Theory of NMR as a local probe for the electronic structure in the mixed state of the high-$T_c$ cuprates

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We argue that nuclear magnetic resonance experiments are a site-sensitive probe for the electronic spectrum in the mixed state of the high-$T_c$ cuprates. Within a spin-fermion model, we show that the Doppler-shifted electronic spectrum arising from the circulating supercurrent changes the low-frequency behavior of the imaginary part of the spin-susceptibility. For a hexagonal vortex lattice, we predict that these changes lead to (a) a unique dependence of the $^{63}$Cu spin lattice relaxation rate, $1/T_1$, on resonance frequency, and (b) a temperature dependence of $T_1$ which varies with frequency. We propose a nuclear quadrupole experiment to study the effects of a uniform supercurrent on the electronic structure and predict that $T_1$ varies with the direction of the supercurrent.

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The form of the fermionic excitation spectrum in and around a vortex in the mixed state of high-$T_c$ cuprates has been the subject of intense research in the last few years (1,2). It has been known for a long time that the supercurrent circulating a vortex gives rise to a Doppler-shift in the fermionic excitation spectrum (3). For a d-wave order parameter, Volovik pointed out (4), this shift leads to the scaling relation $D(\epsilon_F) \sim \sqrt{H}$, where $D(\epsilon_F)$ is the density of states (DOS) at the Fermi energy, and $H$ is the applied field. The main contribution to the DOS comes from regions close to the nodes, but in real space far from the vortex cores. Specific heat (SH) measurements by Moler et al. (5) on YBa$_2$Cu$_3$O$_{6.95}$ confirmed this scaling behavior, however, these experiments only provide information on the sample averaged DOS.

In this communication we argue that the spin-lattice relaxation time, $T_1$, measured in nuclear magnetic resonance (NMR) experiments, is a more direct probe since it can provide site-specific information on the electronic structure in the vicinity of a vortex. Our conclusions are based on two important results. First, it was recently pointed out by Curro and Slichter (CS) that the dependence of resonance frequency on the local field allows one to measure $T_1$ as a function of the nucleus position in a vortex lattice (6). In addition, $T_1$-measurements are a purely electronic probe, in contrast to SH experiments, which are dominated by phononic contributions. Second, within a spin-fermion model (7), we find that the Doppler-shifted electronic spectrum significantly changes the low-frequency behavior of the imaginary part of the spin susceptibility, $\chi''(q, \omega)$. Since $\chi''(q, \omega)$ changes with the spatially varying supercurrent, $1/T_1$ which is a weighted momentum average of $\chi''$ in the zero frequency limit, acquires a frequency, i.e., spatial dependence.

Experimentally, CS found for the $^{17}$O resonance in YBa$_2$Cu$_3$O$_7$ that $1/T_1$ increases with decreasing distance from the vortex core (8), i.e., with increasing frequency. Preliminary results of Milling et al. (9) show a qualitatively similar behavior for the $^{63}$Cu relaxation rate. We demonstrate that the experimentally observed frequency dependence of $T_1$ can be qualitatively explained by the spatial variation of the supercurrent and the resulting Doppler-shift in the electronic spectrum and predict a temperature dependence of the relaxation rate which changes uniquely with frequency. Both results provide insight into the local structure of electronic and magnetic excitations in the mixed state, in contrast to the sample averaged information provided by SH experiments.

In what follows we consider for simplicity a single-layer system; an extension of our results to the case of a double-layer system, i.e., YBa$_2$Cu$_3$O$_{6-\delta}$, is straightforward. The $^{63}$Cu spin lattice relaxation rate, $1/T_1$, for an applied field parallel to the $c$-axis is given by

$$\frac{1}{T_1T} = \frac{k_B}{2\hbar} (\hbar^2 \gamma_n \gamma_e)^2 \frac{1}{N} \sum_q F_c(q) \lim_{\omega \to \nu} \frac{\chi''(q, \omega)}{\omega} ,$$

