as a function of T for different values of the frequency ω. For small temperatures $[M_2/(K/\beta)]^2 \ll \omega^2$ the theoretical result is given by (13) whereas for large temperatures $[M_2/(K/\beta)]^2 \gg \omega^2$ the expression

$$\delta H = \frac{K}{\gamma\chi_\perp}\frac{\omega}{[M_2/(K/\beta)]^2}$$

should be valid. Thus one expects a qualitative behavior of the line shift as a function of T as is shown in Fig. 2 for two different frequency values ω_1 and ω_2 ($>\omega_1$): For the lower-temperature regime the line shift with ω_1 should be larger than that with ω_2, whereas at higher temperatures the situation should be reversed. Again the theoretical result agrees with the experimental findings.[14]

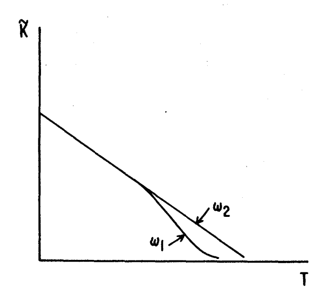

FIG. 1. Qualitative behavior of $\tilde{K}$, defined by (14), as a function of T. For small measuring frequencies and high temperatures $\tilde{K}$ is expected to deviate from the anisotropy constant K ($\omega_2 > \omega_1$).

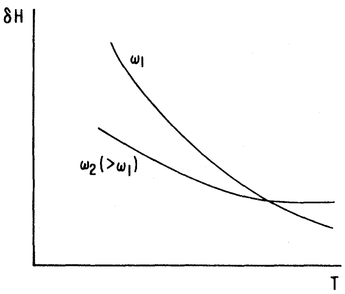

FIG. 2. Qualitative behavior of the lineshift δH as a function of T for two different frequencies ω_1 and ω_2 $(>\omega_1)$.

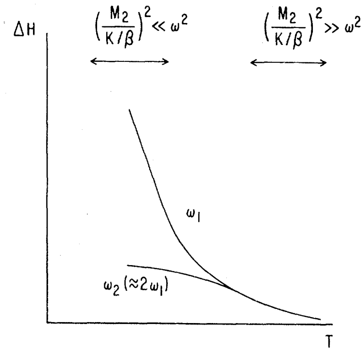

FIG. 3. Qualitative behavior of the linewidth ΔH as a function of T for two different frequencies ω_1 and ω_2 $(\approx 2\omega_1)$.

IV. LINEWIDTH EFFECTS

Again we discuss the cases of small and large temperatures separately. For high temperatures, when $[M_2/(K/\beta)]^2$ is very large compared to ω^2, we have

$$\Delta H \approx \frac{K}{\gamma \chi_\perp} \frac{1}{M_2/(K/\beta)}, \quad [M_2/(K/\beta)]^2 \gg \omega^2 . \tag{15}$$

Thus, the linewidth should be independent of the measuring frequency. One also expects ΔH to become very small because K is small and $M_2/(K/\beta)$ in the denominator of (15) is very large. However, in the opposite case, when $[M_2/(K/\beta)]^2 \ll \omega^2$, ΔH is no longer frequency independent. Instead of (15) we find

$$\Delta H \approx \frac{K}{\gamma \chi_\perp} \frac{M_2/(K/\beta)}{\omega^2}, \quad [M_2/(K/\beta)]^2 \ll \omega^2 . \tag{16}$$

That means in this regime the linewidth ΔH is inversely proportional to ω^2, i.e., $\Delta H \sim 1/\omega^2$, so that the linewidth can be dramatically increased by reducing the measuring frequency (compare Fig. 3). Therefore, we believe that this theory can explain the above-mentioned enormous increase of the ESR linewidth found recently.[8–12] It was claimed that this increase was a characteristic feature of the occurrence of a phase transition in a spin-glass and was also theoretically explained in this way. Indeed, a reduction of the measuring frequency, for instance, by a factor of 9 should lead to an increase of the ESR linewidth by a factor of 81, so that it looks like a phase transition occurs. However, the theory used here is a conventional theory without any anomalous behavior of the self-energy at or near the spin-glass transition temperature T_f. Note that the resonance field H_r, Eq. (10), goes to zero for small temperatures when $K/\chi_\perp \simeq \omega^2$.

