Phonon Density of States and Superconductivity in $Nd_{1.85}Ce_{0.15}CuO_4$

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Inelastic-neutron-scattering techniques have been used to measure the generalized phonon density of states in superconducting $Nd_{1.85}Ce_{0.15}CuO_4$. There is reasonable agreement at low energies between our measurements and recent point-contact-tunneling density-of-states measurements reported by Huang *et al.*, which suggests that phonons play a major role in pairing in the electron-superconductor systems. The correspondence at higher energies, however, is not particularly good, but probably reflects the difficulty in obtaining quantitative tunneling data.

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Since the discovery of the high-T_c oxide superconductors, the question of the origin of the electron pairing has remained elusive. In conventional superconductors, there are three direct experimental tests of the electron-phonon mechanism: the isotope effect, phonon structure in the tunneling density of states, and phonon-lifetime anomalies for phonon energies below the superconducting gap energy.[1] For the oxide systems,[2] a substantial isotope effect has been observed for the "moderate"-T_c ($\sim$30 K) $(La\text{-}Sr)_2CuO_4$ and $(Ba\text{-}K)BiO_3$ systems,[3] while no isotope studies have been reported for the electron superconductors such as $(Nd\text{-}Ce)_2CuO_4$. In the Cu-containing high-T_c ($\gtrsim$77 K) systems, on the other hand, isotope-effect studies show no substantial shift in T_c.[3] Very recently, Huang *et al.*[4] have reported point-contact-tunneling data for both the $(Ba\text{-}K)BiO_3$ and $(Nd\text{-}Ce)_2CuO_4$ moderate-T_c systems. The $\alpha^2F(\omega)$ obtained from these data show structure which is reminiscent of the phonon structure observed in phonon-mediated superconductors. In the $(Ba\text{-}K)BiO_3$ system, in particular, the structure was found to be in reasonable agreement with the measured phonon density of states for this material, especially at the lower energies where the tunneling data are most reliable. Their results thus provide direct evidence for phonon-mediated pairing in this system. Here we report measurements of the generalized phonon density of states for the $(Nd\text{-}Ce)_2CuO_4$ system, which can be directly compared with the $\alpha^2F(\omega)$ obtained by Huang *et al.*[4] for this system. We find that there is a similar correspondence at lower energies between the two techniques, which suggests that phonons are playing a major role in the T' electron-superconductor materials as well. At higher energies the correspondence between the two techniques is presently not good, but we believe that this likely reflects the severe difficulty in obtaining tunneling data in these oxide systems with the precision necessary to extract accurate $\alpha^2F(\omega)$ information.[5]

The neutron data were obtained on the BT-4 and BT-2 triple-axis spectrometers at the National Institute of Standards and Technology research reactor. At lower energies, the usual triple-axis technique was employed, with either a Cu(220) or pyrolytic graphite PG(002) monochromator. At higher energies, these two monochromators were used with either a cold (77 K) Be filter in front of the detector, or a combination of a graphite (C) and a Be filter. This is an ideal technique to obtain density-of-states information,[6] and the experimental conditions and approximate energy resolutions are given in Table I. Data were obtained on a 43-g polycrystalline sample of superconducting $Nd_{1.85}Ce_{0.15}CuO_4$. A limited set of data was also obtained on an 11-g Nd_2CuO_4 sample for comparison. All data were taken at 4.2 K. The details of the sample preparation and properties are given elsewhere.[7]

The measured spectrum up to an energy of $\sim$100 meV is shown in Fig. 1. The scattering is dominated by two strong peaks at 20.7 and 26.4 meV. This is magnetic scattering which originates from single-ion crystal-field excitations of the Nd ions, and the energies we obtain are in good agreement with the data already reported by other groups.[8] There is an additional crystal-field excitation which we observe at 94.2 meV. These magnetic peaks are of no direct interest here, but they do provide a convenient method for comparing data obtained under varying instrumental conditions as listed in Table I. Unfortunately, they also masked the phonon

TABLE I. Instrumental configurations employed and the corresponding energy resolution.

