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Nonlinear temperature dependence of the normal-state resistivity in $YBa_2Cu_4O_{8\pm\delta}$ films

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The *ab*-plane resistivity of $YBa_2Cu_4O_{8\pm\delta}$ (1:2:4) films is measured from 68 to 600 K and found to be a nonlinear function of temperature in contrast to that of $YBa_2Cu_3O_{7-\delta}$ (1:2:3) films. The nonlinearity can be understood in terms of a Bloch-Grüneisen-type behavior with a transport Debye temperature of $\Theta_D^* \simeq 500$ K, as compared to $\Theta_D^* \simeq 200$ K for 1:2:3. This difference is attributed to the larger number of charge carriers per unit cell in 1:2:4.

A distinct feature of the normal state of the high-T_c superconductors is the linear increase in the resistivity over a large temperature range.[1-3] In $YBa_2Cu_3O_{7-\delta}$ (1:2:3) a deviation from linearity was measured only for $T \gtrsim 600$ K due to loss of oxygen.[1] Several novel types of transport mechanisms have been proposed to account for the observed linear behavior.[4-6] We present here experimental data which suggest that the classical Bloch-Grüneisen (BG) theory of metallic conduction is quite adequate to describe the normal-state *ab*-plane resistivity. The BG model which treats scattering of electrons by phonons predicts a nonlinear increase of the resistivity for T smaller than a characteristic temperature T^* and linear resistivity for $T > T^*$. An examination of the BG dependence shows that $T^* = \eta\Theta_D$, where Θ_D is the Debye temperature and $\eta \sim 1$-2.

A $\Theta_D \simeq 360$ K has been determined from specific-heat measurements on 1:2:3 bulk samples,[7] indicating that nonlinear temperature dependence should be readily seen in 1:2:3 above T_c. However, following Sondheimer's argument for bismuth,[8] we suggest that the relevant temperature is an effective transport Debye temperature $\Theta_D^* = (2k_F/G)\eta\Theta_D$; the latter can be substantially smaller than T^* if the Fermi wave vector k_F is sufficiently small compared to the reciprocal-lattice vector G. Consequently, the high superconducting transition temperature of 1:2:3 would make it difficult to observe the nonlinear behavior predicted by BG theory.

The recently discovered $c = 27$-Å phase of Y-Ba-Cu-O with the stoichiometric composition $YBa_2Cu_4O_{8+\delta}$ (1:2:4) has been found to have a larger number of carriers per unit cell than the 1:2:3 phase.[9-12] This implies a larger Θ_D^* and hence the possibility of an observable nonlinear BG-type resistivity behavior. Kapitulnik and co-workers[13] have discussed the relevance of the BG theory for 1:2:4 films and reported a negative intercept of the zero-temperature resistivity. We present here measurements performed on high-quality 1:2:4 films from T_c up to 600 K which show that nonlinear temperature dependence for 1:2:4 is consistent with a fit to BG theory using $\Theta_D^* = 500$ K. The linear temperature dependence for 1:2:3 is consistent with a corresponding $\Theta_D^* = 200$ K. The measurements on 1:2:4 were extended down to 68 K by suppressing T_c with a magnetic field perpendicular to the *ab* basal plane. The low-temperature results further support the fit to BG theory obtained from the high-temperature data within experimental error ($\sim 5\%$).

The films (thickness 2000–3000 Å) were grown epitaxially on (100)-oriented $SrTiO_3$ substrates using a vacuum deposition system with two electron guns and two thermal evaporation sources, as described elsewhere.[14] The quality of the films was found to be good as evidenced by large critical current densities of $J_c = 1\times10^5$ A/cm^2 (1:2:4) and $J_c = 3\times10^6$ A/cm^2 (1:2:3) at 77 K. X-ray-diffraction analysis confirmed a single-phase $YBa_2Cu_4O_{8\pm\delta}$ with an impurity level of less than $\sim 3\%$ $YBa_2Cu_3O_{7-\delta}$. Au wires (25 μm diam) were attached to the surface using an Ag paint and paste mixture which was cured at 300 °C in a flow of pure O_2. Measurements were performed in an oven cryostat with a flow of O_2 above 300 K and He below 300 K. Resistances were measured using a standard ac lock-in technique with $I = 350$ μA and $f = 11.3$ Hz.

Figure 1(a) shows the measured resistivities and Fig. 1(b) shows the corresponding temperature derivatives of a 1:2:4 (squares) and a 1:2:3 film (triangles). The superconducting transitions ($R = 0$) occur at $T_c = 82$ K (1:2:4) and $T_c = 91.5$ K (1:2:3), with 10–90% widths of 2.8 and 2.5 K, respectively. For $T \gtrsim 580$ K, loss of oxygen in 1:2:3 leads to a sharp increase in resistivity. For 1:2:4, there is no evidence of oxygen loss up to 600 K, but rather a slight indication of saturation. Measurements on 1:2:4 were restricted to $T < 600$ K due to concern of possible phase decomposition.

