Electronic Raman Scattering of Overdoped $Tl_2Ba_2CuO_{6+\delta}$ in High Magnetic Fields

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(Received 13 September 1996)

The excitations across the superconducting gap (2Δ-peak in B_{1g}-symmetry Raman spectra) in an overdoped $Tl_2Ba_2CuO_{6+\delta}$ single crystal have been studied as a function of temperature and field. The 2Δ-peak intensity, which we interpret as the density of the superconducting condensate around the gap antinodes, is a linear function of temperature, but a nonlinear function of the field. The latter is attributed to renormalization of quasiparticle spectra in the vicinity of vortex lines in the mixed state. The 2Δ-peak is present above the irreversibility line and yields a conventional temperature dependence of the effective upper critical field. [S0031-9007(97)02748-8]

PACS numbers: 74.25.Ha, 74.60.Ec, 74.72.Fq, 78.30.Er

One of the properties of the cuprate high temperature superconductors is their high upper critical fields, H_{c2}, governed by a short in-plane superconducting coherence length $\xi \approx 10 - 20$ Å and a large Landau-Ginzburg parameter λ/ξ (the in-plane London penetration depth $\lambda \approx 10^3$ Å). These high critical fields have been obstacles to the study of the field-induced superconducting-normal state transitions at low temperatures. In two recent Letters, Mackenzie *et al.* [1] and Osofsky *et al.* [2] have observed that in the overdoped cuprates a critical field $H^*(T)$, associated with the onset of dissipation, is depressed to low values around T_c, but displays a steep rise with no sign of saturation as the temperature is decreased. $H^*(T)$ has been interpreted as the upper critical field. The low values of $H^*(T)$ enable a study of the renormalization of the quasiparticle spectra in the field-induced transitions by spectroscopic methods. In this Letter we use electronic Raman scattering from overdoped $Tl_2Ba_2CuO_{6+\delta}$ to study the redistribution of quasiparticle spectra near the superconducting transition in the presence of magnetic fields up to H_{c2}. We find that a local superconducting order is present at temperatures and fields above the irreversibility field obtained from the magnetization hysteresis curves. We extract the temperature dependence of an effective upper critical field $H^*_{c2}(T)$ that differs qualitatively and quantitatively from $H^*(T)$.

Electronic Raman scattering in metals arises from electron-hole or multi-quasi-particle excitations around the Fermi surface and provides spectroscopic information about the quasiparticle excitation spectra. For strongly correlated systems incoherent quasiparticle scattering can lead to finite Raman intensity over a broad region of frequency [3,4] and, indeed, for the superconducting cuprates a flat continuum extending to at least 2 eV has been observed. In the superconducting state the frequency distribution of the low-frequency tail of the continua changes to reflect the quasiparticle spectra renormalization due to superconducting fluctuations. Opening of a superconducting gap, $\Delta(\mathbf{k})$, causes suppression of the low-frequency part of the continua, and the scattering across the superconducting gap (breaking the Cooper pairs) develops a peak, the so called 2Δ-peak [5], in the Raman spectra. The recent resonance Raman study of the 2Δ-peak and the normal state continuum concludes that the peak truly results from renormalization of the continuum in the superconducting state [6]. The shape and intensity of the 2Δ-peak in cuprate superconductors where the gap function $\Delta(\mathbf{k})$ is believed to be anisotropic have been discussed in a number of theoretical studies [7] based on earlier work on conventional superconductors [8]. These calculations produce a peak similar to the experimental 2Δ-peak in the superconducting state without considering the normal state continuum, but no one yet has suggested a model that describes the continuum and its renormalization starting from the normal state intensity. There is a general consensus, however, (1) that the position of the peak is proportional to a weighted average of the superconducting gap value near the Fermi surface and that the shift of the peak position for various scattering geometries contains information about the anisotropy of the superconducting order parameter; (2) that the peak intensity is proportional to the density of the superconducting condensate (Cooper-pair density).