	Monochromator	Collimation (min FWHM)	Analyzer	Energy resolution (meV)
1	Cu(220)	60-40-20-40	PG(002) [a]	1.7 at $\Delta E=10$
2	PG(002)	40-40-40-40	PG(002) [a]	3.3 at $\Delta E=10$
3	Cu(002)	40-40	Be	6.3 at $\Delta E=90$
4	PG(002)	20-20	Be+C	1.7 at $\Delta E=20$
5	Cu(002)	40-40	Be+C	4.1 at $\Delta E=90$

[a]$E_f=28$ meV.

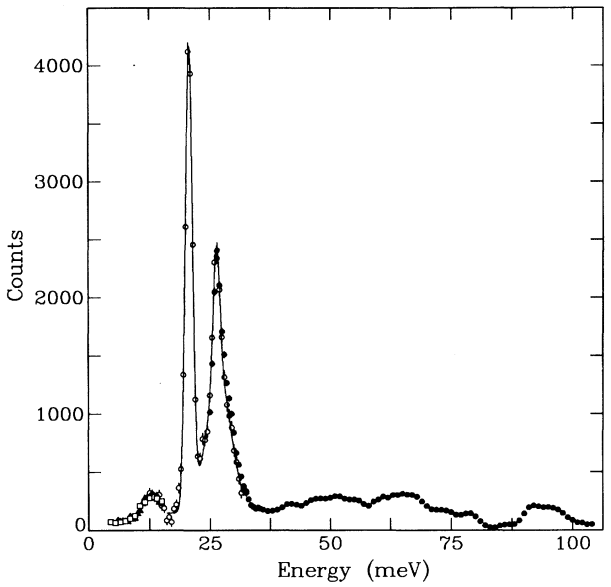

FIG. 1. Density-of-states scattering observed in $Nd_{1.85}Ce_{0.15}CuO_4$. The two strong peaks are (magnetic) crystal-field excitations of the Nd. The symbols denote the various instrumental configurations as given in Table I: ▲, 1; □, 2; ○, 4; ●, 5 and 3.

scattering at these energies, and thus prohibited us from determining the phonon density of states $F(\omega)$ in these regions.

At low temperatures the scattering cross section for phonons is given in the incoherent approximation by

$$\frac{d^2\sigma}{d\Omega\, d\omega} \propto \sum_{i=1} \frac{\sigma_i}{m_i}\left[\frac{\langle(\mathbf{Q}\cdot\boldsymbol{\varepsilon}_i)^2 e^{-2W_i}\rangle}{\omega}\right] F(\omega)\,,$$

where i represents the ith atom, σ_i is the total nuclear bound scattering cross section, m_i is the mass, $\mathbf{Q}$ is the wave-vector transfer, $\boldsymbol{\varepsilon}_i$ is the polarization vector, W_i is the Debye-Waller factor, and the sum is over all atoms in the unit cell.[6] The data have been corrected by the factor in square brackets to obtain $\sum(\sigma_i/m_i)F(\omega)$, the generalized phonon density of states (GDOS). Note that this is weighted by the factor σ_i/m_i which represents how strongly each atom scatters. For Nd, Ce, Cu, and O this factor is 0.11, 0.02, 0.12, and 0.26 b/amu, which means that we will be most sensitive to vibrations of the oxygen.[9]

The phonon scattering of interest is shown in Fig. 2 (top), plotted on an expanded scale. There are obvious peaks at 13, 51, and 65 meV, and additional smaller peaks or shoulders at 42 and 79 meV. The peak at 13 meV originates from the acoustic-phonon modes bending over as they approach the zone boundary, while the peaks at 51 and 65 meV are likely due to oxygen bending and stretching modes. The peak at ~95 meV we believe may be partly phonon in origin, although there is a crystal-field excitation at this energy as well.[8] The

