EPR linewidth ($T \gtrsim T_g$) in amorphous transition-metal—metalloid spin glasses: Theory

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A microscopic theory of the paramagnetic linewidth and line shift for an amorphous transition-metal—metalloid alloy spin glass has been given in the Mori-Kawasaki formalism. The linewidth has been calculated as being due to random single-ion anisotropy and the magnetic dipole-dipole anisotropic interaction, which are considered to be small perturbations. The contribution of the single-ion anisotropy has been found to be smaller. While it is noted that for $T > T_g$ there is no static component of the random dipolar magnetic field the dynamic component of this field does exist. Taking the point of view of Souletie and Tholence [Phys. Rev. B 32, 516 (1985)] that the autocorrelation time of the dynamic component increases as the temperature is lowered to T_g and attains the saturation value of $2\pi/w$ at $T = T_g(w)$, we predict that at this temperature the EPR response should be an inhomogeneously broadened Voigt line shape. The very limited data available have been compared with a rough numerical estimate of the linewidth at $T_g(w)$ and reasons have been suggested for the lack of good agreement. At higher temperature the dynamic component will be partially averaged out by the EPR probe and the linewidth is expected to be less and the line shape more Lorentzian. It has been found that the critical part of the linewidth and line shift for all frequencies should obey dynamic scaling down to and including $T_g(w)$. Also their temperature and frequency dependence shows trends similar to those for the canonical spin glasses. It is noted that more detailed data on the line shape close to $T_g(w)$ and accurate determination of the single-ion anisotropy constant are needed for a check on the above predictions. A justification has been given for the applicability of the Mori-Kawasaki formalism at least for the higher frequencies. The regime of applicability of the theory is expected to be about $T \gtrsim 1.25T_g$.

I. INTRODUCTION

A formalism developed by Mori and Kawasaki[1] (MK) has been adapted by Levy *et al.*[2] to explain the position and width of EPR lines of crystalline transition-metal spin glasses (SG). Such SG are believed to acquire magnetic anisotropy due to the Dzyaloshinski-Moriya (DM) interaction on experimental grounds. Huber[3] has used this theory with the input of neutron spin-echo data of spin-correlation functions to analyze the recent experiments on Ag:Mn SG.[4,5] He has found that the critical part of the linewidth follows the dynamic scaling law down to but possibly not exactly at T_g. He has also pointed out that this is possible only because of the insensitivity of the spin-correlation function to the applied magnetic field. The important point to note is that it was uncertain if scaling would be obeyed at T_g from the point of view of his theory. The experimental finding was that the linewidth did not diverge at T_g.

The detailed experimental work of Uemura *et al.*[6] has shown, *inter alia*, that for $T > T_g$ the depolarization of muon spins in Cu:Mn SG, which is due to dynamic random dipolar fields does not change due to an applied magnetic field up to 640 G. This is an important finding in relation to our work. Another important experimental observation was that the spin-autocorrelation time did not diverge but only saturated at T_g. Further, as has been pointed out by Souletie and Tholence,[7] if one accepts that the SG transition is a phase transition then it is natural to expect that the usual power-law variation will be valid down to T_g. They have pointed out how this is consistent with the finite value of the spin-autocorrelation time, τ, at T_g because of the frequency dependence of T_g. Thus, in effect, they proposed,

$$\tau = \tau_0 \left[\frac{T}{T - T_g(0)} \right]^{z\nu} \tag{1}$$

with $\tau = 2\pi/w$ at $T = T_g(w)$. By analysis of known data they have obtained τ_0 and $z\nu$ for various SG. ν and z are the usual critical exponents. Anticipating our results, we shall see in the following that near $T_g(w)$ the linewidth scales as τ and so the ideas of Refs. 6 and 7 justify the opinion that in spite of the saturation of linewidth, dynamic scaling law may be considered to be valid down to and including $T_g(w)$.

