Dynamic narrowing of inhomogeneously broadened EPR lines in metals

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The line parameters of the EPR on magnetic impurities in metals (i.e., the line shape, width, the signal shape, and their temperature dependence) are computed with allowance for the inhomogeneous broadening and the "electron bottleneck" effect, i.e., the strong dynamic coupling of the impurities to the conduction electrons. For this purpose the kinetic equations for the transverse components of the conduction-electron magnetization and the spectral density of the transverse components of the magnetization of the impurities producing the inhomogeneously broadened EPR line are derived. The system of equations is solved for the case of an arbitrary local-field distribution function $g(\Omega)$. The solution is analyzed analytically and numerically for specific $g(\Omega)$ functions. The conditions under which the electron bottleneck effect leads to the narrowing of the inhomogeneously broadened lines are found, and the residual inhomogeneous contribution to the total line width is computed. It is shown that a minimum appears on the temperature dependence of the EPR line width as a result of the disappearance of the dynamic narrowing as the temperature is lowered; the line has a significantly different shape in this temperature region. The experimental investigations are discussed.

INTRODUCTION

The temperature dependence of the line width $\Gamma(T)$ of the electron paramagnetic resonance (EPR) on magnetic impurities in metals is approximated by the linear function

$$\Gamma(T)=a+bT \tag{1}$$

and is well corroborated by measurements at high temperatures. The coefficient b is called the temperature slope of the line; a, the residual width. The dominant contribution to the latter is made by the inhomogeneous broadening.[1,2] There are, however, a large number of experiments in which the linear decrease of the width $\Gamma(T)$ at low temperatures is replaced by a nonlinear increase, i.e., the line width as a function of the temperature has a minimum (see, for example, Refs. 1–7). As the possible causes of the EPR line broadening at low temperatures, there have been discussed mechanisms connected with the appearance of magnetic order (presumably of the spin-glass type) in the system of impurities; the disappearance of the exchange narrowing of the dipole width of the line as a result of the critical slowing down of the exchange fluctuations of the local fields[8,9]; the nonmonotonic temperature dependence of the homogeneous relaxation contribution to the EPR line width, that stems from the presence of a maximum in the susceptibility of the impurities in the region of the magnetic transition.[7] It is, however, noteworthy that in the overwhelming majority of experiments in which the nonmonotonic behavior of $\Gamma(T)$ was observed (see, for example, Refs. 1–6), the more rigid the regime of "electron bottleneck" (the appearance of coupled oscillations of the spin magnetizations of the impurities and conduction electrons if the latter are bound more weakly to the lattice than to the impurities[10]) is, the sharper is the minimum. This indicates the existence of a connection between the bottleneck phenomenon and the nonmonotonicity of $\Gamma(T)$. It is well known[1,2,10] that the effect of the bottleneck on the homogeneous width of the EPR line is to reduce the temperature slope b in (1); therefore, it is necessary to investigate the dynamic narrowing of the inhomogeneous width as a possible cause of the nonmonotonicity of $\Gamma(T)$.

The present paper is devoted to the systematic investigation of the spin dynamics of impurities having an inhomogeneously broadened EPR line a metal. The equations describing the coupled oscillations of the spectral density of the magnetization of the impurities and the magnetization of the electrons are derived and solved for an arbitrary local-field distribution function. Analysis of the solution shows, in particular, that the gradual narrowing down, as the temperature increases, of the inhomogeneous contribution to the line width, together with the monotonic increase of the homogeneous width b^*T, leads to the appearance of a minimum in the dependence $\Gamma(T)$ even when the broadening mechanisms connected with the magnetic order are not considered. Under the experimentally realizable bottleneck conditions, the minimum width of $\Gamma(T)$ can be several times smaller than the width of the original local-field distribution. Since the resonance line width in a metal is a mixture of χ' and χ'' (where $\chi=\chi'+i\chi''$ is the dynamic susceptibility), knowledge of the formula for χ'' is necessary for the correct extraction of the width. Accordingly, we investigate the dependence of the EPR line shape on temperature and bottleneck conditions.

