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# Far-infrared measurement of the energy gap of $La_{1.8}Sr_{0.2}CuO_4$

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We report the far-infrared transmission of the high-$T_c$ superconductor $La_{1.8}Sr_{0.2}CuO_4$ at temperatures above and below $T_c$ and for magnetic fields up to 10 T. At 7 K our data are best described by a distribution of superconducting energy gaps with a representative value of about 50 $cm^{-1}$ (i.e., 72 K).

Recently, Bednorz and Müller[1-3] discovered a new family of high-$T_c$ oxide superconductors ($T_c > 30$ K) with composition $La_{2-x}A_xCuO_4 (x \lesssim 0.3, A = Ba,Sr,Ca)$. Since the original work,[1] a number of papers[4-7] have appeared confirming the high $T_c$ and reporting resistivity, low-field dc magnetization (Meissner) and pressure experiments with the onset of the superconducting transition as high as 52 K. To elucidate the mechanism responsible for such high transition temperatures, it is important to measure normal- and superconducting-state parameters such as the electron and phonon density of states, the electron-phonon coupling constant, and the energy gap. In this paper we demonstrate the existence of an energy gap in the superconducting state of a member of the $La_{2-x}A_xCuO_4$ class of high-$T_c$ superconductors.

The sintered samples of $La_{1.8}Sr_{0.2}CuO_4$ were prepared by a high-temperature solid-state reaction from $SrCO_3$, CuO, and $La_2O_3$. These samples are pressed into pellets and then annealed in air as discussed elsewhere.[3] X-ray analysis indicates an essentially single phase of the $K_2NiF_4$-type tetragonal structure. A four-probe resistivity measurement gives a $T_c$ of 35 K (midpoint determination) with a transition width (10–90%) of ~8 K. A dc magnetic susceptibility measurement[8] gives 80% of the full diamagnetic shielding $(-\frac{1}{4}\pi)$ below 20 K, a midpoint $T_c$ of 27 K, a transition width of ~15 K, and a 30% Meissner effect at 4.2 K. Thus, at least 30% of our sample is superconducting but with a rather wide distribution of $T_c$'s, presumably caused by some inhomogeneity in the preparation process.

To prepare the sample for the infrared transmission measurement, we grind a small piece of the pellet into a powder, with roughly one- to ten-micron particle sizes. This powder is mixed with vacuum grease and this composite is pressed between two wedged sapphire windows. We place this sample in a Dewar, where it is studied as a function of temperature and magnetic field. Broadband radiation from a Michelson interferrometer is transmitted through the sample at roughly normal incidence and detected by a bolometer well below the sample. The standard Fourier transform technique is used to obtain the far-infrared transmission spectra.

In Fig. 1, we show the normalized difference in the transmission ($\mathcal{T}$) between the sample in the superconducting ($s$) state ($T = 10$ K) and the normal ($n$) state ($T = 50$ K)

$$\Delta\mathcal{T}(\omega) = [\mathcal{T}_s(\omega) - \mathcal{T}_n(\omega)]/\mathcal{T}_n(\omega) . \quad (1)$$

Below 60 $cm^{-1}$, the $s$ state transmits better than the $n$ state, while above 60 $cm^{-1}$, the $s$ state transmits less, and the difference goes to zero at higher frequency. This demonstrates the existence of a superconducting energy gap in $La_{1.8}Sr_{0.2}CuO_4$.

In Fig. 2, we show the same normalized difference spectra

$$\Delta\mathcal{T}(\omega;T) \equiv \frac{\mathcal{T}(\omega;T) - \mathcal{T}(\omega;50\text{ K})}{\mathcal{T}(\omega;50\text{ K})} \quad (2)$$

for several temperatures $T$. The inset shows the values of $\Delta\mathcal{T}(\omega;T)$ at 86 $cm^{-1}$ as a function of temperature. We see that the difference decreases gradually between 10 and 30 K, above which there is no further change in the transmission. This gradual growth is consistent with the existence of a broad distribution of $T_c$'s. If our sample had a sharp $T_c$, and hence a single energy gap, we would ex-

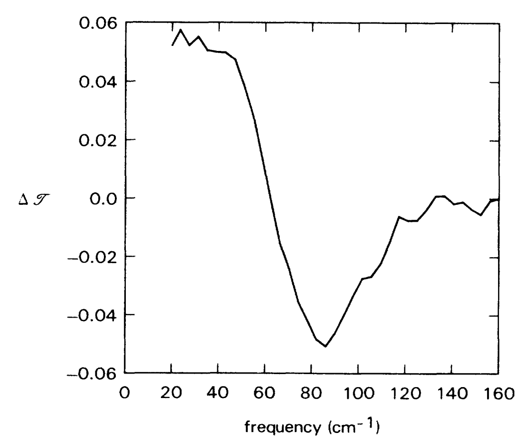


FIG. 1. We show the normalized transmittance difference $\Delta\mathcal{T} = (\mathcal{T}_s - \mathcal{T}_n)/\mathcal{T}_n$ between the superconducting (10 K) and normal (50 K) $La_{1.8}Sr_{0.2}CuO_4$ as a function of frequency. This behavior demonstrates the existence of a superconducting energy gap.

