Structural features in the ac conductivity and the static dielectric constant of $La_2CuO_{4+\delta}$ at the metamagnetic transition

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It has been found that the metamagnetic transition in La_2CuO_4, which produces a jump in the ac conductivity $\sigma=\sigma_0+\sigma(\omega)$, has quite different effects on the components σ_0 and $\sigma(\omega)$. The behavior of the static dielectric constant ϵ_0 upon this transition is discussed. It is suggested that the changes observed in σ_0, $\sigma(\omega)$, and ϵ_0 may be due to a change in the localization length of impurities.

The compound $La_2CuO_{4+\delta}$ is distinguished by a strong correlation between magnetic and transport properties in the hopping-conductivity region. This correlation is seen in an anomalously large jump in the resistance upon the metamagnetic transition.[1] Gogolin and Ioselevich's theory[2] explains this effect on the basis of a change in the spin projection of localized states in the course of this transition. Tineke Thio *et al.*[1] have discussed the possibility of explaining the observed jump on the basis of a change in the effective localization length of impurity states, a_B^*. Whether a particular approach is valid for explaining the anomalies in the kinetic characteristics at the metamagnetic transition can be resolved by measuring the ac conductivity σ and the static dielectric constant ϵ in a magnetic field. In the hopping-conductivity region we would have[3]

$$\sigma=\sigma_0+\sigma(\omega)=\sigma_0+A\omega^s, \tag{1}$$

$$\epsilon = \epsilon_0 + \epsilon(\omega) = \epsilon_0 + A\omega^{s-1}\tan(s\pi/2)\,, \qquad (2)$$

where σ_0 is the dc conductivity, A is a constant, $s \leqslant 1$, $\omega = 2\pi f$ is the measurement frequency, and ϵ_0 is the static dielectric constant, due to the polarizabilities of the lattice and of the localized impurity states. According to Mott and Davis,[4] the terms σ_0 and $\sigma(\omega)$ depend on a_B^* in different ways, and when the latter changes at the metamagnetic transition, they should behave differently. There should also be a jump in ϵ_0. In the present letter we separate the contributions to σ and ϵ, and we examine the effect of the metamagnetic transition on σ_0, $\sigma(\omega)$, and ϵ_0.

The capacitance C and the conductance G of an La_2CuO_4 single crystal were measured over the frequency range $300 < f < 10^5$ Hz at $T = 4.2$ K in magnetic fields up to 80 kOe. The test sample was a platelet with typical dimensions of $3 \times 3 \times 0.5$ mm. Copper films $\simeq 0.2$ μm thick were applied to two sides of the sample by magnetron sputtering. The values of C and G of the resulting plane capacitor were measured with the help of a circuit which suppressed the effect of the capacitance of the connecting cables. The conductivity σ and the dielectric constant ϵ were calculated from $\sigma = Gd/S$ and $\epsilon = Cd/S\epsilon_v$, where S and d are the area of the capacitor plates and the thickness of the platelet crystal, respectively, and ϵ_v is the permittivity of free space. The platelet was cut from a single-crystal block in such a way that the tetragonal axis, c, was parallel to the planes of the platelet. In this geometry the electric field was parallel to the CuO_2 planes in the crystal. The magnetic field was in the orientation $\mathbf{H} \| \mathbf{c}$. The Néel temperature of this crystal was $T_N \simeq 250$ K, according to data on the magnetic susceptibility.