The purpose of this work is to investigate the EPR linewidth and line-shift variation with temperature in the critical regime for amorphous transition-metal—metalloid (TM-*M*) SG and to theoretically demonstrate, if possible, dynamic scaling in this case. Dynamic scaling, as in the canonical SG, is expected as there is no basic difference from the point of view of spin dynamics except in the range of interaction and the crystal structure. What is more, any uncertainty regarding the validity of scaling is now removed. For macroscopically homogeneous material one does not expect a different universality class for the amorphous SG (Ref. 8) but both short-range interaction and the presence of random local fields (which we shall discuss later) will reduce the effective dimensionality (Ref.

9). This will change the critical exponents which we have not tried to calculate here. In Sec. II we have calculated the linewidth and line shift in terms of τ, among others. In Sec. III there is a numerical estimate of the theoretical expression for the linewidth. Section IV contains a discussion of the methodology and results of the calculation and, in particular, comments on the effect of the random dipolar fields. Also, in this section we have compared the numerical estimates with experimental linewidth data and have justified the use of the MK formalism used in Sec. II. There is a short summary in Sec. V.

II. CALCULATIONS

As usual, we consider the relaxation of the macroscopic transverse magnetization vector to be due to anisotropic magnetic interaction. This anisotropy has to be a small perturbation on the Hamiltonian made up of the exchange and Zeeman terms. In the case of amorphous materials DM anisotropy is ruled out.[10] The magnetic dipole-dipole interaction is always present in magnetic systems. A candidate typically suited for the amorphous SG is the local random single-ion magnetocrystalline anisotropy.[11] In the case of TM-M alloys this anisotropy is of the order of 10^{-24} J/atom (Ref. 12) and is small enough to be considered a perturbation. The applied field may also contribute to the relaxation. However, if we assume that the result of Ref. 6 mentioned before is true for substantially higher fields than had been applied actually, i.e., about 3–4 kG, we can ignore this possibility. H can cause relaxation directly also in addition to via τ. This possibility we exclude at this time though the calculations can be done without great difficulty.

In this calculation we set N, V, $g\mu_B$, and $\hbar$ all equal to unity. We take,

$$H = H_0 + H_{\text{aniso}} , \tag{2}$$

where

$$H_0 = -\sum_{\substack{i,j \\ j>i}} J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j - h_{\text{ext}}\sum_i S_i^z$$

and

$$H_{\text{aniso}} = -K_2\sum_i(\mathbf{S}_i\cdot\hat{\mathbf{n}}_i)^2 - 3\sum_{\substack{i,j \\ j>i}}(\mathbf{r}_{ij}\cdot\mathbf{S}_i)(\mathbf{r}_{ij}\cdot\mathbf{S}_j)r_{ij}^{-5} . \tag{3}$$

The first term on the right is the single-ion anisotropic term and the second is the anisotropic part of magnetic dipole-dipole interaction. $\hat{\mathbf{n}}_i$ is the direction of the local easy axis at site i and K_2 is the appropriate anisotropy constant. The single-ion term is suitable for amorphous SG and has been found to give good results in realistic cases.[13] We consider the dynamics of the total transverse magnetization vector,

$$\dot{S}_T^{\pm}(t) = i[H_{\text{aniso}}, S_T^{+}(t)] ,$$

where $S_T^{+} = \sum_i S_i^{+}$ and $S^{+} = S^x + iS^y$ etc. Using the usual commutation relations,

$$\dot{S}_T^{\pm} = -iK_2\sum_i\{2(S_i^{+})^2\alpha_i\gamma_i - [S_i^{+},S_i^{-}]_{+}\beta_i\gamma_i - [S_i^{+},S_i^{z}]_{+}\gamma_i^2 + [S_i^{z},S_i^{+}]_{+}\alpha_i\beta_i + [S_i^{z},S_i^{-}]_{+}\beta_i^2 + 2(S_i^{z})^2\beta_i\gamma_i\} - 3\sum_{\substack{i,j \\ j>i}}(\mathbf{r}_{ij}\times\mathbf{S}_i)^{+}(\mathbf{r}_{ij}\cdot\mathbf{S}_j)r_{ij}^{-5} . \tag{4}$$

