Study of spin-wave resonance in a superconductor with paramagnetic impurities

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The dynamics of paramagnetic impurities in a superconductor in an external magnetic field was studied theoretically and experimentally. It is shown that long-wavelength ($q \leqslant \xi^{-1}$, where q is the wave vector and ξ is the coherence length) excitations, in contradistinction to diffusion of magnetization in a normal metal, have a coherent spin-wave character, even in the paramagnetic phase. This phenomenon is connected with the long-range ($R \sim \xi$) character of indirect exchange interaction between spins in a superconductor, thanks to which the internal exchange field fluctuates slightly. The damping of spin waves is calculated and the region of existence of spin waves is determined. Manifestation of spin-wave effects in EPR of erbium magnetic impurities in superconducting lanthanum films was observed in experiment. Additional absorption is observed on the high-field side of the principal resonance. This absorption distorts the resonance line, the distortion depending on the film thickness, the direction of the external magnetic field, the temperature, and the impurity concentration. Modeling of the spectra, taking into account microwave absorption by nonuniform spin-wave excitations, gives a satisfactory description of this behavior. The spin rigidity coefficient and the temperature dependence of the uniform spin susceptibility of lanthanum were determined.

INTRODUCTION

The role of magnetic-resonance methods in the study of the physics of the superconducting state is well known. In particular, nuclear magnetic resonance has been used successfully to study subtle questions concerning the quasiparticle spectrum and the symmetry of Cooper pairs. The method of electron paramagnetic resonance (EPR), in turn, is found to be especially effective for studying spin-spin exchange interactions and associated dynamical and static correlations of localized spins.[1] The latter capability is important for studying the interrelation of magnetic order and the superconducting state—a well-known problem which is still important today.

The present work is concerned with the study of the spectrum of long-wavelength spin excitations of magnetic impurities in a singlet BCS superconductor.[1)] Our work was motivated by the following considerations. The EPR study made in Ref. 3 of erbium impurities in lanthanum revealed an anomalous, almost jumplike narrowing of the absorption line at the superconducting transition of the sample. This behavior contrasted sharply with the expected change in the EPR linewidth. Alekseevskiĭ *et al.*[3] attributed the effect which they observed to a new exchange interaction appearing in the superconducting phase via the superconducting system of electrons. Owing to long-range correlations in the Cooper condensate, the interaction radius of this new interaction is large. This idea of an additional indirect interaction of local spins in a superconductor, first proposed by Anderson and Suhl[4] in connection with a possible modification of magnetic ordering in the superconducting phase, was confirmed by microscopic calculations for both "pure"[3,5] and "dirty"[6] superconductors.

The spin exchange-interaction integral[6]

$$H_{\mathrm{ex}} = \sum_{\langle ij \rangle} J(R_{ij})(S_i S_j) \tag{1}$$

can be represented approximately as a sum of two parts $J_n(R) + J_s(R)$, the first one, acting at short distances,

$$J_n(R) = -J_{\mathrm{sf}}^2 \chi_n(R) \approx \frac{J_0 v_0}{4\pi R^3}\left[\cos(2k_F R) - \frac{\sin(2k_F R)}{2k_F R}\right], \tag{2}$$

being the conventional Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction, and the second one

$$J_s(R) = -J_{\mathrm{sf}}^2 \Delta\chi_s(R) \approx \frac{J_0 v_0}{4\pi \xi^2 R}\,\delta\chi(T)\exp\left(-\frac{R}{\xi}\right) \tag{3}$$

being the "superconducting" correction. In Eqs. (2) and (3) v_0 is the volume per lattice site, ξ is the coherence length in the "dirty" superconductor, and k_F is the Fermi momentum. The quantity

$$\delta\chi(T) = (\chi_p - \chi_s)/\chi_p \leqslant 1$$

characterizes the decrease in the static spin susceptibility χ_s of conduction electrons in the singlet BCS state as compared with the Pauli susceptibility χ_p. The constant J_0 is given by the expression

$$J_0 = J_{\mathrm{sf}}^2 \rho(\varepsilon_F) v_0/2,$$

where J_{sf} is a parameter characterizing the exchange interaction of a local spin with a conduction electron and $\rho(\varepsilon_F)$ is the electron density of states at the Fermi level. In Eq. (2) the quantity $\chi_n(R)$ is the normal-metal nonlocal spin susceptibility and $\Delta\chi_s(R)$ in Eq. (3) is the superconducting correction to the susceptibility.

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The additional interaction (3) is of an antiferromagnetic character and is small: For example, at average distances between impurity spins $r_{av} \ll \zeta$ we have

$$|J_s(r_{av})/J_n(r_{av})| \propto r_{av}^2/\zeta^2 \ll 1.$$

However, owing to the long range ($R \leqslant \zeta$) and sign alternation, its contribution to the total molecular field is of the same order of magnitude as the normal RKKY contribution (2). It is now known[7] that the potential (3) is one reason why helicoidal magnetic structures form in superconductors, as was predicted in Ref. 4.

The expression (3) means that even when the concentration x of magnetic impurities is low the number of spins $N \approx x\zeta^3/v_0$ in the interaction region is anomalously high, since the coherence length ζ is much longer than the lattice constant. This introduces a new factor into spin dynamics in an external magnetic field: the mean exchange molecular field $\langle H_{ex} \rangle \sim N\langle S^z \rangle$ induced by the external magnetic field H. At the same time, the rms fluctuation of the exchange field in the para-phase is

$$\langle H_{ex}^2 \rangle^{1/2} \sim [NS(S+1)]^{1/2} \sim SN^{1/2}.$$

This means that if the coherence length ζ is sufficiently large, then even in the paramagnetic region the fluctuations of the exchange field are small compared with the average value:

$$\langle H_{ex}^2 \rangle^{1/2}/\langle H_{ex} \rangle \approx \frac{k_B T}{\beta H} N^{1/2} \ll 1. \tag{4}$$

The smallness of the fluctuations suggests[8] that the long-wavelength excitations of the impurity spins in an external magnetic field exhibit magnon behavior, i.e., transport of spin excitations is of a weakly dissipative wave character.

The foregoing considerations concerning the possibility of spin-wave oscillations of magnetic impurities in a superconductor actually require a more accurate argumentation. Local field fluctuations which are connected with the short-range part of the potential (2) and the dipole interactions, as well as with the spatial disordering of impurities, can in principle give significant damping of magnons and limit the range of temperatures, magnetic fields, and wave vectors where a spin-wave description is possible.

Thus, the object of the present work can be formulated as follows: to investigate, theoretically and experimentally, spin-wave oscillations in a system of paramagnetic impurities in a superconductor in an external field. The conditions under which magnon excitations exist are determined in Sec. 1. The results of an experimental search for spin waves in superconducting $La_{1-x}Er_x$ films are presented in Sec. 2. Data that make it possible to estimate the required parameters of the experimental samples are also given in Sec. 2. In Sec. 3 the experimental results are compared with theory and the results obtained are discussed.

1. THEORY

We are interested in the dynamics of the transverse magnetization S^+ of magnetic impurities coupled by the Heisenberg interaction (1). Since the latter interaction conserves the total spin of the system, the magnetization averaged over larger distances than the average distance r_{av} between spins is a quasi-integral of the motion and the long-wavelength excitations are, evidently, hydrodynamic diffusion modes and are described by Bloch equations with the diffusion term iDq^2, where q is the wave vector. The spatial disordering of magnetic impurities introduces well-known difficulties in the calculation of the diffusion coefficient D. Care must be exercised in averaging the kinetic transport equation for the nonequilibrium magnetization, in order that a physically reasonable result be obtained: The diffusion coefficient is determined by the interaction of the spins at average distances r_{av},[9,10] and not by the interaction of close random exchange pairs.