where

$$F_c(q) = [A_{ab} + 2B(\cos(q_x) + \cos(q_y))]^2 ,$$

and $A_{ab}$ and $B$ are the on-site and transferred hyperfine coupling constants, respectively (10). To describe the effects of a supercurrent on $\chi''$ we use the spin-fermion model (11) in which the damping of the spin mode is determined by its coupling to the planar quasi-particles. Within this model, the spin propagator, $\chi$, is given by

$$\chi^{-1} = \chi_0^{-1} - \Pi ,$$

where $\chi_0$ is the bare propagator, and $\Pi$ is the bosonic self-energy given by the irreducible particle-hole bubble. It was earlier suggested (12) that $\text{Re} \chi^{-1}$ should possess a hydrodynamic form such that for $\omega \to 0$

$$\text{Re} \chi^{-1} = \chi_0^{-1} - \text{Re} \Pi = \frac{\xi_{AF}^2}{\alpha} + \frac{(q - Q)^2}{\alpha} ,$$

where $\xi_{AF}$ is the magnetic correlation length, $\alpha$ is a temperature independent overall constant, and $Q = (\pi, \pi)$ is the position of the magnetic peak in momentum space.
where we assume to be commensurate \[\xi\]. The description of \(\text{Re} \chi^{-1}\) in Eq. (4) as a relaxational, non-dispersing spin mode at low frequencies is in agreement with the analysis of NMR and inelastic neutron scattering experiments on several La\(_{2-x}\)Sr\(_x\)CuO\(_4\) and YBa\(_2\)Cu\(_3\)O\(_{6+x}\) compounds \[11, 12\].

We are thus left with the calculation of the imaginary part of \(\Pi\) which describes the spin damping brought about by the decay of a spin excitation into a particle-hole pair. In the superconducting state we find lowest order in the spin-fermion coupling \(g\)

\[-\Pi(q, i\omega_n) = g^2 T \sum_{k,m} \left\{ G(k, i\Omega_m)G(k + q, i\Omega_m + i\omega_n) + F(k, i\Omega_m)F(k + q, i\Omega_m + i\omega_n) \right\}, \quad (5)\]

where \(G\) and \(F\) are the normal and anomalous Green’s functions. Assuming that the supercurrent momentum, \(p_s\), possesses only a weak spatial dependence, we find in semiclassical approximation \[3\]

\[G(k, i\Omega_m) = \frac{v_k^2}{i\Omega_m - E_k + i\Omega_m + E_{-k}};\]
\[F(k, i\Omega_m) = -u_k v_k \left\{ \frac{1}{i\Omega_m - E_k} - \frac{1}{i\Omega_m + E_{-k}} \right\}. \quad (6)\]

Here, \(E_k\) is the dispersion of the fermionic quasi-particles (Bogoliubons), which up to linear order in \(p_s\) is given by \[3\]

\[E_k = \sqrt{c_k^2 + |\Delta_k|^2 + v_F(p_s) \cdot p_s}, \quad (7)\]

where \(c_k\) is the electronic normal state dispersion and \(v_F(k) = \partial E_k / \partial k\). Any changes in the \(d\)-wave gap \(\Delta_k = \Delta_0 (\cos(k_x) - \cos(k_y))/2\) and the BCS coherence factors \(u_k, v_k\) due to a supercurrent appear only to order \(p_s^2\) and can thus be neglected.

Since the dominant contribution to \(\text{Im} \Pi\) for \(\omega \to 0\) comes from quasi-particle excitations in the vicinity of the nodes, we expand \(E_k\) around the nodes to linear order in \(p_s\) and perform the momentum and frequency integrations in Eq. (5) analytically. Combining the results with Eqs. (1), (3) and (4) we find that the dependence of \(\text{Im} \Pi\) on temperature \(T\) and on \(p_s\) is determined by the set \(\{D_m/T\}\), where \(D_m = v_F^{(m)} \cdot p_s\) and \(v_F^{(m)}\) is the Fermi velocity at the node \(m\) in the \(m\)th quadrant of the Brillouin zone. The full expression for \(1/T_1\) for arbitrary \(\{D_m/T\}\) is too cumbersome to be presented here, however, in the low temperature limit where \(|D_m/T| \gg 1\) for \(m = 1..4\), it simplifies and we obtain up to order \((T/D_m)^2\)