The last term on the right had been obtained in this form by Huber.[14] Here $\alpha_i = (u_i - iv_i)/2$, $\beta_i = (u_i + iv_i)/2$, and $\gamma_i = w_i$ where u_i, v_i, and w_i are the direction cosines of $\hat{\mathbf{n}}_i$. $[\ldots,\ldots]_{+}$ indicates an anticommutator. Here we have to average the Kubo-Mori inner product[1] $(\dot{S}_T^{\pm}(t)\,|\,\dot{S}_T^{\mp})$ over the random directions $\hat{\mathbf{n}}_i$. In this connection one notes the following nonzero values obtained by this averaging,

$$\langle\,|\alpha_i\gamma_i|^2\rangle = \langle\,|\beta_i\gamma_i|^2\rangle = \tfrac{2}{5} ,$$

$$\langle\,|\alpha_i\beta_i|^2\rangle = \langle\,|\beta_i|^4\rangle = \tfrac{2}{3} ,$$

and

$$\langle\gamma_i^4\rangle = \tfrac{1}{5} .$$

(Unlike the preceding equations, in the following the angular bracket indicates average over canonical ensembles.) All the other averages over $\hat{\mathbf{n}}_i$ vanish. After some algebra one gets the averaged single-ion term in the Kubo-Mori inner product,

$$K_2^2\sum_{\substack{i,j \\ j>i}}([S_i^{+}(t),S_i^{z}(t)]_{+}\,|\,[S_j^{-}(t),S_j^{z}(t)]_{+}) . \tag{5}$$

This expression has four terms each of which, by definition, has the form

$$(A(t)\,|\,B) = \int_0^{\beta} d\lambda\langle e^{\lambda H}A(t)e^{-\lambda H}B\rangle \simeq \beta\langle A(t)B\rangle , \tag{6}$$

with $\beta^{-1} = k_B T$. The last form is the high-temperature approximation. Thus each of the four terms is a four-spin correlation function. One decomposes them in the following manner:[15]

$$\langle S_i^{+}(t)S_i^{z}(t)S_j^{-}S_j^{z}\rangle \approx \langle S_i^{+}(t)S_j^{-}\rangle\langle S_i^{z}(t)S_j^{z}\rangle + \langle S_i^{+}(t)S_j^{z}\rangle\langle S_i^{z}(t)S_j^{-}\rangle . \tag{7}$$

All the four terms give the same result on decomposition. The second term on the right side of Eq. (7) disappears if we assume the total z component of magnetization to be a constant of motion as is usual in magnetic resonance. Using Eq. (6) the expression in (5) then becomes,

$$4K_2^2\beta\sum_{\substack{i,j \\ j>i}}\langle S_i^{+}(t)S_j^{-}\rangle\langle S_i^{z}(t)S_j^{z}\rangle . \tag{8}$$

Using transverse macroscopic symmetry,

$$\langle S_i^{+}(t)S_j^{-}\rangle = 2\langle S_i^{x}(t)S_j^{x}\rangle . \tag{9}$$

Ignoring spatial correlation for $T > T_g$ the expression in (8) then becomes,

$$8K_2^2\beta\sum_i(\langle S_i^x(t)S_i^x\rangle)^2 . \tag{10}$$

Here we have assumed that autocorrelation in the z direction is independent of h_{ext} and is equal to those in the transverse direction.

We now calculate the dipole-dipole anisotropic interaction term in the Kubo-Mori inner product. This term is given by,

$$\left[-3\sum_{\substack{i,j\\ j>i}}(\mathbf{r}_{ij}\times\mathbf{S}_i)^+(\mathbf{r}_{ij}\cdot\mathbf{S}_j)r_{ij}^{-5}\,\middle|\,-3\sum_{\substack{i,j\\ j>i}}(\mathbf{r}_{ij}\times\mathbf{S}_i)^-(\mathbf{r}_{ij}\cdot\mathbf{S}_j)r_{ij}^{-5}\right] . \tag{11}$$

After some algebra involving detailed expansion of the expression and using Eqs. (6)—(9) above this becomes,

$$9\beta\sum_{\substack{i,j\\ j>i}}(\mathbf{r}_{ij}^{-6}+z_{ij}^2r_{ij}^{-8})\langle S_i^x(t)S_i^x\rangle\langle S_j^x(t)S_j^x\rangle .$$

