Dependence of T_c on the density of states in $La_{2-x}Sr_xCuO_4$ according to ESR data

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Analysis of the ESR spectra of Gd^{3+} ions serving as spin probes in samples of a lanthanum–strontium metal oxide with various Sr concentrations yields an empirical relationship between T_c and the density of carrier states at the Fermi level, $N(E_F)$.

Experimental studies of the relationships between the critical characteristics of a superconductor, on the one hand, and its electronic properties in the normal state, on the other, make it possible to draw conclusions about the mechanism for superconductivity in the given material. Experiments of this sort are of particular interest in research on the high-T_c superconductors, in which testing various theoretical models

for superconductivity is a particularly acute problem. One such experiment is to measure the dependence of the superconducting transition temperature T_c on the carrier density n and on the density of electron states at the Fermi surface, $N(E_F)$.

The n dependence of T_c has by now been determined quite reliably for most of the high-T_c compounds, although the relationship between the transition temperature and the density of states for several compounds, with various carrier densities, remains an open question. Measurements of the ESR of localized magnetic moments added to the system of interest may prove useful for answering this question. The temperature-dependent contribution to the width of their absorption line is determined by specifically the density of electron states. For example, an ESR study[1] of the relaxation of dopant Cd^{3+} ions in a lanthanum–strontium ceramic has made it possible to trace the changes in the density of states $N(E_F)$ upon the transition to the superconducting state and to evaluate the properties of the superconducting gap.

In this letter we are reporting an experimental study, by the method of ESR involving dopant gadolinium ions, of the behavior of the density of electron states in the normal phase as a function of the carrier density in the polycrystalline compound $La_{2-x-y}Gd_ySr_xCuO_4$. We were also interested in establishing the correspondence between $N(E_F)$ and the superconducting transition temperature T_c.

Two groups of samples were synthesized independently by the standard solid-phase synthesis procedure. The strontium concentration x was varied from 0 to 0.15, while the gadolinium concentration y was fixed at 0.01. The oxygen content corresponded to an oxygen content of 4. The superconducting transition temperature of the samples was determined from measurements of the electrical resistance, the ac susceptibility, and the microwave absorption in weak magnetic fields. A fine-grain powder ($5–10\mu$) mixed with paraffin was used for the ESR measurements. The ESR signal and the microwave-absorption signal were measured by a Bruker-418s spectrometer operating at a frequency of 9300 MHz.

The ESR spectrum of all the test samples consists of a set of partially overlapping absorption lines in the magnetic-field interval 0–7 kG. The widths of the individual components of the spectrum depend on both the the strontium concentration x and the temperature. Figure 1a shows the temperature dependence of the linewidth dH of the best-resolved component of the spectrum, with a resonant field $H_{res}\approx 3000$ G, for four samples, with $x=0.08$, 0.10, 0.12, and 0.15. The solid lines in Fig. 1a are fits of a linear expression $dH=a+bT$ to the high-temperature parts of the $kH(T)$ curves. Figure 1b shows the corresponding results on the temperature dependence of the electrical resistance R.

The structure of the observed spectrum is extremely typical of the ESR of the Gd^{3+} ion. It results from a fine splitting of the energy ground state of this ion in the crystal electric field.[2] The basic qualitative features of the experimental spectrum (the positions and relative intensities of the lines) can be reproduced successfully by numerical simulation based on a simple, axisymmetric, phenomenological Hamiltonian

$$\mathscr{H}=D[S_z^2+1/3S(S+1)]+g\mu_B HS, \qquad (1)$$

where $S=7/2$ and $g=2$ for the Gd^{3+} ion, with the single adjustable parameter

