# Electron spin resonance on localized magnetic states in the super-conducting system La-Er

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Electron spin resonance on the localized magnetic moments of Er in La in the normal and superconducting state is investigated. A strong narrowing of the resonance line following the superconducting transition is observed. We separate the contribution made to the ESR lines by the inhomogeneous distribution of the magnetic field H in a sample whose state is mixed at  $H_{c1} \le H \le H_{c2}$ . The results are interpreted on the basis of the premise that the conditions for the "electron bottleneck" in indirect exchange interactions become enhanced on going to the superconducting state.

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# INTRODUCTION

The influence of paramagnetic impurities on the properties of superconductors is one of the timely and intensively investigated problems of superconductivity physics. Observation of electron spin resonance absorption in type-II superconductors<sup>(11)</sup> has added the ESR method to the number of physical methods used to study this problem. Being a direct detector of the formation of an electronic localized magnetic state, this method yields information on a number of important properties of a superconductor doped with a magnetic impurity. All the possible applications of ESR to superconductors have not yet been clarified completely, but the following can already be noted at present.

The ESR method makes it possible to measure directly the exchange interaction of conduction electrons and the localized states, to obtain detailed information on the spin scattering of the conduction electrons individually for a given sort of impurity, to investigate the collective spin-density oscillations produced at sufficiently high concentrations of the paramagnetic impurity (the "electron bottleneck" effect<sup>[2]</sup>). Finally, and apparently most significantly, ESR makes it possible to investigate directly in the superconducting phase the character and the strength of the interactions between the electronic localized states (in particular, the indirect exchange interaction via the conduction electrons, of the Ruderman-Kittel-Kasuya-Yosida type), and consequently to investigate the problem of the coexistence of magnetic order and superconductivity. The latter is also of great practical interest when it comes to the development of new superconducting materials with high critical fields, inasmuch as introduction of a magnetic impurity can lead under certain conditions<sup>[3]</sup> to an increase of the paramagnetic limit of the superconductor.

Both in the first paper<sup>[1]</sup> and in the subsequent foreign paper<sup>[6]</sup> on ESR in superconductors, the investigations were carried out on intermetallic compounds with small admixtures of gadolinium. The choice of intermetallic compounds for the investigations in<sup>[1,4-6]</sup> was dictated by the need for obtaining relatively high critical parameters for the samples doped with paramagnetic impurities. In the investigations of such samples by the ESR

method, however, there is always the danger of the appearance of parasitic signals, which mask the true effect. This may be caused either by the increased chemical activity of these compounds, or by the difficulty of attaining the necessary homogeneity and stoichiometry. It is therefore of interest to carry out the ESR investigations on paramagnetic impurities in a "pure" metallic type-II superconductor. In this respect, metallic lanthanum is very attractive, in view of the good solubilities of rare-earth metals in it. The study of lanthanum is undoubtedly also of great fundamental interest, since the question of the nature of its superconductivity still remains open (see, e.g.,<sup>[7]</sup>). Although the critical parameters of pure single-component superconductors are usually lower than the corresponding values for compounds, this difficulty can be circumvented by choosing a paramagnetic impurity with a large g factor, for example erbium, which in addition suppresses the superconductivity of the lanthanum much less than gadolinium.<sup>[8]</sup> Thus, the measurements can be performed up to high concentrations of the paramagnetic impurities, when the interaction between them becomes substantial. It is known that metallic lanthanum crystallizes in the form of a mixture of two modifications with hexagonal  $(\alpha-La)$  and face-centered cubic  $(\beta-La)$  lattices. However, the choice of erbium as the paramagnetic impurity makes it possible to disregard the presence of the hexagonal phase of the lanthanum, since the strong anisotropy of the g factor of the  $Er^{3+}$  ion in a hexagonal crystal makes the ESR signal unobservable in the  $\alpha$  phase in a polycrystalline sample.

We present here the results of investigations of ESR on the localized moments of erbium in metallic lanthanum with relatively high concentrations of the magnetic impurity (up to 6 at.%), and discuss some possibilities and singularities of ESR in superconductors. Preliminary results were published in<sup>[9,10]</sup>.