In an external magnetic field the diffusion coefficient becomes complex: $D' + iD''$; the imaginary part is related to the appearance of a static molecular field, proportional to the spin polarization $\langle S^z \rangle$. As far as short-range potentials of the RKKY type (2) in normal metals are concerned, thermal fluctuations of the exchange field are much larger than its average value (we emphasize that it is the paramagnetic phase that is referred to everywhere below). For this reason $D''/D' \sim \langle S^z \rangle \ll 1$ and the diffusion regime is preserved. The basic idea of the calculations presented below is that the "superconducting" correction (3) to the potential makes a large contribution to D, and for certain values of the parameters the coefficient D becomes almost imaginary: $D''/D' \gg 1$; this indicates the appearance of transverse rigidity and of real dispersion of spin excitations.

Dipole–dipole interactions between impurities must also be included in the calculations: they determine the damping of excitations in the limit of small q, and they limit the "window" for spin waves at low momenta. A good calculation of this damping, which determines the EPR linewidth, in magnetically dilute systems has not yet been performed. The classical work of Anderson and Weiss[11] on exchange narrowing of the EPR line cannot be applied directly to disordered systems, and the standard method of moments simply leads to incorrect results. The qualitative conclusion[12] is that the interaction of spins at average distances determines how effectively the dipole contribution to damping due to exchange fluctuations is suppressed.

The kinetic equations can be derived by Zubarev's nonequilibrium statistical operator method.[13] For this, the quantum-mechanical equation of motion for the local spin S_i^-

$$i\frac{\partial S_i^-}{\partial t} = \left[\omega_0 + \sum_j (J_{ij} + A_{ij})\langle S_j^z \rangle\right] S_i^- - \sum_j j_{ij}\langle S_i^z \rangle S_j^- + \sum_j \{(J_{ij} + A_{ij})\delta S_j^z S_i^- - J_{ij}\delta S_i^z S_j^-\} \tag{5}$$

must be averaged with the help of the nonequilibrium statistical operator

$$\rho=Z^{-1}\exp\left\{-\frac{\mathscr{H}}{kT}-\sum_i\left[F_i^+(t)S_i^- - \int_{-\infty}^0 dt e^{\varepsilon t}\frac{\partial}{\partial t}F_i^+(t+t)S_i^-(t)+\text{h.c.}\right]\right\},\quad \varepsilon\to+0. \tag{6}$$

The notation is as follows: $\omega_0=g\beta H$ is the Zeeman frequency, g is the g-factor, $\delta S^z=S^z-\langle S^z\rangle$, J_{ij} is the exchange-coupling potential (1), Z is the partition function, and

$$A_{ij}=\frac{3}{2}\frac{g^2\beta^2}{R^3}(3\cos^2\vartheta_i-1) \tag{7}$$

is the dipole interaction constant at distance r_{ij}. The Hamiltonian $\mathscr{H}$ in (6) includes the Zeeman, dipole, and exchange interactions of the spins; $F(t)$ are conjugate thermodynamic parameters.[13] The summation in Eq. (5) and in what follows below extends over the impurities. It is assumed that at average distances between impurities the exchange interaction is stronger than the dipole interaction ($J_{ij}>A_{ij}$), and this results in averaging of the dipole fields (exchange narrowing of the EPR line). This condition is quite typical for metals. To avoid details which are not important for our purposes, terms resulting in the standard Korringa relaxation of local spins into conduction electrons are dropped from Eq. (5). The corresponding contribution to the damping of spin excitations in the paraphase does not depend on the wave vector and can be simply taken into account as natural broadening.

We note, finally, that our choice in Eq. (6) of the transverse components of the spins as the parameters for describing the weakly nonequilibrium state of the system is motivated by the following qualitative considerations. We are studying the response of spins to a weak transverse magnetic field, having in mind magnetic resonance measurements. Owing to the presence of a quite high spin-lattice (Korringa) relaxation rate of local moments, there are no saturation effects in EPR experiments in metals. As a result, the possible nonequilibrium of the longitudinal component of the spins and the spin-spin subsystem can be neglected, and the problem can be studied within the linear-response theory.

Averaging Eq. (5) with the help of Eq. (6) by the standard method,[13,14] we arrive at the following kinetic equations for the local nonequilibrium magnetization $S_i^-(t)$:

$$i\frac{\partial S_i^-(t)}{\partial t}=\omega_0 S_i^-(t)+\sum_j(W_{ij}+V_{ij})S_i^-(t) - \sum_j W_{ji}S_j^-(t). \tag{8}$$

Here the second and third terms on the right-hand side describe the efflux and influx of nonequilibrium magnetization because of the coupling between the spins. The corresponding transport coefficients

$$W_{ij}=W_{ji}=J_{ij}\langle S^z\rangle-i2J_{ij}^2\tau_{ij},\qquad V_{ij}=-iA_{ij}^2\tau_{ij} \tag{9}$$

are expressed in terms of equilibrium correlation functions

$$\tau_{ij}\approx\int dt\exp(-i\omega_i t)\frac{\langle S_i^+(t)\delta S_j^z(t)S_i^-(0)\delta S_j^z(0)\rangle}{\langle S_i^+S_i^-\rangle} \tag{10}$$

and the spin polarization $\langle S^z\rangle$ in the local field

$$\omega_i=\omega_0+\sum_j J_{ij}\langle S^z\rangle.$$

We are studying the para-phase, where $\omega_i\sim\omega_0$ and the site-number dependence of $\langle S^z\rangle$ is not important. The scale of W in Eq. (9) is evidently determined by the exchange-interaction energy. The quantity τ (10) is the exchange-fluctuation time of the spins. The equation (8) in the frequency representation is

$$\left[\omega-\omega_0-\sum_j(W_{ij}+V_{ij})\right]S_i^-(\omega)+\sum_j W_{ji}S_j^-(\omega)=0, \tag{11}$$

or, which is the same thing,

$$S_i^-+\sum_j S_j^- W_{ij}\Big/\left[\omega-\omega_0-\sum_k(W_{ik}+V_{ik})\right]=0. \tag{12}$$

The solution of Eq. (12) is a directly observable physical quantity, and it is this quantity that must be averaged over the random distribution of the magnetic impurities in the sample. There is no known rigorous procedure for performing this averaging exactly; here we confine ourselves to the simplest approximation, uncoupling the configurational averaging (designated below by the symbol $\langle\ldots\rangle_{\text{imp}}$) of the second term:

$$\langle S_j^- W_{ji}/(\ldots)\rangle_{\text{imp}}\approx\langle S_j^-\rangle_{\text{imp}}\langle W_{ji}/(\ldots)\rangle_{\text{imp}}. \tag{13}$$

After this, we can transform in Eq. (12) into momentum space:

$$\langle S_q^-\rangle_{\text{imp}}\left\{1+\left\langle\sum_j W_{ij}\exp(iqR_{ij})\Big/\left[\omega-\omega_0-\sum_k(W_{ik}+V_{ik})\right]\right\rangle_{\text{imp}}\right\}=0. \tag{14}$$

It should be noted that the approximation (13) and the subsequent equation (14) make sense, strictly speaking, only in the long-wavelength limit $qr_{\text{av}}\ll 1$.

