# Coherence effects in the spin relaxation of lanthanum-strontium ceramics

V. E. Kataev, E. F. Kukovitskiĭ, Yu. I. Talanov, and G. B. Teĭtel'baum
*Physicotechnical Institute, Kazan' Branch of the Academy of Sciences of the USSR, Kazan'*

(Submitted 21 September 1988)
Pis'ma Zh. Eksp.Teor. Fiz. **48**, No. 8, 433–436 (25 October 1988)

Study of the critical behavior of the ESR line of the gadolinium ions used for spin probing of a lanthanum-strontium superconductor has made it possible to determine the magnitude of the gap and its temperature dependence in the charge-excitation spectrum.

It has now been established that high-temperature superconductivity occurs as a result of charged-quasiparticle pairing, although the actual pairing mechanism is not yet known. An important step in understanding this mechanism is the determination of the magnitude and the temperature dependence of the gap in the charge excitation spectrum. The contribution of research on nuclear spin relaxation to the study of ordinary superconductors is known.[1,2] In high-$T_c$ superconductors the critical anomalies of the nuclear spin relaxation, which are associated with the establishment of coherence of paired electrons, are barely discernable,[3] which accounts for the lower analytical capabilities of the method. The study of electron spin relaxation in the superconducting state is, however, fundamentally difficult because of the singlet nature of the pairing. We have therefore studied the coherence effects near the superconducting transition in a lanthanum-strontium system by means of ESR, using $Gd^{3+}$ ions which were implanted in this compound as spin probes in a concentration small enough not to lower the temperature $T_c$.

  

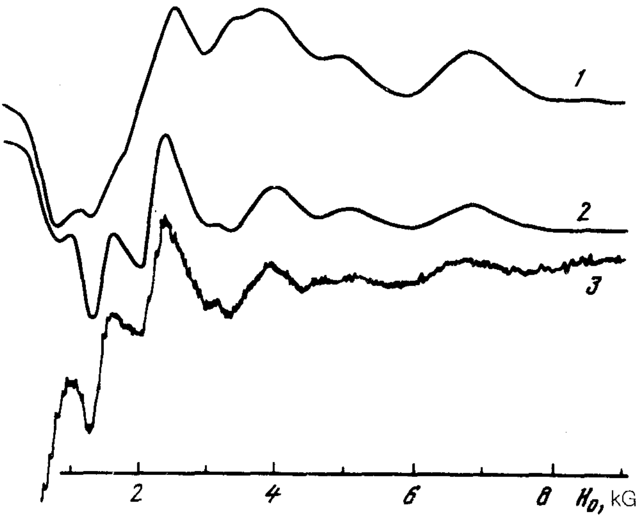


FIG. 1. Fine structure of the ESR spectrum of $Gd^{3+}$ ion in $La_{1.98}Gd_{0.02}CuO_4$ (1) and $(La_{1.98}Gd_{0.02})_{1.85}Sr_{0.15}$ $CuO_4$ (2) at $T = 41$ K; the signal is the same in the superconducting state at $T = 25$ K (3).

To determine the effect of strontium doping on ESR of the gadolinium ions, we studied the compound $La_{1.98}Gd_{0.02}CuO_4$, in addition to the system $(La_{1.98}Gd_{0.02})_{1.85}$ $Sr_{0.15}CuO_4$. All measurements were carried out at a frequency 9.4 GHz over a broad temperature interval 6–300 K. To increase the signal-to-noise ratio and to reduce the inhomogeneous broadening due to the finite penetration depth of the magnetic field, we ground the original ceramics into a powder. The powder particles (crystallites with dimensions on the order of several microns) were set in paraffin.

The ESR spectrum of the $La_{1.98}Gd_{0.02}CuO_4$ sample, consists of seven resonance absorption lines (Fig. 1), which are the fine structure of the ESR spectrum of the $Gd^{3+}$ ion. The distance between the outermost components is ~7 kG and the individual widths of the single lines lie in the interval 300–900 G. The positions of these lines do not depend on the temperature and their widths increase slightly as the temperature decreases and at 60–80 K they peak smoothly in the interval ~80–100 K. The characteristic variation of the width in this region is < 100 G. This variation apparently is attributable to the temperature dependence of the scatter of the local magnetic fields by the gadolinium ions.