THE KINETIC EQUATIONS AND THE DYNAMIC SUSCEPTIBILITY

Let us consider a dilute magnetic alloy located in an external constant magnetic field $\mathbf{H}_0$ and a weak external variable magnetic field $\mathbf{h}(t)$ perpendicular to $\mathbf{H}_0$. Let us assume that the magnetic impurities are distributed randomly at the lattice sites j of the metal with concentration c and local precession frequency Ω_j. The impurities and conduction electrons interact with each other through the s–f exchange $\mathscr{H}_{es}$; furthermore, the electrons interact with the

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lattice through the momentum and spin-orbit scattering $\mathcal{H}_{eL}$ by the nonmagnetic impurities, defects, etc. The total Hamiltonian of the system is written in the standard—for EPR in metals—form (see, for example, Refs. 10 and 11), except for the terms connected with the magnetic impurities:

$$\mathcal{H}(t)=\mathcal{H}_0{}^s+\mathcal{H}_0{}^e+\mathcal{H}_k+\mathcal{H}_1{}^s(t)+\mathcal{H}_1{}^e(t)+\mathcal{H}_{es}+\mathcal{H}_{eL},$$

$$\mathcal{H}_0{}^s=\hbar\int S^z(\Omega)\,(\omega_s+\Omega)\,d\Omega,$$

$$\mathcal{H}_1{}^s(t)=-\frac{g_s\mu_B h}{2}\int[S^+(\Omega)\exp(-i\omega t)+S^-(\Omega)\exp(i\omega t)]\,d\Omega, \tag{2}$$

$$\mathcal{H}_{es}=-J_{sf}\int\sum_j \mathbf{S}_j\boldsymbol{\sigma}(\mathbf{r}_j)\,\delta(\Omega-\Omega_j)\,d\Omega,$$

$$S^\alpha(\Omega)=\sum_j S_j{}^\alpha\delta(\Omega-\Omega_j),\quad \hbar\Omega_j=-g_s\mu_B H_j,$$

$$\hbar\omega_s=-g_s\mu_B H_0,\quad \hbar\omega_e=-g_e\mu_B H_0,$$

$\mathcal{H}_0^{s(e)}$ and $\mathcal{H}_1^{s(e)}(t)$ are the Zeeman-energy operators for the impurities (electrons), $\mathcal{H}_k$ is the Hamiltonian for the electron kinetic energy,

$$\boldsymbol{\sigma}(\mathbf{r}_j)=\frac{1}{N}\sum_{\mathbf{q}}\exp(-i\mathbf{q}\mathbf{r}_j)\boldsymbol{\sigma}_{\mathbf{q}}$$

is the electron spin density at the jth lattice site, $\mathbf{r}_j$ is the radius vector of the site, $\boldsymbol{\sigma}_{\mathbf{q}}$ is the Fourier transform of the spin density, S_j^α is the α component of the spin angular momentum of the impurity at the site j, J_{sf} is the exchange integral, N is the number of lattice sites per unit volume, $g_s(g_e)$ and μ_B are the impurity (electron) g factor and the Bohr magneton, h and ω are the amplitude and frequency of the variable field $\mathbf{h}(t)$, and H_j is the local magnetic field at the jth site.

As can be seen from (2), the system is both spatially and spectrally inhomogeneous. It is well known[12] that bounded motion occurs under bottleneck conditions over distances of the order of $\delta=(D_eT_e)^{1/2}\sim10^{-5}$ cm, where D_e and T_e are respectively the diffusion coefficient and the spin-lattice relaxation time for the electrons. The length δ is significantly greater than the spatial scale of the inhomogeneities in the distribution of the local fields, the principal sources of which are the straggling of the parameters of the crystalline field, the unresolved fine and hyperfine structures, the dipole field, etc. Therefore, we can first carry out the averaging over the space variables. After the averaging, the system remains spectrally inhomogeneous, with a local-field distribution function $g(\Omega)$. The spins having the same frequency are most strongly coupled to each other in the bottleneck regime, and therefore kinetic equations can be derived for the spectral density of the homogeneous (in space) magnetization of the impurities. Because the electrons move rapidly (the Fermi velocity $v_F\sim10^8$ cm/sec), the local fields acting on them average out to negligible values. All this allows to us choose as the dynamic variables for the description of the spin dynamics the spectral-density operators for the impurity and electron magnetizations, $\mathbf{M}_s(\Omega)$ and $\mathbf{M}_e$, respectively, and the electron kinetic energy operator $\mathcal{H}_k$. The kinetic equations are derived by averaging the quantum-mechanical equations of motion for the corresponding operators with a nonequilibrium statistical operator. We omit the corresponding computations, since the literature contains a detailed description of the computational procedure (see, for example, Refs. 11, 13,–15). In second order in $\mathcal{H}_{eL}$ and $\mathcal{H}_{es}$ the equations for the transverse components of the mean spectral density of the magnetization of the impurities and the transverse components of the mean magnetization of the electrons have the following form:

