Nuclear relaxation in a superconductor with strong electron-electron correlations

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If there is a strong repulsion of electrons with opposite spins at a common site, the rate of nuclear magnetic relaxation in the superconducting phase will become a monotonic function of the temperature, in contradiction to the prediction of the BCS theory.

1. We know quite well (Ref. 1, for example) that the primary mechanism for nuclear magnetic relaxation in superconductors is a relaxation involving conduction electrons. In the BCS theory, conduction electrons are treated in the approximation of nearly free electrons, for which the relaxation rate is a linear function of the tempera-

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ture in the normal phase. In the superconducting phase the relaxation rate $R_s(T)$ increases immediately below the superconducting transition temperature T_c, because of a coherence factor. As the temperature is lowered further, $R_s(T)$ decreases rapidly because of a decrease in the number of one-particle excitations.

In the present letter we show that if there is a large potential energy U for the repulsion of electrons with opposite spins at a site, the coherence factor in the relaxation rate $R_s(T)$ will be close to unity, and $R_s(T)$ will fall off montonically below T_c.

2. We assume that nuclei with a spin **I** are coupled with electrons through a hyperfine contact interaction

$$H_{hf} = A\sum_i \mathbf{I}_i \vec{\sigma}(\mathbf{R}_i), \tag{1}$$

where $\vec{\sigma}(\mathbf{R}_i)$ is the spin density of conduction electrons at the nucleus. The relaxation rate

$$R_s(T) \equiv T_{1s}^{-1} = \frac{-\int_{-\infty}^{\infty} dt \langle [H_{hf}, I^z](t)[I^z, H_{hf}] \rangle}{\langle I^z I^z \rangle} \tag{2}$$

is expressed in terms of the equilibrium correlation function of the commutators of the z component of the total spin, $I^z = \Sigma_j I_j^z$, with Hamiltonian (1). Calculating the commutators, we conclude that the numerator in (2) contains a product of nuclear and electron operators, taken at a common site, of the type

$$\langle I_i^-(t) I_i^+ \rangle \{ \langle a_{i\downarrow}(t) a_{i\downarrow}^+ \rangle \langle a_{i\uparrow}^+(t) a_{i\uparrow} \rangle + \langle a_{i\uparrow}^+(t) a_{i\downarrow}^+ \rangle \langle a_{i\uparrow} a_{i\downarrow}(t) \rangle \}, \tag{3}$$

where $a_{i\sigma}^+$, $a_{i\sigma}$ are the electron second-quantization operators. Since correlation effects turn out to be important for several high-temperature superconductors, to reach an understanding of the influence of these effects on the nuclear relaxation we will carry out a calculation in the Hubbard model, assuming below that the quantity U is the largest parameter:

$$H = E\sum_{i\sigma} n_{i\sigma} + \sum_{ij\sigma} t_{ij} a_{i\sigma}^+ a_{j\sigma} + U\sum_i n_{i\uparrow} n_{i\downarrow}, \tag{4}$$

where $n_{i\sigma} = a_{i\sigma}^+ a_{i\sigma}$, and t_{ij} is the integral representing a hop from site i to j. A superconductivity problem can be formulated for model (4) (Refs. 2 and 3). Postponing a discussion of whether the superconducting state can be realized, we wish to call attention to the circumstance that for (4) there is an exact relationship[2] between the order parameter $\Delta = 2\Sigma_{ij} t_{ij} \langle a_{i\uparrow} a_{j\downarrow} \rangle$, which is nondiagonal with respect to sites, and the single-site anomalous expectation value

$$\langle a_{i\downarrow}(0) a_{i\uparrow}(0) \rangle = \Delta/(2E + U), \tag{5}$$

which is small if U is large. Relation (5) is a formal expression of the disadvantage of a situation in which there are two quasiparticles with opposite spins at a common site. This is a more general statement of model (4), rather than its corollary. Calculations of the dynamic average expectation values of the type $\langle a_{i\uparrow}(t) a_{i\downarrow}(t') \rangle$ in the second term in (3) (we will omit the details of these calculations, because of their complex-

ity) show that these expectation values are also small, to the extent that relation (5) is small. Consequently, only expectation values of the normal type, $\langle a_{i\downarrow}^{+}(t)a_{i\downarrow}(0)\rangle$, contribute to the nuclear relaxation rate in (2) in the limit $U\to\infty$. At large U, these expectation values assume the form customary for the BCS theory and contain a spectrum with a gap Δ, which is a consequence of heterosite anomalous expectation values. Standard calculations for the relaxation rate, divided by that in the normal phase, then yield the expression

