EPR line shape in a type II superconductor

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The absorption line shape for paramagnetic impurities in a type II superconductor is calculated. It is the convolution of homogeneously broadened line with the local-field distribution function in the vortex lattice. The homogeneously broadened line is determined by the combination $\chi' + \alpha\chi''$ of Lorentz dispersion (χ') and absorption (χ'') curves with $\alpha > 1$ (in contrast to the normal metal, for which $\alpha = 1$). Together with the inhomogeneous broadening mechanism, this also leads to a reduction in the line asymmetry parameter (the ratio A/B) observed in the experiments. The theory is compared with available experimental data.

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1. INTRODUCTION

The method of electron paramagnetic resonance (EPR) in metals allows us to extract important information concerning the static and dynamic properties of localized moments and conduction electrons. Here we primarily refer to the measurements of the shift in the resonance field (the g-factor shift), of the rates of relaxation between subsystems of localized spins and of the conduction electrons, and also between them and the lattice. One of the most important characteristics of EPR in metals is the paramagnetic absorption line shape. As is well known, in contrast to dielectrics (where the line shape is determined only by the imaginary part of the dynamic susceptibility $\chi = \chi' + i\chi''$), in metals the resonance line shape is a mixture of χ' and χ'' (Refs. 1,2) due to the fact that the electromagnetic field penetrating deep into the metal changes both in amplitude and in phase. These changes occur with the same characteristic length δ_0 (δ_0 is the skin depth of the normal metal); therefore χ' and χ'' enter into the expression for the line shape with equal weight, as has been repeatedly observed in experiments.[2,3]

Recently the EPR method has been widely used to study type-II superconductors.[2,4,5] In particular, it has been observed that the g-factor shift depends on the static magnetic field, and the line shape substantially changes on going from the normal phase to the superconducting phase. Thus, for example the line asymmetry parameter (the ratio of the low-field peak to the high-field peak in the derivative of absorption with respect to magnetic field: the ratio A/B) decreases from 2.55 in the normal metal down to ~1 in the superconducting metal (for $T \ll T_c$, where T_c is the critical temperature). In order to interpret these properties of the paramagnetic state in type-II superconductors, a theoretical calculation of the EPR line shape is necessary. Orbach[6] and also Alekseevskii *et al.*[7] have calculated the absorption line shape in the form of a convolution of a homogeneously-broadened line (taken to be the line shape of the normal metal) with the distribution function of the local fields in the unit cell of the vortex lattice. However, this approach completely disregards the fact that the microwave field penetrates into the superconductor differently than into the normal metal, due to screening by superconducting alternating currents (see, for example, Ref. 8). Therefore it is not surprising that such calculations seldom agree with experiments.[6] Furthermore, in recent experiments Barberis *et al.*[5] have observed that the EPR line shape of Nd^{3+} in superconducting $CeRu_2$ and $ThRu_2$ may be described simply by a combination of χ' and χ'' with different weights, without invoking the mechanism of inhomogeneous broadening.

In connection with what has been said above, this paper is devoted to constructing a systematic theory for the EPR line shape, taking into account both the inhomogeneous broadening of the static field in the sample and the characteristic features of penetration of the alternating field into the superconductor.

2. LINE-SHAPE CALCULATION

Let us consider a "dirty" superconductor with paramagnetic impurities occupying the $z \geq 0$ half-space and located in a constant magnetic field H_0. Let a microwave electromagnetic field of frequency ω and with components $\mathbf{H}_1 \| x$ and $\mathbf{E}_1 \| y$ be applied to the sample. We are interested in resonant absorption of energy from the alternating field by the paramagnetic impurities; therefore $\mathbf{H}_1 \perp \mathbf{H}_0$, and $\mathbf{H}_0$ can be directed either perpendicular to the surface of the sample ($\mathbf{H}_0 \perp \mathbf{E}_1$), or parallel to the surface ($\mathbf{H}_0 \| \mathbf{E}_1$). As a rule, the EPR experiments are carried out in the field region $H_{c1} \ll H_0 < H_{c2}$ and in the temperature region $T \ll T_c$ (where H_{c1} and H_{c2} are respectively the lower and upper critical fields). This case corresponds to a high density of vortex filaments in the sample, so that the separation d between their centers is much smaller than the penetration depth λ of the d.c. field.[9]

