Strong-coupling effects in cuprate high-$T_c$ superconductors by magneto-optical studies

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(Received 25 July 2005; published 23 November 2005)

Signatures of strong coupling effects in cuprate high-$T_c$ superconductors have been authenticated through a variety of spectroscopic probes. However, the microscopic nature of relevant excitations has not been agreed upon. Here, we report on magneto-optical studies of the CuO$_2$ plane carrier dynamics in a prototypical high-$T_c$ superconductor YBa$_2$Cu$_3$O$_y$ (YBCO). Infrared data are directly compared with earlier inelastic neutron scattering results by Dai et al. [Nature (London) 406, 965 (2000)] revealing a characteristic depression of the magnetic resonance in H$\parallel$c field less than 7 T. This analysis has allowed us to critically assess the role of magnetic degrees of freedom in producing strong-coupling effects for YBCO system.

Electron pairing in conventional superconducting metals is mediated by phonons. Strong interaction of electrons with the lattice also manifests itself through self-energy effects yielding fingerprints of the electron-phonon spectral function $\alpha^2 F(\omega)$ in the tunneling density of states,$^1$ infrared (IR) conductivity,$^2,3$ or energy band dispersion probed in the angle resolved photoemission spectroscopy (ARPES).$^4,5$ A similar clear understanding of the cuprate high-$T_c$ superconductors is yet to be achieved. Spectroscopic probes of self-energy unequivocally prove the relevance of strong-coupling effects.$^6,7$ However, the microscopic origin of the pertinent spectral function is still debated. Numerous experiments are suggestive of quasiparticle (QP) interaction with a magnetic resonance mode$^8$–$^{14}$ seen in inelastic neutron scattering (INS) experiments.$^{15,16}$ An issue of whether or not the magnetic mode is capable of having a serious impact on the electronic self-energy, in view of the small intensity of the resonance, has been contested on theoretical grounds.$^{17–19}$ Moreover, a recent reexamination of ARPES results$^{20–22}$ has suggested that the totality of data is better described in terms of coupling to phonons and not to magnetic excitations. However, this latter claim is not supported by IR studies of isotopically substituted YBa$_2$Cu$_3$O$_y$ (YBCO), which show no isotope effect for the feature in question.$^{23,24}$ Thus, currently available data leaves ambiguities regarding the roles of lattice and magnetic degrees of freedom in carrier dynamics as well as in the superconductivity of cuprates.

Insights into strong coupling effects may be gained from studies of the QP dynamics in the magnetic field. The rationale for this approach is provided by the work of Dai et al. who discovered that the intensity of the magnetic resonance in the $y=6.6$ YBCO crystal ($T_c=62.7$ K) is suppressed by 20% in a 6.8-T field applied along the c axis.$^{25}$ Other candidate excitations including phonons, or the continuum of spin fluctuations, are unlikely to be influenced by a magnetic field of similar modest magnitude. For this reason, an exploration of the field-induced modifications of the electronic self-energy enables a direct experimental inquiry into the role of the magnetic resonance in QP properties and on a more general level, into an intricate interplay between superconductivity and magnetism in cuprates. Here, we report on studies of a magnetic field dependent QP response in a series of YBCO crystals using IR spectroscopy. Changes of the optical conductivity and of the $\alpha^2 F(\omega)$ spectrum extracted from the data in a 7-T field are found to be within the uncertainty of our measurements. This null result nevertheless allows us to critically assess the strong-coupling scenario in high-$T_c$ superconductors.

We investigated YBCO single crystals with $y=6.50$ ($T_c \sim 31$ K) and 6.65 ($T_c \sim 60$ K) grown by a conventional flux method and detwinned under uniaxial pressure at Central Research Institute of Electrical Power Industry (CRIEPI).$^{26}$ Detwinned crystals are necessary to perform reflectivity measurements with the polarization of IR light along the a axis. In this geometry, the properties studied here unveil the intrinsic dynamics of the CuO$_2$ planes without contamination by the chain segments extending along the b axis.$^{27}$ Field-dependent reflectivity spectra were recorded at University of California at San Diego (UCSD) in the frequency range 20–5000 cm$^{-1}$,$^{28}$ and were supplemented by zero-field reflectance and ellipsometry data up to 5 eV. The magnetic field aligned along the c axis was applied using a superconducting split-coil magnet. Our magneto-optical apparatus enables absolute measurements of reflectivity. The 0 T data obtained using this instrument were found to be consistent with the spectra obtained by means of our compact reflectometer for temperature-dependent reflectance. Owing to the small cyclotron frequency of YBCO, it is appropriate to extract the complex conductivity $\sigma(\omega)=\sigma_r(\omega)+i\sigma_i(\omega)$ from reflectance spectra using the standard Kramers-Kronig equations.