\[\frac{1}{T_1} = \frac{C}{N} \left( \frac{1}{v_F v_\Delta} \right)^2 \sum_{n,m} \left\{ F(q_{n,m}) \left( D_m D_n + \frac{\pi^2 T^2}{3} \right) + O(e^{D_m/T}) \right\} \quad (8)\]

where the sum runs only over those nodes with \(D_m, D_n < 0\), \(C = (\alpha g)^2 k_B h^2 \gamma_n \gamma_c)/(2\hbar)\), \(v_\Delta = |\partial \Delta_k / \partial k|\) at the nodes, and

\[F(q_{n,m}) = \frac{F_c(q_{n,m})}{(\xi_A^2 + (q_{n,m} - Q)^2)^2}. \quad (9)\]

Here, \(q_{n,m}\) is the momentum connecting nodes \(n\) and \(m\). The opposite signs of the \(T^2\)-terms on the r.h.s. of Eq. (8) arises from the combination of Fermi functions involved in different relaxation processes. While the first term describes a process in which a Bogoliubon is simply scattered at the spin-fermion vertex, the second one involves processes in which two Bogoliubons are simultaneously created or destroyed at the vertex. Since the dominant contribution to the relaxation rate in the low temperature regime comes from the second term in Eq. (8), we find the unexpected result that in this limit \(1/T_1\) actually decreases with increasing temperature. Note, that for \(T \to 0\) it follows from Eq. (8) that \(1/T_1 \sim T^2\).

In the high-temperature limit \((|D_m/T| \ll 1\) for all \(m\)), we obtain to leading order in \((D_m/T)^2\)

\[\frac{1}{T_1} = \frac{\pi C}{6N} \left( \frac{T}{v_F v_\Delta} \right)^2 \sum_{n,m} \left\{ F(q_{n,m}) + F(q_{n,m+2}) \right\} \quad (10)\]

where the sum runs over all nodes. For \(T \gg |D_m|\) we thus recover as expected the temperature dependence of the relaxation rate in the absence of a supercurrent.

We now turn to NMR experiments in the mixed state of the high-\(T_c\) cuprates which possess a hexagonal vortex lattice. We consider for definiteness YBa\(_2\)Cu\(_3\)O\(_7\), where \(v_F \approx 0.4\) eV, \(v_\Delta \approx 20\) meV, \(A_{ab}/B = 0.7\), and \(\xi_A \approx 2\) is temperature independent in the superconducting state \[9\]. Each nucleus in the sample is characterized by a resonance frequency \(\Delta_{r}(\mathbf{r}) = \gamma_n h E_{\zeta_0}(\mathbf{r})\) and a supercurrent momentum \(p_s(\mathbf{r})\). Here \(E_{\zeta_0}(\mathbf{r})\) is the local magnetic field and \(\gamma_n\) is the nuclear gyromagnetic ratio. Since the electronic Zeeman-splitting for typical applied fields is of the order \(10^{-1}\) meV, and thus smaller than the Doppler-shift for most of the nuclei, it can be neglected.

Our scenario for the calculation of \(T_1\) is only applicable to a given nucleus, if in its vicinity \(\Delta(\mathbf{r})\) is uniform and \(p_s(\mathbf{r})\) varies sufficiently slowly. Since both assumptions are violated in the vicinity of a vortex core we exclude in what follows all nuclei within a radius \(R = 2\xi_{ab} \approx 5\)\(\sigma_0\) of the vortex core. Here, \(\xi_{ab}\) and \(\sigma_0\) are the superconducting in-plane coherence length and the crystal lattice constant, respectively, and \(H_z(\mathbf{r})\) is given by \[9\]