$$\frac{dM_s{}^\pm(\Omega,t)}{dt}=\pm i\{\Omega M_s{}^\pm(\Omega,t)+\omega_s(1+\lambda\chi_e)\nu[M_s{}^\pm(\Omega,t)$$

$$-\chi_s(h^\pm(t)+\lambda M_e{}^\pm(t))\,g(\Omega)]\}$$

$$-\int_{-\infty}^{0}dt_1[M_s{}^\pm(\Omega,t+t_1)-\chi_s(h^\pm(t+t_1)$$

$$+\lambda M_e{}^\pm(t+t_1))\,g(\Omega)]\Sigma_{se}{}^\pm(t_1)$$

$$+\varkappa\int_{-\infty}^{0}dt_1[M_e{}^\pm(t+t_1)-\chi_e(h^\pm(t+t_1)$$

$$+\lambda M_s{}^\pm(t+t_1))]\,g(\Omega)\Sigma_{es}{}^\pm(t_1); \tag{3}$$

$$\frac{dM_e{}^\pm(t)}{dt}=\pm i\omega_e(1+\lambda\chi_s)\nu[M_e{}^\pm(t)-\chi_e(h^\pm(t)+\lambda M_s{}^\pm(t))]$$

$$-\int_{-\infty}^{0}dt_1[M_e{}^\pm(t+t_1)-\chi_e(h^\pm(t+t_1)+\lambda M_s{}^\pm(t+t_1))]$$

$$\times(\Sigma_{es}{}^\pm(t_1)+\Sigma_{eL}{}^\pm(t_1))$$

$$+\varkappa^{-1}\int_{-\infty}^{0}dt_1[M_s{}^\pm(t+t_1)-\chi_s(h^\pm(t+t_1)+\lambda M_e{}^\pm(t+t_1))]\Sigma_{se}{}^\pm(t_1),$$

where

$$M_s{}^\pm(t)=\int_{-\infty}^{\infty}M_s{}^\pm(\Omega,t)\,d\Omega$$

is the total transverse magnetization of the impurities;

$$\int_{-\infty}^{\infty}g(\Omega)\,d\Omega=1;$$

$$\nu=(1-\lambda^2\chi_e\chi_s)^{-1},\quad \lambda=2J_{sf}/Ng_sg_e\mu_B^2,\quad \varkappa=g_s/g_e;$$

$$\chi_s(T)=\frac{S(S+1)}{3k_BT}(g_s\mu_B)^2cN,\quad \chi_e=\frac{(g_e\mu_B)^2}{2}\rho(\varepsilon_F)$$

are the static susceptibilities of the impurities and electrons respectively, $\rho(\varepsilon_F)$ is the density of states at the Fermi level ε_F, and T is the lattice temperature. The explicit form of the kinetic coefficients $\Sigma_{es}^{\pm}$, $\Sigma_{eL}^{\pm}$, and $\Sigma_{se}^{\pm}$ can be found in Ref. 11. In deriving the equations (3), we took into account the fact that the spectral densities of the quantities χ_s, $\Sigma_{es}^{\pm}$, and $\Sigma_{se}^{\pm}$ are related with the quantities themselves in the following manner:

$$\chi_s(\Omega)=g(\Omega)\chi_s,$$

$$\Sigma_{es}{}^\pm(\Omega,\,t)=g(\Omega)\Sigma_{es}{}^\pm(t),\quad \Sigma_{se}{}^\pm(\Omega,\,t)=\Sigma_{se}{}^\pm(t). \tag{4}$$

The local-field distribution function $g(\Omega)$ is arbitrary.