The spin-excitation spectrum is determined by the poles ω_q of Eq. (14). They are found from the relation

$$\left\langle\frac{\omega_q-\omega_0-\Sigma_j\{W_{ij}[1-\exp(iqR_{ij})]+V_{ij}\}}{\omega_q-\omega_0-\Sigma_k(W_{ik}+V_{ik})}\right\rangle_{\text{imp}}=0. \tag{15}$$

For small momenta the difference $\omega_q-\omega_0$ is much smaller than the characteristic values of the exchange energy $W(R)$; this makes it possible to neglect the frequency de-

pendence in the denominator of the expression (15). Then the dispersion law for spin excitations is determined (for small momenta) by the relation

$$\omega_q \approx \omega_0 + \left\langle \frac{\Sigma_j\{W_{ij}[1-\exp(iqR)]+V_{ij}\}}{\Sigma_k W_{ik}} \right\rangle_{\text{imp}} \left\langle \frac{1}{\Sigma_k W_{ik}} \right\rangle_{\text{imp}}^{-1}. \tag{16}$$

For $q=0$ the contribution of exchange interactions in Eq. (16) vanishes owing to the isotropic character of the Hamiltonian (1); only uniform precession with frequency ω_0 and damping determined by the dipole interaction [the imaginary term $V(R)$ in Eq. (16)] remains.

The expression (16) is still too complicated because of the presence of the nontrivial configurational averaging. It simplifies significantly if the correlation function in (10) is factored and independent configurational averaging of the exchange-fluctuation time τ is performed. Factorization in Eq. (10) is not too rough an approximation, because we are concerned only with the paramagnetic phase, where local high-frequency fluctuations of different spins are incoherent. The presence of long-wavelength low-frequency spin-wave oscillations is insignificant for calculation of the correlation function in Eq. (10) because the corresponding phase volume is small; local correlation functions of the type $\langle S_i^\alpha(t)S_i^\alpha(0)\rangle$ decay exponentially in the para-phase over times determined by the short-range RKKY potential (2). Then

$$\tau_{ij} \approx \tau = \frac{S(S+1)}{3}\int_0^\infty dt \langle f(t)\rangle_{\text{imp}}^2, \qquad f(t) = \frac{3\langle S_i^+(t)S_i^-(0)\rangle}{S(S+1)}. \tag{17}$$

We note that in Eq. (17) the correlation function is evaluated at zero frequency; in the para-phase with $\langle S^z\rangle \ll 1$ the average thermodynamic values of the local fields ω_i in Eq. (10) are significantly smaller than the instantaneous values of the exchange fields. In other words, the correlation function in the integrand in the expression (10) decays, owing to exchange fluctuations, over times much shorter than ω_i^{-1}, and there is not enough time for precession around the local field to appear. The local spin fluctuation times τ (17) are calculated as in Refs. 12 and 15: The function $f(t)$ is assumed to be Gaussian and its decay is calculated by the method of moments:

$$f(t) \approx \exp\left[-\frac{S(S+1)}{3}\sum_R J^2(R)t^2\right]. \tag{18}$$

Then this function is averaged over the random positions of the impurities using the technique employed in the statistical theory of EPR lineshape:[16]

$$\langle f(t)\rangle_{\text{imp}} = \exp\left\{-n\int d^3R\left[1-\exp\left(-\frac{S(S+1)}{3}J^2(R)t^2\right)\right]\right\}. \tag{19}$$

Here $n=x/v_0$ is the spin density. The averaging method adopted gives for the frequency of spin fluctuations τ^{-1} a value of the order of the interaction of impurities at average distances. This is especially important in the case of short-range potentials, when averaging of the moments themselves of the correlation function $f(t)$ results in serious errors. For a specific form of the potential $J(R)$, determined by the sum of the expressions (2) and (3), preliminary averaging of fast oscillations of $J^2(R)$ and the integration in Eqs. (19) and (17) give

$$\tau = [3S(S+1)/2\pi]^{1/2}(xJ_0)^{-1}. \tag{20}$$

Now the expression for ω_q (16) assumes the form $\omega_q = \varepsilon_q - i(\gamma_q^{\text{ex}} + \gamma_q^d)$, where the dispersion ε_q and the damping γ_q are, respectively,

$$\varepsilon_q = \omega_q + \langle S^z\rangle \left\langle \varphi_i \sum_j J_{ij}[1-\exp(iqR_{ij})] \right\rangle_{\text{imp}} \Big/ \langle\varphi_i\rangle_{\text{imp}}, \tag{21}$$

$$\gamma_q^{\text{ex}} + \gamma_q^d = \tau\left\langle \varphi_i \sum_j 2J_{ij}^2[1-\exp(iqR_{ij})] + \varphi_i \sum_j A_{ij}^2 \right\rangle_{\text{imp}} \Big/ \langle\varphi_i\rangle_{\text{imp}}, \tag{22}$$

$$\varphi_i = \left(\sum_k J_{ik}^2\right)^{-1}.$$

Generally speaking, the denominator in the expression for φ_i also contains a term proportional to $\langle S^z\rangle$, but it was dropped because in the para-phase it is numerically small. The algorithm for calculating the averages in these formulas is as follows:

$$\langle\varphi_i\rangle_{\text{imp}} = \int_0^\infty dt \left\langle \exp\left(-\sum_k J_{ik}^2 t\right)\right\rangle_{\text{imp}} \approx \int dt\varphi(t), \tag{23}$$

$$\varphi(t) = \exp\left\{-n\int d^3R[1-\exp(-J^2(R)t)]\right\}, \tag{24}$$

$$\left\langle \varphi_i \sum_j \lambda_{ij}\right\rangle_{\text{imp}} = \int_0^\infty dt \left\langle \sum_j \lambda_{ij}\exp(-J_{ij}^2 t)\exp\left(-\sum_{k\neq j} J_{ik}^2 t\right)\right\rangle_{\text{imp}} \approx n\int d^3R\int_0^\infty dt\lambda(R)\exp[-J^2(R)t]\varphi(t). \tag{25}$$

The main approximation in the derivation of the relations (23)–(25) reduces to independent averaging over the positions of different impurities and ignoring "excluded volume" effects, a procedure admissible for low spin density.

We note that the description provided by the kinetic equation (8) is too "abbreviated" to contain the spectra of strongly coupled close exchange pairs. However, there are few such random pairs; moreover, under the averaging procedure adopted the contribution of short distances $r \ll r_{\text{av}}$ is insignificant and the long-wavelength kinetics is controlled by the interaction at average distances.

For specific calculations, using exchange potentials of the form (2) and (3), it is found that the damping γ^{ex} (22) of spin excitations due to exchange fluctuations and the correlation time τ (19) are mainly related with the short-range RKKY potential (2); conversely, the dispersion ε_q (21) for small q is determined virtually completely by the interaction (3) arising at long distances in the superconducting phase. This is a consequence of the different scales of the interaction region: the Fourier transform

$$J_s(q) \propto \Delta\chi_s(q) \propto (1+q^2\zeta^2)^{-1}$$

changes sharply over the scale $q \sim \zeta^{-1}$, while $J_n(q) \propto \chi_n(q)$ changes sharply over the scale $q \sim 2k_F$.