The Raman intensity is related to the Raman response function via the fluctuation-dissipation theorem,

$$I(\omega, T) \propto [1 + n(\omega, T)]\chi''(\omega, T), \qquad (1)$$

where $n(\omega, T)$ is the Bose factor. In the conventional calculations extended for an anisotropic gap function, the Raman response function is [7,9]

$$\chi''_{sc}(\omega, T) = \sum_{\mathbf{k}} \gamma(\mathbf{k})^2 \zeta''(\mathbf{k}, \omega, T). \qquad (2)$$

Here $\gamma(\mathbf{k})$ is a light scattering vertex (or a Raman form factor) and $\zeta(\mathbf{k}, \omega, T)$ is a Tsuneto function

$$\zeta''(\mathbf{k},\omega,T) = \frac{\Delta(\mathbf{k})^2}{E(\mathbf{k})^2}\tanh\left[\frac{E(\mathbf{k})}{2k_BT}\right][\pm\delta(2E(\mathbf{k})\mp\omega)]. \tag{3}$$

In Eq. (3) $E(\mathbf{k})^2 = \epsilon(\mathbf{k})^2 + \Delta(\mathbf{k})^2$ is the square of the quasiparticle energy and $\epsilon(\mathbf{k})$ is the fermionic band dispersion relative to the Fermi energy. The Raman form factor $\gamma(\mathbf{k})$ in Eq. (2) depends on scattering geometry and the specific model for photon coupling to the quasiparticle excitations. For excitations of B_{1g} $(x^2 - y^2)$ symmetry, which are the subject of the current work, the form factor has nodes for wave vectors along the diagonals $|k_x| = |k_y|$ and is peaked around antinode wave vectors $\{\mathbf{k}_{\max}\} = \{(0, \pm\frac{\pi}{a})$ and $(\pm\frac{\pi}{a}, 0)\}$, where the superconducting gap magnitude is believed to reach its maximum value $2\Delta_{\max}$. Thus for the B_{1g} symmetry $\chi''_{sc}(\omega,T)$ is peaked about $2\Delta_{\max}(T)$. The temperature and/or magnetic field dependence of the 2Δ-peak position and intensity probes the magnitude of the superconducting gap and the Cooper-pair density.

The overdoped $Tl_2Ba_2CuO_{6+\delta}$ single crystal used in this study was grown as described in Ref. [10] and has superconducting transition at $T_c = 26$ K ($\Delta T_c = 2$ K).

The magnetization measurements have been performed with a 7 T SQUID magnetometer (Quantum Design). The magnetization curves as a function of field perpendicular to the *ab* plane of the sample surface at temperatures between 2 and 15 K are shown in Fig. 1. In the inset the temperature dependence of the irreversibility field, $H_{\rm irr}(T)$, extracted from the magnetization hysteresis curves is presented. Analogously to the $H^*(T)$ data (see Ref. [1]), $H_{\rm irr}(T)$ starts from low values around T_c, has a positive curvature, and continues to rise sharply to the lowest measured temperature (2 K).

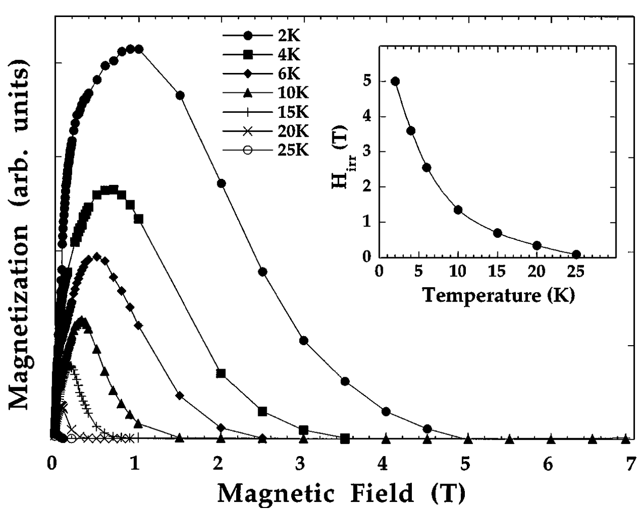

FIG. 1. The field dependence of magnetization (**H** normal to *ab* plane) measured at temperatures between 2 and 25 K. Inset: The temperature dependence of irreversibility field obtained from the magnetization hysteresis curves.

