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Optical excitations of a few charges in cuprates

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In studies of the optical spectrum of lightly doped semiconducting crystals containing CuO_2 planes, we have observed absorption maxima that occur at the same energies as excitations in the superconducting phase of these materials. One structure occurs just above the phonon energies and near the antiferromagnetic exchange energy.

We have used optical reflectivity to study the properties of a few charges in CuO_2 planes. The charges are introduced into crystals by adding or removing relatively small numbers of O atoms at concentrations well below those that allow the appearance of superconductivity in these same materials.[1,2] The motivation for studying this range of charge density is that the interactions among charges are relatively weak and, therefore, perhaps easier to understand.[3] We also wish to provide data that might test theories of the internal structure of a charge in an antiferromagnet[4] and the binding of the charge to an impurity.[5,6]

We have produced crystals of several materials containing CuO_2 planes for which the added charges remain bound, so that the materials remain semiconducting. We find that the (presumably large number of) excited states of these charges are broadened into a few resolved bands. We focus here on two of these absorption peaks. The one at higher energy is similar to the impurity band in a nonmagnetic, elemental semiconductor[7,8] and is near one estimate of the bound state to continuum transition energy of the impurity. The second band peaks above the phonon energies and near the measured[9] antiferromagnetic exchange energy J. We speculate that this excited state involves electrons coupled to the spin and lattice excitations of the crystal.

Our clearest example of the structure in the optical spectrum at low density is shown in Fig. 1. The doped sample is a single crystal of Nd_2CuO_{4-y} grown from a Cu_2O flux in a closed Pt crucible (solid curve). We found that, using the same procedure with a CuO flux, we were able to grow an "undoped" crystal with nearly stoichiometric O concentration (dashed curve). The mixture was melted at 1430 °C for 2 h, then cooled at 3 °C/h to 1030 °C, and finally cooled with the furnace off. The crystal was in the form of a flat plate of dimensions $3\times3\times0.2$ mm^3 with the large surface parallel to the CuO_2 planes in the T' crystal structure. The large surface was optically flat as grown and allowed us to carry out specular reflectivity measurements with near-normal incidence. We extended our measurements over a frequency range from 0.004 to near 3 eV using a Michelson interferometer, with a set of appropriate of sources, detectors, windows, and beam splitters. We cooled the crystals to temperatures as low as 10 K with a helium-flow cryostat. Absolute reflectivities were obtained by moving a calibrated Au mirror into the beam between repeated measurements of the sample. We combined our measurements of the reflectivity amplitude, shown in Fig. 1, with conventional extrapolations outside our frequency

