Dynamics of an Excited Electron in a Cuprate Antiferromagnet

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We find that the linewidth and energy of an electronic excited state in a cuprate semiconductor increase linearly with increasing temperature. We model this change as due to interactions of the bound electrons with bosons in the antiferromagnetic lattice. The coupling constant between the relaxation rate and the thermal energy is of order 0.5, similar to that for free carriers in superconducting cuprates.

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The frequency-dependent conductivity of high-temperature superconductors in the metallic state has been recently modeled as a sum of two components [1-5]: first, the mobile carriers and, second, a direct absorption. The mobile carriers are scattered by static and dynamic disorder arising from fluctuations in the electronic, lattice, and spin systems. This same dynamic disorder can lead to direct absorption (if its symmetry gives rise to finite matrix elements). Some analyses have modeled the conductivity without including the direct absorption [6-9], and some have argued that it is a large contribution [1-5]. None has determined its temperature dependence, although there are hints that some is present [3].

We have investigated the direct absorption by lowering the charge density to eliminate the mobile carriers. We find that one component of the absorption has a relaxation rate that increases linearly with temperature. This result is important because both its magnitude and functional form are similar to the behavior in the metallic state of the mobile carriers of similar materials as seen [1-8] in the linear dc resistivity and linear scattering rate. Our observations do not, by themselves, settle the question of the origin of the linear relaxation in either the metal or the insulator. We are aware that the behavior in the metal and insulator will be different. However, we argue that the similarity is suggestive, and that the spectral density of fluctuations in the insulating state may be helpful in understanding the high-temperature superconductors.

The optical reflectivity of single crystals of Nd_2CuO_{4-y} with y of order 0 and 0.03 has been measured. Unpolarized light has been used at an angle of incidence nearly normal (12°) to the Cu-O planes of the crystals, and the frequency has been varied over a range from about 3 to 3000 meV using a Michelson interferometric spectrometer. The temperature of the sample and a calibrated Au reference mirror has been varied from 10 to 300 K.

In the doped sample ($y \sim 0.03$) new absorption appears in the energy region below the semiconducting band gap due to the O vacancies added to the Nd_2CuO_4 system. We attribute this absorption to excitations of the electrons (bound to the vacancies) from their ground state to excited states and the continuum. One of these lower-energy transitions has a strong temperature dependence, while the other features in the spectrum do not. This feature is also particularly interesting because a resonant absorption at about the same energy (100 to 200 meV) persists into the metallic state of similar materials [1-5].

Figure 1 shows one set of the raw-data curves. The reflectivity R is plotted on an expanded scale as a function of energy E (or frequency ω) for a series of temperatures, T. We shall concentrate on the change with T of this peak. We have reproduced this behavior in four samples. While the measurement of R establishes that the peak is T dependent, we have extracted the real part of the conductivity to fit the spectrum more easily. To do this, R was measured over a wide frequency range, extrapolated at frequencies below 3 meV to our estimate of the dc conductivity [$\lesssim 10^{-4} (\Omega\,\mathrm{cm})^{-1}$], and extrapolated above 3 eV from our high-frequency R with the conventional (ω^{-4}) form. The real parts of the conductivity σ and dielectric constant ε were then calculated using the Kramers-Krönig relation, and representative curves are shown in Fig. 2.

The conductivity spectrum was fitted with a set of charged harmonic oscillators, with index $i = I, J, K$, resonant energy E_i, half width Γ_i, and strength ω_{Pi}^2, with the

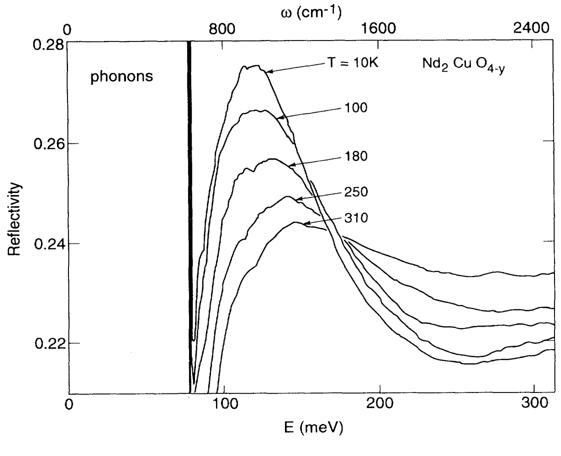

FIG. 1. Optical reflectivity R as a function of photon energy E at several temperatures T, as indicated. The sample is a crystal of Nd_2CuO_{4-y} for a small density of O defects ($y \sim 0.03$).