It may be instructive to compare the relaxation mechanism leading to the ESR linewidth (11) with that of an ordinary T_2 relaxation process. Consider the first self-energy $M_1(\omega)$ defined in (B5) of I, which describes the relaxation of the magnetization operator due to anisotropic interactions. In an ordinary T_2 process $M_1(\omega)$ is usually considered as ω independent, i.e., $M_1(\omega) \simeq iM_1$. This leads to an ω-independent ESR linewidth $\Delta H = M_1/(2\chi_\perp/\beta)$ if the H^{ex} dependence of M_1 and $\chi_\perp$ is negligible. For the damping mechanism considered here it is essential to extract the ω dependence from $M_1(\omega)$ as was done in (2b). Note the final expression (7) for $\chi(\omega)$ is equivalent to the description of the resonance properties in spin-glasses by two coupled modes in agreement with Schultz *et al.*[6] In (7) the second self-energy determines the ESR linewidth. M_2, defined by (3.8) of I describes the relaxation of the two-spin variable $[H_A, \mathscr{M}^-]$ (which is the second dynamical variable for the zero-remanence case) due to the dominant exchange interaction. Note in the high-temperature regime, $[M_2/(K/\beta)]^2 \ll \omega^2$, the first self-energy M_1 and thus the ESR linewidth (11) become ω independent, and the relaxation mechanism can again be considered as an ordinary T_2 process.

One should mention that the experimental ESR linewidth ΔH shows an ω-independent background contribution[14] instead of decreasing to zero for large measuring frequencies as expected by Eq. (11). This behavior could be caused by additional relaxation mechanism of the localized moments [e.g., to conduction electrons or to the lattice which are not included in the model Hamiltonian (1)]. The outlined theory is easily generalized to include such new relaxation channels leading to additional contributions to $M_1(\omega)$ in (2b). For instance, for an *sd* interaction between the localized moments and the conduction electrons the resulting extra linewidth is the usual Korringa rate (for the so-called isothermal case). However also each different additional relaxation contribution to $M_1(\omega)$ with negligible ω dependence leads to an ω-independent background in ΔH.

Finally note that ordinary T_1 processes (longitudinal relaxation) do not enter the linear transverse response function $\chi(\omega)$ for the considered case of dominant isotropic exchange and small anisotropic interaction. T_1 processes should appear if contributions nonlinear in the applied transverse field $h_x \cos\omega t$ [compare (3.1) in I] are included in the response function.

IV. CONCLUSION

It was shown that the dramatic increase of the ESR linewidth when the temperature approaches T_f from above can be explained as an effect of the measuring frequency. The essential assumption for the understanding of this effect is the temperature dependence of the quantity $M_2/(K/\beta)$. Since, however, characteristic features of the experimental line shift can also be consistently explained by the same theory, this assumption can be considered as proven by experiment. A theoretical justification by evaluating the microscopic expressions for K and M_2, however, still remains to be done.

ACKNOWLEDGMENTS

I thank Professor D. Fredkin and Mr. E. Gullikson for helpful discussions. I am also indebted to the latter for making available his experimental results prior to publication and for critical reading of this manuscript. The work was supported by the Deutsche Forschungsgemeinschaft.

---

*On leave from the Universität Regensburg, Fachbereich Physik, D-8400 Regensburg, West Germany. Requests for reprints should be sent to this address.

[1]J. Owens, J. E. Browne, V. Arp, and A. F. Kip, J. Phys. Chem. 2, 85 (1957).
[2]D. Griffiths, Proc. R. Soc. London 90, 707 (1967).
[3]K. Okuda and M. Date, J. Phys. Soc. Jpn. 27, 839 (1969).
[4]F. W. Klainhaus, J. P. Long, and P. E. Wigen, Phys. Rev. B 11, 2638 (1975).
[5]P. Monod and Y. Berthier, J. Magn. Magn. Mater. 15-18, 149 (1980).
[6]S. Schultz, E. M. Gullikson, D. R. Fredkin, and M. Tovar, Phys. Rev. Lett. 45, 1508 (1980).
[7]K. W. Becker, preceding paper, Phys. Rev. B 26, 2394 (1982).
[8]M. B. Salamon and R. M. Herman, Phys. Rev. Lett. 41, 1506 (1978).
[9]M. B. Salamon, Solid State Commun. 31, 781 (1979).
[10]M. B. Salamon, J. Magn. Magn. Mater. 15-18, 147 (1980).
[11]E. D. Dahlberg, M. Hardiman, R. Orbach, and J. Souletie, Phys. Rev. Lett. 42, 401 (1979).
[12]J. Elliott, M. Hardiman, R. Chamberlin, and R. Orbach (unpublished).
[13]R. V. Chamberlin, M. Hardiman, and R. Orbach, J. Appl. Phys. 52, 1771 (1981).
[14]E. Gullikson (private communication) and (unpublished).