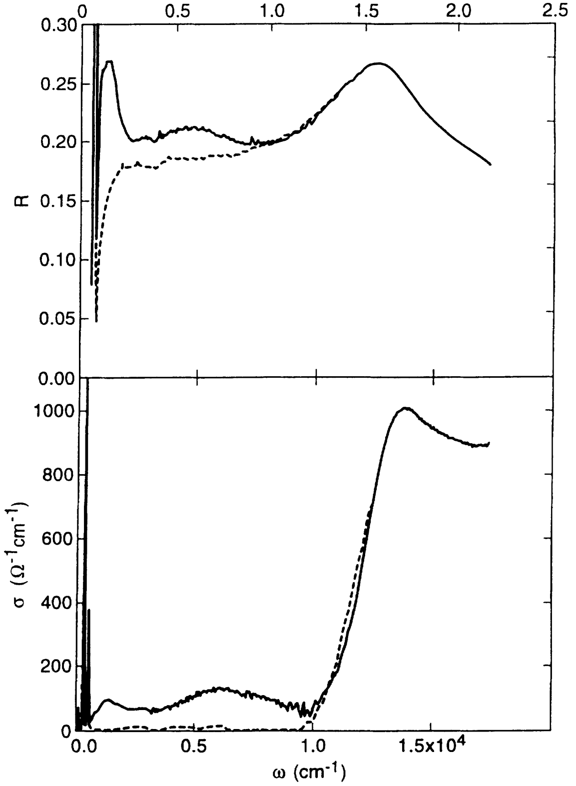


FIG. 1. Optical spectra of two insulating crystals of Nd_2CuO_{4-y} with incident electric field in the a-b plane, at a temperature of 10 K. The upper part shows the measured reflectivity R as a function of frequency ω, and the lower part shows the conductivity σ obtained from a Kramers-Kronig transformation of R. The dashed curves are data for a crystal with $y=0.002\pm0.002$, and the solid curves are for $y=0.017\pm0.002$. These values of y are based on the measured spectral weight with important assumptions that are discussed in the text.

range and carried out a Kramers-Kronig transformation to obtain the imaginary part of the reflectivity. From the complex reflectivity, we calculated the real part of the conductivity σ and the real part of the dielectric constant ϵ. These two quantities are plotted in Figs. 1 and 2.

The curve for σ in Fig. 2 shows two peaks labeled E_I and E_J and a third weak peak labeled E_K. The changes in reflectivity associated with these features reproduced in three separate experiments on the same crystal. The reflectivity (Fig. 1) shows structure very similar to ϵ (Fig. 2) since the absorption from σ is small in this region of the spectrum. The plot of σ in Fig. 1 shows the phonons below about 75 meV and the charge-transfer absorption band rising for energies above 1.3 eV. The systematic uncertainties in these measurements are greater toward higher energy, because this range is somewhat sensitive to our extrapolation outside the measured frequency range. At low frequency we constrained our extrapolation to be consistent with the dc conductivity of $\sim 10^{-4}$ $\Omega^{-1}\,\text{cm}^{-1}$. We estimate the absolute uncertainty in σ to be less than a factor of 2 at 2 eV, decreasing to $\sim 20\%$ near 0.3 eV. The peak positions in σ and ϵ are relatively unaffected by the absolute uncertainty.

In order to describe empirically the absorption bands produced by doping, we have fit each peak in σ to the usual Lorentzian form with three constants adjusted. The fit is shown as the heavy, smooth curve in Fig. 2. The values of the peak energies E_J and E_I are listed in Table I. The linewidths are $\Gamma_J = 184 \pm 10$ meV and $\Gamma_I = 680 \pm 60$ meV. The small peak labeled K has the parameters $E_K = 335 \pm 10$ meV and $\Gamma_K = 87 \pm 25$ meV, where here and below we quote the random errors. The third fitting parameter is related to the spectral weight, which we can express as a fractional number of electrons per Cu atom, f, using the sum rule

$$f = (2m^* V_{\text{cell}})/(\pi e^2 N_{\text{Cu}}) \int_0^\infty \sigma(\omega)\, d\omega \; . \qquad (1)$$

For Nd_2CuO_4 we use the unit-cell volume[10] $V_{\text{cell}} = 187$ Å^3 and the number of Cu atoms per unit cell, $N_{\text{Cu}} = 2$. For an initial estimate, we use an effective mass m^* equal to the free-electron mass. (The numbers can be scaled if a different m^* is considered.) With these parameters we have effective electron densities for the three peaks:

$$[f_I, f_J, f_K] = [(2.9 \pm 4), (0.6 \pm 0.1), (0.06 \pm 0.03)] \text{ electrons}/(100 \text{ Cu atoms}) \; .$$