In the first equation of the system (3) the integral terms describe the drift of the magnetization of the impurities from the frequency interval $(\Omega,\Omega+d\Omega)$ with velocity $\Sigma_{se}^{\pm}(t)$ to the electrons and the return of the magnetization with velocity $\Sigma_{se}^{\pm}(\Omega,t)$ to a fraction $g(\Omega)$ of the spins with frequency lying in the indicated interval. The integral terms in the equation for $M_e^{\pm}(t)$ have a similar meaning, with the only difference that, for the electrons, relaxation of the magnetization into the lattice at a rate of $\Sigma_{eL}^{\pm}(t)$ is another channel. Notice that the equations of the system (3), with the kinetic coefficients $\Sigma_{ij}^{\pm}$ computed for arbitrary temperatures, are valid also for the low-temperature regime ($k_BT \lesssim \hbar\omega_s \approx 0.45$ K for the 9-GHz frequency band). To solve the equations (3), we perform a temporal Fourier transformation, defined as

$$\Phi(t)=\frac{1}{2\pi}\int_{-\infty}^{\infty}\Phi(\omega)\exp(-i\omega t)\,d\omega, \tag{5}$$

and go over to the dynamic susceptibilities

$$\chi_e^{\pm}(\omega)=M_e^{\pm}(\omega)/h^{\pm}(\omega),\ \chi_s^{\pm}(\Omega,\omega)=M_s^{\pm}(\Omega,\omega)/h^{\pm}(\omega). \tag{6}$$

We solve the resulting system of equations for $\chi_e^{\pm}(\omega)$ and $\chi_s^{\pm}(\Omega,\omega)$. Then, integrating both parts of the solution obtained for $\chi_s^{\pm}(\Omega,\omega)$ over Ω, we obtain for $\chi_e^{\pm}(\omega)$ and

$$\chi_s^{\pm}(\omega)=\int_{-\infty}^{\infty}\chi_s^{\pm}(\Omega,\omega)\,d\Omega,$$

a new system of equations whose solution for the total response of the system of impurities and electrons gives the complex dynamic susceptibility

$$\chi^{\pm}(\omega)=\chi'(\omega)+i\chi''(\omega)=\chi_e^{\pm}(\omega)+\chi_s^{\pm}(\omega)=\frac{\eta_s^{\pm}(\varepsilon_e^{\pm}+\xi_e^{\pm}-\omega)+\eta_e^{\pm}(\varepsilon_s^{\pm}+\xi_s^{\pm}-\omega)}{(\varepsilon_s^{\pm}-\omega)(\varepsilon_e^{\pm}-\omega)-\xi_s^{\pm}\xi_e^{\pm}}, \tag{7}$$

where

$$\varepsilon_s^{\pm}=[D^{\pm}(\omega)]^{-1}+\omega-i\lambda\varkappa\chi_e\Sigma_{es}^{\pm}(\omega),$$

$$\varepsilon_e^{\pm}=\mp\omega_e(1+\lambda\chi_s)\nu-i(\Sigma_{es}^{\pm}(\omega)+\Sigma_{eL}^{\pm}(\omega))-i\lambda\varkappa^{-1}\chi_s\Sigma_{se}^{\pm}(\omega),$$

$$\xi_s^{\pm}=\mp\lambda\omega_s\chi_s(1+\lambda\chi_e)\nu-i\varkappa\Sigma_{es}^{\pm}(\omega)-i\lambda\chi_s\Sigma_{se}^{\pm}(\omega),$$

$$\xi_e^{\pm}=\mp\lambda\omega_e\chi_e(1+\lambda\chi_s)\nu-i\varkappa^{-1}\Sigma_{se}^{\pm}(\omega)-i\lambda\chi_e(\Sigma_{es}^{\pm}(\omega)+\Sigma_{eL}^{\pm}(\omega)),$$

$$\eta_s^{\pm}=\mp\omega_s\chi_s(1+\lambda\chi_e)\nu-i\varkappa\Sigma_{se}^{\pm}(\omega)+i\varkappa\chi_e\Sigma_{es}^{\pm}(\omega),$$

$$\eta_e^{\pm}=\mp\omega_e\chi_e(1+\lambda\chi_s)\nu-i\chi_e(\Sigma_{es}^{\pm}(\omega)+\Sigma_{eL}^{\pm}(\omega))+i\varkappa^{-1}\chi_s\Sigma_{se}^{\pm}(\omega).$$