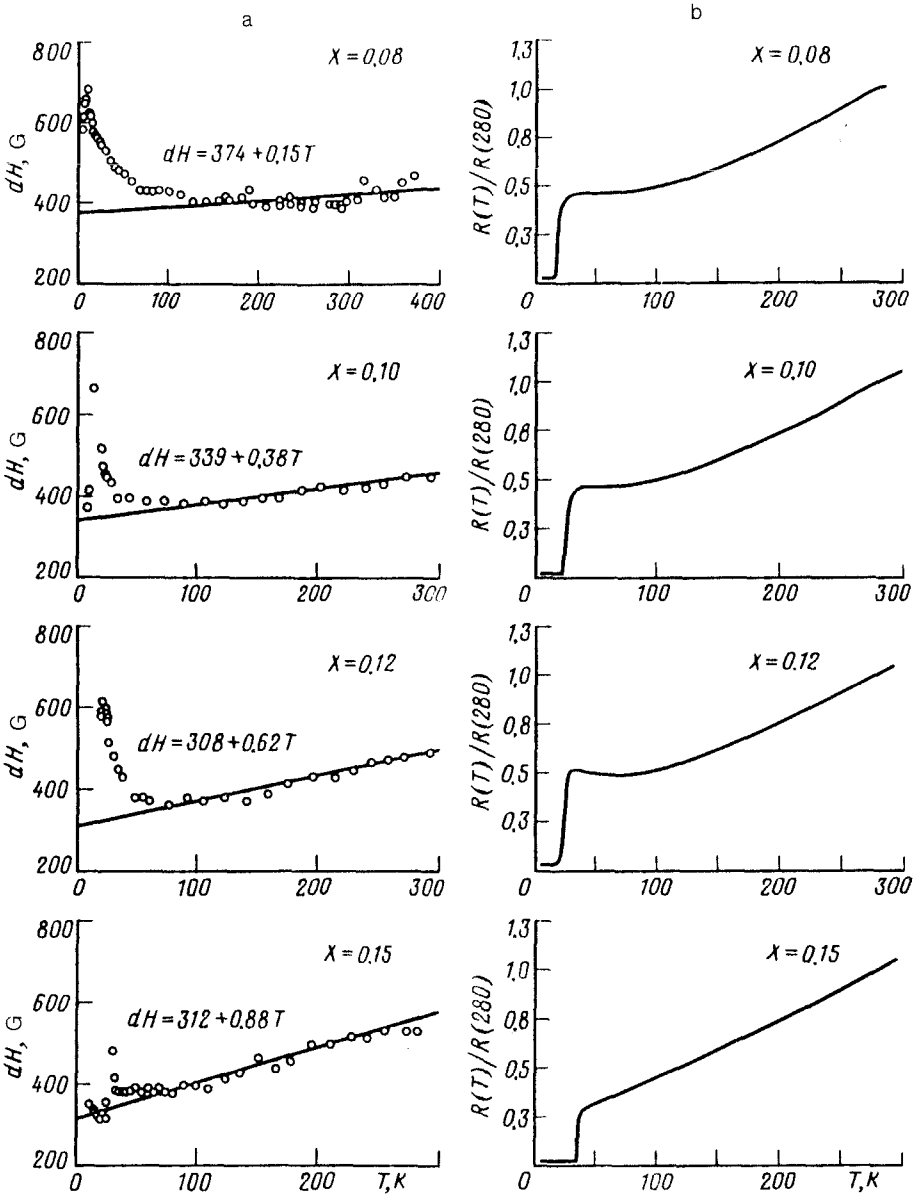

FIG. 1. *a*—Temperature dependence of the linewidth of one of the fine-structure components of the Gd^{3+}ESR spectrum; *b*—temperature dependence of the electrical resistance of $La_{1.99-x-y}Gd_{0.01}Sr_xCuO_4$ samples (x=0.08, 0.10, 0.12, and 0.15).

D=0.28 cm^{-1}($\pm$10%). This numerical simulation included a numerical diagonalization of the energy matrix for each value of the magnetic field, a calculation of the resonant fields and the intensities of the resonant transitions on the basis of the energy

levels found for the Gd^{3+} ion, and an averaging of the spectrum over all orientations of the powder grains with respect to the direction of the magnetic field H. The temperature dependence of the spectrum was simulated by fitting a linear dependence of the width of the individual components. In Fig. 1a we can distinguish two regions on the temperature dependence of the observed width of the ESR line. The dependence is linear in the high-temperature region, while dH is described by a more complicated law in the low-temperature region. Interestingly, the deviation of $dH(T)$ from linearity with decreasing temperature correlates with a characteristic minimum (or shelf) on the plot of $R(T)$, as can be seen by comparing Figs. 1a and 1b.