\[H_z(\mathbf{r}) = \frac{2\Phi_0}{\sqrt{3} \lambda^2} \sum_{\mathbf{q}} \frac{\exp(i\mathbf{q}\mathbf{r})}{1 + \lambda^2 q^2} \quad (11)\]

where \(\lambda\) is the magnetic penetration depth, \(\Phi_0\) is the flux quantum, and \(\mathbf{q}\) runs over the reciprocal lattice of the 2D vortex array. Since non-local as well as non-linear effects can effectively be accounted for by a renormalization of \(\lambda\)
we neglect them in the following. We can then calculate the supercurrent momentum via \( \nabla \times \mathbf{H}(\mathbf{r}) \sim \mathbf{p}_s(\mathbf{r}) \), and thus obtain a spatial relation between \( H_s(\mathbf{r}) \) and \( \mathbf{p}_s(\mathbf{r}) \). In Fig. 1 we plot the resulting \( ^{63}\text{Cu} \) spectrum (solid line) and \( 1/T_1T \) (open squares) as a function of the local field \( H_s \) at \( T = 1 \) K. Nuclei at the highest frequencies are located at a distance \( 2\xi_{ab} \) from the center of the vortex, nuclei at the lowest frequencies are in the center of a vortex triangle, those at the maximum of the spectrum are at the midpoint between two vortices. For \( T = 1 \) K, the relaxation rate for practically all nuclei is determined by the low-temperature expression, Eq.(8). In this case, \( 1/T_1T \sim p_s^2 \) and hence reflects the frequency dependence of \( p_s \), which increases with decreasing distance from the vortex core. The Doppler-shifted fermionic spectrum thus gives rise to a unique dependence of \( 1/T_1T \) on frequency within the vortex-lattice spectrum of \( ^{63}\text{Cu} \). This result is in qualitative agreement with preliminary measurements by Milling et al. [5] who indeed find that \( 1/T_1T \) increases with increasing local field.

Another unique signature of Doppler-shifted excitations appears in the temperature dependence of the relaxation rate. In Fig. 2, we present \( 1/T_1T \) as a function of field for three different temperatures (the 30 K and 60 K curves are horizontally offset for clarity). For nuclei at the low-field end of the curve, \( p_s \) is small, \( |D_m/T| \ll 1 \) already at low \( T \), and hence \( (T_1T)^{-1} \sim T^2 \) which increases with increasing temperature. On the other hand, for nuclei closer to the vortex core, and particularly for those at the high-field end of the curve, \( p_s \) is large and \( |D_m/T| \gg 1 \) even for \( T = 60 \) K (which is still below the lattice melting temperature at \( T_m = 70 \) K). The temperature dependence of \( 1/T_1T \) is therefore described by Eq.(8), and thus decreases with increasing temperature, as discussed earlier. Between these two extrema there exists a cross-over region, characterized by a minimum in the relaxation rate. This non-monotonic behavior of \( 1/T_1T \) arises from two competing contributions: while quasi-particle excitations involving nodes with \( D_m > 0 \) become exponentially suppressed with increasing \( H_s \), the contributions from the remaining (un-suppressed) excitations scale as \( \sim p_s^2 \) and thus increase. The cross-over occurs approximately at those nuclei for which \( \text{max}|D_m| \sim O(T) \); the minimum thus shifts towards higher fields, i.e., larger values of \( |D_m| \), with increasing temperature, as shown in Fig. 2.

While the absolute scale of our results in Figs. 1 and 2 depends on a number of parameters, it is practically independent of \( \xi_{AF} \). In contrast, the predicted temperature and frequency dependence of \( T_1 \) depends only on \( \{D_m/T\} \) and thus presents a generic feature of a Doppler-shifted electronic spectrum due to the distribution of supercurrents in a hexagonal vortex lattice.