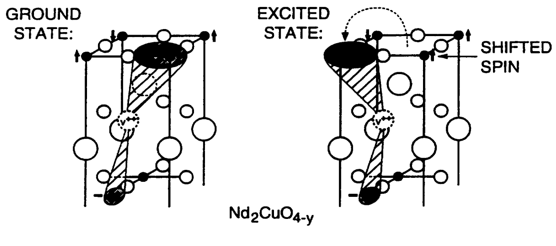
An estimate of the added electron density in this doped sample is given by the sum of the three values of f. To obtain the density of vacancies, we divided this by 2 under the assumption that each vacancy arises from the absence of an O atom and thus contributes two valence electrons. For this sample the estimated vacancy density N_{tot} is thus

$$N_{\text{tot}} = (1.8 \pm 0.3) \text{ vacancies}/(100 \text{ Cu atoms}) \; .$$

Based on this density and a comparison with other measurements[11] of doped Nd_2CuO_4, we conclude that this crystal is well within the antiferromagnetic semiconducting state with a Néel temperature near 250 K. Because of this low density, we consider a picture of the vacancy and

TABLE I. Peak energy E_I of the upper band in Figs. 1–3 is near the ionization energy of a charge bound to an impurity center as estimated by E_C using Eq. (2). The magnitude of E_I indicates a deep impurity at a substantial fraction of the semiconducting energy gap E_G reported in Ref. 15. The peak energy E_J of the lower band in Figs. 1–3 may arise from an excitation as shown schematically in the picture of Nd_2CuO_{4-y}. E_J is near half of the two-magnon Raman scattering peak energy, $E_R/2$, and near the antiferromagnetic exchange energy J, both reported in Refs. 9 and 15. The distance between the impurity center and a nearby, planar Cu atom, r, reported in Refs. 10 and 15 is illustrated in the picture. The local dielectric constant ϵ_L is the value that we measure in undoped crystals in the frequency range near E_J and E_I. The value f of the fractional number of added charges is measured from the spectral weight added by doping (assuming a free-electron mass).

	E_I (meV)	E_C (meV)	E_G (meV)	E_J (meV)	$E_R/2$ (meV)	J (meV)	r (Å)	ϵ_L	f (per 100 Cu)
Nd_2CuO_{4-y}	760±10	560	1450±50	162±5	144±6	107±6	3.6	6.3±0.3	3.5±0.5(0.5±0.5)
$Ba_2YCu_3O_{6+y}$	620±50	740	1600±50	160±30	132±6	98±6	4.7	4±0.5	7±1
La_2CuO_{4+y}	600±20	820	1800±50	130±20	170±6	126±6	3.4	5±0.3	2±(0.5±0.5)
$La_{2-x}Sr_xCuO_{4+y}$	530±50	850	1800±50	160±30	170±6	126±6	3.3	5±0.3	6±2



its charge distribution as a relatively isolated entity.

One question we can try to address about the features in Figs. 1 and 2 is whether we can see similar structures in other cuprates. Figure 3 indeed shows similar features in a comparison of measurements of single crystals of $Ba_2YCu_3O_{6+y}$, Nd_2CuO_{4-y}, and La_2CuO_{4+y}. In addition, similar midinfrared absorption has been seen[12] in $La_{2-x}Sr_xCuO_{4-y}$ crystals.

The first aspect of the data that we would like to emphasize is the presence of structure in the conductivity spectra of all the compounds shown in Fig. 3, although they differ in detail. In Nd_2CuO_{4-y} and La_2CuO_{4+y}, two peaks are resolved, and in $Ba_2YCu_3O_{6+y}$ two similar peaks describe the principal spectral weight in the data. (Sketches of some peaks are shown as the dashed lines in Fig. 3).

The second important experimental result illustrated in Fig. 2 is that when we changed the O concentration toward O_4 for both La_2CuO_{4+y} and Nd_2CuO_{4-y} by annealing and varying the growth conditions, the conductivity was drastically reduced in this mid infrared frequency range. This result is shown by the lower curves for both the Nd and La compounds and indicates that the absorption peaks definitely arise from the changes in O doping away from stoichiometric amounts.