The Raman spectra were measured in the backscattering geometry from the crystal mounted in a continuous helium flow optical cryostat (Oxford Instruments). The cryostat was installed in a bore of a superconducting magnet (Oxford Instruments). The applied magnetic field was normal to the *ab* plane of the sample surface. Circularly polarized light of different handedness for the excitation and the scattered beams were used to select mainly the B_{1g} symmetry spectral component [assuming $D_{4h}(I4/mmm)$ symmetry]. The B_{2g} component is known to be quite weak. A resonance Raman study of $Tl_2Ba_2CuO_{6+\delta}$ has shown that phononic scattering resonates much stronger toward violet excitation than the electronic scattering [6]. To minimize the strength of the phononic scattering and of the luminescence contribution [11] superimposed on top of electronic scattering continuum, we have used the red 6471 Å excitation from a Kr^+ laser. To reduce the heating by laser illumination, we used only about 1 mW of the incident laser power focused onto a 50 μm diameter spot on the *ab* plane of the as grown mirrorlike crystal surface [12]. In the data analysis we account for about 2 K laser heating. The spectra were analyzed by a triple grating spectrometer with a liquid nitrogen cooled CCD detector.

Figure 2 shows the low-frequency electronic Raman continuum and its redistribution as a function of the

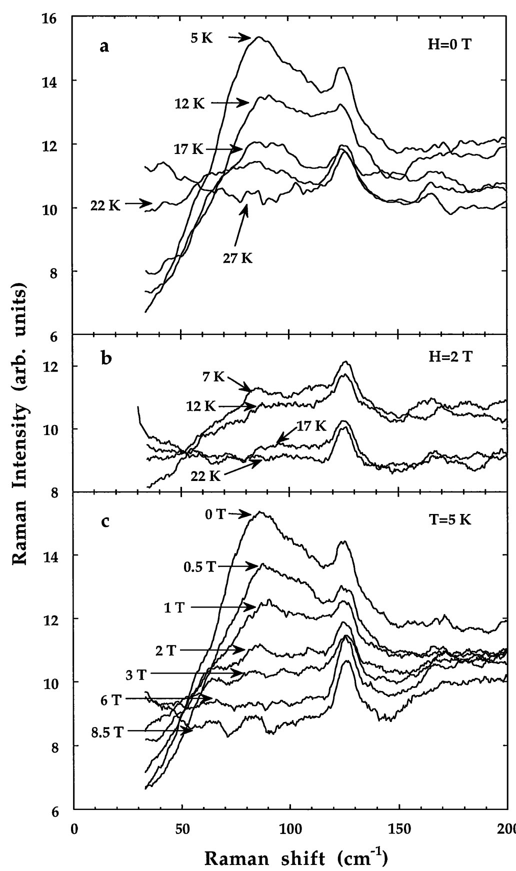

FIG. 2. The B_{1g}-symmetry Raman spectra as a function of temperature (a) at zero field, (b) at 2 T field, and (c) as a function of field normal to the *ab* plane at 5 K. Note offset of the intensity scale.

temperature at zero and 2 T field and of the field at 5 K [13]. In the normal state (above the superconducting transition) the continuum is flat with a slight rise below 50 cm^{-1} [14]. The spectral shape at 27 K and zero field matches well the shape at 5 K and 8.5 T. Cooling or field reduction drives the crystal into the superconducting state, and the spectra exhibit an intensity reduction below 60 cm^{-1} simultaneously with development of a 2Δ-peak. As a result of the intensity redistribution, all spectra cross around 60 cm^{-1}. For zero field and 5 K the peak position is at $\approx$85 cm^{-1} ($\approx 4.9 k_B T_c$). Below the superconducting transition the position of the peak that is proportional to $2\Delta_{\max}(T,H)$ weakly depends on both conditions, temperature and field, showing a slight softening at higher temperatures and/or fields where the peak intensity becomes weak. This implies that the superconducting gap opens up very rapidly near the boundaries of the superconducting phase diagram (below T_c and H_{c2}).

In Fig. 3 we show the 2Δ-peak intensity as a function of temperature for fields between 0 and 3 T. For a lack of a fitting function, we determine the intensity as a difference between the intensity in the maximum of the peak and the spectral crossing point ($\approx$60 cm^{-1}) intensity (see Fig. 2). The peak is associated with Cooper pair breaking excitations; thus the intensity should be proportional to the density of the superconducting condensate. Quite surprisingly, the intensity shows a linear temperature dependence down to almost the lowest measured temperature for all measured fields. This temperature dependence cannot be obtained within the framework of the conventional model calculations [Eqs. (1)–(3)], and it points out the importance of the incoherent processes.