# I. MAGNETIC AND SUPERCONDUCTING PROPERTIES OF SAMPLES

1) Preparation and analysis of the samples. The samples were prepared from the initial components (lanthanum and erbium purities 99.88 and 99.5 wt.%,



FIG. 1. Dependence of the upper critical field on the temperature for samples with different erbium contents:  $\circ -0.5$  at. %,  $\Delta -1.5$  at. % at. %,  $\bullet -2$  at. %.

respectively) using an induction furnace in tantalum and niobium crucibles in an atmosphere of pure helium, followed by quenching in a stream of cold helium. The samples for the measurements were cut in the form of plates measuring  $15 \times 4 \times 0.5$  mm. An x-ray analysis has shown that they consist predominantly of the  $\beta$  phase of lanthanum with a lattice parameter a = 5.3 Å. Data on the phase composition of certain samples, together with the results of measurements of the resistance, are presented below ( $c_{\rm Er}$  is the concentration of the erbium in the sample):

c <sub>Er</sub> , at. %:	0.5	1.0	1.5	1.75
Amount of $\beta$ -phase:	95	60	95	80
$\rho_{300 \text{ K}} / \rho_{6 \text{ K}}$ :	16.5	11.5	•••	8.5
$ \rho_{6 \text{ K}}, \mu\Omega\text{-cm} $	3.6	4.4	• • •	6.6

2) Measurements of the critical temperature  $T_c$  and of the upper fields  $H_{c2}$ . The measurements of  $T_c$  in a zero field were carried out by the induction method. The character of the transition curves for samples containing noticeable amounts of the  $\alpha$  phase (more than 10%) made it possible to estimate the ratio of the phase. The volume of the phase in the samples agreed with the results obtained from the x-ray analysis. The decrease of the critical temperature from its value in pure lanthanum for the  $\beta$  phase (in our case,  $T_c = 5.9 \pm 0.1$  K) for 1% of erbium impurity was  $\Delta T_c = 0.4$  K.

For a number of samples, we have measured also the upper critical field  $H_{c2}$  by determining the change of the dc resistance. The results are shown in Fig. 1 ( $H_{c2}$  was measured by determining the start of the transition to the normal state<sup>30</sup>). As seen from the figure,  $H_{c2}$  depends weakly and nonmonotonically on the impurity concentration. It can be assumed that this dependence is determined by the competing influences of the orbital and spin contributions of the magnetic impurity. An



FIG. 2. Magnetization curve (solid line) and resistance transition from the superconducting to the normal state (dashed) for a sample containing 0.5 at. % Er: T=4.2 K.



FIG. 3. Temperature dependence of the susceptibility of a sample La +0.5 at. % Er.

actual calculation, however, carried out using our data on the resistance and the dependence of  $T_c$  on the erbium concentration does not agree quantitatively with this assumption. Furthermore, the plot of  $H_{c2}$  against temperature shows clearly a break that is obviously due to the fact that the sample has two phases. Extrapolation of the curves makes it possible to divide the obtained diagrams into two parts corresponding to the  $\alpha$  and  $\beta$ phases. (It is known that the  $\beta$  phase has a higher value of  $T_c$ .<sup>[11]</sup>) The relatively large values of  $H_{c2}$  for the  $\alpha$ phase may be due to the fact that this phase is finely dispersed and to the strong mechanical stresses in this phase.

3) Magnetization of samples in the superconducting state. From the phase diagram (Fig. 1) it is seen that correct measurements of the magnetization of the  $\beta$  phase can be carried out only at temperatures above  $T_c$  of the  $\alpha$  phase. The results, shown in Fig. 2, of the measurements at T = 4.2 K of the magnetization of a superconducting sample containing 0.5 at.% Er make it possible to determine the Ginzburg-Landau coefficients for this sample:  $k_1 = H_{c2} / \sqrt{2} H_{c1} = 3.2$  and  $k_2 = 3.45$  ( $k_2$  is calculated from the values of  $(dM/dH)_{H_{c2}}$ ).

