Sum Rules and Interlayer Infrared Response of the High Temperature $YBa_2Cu_3O_y$ Superconductor in an External Magnetic Field

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We present infrared magneto-optical measurements of the c-axis conductivity of $YBa_2Cu_3O_y$ in both the underdoped ($y = 6.67$ and 6.75) and optimally doped ($y = 6.95$) regimes. We show that modest c-axis magnetic fields radically modify the condensate formation and restore conventional BCS-like energetics. Additionally, we demonstrate the pivotal role of interplane coherence in the anomalous high-energy contribution to the superfluid density.

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The theory of Bardeen, Cooper, and Schreiffer (BCS), undisputed for metallic superconductors, dictates that the transition involves a narrow interval of energies on the order of k_BT_c. Arguably, the most significant departure from the BCS scheme in high-T_c cuprate superconductors is revealed by optical studies [1,2], which indicate that electronic processes occurring on the energy scale $(10^2–10^3)k_BT_c$ are often involved in the formation of the superconducting condensate [2–9]. These high-frequency optical effects can be interpreted in terms of electronic kinetic energy savings at $T < T_c$ [10,11], at odds with predictions of the BCS theory.

The infrared (IR) optics technique offers an unparalleled window into the formation of the condensate in superconductors due to a well-known relationship between the integral of the real part of the complex optical conductivity $\hat{\sigma}(\omega) = \sigma_1(\omega) + i\sigma_2(\omega)$ and the electronic kinetic energy $K_\mathbf{r}$ in the direction $\mathbf{r}$ [10–13],

$$\int_0^\Omega d\omega\sigma_1(\omega) = \frac{\pi e^2 a_\mathbf{r}^2}{2\hbar^2}K_\mathbf{r}. \quad (1)$$

Here $a_\mathbf{r}$ is the lattice spacing in the $\mathbf{r}$ direction. The conductivity of a superconductor at $T < T_c$ has two contributions: $\sigma_{1,\mathbf{r}}^{SC}(\omega) = \rho_{s,\mathbf{r}}\delta(w) + \sigma_{1,\mathbf{r}}^{reg}(\omega)$, where the first term accounts for the response of the condensate and the second stands for the response of charges not participating in the pairing. Low-frequency spectral weight lost across the transition is transferred to the superfluid density $\rho_{s,\mathbf{r}} = \pi e^2 n_s/2m_\mathbf{r}^*$, where n_s is the superconducting carrier density and $m_\mathbf{r}^*$ is the pair effective mass. Provided changes of $K_\mathbf{r}$ are small, the conductivity follows the Ferrel-Glover-Tinkham sum rule [14]:

$$\int_0^\Omega d\omega\sigma_{1,\mathbf{r}}^N(\omega) = \rho_{s,\mathbf{r}} + \int_{0^+}^\Omega d\omega\sigma_{1,\mathbf{r}}^{SC}(\omega). \quad (2)$$

N denotes the normal state, and the cutoff Ω is usually comparable to few energy gap values. In many cuprates, however, the suppression of the conductivity in this frequency range is insufficient to fully account for the superfluid density [2–9]. The Hirsch kinetic energy sum rule [10] then allows one to interpret this additional contribution as a lowering of the electronic kinetic energy:

$$\rho_{s,\mathbf{r}} = \int_{0^+}^\Omega d\omega[\sigma_{1,\mathbf{r}}^N(\omega) - \sigma_{1,\mathbf{r}}^{SC}(\omega)] + \Delta K_\mathbf{r}. \quad (3)$$

An experimental survey of the energy scales associated with the superconducting condensate requires a perturbation that destroys or suppresses superconductivity, such as temperature, doping, and/or magnetic field. The former two approaches are easily achieved but may lead to artifacts in the analysis since they impact multiple coexisting interactions, thus obscuring processes directly related to the condensate formation [15]. The magnetic field competes with superconductivity without introducing disorder; therefore, magneto-optical experiments are uniquely suited for exploring the condensate formation without significantly altering other properties.