In the microwave frequency region, the skin depth in a normal metal usually exceeds λ.[4,10] Therefore we can expect the average penetration depth for the alternating field in a type-II superconductor to be much greater than d. In this case, reflection of the electromagnetic wave from the surface of the type-II superconductor can be studied using the system of macroscopic Maxwell equations

$$\mathrm{rot}\,\mathbf{H}_1 = \frac{4\pi}{c}\mathbf{j}, \quad \mathrm{rot}\,\mathbf{E}_1 = -\frac{1}{c}\left(\mu\frac{\partial \mathbf{H}_1}{\partial t} + 4\pi\frac{\partial \mathbf{M}}{\partial t}\right), \tag{1}$$

obtained from the corresponding microscopic equations by averaging over the vortex lattice. In Eq. (1), E_1

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and H_1 are the macroscopic strengths of the alternating electric and magnetic fields; μ is the effective magnetic permeability of the type-II superconductor, and reflects the distortion of the vortex structure under the action of the alternating field and depends on the relative orientation of $\mathbf{H}_1$ and $\mathbf{H}_0$ (Ref. 11); $\mathbf{M}$ is the macroscopic magnetization of the localized spins of the impurities; $\mathbf{j}$ is the alternating-current density. All the parameters in Eq. (1), except for the constant μ, are now functions of the single coordinate z.

In addition to Eqs. (1), we still need the material equations connecting $\mathbf{j}$ with $\mathbf{E}_1$ and $\mathbf{M}$ with $\mathbf{H}_1$. In our case of a dirty superconductor with $l \ll \xi_0$ (where l is the mean free path of the conduction electrons, and ξ_0 is the coherence length of the pure superconductor), $\mathbf{j}$ and $\mathbf{E}_1$ are connected by the local relationship

$$\mathbf{j}=\sigma_s(\omega,\ \mathbf{H}_0,\ T)\mathbf{E}_1, \tag{2}$$

where $\sigma_s = \sigma_1 + i\sigma_2$ is the effective complex conductivity of the type-II superconductor, and depends on the frequency of the alternating field ω, on the strength of the constant field H_0, on the temperature T, and also on the relative orientation of $\mathbf{H}_0, \mathbf{E}_1$, and the surface of the sample (see the review in Ref. 12). Calculations of σ_s have been performed earlier both on the basis of the linear response theory[12] as well as by using the nonstationary Ginzburg–Landau equations.[13,14] Unfortunately, the analytical expressions obtained for the complex conductivity from these calculations are valid only in the vicinity of the transition to the normal state: either at $H_0 \approx H_{c2}$ (Refs. 12, 13) or at $T \approx T_c$ (Ref. 14). We are interested in the region $H_{c1} \ll H_0 < H_{c2}$ and in T substantially less than T_c. Therefore we will consider σ_s as a parameter which can be determined by experimental investigation of the nonresonant part of the surface impedance.

As is well known, the magnetization of the localized spins in a normal metal obey a Bloch-type equation that describes the relaxation of the magnetization to its instantaneous equilibrium value.[15] A similar equation of motion for the magnetization was obtained for a gapless type-II superconductor ($H_0 \simeq H_{c2}$) in Ref. 16. It is inapplicable in our case. Therefore we proceed in the following way. To be specific, let us consider the perpendicular orientation ($\mathbf{H}_0 \perp \mathbf{E}_1$ and to the surface of the sample). In a plane perpendicular to the direction of the static field $\mathbf{H}_0$ we isolate a region from $\mathbf{r}$ to $\mathbf{r}+d\mathbf{r}$ (where $\mathbf{r}$ is a two-dimensional vector in the vortex lattice) with dimensions which are significantly smaller than the separations between vortices, so that the magnetic field is practically homogeneous in this region.

The magnetization density $\mathbf{m}(\mathbf{r},t)$ of this region obviously satisfies the equation

$$\frac{\partial \mathbf{m}}{\partial t}=g\mu_B[\mathbf{m},(\mathbf{h}_0(\mathbf{r})+\mathbf{h}_1(\mathbf{r},t))]-\frac{1}{T_2(\mathbf{r})}\{\mathbf{m}-\chi_0(\mathbf{h}_0(\mathbf{r})+\mathbf{h}_1(\mathbf{r},t))\}, \tag{3}$$

where μ_B is the Bohr magneton; g and χ_0 are respectively the g factor and the static susceptibility of the localized spins; $\mathbf{h}_0(\mathbf{r})$ and $\mathbf{h}_1(\mathbf{r},t)$ are the local (microscopic) values of the constant and alternating fields at the point $\mathbf{r}$ of the vortex lattice; $T_2(\mathbf{r})$ is the local relaxation time of the paramagnetic impurities, reflecting the fact that the localized spins located at the core of the vortex relax faster than the spins outside the core (due to the fact that the excitation spectrum of conduction electrons within them is gapless—see, for example, Ref. 17.)