We note in Fig. 1(b) an anomalous change in slope for 1:2:4 in the temperature region around $T \simeq 250$ K. This feature has been previously observed both in resistivity and in Hall-effect measurements[12,15] and is suggestive of a phase transition as reported earlier for mixed-phase ceramic samples by several groups.[16,17] However, we performed x-ray scattering as a function of temperature and have not found any structural evidence of a phase transition. The linear extrapolation of the low-temperature resistivity to $T = 0$ leads to a negative intercept, which was also reported previously.[12,15,18] We find that the negative intercept results from the BG temperature dependence for sufficiently small residual resistivities.

The semiclassical transport theory leads to the Drude formula for the resistivity $\rho_{BG} = mv_F/ne^2\Lambda$, where the mean free path Λ can be calculated assuming electron-phonon N processes:[19] $1/\Lambda \propto (T^5/\Theta_D^{*3})F(\Theta_D^*/T)$. The function F is evaluated numerically and exhibits a simple asymptotic behavior. For $T \ll \Theta_D^*$, F becomes constant

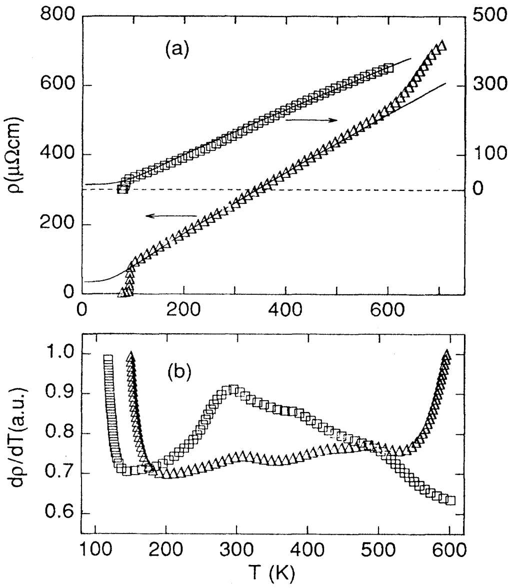

FIG. 1. (a) Measured temperature dependence of the *ab*-plane resistivity of 1:2:3 (△) and 1:2:4 (□) phases of Y-Ba-Cu-O. The solid lines are BG fits with effective Debye temperatures $\Theta_D^* = 200$ K for 1:2:3 and $\Theta_D^* = 500$ K for 1:2:4. (b) Corresponding temperature derivatives of the resistivity.

and at high temperatures $F \rightarrow (\Theta_D^*/T)^4$. The result is an initial T^5 increase of the resistivity and a turnover into linear behavior at sufficiently high temperatures. In order to compare the data with theory, we introduce a residual resistivity $\rho_0 = \rho(T=0)$ and a saturation resistivity $\rho_s = \rho(T \rightarrow \infty)$. A temperature-dependent resistivity of the form $\rho = [1/(\alpha\rho_{\mathrm{BG}} + \rho_0) + 1/\rho_s]^{-1}$, where α is a scaling factor and ρ_{BG} is the BG formula given above, is then fit to the data.

We obtain consistent fits [shown as solid lines in Fig. 1(a)], for 1:2:4 (1:2:3) with $\alpha = 27$ (10.5), $\rho_0 = 14$ (36) $\mu\Omega$ cm, $\rho_s = 0.06$ (∞) Ω cm, and $\Theta_D^* = 500$ K (200 K). The deviations of the data from the BG curves are less than 2% except in the region around 250 K for 1:2:4 where the anomaly is observed. The main conclusions drawn here are that nonlinear resistivity of 1:2:4 is indicative of BG behavior with a large effective Debye temperature and linear resistivity for 1:2:3 can be described by the BG theory with a smaller Θ_D^*. We should note that the values of Θ_D^* are not the results of a multiparameter fit but were obtained by a self-consistent fitting procedure. We chose this procedure in order to give Θ_D^* a higher weight than the other parameters.

The temperature Θ_D^*, where nonlinear BG resistivity crosses over to an essentially linear behavior, can be estimated as follows.[8] At the thermodynamic Debye temperature Θ_D, the maximum phonon wave vector $q_{\max}$ is equal to half the reciprocal-lattice vector $G/2$. The dispersion relation for acoustic phonons implies a linear temperature dependence $q \propto T$ resulting in $q = (G/2)(T/\Theta_D)$. At the crossover temperature Θ_D^*, the thermally excited phonons should have wave vectors large enough to randomize the electron momentum in all directions of k space, stated as $q(T=\Theta_D^*) = \eta k_F$ with $\eta \sim 1$–2, which yields $\Theta_D^* = (2k_F/G)\eta\Theta_D$.