Here, we use IR magneto-optical reflectance measurements to characterize the interplane electrodynamics across the phase diagram of $YBa_2Cu_3O_y$, a prototypical high-T_c cuprate. We focused on the interplane (c-axis) response, where the condensate formation anomalies are most prominent. Doping trends seen in the c-axis energetics mirror in-plane results [2–4,6–8], supporting the notion that it is the same phenomenon which is probed in two different polarizations of IR experiments [16]. Our experiments show that high-energy contributions to the c-axis superfluid density in underdoped YBCO are eliminated with the application of modest magnetic fields, suggesting that kinetic energy lowering is not essential to high-temperature superconductivity.

Very clean YBCO single crystals studied in this work were grown in pure Y_2O_3 crucibles using a flux method [17]. They were annealed to oxygen dopings of $y = 6.67$, 6.75, and 6.95, leading to sharp superconducting transitions at temperatures $T_c = 60$, 65, and 93 K, respectively [18]. c-axis polarized magnetoreflectance measurements were performed over a broad range of frequency (18–35 000 cm^{-1}) and magnetic field (0–8 T) and are described in greater detail in Refs. [19,20]. Reflectance data with appropriate extrapolations were transformed using the Kramers-Kronig relations to obtain the optical conductivity $\hat{\sigma}(\omega)$. Figures 1(a)–1(c) show the real part $\sigma_1(\omega)$. At optimal doping [Fig. 1(c)] the optical conductivity at T_c is flat in the far IR, followed by a series of optically active phonons from 100–700 cm^{-1}. As temperature is lowered below T_c, far-IR conductivity levels drop and spectral weight (SW) is transferred into the superconducting δ peak at $\omega = 0$. In underdoped samples [Figs. 1(a) and 1(b)], however, the suppression of the far-IR conductivity begins in the pseudogap state at T above T_c and is accompanied by transfer of spectral weight at mid-IR frequencies [21]. The asymmetrical mode near 450 cm^{-1} [22,23], which is already visible in the pseudogap state, grows dramatically below T_c. All these findings are consistent with earlier systematic studies of YBCO [24–26].

The dominant effects of the c-axis magnetic field H on the 8 K conductivity of both underdoped samples [Figs. 1(a) and 1(b)] are the simultaneous softening or weakening of the asymmetric mode near 450 cm^{-1} and the growth of the high-frequency tail of the phonon at 320 cm^{-1}. In the optimally doped sample, background conductivity levels increase with field, and the resonance at 800 cm^{-1} is suppressed.

Next we present, in Figs. 1(d)–1(f) and 2, the key experimental results of this work revealing the field-induced redistribution of the electronic spectral weight in the conductivity. It is instructive to introduce the integral spectral function $\Delta N_{T_c}(\omega, H) = \int_{0^+}^{\omega} d\omega' [\sigma_1(\omega', T_c, 0\ \mathrm{T}) - \sigma_1(\omega', 8\ \mathrm{K}, H)]$ quantifying the spectral weight transferred