A third question is how these structures evolve at higher doping density and if similar absorption occurs with other than O impurities in cuprates. Spectra[13] for a series of crystals of $Ba_2YCu_3O_{6+y}$ (with O concentration indicated by the values of T_c) have been published. We interpret the results as a growth of the band near E_I up to the metal-insulator transition, above which it saturates. For the barely metallic crystal, with $T_c = 30$ K, the peak near E_I dominates. The peak near E_J continues to grow at higher density and is still resolved for the crystal with $T_c = 80$ K. For crystals with $T_c \sim 90$ K, absorption is still present at E_I and E_J, but the Drude-like peak at $\omega = 0$ dominates. Spectra have also been reported[12] for $La_{2-x}Sr_xCuO_4$ with no intentional O doping. In the barely metallic sample with Sr concentration $x = 0.02$, the energy of the dominant peak is near the bands labeled E_I in Fig. 3. While this E_I peak saturates at higher x, the absorption near E_J continues to grow, and in the sample with $x = 0.10$, a peak is resolved near E_J. We argue that the band near the energy E_J is present at low x in $La_{2-x}Sr_xCuO_4$ and grows with doping up to $x = 0.15$.

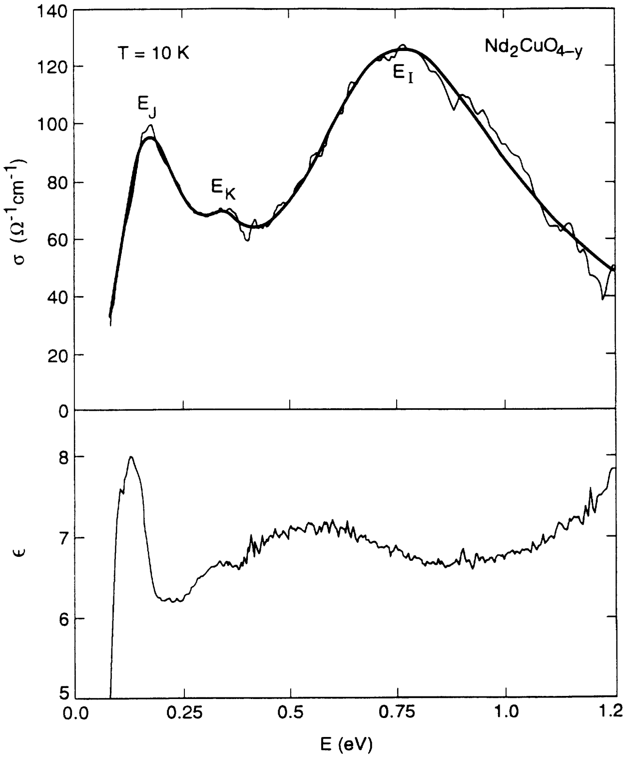


FIG. 2. Optical spectra of the electronic bands that arise with doping for the insulating crystal of Nd_2CuO_{4-y} with $y \sim 0.018$. The upper part shows the measured conductivity in the a-b plane, σ, as the light line showing noise, and the fitted curve as the heavy line. The fit is a sum of three Lorentzians centered at energies E_J, E_K, and E_I with parameters listed in Table I and the text. The lower part of the figure plots the dielectric constant ϵ for the same crystal showing the same electronic bands and also the typical value of ϵ in this part of the spectrum, $\epsilon_L \sim 6-7$, as listed in Table I.