Here

$$D^{\pm}(\omega)=\int_{-\infty}^{\infty}d\Omega g(\Omega)\{-\omega\mp\Omega\mp\omega_s(1+\lambda\chi_e)\nu-i\Sigma_{se}^{\pm}(\omega)\}^{-1},$$

$$\Sigma_{ij}^{\pm}(\omega)=\int_{-\infty}^{0}\Sigma_{ij}^{\pm}(t)\exp(-i\omega t)\,dt.$$

In the high-temperature approximation ($\hbar\omega_s/k_BT \ll 1$) the kinetic coefficients $\Sigma_{ij}^{\pm}(\omega)$ do not depend on the frequency, and have the following form:

$$\Sigma_{se}=\frac{4\pi}{\hbar}\left[\frac{\rho(\varepsilon_F)J_{sf}}{N}\right]^2 k_BT,$$

$$\Sigma_{es}=\frac{8\pi}{3\hbar}cS(S+1)\frac{\rho(\varepsilon_F)J_{sf}^2}{N}, \tag{8}$$

$$\Sigma_{eL}=\frac{16}{9}\frac{\pi c'}{\hbar}\frac{\rho(\varepsilon_F)}{N}|V_{s0}|^2.$$

Here c' is the concentration of the nonmagnetic impurities and V_{s0} is the potential describing the scattering by them. Under normal—for EPR—conditions, $\Sigma_{se}\sim 10^9$ sec^{-1} and $\Sigma_{es}, \Sigma_{eL}\sim 10^{10}$–$10^{12}$ sec^{-1}.

As can be seen from (7), the solution to the equations (3) describing the dynamics of a spectrally inhomogeneous system of impurities can be written in the same form as for the case of a spectrally homogeneous system,[16] but the line shape is not Lorentzian, and depends on the shape of $g(\Omega)$ and the temperature (see below). If there is no inhomogeneous broadening of the spectrum of the impurities, the distribution degenerates into a delta function: $g(\Omega)=\delta(\Omega)$. In this case we obtain the well-known result for $\chi^{\pm}(\omega)$ (Ref. 16). If the local-field source is the intrinsic nuclear spin $\hat{I}$ of the impurity, then

$$g(\Omega)=(2I+1)^{-1}\sum_{I_z=-I}^{I}\delta(\Omega-\omega_{hf}I_z)$$

and the virtually infinite system of equations (3) becomes a system of $2I+2$ equations describing the dynamics of the hyperfine structure of an EPR line with a splitting of magnitude ω_{hf} between neighbors of the $2I+1$ components.[17–19]

If there is no bottleneck (i.e., if $\Sigma_{eL}\gg\Sigma_{es}$), then the electrons are in equilibrium, and the impurities having different frequencies inside the $g(\Omega)$ profile relax independently of each other. In this case the line shape should evidently be the convolution of the homogeneously broadened line with the local-field distribution $g(\Omega)$; this can be shown from (7), using the above indicated inequality. Thus, all the particular and limiting cases investigated earlier follow from the equations (3) and their solutions (7).

WIDTH OF THE DYNAMICALLY NARROWED EPR LINES

Let us consider the situation in which there is a bottleneck: $\Sigma_{es}\gg\Sigma_{eL}$. In this case the nonequilibrium magnetization transferred from one part of the $g(\Omega)$ spectrum to the electrons is more likely to return to the impurities than to relax into the lattice. And, as can be seen from the equations (3), the magnetization will return not necessarily to the former frequency interval $(\Omega,\Omega+d\Omega)$, but to any other interval $(\Omega',\Omega'+d\Omega')$ with probability proportional to the number of spins having the frequency Ω'. Thus, the impurities having different local frequencies turn out to be coupled via the electrons by the cross-relaxation. If the rate of the cross-relaxation is greater than the magnitude of the local-field spread, then we should expect, as in the Anderson-Weiss theory of exchange narrowing,[20] a narrowing down of the EPR line. Let us assume that the relation $\Sigma_{se}\gg\Delta$, where Δ characterizes the width of the $g(\Omega)$ distribution, is fulfilled. Then the denominator in the $D^{\pm}(\omega)$ integral in (7) can be expanded in a power series in the small ratio