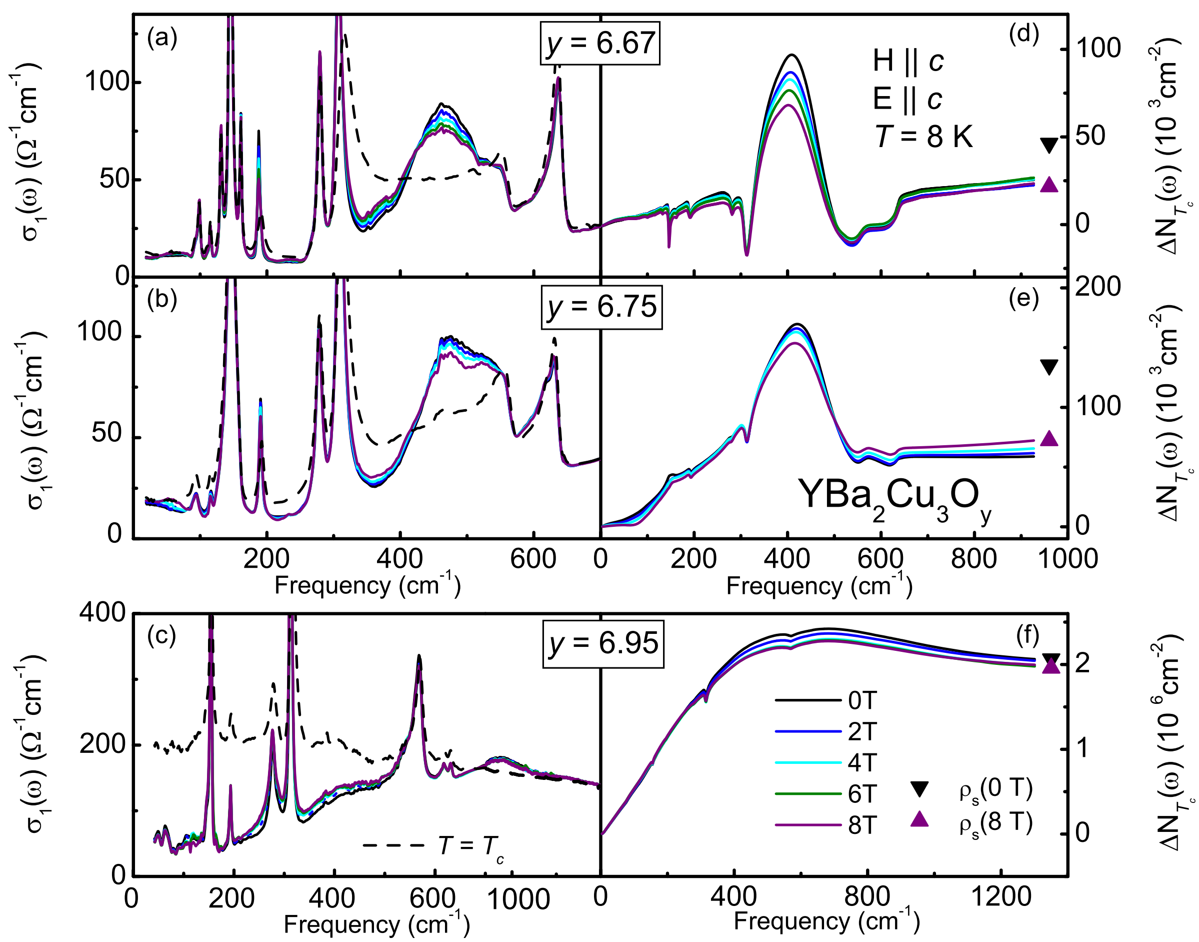

FIG. 1 (color online). Condensate formation revealed by IR magneto-optics for $YBa_2Cu_3O_y$ crystals with oxygen content $y = 6.67$ (top), $y = 6.75$ (middle), and $y = 6.95$ (bottom). Magnetic field is oriented parallel to the c axis. Left panels: Optical conductivity at $T = 8$ K in magnetic field (solid curves) and at T_c (dashed curves). Right panels: Difference in integrated SW between normal ($T > T_c$) and superconducting state $\Delta N_{T_c}(\omega, H) = \int_{0^+}^{\omega} d\omega' [\sigma_1(\omega', T_c, 0\ \mathrm{T}) - \sigma_1(\omega', 8\ \mathrm{K}, H)]$. Line legend is common for all panels.

to the $\delta(\omega)$ peak from the frequency region confined to ω. Values of ρ_s [calculated from $\sigma_2(\omega)$ [27]] are plotted for 0 and 8 T on the right-hand axes of the main panels of Fig. 1 in the same units as the SW change, and they appear for all fields and dopings in Fig. 2. Figure 2 also displays the values of $\Delta N_{T_c}(\Omega_c)$ for $H = 0$–8 T. We used cutoff frequencies $\Omega_c = 1000\ \mathrm{cm}^{-1}$ for underdoped samples and $1300\ \mathrm{cm}^{-1}$ for the optimally doped crystal because field-induced changes of raw reflectance become negligibly small beyond these values. Lastly, we have identified the difference between ρ_s and $\Delta N_{T_c}(\Omega_c)$ as the high-frequency contribution to the superconducting condensate. This term is labeled as ΔK_c to underscore the proposed link to the kinetic energy change.

