I. INTRODUCTION

Infrared spectroscopy is a powerful tool for studying condensed-matter systems. It provides numerous insights due to its sensitive resolution of subtle spectral characteristics, but also for its ability to utilize data from a wide frequency range to determine system properties based on conservation laws and sum rules. For example, one of the Kubo conductivity sum rules relates the integrated real part of the optical conductivity to the strength of the zero-frequency electronic kinetic energy during the superconducting transition. The spectral weight shift during the transition is then described by

$$\rho_{\omega} = \int_0^{\Omega} d\omega \sigma_1(\omega) = \frac{\pi e^2 a_c^2}{2 h^2} K_r. \tag{2}$$

A sum rule which is quintessential for the understanding of superconductivity was formulated by Ferrer, Glover, and Tinkham (FGT); it equates the spectral weight lost in the superconducting gap to the strength of the zero-frequency superconducting condensate, quantified by the superfluid density $\rho_s$,

$$\rho_s = \int_0^{\Omega} d\omega \left[ \sigma_1^N(\omega) - \sigma_1^{SC}(\omega) \right]. \tag{3}$$

The FGT sum rule is valid for elemental superconductors which are well-described by the theory of Bardeen, Cooper, and Schrieffer (BCS). In this case the difference in spectral weight between the normal and superconducting states reaches the full value of $\rho_s$ by an integration cutoff of only a few gap values. However, for many high-$T_c$ cuprate superconductors, optical experiments have revealed that the difference in conductivity only constitutes a portion of the superfluid when integrated up to several gap values. This behavior implies that the extra spectral weight in the superconducting condensate must be transferred from much higher energies, and also that the system experiences a lowering of electronic kinetic energy during the superconducting transition. The spectral weight shift during the transition is then described by

$$\rho_{\omega} = \int_0^{\Omega} d\omega \sigma_1(\omega) = \frac{\pi e^2 a_c^2}{2 h^2} K_r, \tag{4}$$

and is illustrated schematically in Fig. 1.

High-energy effects are common to at least four different families of cuprates, appearing in both $ab$-plane and $c$-axis measurements. Typically, they are only observed at doping levels below the optimal value, and not in the overdoped regime. Such broad occurrence and similarity in doping dependence prompted many to consider whether the lowering of electronic energy was an essential component to the superconducting mechanism in this class of materials. These questions sparked a need for experiments that could unambiguously identify the high-energy spectral weight transfer and access a weakened superconducting state to verify if the condensation scheme remained intact when the order parameter was suppressed. Infrared measurements in which the incident electric field is polarized parallel to the $c$ axis are highly sensitive to changes in kinetic energy and therefore uniquely suited to address this experimental need. Further, the desired perturbation can be provided by application of an
external magnetic field, which competes with superconductivity without promoting disorder. Consequently, the technique of infrared optics in magnetic field is an ideal tool for probing these phenomena.

We have previously reported on infrared measurements\(^\text{19}\) of underdoped YBa\(_2\)Cu\(_3\)O\(_y\), a prototypical high-\(T_c\) superconductor, recording the evolution of the spectral weight balance in magnetic field \(H\parallel c\). We found that, from a sum-rule–analysis point of view, the high-field data were less anomalous than at zero field. Here, we extend these results to include sum-rule analysis in magnetic field for data recorded at higher temperatures, as well as for magnetic fields oriented parallel to the CuO\(_2\) planes. Section II provides details regarding our infrared magneto-optical experiment and reflectance data, and Sec. III presents the calculated optical conductivity. The sum-rule analysis of the conductivity data is described in Sec. IV and, lastly, connections to interlayer phase coherence and vortex lattice resonance modes are discussed in Sec. V.

II. INFRARED REFLECTANCE EXPERIMENT IN MAGNETIC FIELD

High-quality \(ac\)-face single crystals of YBa\(_2\)Cu\(_3\)O\(_y\) (YBCO) were grown using a flux method\(^\text{20}\) and annealed to achieve doping levels of \(y\approx 6.67, 6.75\) (both underdoped), and 6.95 (optimally doped). Transport measurements\(^\text{21}\) reveal sharp transitions to the superconducting state at 60, 65, and 93 K, demonstrating the high purity of the crystals. For each doping several single crystals from a single batch were assembled to form mosaics approximately \(3 \times 6 \text{ mm}^2\) in size.