4) Susceptibility of samples in the normal state. The magnetic moments were measured with a strong magnetometer<sup>[12]</sup> in the temperature range 4.5-300 K in magnetic fields up to 80 kOe, on samples containing 0.5 and 1.5 at.% Er.<sup>4</sup> These samples were practically pure (95%)  $\beta$  phase. At low temperatures, the susceptibility (Fig. 3) deviates from the Curie law. These deviations make it possible to determine the Stark splitting of the levels of the Er<sup>3+</sup> in the cubic field of the lanthanum. The appropriate calculation procedure is given in Appendix I. In all cases when the calculation results could be reconciled with the susceptibility measurements, the ground state turned out to be the doublet  $\Gamma_7$ , and the distances between the ground and nearest excited levels ranged from 13 to 22 K (see Table II in Appendix I).

#### **II. ESR MEASUREMENTS. EXPERIMENTAL RESULTS**

The ESR measurements were performed with a BER218s modulation spectrometer in the 3-cm band and in the temperature interval 1.6-25 K. Besides the usual limit imposed on the real sensitivity of the ESR spectrometer for a metal as a result of the presence of the skin layer in the superconducting state, it is necessary to take into account also the increased noise to the motion of the vortices and a strong dependence of the surface impedance of the superconductor on the strength of the constant magnetic field. The latter circumstance is particularly significant near the transition temperature, where the ESR measurements are very



FIG. 4. ESR signal of La+0.5 at.% Er sample in the superconducting state at T=2.1 K. The points were calculated with allowance for the vortex lattice with  $\Delta H=35$  Oe and  $H_v-H_c$ = 84 Oe.

difficult. The minimum erbium concentration that could be observed in the superconducting state was 0.5 at.%. The maximum concentration was 6 at.%.

1) Line shape. The ESR line shape in the normal state had the asymmetric form usually observed for the bulk metal and was well described by a superposition of Lorentzian dispersion and absorption curves. The line width  $\Delta H$  and the g factor were determined in accordance with<sup>[13]</sup>.

Figure 4 shows a plot of the derivative of the resonant-absorption power of a sample in the superconducting state. The line shape, especially for small erbium concentrations, differs substantially from a pure Lorentzian, principally as a result of the inhomogeneous distribution of the magnetic field inside the sample, caused by the appearance of the vortices. The shape of the resonance signal in the superconducting phase with allowance for the vortex lattice is given in Appendix II. As seen from Fig. 4, the calculated curve describes well the experimental spectrum. By reconciling the theoretical curves with the experimental data it is possible to obtain the true line width  $\Delta H$  and the difference between the maximum  $H_v$  and the minimum  $H_c$  fields in a sample that is in the mixed state at  $H_{cl} \leq H \leq H_{c2}$ .

2) The g factor. In the normal state, the g factor was equal to  $6.80 \pm 0.05$ . The proximity of the observed value of the g factor to the theoretical one for the doublet  $\Gamma_7$  (for which g = 6.77 in a cubic field when account is taken of the intermediate coupling) is evidence that the ground state in our case is the doublet  $\Gamma_7$ , as follows also from the susceptibility. This is an unequivocal confirmation of the fact that the resonance signal is due to the cubic phase of the lanthanum.



FIG. 5. Temperature dependence of the line width  $\Delta H$  for the following samples: O-La+0.5 at. % Er;  $\Delta-La+1$  at. % Er;  $\bullet-La+2$  at. % Er.



FIG. 6. Dependence of the line width and of the difference of the limiting values of the magnetic field in the vortex lattice for the sample La+0.5 at.% Er:  $\bullet -\Delta H$ ;  $\bullet -H_v - H_c$ .

For the superconducting phase, when calculating the line shape by the scheme described in Appendix II, it was assumed that the magnetic field at the center of the vortex is equal to the external magnetic field. Actually, as shown, for example, by measurements of NMR on single-crystal niobium samples of high quality, this field can noticeably exceed the external field.<sup>[14]</sup> Therefore the values of the g factors, calculated from the spectrum distorted by the vortex lattice, may differ from the true values. For this reason, we do not analyze in this paper the g factors in the superconducting region.