The relaxation of localized spins in metals is known to be governed by the Korringa mechanism, which makes the ESR linewidth a linear function of the temperature. For the fine-structure spectrum of the Gd^{3+} ion, the temperature-dependent term in the expression for the linewidth of an individual component, $dH=a+bT$, is given by[3]

$$b=M^2[N(E_F)J_{sf}]^2T. \tag{2}$$

The matrix element M relates the states of the Gd^{3+} ion to various spin projections S_z (M^2 also determines the total intensity of the corresponding absorption line), and J_{sf} is the exchange–interaction integral representing the exchange interaction between a localized spin and current carriers (for Gd^{3+} in $La_{2-x}Sr_xCuO_4$ we have[4] $J_{sf}\approx 2.5$ meV).

The simultaneous deviation of $dH(T)$ and $R(T)$ from linearity suggests that the deviation is caused by the same factor in the two cases. A similar aspect of the behavior of the resistance is observed (see Ref. 5, for example) in lanthanum–strontium high-T_c compounds with various dopants. Presently under discussion as possible reasons are the appearance of a semiconducting gap, a hopping conductivity, and Kondo and localization effects. Since the $R(T)$ curves which we observed cannot be approximated by an expression $R\propto\exp(1/T^{\alpha})$, where $\alpha=1$, 0.5, or 0.25, the first two of these mechanisms can apparently be ruled out. These curves are described considerably better by the function proposed in Ref.5, which contains a logarithmic term:

$$R(T)=A+BT-C\ \ln(T). \tag{3}$$

This circumstance is evidence for a Kondo effect or a weak localization. A change in the density of carrier states accompanying these effects could change the coefficient b in the temperature-dependent part of dH [see (2)]. The deviation of the Gd^{3+}ESR linewidth from linearity at temperatures on the order of 100 K and below can be described fairly well by adding a logarithmic term $-c\ln T$ to the expression $dH(T)=a+bT$ (Fig. 2). This circumstance can serve as evidence in favor of localization, since the effective increase in the density of states $N(E_F)$ upon a weak localization of carriers should accelerate the relaxation of the Gd^{3+} ion with decreasing temperature. A corresponding logarithmic term of course arises in the $dH(T)$ dependence in the case in which the ESR in observed at a Kondo ion,[3] but the Gd^{3+} ion, with its spherically symmetric, deep-lying f shell, is an extremely unlikely candidate for such a role.

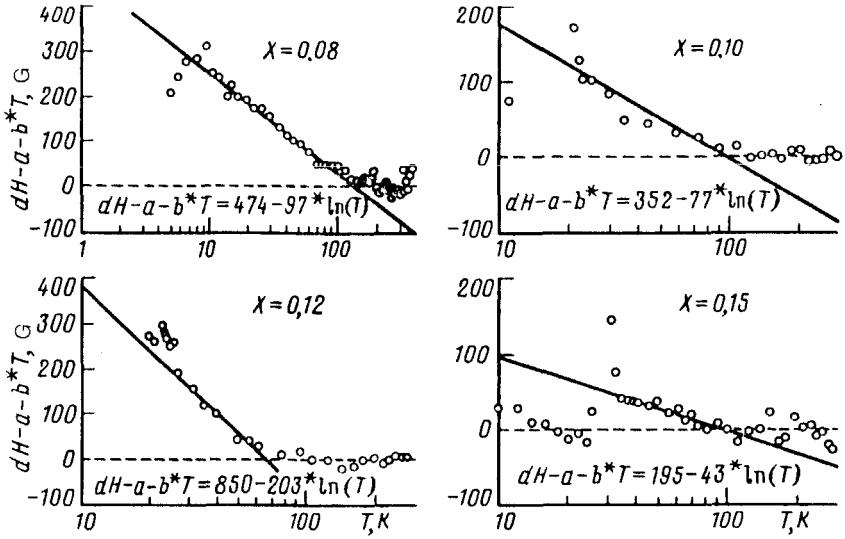

FIG. 2. Nonlinear part of the linewidth $dH(T)=a-bT$ versus $\ln T$ for $La_{1.99-x-y}Gd_{0.01}Sr_xCuO_4$ samples (x=0.8; 0.10; 0.12; 0.15).