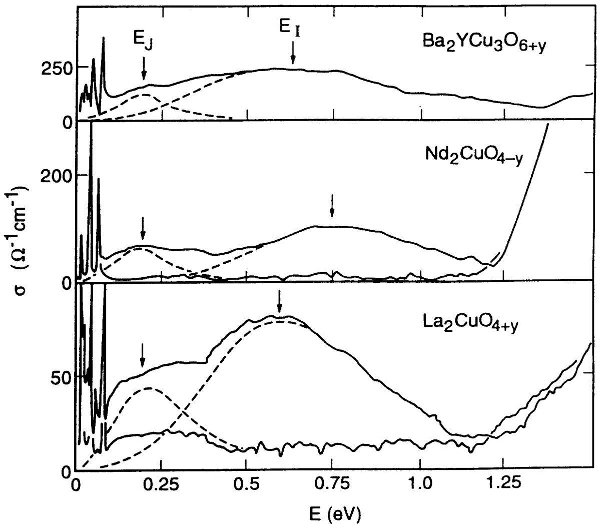


FIG. 3. Comparison of the optical conductivity σ for five crystals containing Cu-O planes in their structure. The sample temperatures were 300 K for $Ba_2YCu_3O_{6+y}$ and 10 K for the others. We argue that the qualitative similarity of the bands E_J and E_I indicates a similar origin in spite of the differences in crystal structure. The lower four curves show that the bands E_J and E_I grow with increasing doping. The lower curves for both Nd_2CuO_{4-y} and La_2CuO_{4+y} are for crystals grown to have no doping, and both have spectral weights corresponding to $y = 0.002 \pm 0.002$. The upper curves are for crystals that were intentionally doped and have larger spectral weights in the bands E_J and E_I as can be seen in the figure and as listed in Table I. The dashed curves are sketched to indicate the band positions.

Combining the results for the three sets of cuprate crystals shown in Figs. 1–3 with those for $La_{2-x}Sr_xCuO_4$ we list the energies of the peaks in Table I. Studies[8] of very lightly doped Si:P have determined the ionization energy E_I' of the electron bound to the P^+ donor to be 45.5 meV. With increasing doping a band is formed from the broadened states near E_I'. The spectrum of this band is similar in appearance to the band we have labeled E_I. Based on this similarity, we suggest that the optical ionization energy is somewhat ($\sim 14\%$) above the peak labeled E_I in the figures. However, unlike Si:P, the thermal ionization energy[3] in La_2CuO_{4+y} (~ 30 meV) is much smaller than this estimate of the optical ionization energy. The thermal energy has been measured[3] from the variation of the dc conductivity and Hall coefficient in the range $50 < T < 300$ K. The difference between the thermal and optical characteristic energies is important and argues for further study.

These energies for the peaks labeled E_I do not occur at half the energy of the optical gap formed by the charge-transfer excitation band in the Cu-O planes. Previously, we have defined the optical gap E_g as the midpoint of the conductivity rise above the absorption edge and obtained the values[14] listed in Table I. Our peaks at E_I are as low as $\frac{1}{3}$ of E_G and do not appear to scale in any simple way with the values of E_g.

A second energy that can be compared with E_I is a crude estimate of the Coulomb contribution to the binding energy of the impurity ground state. This energy is given by the local dielectric constant ϵ_L and the distance r between the position of the core charge (the vacancy in Nd_2CuO_{4-y}) and an assumed position of the bound charge (the electron in Nd_2CuO_{4-y}). This Coulomb energy E_C is given by

$$E_C = e^2/\epsilon_L r \ . \tag{2}$$

We expect that the charge will be distributed over various sites near the impurity, but some will accumulate nearby in simple positions in the Cu-O planes. One such position is the site of a nearby Cu atom, as illustrated in the inset to Table I. This schematic picture of an enlarged, missing O atom is based on previous analysis of doped semiconductors[7,8] and on calculations[6] for $La_{2-x}Sr_xCuO_4$. We expect that the charge will not reside symmetrically between the two Cu atoms because it will have lower energy when it is aligned antiparallel to the nearest Cu spin. This breaking of the ground-state symmetry will be enhanced by a Jahn-Teller distortion.[7] We take r to be the distance[10,15] from the impurity core to the nearest planar Cu site as listed in Table I. For the local dielectric constant ϵ_L, we use our measurements of ϵ near E_I (e.g., Fig. 2) since this frequency range involves short-range vibrations of the charges. Equation (2) then gives values of E_C as listed in the table.