Representative results are displayed in Fig. 1. Here, we plot the raw reflectance spectra measured at $T=5$ K for $y=6.50$ and 6.65 crystals. The spectra for the latter material are in good agreement with the earlier studies of YBCO with similar oxygen content.$^{29}$ Notably, we found that the field-induced changes of the reflectivity are negligibly small either under zero-field cooling or under in-field cooling conditions. In order to quantify the experimental accuracy achievable in our magneto-optical apparatus, in the bottom panel of Fig. 1,
we also show spectra for highly oriented pyrolytic graphite (HOPG) measured with the polarization of the $E$ vector along the graphene sheets. A comparison is warranted by the similarity in the zero-field optical properties of graphite and high-$T_c$ cuprates. In the former system, we are capable of resolving small (less than 1%) changes of the overall reflectivity level as well as weak structure associated with the Landau level transitions triggered by the magnetic field perpendicular to graphene planes.\textsuperscript{30} No such changes are detectable for YBCO.

We proceed by briefly outlining the fundamentals of an IR probe of the electronic self-energy. Interaction of the mobile charges with bosonic excitations leads to a frequency dependence of the scattering rate $1/\tau(\omega)$ in accord with the Allen formula\textsuperscript{31}

$$\frac{1}{\tau(\omega)} = \frac{2\pi}{\omega} \int_0^\omega d\omega' (\omega - \omega') \alpha^2 F(\omega') + \frac{1}{\tau_{\text{imp}}},$$

(1)

where $1/\tau_{\text{imp}}$ is the impurity scattering. Experimentally, the frequency dependence of $1/\tau(\omega)$ can be inferred from the analysis of the complex optical conductivity $\sigma(\omega)$ within the extended Drude model.\textsuperscript{6} $1/\tau(\omega) = \omega_p^2/4\pi \text{Re}[1/\sigma(\omega)]$, where a total plasma frequency $\omega_p^2$ is determined by integration of $\sigma(\omega)$ up to the charge transfer gap. Equation (1) is commonly applied to the analysis of the data for cuprates and provides support for an idea of QPs coupling to a magnetic resonance.\textsuperscript{9,14} Nevertheless, Eq. (1) is not entirely adequate for a superconductor since it completely ignores the effect of the superconducting energy gap $2\Delta$ on the form of the $1/\tau(\omega)$ spectra. In order to treat the impact of the gap and of strong coupling to bosonic modes on equal footing, we used the following result also derived by Allen:\textsuperscript{31}

$$\frac{1}{\tau(\omega)} = \frac{2\pi}{\omega} \int_0^{\omega-2\Delta} d\omega' (\omega - \omega') \alpha^2 F(\omega') \times E \left[ \left(1 - \frac{4\Delta^2}{(\omega - \omega')^2}\right)^{1/2} \right],$$

(2)

where $E$ is the complete elliptic integral of second kind. Although the utility of Eq. (2) is obvious, it is nontrivial to employ this formula for the extraction of $\alpha^2 F(\omega)$ from experimental data since simple inversion prescriptions do not apply in this case.\textsuperscript{32} To circumvent this limitation, Dordevic \textit{et al.} developed a numerical procedure based on the inverse theory that is described in detail elsewhere.\textsuperscript{33} In the bottom panels [(e) and (f)] of Fig. 2, we show the $\alpha^2 F(\omega)$ spectrum extracted in this fashion from the $\mathbf{H}=0$ data. We wish to point out an excellent agreement with INS results for the spin susceptibility $\chi(\omega)$ [open symbols in Fig. 2(e)]\textsuperscript{34} without introducing a frequency offset.\textsuperscript{9} Indeed, both a sharp resonance and a broad incoherent background of the spin susceptibility appear to be reproduced in the $\alpha^2 F(\omega)$ spectrum.\textsuperscript{35}