Proceeding with discussion of the field-induced redistribution of the electronic SW, we will consider first the $y = 6.67$ crystal. The data in Fig. 1(d) show that at $H = 0$ (black curves) the background level of $\Delta N_{T_c}(\omega)$ increases with frequency, with changes to phonons and the asymmetrical mode appearing as sharp features. Importantly,

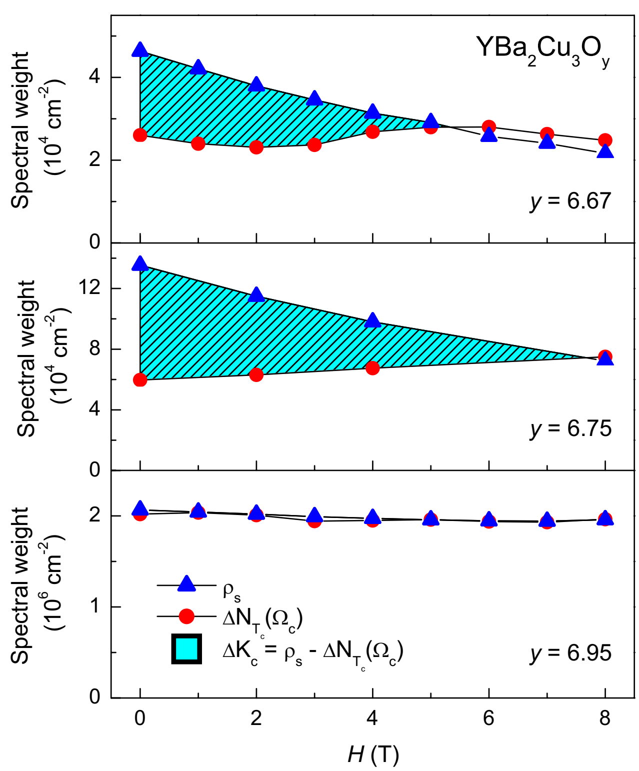

FIG. 2 (color online). Comparison of spectral weight redistribution in c-axis magnetic field for $YBa_2Cu_3O_y$ at $y = 6.67$ (top panel), 6.75 (middle), and 6.95 (bottom) doping levels. Red circles represent the value of $\Delta N_{T_c}(\Omega_c, H)$ at cutoff frequencies $\Omega_c = 1000\ \mathrm{cm}^{-1}$ for the underdoped crystals and $\Omega_c = 1300\ \mathrm{cm}^{-1}$ for the optimally doped system. Blue triangles indicate total superfluid density at each value of magnetic field. High-energy contributions (shaded regions) were inferred from $\rho_s - \Delta N_{T_c}(\Omega_c, H)$ and can be interpreted as kinetic energy change ΔK_c via Ref. [10].

$\Delta N_{T_c}(\Omega_c)$ is nearly unchanged for $H \parallel c$. The asymmetrical mode and the $320\ \mathrm{cm}^{-1}$ phonon in the conductivity are significantly modified by field, but the majority of the SW transfer takes place between these two features, conserving the finite-frequency SW in the far-IR region. Figure 2 illustrates trends in the field dependence of $\Delta N_{T_c}(\Omega_c)$ and ρ_s; the latter is suppressed by about 50% at the highest field in both underdoped samples. At zero field the finite-frequency SW lost below Ω_c only constitutes half of that gained by the superconducting condensate, implying that the remaining portion of the condensate must be transferred from higher energies. As magnetic field is applied we see in greater detail the effect apparent in Fig. 1: $\Delta N_{T_c}(\Omega_c)$ is roughly constant, while ρ_s is suppressed nearly linearly with H. Thus, the shrinking shaded regions in Fig. 2 represent the waning contribution of higher-energy SW to the condensate. This result is significant since, in view of Eq. (3), it implies that the energetics of the superconducting transition are dramatically modified by magnetic field.