The analysis above shows that in order to extract unambiguous information about the density of states in the normal phase we must use specifically the high-temperature parts of the plot of the ESR linewidth (Fig. 1a). It seems natural to link the observed increase in the Korringa slope $b=dH/dT$ in (2) with increasing strontium content x in the samples as the density of carrier states at the Fermi surface increases, since we have $N(E_F)\sim\sqrt{b}$ according to (2). Measurements of the resistance, the susceptibility, and the microwave absorption show that the transition temperature T_c changes along with the increase in x in these samples. It is thus possible to relate two quantities which can be measured experimentally: T_c and dH/dT. The result is shown in Fig. 3, where

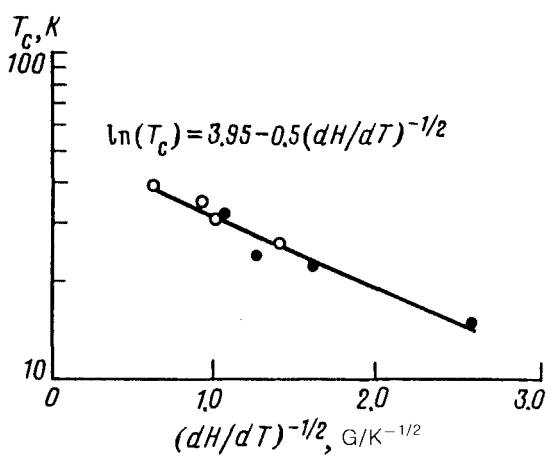

FIG. 3. Superconducting transition temperature as a function of the square root of the reciprocal slope of the temperature dependence of the Gd^{3+}ESR linewidth. The open and filled circles correspond to samples of different lots.

the transition temperature is plotted in logarithmic scale along the abscissa, and $b^{-1/2}$ is plotted along the ordinate. The symbols of one type in this figure correspond to samples of lot 1 (a preliminary discussion of the results found in measurements on these samples was published in Ref. 4); the symbols of the other type correspond to samples of lot 2, prepared independently. Experimental data for this second lot are shown in Fig. 1. It can be seen from Fig. 3 that all the points plotted here can be approximated well by

$$T_c = \alpha \exp(-\beta b^{-1/2}), \tag{4}$$

where α and β are adjustable parameters. In view of the relationship between b and $N(E_F)$, we can assert that a universal relation holds between the transition temperature and the density of states in this compound: $d(\ln(T_c))/d(1/N(E_F)) = \text{const}$. This constant can serve as a characteristic of the binding of the quasiparticle, while the pre-exponential factor in (4) specifies the energy scale.

As one possible example of the use of the relation found here to evaluate microscopic properties, we consider the BCS expression $T_c = \alpha \exp(-1/L_{\text{eff}})$. From our data (Fig. 3) we find $\alpha = 52$ K and an effective coupling constant $\lambda_{\text{eff}} = 0.78$–$1.88$ for the samples of lot 2, with strontium concentrations from 0.08 to 0.15. The plausibility of these estimates does not, however, constitute a decisive argument in favor of a phonon model, since expressions for T_c with a corresponding structure arise in several other theories of high-T_c superconductivity.

In summary, these measurements of the ESR of Gd^{3+} ions added to $La_{2-x-y}Sr_xCuO_4$ samples doped with strontium to various extents have established the relationship $d[\ln(T_c)]/d[1/N(E_F)] = \text{const}$ between the superconducting transition temperature and the density states at the Fermi surface. The simple structure of $La_{2-x}Sr_xCuO_4$ raises the hope that this relationship is characteristic of specifically the CuO_2 planes. The presence of these planes in other superconducting metal oxides suggests that this relationship holds in these other cases, also, since the primary differences among the various superconductors (if we ignore the interaction between planes) are all in the nature of the doping and are manifested in different densities of states. Unfortunately, a study of one compound alone does not tell us whether this relationship holds over the entire range of transition temperatures which have been reached or whether this relationship is a low-temperature asymptotic form of a more complex law. Another problem, which we regard as no less interesting, is the behavior of the density of electron states as a function of the carrier density in samples of a lanthanum metal oxide which are overdoped with strontium, in which the conductivity is observed to increase while T_c decreases. Preliminary ESR experiments on two $La_{1.99-x-y}Gd_{0.01}Sr_xCuO_4$ samples ($x = 0.2$ and 0.25; see also Ref. 4) show that T_c and $N(E_F)$ are related by the same empirical relationship in these cases.

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