Because of the random location of the magnetic atoms $z_{ij}^2=r_{ij}^2/3$ on averaging. Also, as pointed out in a similar case in Ref. 2 the above summations over the "lattice" and the spin correlations can be performed separately as an acceptable approximation in this kind of perturbational calculations. Thus the magnetic dipole-dipole contribution to the Kubo-Mori inner product is

$$12\beta\sum_j r_j^{-6}\sum_i(\langle S_i^x(t)s_i^x\rangle)^2 . \tag{12}$$

It can be easily seen that the cross terms in the inner product disappear on averaging over $\hat{\mathbf{n}}_i$. One notices that in expressions (10) and (12) the quantity under site summation is the same.

It is known that the spin-autocorrelation (or relaxation) time has a hierarchy of values. We, however, assume a single effective correlation time τ and set,

$$\langle S_i^x(t)S_i^x\rangle=\langle(S_i^x)^2\rangle e^{-t/\tau} . \tag{13}$$

Again, ignoring spatial correlation,

$$\sum_i(\langle S_i^x(t)S_i^x\rangle)^2=\left[\sum_i\langle(S_i^x)^2\rangle\right]^2e^{-2t/\tau} . \tag{14}$$

From Eq. (6) and transverse symmetry,

$$(S_T^+\,|\,S_T^-)=2\beta\sum_i\langle(S_i^x)^2\rangle . \tag{15}$$

Now, in the first approximation in the MK formalism the self-energy is[1]

$$\Gamma(w,T)=\int_0^\infty dt\,e^{-iwt}(\dot{S}_T^+(t)\,|\,\dot{S}_T^-)/(S_t^+\,|\,S_T^-) , \tag{16}$$

where w is the resonance frequency. From Eqs. (10) and (12)—(16),

$$\Gamma(w,T)=\left[4K_2^2+6\sum_j r_j^{-6}\right]\sum_i\langle(S_i^x)^2\rangle\int_0^\infty dt\,e^{-iwt-2t/\tau} ,$$

which on integration gives,

$$\mathrm{Re}\Gamma(w,T)=\tfrac{4}{3}(2K_2^2+3\sum_j r_j^{-6})S(S+1)\tau/(4+w^2\tau^2) , \tag{17a}$$

$$\mathrm{Im}\Gamma(w,T)=-\tfrac{2}{3}\left[2K_2^2+3\sum_j r_j^{-6}\right]S(S+1)w\tau^2/(4+w^2\tau^2). \tag{17b}$$

Eqs. (17a) and (17b) give, respectively, the linewidth and line shift. We notice that both obey the dynamical scaling hypothesis in the whole critical regime for all frequencies as τ is expected to do so down to $T_g(w)$. The particular form of Eq. (17) arises because of the form assumed in Eq. (13) which is an approximation.

For $T\gg T_g$ it is expected from Eq. (1) that $w\tau\ll 1$ and Eq. (17) becomes

$$\mathrm{Re}\Gamma(w,T)\simeq\left[\tfrac{2}{3}K_2^2+\sum_j r_j^{-6}\right]S(S+1)\tau , \tag{18a}$$

and

$$\mathrm{Im}\Gamma(w,T)\simeq-\left[\tfrac{1}{3}K_2^2+\tfrac{1}{2}\sum_j r_j^{-6}\right]S(S+1)w\tau^2 . \tag{18b}$$

For $T\gtrsim T_g$ from Eq. (1) $w\tau\approx 2\pi$ and Eq. (17) becomes

$$\mathrm{Re}\Gamma(w,T)\simeq\left[0.061K_2^2+0.092\sum_j r_j^{-6}\right]S(S+1)\tau , \tag{19a}$$

and

$$\mathrm{Im}\Gamma(w,T)\simeq-\left[0.188K_2^2+0.29\sum_j r_j^{-6}\right]S(S+1)\tau . \tag{19b}$$