Near-normal reflectance measurements were performed in a broadband Fourier transform spectrometer over a frequency range of 18–35 000 cm\(^{-1}\). First, absolute reflectance was obtained at temperatures \(T=8–295\) K by measuring sample reflectance relative to a stainless-steel reference mirror and normalizing by the reflectance of the sample coated with Au.\(^\text{22}\) Then, changes in reflectance induced by magnetic field \(H\) were recorded via the ratio \(R(T, H)/R(T, H=0)\) in a split-coil magnet\(^\text{23}\) for field magnitudes up to 8 T. This step utilized an Al reference mirror to correct for minor spurious effects in the magnet system.

Reflectance spectra for the YBCO system (Fig. 2) are weakly metallic near room temperature, with an upturn toward \(R=1\) as \(\omega\to 0\). The sharp peaks in the far infrared correspond to phonons. As temperature is decreased to \(T_c\), the reflectance of the most metallic, optimally doped sample increases, while that of the underdoped crystals decreases, due to the formation of the pseudogap.\(^\text{24–27}\) At temperatures below \(T_c\), the Josephson plasma edge develops, corresponding to coherent oscillation of the nondissipative superconducting condensate. This feature is characterized by very high reflectance at low frequencies followed by a sharp dip at the Josephson plasma resonance (JPR) frequency \(\omega_p = \sqrt{\mu/\epsilon_c}\). This frequency, a direct measure of the superfluid density, softens with oxygen reduction, (from \(\omega_{p\text{JPR}}\approx 250\) cm\(^{-1}\) at \(y\approx 6.95\) to \(\omega_{p\text{JPR}}\approx 60\) cm\(^{-1}\) at \(y\approx 6.67\)) and stiffens at lower temperatures. Also, in the underdoped crystals a broad, asymmetric feature near 450 cm\(^{-1}\) which is weakly visible above \(T_c\) becomes significantly more prominent in the superconducting state. These results are consistent with previous studies of similar YBa\(_2\)Cu\(_3\)O\(_y\) compounds.\(^\text{24–31}\)

Application of magnetic field parallel to the \(c\) axis in many respects reverses the trends of lowering temperature. As seen in Fig. 3, for underdoped crystals the field softens the JPR and reduces the magnitude of the asymmetric feature. For \(H\parallel c\) no new features are observed in \(R(\omega)\). Magnetic fields \(H/\text{CuO}_2\) (Fig. 4), however, do introduce new absorption features at frequencies below the JPR. The frequency of the dip in \(R(\omega)\) increases with field, moving from...
MAGNETIC FIELD INDUCED MODIFICATION OF...

Reflectance data were transformed via the Kramers-Kronig relations to obtain the optical conductivity $\sigma(\omega)$ over the full frequency half-space, we augment the raw data with appropriate low- and high-frequency extrapolations. In the normal state we assumed a Hagen-Rubens metallic response of the form $(1-R) \propto \omega^{\frac{1}{2}}$ for frequencies below the lowest measured data. A two-fluid form was assumed in the superconducting state. High frequency data were extended to $+\infty$ with a combination of linear and $\omega^{-4}$ asymptotic extrapolations.

The real part of the optical conductivity $\sigma_r(\omega)$ at zero magnetic field is displayed in Fig. 5. The room temperature conductivity is flat overall, interrupted only by a series of infrared-active phonons. For the optimally doped crystal, the background conductivity increases with decreasing temperature to $T_c$, consistent with a metallic system. The underdoped crystals, however, become less conductive upon lowering to $T=T_c$, and reach maximum conductivity levels roughly an order of magnitude smaller than those of the optimally doped case. Further cooling reveals a partial gapping of the Fermi surface, characteristic of the pseudogap. In oxygen-reduced crystals spectral weight (SW) is removed from phonons to create a broad band near 450 cm$^{-1}$. This feature has been previously studied in detail and may be consistent with either a bilayer transverse plasmon mode or a bilayer splitting.