Solving Eq. (3) relative to $m_+ = m_x + im_y$ (the components of the local magnetization) using the time dependence of the alternating field in the form $e^{-i\omega t}$, and then averaging the solution obtained over the unit cell Σ of the vortex lattice, we obtain the following expression for the macroscopic magnetization of the localized spins:

$$M_+=\langle m_+\rangle_\Sigma=\mu H_1\int f(\omega')\chi(\omega',\omega)\,d\omega', \tag{4}$$

where $\mu H_1 = \langle h_1\rangle_\Sigma$, $\chi(\omega',\omega)$ is the dynamic susceptibility of the paramagnetic impurities

$$\chi=\chi'+i\chi''=\chi_0\frac{\omega' T_2(\omega')-i}{(\omega'-\omega)T_2(\omega')-i}, \tag{5}$$

and $f(\omega')$ is the distribution function for local frequencies (fields) in the unit cell of the vortex lattice (see Ref. 12)

$$f(\omega')=\int_\Sigma d^2r\,\delta(\omega_0)(\mathbf{r})-\omega')\Big/\int_\Sigma d^2r; \tag{6}$$

here $\omega_0(\mathbf{r}) = g\mu_B h_0(\mathbf{r})$ is the local Zeeman frequency of the paramagnetic impurity. The function $f(\omega')$ determines the number of paramagnetic impurities (the distribution of which over the sample we assume to be statistical) having the resonance frequency ω'. In obtaining Eq. (4) we used the fact that $h_1(\mathbf{r},t)$ (as can be easily established from analysis of the nonstationary Ginzburg–Landau equations cited in Ref. 14) are practically unchanged within the boundaries of the unit cell.

When $\omega' T_2(\omega') \gg 1$, which is the condition for observability of the paramagnetic resonance, the function $\chi(\omega',\omega)$ in Eq. (5) has a high peak of width $T_2^{-1}(\omega)$ at $\omega' = \omega$. In what follows, we will be interested specifically in the resonant part of the function $\chi(\omega'-\omega)$, especially since $T_2^{-1}(\omega')$ is a smooth function[18] and ω' changes insignificantly within the line width.

Using the Maxwell equations (1), the material equations (2) and (4), and also the boundary conditions which include the continuity of the tangential components of $\mathbf{H}_1$ and $\mathbf{E}_1$, it is not difficult to calculate the surface impedance for the superconducting half-space

$$Z=\frac{4\pi}{c}\frac{\mathbf{n}[\mathbf{E}_1\times\mathbf{H}_1]}{|\mathbf{H}_1|^2}\Big|_{z=0}=Z_0+Z_M,$$

$$Z_0=R-iX=\frac{\mu^{1/2}R_n}{G}\left[\left(G-\frac{\sigma_2}{\sigma_n}\right)^{1/2}-i\left(G+\frac{\sigma_2}{\sigma_n}\right)^{1/2}\right], \tag{7}$$

$$G=[(\sigma_1/\sigma_n)^2+(\sigma_2/\sigma_n)^2]^{1/2},$$

$$Z_M=2\pi Z_0\int f(\omega')\chi(\omega'-\omega)\,d\omega',$$

where Z_0 is the nonresonant part of the surface impedance of the type-II superconductor, and R and X are the corresponding surface resistance and reactance (see, for example, Ref. 19); in the orientation considered ($\mathbf{H}_0 \perp \mathbf{E}_1, \mathbf{H}_1$), we have $\mu = B_0/H_0$ (Ref. 11) [where $B_0 = \langle h_0(\mathbf{r})\rangle_\Sigma$ is the static magnetic induction]; σ_n, $R_n = (\sigma_n\delta_0)^{-1}$, and δ_0 are respectively the conductivity, the surface resistance, and the skin depth in the normal phase; and finally, Z_M is the resonant part of the surface impedance, calculated to first order in χ_0, since in the paramagnetic state $\chi_0 \ll 1$.

The EPR line shape is determined by the microwave power absorbed by the paramagnetic impurities per unit area of the surface of the metal, i.e.,

$$P=\left(\frac{c}{4\pi}\right)^2 H_1^2(0)\,\mathrm{Re}\,Z_M. \tag{8}$$

Substituting here Z_M from Eq. (7), we obtain

$$P=2\pi P_0 \int f(\omega')\,[\chi'(\omega'-\omega)+\alpha\chi''(\omega'-\omega)]\,d\omega'. \tag{9}$$

Here $P_0=(c/4\pi)^2 H_1^2(0)R$ is the power nonresonantly absorbed by the type-II superconductor and $\alpha = X/R$ is the ratio of the surface reactance to the surface resistance, a quantity larger than unity, as is evident from Eq. (7), and dependent on ω, H_0, and T.