$$\frac{R_s}{R_n} = \frac{2}{kT}\int_{\Delta}^{\infty} \frac{\epsilon(\epsilon+\omega_N)f(\epsilon)[1-f(\epsilon+\omega_N)]\,d\epsilon}{\sqrt{\epsilon^2-\Delta^2}\sqrt{(\epsilon+\omega_N)^2-\Delta^2}}, \tag{6}$$

where ω_N is the precession frequency of the nuclear moment, and $f(\epsilon)$ is the Fermi function. Expression (6) can be derived from the corresponding expression of the BCS theory[1] if we set the coherence factor equal to unity in the latter expression. Under the conditions $\omega_N \ll kT$, Δ, an integration in (6) leads to

$$\frac{R_s}{R_n} \approx 2f(\Delta)\{1+\frac{\Delta}{2kT}f(-\Delta)\ln(\Delta/\omega_N)\}. \tag{7}$$

Assuming for definiteness that Δ depends on the temperature in the same way as in the BCS theory, we can construct temperature dependence (7) (Fig. 1). We see from this figure that below T_c the value of $R_s(T)$ does not exceed the value in the normal phase, $R_n(T)$.

3. A recent preprint by Katayama-Yoshida *et al.*[4] reports results of a study of the relaxation of ^{63}Cu and ^{17}O nuclei which was carried out in a common $YBa_2Cu_3O_{7-\delta}$ sample. The temperature dependence of the relaxation rate of ^{17}O nuclei is nonmonotonic, as in BCS superconductors.[1] In contrast, the relaxation rate of ^{63}Cu nuclei in the CuO_2 plane of the structure falls off rapidly just below T_c (as has been seen previously in many experiments). If we assume that the repulsion energy at a site is significantly

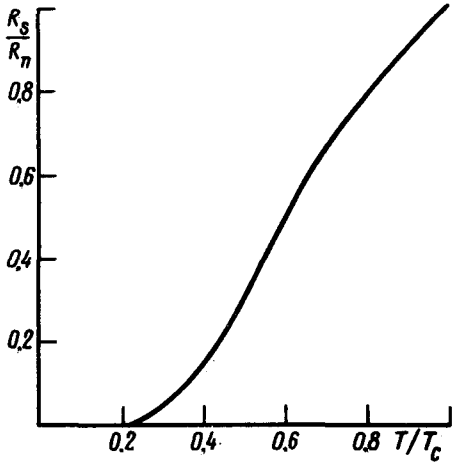

FIG. 1.

greater than the energy of the band motion in the copper system, while in the oxygen system these energies are comparable in magnitude, we would expect that the peak in the relaxation rate below T_c would be suppressed for copper and weakened for oxygen—in accordance with experiments.[4] For an isotropic hyperfine interaction the mechanism which we are discussing here should make a contribution of the same magnitude to the transverse relaxation rate T_2^{-1}. Indeed, a monotonic decrease just below T_c has been observed[5] for copper; the magnitude of the change in T_2^{-1} is about 3.5×10^3 s^{-1}. This figure is comparable to the change $\sim2.5\times10^3$ s^{-1} seen in T_1^{-1} (Ref. 4).

According to the prevalent opinion, which is based one experimental data, a band description of the subsystem of oxygen electrons is valid as a zeroth approximation, and the conductivity and superconductivity play out primarily through oxygen states. Experiments on photoemission, x-ray absorption, and x-ray emission and calculations by Hirsch *et al*[6] show that the number of Cu^+ ions in the system increases with increasing hole density upon doping. In the Hubbard model, this increase corresponds to a partial filling of the upper copper subband by the electrons and means that this subband has an energy overlap with an oxygen band which has a hole conductivity. Consequently, the order parameter Δ in the copper subsystem should be sought by solving the system of self-consistency equations, which is coupled to the extent that there is hybridization. Although we will not take up that problem in the present letter, we believe that it could influence only the particular features of the temperature dependence $R_s(T)$, causing no qualitative change in the result.

It follows from the structure of $YBA_2Cu_3O_7$ that the overlap of wave functions between layers is more pronounced for copper than for oxygen. Consequently, the sign of the Hall constant for the magnetic field in the CuO_2 plane is opposite that in the case in which the field is directed perpendicular to the CuO_2 layers (Ref. 7), in which the overlap primarily involves oxygen.

In principle, a band picture is not devoid of meaning as an initial approximation for the copper electrons. There is a reasonable procedure for describing electron-electron correlations in a band in terms of magnetic fluctuations[8] whose length scale ξ_M decreases to the atomic level as the correlation strengthens. The result of this study will apparently remain valid if the length ξ_M does not exceed the superconducting correlation length.

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