As found in the reflectance, modifications to the conductivity by the magnetic field are strikingly dissimilar in the different field orientations. For H$\parallel$CuO$_2$ (Fig. 6) no new modes appear in $\sigma_r(\omega)$, but substantial changes occur in the phonon region. In the underdoped crystals the field initiates a pronounced shift of SW from the asymmetrical mode back into the phonon at 320 cm$^{-1}$, mirroring the effect of raising tem-
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Resonance appears at low frequencies. With increasing temperature. Direct evidence for the equivalence of increasing magnetic field and temperature is found in Fig. 7, where we plot for the magnetic field and temperature is found in Fig. 7, where we plot for the $y=6.67$ crystal $\sigma_1(\omega)$ at $8$ K (0 and 8 T), 45 K (0 and 8 T) as well as just above $T_c$ at 70 K. Using the 8 K, 0 T result as a starting point, it is clear that both $H$ and $T$ drive the spectrum toward the 70 K curve. The result of raising $T$ to 45 K at zero field is nearly identical to that of raising $H$ to 8 T and keeping $T$ fixed.

The data for fields applied parallel to the CuO$_2$ planes, shown in Fig. 8, exhibit fundamentally different behavior than was observed for $H$ parallel to the CuO$_2$ planes. For magnetic fields $H$ parallel to the CuO$_2$ planes, a resonance appears at low frequencies. With increasing magnetic field the resonance hardens and gains SW. This feature has been shown to have its origin in Josephson vortex lattice dynamics. Other than minor modifications to the electronic background which affect phonon features, minimal field-induced changes occur at higher frequencies.

IV. SUM RULES AND SPECTRAL WEIGHT TRANSFER

To investigate in more detail the energy scales governing the superconducting transition we now compare the low-frequency integrated SW with the superfluid density at key values of temperature and magnetic field. We will show that magnetic fields $H$ parallel to the CuO$_2$ planes, shown in Fig. 8, exhibit fundamentally different behavior than was observed for $H$ parallel to the CuO$_2$ planes. For magnetic fields $H$ parallel to the CuO$_2$ planes, a resonance appears at low frequencies. With increasing magnetic field the resonance hardens and gains SW. This feature has been shown to have its origin in Josephson vortex lattice dynamics. Other than minor modifications to the electronic background which affect phonon features, minimal field-induced changes occur at higher frequencies.

FIG. 6. (Color online) Optical conductivity of YBa$_2$Cu$_3$O$_{6.67}$ single crystals for dopings $y=6.67$ (top), 6.75 (middle), and 6.95 (bottom) at temperatures $T=8$ K (left) and 45 K (right). Magnetic fields up to $H=8$ T are applied parallel to the $c$ axis.

FIG. 7. (Color online) Optical conductivity for YBa$_2$Cu$_3$O$_{6.67}$ single crystal at 8 K (0 and 8 T), 45 K (0 and 8 T) and just above $T_c$ at 70 K. Similarity between 8 K, 8 T curve and 45 K, 0 T curve demonstrates equivalence of temperature and magnetic field $H$ parallel to the CuO$_2$ planes for modification of optical properties.

FIG. 8. (Color online) Optical conductivity of YBa$_2$Cu$_3$O$_{6.67}$ single crystals for dopings $y=6.67$ (top), 6.75 (middle), and 6.95 (bottom) at temperatures $T=8$ K (left) and 45 K (right). Magnetic fields up to $H=8$ T are applied parallel to the CuO$_2$ planes.
Superfluid density closely matches the missing SW at zero field as being driven to lower fields as temperature is increased to 45 K. Comparing SWs in Fig. 10, we see that the impact of magnetic field is simply to redistribute SW within the phonon region that are unchanged by magnetic field. The primary contrast to the mostly flat behavior of $\Delta N_T(\omega)$ is the low-frequency region. We infer that the extra SW in the superfluid must have been transferred from high frequencies. As the magnetic field is increased, the superfluid density is strongly suppressed, in contrast to the mostly flat behavior of $\Delta N_T(\Omega_c)$. Eventually the two curves cross or merge, with the intersection being driven to lower fields as temperature is increased to 45 K. In the data recorded for the optimally doped crystal, the superfluid density closely matches the missing SW at zero field, and neither quantity is changed by magnetic field.