The approach presented, which employ averaging of the microscopic Maxwell equations and the equations of motion of the local magnetization of the impurities, is valid if the actual penetration depth of the alternating field $\delta_{act}(\omega)=\delta_0[\mu(G+\sigma_2/\sigma_n)]^{-1/2}$ significantly exceeds the period d of the vortex lattice, and the concentration of paramagnetic impurities is such that the average separation between them is much less than d and magnetic ordering effects are insignificant.

An analogous calculation may be performed for parallel orientation ($H_0 \| E_1$ and to the surface of the sample). In our case, when the penetration depth of the alternating field significantly exceeds the period of the vortex lattice, we obtain the same result as in Eq. (9) above. However, we should note that the parameter α in Eq. (9) strongly depends on the relative orientation of the vectors H_0 and E_1. In the $H_0 \perp E_1$ geometry the dominant contribution to the surface resistance is from the fluctuating motion of the vortex lattice under the action of the Lorentz force exerted by the superfluid component of the alternating current. In the $H_0 \| E_1$ orientation, the vortices are at rest (the Lorentz force is equal to zero) and the surface resistance is significantly smaller than in the perpendicular orientation, up to fields $H_0 \approx H_{c2}$ (Refs. 10, 12). Thus, due to the anisotropy of the surface impedance Z_0, the line shape of the paramagnetic resonance also depends on the relative orientation of the vectors H_0 and E_1.

3. DISCUSSION OF RESULTS

Equation (9) allows us to describe the basic changes in the shape and position of the EPR line that occur on going from the normal phase to the superconducting phase. In this equation, we take into account the two important (for EPR in conductors) differences between the type-II superconductor and the normal metal. First of all, due to the inhomogeneous distribution of the constant magnetic field in the sample, the line shape is a convolution of the homogeneously-broadened line $\chi' + \alpha\chi''$ with the distribution function for the local fields in the unit cell of the vortex lattice. Secondly, the homogeneously broadened line itself differs from the line shape in the normal metal because the amplitude of the alternating field in the superconductor drops off over a distance smaller than that over which its phase changes; therefore $X > R$ in the superconductor and χ'' enters in Eq. (9) with greater weight than χ'. Physically, this stems from the purely screening character of the "superconducting" component of the alternating current $\mathbf{j}_s = i\sigma_2\mathbf{E}_1$, whereas the "normal" component of the current $\mathbf{j}_n=\sigma_1\mathbf{E}_1$ changes not only the amplitude of the alternating field $\mathbf{H}_1$, but also its phase.

The function $f(\omega')$ in Eq. (9) is the probability of encountering a given local Zeeman frequency in the vortex lattice (usually triangular). It is characterized by maximum ($\omega_v = g\mu_B h_v$) and minimum ($\omega_c = g\mu_B h_c$) local frequencies at which it becomes discontinuous, and also by a local frequency $\omega_s = g\mu_B h_s$ corresponding to a saddle point where $f(\omega')$ diverges logarithmically.[2] This means that most of the paramagnetic impurities have a local frequency $\omega_s < \omega_0 = g\mu_B H_0$, and accordingly the resonant field at a given frequency ω of the external field is shifted toward higher fields, as is indeed observed in experiments.[2,4,5] We should point out that the decrease in the static susceptibility of the conduction electrons on going into the superconducting state also introduces a contribution to the g-factor shift (see, for example, Ref. 4). This contribution may be easily separated from the preceding contribution by performing the experiments at different frequencies.[4,5]

Another important consequence of Eq. (9) is the change in the resonance line shape on going from the normal phase to the superconducting phase. In EPR experiments, what is usually observed directly is not the power actually absorbed by the spins but rather its derivative with respect to magnetic field dP/dH_0. The dP/dH_0 line shape is customarily characterized by the ratio of the low-field peak to the high-field peak (the ratio A/B). As has already been pointed out in Sec. 1, a decrease in this ratio from 2.55 in the normal metal down to ~1 in the superconducting metal has already been observed experimentally ($T \ll T_c$). Explanations of this behavior of the line shape have been given thus far on the basis of an inhomogeneous broadening mechanism. In this case it was shown that the observed g-factor shift and the decrease in the ratio A/B may be partially explained by a spread in local fields sensed by the paramagnetic impurities.[6] However, the calculated shape of the peaks themselves in dP/dH_0 in this case did not agree with the experimental peaks. On the other hand, Barberis *et al.*[5] could describe the observed shape of the EPR signal by a combination of $d\chi'/dH_0 + \alpha d\chi''/dH_0$ without calling on an inhomogeneous broadening mechanism, although they observed in this case a field-dependent shift in the g factor. It became clear therefore that inhomogeneous broadening is not the sole reason for the change in line shape in a type-II superconductor. Indeed, recognizing that the alternating field penetrates into the sample while undergoing additional screening from the superconducting alternating currents, we obtained for the line shape Eq. (9), where χ'' indeed enters with a greater weight ($\alpha > 1$). This also leads to a decrease in the ratio A/B, since the ratio $A/B = 1$ for $d\chi''/dH_0$. Thus, the EPR line shape changes in the general case as a consequence of the inhomogeneous broadening, and thus also as a consequence of the fact that the homogeneously broadened line differs substantially from the line shape in the normal metal.