Substituting Eqs. (2) and (3) into Eqs. (21)–(25) gives ($q \ll \zeta^{-1}$)

$$\varepsilon_q = \omega_0 - Dq^2, \quad D \approx -x\langle S^z\rangle \delta\chi J_0 \zeta^2, \tag{26}$$

$$\gamma_q^{ex} = Dq^2, \quad D \approx x^{1/3}\langle S^z S^z\rangle^{1/2} J_0 a^2/4, \tag{27}$$

$$\gamma^d = \frac{3}{5}(2\pi)^{5/2} \frac{x\langle S^z S^z\rangle^{1/2}}{J_0} \left(\frac{g^2\beta^2}{v_0}\right)^2, \tag{28}$$

where a is the lattice constant. The coefficient D'' in the dispersion relation (26) is defined so that $D'' > 0$. (Note that $\langle S^z\rangle < 0$ if $\omega_0 > 0$). Comparing the spin rigidity coefficient D'' and the diffusion coefficient D' we find that the condition for the existence of coherent spin motion

$$\frac{D''}{D'} = -\left(\frac{x^{1/3}\zeta}{a}\right)^2 (4\delta\chi) \frac{\langle S^z\rangle}{\langle S^z S^z\rangle^{1/2}} \approx \left(\frac{\zeta}{r_{av}}\right)^2 \frac{\omega_0}{k_B T} \gg 1 \tag{29}$$

is easily satisfied in superconductors with long coherence length ζ (compared with r_{av}) in magnetic fields of the order of a kilogauss and at liquid-helium temperatures. For $q > \zeta^{-1}$ spin excitations are incoherent. The minus sign in the dispersion relation (26) reflects the antiferromagnetic nature of the potential (3); as the temperature decreases, the spectrum ε_q will soften and magnetic order will appear at nonzero wave vectors $q \sim \zeta^{-1}$, as predicted by Anderson and Suhl.[4] For small q the damping is determined by the dipole contribution γ^d; a "window" for spin-wave excitations

$$(\gamma^d/D'')^{1/2} < q < \zeta^{-1} \tag{30}$$

exists if the width of the magnon band is greater than the damping γ^d:

$$I = x\langle S^z\rangle(\delta\chi)J_0 > \gamma^d. \tag{31}$$

This condition can also be satisfied, since in metals exchange interactions are usually significantly stronger than the dipole-dipole interactions. Thus when the conditions (29) and (31) are satisfied, the long-wavelength spin dynamics has a magnon character. Of course, the contribution of these excitations to the thermodynamic parameters is negligible; the main fraction of the spectral density of spin fluctuations in the para-phase is incoherent and determined by the short-range RKKY potentials. Spin-waves can appear in the long-wavelength response of a superconductor with magnetic impurities.

We present below the experimental evidence, obtained in EPR studies performed in $La_{1-x}Er_x$ thin films, for spin-wave effects in a superconductor with magnetic impurities.

2. EXPERIMENT

2.1. Sample preparation

Films of $La_{1-x}Er_x$ with $x = 0.008$–0.02 and thickness $L = 2500$–5000 Å were prepared by separate thermal evaporation of the components from tantalum crucibles. The initial components were 99.88 wt.% lanthanum and 99.95 wt.% erbium. The substrates consisted of polished 15 $\times 4 \times 1$ mm^3 wafers of optically pure quartz. Just before each film is sputtered, a 100 Å thick sublayer of silicon monoxide was deposited on the substrate. The prepared film was coated, immediately after sputtering, with a protective layer of silicon monoxide with a thickness of the order of 3000 Å. The thickness of the films and the deposition rate were monitored with the help of a KIT-1 quartz thickness meter. The distance from the evaporator to the substrate was 15 cm. In order to decrease the content of volatile impurities in the initial material the crucibles were heated, prior to sputtering, for a long time at temperatures close to the evaporation temperatures. Sputtering was started after the evaporation chamber was outgassed and when a vacuum of $5 \cdot 10^{-7}$ Torr was obtained. The erbium evaporation rate required in order to obtain a given impurity concentration was established first. Next, the lanthanum evaporator was heated, and sputtering was conducted after the time required for establishing a constant evaporation rate had elapsed. In a number of cases the vacuum achieved could not be maintained during the sputtering process, and by the end of film deposition the pressure in the chamber rose to the value $5 \cdot 10^{-6}$ Torr. The film sputtering rate was $\sim 2 \cdot 10^3$ Å/min.

It is well known that lanthanum crystallizes as a mixture of two modifications with face-centered cubic (β-La) and hexagonal (α-La) lattices. X-ray structural analysis of the samples showed that the $La_{1-x}Er_x$ films have mainly ($\geqslant 70\%$) an fcc structure with lattice parameter $a \approx 5.304$ Å, corresponding to β-phase lanthanum. As far as the hexagonal α-phase is concerned, its presence is not revealed by EPR measurements, since the EPR signal of Er^{3+} ions in the polycrystalline α-phase is suppressed by the strong g-factor anisotropy in a hexagonal crystal field.[3]

Twenty samples with different erbium concentrations and thicknesses were prepared. For detailed studies six samples were chosen: C7 ($L = 2500$ Å, $x = 0.014$), C12 ($L = 2900$ Å, $x = 0.02$), H5 ($L = 3380$ Å, $x = 0.02$), C9 ($L = 4030$ Å, $x = 0.009$), N6 ($L = 4800$ Å, $x = 0.016$), N9 ($L = 4800$ Å, $x = 0.014$), which were prepared without destroying the vacuum, and H1 ($L = 2400$ Å, $x = 0.008$) from the group sputtered with a vacuum of $5 \cdot 10^{-6}$ Torr. The remaining samples were rejected, because they were prepared under variable conditions.

TABLE I. Technological and superconducting parameters of film and bulk samples [P—pressure in the evaporation chamber, ΔT_c—width of the superconducting transition, $\rho(T_c)$—residual resistivity].

Sample	P, Torr	L, Å	x	H_{c2}, Oe at 4.2 K	T_c, K	ΔT_c, K	$\rho(T_c)$, $\mu\Omega \cdot$ cm
H1	$5 \cdot 10^{-6}$	2400	0.008	3700	4.54	0.1	19.2
H5	$5 \cdot 10^{-7}$	3380	0.02	4900	4.80	0.05	4.8
M-1*	—	bulk	0.01	2400	5.45	—	4.4
M-1.75*	—	bulk	0.0175	1500	5.10	—	6.6

*The data for M-1 and M-1.75 were taken from Ref. 3.

2.2. Electric resistance and superconducting critical parameters

The resistivity ρ of the sample films was measured by the four-contact method using dc and clamped contacts. The critical temperature T_c and the upper critical field H_{c2} were determined at the points where the resistivity was half its value at the start of the superconducting transition. It was established that the residual resistivity and the critical temperature T_c are determined more by the sputtering conditions than the erbium concentration. This can be verified by analyzing the data given in Table I, where, as an example, data are presented for two typical representatives of groups of samples obtained with low ($5 \cdot 10^{-6}$ Torr) and high ($5 \cdot 10^{-7}$ Torr) vacuum. Table I also gives data from Ref. 3 for bulk samples with close erbium concentrations. Figure 1 displays the temperature dependence of the upper critical field H_{c2} for the sample H1 and H5 with the magnetic field oriented parallel to the plane of the film. The values of H_{c2} obtained for the perpendicular orientation were found to be 10% lower.