$\Omega/(-\omega \mp \omega_s - i\Sigma_{se})$, and integrated. For clearness of exposition, let us set $g_s = g_e$, which gives rise to a situation favorable to the formation of a bottleneck,[2] and neglect the molecular fields, i.e., set $\lambda = 0$. Then

$$D^{\pm}(\omega) = \frac{1}{(-\omega \mp \omega_s - i\Sigma_{se})} \times \left[1 \pm \frac{M_1}{(-\omega \mp \omega_s - i\Sigma_{se})} + \frac{M_2}{(-\omega \mp \omega_s - i\Sigma_{se})^2}\right], \quad (9)$$

where

$$M_1 = \int_{-\infty}^{\infty} \Omega g(\Omega)\, d\Omega, \quad M_2 = \int_{-\infty}^{\infty} \Omega^2 g(\Omega)\, d\Omega$$

are the moments of the function $g(\Omega)$. If $M_1/\Sigma_{se} \ll 1$ and $M_2/\Sigma_{se}^2 \ll 1$, the terms of the expansion can be returned to the denominator. The substitution of $D^{\pm}(\omega)$ into the expression for $\varepsilon_s^{\pm}$ in (7) and the analysis of the susceptibility show that $\chi^{\pm}(\omega)$ possesses a simple pole, which determines the position and width of the collective mode occurring in the situation with the bottleneck:

$$\omega^{\pm} = \mp(\omega_s + M_1) - iB\Sigma_{se} - iM_2/\Sigma_{sc}, \quad (10)$$

where the bottleneck factor B, which lies in the interval (0,1), is given by the expression

$$B = \Sigma_{eL}/(\Sigma_{eL} + \Sigma_{es}). \quad (11)$$

As a result of the simple pole of the form $\chi^{\pm}(\omega)$, the shape of the collective mode is Lorentzian for any shape of $g(\Omega)$ if the latter fulfills the requirement that the second moment M_2 be finite and the ratio M_2/Σ_{se}^2 be small. The ratio M_2/Σ_{se} is the residual contribution from the local-field distribution $g(\Omega)$, which is narrowed down by the cross-relaxation under bottleneck conditions. As can be seen from (10), the contribution (Σ_{se}) of the Corringa mechanism to the EPR line width is reduced by the dynamic character of the interaction between the impurities and the electrons (lower B values correspond to a narrower bottleneck). It can also be seen from (10) that the extent of the narrowing is regulated by the Corringa rate $\Sigma_{se} \propto T$ (8) (when $\Sigma_{se} \gg \Sigma_{se}$, which is almost always the case, the narrowest bottleneck in the two-stage cross-relaxation process impurities$\rightarrow(\Sigma_{se})\rightarrow$electrons$\rightarrow(\Sigma_{es})\rightarrow$ impurities is the first stage, which proceeds at a rate of Σ_{se}, and therefore it determines the effective cross-relaxation rate). Thus, we should expect the inhomogeneous contribution to the line width to increase as the temperature is lowered. Now, instead of (1), we can approximate the temperature dependence of the width by the following expression:

$$\Gamma(T) = a^* + b^*T + cT^{-1}, \quad (12)$$

where $b^* = Bb$ and $c = 2TM_2/\Sigma_{se}$. Evidently, $\Gamma(T)$, (12), has a minimum at

$$T_m = (2TM_2/bB\Sigma_{se})^{1/2} \quad (13)$$

[but this is not necessarily the case for the width of $\chi''(\omega)$, (7), since the temperature region of validity of (12) is bounded from below]. It is easy to see from (13) that the narrower the bottleneck (i.e., the smaller B is) and the broader the $g(\Omega)$ distribution, the higher the temperature T_m at which $\Gamma(T)$ has its minimum. The behavior of $\Gamma(T)$ in the region of intermediate and slight narrowings can be investigated in detail only by numerical methods.

Before proceeding to the numerical analysis, let us note that, for a distribution of the Lorentzian form

$$g_L(\Omega) = \Delta/\pi(\Omega^2 + \Delta^2)$$

the second moment $M_2 = \infty$, and the criterion for narrowing $M_2/\Sigma_{se}^2 \ll 1$ is not fulfilled. Therefore, the Lorentzian distribution does not narrow down, and its shape does not change. Salikhov *et al.*[21] and Silsbee and Hone[22] have also concluded on the basis of model calculations that a distribution with the Lorentzian shape does not narrow down in the course of spectral diffusion. It is, however, clear that a real $g_L(\Omega)$ distribution has finite wings. Computer calculations have shown that, when the range of the integration in the expression (7) for $D^{\pm}(\omega)$ is cut off after, say, ten halfwidths (with $g(\Omega)$ normalized to unity beforehand in the indicated range), the EPR line undergoes insignificant narrowing.