In order to compare specific experimental lines with the theoretical lines from Eq. (9), we need additional data concerning the geometry of the experiment and the corresponding complex conductivity σ_s (or on the quantity $\alpha = X/R$), as well as concerning the homogeneous width of the absorption line $T_2^{-1}(\omega')$ and its relation to the maximum spread in local frequencies $\omega_v - \omega_c$. In a number of cases (impurities with spin $S > \frac{1}{2}$) additional broadening due to the unresolved fine structure is possible[4]; this greatly complicates a detailed comparison of the theory with experiment. Therefore, for simplicity we limit ourselves to the case $S = \frac{1}{2}$ and consider two limiting situations, connected with the different relationships between $\omega_v - \omega_c$ and $T_2^{-1}(\omega')$.

Let $T_2^{-1}(\omega') \ll (\omega_v - \omega_c)$ initially; such a relationship is typical for nuclear magnetic resonance (NMR) in type-II superconductors. In this case the integrand in Eq. (9) is the product of a slowly changing function $f(\omega')$ and a rapidly changing function $\chi' + \alpha\chi''$. The first term, proportional to χ', averages out to zero on integration in the second term χ'' can be approximated (apart from a factor) by a δ function. As a result, we obtain the following for the line shape:

$$P = 2\pi^2 P_0 \chi_0 \frac{\omega}{\omega_v - \omega_c} \alpha f\left(\frac{\omega - \omega_s}{\omega_v - \omega_c}\right). \qquad (10)$$

Thus, in this case the shape of the absorption curve is completely determined by the distribution function of the local frequencies in the vortex lattice. Indeed, as has been shown earlier (see Refs. 2, 17, and the citations in these articles), the NMR method allows direct study of the distribution of the constant field in a type-II superconductor.

In the other limiting case, when $T_2^{-1}(\omega') \gg (\omega_v - \omega_c)$, even $f(\omega')$ is a rapidly changing function, and $\chi' + \alpha\chi''$ is practically constant. In the integration of Eq. (9) in this situation, it is convenient to use the fact that $f(\omega')$ has a sharp peak at $\omega' = \omega_s$, and to take $\chi' + \alpha\chi''$ out from under the integral sign at this point. As a result, we obtain the following expression for the absorbed power:

$$P = 2\pi P_0 [\chi'(\omega_s - \omega) + \alpha\chi''(\omega_s - \omega)]. \qquad (11)$$

Such a line shape, characterized by a field-dependent g-factor shift $[\omega_s(H_0) < \omega_0]$ and at the same time described by a linear combination of Lorentz dispersion χ' and absorption χ'' curves, is in good qualitative agreement with the experiments of Barberis *et al.*[5] on Nd^{+3} in $CeRu_2$ and $ThRu_2$.

For an arbitrary relationship between the homogeneous width $T_2^{-1}(\omega')$ and the width of the field distribution $\omega_v - \omega_c$, the convolution in Eq. (9) may be calculated with a computer using an analytic approximation of the function $f(\omega')$, given, for example, in Ref. 7. It is interesting to note that for $\alpha \gg 1$ $(\sigma_2 \gg \sigma_1)$ the shape of the homogeneously broadened EPR line in a superconductor will resemble the shape of the absorption curve in dielectrics, χ''. In this case, the inhomogeneous broadening, more strongly reducing the amplitude of the low-field peak in the absorption derivative than the amplitude of the high-field peak (see Ref. 6), may cause the ratio A/B become less than 1.

Thus, the characteristic differences in the electrodynamics of type-II superconductors lead to a substantial change in the shape and position of the EPR line compared with normal metals. Also, consequently the EPR (NMR) method may be used to obtain information on the electrodynamic characteristics of superconductors, such as the distribution of the static field in the sample, the magnitude of the complex conductivity σ_s, or the value of the surface reactance X for a known surface resistance R (the resonance signal is observed on the background dR/dH_0).

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