Spin-wave resonance in a superconductor with paramagnetic impurities

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An anomalous distortion of the ESR lineshape has been observed in thin $La_{1-x}Er_x$ films with a transition to the superconducting state. This effect is explained by the appearance of collective spin-wave excitations of paramagnetic impurities in a magnetic field.

1. When a sample undergoes a transition to the superconducting state, the radius of the RKKY interaction between the magnetic impurities becomes anomalously large, on the order of the coherence length ξ (Refs. 1 and 2). In view of this circumstance, we can expect that the long-wave excitations of the magnetic impurities in an external magnetic field do not have a diffusion nature, but rather a spin-wave nature[3] similar to that of the excitations in a paramagnetic Fermi liquid in a magnetic field[4] or in a system of nuclear spins with a Shul–Nakamura interaction.[5] We have carried out ESR studies of thin superconducting $La_{1-x}Er_x$ films in order to measure the spin-wave oscillations of the magnetic impurities.

2. The $La_{1-x}Er_x$ films with $x = 0.008$–0.016 and thickness $L = 3000$–5000 Å were synthesized by a separate thermal evaporation of the components from a tantalum crucible in a vacuum of 5×10^7 Torr. The ESR measurements were carried out at a frequency of 9.41 GHz at temperatures of 1.5–15 K. The same samples were used to measure the critical fields H_{c2} and the magnetization curves.

In the normal state, we observed the ESR line of the Er^{3+} ions with a g factor of ~ 6.8. A change in the angle between the plane of the sample and the direction of the static magnetic field had virtually no effect on the position and shape of the ESR line (Fig. 1a). As the samples underwent a transition to the superconducting state, the "zero line," which reflects the dependence of the nonresonant part of the surface impedance on the magnetic field, behaved in a nonmonotonic manner and the noise level rose rapidly, as is usually the case in bulk superconductors (Fig. 1b).

The behavior of the lineshape of the absorption signal in the superconducting state deserves more attention. When the magnetic field is perpendicular to the film surface, the width and shape of the ESR line is nearly independent of the temperature for all the test samples, as in the case of bulk $La_{1-x}Er_x$ (Ref. 2). In a longitudinal magnetic field, however, the shape of the resonance signal became drastically distorted in the superconducting state as the temperature was lowered (see Fig. 1b). The distortion was such that the signal began to resemble a spectrum of a superposition of two lines: the basic line with a constant field H_0 and a broader auxiliary line with a resonance at $H_0' > H_0$. An analysis showed that the shift $\delta H = H_0' - H_0$ depends on the composition (as $\delta H \sim x$), on the film thickness (Fig. 2), and on the temperature (Fig.

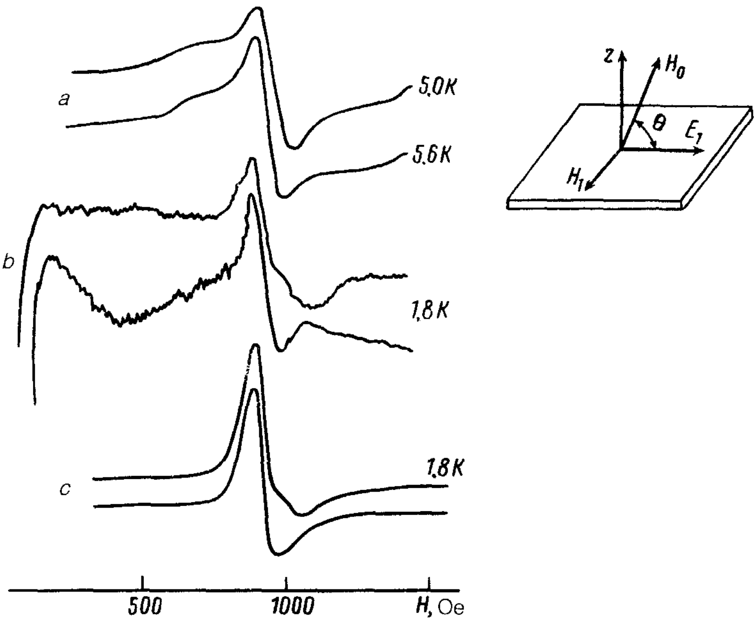

FIG. 1. The ESR spectra of Er^{3+} ions in the samples of a $La_{1-x}Er_x$ film with $x = 0.016$ and $L = 4800$ Å in the normal state (*a*) and in the superconducting state (*b*); *c*—calculated spectra. In each pair, the upper spectra correspond to a parallel orientation of the plane of the sample relative to the static magnetic field and the lower spectra correspond to the perpendicular orientation.