3) Temperature and concentration dependence of the line width. Figure 5 shows the dependence of  $\Delta H$  on T for several samples in the investigated temperature range, while Fig. 6 shows in greater detail the low-temperature of this dependence for one of the samples. For the superconducting region, the difference between the limiting values of the magnetic fields in the sample is indicated.

These figures show the following: a) at temperatures below 14 K in the normal state, the line width depends linearly on the temperature and can be described by the functional relation  $\Delta H = a + bT$ ; b) above 14 K, the temperature dependence of  $\Delta H$  is no longer linear; c) the coefficients a and b depend on the concentration (Table I lists the values of these coefficients for four samples that cover the entire range of investigated concentrations); d) on going to the superconducting state, the line width decreases sharply, and the jump is proportional to the erbium concentration; e)  $\Delta H$  is constant in the superconducting state.<sup>50</sup>

Figure 7 shows the concentration dependence of  $\Delta H$  in the normal and superconducting states for all the investigated samples.

## **III. DISCUSSION OF RESULTS**

1) Normal state. That part of the ESR line width which depends linearly on the temperature is due to thermal fluctuations of the exchange interaction of the localized f electrons with the conduction electrons (the

TABLE I.

°Er, at.%	a, Oe	b, Oe/deg	<sup>c</sup> Er, at.%	a, Oe	b, Oe/deg
0.5	26±3	5.3±0.4	2.0	75±8	3.7±0.4
1.0	38±4	5.3±0.4	3.0	102±10	3.7±0.4



Korringa mechanism). If we introduce for the lower doublet  $\Gamma_7$  an effective spin S = 1/2, then the Hamiltonian of the exchange interaction can be written in the form  $g(g_L - 1)JSs/g_L$  (s is the spin of the conduction electrons), and the exchange integral J is determined from the temperature slope of the line width<sup>[15]</sup>

$$b = \frac{\pi g k_B}{\mu_B g_{L^2}} (g_L - 1)^2 J^2 N^2, \tag{1}$$

where  $\mu_B$  is the Bohr magneton,  $k_B$  is the Boltzmann constant,  $g_L$  is the Lande factor, and N is the density of states of the conduction electrons with given spin orientation on the Fermi surface. As seen from expression (1), the measured quantity is the product of the exchange integral by the density of states. If we use for the state density the value<sup>6</sup>  $N = 2 \text{ eV}^{-1} \text{ atom}^{-1} \text{ spin}^{-1}$  known from measurements of the heat capacity,<sup>[16]</sup> then in the case of the sample with the smallest erbium concentration, when the effect due to the collective oscillations of the spin density of the localized moments and conduction electrons are each significant, we obtain from (1) the value J = 0.13 eV.

It is of interest to compare this quantity with the data obtained from the dependence of  $T_c$  on the concentration of the paramagnetic impurity. The presented ESR and susceptibility results show that at temperatures  $\sim T_c$  we can neglect the scattering of the electrons by the excited levels of erbium. To obtain J in the presence of the crystal-field effects we can therefore use the formula of Abrikosov and Gor'kov,<sup>[7]</sup> which we express in terms of the effective spin:

$$\Delta T_c = -\frac{\pi^2}{8k_B} cNJ^2 S(S+1), \qquad (2)$$

where  $\Delta T_c$  is the change of  $T_c$  due to the introduction of the magnetic impurity. This yields a value J = 0.04 eV, which differs noticeably from the corresponding value obtained from ESR. The observed discrepancy can be connected with the fact that in lanthanum the conduction electrons belong mainly to the s and d bands, and the contributions from these bands to the spin relaxation rate (the Korringa and Overhauser relaxations) and to the heat capacity may not be fully equivalent.

The proximity of the excited level to the ground level causes the temperature dependence of the line width to deviate from linearity at temperatures ~14 K, as a result of the relaxation process of the Orbach-Aminov type.<sup>[18]</sup> Allowance for this mechanism leads to the temperature dependences of the line width, shown by the solid lines in Fig. 6. It is interesting to note that all

three sets of crystal-field parameters obtained in the calculation of the susceptibility make the same contribution to the relaxation.