These values of E_C do not provide an adequate explanation of the peak positions E_I, but they are of the same order. An estimate of characteristic energies for $La_{2-x}Sr_xCuO_4$ has been made[6] by considering a positive charge in a Cu-O plane, described by the t-J model and bound to a Sr impurity. Using a value of ϵ_L similar to ours, this calculation finds that the charge is distributed primarily around the square planar plaquettes of Cu atoms, and obtains an ionization energy near 0.7 eV. We argue that this estimate, the analogy with Si:P, and our estimates of E_C support the identification of the peak labeled E_I as arising from an impurity band near the optical ionization energy of the isolated impurity.

Proceeding further with this set of assumptions, the absorption labeled E_J may arise from a band near the energy of a strong transition between the ground state and an excited state of the same impurity. We expect that the low-lying states with strong matrix elements will be those with a dipole moment and with excitation of the valence charge to nearby planar sites. The sketch in the inset to Table I, made within an Ising representation, shows a valence electron initially in a singlet state and the surrounding Cu spins antiferromagnetically aligned. The electron can be excited by the incident photon to the adjacent Cu atom with negligible change in E_C (in a rigid lattice). The lowest-energy final state of the electron will be a singlet, and since the photon cannot change the net spin, the Cu left behind will have its spin aligned opposite to the surrounding antiferromagnetic order. As indicated in the picture accompanying the table, the electron will hop one way and will, in effect, induce the spin to hop the other way. The energy of the transition is then due primarily to the spin on the wrong sublattice and is of the order of the Cu-Cu exchange energy J. For the three systems we consider here, the values of J from two-magnon Raman scattering[9,14] are listed in Table I and are of the same order as E_J. Just as the charge motion will disturb the spin order, it will also disturb the lattice.[16] In either case one can describe the peak labeled E_J as a polaronic excitation of the bound charge.

For the four doped cuprates discussed above, the energies of the strong, low-frequency peaks near J are given by the values E_J listed in Table I. (The measurements were carried out at room temperature for $Ba_2YCu_3O_{6+y}$ and $La_{2-x}Sr_xCuO_4$ and at 10 K for the others.) The uncertainty in E_J is small for Nd_2CuO_{4-y} because the peak in its spectrum is clearly resolved. In general, there will be a number of overlapping bands formed from various excited states. The peak labeled K in Nd_2CuO_{4-y} is probably one example, but it does not strongly affect the measurement of E_J. In other cases we find that nearby transitions significantly reduce our ability to parametrize a single peak near J with as much certainty.

The comparison between the values of E_J and J indicates that the simple argument given above does not provide an adequate explanation, but we argue that the correspondence is suggestive and that J might play a role in determining the transition energies in a realistic theory. In addition to this coupling to the antiferromagnetism, the impurity state will also couple to the lattice (see, e.g., Refs. 16 and 17), affecting the transition energies. These interactions will also affect the line shape, which we have assumed to be simply a Lorentzian with a constant width parameter Γ. For instance, the interaction of the phonons with the low-energy tail of the band near J could lead[18] to an anomaly (seen as a knee in the reflectivity) at an energy that is independent of doping density.[13] Another contribution to the broadening, studied extensively in elemental doped semiconductors,[7,8] is near-neighbor impurity interactions.

We conclude by reconsidering the results at higher density[12,13] in view of the band near J discussed above. One way of modeling the spectra[19] for crystals with higher doping is suggested by the data for $Ba_2YCu_3O_{6+y}$ with $T_c = 80$ K and the $La_{2-x}Sr_xCuO_4$ with Sr concentration $x = 0.10$. Here two components are resolved:[12,13] a large (Drude-like) peak at zero frequency and a second peak near J. A model[20] in which a band near J produces scattering, absorption and pairing interactions provides a good fit to the optical spectrum for the crystal with $T_c = 80$ K.

We argue that the changes in the spectrum arise from bands that grow with doping. Independent of models, our results indicate that these bands result from the bound charges. One of these bands, which is near J, appears to continue to grow in the metallic state and may have similarities in its origin in the metallic and insulating states.

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