In Fig. 4 we plot the 2Δ-peak intensity in the mixed state as a function of the field for temperatures between 5 and 22 K. The intensity exhibits nonlinear behavior. The 2Δ-peak is clearly present at values of temperature and field above the irreversibility line $H_{\text{irr}}(T)$ [see, for example, the 12 K, 2 T spectrum in Fig. 2(b)].

We interpret the nonlinear intensity drop as a result of the quasiparticle density renormalization in the vicinity of Abrikosov vortex lines in position space and in the region close to the antinodes $\mathbf{k}_{\max}$ in momentum space. In the semiclassical approach and for fields $H_{c1} \ll H \ll H_{c2}$ the local quasiparticle energy, $E_{\text{mixed}}(\mathbf{k},\mathbf{r})$, is Doppler shifted by the local superfluid velocity $\mathbf{v}_s(r) = \hbar/2m_e r$ at a distance $\mathbf{r}$ from the vortex core [15,16]

$$E_{\text{mixed}}(\mathbf{k},\mathbf{r}) = E(\mathbf{k}) + \hbar \mathbf{v}_s(r) \cdot \mathbf{k} . \tag{4}$$

As a result, the Tsuneto function, $\zeta_{\text{mixed}}(\mathbf{k},\mathbf{r},\omega,T)$, is $\mathbf{r}$ dependent in the mixed state and an averaging over the area in the vortex vicinity for the Raman response function is required:

$$\chi''_{sc}(\omega,H,T) = \sum_{\mathbf{k}} \frac{\gamma(\mathbf{k})^2}{\pi R^2} \times \int_\xi^R \int_0^{2\pi} \zeta''_{\text{mixed}}(\mathbf{k},\mathbf{r},\omega,T)\, d\phi\, r\, dr . \tag{5}$$

Here $R(H) \sim \xi\sqrt{H^*_{c2}/H} < \lambda$ is half of the intervortex distance. The Doppler shift results in partial redistribution of the 2Δ-peak intensity to other frequency regions, causing the peak broadening. The remaining intensity at $\omega = 2\Delta$ is governed by that quasiparticle density that does not undergo the shift; for each momentum $\mathbf{k}$ the nonshifted quasiparticle density is collected from a line in position space where the superfluid velocity $\mathbf{v}_s$ is normal to the momentum $\mathbf{k}$. On the other hand, the Doppler shift is in inverse proportion to the distance from the vortex core. Bearing in mind that the Raman form factor

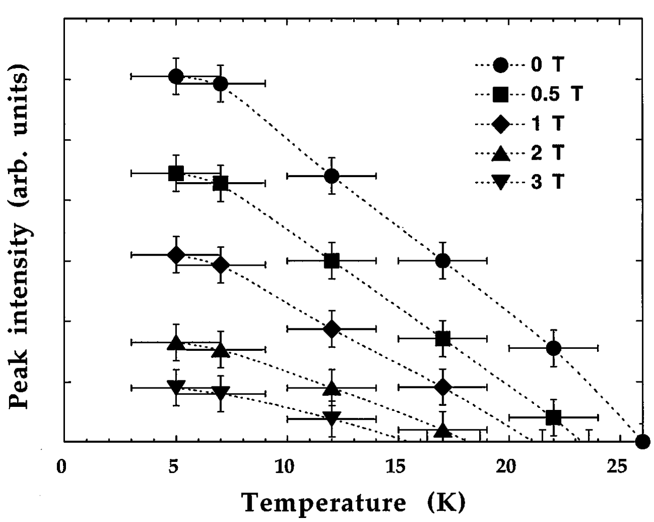

FIG. 3. The temperature dependence of the 2Δ-peak intensity for the fields between 0 and 3 T.