Doping of the system under study with strontium leads to a narrowing of single lines to 100–700 G, while keeping the width of the fine structure of the ESR spectrum of $Gd^{3+}$ constant (Fig. 1). It also leads to the appearance of the temperature dependence of the width of the individual components of the spectrum from 0.3 to 1.15 G/K (Fig. 2). We have observed an ESR signal even in the superconducting state. At $T < T_c = 35$ K the structure of the $Gd^{3+}$ spectrum remains the same, but the widths of the individual lines behave critically in a manner characteristic of the phenomena associated with the appearance of coherence in the electronic system.

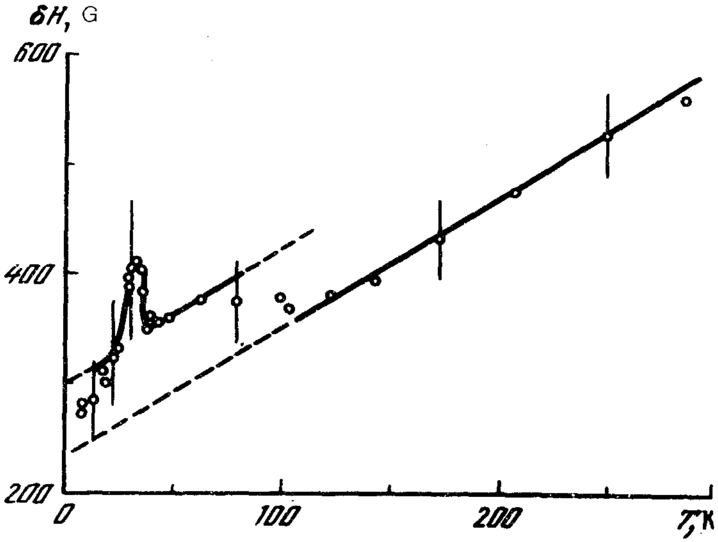


FIG. 2. Temperature dependence of the width of one of the components of the fine structure of the $Gd^{3+}$ spectrum in $(La_{1.98}Gd_{0.02})_{1.85}Sr_{0.15}CuO_4$ ($T_c = 35$). Solid line—The theoretical curve constructed for $2\Delta_0/kT_c = 6$, $\alpha = 0.25$ [Eq. (3)].

In interpreting the results we should take into account that the $Gd^{3+}$ ion, which is in the $S$ state ($S = 7/2$), in the crystal structure of the lanthanum ceramic has a local tetragonal symmetry (if the slight orthorhombic nature is ignored) and its ground state in the magnetic field **H** can be described, in a first approximation, by a Hamiltonian[4]

$$\delta H_n = 4\pi[S(S+1) - M(M-1)](\rho_F J)^2\, kT. \qquad (1)$$

Only the axial anisotropy is taken into account here. Analytic expressions for the angular dependence of the resonant fields of the allowed transitions with $\Delta M = \pm 1$ can be obtained only for the limiting values of the fine structure constant $D(D \gg H$ or $D \ll H)$. Judging from the observable width of the fine-structure spectrum, the most difficult situation, in terms of the calculation of the energy levels and wave functions of the $Gd^{3+}$ ion, of the intermediate fields, which requires an exact diagonalization of the energy matrix, is realized in this case. For this reason, in contrast with the limiting cases, the energy states of the $Gd^{3+}$ ion are mixed. This situation may cause a possible confusion in the sequence of the resonance magnetic-field transitions. To estimate the constant $D$, we have therefore numerically modeled the spectrum of the polycrystal under study. The results of this numerical simulation of the spectrum are in agreement with the observation at $D = 3400$ G.

The modification of the ESR spectrum of $Gd^{3+}$ upon strontium doping of the system is associated with the appearance of the current carriers. A decrease in the width of the individual components can be attributed to the RKKY exchange contraction[5] and the appearance of a linear temperature dependence in their behavior is associated with the Korringa relaxation of the $Gd^{3+}$ spins through the current carriers. It should be pointed out that the temperature slope depends on the spin projection M (Ref. 5):

$$\mathcal{H} = D[S_z^2 - {}^1/_3 S(S+1)] + g\beta \mathbf{HS}. \tag{2}$$

Here $\rho_F$ is the state density of the current carriers at the Fermi level, and $J$ is the integral of their exchange interaction with the magnetic moments of $Gd^{3+}$. A similar difference in the temperature dependences of the widths of the individual components has been observed experimentally.