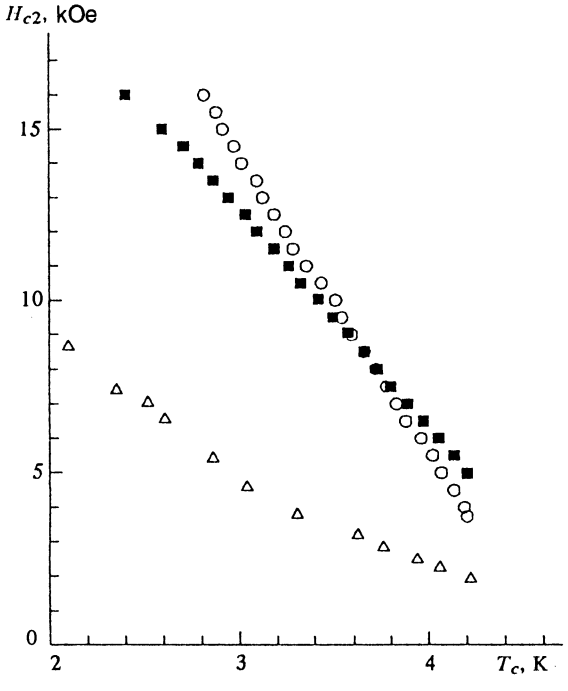

FIG. 1. Temperature dependences of the upper critical field with the magnetic field oriented parallel to the plane of the film for samples H1 (○) and H5 (■). Δ—data taken from Ref. 3 for a bulk sample with $x=0.02$.

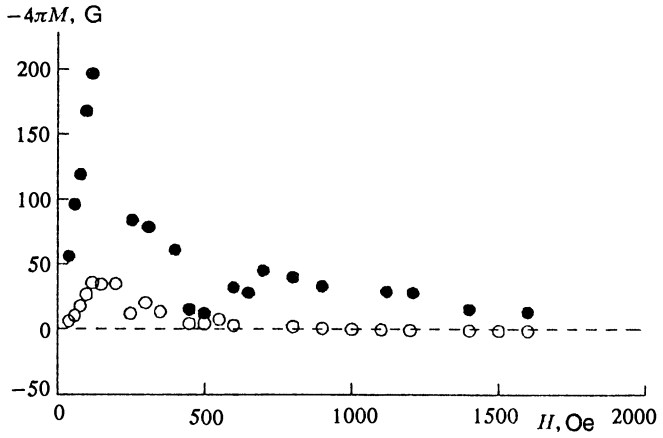

FIG. 2. Magnetization curves for sample H5 with the magnetic field oriented parallel to the sample plane with $T=4.2$ K (○) and $T=2.64$ K (●).

2.3. Magnetization in the superconducting state

Apparatus for measuring magnetic susceptibility by the Faraday method was used to obtain the magnetization curves. When the magnetic field was perpendicular to the plane of the samples the magnetization curves were found to be significantly irreversible: Increasing the magnetic field and then decreasing it to zero resulted in significant magnetic-flux trapping by the sample. In the case of parallel orientation the magnetization curves, as a rule, were found to be reversible. The form of the magnetization curves in the case of parallel orientation is displayed in Fig. 2 for one of the samples. In a superconductor, after reaching a maximum near the lower critical field H_{c1} the magnetic moment usually decreases continuously to H_{c2}. In our films, however, at low temperatures an anomaly is observed in the magnetization curves in fields $H_{c1} < H < H_{c2}$. This anomaly could be due to the presence of a large number of pinning centers in our samples. The same picture was observed previously in bulk lanthanum samples doped with erbium.[17]

2.4. Electron paramagnetic resonance

The EPR measurements were performed on a Brueker B-ER 418[s] spectrometer at 9.5 GHz at temperatures in the range 1.5–20 K. A rectangular cavity of the TE102 type was used for the experiments. The magnetic component H_1 of the microwave field was oriented perpendicular to the constant external magnetic field **H**. In order to study the angular dependence of the EPR spectrum, the angle ϑ between the field **H** and the plane of the sample was varied. In so doing, the alternating field H_1 always remained in the plane of the sample.

A single EPR line with g-factor close to 6.8 was observed for all experimental samples in the normal state.

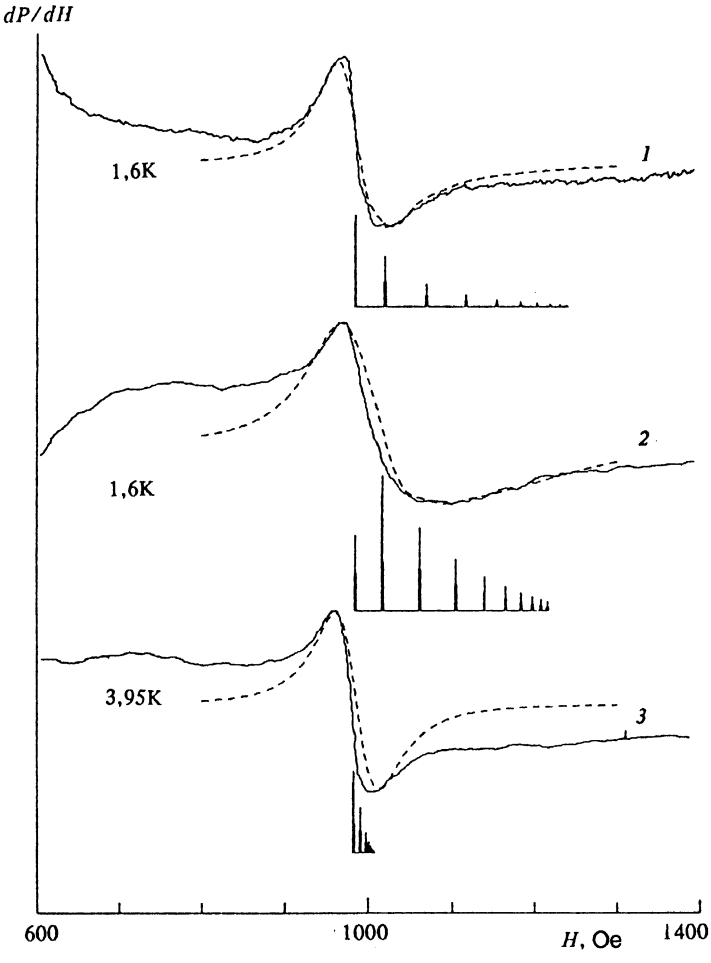

FIG. 3. EPR signal traces for sample N9 at different temperatures with the magnetic field oriented perpendicular (*1*) and parallel (*2* and *3*) to the plane of the film. The dashed lines are the computed spectra. The relative intensities and arrangement of the spin-wave resonance lines in the computed spectra are indicated beneath each spectrum.

The derivative of the absorbed power dP/dH was symmetric. As in the case of bulk samples,[3] the EPR line width increased linearly with temperature, evidently owing to the Korringa relaxation mechanism, and its concentration dependence at $T=\text{const}$ was found to be linear. The position and shape of the EPR line remained virtually unchanged as a function of the angle ϑ between the sample plane and the direction of the constant magnetic field.

As the samples passed into the superconducting state, the "zero line," reflecting the magnetic-field dependence of the nonresonance part of the surface impedance, behaved nonmonotonically and the noise level increased sharply, as is usually observed also in bulk superconductors. The most interesting observation in the superconducting state is the behavior of the line shape of the absorption signal. When the constant magnetic field is perpendicular to the plane of the films, the width and shape of the lines was virtually temperature independent. In a longitudinal magnetic field, however, the high-field peak of the resonance signal became sharply distorted as the temperature decreased. This effect is demonstrated by the EPR spectra of sample N9, which are displayed in Fig. 3. The same pattern is also observed for all other samples prepared in high vacuum (see Fig. 4, curves *1–4*). The degree of distortion of the high-field wing of the absorption signal depended on the temperature, on the thickness of the film, and on the erbium concentration in the sample. As for sample H1 (prepared in low vacuum), its absorption line shape remained virtually unchanged right down to the lowest temperatures (see Fig. 4, curve *5*).