At $T=T_g(w)$ from Eq. (1) $w\tau=2\pi$ and if we assume Eq. (17) still valid, we have

$$\mathrm{Re}\Gamma(w,T)\simeq\left[0.38K_2^2+0.58\sum_j r_j^{-6}\right]S(S+1)/w , \tag{20a}$$

and

$$\mathrm{Im}\Gamma(w,T)\simeq-\left[1.18K_2^2+1.82\sum_j r_j^{-6}\right]S(S+1)/w . \tag{20b}$$

Thus at $T=T_g(w)$ both linewidth and line shift vary inversely as w. Also, from Eq. (17) for $w=0$ in all relevant temperature regimes,

$$\mathrm{Re}\Gamma(w,T)=\left[\tfrac{2}{3}K_2^2+\sum_j r_j^{-6}\right]S(S+1)\tau , \tag{21a}$$

and

$$\mathrm{Im}\Gamma(w,T)=0 . \tag{21b}$$

From Eq. (17b) it follows that there is a minimum in the line shift versus w curve. Equations (20a) and (20b) indicate frequency-dependent saturation of linewidth and

line shift. These characteristics have been already found in canonical SG (Refs. 4 and 5) and are now expected in TM-M alloy SG also.

III. NUMERICAL ESTIMATES

It is in order to make an estimate of the numerical values of Γ as given by Eq. (17) and also the relative importance of the single-ion and dipole-dipole contributions. Due to our initial choice of units, the single-ion term of Eq. (17) has to be divided by $g\mu_B\hbar = 1.95\times10^{-57}$ J^2 s T^{-1} and the dipole-dipole term multiplied by $(\mu_0/4\pi)^2(g\mu_B)^3\hbar^{-1} = 6.07\times10^{-49}$ J^2 T^{-3} s^{-1}. Because of the small $l\cdot s$ coupling expected in this case we ignored the pseudodipolar interaction in comparison with the pure dipole-dipole interaction; hence g has been set equal to 2.

K_2 can be estimated experimentally by measuring the approach to saturation of the magnetization.[16] Fähnle and Egami[17] have estimated K_2 for $Fe_{40}Ni_{40}B_{20}$ to be of the order of 10^{-24} J/atom. Other TM-M amorphous alloys are also expected to have K_2 of the same order but it is necessary to perform individual measurements.

We have next to perform the "lattice" sum $\sum_j r_j^{-6}$. To be specific, let us take the typical TM-M SG alloy $Fe_{10}Ni_{70}P_{20}$. Direct calculation of the sum does not seem possible as neither its density nor its structure is known. We can, however, make a rough estimate on the basis of general ideas about the amorphous structure.[18] The short-range structure of amorphous materials is icosahedral with the number of nearest neighbors of increasing order 12,20,24,... (Ref. 19). So the average number of nearest neighbors (NN) and next nearest neighbors (NNN) of the same type for an Fe atom in our typical material is ~1.2 and 2.0, respectively. The TM-TM NN distance found by x-ray scattering in $Fe_{75}B_{25}$ and $Fe_{75}Si_{15}B_{20}$ is ~2.4 Å. This distance should be more with larger metalloid atoms and their higher concentration. Notice, however, that the difference in regard to this distance between these two materials is almost negligible even though $r_{Si} = 1.10$ Å and $r_B = 0.91$ Å. The concentration dependence also works up to a point. Noting $r_P \approx r_{Si}$ we might have concluded that TM-TM NN distance in $Fe_{10}Ni_{70}P_{20}$ would be slightly less than that in $Fe_{75}Si_{15}B_{10}$. By the same token we might conclude similarly for $Fe_{40}Ni_{40}P_{14}B_6$. But the relevant distance for this material is ~2.5 Å rather than something less than 2.4 Å. A safe conclusion would be to take the TM-TM NN distance in our material also to be ~2.4 Å.