For the subsequent numerical analysis, we choose a symmetric Gaussian distribution function $g_G(\Omega)$ and certain symmetric functions that decrease according to a power law at the wings. Since the results are qualitatively similar, we limit ourselves to one example:

$$g_r(\Omega) = \frac{1}{\sigma\sqrt{2\pi}} \exp\left(-\frac{\Omega^2}{2\sigma^2}\right), \quad \sigma = \frac{\Delta}{(2\ln 2)^{1/2}}. \quad (14)$$

The program was written in the FORTRAN language and computes: the complex susceptibility $\chi^{\pm}(\omega)$, (7), the width of $\chi''(\omega)$ at half-maximum, the absorbed power $P(H_0,\omega)$, and the field derivative dP/dH_0 of the absorbed power. Figure 1 shows plots of the dependence of the line width on temperature (in units of $\Sigma_{se}/\Delta \propto T$) for different values of the factor B. It can be seen from the figure that, as the bottleneck narrows (i.e., as B decreases), there appears a distinct minimum in the dependence $\Gamma(T)$, and that this minimum moves into the region of higher temperatures, as follows from the approximate formula (12). It can be seen from Fig. 1 that, for the factors $B = 0.1$ and 0.03, the total width of the $\Gamma(T)$ line in the region of the minimum is significantly smaller than the width of the distribution $g_G(\Omega)$. The line width increases at low temperatures because of the removal of the dynamic nar-

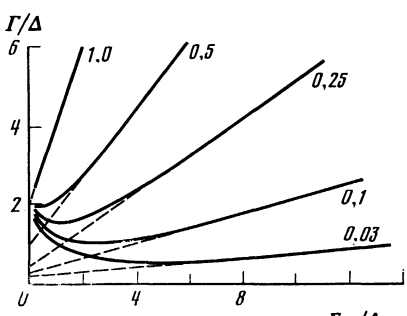

FIG. 1. Dependence of the EPR line width at half-maximum of $\chi''(\omega)$ on the temperature (in units of $\Sigma_{se}/\Delta \propto T$) for $g_G(\Omega)$ [see (14)]; Δ is the halfwidth of the function $g_G(\Omega)$. The values of B are indicated on the curves.

rowing as a result of the decrease of the cross-relaxation rate $\propto T$.

Let us now discuss in greater detail the experimental investigations reported in Refs. 3–7, in which the line broadening at low temperatures is attributed to magnetic ordering, and let us look at them from the standpoint of the present investigation. In experiment, in order to change the factor B, we must change the concentration of the magnetic impurities, i.e., Σ_{es} (8), or the concentration of the nonmagnetic spin-orbit scatterers, i.e., Σ_{eL} (8), through, for example, the addition of impurities prepared from the actinide series; in other words, we must prepare a new sample. Thus, when the impurity concentration is changed, the factor B decreases by more than a factor of 13 in the case of Gd in LaB_6 (Ref. 3), by almost a factor of four in the case of Gd in $LaAl_2$ (Ref. 4), by almost a factor of 10 in the case of Gd in amorphous YAl_2 (Ref. 5), and by more than a factor of 10 in the case of Mn in the semiconductor PbTe (Ref. 6). Thus, the B values for which the curves in Fig. 1 were constructed are typical values for experiments of that sort. It is clear that it is impossible to secure the same shape and width of the distribution $g(\Omega)$ in different samples; nonetheless, the similarity between the graphs in Fig. 1 and the corresponding curves given in Refs. 3–7 is apparent. As the bottleneck narrows (B decreases), there appears on the width-versus-temperature curves a minimum that shifts into the region of higher temperatures, as in Fig. 1. We should expect that in $Gd_xY_{1-x}Al_2$ (Ref. 5) the distribution function $g(\Omega)$ will be least sensitive to changes in the Gd concentration ($0.0059 \leqslant x \leqslant 0.33$) because of the amorphism of the compound. Malozemoff *et al.*[5] have noted as incomprehensible the fact that the residual width a obtained by extrapolating the high-temperature data to $T = 0$ K decreases as the bottleneck narrows. If we try to carry out a similar extrapolation (see the dashed lines in Fig. 1), then, as in Ref. 5, we obtain a systematically decreasing positive residual width (1→5). This procedure is nonunique, since, according to (12), the function $\Gamma(T)$ is nonlinear in the high-temperature region as well, but the error is not large, since the contribution of c/T is small. In experiment the nonlinearity is not noticeable because of the statistical spread of the measurement data. It is clear from the foregoing that allowance for the dynamic narrowing in the interpretation of experiments is not less important than allowance for the effects connected with the appearance of magnetic order.