Application of the magnetic field parallel to the CuO$_2$ planes results in a qualitatively different picture of SW transfer. For underdoped crystals, the superfluid density behaves similarly to the $H||c$ orientation, with a strong suppression in field. The low-frequency finite SW change, seen in the bottom panels of Fig. 9, as well as in Fig. 11, is no longer field independent: the limiting values of $\Delta N_T(\omega, H)$ decrease monotonically with magnetic field, mirroring the $\rho_s(H)$ curve. Thus, the high-frequency SW transfer [the difference between $\rho_s$ and $\Delta N_T(\Omega_c, H)$] is not entirely suppressed in the superconducting state. Rather, it is gradually diminished, trending toward zero along with the superconducting order parameter. This behavior continues at higher temperatures for the $y=6.75$ crystal. The optimally doped crystal again shows no SW anomaly, with $\Delta N_T(\omega, H)=\rho_s(H)$ at all fields, even as both are reduced. It should be noted that the field dependence of the magnitudes of the changes for the $y=6.95$ crystal are somewhat extrapolation dependent. However, their equality at all fields holds for any reasonable extrapolation.

The fundamental empirical difference between the results of the two orientations of the magnetic field is the final destination of the SW which is removed from the superfluid density. For $H||c$ the SW is returned to higher frequencies, thus implying that the energy scale of the condensate formation evolves toward a BCS-like regime. Fields applied $H||CuO_2$, on the other hand, reduce the high-frequency SW transfer proportionately to the superfluid density, maintaining a discrepancy between $\Delta N_T(\omega, H)$ and $\rho_s(H)$. This behavior suggests that the mechanism of condensate formation involving transfer of SW from high frequencies remains intact for all values of magnetic field.
V. VORTEX MEANDERING AND INTERLAYER PHASE COHERENCE

The dramatic change in the infrared response at relatively small fields $H || c$ is surprising in several respects. Most notable are the modification of the SW redistribution scheme over an anomalously large energy scale, and the substantial (50\%) reduction in superfluid density seen in underdoped samples at fields much smaller than the pair breaking field.\textsuperscript{39,40} Both of these effects may be consistent with a theoretical perspective involving the wandering of pancake vortices and the subsequent suppression of interlayer phase coherence.\textsuperscript{41,42} In a layered type-II superconductor material with no disorder, it is known that pancake vortices will be well aligned along the $c$ axis, maintaining phase coherence between adjacent planes. A disordered pinning potential, however, will produce a random displacement of vortices from layer to layer. As magnetic field is increased this pinning eventually destroys vortex lines and interlayer phase coherence. Since the interlayer phase difference $\phi_{n+1}$ is intimately related to the interlayer coupling $J$ and superfluid density $\rho_s$ [\(J \approx J_0 \cos(\phi_{n+1}) \approx \rho_s\)], the effect is visible in the infrared data.

The finite interplane phase difference is central to a model of bilayer dynamics proposed by Ioffe, Millis, and Shah (IMS) in which SW derived from energies far above $k_BT_c$ contributes to $\rho_s$.\textsuperscript{43,44} High-energy SW transfer is expected within the IMS picture when the transition occurs between a normal state above $T_c$ characterized by pairing, but no phase coherence, and a SC state with well-defined phase coherence. One can then extend this description to attribute the elimination of high-energy SW transfer to the competition between vortex meandering and restoration of interlayer phase coherence in magnetic field.

FIG. 11. (Color online) See caption for Fig. 10. Magnetic field is applied parallel to CuO$_2$ planes.