Continuation of this analysis across the phase diagram reveals other interesting trends. Features of the $H \parallel c$ data for the second underdoped sample ($y = 6.75$) are generally similar to those discussed for $y = 6.67$, but data for the optimally doped crystal, shown in Fig. 2 (bottom), exhibit important differences. First, there is no high-energy contribution to the condensate observed at zero field: $\Delta N_{T_c}(\Omega_c)/\rho_s \simeq 1$ with high accuracy. Second, this condition is maintained up to the highest measured fields, as changes in ρ_s consistently match those in $\Delta N_{T_c}(\Omega_c)$. Thus, at optimal doping the superconducting transition involves no high-energy transfer of spectral weight.

In light of the introductory comments on the energetics of condensate formation, it is clear that the magneto-optics data presented in Figs. 1 and 2 for $H \parallel c$ seriously undermine the notion of kinetic-energy-driven superconductivity. We see that the high-energy contribution to the condensate, which can be interpreted in terms of kinetic energy change in underdoped samples, is mostly eliminated by 5–8 T. This evidence for a robust superconducting state in the absence of a substantial kinetic energy change weakens the case for this type of condensation mechanism. Furthermore, effects attributable to kinetic energy change never appear at optimal doping.

Remarkably, the conventional condensation scheme has been restored in underdoped samples via field-induced modification of processes occurring at energies exceeding the energy scale of the magnetic field by several orders of magnitude. Equally surprising is the giant (50%) depression of the superfluid density observed in underdoped samples despite the fact that the maximum field in our experiments is much smaller than the pair-breaking field [28,29]. We propose that both effects share a common origin related to the interlayer phase coherence. First, note that pancake vortices initiated by the $H \parallel c$ field are likely to be misaligned in the c direction due to pinning [30]. This vortex meandering is known to introduce a phase difference $\phi_{n,n+1}(r)$ between neighboring layers [31,32]. An immediate consequence is that the average strength of the interlayer Josephson coupling $J \propto J_0 \cos(\phi_{n,n+1})$ is reduced, leading to suppression of the c-axis superfluid density governed by J. The finite interplane phase difference in the vortex state is of direct relevance to a scenario for a contribution to ρ_s originating from energies much larger than $k_B T_c$ proposed by Ioffe, Millis, and Shah (IMS) [33,34]. According to these calculations the high-energy contribution is indeed expected, provided the superconducting transition occurs from a state where pairing already exists but phase coherence between the planes is still missing and is only established below T_c. Importantly, vortex meandering is in direct competition with the restoration of interlayer phase coherence. Extending this argument to the IMS picture one can conclude that the above competition reduces and eventually eliminates the high-energy contribution to the δ peak, in accordance with our findings. It is important to note that these theoretical results in Refs. [34,35] are sensitive to the details of the bilayer coupling [34], and that a different behavior may be expected for single-layer cuprate compounds. The central role of phase fluctutation is underscored by the fact that these enigmatic field-induced effects are observed only in the underdoped region of the phase diagram, where accurate terahertz experiments [35] and Nernst measurements [36] have established superconducting pairing above T_c.

In summary, our data demonstrate that relatively small magnetic fields can modify spectral weight redistribution in a prototypical family of cuprate superconductors over an anomalously large energy scale. Fields $H \parallel c$ of 8 T suppress the high-frequency contribution to the zero-ω $\delta(\omega)$ peak but do not completely destroy the superfluid density, leaving a robust superconducting state. These findings point toward a more BCS-like formation of the superconducting condensate even in underdoped samples where earlier zero-field data pointed to a highly exotic condensation process. The large energy scale of the associated electronic processes then seems to be a generic property of correlated electron systems, but only peripherally related to superconductivity. Furthermore, we have proposed a scenario in which the changes in spectral weight redistribution are linked to a reduction in phase coherence in the superconducting state, primarily due to vortex meandering.

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