Figure 1 shows the frequency dependence of σ (a) and ϵ (b) at $H = 0$ (the open points). The curves are drawn from Eqs. (1) and (2). In drawing these curves we first determined σ_0, A, and s from (1) and then ϵ_0 from (2). We see that at $f > 10^3$ Hz the frequency dependence of σ and ϵ is described by Eqs. (1) and (2). Figure 2a shows curves of $\sigma(H)$ for two measurement frequencies, 300 and 10^5 Hz. We see that ω has a significant effect on both the size of the jump at the metamagnetic transition, $\Delta\sigma = \sigma^{H=80\ \mathrm{kOe}} - \sigma^{H=0}$, and the relative size $\Delta\sigma/\sigma$. This behavior is possible if the metamagnetic transition affects σ_0 and $\sigma(\omega)$ in different ways. Similar curves are found for $\epsilon(H)$. Also shown in Fig. 1, a and b (by the filled points), are plots of σ and ϵ versus f in the strongest magnetic field, $H = 80$ kOe. The curves were fitted to the experimental points by the same procedure as was used at $H = 0$. Expression (1) for σ holds over the entire frequency range, and at any H, within better than 0.7%, which corresponds to the experimental error (Fig. 1a). The values found for $\sigma_0^{H=0}$ and $\sigma_0^{H=80\ \mathrm{kOe}}$ can be used to distinguish the contributions of the metamagnetic transition to the values of σ_0 and $\sigma(\omega)$. Figure 2b shows the frequency dependence of the total relative change in the conductivity at the transition, $\Delta\sigma/\sigma = (\sigma^{H=80\ \mathrm{kOe}} - \sigma^{H=0})/\sigma^{H=0}$ (curve 1), along with $\Delta\sigma(\omega)/\sigma(\omega) = [\sigma^{H=80\ \mathrm{kOe}}(\omega) - \sigma^{H=0}(\omega)]/\sigma^{H=0}(\omega)$, where $\sigma(\omega) = \sigma - \sigma_0$ (curve 2). We see that $\Delta\sigma/\sigma$ depends significantly on ω and that we have $\Delta\sigma_0/\sigma_0 = \lim(\Delta\sigma/\sigma) = 0.75 \pm 0.04$ as $f \to 0$. We find $\Delta\sigma(\omega)/\sigma(\omega) = 1.18 \pm 0.04$ over the entire frequency interval except at low frequencies, where we have $\sigma \simeq \sigma_0$, so the error in $\sigma(\omega) = \sigma - \sigma_0$ is large. It is quite difficult to distinguish the various contributions of the metamagnetic transition to ϵ because (first) $\Delta\epsilon/\epsilon$ is much smaller than $\Delta\sigma/\sigma$ (Fig. 1) and (second) the relation $\epsilon(\omega) > \epsilon_0$ holds over the entire

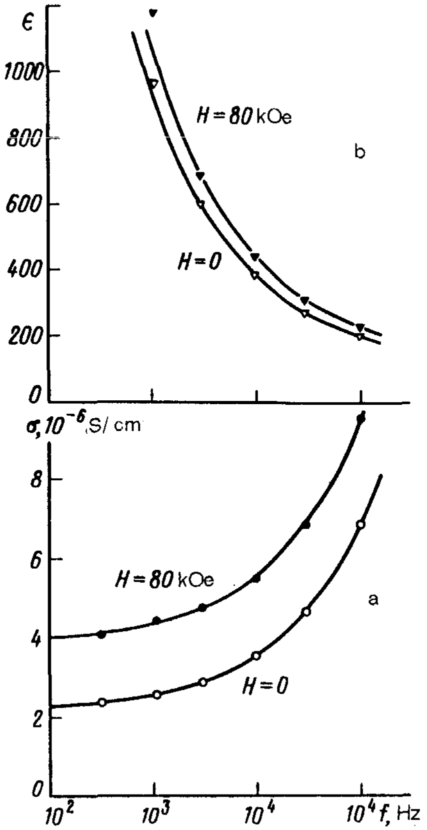

FIG. 1. The frequency dependence of the conductivity σ (a) and that of the dielectric constant ϵ (b) at $H=0$ (open points) and in the magnetic field $H=80$ kOe (filled points).

ω interval, so it is not possible to determine $\Delta\epsilon_0$ accurately by the method used for $\Delta\sigma_0$. To determine $\Delta\epsilon_0$ we use the following relation, which follows from (1) and (2):

$$\epsilon=\epsilon_0+\tan(s\pi/2)\sigma(\omega)/\omega\,, \tag{3}$$

with $\epsilon_0=\lim(\epsilon)$ as $[\sigma(\omega)/\omega]\to 0$. Since we have $\sigma(\omega)=A\omega^3$ and $s\simeq 0.53<1$, this limit corresponds to high frequencies, where the ω dependence of ϵ and σ is described well by expressions (1) and (2) (Fig. 1). Figure 3 shows the experimental data as a plot of ϵ versus $\sigma(\omega)/\omega=(\sigma-\sigma_0)/\omega$ at $H=0$ and $H=80$ kOe. In accordance with (3), we see a linear dependence; an extrapolation to the point $\sigma(\omega)/\omega=0$ yields $\epsilon_0=106\pm 7$. We see that the value of $\Delta\epsilon_0$ lies within the error, which corresponds to the size of the symbols in Fig. 3 and which is due primarily to the error in the determination of σ_0. These results can thus be used to find an upper estimate, $\Delta\epsilon_0/\epsilon_0\leqslant 0.07$.