3), and that the width of the auxiliary line increases with increasing shift.

3. It is unlikely that the unusual behavior of the ESR spectrum in the lanthanum films, which we have observed, could be attributed to a magnetic field distribution in the vortical lattice different from that in a bulk sample. First, the amplitudes of the

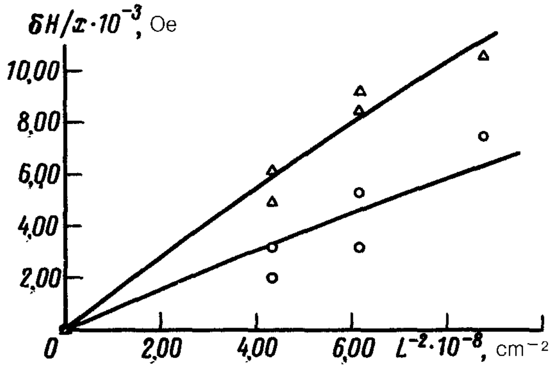

FIG. 2. Shift of the satellite line versus the thickness of the sample (Δ—T = 1.8 K; 0—T = 2.5 K). Solid lines—The result of a fit top expression (1).

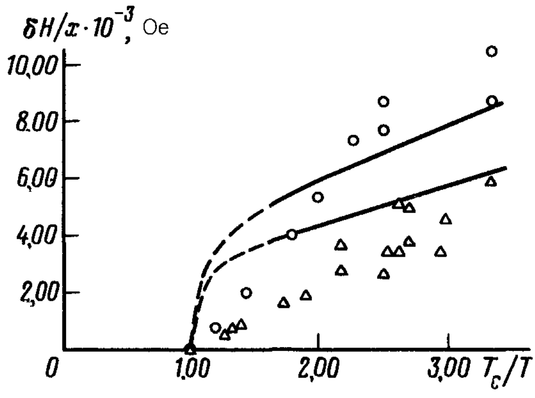

FIG. 3. Temperature dependences of the shift of the satellite line (0—$L = 4030$ Å; Δ—$L = 4800$ Å). Lines—The result of a calculation.

variation of the magnetic field inside the film, $A = (H_{max} - H_{min})/2$, and of the London penetration depth λ, which we estimated from the measurements of the critical fields and the diamagnetic moment of the films, turned out to be on the order of 50 Oe and 800 Å, respectively. These values are essentially the same as those obtained for the bulk $La_{1-x}Er_x$ samples.[2] Secondly, it was shown in Ref. 2 that the magnetic field nonuniformity in the vortical lattice gives rise to a nonuniform broadening of the resonance line, but does not noticeably distort its shape, $dP(H)/dH$, in contradiction of the results of our experiment.

4. We believe that the shape of the signal in the high-field wing is distorted because of the additional absorption of the microwave energy by the nonuniform spin-wave oscillations of the extrinsic magnetic moments, as was predicted in Ref. 3. In the normal phase the spin excitations of the impurities are, as usual, of a diffusion nature. In the superconducting state, however, the number of spins $N \approx x\xi^3/V_0$, which fall into the region of the RKKY interaction, is anomalously large even when the erbium impurity concentration is low, since the coherence length of lanthanum is $\xi \approx 200$ Å (V_0 is the volume per atom). If the magnetic field strength in this case is such that $\langle S_Z \rangle \sqrt{N} \gg 1$, then the long-wavelength spin dynamics will be of a magnon nature, since the fluctuation corrections to the excitation energy and to the magnon damping with $q < \xi^{-1}$ are small to the extent that $1/\langle S_Z \rangle \sqrt{N}$ is satisfied. The resonance field H_n for the excitation with the wave vector $Q_n = 2\pi n/L$ can be represented in the form[3]

$$H_n = H_0 + H_0 \frac{|\theta(x)| \delta\chi(T)}{T} \frac{(\xi q_n)^2}{[1 + (\xi q_n)^2]}, \tag{1}$$

where $\theta(x) \sim x$; this quantity is on the order of the paramagnetic Curie temperature, and $\delta_\chi(T) = (\chi_p - \chi_s)/\chi_p \lesssim 1$ characterizes the decrease of the static spin susceptibility χ_s of the conduction electrons in the singlet BCS state compared with the Pauli susceptibility χ_p.