Similar shrinking of the energy scale for condensate formation is not seen in the $H // CuO_2$ data, consistent with the above considerations of phase coherence. The Josephson vortices created in this field geometry affect the phase of the superconducting order parameter in a more complicated way than pancake vortices do.\textsuperscript{37} Oscillations of the Josephson vortex lattice result in an interlayer phase relationship which is dynamic and highly frequency dependent, a departure from the simple linear suppression of coherence expected for pancakes. Furthermore, additional complications arise in the analysis of the IR data obtained in this geometry due to the new resonances in the conductivity spectra which are produced by the motion of Josephson vortices. For vortex-dynamics-related features in $\sigma(\omega)$ the distinction between “normal” and “superconducting” SW is no longer as clear as in the zero-field data. Indeed, these resonances are believed to result from oscillations of Josephson vortices and therefore are of superfluid origin. Yet the features appear in the dissipative part of the conductivity at the expense of the suppression of the superconducting $\delta$ peak. Since both the vortex resonance and the Josephson plasma resonance are modified significantly by magnetic field, the SW shifts and possible changes in kinetic energy may be related in a more subtle manner than this analysis allows. Regardless of these complications, the linear scaling of $\Delta N_s(\Omega_s)$ and $\rho_s$ informs us that high-energy SW transfer is not as easily stifled by $H // CuO_2$.

Reflectance measurements were also recorded for both magnetic field geometries at temperatures just above the superconducting transition. In both cases no field-induced modifications to the infrared reflectance were observed within the signal to noise of our data. This result has important implications in relation to the subject of preformed pairing. In this theoretical description of the pseudogap, for temperatures between $T_c$ and the pseudogap temperature $T^*$, Cooper pairs are believed to exist but do not have long-ranged phase coherence.\textsuperscript{45,46} The null result above $T_c$ is consistent with the preformed pairs picture since the primary action of magnetic field is to destroy phase coherence, rather than to break Cooper pairs. For this reason, the magnetic field only impacts the optics when phase coherence is appreciable, below $T_c$. As a result, features in the data which are connected to superconductivity but appear above $T_c$, such as the asymmetric mode, are not modified by field in this temperature range. It would be highly instructive to extend these measurements to higher magnetic fields to determine if the low-field trends are continued.

It should be noted that the kinetic-energy change observed in $c$-axis polarized experiments is not a phenomenon constrained to the interplane conductivity. In fact, kinetic energy lowering at zero field is consistent with angle-resolved photoemission spectroscopy (ARPES) data measured with \textit{ab}-face crystals (see Ref. 10 and references therein). ARPES measurements of underdoped cuprates at the antinodal $[(\pi,0)$ and $(0,\pi)]$ regions of $k$ space reveal indicators of quasiparticle coherence consistent with the IMS picture described above: coherence at temperatures below $T_c$, but not above.\textsuperscript{47,48} Comparison of IR and ARPES data confirms that kinetic-energy change only occurs when this pattern of coherence is observed. Since the $c$-axis electrodynamics are
thought to be strongly determined by the properties of the Fermi surface at the antinodal regions, free of the strong nodal contributions inherent to the CuO$_2$ planes,\textsuperscript{49} interplane measurements are especially sensitive to modifications of kinetic energy. In this way, c-axis experiments can be regarded as a probe of superconductivity in the planes.

This study is not unique in its approach of using magnetic field to tune anomalous properties of the high-$T_c$ superconductors. Recent transport measurements\textsuperscript{50} of the normal state uncovered by magneto-optical studies\textsuperscript{51} of La$_{2-x}$Sr$_x$CuO$_4$ in which c-axis magnetic fields were shown to promote antiferromagnetism in the CuO$_2$ planes.

**VI. CONCLUSION**

The primary finding of this work is that the application of an external magnetic can initiate profound redistribution of spectral weight from the superfluid density to the finite-frequency spectrum. The character of these effects differs depending on the orientation of the field with respect to the CuO$_2$ planes. Fields $H \parallel c$ return weight to high-energy regions of the spectrum, undoing the lowering of kinetic energy observed at zero field. Fields $H \parallel \text{CuO}_2$ place the weight at frequencies on the order of the superconducting gap, maintaining the reduction in kinetic energy. Since it is possible to reduce the interlayer phase coherence to the point where high-energy spectral weight transfer ceases, but a robust superconducting state remains, we must conclude that the reduction in kinetic energy seen at zero field is not a necessary condition for superconductivity. The importance of the phase coherence to this process is supported by the data for fields $H \parallel \text{CuO}_2$: these fields are less destructive to the interlayer phase relationship and leave intact the kinetic-energy reduction intact.

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