3. ANALYSIS OF RESULTS

3.1. Normal state

At temperatures above T_c the derivative dP/dH for the resonance line with $g\approx 6.8$ was approximately symmetric in all of the experimental samples. In metals the shape of the EPR line is, generally speaking, asymmetric because the absorbed power is determined by a mixture of dispersion χ' and absorption χ'' curves, because as it penetrates into the metal the electromagnetic field changes in both amplitude and phase. As the film thickness decreases to values less than the skin depth δ, the contribution of χ' to the resonance signal decreases and the line becomes more symmetric. At temperatures close to T_c in our samples δ is of the order of 5 μm and $L/\delta\leqslant 0.1$. In this case the contribution of dispersion is negligible,[18] as is observed in experiment.

The value of the g-factor of the EPR signal and the absence of angular dependence of the spectrum indicate that the resonance signal is due to Er^{3+} ions in a cubic environment, and the ground state is the isotropic doublet Γ_7 with $g=6.77$, just as in bulk samples.[3] The dependence of the linewidth on the erbium concentration was found to

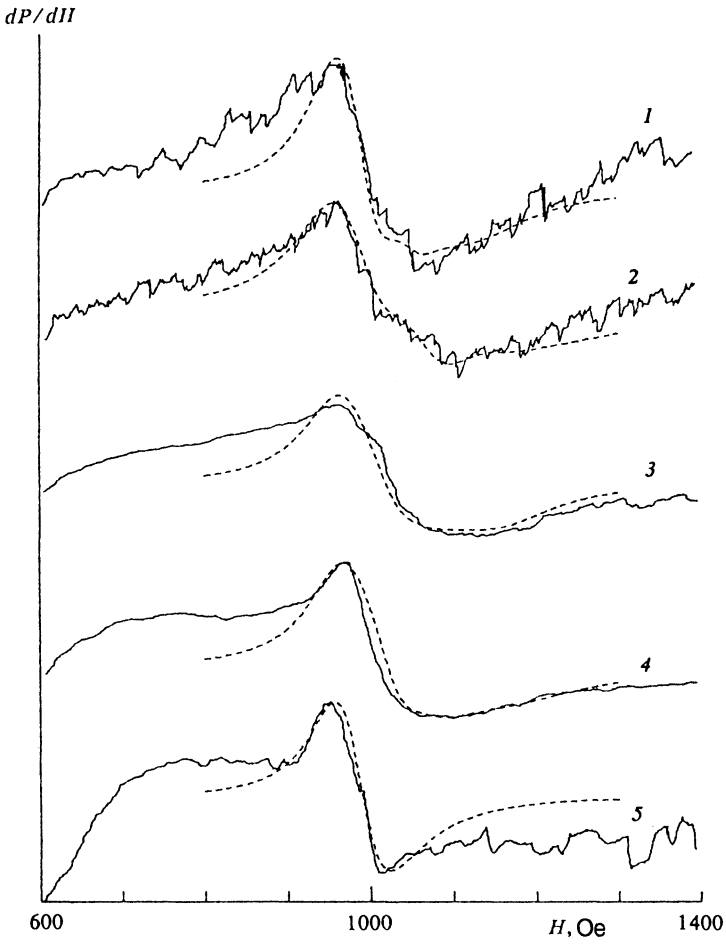

FIG. 4. EPR signal traces for samples C7 (*1*), H5 (*2*), C9 (*3*), N9 (*4*), H1 (*5*) at a temperature of 1.6 K with the magnetic field oriented parallel to the surface of the film. The dashed lines were computed.

be close to that observed previously in bulk samples. The latter observation confirmed further that the computed erbium content in the samples was close to the real one.

3.2. Superconducting state

It is well known that the EPR line shape changes significantly on going from the normal into the superconducting state. First, the amplitude of the alternating field in the superconductor decreases over a shorter distance than that over which the phase changes, and for this reason in a superconductor χ'' determines the absorbed power to a larger extent than does χ'.[19] Second, since the distribution of the constant magnetic field in the sample is nonuniform, the lineshape is a convolution of a homogeneously broadened line $\alpha\chi'+\chi''$ and the distribution function of the local fields in a unit cell of a vortical lattice.[3] We shall discuss the possibility that in our experiments this distorts the EPR lineshape.

Judging from the magnetization curves (Fig. 2) the anomalous shape of the EPR signal in the superconducting state with the external magnetic field oriented parallel to the sample plane cannot be due to the nonuniform distribution of the magnetic field in the vortex lattice. The point is that the degree of nonuniformity can be estimated from the values of the magnetization in the corresponding magnetic fields. An estimate of the amplitude of the variation of the magnetic field with the external magnetic field close in magnitude to the resonance field ($H_0 \approx 1000$ Oe) in accordance with the expression $H_{\max}-H_{\min}=1.46\cdot 4\pi M$ (Ref. 20) shows that, for example, $H_{\max}-H_{\min}\approx 50$ Oe at $T=2.6$ K (see Fig. 2). This is approximately 1.5 times smaller than the corresponding value obtained for bulk samples.[3] In bulk samples, however, the nonuniformities of the magnetic field in the vortex lattice result in inhomogeneous broadening of the resonance line but do not appreciably distort its shape. It is thus difficult to expect smaller nonuniformities of the magnetic field in the film samples to lead to the observed distortion of the EPR signal.

It remains to examine the possibility that the distortion of the signal shape in the high-field wing is caused by additional absorption of microwave energy by nonuniform spin-wave oscillations of the impurity magnetic moments. The energy of a spin-wave with momentum q_n is determined by the expression (26):

$$\varepsilon_q=g\beta H-D''q_n^2.$$

Hence it is evident that the magnon absorption lines for magnons with $q_n\neq 0$ should appear on the high-field wing of the principal resonance:

$$g\beta H_n=g\beta H_0+D''q_n^2,$$

where $g\beta H_0$ is the working frequency of the spectrometer.

Using the expression (26) for D and the more accurate expression for the Fourier transform $J_s(q)$, the resonance field H_n can be written as

$$H_n=H_0+H_0\frac{xJ_0\delta\chi(T)}{k_BT}\frac{S(S+1)}{3}\frac{(\zeta q_n)^2}{1+\zeta^2q_n^2}. \quad (32)$$

As far as the allowed values of the momentum q_n and the intensity of the spin-wave satellites are concerned, they are determined by the boundary conditions at the surface of the film and the degree of nonuniformity of the microwave field $H_1(z)$. In the case of symmetric bilateral excitation

$$H_1(z)=H_1\frac{\mathrm{ch}(k_0z)}{\mathrm{ch}(k_0L/2)},\quad k_0^2=i\frac{4\pi\omega}{c^2}\sigma(\omega), \quad (33)$$

where H_1 is the amplitude of the incident wave and $\sigma(\omega)$ is the complex conductivity at the resonance frequency. In order to find the power absorbed by the spins, it is necessary to solve the Bloch equation ($D=D'-iD''$)

$$\left(\omega-\omega_0+i\gamma^d-iD\frac{\partial^2}{\partial z^2}\right)S^-(\omega,z)=-\langle S^z\rangle H_1(z) \quad (34)$$

with the boundary condition

$$\frac{L}{S^-}\frac{\partial S^-}{\partial z}\Big|_{\pm L/2}=\mp\varepsilon. \quad (35)$$

The dimensionless phenomenological parameter $0\leqslant\varepsilon<\infty$ on the right-hand side of Eq. (35) accounts for the loss of nonequilibrium magnetization at the boundary. In this case the main physical reason for this loss is unavoidable lattice deformations in the surface layer, since the thermal expansion coefficient of the lanthanum film differs strongly from that of the silicon monoxide layers coating the film on both sides. Lattice distortions result in admixing of the upper-lying states of the erbium ion to the ground-state doublet Γ_7 in a crystal field and they shift the g-factor. Such random shifts of the g-factor of erbium ions at the surface detune the ions from resonance, and this can be represented phenomenologically as suppression of response due to effective surface relaxation (35). Large values of ε actually correspond physically to complete detuning of resonance at the boundary: $S^-(\pm L/2)=0$, and they presuppose strong nonuniform lattice distortions near the film boundary.