In some of these amorphous materials because of the TM-TM-TM collinear configuration the TM-TM NNN distance is about twice the NN distance. We do not know if there is a collinear configuration in this case. By ignoring the NNN term in the "lattice" sum we change the latter by only $\sim2.5\%$. Hence we calculate here by assuming such a configuration. Thus the "lattice" sum is,

$$\sum_j r_j^{-6} \approx 10^{60}(1.2\times2.4^{-6}+2\times4.8^{-6}+\cdots)$$
$$\approx 6.4\times10^{57} \text{ mks} . \quad (22)$$

The alloys under consideration are intermetallic compounds and S, the total angular momentum quantum number of the magnetic species is determined by the band structure. It is known that in these materials the magnetic moment of Fe, Co, and Ni atoms are, respectively, 2, 1, and 0 Bohr magnetons.[12] Taking $g=2$, $2[S(S+1)]^{1/2}=2$ and 1 for Fe and Co, respectively. Thus these materials behave like Heisenberg magnets with nonintegral spins. For Fe we find $S=0.618$.

We have not found the temperature variation of τ here. Using linearized Glauber dynamics for the well-known Sherrington-Kirkpatrick (SK) model in the case of an Ising system a power-law variation for τ was derived with $z\nu=2$.[20] This approach cannot be carried through mathematically at this time for the materials in question, which are believed to have short-ranged interaction, ferromagnetic with NN and antiferromagnetic with the NNN.[12] In addition, we have Heisenberg systems here. Because of the topological disorder of the amorphous structure these interactions will have a random component also. It has been shown that short-range materials cannot have an SG transition and additional symmetry-breaking interaction is needed to effect such a transition.[21,22] No way seems to have been found yet to take account of these terms and the short-range interaction while finding $z\nu$. Using an elaborate dynamical approach, Zippelius[23] has found for an Ising system with only nearest-neighbor interaction with a Gaussian distribution about zero mean that $z\nu=1$ for mean-field theory. When this calculation is extended by renormalization-group analysis an ambiguous value is found for z.

It thus appears that at this time experimental methods, e.g., that using Eqs. (17)—(21) or that of Ref. 6 would be helpful in determining $z\nu$ and τ_0 for real systems.

Thus while at higher temperatures there is the problem of lack of knowledge of τ, at $T=T_g(w)$ taking the cue from Ref. 7 we set $w\tau=2\pi$. The linewidths at two frequencies at $T=T_g(w)$ evaluated from Eq. (20) are the following.

(i) For 9.3 GHz the single-ion contribution is 33.8 G and the dipole-dipole contribution is 390 G; (ii) for 1.1 GHz the two respective contributions are 286 and 3260 G. It is thus found that the dipole-dipole contribution is quite significant here.

IV. DISCUSSION

The question whether Eq. (1) holds down to and including $T=T_g$ has been debated as experimentally τ has been found to saturate instead of diverging. Since phase transition at SG transition and dynamic scaling hypothesis are now generally accepted, and T_g is known experimentally to be a function of w, the viewpoint of Ref. 7 seems reasonable. If, as expected, Eqs. (17)—(21) are found applicable to a real amorphous TM-M SG, EPR measurements can also help in the confirmation or otherwise of this view. In this connection it may be of interest to note that in the course of EPR experiments one can easily determine $T_g(w)$.[24] If one knows $T_g(0)$ this gives a way of finding τ_0 and $z\nu$.

We have implied that Eq. (17) could be used down to $T_g(w)$. We have restricted ourselves to the first order in the MK perturbation approach. The use of four-spin

decomposition and other approximations is estimated[2] to define the regime of validity of this kind of theory to about $T \gtrsim 1.25T_g$. This decomposition is equivalent to a mean-field approximation which underestimates the fluctuations. Changes in fluctuation will only change the critical exponents but, as already mentioned, we have not calculated $z\nu$ here. One could carry forward the MK perturbation to higher order also. Though we have not done the calculation we have doubts if Eq. (17) with a suitable $z\nu$ will be valid down to $T_g(w)$.