For a simple cubic monovalent metal, $k_F^{3\mathrm{D}} = (3\pi^2 a^{-3})^{1/3} \simeq \pi/a$, and we get $\Theta_D^* \simeq \eta\Theta_D$ which is the crossover point T^* often observed experimentally. However, in 1:2:3 the reduction factor could be smaller than 1 because of a small Fermi surface. Assuming conduction in the *ab*-planes due to one carrier per unit cell, we obtain a 2D Fermi wave vector $k_F^{2\mathrm{D}} \simeq 3 \times 10^7$ cm^{-1}. Taking $G \simeq 1.6 \times 10^8$ cm^{-1} from structural data[20] we estimate $\Theta_D^*/\Theta_D \simeq 0.37\eta$. Specific-heat measurements on 1:2:3 bulk samples yield a thermodynamic $\Theta_D \simeq 360$ K which combined with our BG fit leads to a reduction factor of $\Theta_D^*/\Theta_D \simeq 0.55$, in reasonable agreement with the above estimate.

In order to further test the validity of the BG fit to the 1:2:4 data, we applied a magnetic field perpendicular to the film plane, depressing the superconducting transition, and measured the resistivity down to 68 K. Fields varying from 1 to 13 T were applied. The normal-state resistivity was obtained by extrapolating $(1/\rho)$ vs $(1/H)$ for $(1/H) \rightarrow 0$, a procedure which eliminates the contribution from magnetoresistance.[21] The results are plotted in an expanded scale in Fig. 2 with the data as symbols (▽) and the BG fit as a solid line. We see here clearly an onset of curvature in the data which can be satisfactorily described by the nonlinear decrease of the BG resistivity at low temperatures. This behavior has also been found for 1:2:3 by Tajima *et al.*,[22] where higher magnetic fields had to be applied in order to obtain a similar T_c suppression.

Although specific-heat measurements have not yet been performed on 1:2:4 due to lack of bulk material, we expect the thermodynamic Debye temperature to be close to the value obtained for 1:2:3. Hall-effect measurements[15] on 1:2:4 phase rich films have shown that the carrier concentration n is about 4 times larger than in 1:2:3. From $k_F^{2\mathrm{D}} \propto n^{1/2}$ we estimate $\Theta_D^* \sim 400$ K for 1:2:4, which is somewhat smaller than the value obtained from the fit to the BG theory. This discrepancy can be attributed to the 250-K anomaly causing an increased curvature of the measured resistivity and hence leading to a larger estimate of Θ_D^*.

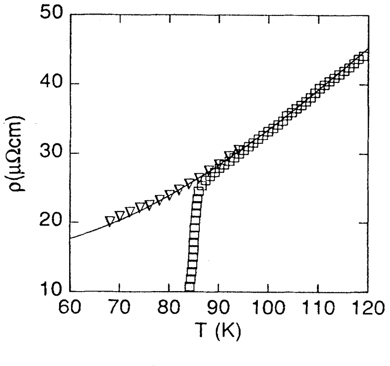

FIG. 2. Temperature dependence of resistivity of the 1:2:4 phase measured down to 68 K (▽) by suppression of T_c in the presence of a magnetic field ($H_{\max} = 13$ T) perpendicular to the *ab* plane. For comparison, the resistivity at zero field (□) and the BG fit (solid line) are shown.

We note that the understanding of the normal-state behavior of $\rho(T)$ should have serious implications for those mechanisms of superconductivity which treat high-T_c oxides as novel, non-Fermi-liquid systems. For example, in the resonating-valence-bond (RVB) theory,[4] normal-state resistivity comes from the scattering of holons by spinons and was predicted to be either linear in T or to have a $T^{3/2}$ dependence.[23] We attempted fits of the 1:2:4 data to a power-law temperature dependence and found that an exponent ~ 1.1 reproduces the data reasonably well ($\approx 2\%$). Therefore, a description of the normal-state resistivity in terms of the RVB theory cannot be ruled out on the basis of the fits alone. The main point we would like to stress, however, is that a "conventional" semiclassical treatment seems to be quite adequate to account for the data presented here.

We further note that linearity of the $\rho(T)$ at high temperatures can be used to estimate an upper bound on the electron-phonon coupling constant λ.[1] It is interesting to do this for the 1:2:4 data, especially since the best fit has been obtained with a small amount of saturation, as discussed above. Using Eq. (5) of Ref. 1, we find a ratio of the mean free path to the interatomic distance of ~ 55 at 600 K as compared to a value of ~ 5 taken as a lower bound in Ref. 1. As a result we obtain an order of magnitude smaller λ than the value of 0.3 estimated for 1:2:3. Clearly this value is too small to explain the high $T_c = 82$ K in agreement with the similar estimate for 1:2:3 samples.[1]

In conclusion, resistivity measurements on high-quality superconducting films of 1:2:4 show that classical BG theory of metallic conduction can account for the observed nonlinearity with an effective $\Theta_D^* \simeq 500$ K. In 1:2:3 films the linear temperature dependence is indicative of a reduced $\Theta_D^* \simeq 200$ K because of the smaller number of carriers per unit cell. Measurements of the resistivity of 1:2:4 down to 68 K in the presence of a magnetic field confirm the BG-type behavior.

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