The principal part of the ESR line width, which does not depend on temperature, can be ascribed to magnetic dipole-dipole interactions. A contribution to the line width can also be made by the distortion of the spatial distribution of the charge density of the conduction electrons, due to lattice defects. They lead to the appearance of a low-symmetry contribution to the crystal field and cause the g factor to be shifted as a result of mixing of excited states with the wave functions of the doublet  $\Gamma_7$ . It is important to emphasize here that whereas the scatter of the g factor is due to the mutual influence of the paramagnetic impurities, the line width will increase with increasing concentration, just as in the dipole-dipole broadening mechanism. On the other hand, the mutual distortion of the spin density of the conduction electrons by paramagnetic impurities leads to the known indirect Ruderman-Kittel-Kasuya-Yosida (RKKY) exchange, which suppresses the indicated two-particle mechanisms of ESR line broadening.

The value of the RKKY exchange integral can be easily estimated in the free-electron approximation (see, e.g.,  $^{(19)}$ ):

$$J_{\rm RKKY} = \frac{9\pi Z^2 J_0^2}{2E_F} \varphi(k_F R), \quad \varphi(x) = \frac{\sin x - x \cos x}{x^*}$$

where Z is the number of conduction electrons per atom,  $E_F$  is the Fermi energy, and  $k_F$  is the corresponding wave vector. To this end it is necessary to calculate the value of the exchange integral  $J_0$ , using the density of states of the conduction electrons  $N_0 = 0.33 \text{ eV}^{-1} \text{atom}^{-1}$ spin<sup>-1</sup> calculated in the free-electron model for lanthanum. From the ESR data we obtain  $J_0 = 0.08 \text{ eV}$ , and from the dependence of  $T_c$  on the erbium concentration we get  $J_0 = 0.1 \text{ eV}$ . The values of  $J_0$  are in fair agreement.<sup>70</sup> The use of  $J_0 = 0.08 \text{ eV}$  yields for the nearest neighbors in the lattice the value  $J_{RKKY} = 2 \text{ K}$ , which is much higher than the energy of the magnetic dipole-dipole interaction (for which in our case the estimate yields ~ 0.05 K). This means that for an exchange narrowing of the ESR line takes place.

Finally, we call attention to the existence of a weak concentration dependence of the temperature slope of the line width on Fig. 6. This dependence points to the possibility of realizing the conditions of the "electron bottleneck" in the normal phase, when the rate of establishment of the equilibrium between the localized moments and the spin system of the electrons exceeds the rate of relaxation of the latter to other degrees of freedom. In this case, the width of the ESR line is determined by some effective relaxation time, and the Korringa mechanism does not manifest itself. Allowance for this circumstance causes the quantitative estimates of  $J_0$  (and accordingly  $J_{RKKY}$ ) from the observed ESR line to be actually somewhat undervalued. At the same time, the procedure itself for extracting the value of the exchange integral from the ESR data is justified because, as shown earlier, [20] the changes of the temperature slope of the line width under the conditions of the "electron bottleneck" are small in the case of spin systems with substantially different g factors.

2) Superconducting state. On going to the superconducting phase, an additional cause for the broadening of the ESR line appears, because of the inhomogeneous distribution of the magnetic field in the sample at  $H_{cl} < H$  $< H_{c2}$ . The experimental observed deviation of the shape of the ESR line from Lorentzian in the phase transition agrees with the assumption that a regular vortex structure is produced in our samples. The theoretical ESR line shape can be calculated by introducing the distribution function of the magnetic field in the superconductor (see Appendix II). The value  $H_v - H_c = 70$  Oe obtained by this method for a sample containing 0.5 at.% Er (see Fig. 4) can be compared with the result obtained from measurements of the magnetic moment M of the superconductor. For a triangular vortex lattice we have  $H_v - H_c = -1.46 \cdot 4\pi M$ ,<sup>[14]</sup> which in our case yields 75 Oe, in good agreement with the value given above.