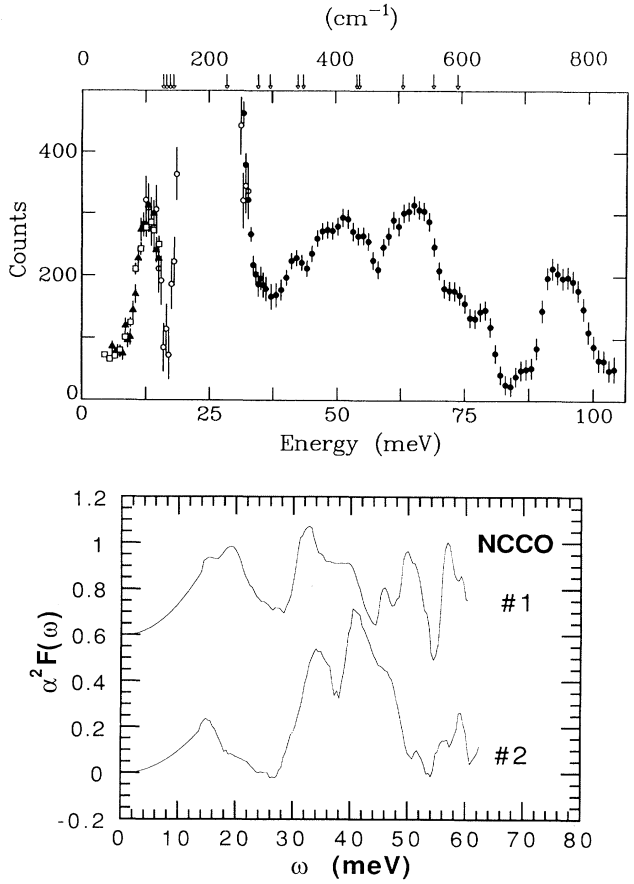

FIG. 2. The generalized phonon density of states (top), compared with the tunneling measurements of $\alpha^2F(\omega)$ from Huang *et al.* (Ref. 4) (bottom). The arrows at the top indicate the Raman and ir active (zone center) phonon modes which have been observed (Refs. 12–14). There is reasonable agreement between the tunneling and neutron data at lower energies.

overall structure is quite similar to the GDOS observed in the related $(La\text{-}Sr)_2CuO_4$ system,[10] though of course the quantitative details in terms of the relative intensities and positions of the peaks are different. We also note that there are a large number of phonon branches (21) in these systems, and in analogy with related systems such as La_2NiO_4 where the dispersion relations have been measured in detail,[11] we anticipate substantial dispersion in all the branches. Hence it is not surprising to find significant GDOS at most energies. The substantial dispersion also makes it difficult to make a direct comparison of the GDOS [or $\alpha^2F(\omega)$ from tunneling] with the lattice-dynamical information obtained from Raman scattering and ir absorption experiments[12–14] since these techniques generally measure the phonon modes only at the zone center. The Raman and ir active modes that have been observed are indicated by arrows at the top of Fig. 2, where we have also included an ener-

gy scale in wave numbers (1 meV = 8.066 cm^{-1}). It can be seen that there is little direct correspondence between the GDOS and the zone-center modes in this case. Finally, we mention that our data have been extended in energy with instrumental condition 3 (Table I) up to 126 meV, and we have found one additional weak peak at ~113 meV. We do not anticipate a phonon peak from the lattice at this energy, and hence tentatively assign this scattering to a small amount of hydrogen in the sample. We note that for hydrogen $\sigma/m = 81$ b/amu, and thus neutrons are especially sensitive to any hydrogen. Our experience has been that there are, in fact, very few polycrystalline oxide samples which do not show some hydrogen contamination, but in the present case we estimate that the hydrogen in the sample is less than 1 at.%.

The bottom portion of Fig. 2 shows the $\alpha^2F(\omega)$ recently obtained by Huang *et al.*[4] from point-contact-tunneling measurements on $Nd_{1.85}Ce_{0.15}CuO_4$ single crystals. The peak at ~14 meV agrees rather well with the observed peak in the GDOS, while the minimum at ~25 meV is masked in the neutron data by the crystal-field excitations. Both tunneling samples show peaks at ~32 and ~40 meV although there are obviously some differences between the samples. In the neutron data we observe structure ~30 meV as discussed below, and a weak peak at ~42 meV as well. At higher energies it is clear from the figure that the $\alpha^2F(\omega)$ data are not yet consistent enough from sample to sample to make a quantitative or semiquantitative comparison with the GDOS.