The question of the effect of dipolar fields on the linewidth and whether in view of the likely strong dynamic random fields the MK formalism is at all applicable is important. We note that the random fields can affect the line shape in three different ways. First, if there is a static component of this field it will increase the linewidth by inhomogeneous broadening. For $T > T_g$ this component of the random dipolar fields is absent.[6] Secondly, the dynamic component of these fields which exists above T_g may require the introduction of the Kubo-Toyabe (KT) approach.[25] Again from Ref. 7 we notice that the characteristic time τ of the dynamic component satisfies $\tau < (w/2\pi)^{-1}$ for known cases at $T > T_g(w)$. Thus in this temperature regime the effect of the dynamic component will be at least partially averaged out by the EPR probe. So a typical KT approach may not be essential. Only when $T = T_g(w)$, $\tau = (w/2\pi)^{-1}$, and the full amplitude of the dynamic field will be sampled by the probe. In this regime, therefore, we should see the inhomogeneous broadening of the EPR line. Such a line has the Voigt line shape[26] which is neither Lorentzian nor Gaussian. This is true for all SG whatever the type. Thirdly, the anisotropic part of the dipole-dipole interaction will influence the relaxation and hence the line shape.

The only relevant published work on the EPR spectroscopy of these materials concerns $Fe_8Ni_{72}P_{20}$ and $Fe_{10}Ni_{70}P_{20}$ (Ref. 25) and covers the temperature regimes both above and below T_g. These data have been analyzed by a different approach and the published data are not detailed enough for comparison with the theory presented above. The comments of the authors that at $T = T_g$ the lines are neither Lorentzian nor Gaussian is of interest. It would be desirable to verify if the line shape fits a Voigt function with appropriate inhomogeneous broadening. Above $T_g(w)$ the amplitude of the dynamic random dipolar field at any magnetic atom equals the full amplitude of the magnetic field at the given atom due to the other magnetic atoms. This is what Uemura *et al.*[6] have indicated by $a_d = a_0$ for $T > T_g$. Noting $S(S+1) = 1$ here and by directional averaging of the polar part of the dipolar field, $g\mu_B \sin\theta / r^3$, which leads to the prefactor $\pi/4$, the full amplitude of the dipolar field, a_0, is given by

$$a_0 = \frac{\pi}{4} g\mu_B \sum_j r_j^{-3} (\mu_0/4\pi)\ \mathrm{T}\ . \tag{23}$$

The radial part vanishes on directional averaging. In the same manner as in Eq. (22) we find the "lattice" sum in Eq. (23) $\approx 1.05 \times 10^{29}$ mks which leads to the width corresponding to the amplitude, a_d, about $\frac{2}{3} a_0 \approx 1020$ G. A dynamic field of this width will combine with the Lorentzian width as calculated in Sec. III. The only temperature at which we can make a comparison is $T = T_g(w)$, where we can calculate the Lorentzian width by using Eq. (20). For higher temperatures comparison is not possible as τ is not known in this case.

The authors of Ref. 25 do not take account of the change of T_g with w. However, from Figs. 1 and 2 of Ref. 25 we extract at $T = T_g$ the widths of the lines at both 9.3 and 1.1 GHz to be ~ 2.5 kG. We have not done the appropriate "folding" of the Gaussian width of 1020 G with the Lorentzian width. However, the width of the Voigt line is approximately the sum of the two widths. Thus we find for 9.3 and 1.1 GHz the respective linewidths ~ 1450 and ~ 4570 G. Subtracting a Korringa broadening of about 10% from the experimental linewidth we find no agreement with the theory. In particular, at the higher frequency we are much lower than the mark and vice versa for the other case.

Some comments are in order in this connection. First, as already mentioned, Eq. (20) from which the relaxational part of the width has been calculated is of doubtful validity. Most likely the inverse proportionality to w is not correct and the w dependence is weaker. This seems to us the most important reason for the lack of agreement. Secondly, the fact that the experimental width is almost independent of the frequency indicates that it is the dynamic component of the dipole field which dominates the width at $T = T_g(w)$. This field is independent of frequency. Further, our calculation of the "lattice" sum is rather rough because of lack of knowledge about the structure. We have used only the estimated order of magnitude of K_2 for calculating the single-ion contribution. In fact, this contribution can be substantially different from the calculated value. Lastly, the experimental linewidth ~ 2.5 kG pertains to a temperature $T < T_g(w)$ because of the expected increase of T_g with w. If one were able to keep to the exact $T_g(w)$ the linewidth would be less as seen from Ref. 25 itself.