 

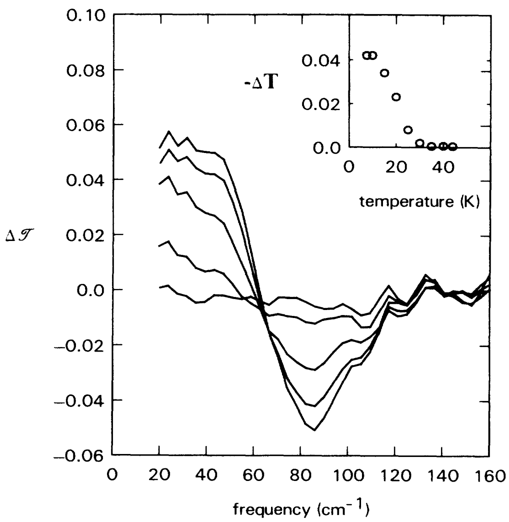


FIG. 2. We show the normalized transmittance difference $\Delta\mathcal{T}(\omega)=[\mathcal{T}(\omega;T)-\mathcal{T}(\omega;50\text{ K})]/\mathcal{T}(\omega;50\text{ K})$ for several temperatures, $T=30$, 25, 20, 15, and 10 K. The inset shows $-\Delta\mathcal{T}(\omega)$ at 86 cm$^{-1}$ as a function of temperature.

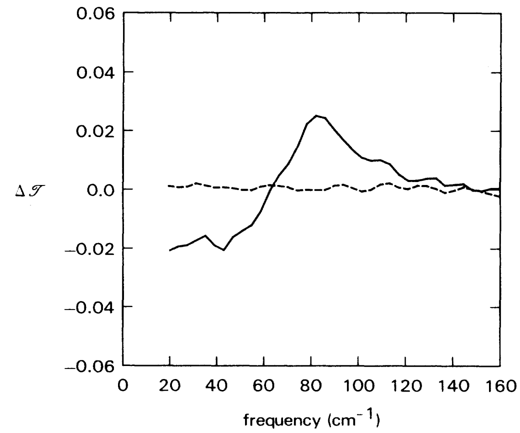


FIG. 3. The effect of magnetic field on the superconducting state is demonstrated. For $T=10$ K we plot the difference $\Delta\mathcal{T}'(\omega)=[\mathcal{T}(\omega;10\text{ T})-\mathcal{T}(\omega;0)]/\mathcal{T}(\omega;0)$. The field alters the transmission (solid curve) in the same way as increasing the temperature. The dashed curve is the normalized difference between two spectra with identical parameters (10 K and 0 T), which indicates the noise level in our measurements.

pect $\Delta\mathcal{T}(\omega;T)$ to increase much faster for $T$ just below $T_c$, in a manner more like the BCS order parameter $\Delta(T)$. The disappearance of $\Delta\mathcal{T}(\omega;T)$ near 30 K is consistent with our susceptibility measurement which shows, at 30 K, a Meissner signal only 5% of its full value. It is important to realize that the far-infrared transmission is probing the bulk of the sample (since we estimate a large penetration depth in these low carrier density materials). The distribution of energy gaps makes it difficult to resolve the expected decrease in $2\Delta$ as $T$ approaches $T_c$, although we find some evidence of a downward energy shift in $\Delta\mathcal{T}(\omega)$ at 25 K.

In Fig. 3, we explore the effect of a magnetic field on the transmittance. Plotted is the normalized difference between the low-$T$ (10 K) transmittance at $B=0$ and $B=10$ T,

$$\Delta\mathcal{T}'(\omega)=\frac{\mathcal{T}(\omega;10\text{T})-\mathcal{T}(\omega;0)}{\mathcal{T}(\omega;0)} . \quad (3)$$

We observe that applying a field produces the same effect as raising the temperature, i.e., the transmittance above 60 cm$^{-1}$ increases while the transmittance below 60 cm$^{-1}$ decreases.[9]

To understand the behavior of our sample, we must consider it not as a thin film but rather as a composite system of metal particles imbedded in a dielectric host. The behavior of superconducting particle composites has been carefully elucidated by Curtin and co-workers.[10,11] Based on both our preparation technique (which involves no heat treatment) and on visual inspection of our samples, one expects that their cluster percolation model is the relevant one. In this model, a real-space renormalization-group technique is used to obtain cluster dielectric functions, which for modest filling factor can be used in a Maxwell-Garnet formalism to obtain an effective dielectric function for the composite. From this effective medium dielectric function, the absorption coefficient and transmittance as a function of frequency are obtained.