Figure 2 shows the temperature dependence of the width of the component of the spectrum, third from the left (Fig. 1), which has the largest value of $d(\delta H)/dT \approx 1.15$ G/K. A larger measurement error at $T < T_c$ stems from the appearance of zero-line drift due to the field dependence of the surface impedance. The width of the component which we are studying can, however, be determined, with an error no greater than that at $T > T_c$, by correctly subtracting the zero-line drift from the observable spectrum. This quantity is indicated in Fig. 2 by a point inside the plotted confidence interval. The state density $\rho_F$ can be estimated from the electron specific heat $\gamma = 0.11$ mJ/(cm$^3$·K$^2$), which was determined for $La_{1.85}Sr_{0.15}CuO_4$ in Ref. 6. Using (2), we can estimate from this value the exchange integral, $J \sim 2.5 \times 10^{-3}$ eV. It can be seen from Fig. 2 that the Korringa dependence which is linear in temperature is augmented by a bell-shaped component ($\sim 80$ G in height), which typically $\sim 100$-K variation range, which has been observed, as noted above, in dielectric samples and which is not related to the superconducting transition.

The anomalous temperature dependence of the linewidths in the superconducting transition, which is seen at scale values of $\sim 15$ K, is caused by the distortion of the Korringa dependence due to the increase in the state density at the Fermi surface as a result of the coherence effects (estimate show that the nonuniform broadening due to the vortex grating can be ignored[7]). The Korringa linewidth in the superconducting state in this case can be written as[8,9] a series of terms

$$\delta H_s = \delta H_n \cdot 2f(\Delta)\{1 + [1 - f(\Delta)](\Delta/kT) \times \ln(2\Delta/\omega_0)\}, \quad f(\Delta) = [\exp(\Delta/kT) + 1]^{-1}, \tag{3}$$

where the logarithmic divergence of the width[1,2] is cut off at the Zeeman frequency $\omega_0$, since the scattering at magnetic impurities is not effective because of the invariance of $T_c$ upon implantation of a few $Gd^{3+}$ ions. Here $\Delta(T)$ is the width of the gap whose temperature dependence is parametrized as $\Delta(T) = \Delta_0 \tau^\alpha$, where $\tau = (T_c - T)/T_c$. A rapid decrease in the linewidth, as $T \to 0$, is the result of exponential decrease in the number of quasiparticles which are responsible for the spin relaxation. As $T \to 0$, the widths of all the components contract to a single value, which corresponds to the residual width. The Korringa superconducting component of the linewidth can be identified by viewing it against a broad bell-shaped background (Fig. 2). The curve which we are seeking can be obtained by subtracting this background, which is nearly constant near $T_c$, from the observed dependence. The description of this curve on the basis of Eq. (3) yields the following parameter values: $2\Delta_0/kT_c = 5$–6 and $\alpha = 0.25$ (Fig. 2). These values differ from the characteristic values of the Bardeen-Cooper-Schrieffer model. They also imply, in particular, that the temperature dependence of the gap is more critical.

We wish to thank B. P. Vodop'yanov and I. A. Garifullin for a useful discussion of the results.

[1]L. C. Hebel and C. P. Slichter, Phys. Rev. **113**, 1504 (1959).
[2]L. C. Hebel, Phys. Rev. **116**, 79 (1959).
[3]M. Lee, M. Judkowsky, W. P. Halperin *et al.*, Phys. Rev. **B36**, 2378 (1987).
[4]A. Abragam and B. Bleaney, Electron Paramagnetic Resonance of Transition Ions, Clarendon Press, Oxford, 1970.
[5]S. E. Barnes, Adv. Phys. **30**, 801 (1981).
[6]D. K. Finnemore, R. N. Shelton, J. R. Clem *et al.*, Phys. Rev. **B35**, 5319 (1987).
[7]N. E. Alekseevskiĭ, I. A. Garifullin, N. N. Garif'yanov, *et al.*, Zh. Eksp. Teor. Fiz. **94**, 276 (1988) [Sov. Phys. JETP **67**, 905 (1988)].
[8]K. Maki and K. J. Nakanishi, Low Temp. Phys. **5**, 55 (1971).
[9]T. S. Al'tshuler, G. G. Khaliullin, and D. I. Khomskiĭ, Zh. Eksp. Teor. Fiz. **90**, 2104 (1986) [Sov. Phys. JETP **63**, 1234 (1986)].

Translated by S. J. Amoretty