An alternative experiment which could observe the effects of a Doppler-shifted fermionic dispersion is a nuclear quadrupole resonance (NQR) measurement in which a uniform supercurrent is applied to the sample. Since in the low-temperature limit, Eq.(8), \( 1/T_1T \) depends on \( D_m = v_F^{(m)} \cdot \mathbf{p}_s \), with \( v_F^{(m)} \) being fixed by the underlying lattice, one would expect that the relaxation rate varies with the angle between the supercurrent momentum, \( \mathbf{p}_s \), and the crystal axes. In Fig. 3 we plot \( 1/T_1T \), for \( |p_s| = 0.02 \) as a function of the angle \( \phi \) between \( \mathbf{p}_s \) and the crystal \( x \)-axis (see inset). For \( \phi = 0 \), i.e., for \( \mathbf{p}_s \) parallel to the \( x \)-axis, we find \( D_{2,3} = -v_F q_s/\sqrt{2} \) while \( D_{1,4} = +v_F q_s/\sqrt{2} \). \( 1/T_1T \) thus decreases with \( T \) for \( |D_m/T| \gg 1 \), and increases as \( \sim T^2 \) for \( |D_m/T| \ll 1 \), in agreement with the results in Fig. 2. On the other hand, \( \phi = \pi/4 \), i.e., \( \mathbf{p}_s \) along the lattice diagonal, is a
special case since $D_{1,3} = \pm v_F g_s$ while $D_{2,4} = 0$. While the contribution to $1/T_1 T$ from quasi-particle excitations involving only nodes 2 and 4 scales as $T^2$ for all temperatures, the contributions involving only nodes 1 and 3 decreases with increasing $T$. As a result, $1/T_1 T$ increases for all temperatures, though only weakly at low $T$. For $T \gg v_F g_s$, the relaxation rate for all $\phi$ scales as $T^2$ and thus becomes angle-independent. Note, that the strong angular dependence of the relaxation rate for intermediate temperatures explains the spread in values of $1/T_1 T$ at a given local field which we found in Fig. 2 particularly in the $T=60K$ curve.

Spin-diffusion and vortex vibrations [14] can potentially contribute to the spin-lattice relaxation and thus smear out the effects described above. However, spin diffusion is strongly suppressed by the inhomogeneity of the magnetic field in a vortex lattice [15]. Moreover, calculations on the effect of vortex vibrations in YBa$_2$Cu$_3$O$_{6+x}$ [15] as well as an experimental comparison of $^{17}$O and $^{63}$Cu relaxation rates [3] conclude that vortex vibrations are irrelevant in the relaxation of $^{63}$Cu but may play an important role in that of $^{17}$O.

Recently, several groups have calculated $T_1$ in the superconducting state starting from a Fermi gas (FG) description of the cuprates, i.e., neglecting antiferromagnetic spin fluctuations [14][17]. We extended our calculations to the FG limit by using $\chi_{full} = \Pi/g^2$ and found two distinct differences between the relaxation rate for a FG and that in the presence of strong spin fluctuations. First, $1/T_1 T$ for a FG always increases with temperature as long as $A_{ab}$ and $B$ possess the same sign, in contrast to our results in Eq. (8). Second, in the FG limit $1/T_1 T$ increases monotonically with increasing local field in a vortex lattice, i.e., it does not exhibit the local minimum we found in Fig. 2. We propose that these differences in the predicted relaxation rates enable NMR experiments to determine which of the two limits applies to the superconducting state of the high-$T_c$ cuprates.

Finally, we found above that the behavior of the relaxation rate in a vortex lattice reflects the presence of a supercurrent, magnetic fluctuations and nodes in the superconducting gap. We are currently studying whether similar effects also occur in Sr$_2$RuO$_4$ for which strong indications of ferromagnetic fluctuations and a p-wave order parameter exist [1].

In summary, we have demonstrated that NMR is a site-specific probe for the electronic structure in the mixed state of the high-$T_c$ cuprates. We have shown that in a hexagonal vortex lattice, the Doppler-shifted electronic spectrum gives rise to a characteristic temperature and field dependence of the $^{63}$Cu relaxation rate. We propose an NQR experiment in which the direction of a uniform supercurrent with respect to the crystal lattice is varied, and predict a unique angular dependence of $T_1$. Finally, we argue that our strong coupling results are qualitatively different from those predicted for a FG.

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