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DIELECTRIC RESPONSE OF La_2CuO_4 AND $EuBa_2Cu_3O_6$

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We have measured the dielectric constant, ε, in the semiconductors La_2CuO_{4+x} and $EuBa_2Cu_3O_{6+x}$. In both cases the crystals are grown by standard techniques and annealed in inert gases, which leads to a semiconductor with $x \simeq 0$, whereas an oxygen anneal produces a superconductor. We have shown earlier that these results are intrinsic to La_2CuO_4 by measuring the temperature dependence of ε at high frequencies. The intrinsic values are $\varepsilon = 23$ for the c-axis of both materials, $\varepsilon = 45$ in the a-b plane of La_2CuO_4, and $\varepsilon = 32$ in the a-b plane of $EuBa_2Cu_3O_6$. The implications of these large dielectric constants for high temperature superconductivity are discussed.

High temperature superconductivity has been well documented in several perovskite-like systems, $K_xBa_{1-x}BiO_3$ ($x \simeq 0.4$), $Sr_xLa_{2-x}CuO_4$ ($x \simeq 0.15$) and $EuBa_2CuO_{6+x}$ ($x \simeq 1.0$). Many experimental studies [1] of the superconductors have concluded that the materials are metals, with typical electron–phonon interactions, and provided no explanation of the atypical transition temperatures. In each case, however, the superconductor is closely related to an insulator with $x \simeq 0$. The doping series from $x \simeq 0$ to larger values is characterized by a monotonically increasing dc conductivity and hall carrier density. The carriers are of hole sign as expected from the nominal valence of the dopants. The close connection between the superconductor and the semiconductor, chiefly differing by carrier density, leads us to study the semiconductor as a possible source of additional insight.

We refer to these materials as perovskite-like because only $BaBiO_3$ is a true perovskite, while the others are a small variation on the perovskite structure. Perovskites often have many structural configurations with nearly equal binding energies and subsequent structural instabilities. In $BaTiO_3$, for example [2], the Ti atom has many positions close together in energy, connected by relative lattice distortions greater than a tenth of an angstrom. This feature has led to proposals by Bednorz and Mueller [3] and others [4, 5], that large lattice displacements occur at low energy cost in the high temperature superconductors and related materials, and may occur in the presence of a carrier. This phenomena could then be the unusual feature that provides the large T_cs in these materials.

In an earlier report [6] we presented substantial evidence for these carrier induced distortions. In a conductivity study over a broad spectral range we found that the carriers in La_2CuO_4 had an anomalously long lifetime, 10^{-11}–10^{-12} s. The spectral weight and the carrier density then implied a dynamical mass of 1100 electron masses. The large mass is expected if lattice displacements of a tenth of an angstrom occur in response to the motion of a hole. In addition to the unusual carrier properties, we have found anomalously large dielectric constants. These large dielectric constants, together with other results, imply that ionic motions are accompanied by substantial electronic polarization. Thus the dielectric constants experimentally demonstrate the converse of our dynamical mass results, and are the focus of this report.

The dielectric constant measurements were performed at 10 kHz with a General Radio capacitance bridge used in a three terminal configuration. A cross-section of the sample cell is shown in fig. 1(a), where the sample is the shaded region. The active leads of the capacitance bridge are connected to the metal plates (1) on each side of the sample. The plates are isolated from the wall with machined ceramic mounts (2 and 5). One of the plates is spring loaded (3) to prevent the sample from slipping during thermal cycling. The third terminal shields both of the active leads and grounds the outer shield of the sample cell. The capacitance measured is described by the equivalent circuit shown in fig. 1(b). The three elements shown are the capacitance between the plates in the volume not including the sample ($C_{\parallel}$), the capacitance between the sample and plates (C_c), and the capacitance of the sample (C_s). $C_{\parallel}$ is measured by using the insulating screw (4) and the threaded sleeve (6) to retract the spring loaded plate. The sample is removed and the plates restored to their original position. The

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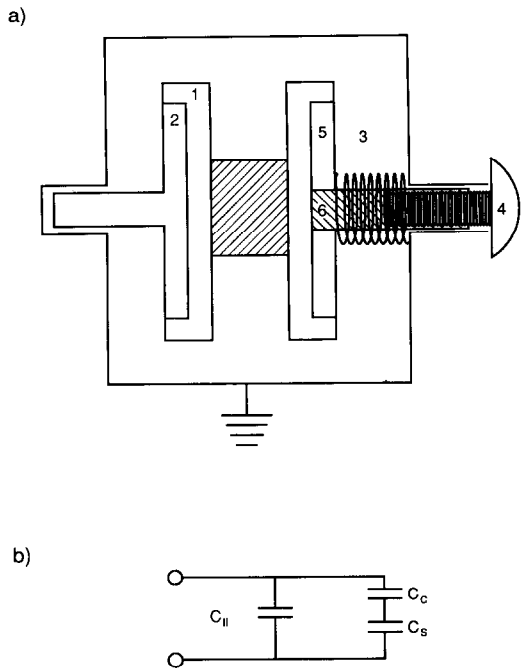