The most interesting feature of the results of the measurements in the superconducting phase is the sharp decrease of  $\Delta H$  immediately following the transition, in contrast to the usually observed increase of the relaxation rate in strongly diluted systems. Let us consider the possible causes of this effect. As a result of the appearance of coherence effects in the scattering of the conduction electrons and of the increase of their state density in the superconductor, the rate of the Korringa relaxation increases sharply near  $T_c$ . At the same time, according to Maki<sup>[21]</sup> the rate of exchange scattering of the conduction electrons below  $T_c$  (the Overhauser relaxation) also increases, and the spin-orbit scattering (the spin-lattice relaxation of the conduction electrons) decreases sharply. This circumstance enhances the conditions of the "electron bottleneck," and leads to a narrowing of the ESR line. This effect, however, can explain only in part the observed narrowing of the line, since the change of  $\Delta H$  at high impurity concentrations exceeds the contribution due to the Korringa mechanism.

The substantial dependence of the jump of the ESR line width on the impurity concentration in the phase transition points to the important role that can be played by two-particle interactions in this effect. If the spinflip frequency due to the Korringa relaxation is increased so much that it exceeds the scatter of the resonance frequencies, then the paramagnetic ion "sees" already the average local field due to the magnetic dipole interactions. In this case the corresponding contribution to the line width decreases by a factor  $S/\langle S_z \rangle$ , whereas the change in the Korringa relaxation does not appear in the ESR line width when the conditions of the "electron bottleneck" are realized.

Let us consider also the possible increase of the exchange narrowing of the ESR line in the superconductor. If the effective RKKY radius in the normal metal is limited to the spin-orbit scattering length, then the sharp increase of this length on going to the superconducting state can lead to the observed effect. In addition, the change of the RKKY exchange interaction on going to the superconducting state is determined by the correlations of the conduction electrons with opposite spin orientations, as a result of which the average spin susceptibility tends to zero. The local spin polarization of the electrons near the paramagnetic impurity is then cancelled out as a result of the indicated correlations over much larger distances, on the order of the coherence length.<sup>[22]</sup> The corresponding change of the exchange integral is

$$\Delta J_{\mathbf{R}\mathbf{K}\mathbf{K}\mathbf{Y}} = \frac{9\pi^2 Z^2 J_0^2}{E_F^2 (2k_F R)^2} k_B T \sum_{\varphi} \frac{\Delta^2}{\Delta^2 + \omega^2} \sin^2 k_F R \exp\left(-\frac{2\sqrt[3]{\Delta^2 + \omega^2}}{v_F} R\right),$$

where  $\hbar \omega = \pi k_B T (2n+1)$ ,  $\Delta$  is the order parameter in the superconductor, and  $v_F$  is the electron velocity on the Fermi surface.

An estimate of this contribution by the method of moments shows that the change of the exchange narrowing of the ESR line is small, since the rate of reorientation of the localized spins depends on  $J^2_{\rm RKKY}$ . It must be noted, however, that the exchange field of spins of one orientation on a paramagnetic ion in a superconductor, at low impurity concentration, can greatly exceed, as a result of the long-range action, the corresponding value for the normal metal.

We note in conclusion that none of the mechanisms discussed above can explain fully the observed discontinuity of the line width and the effect is apparently due to their joint action.

### APPENDIX I

The Hamiltonian of a crystal field of cubic symmetry for the 4f ion can be expressed in terms of the equivalent operators in the form

$$\mathcal{H}=B_{\bullet}F(4)\frac{O_{\bullet}}{F(4)}+B_{\bullet}F(6)\frac{O_{\bullet}}{F(6)},$$

where  $O_4 = O_4^0 + 5O_4^4$  and  $O_6 = O_6^0 - 21O_6^4$  are the equivalent operators,  $B_4$  and  $B_6$  are fourth- and sixth-order crystal-parameters, and F(4) and F(6) are certain positive coefficients.