5. The intensity of the satellite lines with $q_n \neq 0$ is determined by the degree of nonuniformity of the microwave field $H_1(z)$ in the film. In the case of a symmetric bilateral excitation, we have

$$H_1(z) = H_1 \frac{\cos(kz)}{\cos(kL/2)}, \quad k^2 = i\frac{4\pi\omega}{c^2}\sigma(\omega), \tag{2}$$

where H_1 is the amplitude of the incident wave, and $\sigma(\omega)$ is the admittance at the resonance frequency. It is easy to show that the relative absorption rate by a spin-wave with $q = q_n$ is

$$\frac{P_n}{P_0} = \frac{2}{[1 + (q/|k|)^2]^2}, \quad n = 1, 2, \ldots . \tag{3}$$

In type-II superconductors with a vortex lattice the conductivity $\sigma(\omega)$ is known to be an anisotropic quantity which strongly depends on the relative direction of the static magnetic field **H** and the electrical component $\mathbf{E}_1$ of the microwave field in the sample, which is a consequence of the modulation of the order parameter Δ by the microwave field.[6] If, for example, $\mathbf{E}_1 \parallel \mathbf{H}$ (in our case, this corresponds to the orientation of **H** along the length of the film; see the inset in Fig. 1), then at low frequencies $\omega \ll \Delta$ the condition is determined largely by the imaginary part of $\sigma(\omega) \sim -i\pi\sigma_n\Delta/\omega$ and $k^2 \approx 1/\lambda^2$. In this case, the effective depth of the skin layer $\delta_\parallel \sim \lambda$ is small in comparison with L, and the intensity of the first spin-wave satellite line is comparable to the intensity of the main absorption line. If, on the other hand, the static field is perpendicular to the film, then $\mathbf{E}_1 \perp \mathbf{H}$, and the conductivity $\sigma(\omega)$ is small: $\sigma(\omega) \approx \sigma_n H_{c2}^{\perp}/H_0$ (Ref. 7), where σ_n is the conductivity of the normal metal. In this case, the effective penetration depth of the microwave field $\delta_\perp \approx \delta_\parallel (\pi\Delta H_0/\omega H_{c2}^{\perp})^{1/2} \sim 2\delta_\parallel$ is greater and the spin waves are essentially not excited.

6. The spectral calculations carried out using expressions (1) and (3) showed a qualitative agreement with the ESR data for films of various thicknesses, based on the change in the line shape as the temperature was lowered and the sample rotated. Allowance for the second and subsequent satellites did not materially change the calculated spectrum, but only flattened the auxiliary line, bringing it closer to the spectrum observed by us. Figure 1c shows a calculated spectrum with allowance for five satellites for a sample with $L = 4800$ Å, $x = 0.016$, and $T_c = 5$ K for $\delta_\parallel = 600$ Å and $\delta_\perp = 1200$ Å. Since the experimental spectrum is superimposed on the nonmonotonic zero line, its behavior is described well by the theoretical spectrum. The best description of the experimental data on the basis of the curve for the shift δH of the first satellite versus the thickness of the sample and the use of expression (1) (see Fig. 2) was obtained when $\theta(x) = 110x$ (K). Compared with the known value $\theta = 41$ K for pure erbium, this value seems quite reasonable. The coefficient of spin rigidity is $D = g\beta H_0(|\theta|\delta_\chi(T)/T)\xi^2/\hbar \approx 0.1$ cm^2 s at $T = 2$ K and $x = 0.01$. The discrepancy between the theoretical temperature dependence of δH and that measured experimentally (see Fig. 3) is apparently attributable to the use of the $\delta_\chi(T)$ curve for a pure superconductor.

7. We have thus shown experimentally that a superconductor in an external magnetic field has weakly damping spin waves in the paramagnetic impurities. The presence of these waves is a direct proof of the existence of a strongly nonlocal RKKY interaction in a BCS superconductor.

[1]P. W. Anderson and H. Suhl, Phys. Rev. **116**, 898 (1959).

[2]N. E. Aleksevskiĭ, I. A. Garifullin, B. I. Kochelaev, and É. G. Kharakhash'yan, Zh. Eksp. Teor. Fiz. **72**, 1523 (1977) [Sov. Phys. JETP **45**, 795 (1977)].

[3]G. G. Khaliullin and B. I. Kochelaev, Phys. Lett. **106A**, 318 (1984).

[4]S. Schultz and G. Dunifer, Phys. Rev. Lett. **18**, 283 (1967).

[5]P. G. de Gennes, P. A. Pinkus, F. Hurtmann-Boutron, and M. Vinter, Phys. Rev. **129**, 1105 (1963).

[6]C. Caroli and K. Maki, Phys. Rev. **159**, 306 (1967).

[7]E. Linton, *Superconductivity*, Russ. transl., Mir, Moscow, (1971).

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