The field dependence of the absorbed power can be found by determining the function $S^-(\omega,z)$ from Eqs. (34) and (35) and integrating it with the field $H_1(z)$ (33):

$$P(H)=|\langle S^z\rangle|\left(\frac{H_1}{\mathrm{ch}\,\varkappa_0}\right)^2\mathrm{Im}\frac{Z'}{2D(k^2-k_0^2)}, \quad (36)$$

$$Z'=1+\frac{\mathrm{sh}\,2\varkappa_0}{2\varkappa_0}-\frac{2}{\varkappa^2-\varkappa_0^2}\frac{\varepsilon\,\mathrm{ch}\,\varkappa_0+2\varkappa_0\,\mathrm{sh}\,\varkappa_0}{\varepsilon\,\mathrm{ch}\,\varkappa+2\varkappa\,\mathrm{sh}\,\varkappa}$$
$$\times(\varkappa\,\mathrm{sh}\,\varkappa\,\mathrm{ch}\,\varkappa_0-\varkappa_0\,\mathrm{sh}\,\varkappa_0\,\mathrm{ch}\,\varkappa), \quad (37)$$

where

$$\varkappa_0=k_0L/2,\quad \varkappa=kL/2,$$
$$k=[(g\beta H_0-g\beta H+i\gamma^d)/D]^{1/2}. \quad (38)$$

We now consider the position of the satellite absorption lines [determined by the poles of the expression (36)] and their intensity in two opposite situations:

a) In the case of "free" boundaries, $\varepsilon=0$, the poles of the expression (36) are determined by the roots of the equation

$$\mathrm{sh}(kL/2)=0 \quad (39)$$

and the absorption is due to excitations with momenta

$$q_n=\pi n/L,\quad n=0,2,4,...$$

and is given by the relation

$$P=\sum_{n=0,2..}I_n\,\mathrm{Im}(g\beta H_0-\varepsilon_q+i\gamma_q)^{-1}. \quad (40)$$

The relative intensity of the lines of different harmonics is

$$I_n/I_0=(2-\delta_{n,0})[1+(q_n/|k_0|)^2]^{-2}, \quad (41)$$

where I_0 is the intensity of the principal line with $n=0$.

b) In the other limiting case, $\varepsilon=\infty$, the poles of the expression (36) are determined by the zeros of the function $\cosh(kL/2)$ and only the odd harmonics $q_n=\pi n/L$, where $n=1,3,5,...$, participate in absorption; their intensities are given by

$$I_n=2|\langle S_z\rangle|H_1^2\left(\frac{2}{|k_0|L}\right)^2\frac{(q_n/|k_0|)^2}{[1+(q_n/|k_0|)^2]^2}. \quad (42)$$

In this case satellite lines appear even in a uniform microwave field, $k_0=0$. However, they are not very noticeable because the intensity decreases very rapidly with increasing n, since $I_n\propto 1/q_n^2$ when $k_0=0$. The effect of surface relaxation becomes important for $k_0\neq 0$; it results, as is seen by comparing Eqs. (42) and (41), in "transfer" of intensity from the principal line to lines corresponding to magnons with $q\sim k_0$.

The inverse penetration depth k_0(33) of the microwave field is determined by the conductivity $\sigma(\omega)$. It is well known that in type-II superconductors with a vortex lattice the conductivity is anisotropic and depends strongly on the relative orientation of the constant magnetic field $\mathbf{H}$ and the electric component $\mathbf{E}_1$ of the microwave field in the sample. This is connected with the modulation of the order parameter Δ of the microwave field.[21] Thus, if $\mathbf{E}_1\|\mathbf{H}$ (in our case this corresponds to $\mathbf{H}$ parallel to the film), then at low frequencies $\omega\ll\Delta$ the conductivity is determined mainly by the imaginary part: $\sigma(\omega)\approx-i\pi\sigma_n(\Delta/\omega)\times\tanh(\Delta/2T)$, where σ_n is the normal-metal conductivity. In this case the effective skin depth

$$\delta_\parallel=k_0^{-1}\approx\lambda(0)[\Delta(0)/\Delta\mathrm{th}(\Delta/2T)]^{1/2} \quad (43)$$

is small compared to L, and the intensity of the first spin-wave satellite line is comparable to the intensity of the principal absorption line. [Here $\lambda(0)$ is the London screening length at $T=0$.] If, however, the constant field is perpendicular to the plane of the film, then $\mathbf{E}_1\perp\mathbf{H}$ and the conductivity $\sigma(\omega)$ is small:[22]

$$\sigma(\omega)\approx-i(\sigma_nH_{c2}^\perp/H_0).$$

In this case the effective penetration depth of the microwave field is greater than in a longitudinal field:

$$\delta_\perp = k_0^{-1} \approx \delta_\parallel \, [\pi H \Delta \, \mathrm{th}(\Delta/2T)/\omega H_{c2}^\perp]^{1/2} \approx 2\delta_\parallel \, . \tag{44}$$

Thus the distribution of the microwave field in a superconducting film is most nonuniform in the case of a longitudinal field. For this reason it is in this geometry that spin-wave effects can be expected to be strongest. To calculate the spectra with the help of the expressions (32), (41), and (42) it is necessary to know the parameters of the experimental samples: the coherence length ζ and the inverse penetration depth k_0 of the microwave field.

The coherence length ζ can be roughly estimated from measurements of the critical field H_{c2} of bulk samples.[3] In accordance to the interpretation given in Ref. 3 for the β-lanthanum results, the extrapolated value $H_{c2}(T=0) \approx 8$ kOe (see Fig. 1), whence follows the estimate $\zeta(0) \approx 200$ Å.

According to Eqs. (43) and (44), to estimate k_0 with magnetic fields parallel and perpendicular to the plane of the film it is necessary to know the London penetration depth λ of the magnetic field in the sample and the upper critical field H_{c2} in the perpendicular orientation of the magnetic field. As indicated in Sec. 2.2, the latter quantity is 10% smaller than the parallel-orientation H_{c2}, whose temperature dependence is displayed in Fig. 1. The quantity λ can be estimated from our experimental data. First, this can be done using the diamagnetic susceptibility of samples in fields less than H_{c1}. Extrapolation of the λ obtained in this manner gave to low temperatures $\lambda(0) \approx 700$ Å. Second, the penetration depth can be estimated by comparing the values of the thermodynamic critical fields for a film in parallel orientation and for a bulk sample.[22] For the sample H5 this gives $\lambda(0) \approx 500$ Å. Finally, the penetration depth can be estimated directly from the decrease in the EPR signal strength in a transition from the normal into the superconducting state. The obtained $\lambda(0)$ ranged from 650 to 700 Å. Thus the possible values of $\lambda(0)$ range from 500 to 700 Å.