An important feature of the strong coupling formalism [Eqs. (1) and (2)] is the integral relationship between $1/\tau(\omega)$ and $\alpha^2 F(\omega)$. This relationship implies that a depression of the intensity in $\alpha^2 F(\omega)$ necessarily reduces the magnitude of $1/\tau(\omega)$ and consequently enhances the reflectivity level at all frequencies \textit{above} the resonance mode in the spectral function. In order to quantify the magnitude of possible $\mathbf{H}$-induced changes associated with a depression of the INS resonance in the magnetic field, we adopted the following protocol. We first reduced the intensity of the sharp peak near $\sim 270$ cm$^{-1}$ ($\sim 34$ meV) in the $\alpha^2 F(\omega)$ spectrum by 20%: a factor suggested by INS measurements.\textsuperscript{25} The intensity of broad background remained intact [blue (thick dashed) line in Fig. 2(f)]. Evidently, this modification will produce a conservative estimate of the impact of the INS resonance on IR data. Using the spectral function with the suppressed intensity, we calculated $1/\tau(\omega, 7\, \text{T})$ from Eq. (2) and also $m'(\omega, 7\, \text{T})$ with the help of the Kramers-Kronig analysis. Finally, a combination of $1/\tau(\omega, 7\, \text{T})$ and $m'(\omega, 7\, \text{T})$ allowed us to generate the reflectance spectrum $R(\omega, 7\, \text{T})$ [blue (thick dashed) line in Fig. 2(b)]. Comparing this final output of modeling with the experimental curve for $\mathbf{H}=0$, one finds that the effect of the applied magnetic field is rather small in the far IR, but exceeds 5% at frequencies above 800 cm$^{-1}$. This is further detailed in the inset of Fig. 2, where we present the ratio $\Delta R(\omega, \mathbf{H}) = R(\omega, 7\, \text{T})/R(\omega, 0\, \text{T})$ calculated from the model spectra. These anticipated changes of reflectance exceed the uncertainty of $R(\omega, \mathbf{H})$ in our ap-
paratus and, therefore, should be readily detectable. Empowered by modeling of the data, we will now discuss the implications of the lack of magnetic field dependence of IR spectra for underdoped YBCO documented in Figs. 1 and 2. One possible interpretation of the data is that the magnetic resonance is irrelevant to QP dynamics. Within this view, self-energy effects in the data can be assigned to excitations inherently insensitive to the magnetic field, such as phonons or the spin fluctuations continuum.36 However, single-phonon processes have a well-defined high-energy cutoff in cuprates that does not exceed 800 cm\(^{-1}\) for YBCO. For this reason, phonons alone cannot account for a high-frequency background in the \(\alpha^2 F(\omega)\) spectra in Fig. 2. On the contrary, magnetic excitations extend to significantly higher frequencies and, therefore, can naturally account for the form of \(1/\tau(\omega)\) spectra in the mid-IR energy range. Thus, our results are consistent with the viewpoint that distinct phonon modes in concert with the broad spin fluctuations continuum are jointly responsible for strong coupling effects in cuprates.

An intriguing interpretation of the magnetic resonance seen in the INS experiments is offered by the \(SO(5)\) theory also providing a unified view on superconductivity and antiferromagnetism in cuprates.37–39 This interpretation is in accord with our data as we will elaborate below. The \(SO(5)\) theory predicts a \(\pi\) resonance in the particle-particle channel that is present both above and below \(T_c\). Coupling of the \(\pi\) resonance to neutrons is facilitated by the formation of the pair condensate in a \(d\)-wave superconductor. This latter attribute of the \(\pi\) mode is important. First, it allows one to understand the quasiparticles’ self-energy probed in the IR data despite apparent depression of the mode seen in the INS experiment. Thus, the \(SO(5)\) interpretation of the INS peak allows one to reconcile dissimilarities in the magnetic field effects in IR and neutron measurements. The results reported here call for an examination of the self-energy effects seen in cuprates by other spectroscopic methods in the magnetic field. While it may be impossible to carry out, such experiments in the case of photoemission studies, tunneling measurements appear to be well suited for this task. It is also worthwhile to reevaluate the role of interband transitions and other excitations in providing a direct contribution to the optical conductivity in the mid-IR region. The so-called multicomponent analysis of the optical data offers a complementary interpretation of some of the effects discussed here within the self-energy formalism.40,41

We acknowledge G. Blumberg, K. S. Burch, J. P. Carbotte, A. Chubukov, E. Demler, S. Kivelson, T. Timusk, J. M. Tranquada, and S. C. Zhang for helpful discussions. This research was supported by a U.S. Department of Energy grant and NSF.

5 The Eliashberg spectral function \(\alpha^2 F(\omega)\) accounts for an effective (and generally energy dependent) electron-phonon coupling \(\alpha^2\); \(F(\omega)\) is the phonon density of states.
One distinction of the spectral function obtained using the inversion theory from that generated using a simplified procedure $W(\omega) = \frac{1}{2}(\omega^2 I + \tau I)$ is worth of attention. The latter approach always yields regions of negative spectral function which is unphysical. On the contrary, the inversion theory offers a self-consistent treatment of both energy gap and strong coupling effects in the optical data and yields the physically meaningful spectral function throughout the entire frequency range. For details, see Ref. 33.

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