A comparison in the regime of validity can be done only when more detailed data on linewidth and line shift are available. It would be desirable to measure τ also by some independent method, e.g., Ref. 6.

Regarding the condition for the cutoff at the first order of the MK formalism, namely,[1]

$$\mathrm{Re}\Gamma \ll w/\gamma \ll \tau_c^{-1}/\gamma, \quad \gamma \equiv g\mu_B \hbar\ . \tag{24}$$

(τ_c is the correlation time of the appropriate "random force.") We note that unlike the case of Brownian motion the "random forces" in MK formalism do not necessarily have any physical reality except that they are introduced to satisfy a mathematical requirement as has been noted in similar other cases.[27] In particular, there is a hierarchy of "random forces" and one can choose τ_c of the appropriate time scale. We find that the relaxational width ~ 424 G at 9.3 GHz easily satisfies the condition (24) while the same ~ 3550 G at 1.1 GHz does not. In any event, this theory is of doubtful applicability at $T = T_g(w)$ and the linewidths known to be smaller in the regime of applicability are not available. Our conclusion is that this theory should work for $T \gtrsim 1.25T_g$ for frequency ~ 9 GHz and higher and possibly also for ~ 1 GHz.

V. SUMMARY

In summary, we have tried to present a simple microscopic theory for EPR linewidth and line shift for amorphous TM-M SG alloys applicable in the regime $T \gtrsim 1.25T_g$. We are not able to take account of the characteristic exchange interaction for SG. But we have used the anisotropic energy most suitable for an amorphous SG and have included the magnetic dipole-dipole interaction. We have predicted that at $T = T_g(w)$, the line shape is likely to be a Voigt function which is due to inhomogeneous broadening by the dynamic random dipolar field. The general trend of frequency and temperature dependence is the same as that for canonical SG and the critical part of the linewidth and line shift should obey dynamical scaling down to $T_g(w)$ for all frequencies. Detailed experimental data are not available at this time for comparison with the theoretical prediction. Further details about the predicted inhomogeneous broadening are given in the Appendix.

APPENDIX

Because of approximations Eq. (20) of the text may not be accurate. But whatever the width, in this theory we expect a Voigt line shape at $T = T_g(w)$. An individual EPR line has an elementary characteristic line shape which is either Gaussian or Lorentzian. This presupposes that all spins responsible for the line are in identical situations. However, in the case considered in the text, at $T = T_g(w)$ different groups of spins at a given instant of time will sample, in addition to the fixed external field, different values of the dynamic random dipolar field. At every spatial location this field is expected to have a Gaussian distribution in time, and for $T > T_g$ we do not expect any spatial correlation. Thus the total Gaussian distribution of the dynamic random dipolar field will be sampled by the spin system. So, to find the resultant line shape one has to "fold" the elementary line with an appropriate function representing the variation of the total field which has a Gaussian-like distributed component now. As the elementary line shape is considered to be Lorentzian in this case the "folding" will produce the Voigt line shape. The absorption amplitude of this line shape is given by[26]

$$Y(v,b) = \frac{b}{\pi} \int_{-\infty}^{\infty} \frac{e^{-x^2} dx}{b^2 + (v - x)^2} ,$$

where $b = \sqrt{\ln 2}\,{}^L\Delta H_{1/2} / {}^G\Delta H_{1/2}$ and $v = 2\sqrt{\ln 2}(H - H_0)/{}^G\Delta H_{1/2}$. H_0 is the line center and ${}^G\Delta H_{1/2}$ and ${}^L\Delta H_{1/2}$ are the Gaussian and Lorentzian linewidths, respectively. The former is proportional to a_d and the latter is given by $\mathrm{Re}\Gamma$ of the text. If indeed the line shape is a Voigt function by a suitable fitting procedure one could disentangle the two contributions and make separate comparisons. Since the above integral is not integrable its values for various values of b and v have been tabulated. We hope to consider in the near future the details of the problem of obtaining the Y versus H curve for the inhomogeneously broadened line by using these tables.

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