It is shown in Ref. 12, and this is well corroborated by experiment (see, for example, Ref. 23), that the exchange interactions between the impurities lead to the renormalization of the relaxation rate:

$$\Sigma_{se} \to \Sigma_{se}' = \Sigma_{se}(1-\Theta/T).$$

Therefore, the dynamic narrowing will be removed as $T \to \Theta$, where Θ is the Curie paramagnetic temperature, which can be either positive or negative.

THE EPR LINE SHAPE

The numerical computations allow us to construct the field sweep of $\chi''(\omega)$ for different temperatures and B factors.

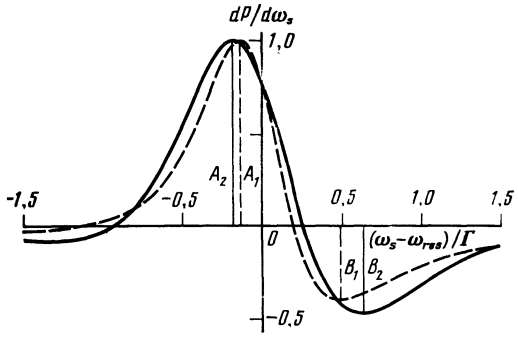

FIG. 2. The field derivative of the microwave power absorbed by a bulk metallic sample, $dP/d\omega_s$ ($\omega_s \propto H_0$); $g(\Omega) = g_G(\Omega)$, (14); $\Sigma_{se}/\Delta = 0.2$; $B = 0.1$; $A_2/B_2 \approx 2.11$. The dashed lines are the corresponding plot for the Lorentzian function $g_L(\Omega)$; $A_1/B_1 = 2.55$. The curves have been reduced to the same flow-field peak amplitude, i.e., $A_1 = A_2$.

It can be seen from them that at high temperatures and (or) low B values the line shape is nearly Lorentzian, as follows from (10). As the temperature is lowered, the line shape changes continuously, tending to the shape of the distribution $g(\Omega)$ at low temperatures. The solution to the equations (3) together with the Maxwell equations shows that the absorbed power $P(H_0,\omega)$ is proportional to the sum of the real and imaginary parts of the dynamic susceptibility $\chi^{\pm}(\omega)$, (7), describing the collective response of the impurities and the electrons. Figure 2 shows the plot of the ω_s derivative $-dP/d\omega_s$ of the power absorbed in the external-field ($H_0 \propto \omega_s$) sweep at a temperature below T_m. For comparison, we also show the $dP/d\omega_s$ for a Lorentzian line of equivalent width Γ. It is well known that, for the Lorentzian shape, the ratio of the amplitudes of the flow-field and high-field signal peaks $A_1/B_1 = 2.55$, and that the halfwidth $\Gamma/2$ of the χ''-line can be obtained by dividing the peak width $\Delta\omega_{pp}^L$ (the distance between the vertices of the $dP/d\omega_s$ signal peaks) by 1.268. It is clear from Fig. 2 that: 1) in the region where the line is broadened the ratio $A_2/B_2 < 2.55$; 2) the peak width $\Delta\omega_{pp}$ for non-Lorentzian lines differ significantly from $\Delta\omega_{pp}^L$ for the same χ''-line width, and the difference is systematic and increases with decreasing temperature. The experimental data on the line shape in the line-broadening region are scanty: it is noted in Ref. 7 that the signal shape is non-Lorentzian, and that the signal has a lower intensity at the wings (the continuous curve in Fig. 2 exhibits the same behavior); in Ref. 5 it is noted that the line shape is non-Lorentzian, and therefore data for the peak width are given. Our calculation shows that a spectral analysis performed under the assumption that the line shape is Lorentzian can lead to a significant error in the determination of the true width of the EPR lines.

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