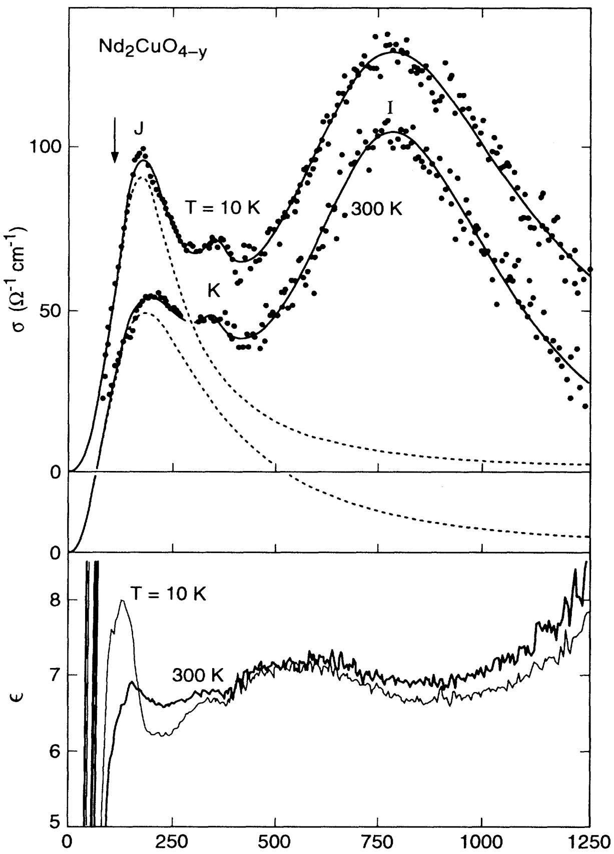

FIG. 2. The real part of the optical conductivity σ as a function of photon energy E at temperatures of 10 and 300 K for the same crystal as in Fig. 1. The dots are the data, the solid lines are fits with a set of Lorentzians, and the dashed lines are the fits for the peak near J only. (The vertical arrow indicates the value of J.) The lower segment shows the real part of the dielectric constant, ε, for the same sample, showing the growth in the peak near J as the crystal is cooled from 300 to 10 K. The changes in ε dominate the changes in R seen in Fig. 1.

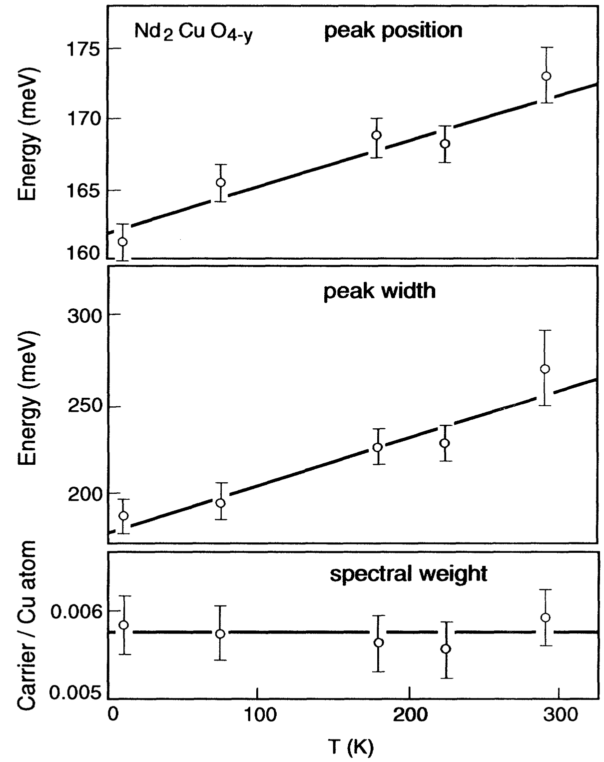

FIG. 3. The parameters extracted from the Lorentzian fits to the peak near J as a function of temperature. The width and peak position increase linearly with T, while the spectral weight is constant within our accuracy.

usual form:

$$\sigma(\omega) = (\Gamma/4\pi)\omega_P^2\omega^2/[\Gamma^2\omega^2+(\omega^2-E^2)^2] , \qquad (1)$$

where ω is the measuring frequency. The two peaks at higher frequency (which we label "I" and "K") only affect the lower-energy peak weakly, and have the parameters $[E_I,E_K,\Gamma_I,\Gamma_K]=[760,335,680,90]$ meV. The strengths, ω_P^2, of these peaks can be expressed as fractional numbers of carriers per 100 Cu atoms: $[f_I,f_K]=[3, 0.06]$. These peaks are only weakly T dependent.