If in analogy with<sup>[23]</sup> we introduce two new quantities X and x:

$$B_{4}F(4) = Wx, \quad B_{6}F(6) = W(1 - |x|),$$

and include the Zeeman energy in the Hamiltonian, then we can write

$$\mathcal{H} = W \left\{ x \frac{O_4}{F(4)} + (1 - |x|) \frac{O_6}{F(6)} \right\} - g_L \mu_B H M.$$

The average magnetic moment along the field direction is then

$$\langle J_z \rangle = \frac{\sum\limits_{n}^{\infty} (\sum\limits_{M} |C_{nM}|^2 M) \exp\left(-E_n/kT\right)}{\sum\limits_{n}^{\infty} \exp\left(-E_n/kT\right)}.$$

Here  $g_L$  is the Lande factor, M is the component of the

TABLE	п.
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x	w	ð, K
-0.4 -0.1 0.4	0.3 0.1 0.2	$\begin{array}{c} \delta^{\Gamma_{\theta}} = 13,  \delta^{\Gamma_{\theta}} = 18\\ \delta^{\Gamma_{\theta}} = 22\\ \delta^{\Gamma_{\theta}} = 22 \end{array}$

angular-momentum operator in the field direction,  $E_n$  denotes the eigenvalues of  $\mathcal{S}_n^2$ , and  $C_{nM}$  are the coefficients of their eigenfunctions.

The diagonalization of the sixteenth-order matrix and the calculation of  $\langle J_z \rangle$  were carried out with a computer. The criterion in the choice of the crystal-field parameter was the quantity

$$d = \frac{(\langle J_z \rangle_{\text{theor}} \langle J_z \rangle_{\text{exp.}})^2}{\langle J_z \rangle^2}.$$

The calculations were performed only for measurements made in weak magnetic fields, since the samples were polycrystals, and the Stark splitting of the multiplet j = 15/2 should contain quadruplets  $\Gamma_8$ , which have an angular dependence. The optimal parameters obtained in the calculation, together with the distances  $\delta$  to the nearest excited level, are given in Table II.

# **APPENDIX II**

The distribution of the probabilities of encountering a given magnetic field in a triangular vortex lattice can be approximated by the analytic function<sup>[14]</sup>

$$f(x) = \begin{cases} 0.837 - 0.500 \ln(-x), & 0.08 < x < 0\\ 0.236 - 0.576 \ln x, & 0 < x < 0.92 \end{cases}$$

Here  $x = (H - H_s)/(H_v - H_c)$ ,  $H_v$  and  $H_c$  are the maximum and minimum fields in the lattice,  $H_s$  is the field at the saddle point of the unit cell of the vortex lattice. If the shape of the homogeneously broadened ESR line in the metal is determined by the Lorentz line

$$I_{\text{norm}}(H) = \frac{1}{\pi} \frac{\Delta H + H}{(\Delta H)^2 + H^2},$$

where  $\Delta H$  is the half-width of the line at half-height and H is the magnetic field relative to the center, then the ESR line shape with allowance for the broadening due to the vortex lattice is determined by the convolution

$$I_{sup}(H) = \int I_{norm}(H_1 - H) f\left(\frac{H_1 - H_s}{H_c - H_c}\right) dH_1.$$

The calculated curve shown in Fig. 4 corresponds to the derivative  $dI_{sup}(H)/dH$ .

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- <sup>3)</sup>It must be noted that the values of  $H_{c2}$  determined from the curve of the surface impedance at  $10^{10}$  Hz are noticeably smaller than the values obtained in dc measurements.
- <sup>4)</sup>The measurements were performed at the International Laboratory for Strong Magnetic Fields and Low Temperatures in Wroclaw.
- <sup>5)</sup>This is in contrast to the published preliminary data, <sup>[9]</sup> where the figures showed the total line width, including also the contribution due to the inhomogeneous distribution of the magnetic field.

- <sup>6)</sup>The estimate of N from  $dH_{c2}/dT$  and the residual resistance yields a value that is of the same order of magnitude.
- <sup>7)</sup>In connection with the already noted possible non-equivalence of the data on the heat capacity and ESR in the presence of two conduction bands in lanthanum, the values of  $J_0$  calculated within the framework of the free-electron model are also of interest in themselves. Together with the values of J previously obtained by using the experimental results on the heat capacity, they can be regarded as limiting values for the real exchange integral in the investigated system.
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