In the calculations it was assumed that ζ and Δ vary with temperature as

$$\zeta = \zeta(0)(1-T/T_c)^{-1/2}, \quad \Delta = \Delta(0)(1-T/T_c)^{1/2}. \tag{45}$$

Calculations of the spectra performed for the two limiting cases of nonequilibrium magnetization at the boundary showed that in the case a ($\varepsilon=0$) the experimentally observed spectra cannot be satisfactorily explained for any reasonable variations of the London length λ and the coherence length ζ about the values indicated above. This concerns especially the case of thin samples. For strong surface relaxation (case b), however, qualitative agreement with the EPR data on the changes in the line-shape with decreasing temperature and rotation of the sample is obtained for films of different thickness. In this case the spectra of all samples can be satisfactorily described by choosing the same values $\zeta(0)=200$ Å, $\lambda(0)=600$ Å, and $\Delta(0)=1.75T_c$. The main adjustable parameter was

$$A = J_0 S(S+1)\delta\chi/3, \tag{46}$$

which entered in the expression (32) and was fitted for each spectrum individually. A much better fit of the spectra was obtained by allowing for the increase in the damping of spin waves with increasing wave vector. For this, the wave-vector dependence of the linewidth of the satellite was incorporated into the computational program in the form

$$\Delta H = \Delta H_0 + \alpha q_n^2.$$

The value of α was chosen for all spectra for each sample and it was unchanged by the choice of A. Analysis showed that the values of the damping constant $\gamma_q^{ex} = Dq^2$ (27) associated with thermal fluctuations of the exchange field are patently inadequate for describing the spectra. Thus for $x=0.01$ and $T=1.6$ K the ratio of the additional broadening of the satellite line to its shift is, according to (29), $D'q^2/D''q^2 \approx 0.01$, which is several times less than required. In reality, the spread in the thickness over the sample, which leads to a momentum uncertainty $\delta q_n \sim \delta L/L$ and to a corresponding broadening of the satellite by an amount $\sim (Dq_n^2)(2\delta L/L)$, apparently makes the main contribution to this behavior of the broadening. Thus it can be expected that in the case when the evaporator and the substrate are separated by a distance ~ 15 cm and the size of the substrate is ~ 1.5 cm the film thickness will differ from the center to the periphery by the amount $\delta L/L \sim 0.05$. The computed values of α varied from sample to sample within the range $2 \cdot 10^{-9}$–10^{-10} Oe $\cdot$ cm^2, which corresponds to broadening of the first satellite lines by several percent of their shift.

Thus, when surface relaxation is strong, the temperature and orientational behavior of the spectra of all experimental samples can be accounted for by varying the single adjustable parameter A. Examples of such fits are displayed in Figs. 3 and 4. Figure 3 illustrates the correspondence of the computed and experimental spectra when the temperature and orientational dependences are described. Figure 4 displays an example of the description of parallel-orientation experimental spectra at the lowest temperature for four samples of different thickness (spectra *1–4*). Bearing in mind superposition of the resonance experimental spectra on the nonmonotonic "zero" line (the latter is associated with the field dependence of the surface impedance of the superconductor), the figure shows that the computed spectra account well for the basic features of the experimental spectra in the region of resonance absorption. Thus all information about the dependence of the spectra on the temperature and orientation is contained in the parameter A (46), whose temperature dependence is shown in Fig. 5. This figure actually reflects the $\delta\chi(T)$ dependence [see the expression (46)]. The solid line in the figure corresponds to the temperature dependence $\delta\chi(T)$ for a BCS superconductor[23] and $J_0 S(S+1)/3 \approx 40$ K. Since the constant J_0 is determined by the spatial integral of the RKKY interaction, the quantity A can be expressed in terms of the paramagnetic Curie temperature Θ as

$$A \approx \Theta(x)\Delta\chi/x, \tag{47}$$

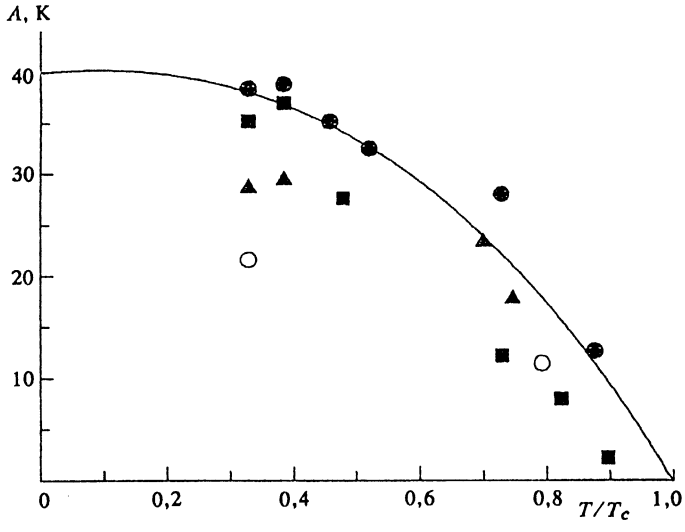

FIG. 5. The parameter A as a function of the reduced temperature for samples N9 (■), C9 (●), H5 (▲), and C7 (○). The solid line corresponds to $\delta\chi(T)$ of a BCS superconductor.

whence follows the estimate $\Theta(x=1)\approx 40$ K. The similar agreement with the paramagnetic temperature for pure erbium ($\Theta_{Er}=41$ K) seems accidental, since it is difficult to expect $\delta\chi$ in the sample with paramagnetic impurity in a magnetic field to come close to unity. Knowing A it is possible to determine the spin rigidity coefficient D'' from Eq. (26): at $T=1.6$ K with $x=0.01$, $D''=0.04$ cm^2/sec.

As far as the low-vacuum sample H1 is concerned, in our opinion no appreciable spin-wave effects are observed in it because, as is evident from Table I, the residual resistivity of this sample is approximately four times that of the high-vacuum sample H5. This means that the mean-free path l of conduction electrons and correspondingly the rigidity coefficient of the spin-wave are four times smaller, since $D''\sim\zeta^2$, and the coherence length of the dirty superconductor is $\zeta\sim(\zeta_0 l)^{1/2}$. This conjecture is confirmed in Fig. 4 (spectrum 5), which shows the computed spectrum with the same interaction constant J_0 as for sample H5 but with a shorter coherence length $\zeta=100$ Å.

Thus we can assert confidently on the basis of the observed anomalous lineshape and its dependence on the temperature and thickness of the samples and on the geometry of the experiment that we have observed spin-wave effects.

In summary, it has been shown above that coherent spin-wave excitations of a system of paramagnetic impurities are present in a superconductor placed in a magnetic field. These excitations are a consequence of the strongly nonlocal character of the spin susceptibility of the superconductor, and their observation is a direct confirmation of the long-range exchange interaction between impurities, which plays an important role in the theory of magnetic superconductors. The short-range "normal" part of the RKKY potential and the dipole interactions as well as the disordering of impurities do not destroy the spin-wave motion, if the coherence length greatly exceeds the average distance between magnetic impurities. The spin-waves studied in the paramagnetic system, where the external-field-induced magnetization plays the role of an order parameter, is completely analogous to magnons in a paramagnetic subsystem of nuclear spins, caused in magnetics by the Suhl-Nakamura interaction,[24] and spin-waves in a paramagnetic Fermi liquid in a magnetic field, as observed in EPR experiments.[25]

[1)]Preliminary results were published in Ref. 2.

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