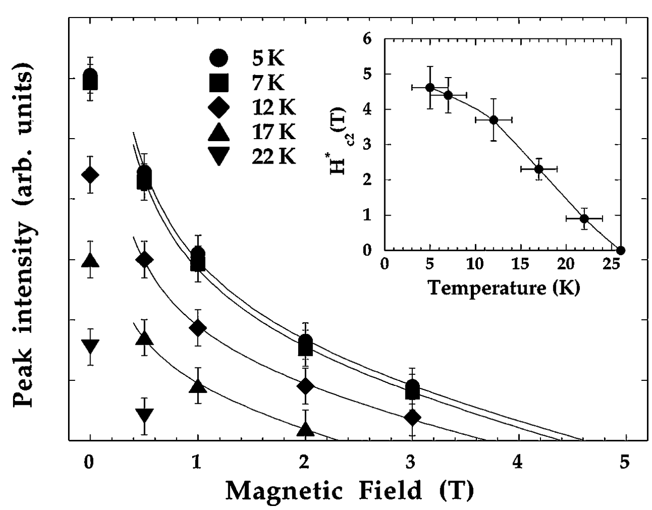

FIG. 4. The field dependence of the 2Δ-peak intensity for the temperatures between 5 and 22 K. Lines present fits of the scaling function in Eq. (6) to the data. Inset: The temperature dependence of the effective upper critical field obtained from the fit.

is peaked at the antinodes' momenta, for high enough fields the response function is in a good approximation proportional to

$$\chi''_{sc}(2\Delta, H) \propto \frac{1}{\pi R^2} \int_{\xi}^{R} \int_{0}^{2\pi} \delta(\hbar \mathbf{v}_s(r) \cdot \mathbf{k}_{\max})\, d\phi\, r\, dr$$
$$\propto \xi \left(\sqrt{\frac{H^*_{c2}}{H}} - \frac{H}{H^*_{c2}} \right). \quad (6)$$

The nonshifted intensity is collected from the points in the vicinity of antinodes $\mathbf{k}_{\max}$ in momentum space and from a line through the vortex core parallel to $\mathbf{k}_{\max}$ with a weight proportional to r^2 in position space. For this model, the vortex cores overlap along the antinode directions at the effective critical field H^*_{c2}. We note that the nonlinear field dependence does not rely on the symmetry of the superconducting order parameter and therefore should be also valid for conventional superconductors.

Figure 4 shows the fit of the scaling function (6) to the experimental peak intensity data. In the inset the temperature dependence of the extracted parameter $H^*_{c2}(T)$ is shown. In contrast to the behavior of $H_{\mathrm{irr}}(T)$ (Fig. 1), $H^*_{c2}(T)$ exhibits a temperature dependence similar to that in the conventional superconductors with a negative curvature. Since $H^*_{c2}(T) > H_{\mathrm{irr}}(T)$ over the measured temperature range, we conclude that the local superconducting order (2Δ-peak) is present at temperatures and fields above $H^*(T)$ and/or $H_{\mathrm{irr}}(T)$. The latter result is in agreement with a recent magneto specific heat study [17] where the mixed state is suggested to exist above the irreversibility transition.

In conclusion, we have studied the field and temperature dependence of magnetization and of low-frequency electronic Raman continuum renormalization in an overdoped $Tl_2Ba_2CuO_{6+\delta}$ single crystal. The superconducting state spectra exhibit a 2Δ-peak about $4.9 k_B T_c$. The position of the peak weakly softens as a function of temperature and/or field around the superconducting transition. The peak intensity shows a linear scaling as a function of temperature below $T_c(H)$, but is a nonlinear function of field below $H^*_{c2}(T)$. We suggest that this nonlinear intensity drop is a result of the quasiparticle density renormalization in the vicinity of Abrikosov vortex lines in position space and close to the antinodes in momentum space. We extract the temperature dependence of the upper critical field from the fit of a suggested model to the field and temperature dependent 2Δ-peak intensity. We find that, in contrast with the nonconventional temperature dependence of the irreversibility field, the effective upper critical field displays conventional temperature dependence with a negative curvature and saturation at low temperatures about 5 T. The latter provides an estimate of the in-plane superconducting coherence length $\xi \approx 100$ Å, considerably longer than for optimally doped superconductors. The local superconducting order parameter was found to be present above the irreversibility temperature and/or field.

We thank A. Carrington, A. van Dalen, K. E. Gray, A. J. Leggett, D. L. Maslov, and M. Zapotocky for valuable discussions and N. N. Kolesnikov for providing the sample. This work was supported by NSF Grant DMR 93-20892 and cooperative agreement DMR 91-20000 through the STCS.

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