In their model calculation they find a $\Delta\mathcal{T}(\omega)$ similar to our data in Fig. 1 and the zero crossing occurs at about $1.2\omega_{\text{gap}}$. Using this as a guideline, we estimate the $\omega_{\text{gap}}$ to be about 50 cm$^{-1}$ from our data (Fig. 1). Using a superconductor transition temperature of $T_c=30$ K (from our Meissner data) we obtain $2\Delta\equiv\omega_{\text{gap}}\approx 2.5k_BT_c$, which is significantly lower than the BCS value of $2\Delta_{\text{BCS}}=3.5k_BT_c$.

We observe, however, that our data suggest that there is an inhomogeneous distribution of gap frequencies. For example, we do not see a clear peak in $\Delta\mathcal{T}(\omega)$ near $\omega=\omega_{\text{gap}}$ as Curtin, Spitzer, Ashcroft, and Sievers[11] find, and the frequency dependence of our $\Delta\mathcal{T}(\omega)$ curve is more gradual than their calculated result. Both indicate that there is a broad distribution of gap energies, consistent with our resistivity and dc magnetization[8] results mentioned above. If we take the midpoint (20 K) of our $\Delta\mathcal{T}$ versus temperature data (inset Fig. 2) as an average $T_c$ for our inhomogeneous powdered sample, we find $2\Delta=3.6k_BT_c$, a value close to the BCS value.

A smaller non-BCS value for the gap could be explained by a combination of the inhomogeneity of $T_c$ values for our sample and the intrinsic anisotropy (2D) of the crystal structure. The 2D anisotropy could lead to an intrinsic anisotropy of the superconducting energy gap as has been found from tunneling measurements in other anisotropic superconductors. For example, in far-infrared measurements on $V_3Si$ single crystals a spread in the energy gap from $2\Delta=1.0k_BT_c$ to $2\Delta=3.8k_BT_c$ was observed.[12] In an inhomogeneous sample of the 2D superconductor $NbS_2$ an energy gap of $2.3k_BT_c$ was observed.[13] Gaps in most materials range from $3.0k_BT_c$ to strong coupling values of $4.5k_BT_c$ and cluster about the BCS value of $3.5k_BT_c$. It is possible that in more homogeneous samples of

$(La,Sr)_2CuO_4$ the energy gap would be given by the BCS value.

The mechanism responsible for the high-$T_c$ superconductivity in the $La_{2-x}A_xCuO_4$ class of oxides is yet to be determined. Our present assessment is that it is likely to be an electron-phonon mechanism similar to what occurs in the $Ba(Pb,Bi)O_3$ system (where $T_c^{max} \approx 12$ K) but with a higher electron density of states and/or a larger electron-phonon (*ep*) coupling. In $Ba(Pb,Bi)O_3$ a BCS value was found[14] for the energy gap by tunneling experiments even though many other measurements show this material to be a strong-coupling superconductor (i.e., $\lambda \sim 1.3$). Our preliminary specific-heat measurements[15] of the Debye temperature and electron density of states suggest that $La_{1.8}Sr_{0.2}CuO_4$ is also a strong coupled superconductor with $\lambda$ between 1 and 2. The increased *e-p* coupling could be caused by soft phonon modes[16] associated with the orthorhombic-tetragonal phase transition that we observe[3] as the concentration ($x$) of the alkaline earth (Sr,Ba) is increased from undoped $La_2CuO_4$. On the other hand, our observed energy gap is also consistent with the predictions of the bipolaron theory of superconductivity[17] in the intermediate coupling regime ($\lambda \sim 1.5$) but not in the strong coupling regime where the theory predicts gapless superconductivity. It is clear that until more properties of the $(La,A)_2CuO_4$ system are measured all theoretical ideas remain unproven, especially when one realizes that the mechanism for superconductivity in the $Ba(Pb,Bi)O_3$ class of superconductors is not yet completely determined.[18]

In summary, our far-infrared transmission experiment gives conclusive evidence for the existence of a superconducting energy gap in $La_{1.8}Sr_{0.2}CuO_4$. Considering the intrinsic 2D anisotropy of the crystal structure and the sample inhomogeneity we believe our measurement of the gap is consistent with BCS superconductivity.

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