We concentrate on the peak near the antiferromagnetic exchange energy [10], $J=107$ meV, indicated by the arrow in Fig. 2. We have fitted the spectrum at a series of temperatures, and obtained the parameters for the peak near J shown in Fig. 3. The two upper parts of Fig. 3 show that the peak position and the width can be described by the linear functional forms:

$$E=E_0+a_Ek_BT , \qquad (2a)$$

$$\Gamma=\Gamma_0+a_\Gamma k_BT , \qquad (2b)$$

where the constants are the following:

$$E_0=162\pm1\text{ meV} , \quad \Gamma_0=180\pm10\text{ meV} ,$$

$$a_E=0.38\pm0.03 , \quad a_\Gamma=3.0\pm0.2 ,$$

and where $k_B=0.0862$ meV/K. The difference between the ground- and excited-state energies, E_0, is of order $\frac{3}{2}J$. E_0 is considerably below (by about a factor of 7) our estimate of the optical semiconducting energy gap, and it is several times a typical phonon energy.

The oscillator strength for the peak near J is independent of temperature within our accuracy as shown in the lower part of Fig. 3. From this we conclude that the state involved in this transition arises from an electron bound to a vacancy site, rather than a thermally activated carrier which might be modeled as a free polaron [11].

The width at $T=0$ K, Γ_0, is slightly larger than E_0, indicating that the mode is overdamped. A few factors may be relevant in determining the origin of this width. First, more than one transition may be present, but unresolved. Measurements at lower density may show such structure, but our motivation is to try to understand the behavior at higher density, and, from this viewpoint, the data here are of greater interest since they are similar to the resonance seen near J in the metal [1-6]. Second, if we treat the line as arising from one transition, the width at 10 K is equivalent to a lifetime of 2.2×10^{-14} sec. One speculation is that the electronic transition may involve a spin rearrangement (with no net spin flip). The lifetime would then be related to the relaxation of the surrounding spins. For comparison, the full width at half maximum of the two-magnon peak seen in Raman scattering [10] in the same crystal is 185 ± 5 meV. Third, the electron is also coupled to the lattice excitations in perhaps a similar fashion to the Frank-Condon effect [12]. The electronic transition near J may, therefore, be describable with a general, polaronic model [12], including both spin and lattice relaxations.

Two additional fits to the line shape have been considered. The first is to use the Lorentzian form but to assume that Γ_0 is frequency dependent. A linear dependence has been chosen by analogy with the linear T dependence. Over the limited range of frequency that we can analyze with reasonable certainty (between the phonon absorption and the higher-energy electronic impurity bands), this fit is indistinguishable from that with a constant Γ, and has the form

$$\Gamma\cong\omega+3k_BT\,. \tag{3}$$

A second fit attempted was with the line shape used for the finite-frequency excitation within a marginal-Fermi-liquid parametrization of the metallic state [6]. This form has a number of qualitative similarities to our observations, including a linear T dependence and a peak at finite frequency. It also treats both temperature and frequency on an equal footing in the electron dynamics. Although the results both here and in the metallic state both seem to argue for a broad excitation spectrum of this generic type, the particular form involving a $\tanh(\omega/2T)$ is noticeably different from our data at low T. This result does not rule out the "tanh" form in the metal because the excitation spectrum may change between the insulating and metallic states. However, the case discussed here provides a stringent test of the functional form in the insulator with small doping because the finite-frequency peak dominates the spectrum. (In the metal, the free-carrier contribution dominates the spectrum [1-9].)

There is a similarity between the behavior of Γ measured here and the width of the peak at zero frequency in metallic $Ba_2YCu_3O_{6+y}$ for $y\sim1$. The Drude-like absorption due to mobile carriers has a half width Γ_M, whose T dependence can be described [1-9] by

$$\Gamma_M\cong\Gamma_1+2.5k_BT\,. \tag{4}$$

This result in the metal is strikingly similar to the width in the insulator described by Eqs. (2) and (3). In the most heavily doped metals, the constant Γ_1 is of order of the peak position, $\Gamma_1\sim0$, while here, Γ_0 is also of order of the peak position E_0. If the metallic case is modeled [4,7] as scattering of the nearly free carriers from a low-energy boson, then the thermal energy term in the scattering is $2\pi\lambda k_BT$, where λ is the electron-boson coupling constant, and Eq. (4) implies $\lambda\sim0.4$. By analogy, we expect that if a similar analysis can be applied to the metallic and bound-carrier cases, the electron-boson coupling constant in the insulator for our observed parameters [Eq. (2)] will be

$$\lambda\equiv a_\Gamma/2\pi\simeq0.5\,. \tag{5}$$

The broadening of the peak with T is qualitatively similar to changes seen in nonmagnetic and magnetic-doped semiconductors [12]. In both cases, the effects have been discussed in terms of polarons. A polaronic type of model can describe a linear T dependence of the linewidth, and such a model including both spin and lattice excitations may prove useful in understanding the cuprates [11,12]. The small shift in E_J with T and the constancy of the spectral weight provide added constraints on such theories.

We conclude that the electronic transition near J involves a bound charge coupled to the nearby spin and lattice excitations. The relaxation of this coupled charge has striking similarities to the scattering of free carriers in the high-T_c superconductors.

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