One important point to address is whether there is any strong phonon structure in the 20–30-meV range, where the tunneling data are more accurate and show a relative minimum. Figure 3 shows the data obtained on the superconducting sample (solid circles) with instrumental condition 4 (Table I). The width of the magnetic transition at 20.7 meV is resolution limited, but the peak at 26.4 meV is obviously broader, and, in fact, it appears that there are two peaks, one narrow and one broader. If we interpret the broader one as phonon in origin, then this would mean that there is a strong phonon peak at this energy, and, in fact, this peak would be ~3 times larger than any other phonon structure in the spectrum. To pursue the origin of this scattering we obtained data on the pure Nd_2CuO_4 sample (open circles). The crystal-field peaks shift to higher energies, and the peak at ~27 meV is considerably sharper. Thus the extra scattering at ~26–27 meV in the superconducting sample is likely not of phonon origin, but results from the Ce substitution (or oxygen defects), with the resultant disorder broadening this particular crystal-field excitation. Note, on the other hand, that there is a shoulder of scattering in both samples in the region of ~30 meV, which *is* likely due to phonons and corresponds well with the structure observed in the tunneling data. It is important to keep in mind, however, that in the pure system

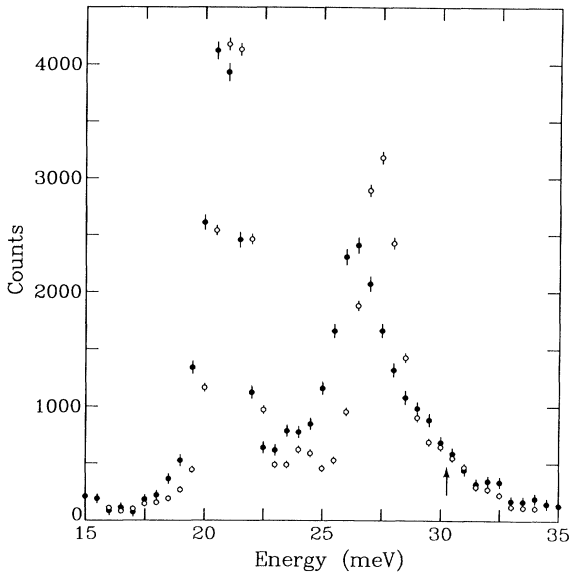

FIG. 3. The scattering observed [configuration 4 (Table I)] in the energy range 20–40 meV for $Nd_{1.85}Ce_{0.15}CuO_4$ (solid circles) and Nd_2CuO_4 (open circles). The shoulder in the scattering in the vicinity of ~30 meV observed in both samples is attributed to phonons.

the Cu spins are antiferromagnetically ordered, and there is substantial coupling between the Nd and Cu sublattices[7] which may also broaden the magnetic scattering in this region. An unambiguous separation of the magnetic and lattice-dynamical scattering in this region would require polarized neutron techniques, but currently the available instrumental intensities are too low to carry out this separation in practice. We can also remark that there is clearly no correspondence between the Nd crystal-field peaks and the tunneling data, and of course none is expected. If there is any substantial coupling of the electrons to the Cu magnetic excitations, we would expect this to be observed at much higher energies (~200 meV),[12,15] and it will certainly be interesting to extend the tunneling data to higher energies.

It is important to keep in mind when comparing the tunneling and neutron data that neither technique directly measures $F(\omega)$; the neutron data are weighted by σ_i/m_i, and the tunneling data by the electron-phonon coupling α^2. Hence for these multicomponent-oxide systems we cannot expect the detailed correspondence that can be obtained for simple systems like lead.[1] In the present case we believe there is reasonable agreement between the $\alpha^2F(\omega)$ and the GDOS, comparable to the agreement found in the (Ba-K)BiO_3 system. The comparison indicates that the electrons and phonons are strongly coupled, and thus suggests that phonons play a major role in pairing in the electron-superconductor systems. However, it does not rule out the possibility that

other excitations are also playing a role in the pairing. Clearly, additional tunneling data, especially at higher energies, would be very desirable.

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