According to the model of Ref. 4, we would have $\sigma(\omega)\propto(\alpha_B^*)^5$, and $\sigma_0\propto\exp-(\mathrm{const}/\alpha_B^*)^n$. These relations give a qualitative explanation of the increase in both components, $\sigma(\omega)$ and σ_0, and of the different sizes of the jumps, $\Delta\sigma(\omega)/\sigma(\omega)\simeq 0.18$ and $\Delta\sigma_0/\sigma_0\simeq 0.75$, in terms of a change in α_B^* in the course of the metamagnetic transition. On the other hand, the nature of the change in the orbital wave function due to the change in the spin state of the system remains unclear. We

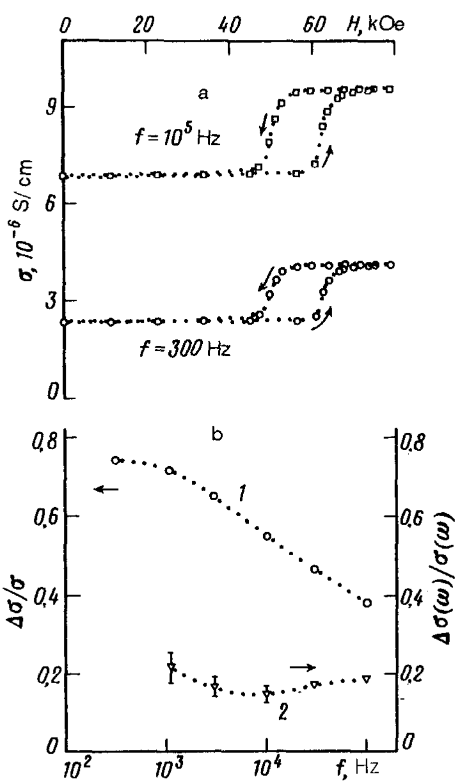

FIG. 2. *a*—Conductivity versus the magnetic field for two measurement frequencies, $f=300$ and 10^5 Hz; *b*—frequency dependence of the relative change in the total conductivity, $\Delta\sigma/\sigma$ (1), and in the contribution $\Delta\sigma(\omega)/\sigma(\omega)$ (2) at the metamagnetic transition.

can work from the size of the jump, $\Delta\sigma(\omega)/\sigma(\omega)\simeq 0.18$, to find the estimate $\Delta\alpha_B^*/\alpha_B^*=0.035\pm0.008$. Since we have $\epsilon_0=\epsilon_h+\epsilon_i$, where $\epsilon_h\simeq 30$ is the static dielectric constant of the stoichiometric La_2CuO_4 lattice,[3] and ϵ_i is the contribution from the polarizability of impurity states [which satisfies $\epsilon_i\propto\epsilon_h(\alpha_B^*)^3$ in the simple Bohr model of an impurity], the value $\Delta\epsilon_0/\epsilon_0\leqslant 0.07$ leads to the estimate $\Delta\alpha_B^*/\alpha_B^*\leqslant 0.03$. This estimate agrees, within the errors, with the value found for $\Delta\alpha_B^*/\alpha_B^*$ from the value $\Delta\sigma(\omega)/\sigma(\omega)$, but it is much smaller than the estimate proposed in Refs. 1 and 3. In

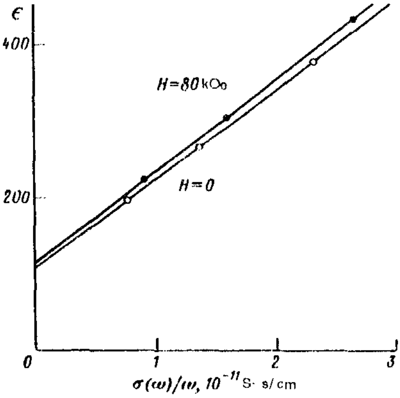

FIG. 3. Frequency dependence of σ and ϵ at $H=0$ (open points) and at $H=80$ kOe (filled points) in the coordinates ϵ, $\sigma(\omega)/\omega=(\sigma-\sigma_0)\omega$.

those papers, the effect of the metamagnetic transition on the terms σ_0 and $\sigma(\omega)$ in Eq. (1) was not analyzed in detail.

The explanation proposed here for the jumps in ϵ and σ at the metamagnetic transition is not certain. All the estimates found here are based on the simple Bohr model for an isolated impurity and Mott's model for the (below-barrier) tunneling of localized electrons. Polaron effects have been ignored, although they apparently play an important role in these compounds. We should also point out that ϵ_h may also vary at the metamagnetic transition, but this possibility has also been ignored. These questions should be resolved by a study of samples with various impurity contents, primarily samples in which the conditions $\epsilon_h \gg \epsilon_i$ and $\epsilon_h \ll \epsilon_i$ hold.

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