Fig. 1. (a) Cross-section of the experimental cell. (b) The equivalent circuit for the measured capacitance.

capacitance measured with the sample out is corrected for the vacuum capacitance of the portion occupied by the sample to yield $C_{\parallel}$. C_c is the capacitance of the pressed contact to the sample. For polished samples with parallel faces this is large, but non negligable and difficult to reproduce. To control this we insert a 1 μm mylar sheet between the plates and the sample. This reduces C_c, but allows us to cycle the cell reproducibly. At room temperature the samples are always conducting and C_s is shorted. The room temperature capacitance and $C_{\parallel}$ then determine C_c. At low temperature ($T \simeq 4.2$ K) C_s is no longer shorted and may be determined from the measured capacitance and knowledge of the other parameters. The dielectric constant is given by $C_s = \varepsilon\varepsilon_0 A/L$ where A is the area of the sample and L is the distance between the plates.

We repeatedly mounted two La_2CuO_4 samples and one $EuBa_2Cu_3O_6$ sample with various numbers of mylar sheets. This is in addition to measurements of samples with known dielectric constants. The sample dimensions were 1–2 mm on each axis, while the plates where 1 cm in diameter. The sample was placed in the center to mitigate possible fringe field effects. In a typical measurement $C_{\parallel}$ and C_s were the same order of magnitude and C_c was an order of magnitude larger. The bridge has parts per million accuracy for capacitance measurements implying that all significant errors are from C_c and $C_{\parallel}$. We earlier estimated [6] an absolute error of 10% and our additional measurements show that this is easily satisfied.

The samples were twinned crystals of La_2CuO_4 and $EuBa_2Cu_3O_6$ prepared by standard flux growth techniques, annealed in inert gasses, and polished. Powder X-ray studies confirmed the correct crystal structures. The La_2CuO_4 samples [6] had $T_N = 300$ K and the correct stoichiometry within the accuracy of microprobe analysis. The $EuBa_2Cu_3O_6$ samples had a slight excess of Eu and a proportionate deficiency of Ba. The materials, however, were good insulators, the Eu excess was apparently charge balanced by excess oxygen.

The dielectric constant results at 4.2 K are displayed in table I along with twinned crystal results from other groups. The results in the first column are ours, the second column is from a study [7] at similiar frequencies with a network analyzer, and the third column is from an infrared study [8] of polarized reflectance. The overall agreement is excellent. The measurements in column two involved painting contacts across the end of the sample. This results in fringe fields near the edge of the contact which angle across the sample and mix the response of the crystallographic axes. The slightly lower values in the plane and higher values out of the plane may result from exactly such a mixing.

The infrared study [8] shows that the unusually large dielectric constants are caused by large spectral weights of the phonons. Phonon spectral weights are typically analyzed by calculating the lattice dynamics and deriving a mass (in these cases of order an oxygen mass), and attributing any excess spectral weight to an effective charge. In La_2CuO_4 the effective charge is nonphysical, i.e. greater than 10. Physically this must arise [9] from the atomic motion being accompanied by displacement of electronic charge. An infrared study [10] of $MBa_2Cu_3O_6$ powders (where M = Y, Ho, Gd, or Sm) identifies two modes near 110 cm^{-1} that

Table I
The dielectric constants of the cuprates.

La_2CuO_4	ε_c	23	27[a]	23[b]
	$\varepsilon_{a,b}$	45	35	44
$EuBa_2Cu_3O_6$	ε_c	23	–	–
	$\varepsilon_{a,b}$	32	–	–

[a]From ref. [7].
[b]From ref. [8].

dominate the spectra. Assuming that the dielectric constant is produced by these modes and assigning a mass of an oxygen atom to the motion, we again obtain an effective charge in excess of 10. Thus in both cases the large dielectric constant is produced by an anomalously large phonon spectral weight.

In conclusion, the large dielectric constants of these materials arise from large phonon spectral weights which, in turn, result from a close coupling of atomic motion to electronic charge transfer.

Acknowledgment

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