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Raman scattering in $YBa_2Cu_4O_8$ and $PrBa_2Cu_4O_8$: Indications of pseudogap effects in nonsuperconducting $PrBa_2Cu_4O_8$

J. Bäckström, M. Rübhausen,* M. Käll, and L. Börjesson
Department of Applied Physics, Chalmers University of Technology, S-412 96 Göteborg, Sweden

A. P. Litvinchuk
Texas Center for Superconductivity, University of Houston, Houston, Texas 77204-5932

M. Kakihana
Materials and Structures Laboratory, Tokyo Institute of Technology, Nagatsuta 4259, Midori-ku, Yokohama 226-8503, Japan

M. Osada
Division of Surface Characterization, The Institute of Physical and Chemical Research, 2-1 Hirosawa, Wako-shi, Saitama 351-0198, Japan

B. Dabrowski
Department of Physics, Northern Illinois University, DeKalb, Illinois 60115

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By analysis of the Raman spectra of superconducting $YBa_2Cu_4O_8$ (Y-124), we observe a suppression of low-energy electronic density of states that we link to changes in the linewidth of the Ba-phonon mode. These effects occur at a temperature below 150 K, where also anomalies in resistivity and other physical properties take place. We argue that these changes in the Raman spectra of Y-124 are connected to the opening of a pseudogap. Analogous anomalies in resistivity and the Ba-phonon width in the nonsuperconducting homologue $PrBa_2Cu_4O_8$ (Pr-124) below 150 K suggest that similar pseudogap effects can occur without a following transition to a superconducting state at a lower temperature.

I. INTRODUCTION

One important aspect of underdoped cuprate superconductors is the observation of a partial opening of a gap, a so-called pseudogap, in the electron excitation spectrum already above the superconducting critical temperature T_c, i.e., in the normal state. The pseudogap has been observed by several different experimental techniques, for instance tunneling spectroscopy,[1] angle-resolved photoemission spectroscopy,[2,3] optical conductivity,[4,5] time domain spectroscopy,[6] and electronic Raman scattering[7–12] in most underdoped high-T_c superconductors. A review of experimental observations can be found in Ref. 13. The observations, which are at variance with observations for conventional superconductors and also with BCS theory,[14] are presently much debated and several theoretical models have been proposed to explain it. Mechanisms suggested to lead to the formation of a pseudogap include the occurrence of precursor pairing without phase coherence,[15] d-wave pairing of spinons above T_c,[16,17] a Fermi surface depletion due to volume changes induced by the antiferromagnetic correlations,[18] or the formation of striped phases.[19]

The double CuO chain system $RBa_2Cu_4O_8$ (124, $R=$Y or rare earth) has several advantages for studies of the physical properties of underdoped high-T_c superconductors; it is intrinsically underdoped, the stoichiometry, e.g., oxygen content, is very well defined, and the degree of disorder is low. In addition, the 124 system is intrinsically free from twins, in contrast to $RBa_2Cu_3O_7$, (123) which facilitates measurements probing the anisotropy of properties in the a/b plane.

Several earlier studies of the temperature dependence of physical properties indicate a crossover temperature $T^* \approx 150\,\mathrm{K}$ for Y-124. For example, ^{63}Cu nuclear quadropole resonance (NQR) measurements[20] have revealed effects around T^* in the NQR frequency and the spin-lattice relaxation rate on the CuO_2 plane copper Cu(2), but not the CuO chain copper Cu(1). Resitivity measurements[21–24] have revealed a deviation from a linear temperature dependence in the direction perpendicular to the metallic[24] chains at about T^*. Hall-effect experiments have shown an anomaly in the Hall angle occurring around T^* when current is driven along the CuO chains.[22] A common explanation for the observations made in Y-124 around T^* is an opening of a (spin) pseudogap.[22] The observations made in Y-124 are all supportive of the hypothesis that the effects on physical properties observed around T^* originate from the CuO_2 planes.

The substitution of Pr for Y in the Y-124 and Y-123 systems is a well-known exception from other substitutions with rare earths since it depresses superconductivity completely.[25] The reason for the effect of Pr substitution is not yet settled. Several explanations have been suggested, e.g., hole filling by +4 valent Pr,[26] hybridization of Pr $4f$ and O $2p$ orbitals leading to charge localization,[27] or partial substitution of Pr on the Ba site.[28] For Pr-124 there is, due to the difficulty in preparing the material, few experiments reported in the literature. Resistivity measurements show a change in behavior at about 150 K, from a semiconductorlike high-temperature behavior to a metal-like low-temperature behavior.[29–31]

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Magnetization data for Pr-substituted Y-124 show a deviation in the susceptibility from a Curie-Weiss dependence at about 150 K.[32] A sign of Cu spin ordering around this temperature has been observed in a neutron-scattering experiment,[33] although no definite conclusion could be drawn. Furthermore, changes in the thermoelectric power of Pr-124 around 150 K have been reported.[31] An earlier Raman-scattering and infrared reflectivity study[34] shows phonon anomalies as well as a sudden increase in the ratio of carrier concentration and effective mass when cooling below 150 K. It is clear that a crossover, visible in the temperature dependencies of several physical properties, occurs at around 150 K also in Pr-124. It is, therefore, of interest to investigate low-energy excitations in the Pr-124 compound and make comparative studies with Y-124.

Raman scattering is an important tool in the investigations of strongly correlated systems, as it gives simultaneous information about lattice, charge, and spin excitations. Another advantage of Raman scattering is that it gives information about the coupling between different excitations. In the high-T_c cuprates, the electronic scattering is observed as an almost energy-independent continuum in the normal state, on top of which the phonon peaks sit. The interaction between a Raman-active phonon and a Raman-active electronic continuum reveals itself by a broadening and an asymmetry of the phonon line profile,[35,36] which can be described by a Fano formula,

$$I(\omega)=A\left[Q+\frac{\omega-\omega_0}{\Gamma}\right]^2\Bigg/\left[1+\left(\frac{\omega-\omega_0}{\Gamma}\right)^2\right], \qquad (1)$$

where ω_0 is the energy of the phonon, Γ the half-width, and Q an asymmetry parameter that can be roughly expressed as

$$Q\approx\frac{T_p}{T_e}\frac{1}{V\rho}. \qquad (2)$$

T_p and T_c are the phononic and electronic Raman matrix elements, respectively. V is the electron-phonon coupling matrix element and ρ is the electronic density of states. The half-width acquired from electron-phonon coupling can be written as

$$\Gamma\approx\pi V^2\rho. \qquad (3)$$

These equations are simplified descriptions compared to more complex approaches considering the anharmonic multiphonon decay and the energy dependence of the electronic continuum explicitly.[37,38] However, we find that this simplified picture accounts well for the coupling between the Ba-phonon mode and the electronic continuum treated in this work.

In this paper, we report on a Raman-scattering study of changes in the phonon spectrum and low-energy electronic excitations around T_c and T^* for a Y-124 single crystal. The results are compared with results for a nonsuperconducting Pr-124 sample. The phonon and electronic Raman scattering of Y-124 clearly show the opening of a pseudogap around 150 K.

Surprisingly, Pr-124 and Y-124 exhibit similar anomalies in the phonon parameters around 150 K, coinciding with the semiconductor-to-metal-like transition in Pr-124 and with T^*.

II. EXPERIMENT

A single crystal of Y-124 with a slight Gd doping was synthesized using a self-flux method at high-oxygen pressure as described in Ref. 39. $T_c=82$ K ($\Delta T_c\approx 1$ K) was measured using a standard ac susceptibility technique.

Polycrystalline Pr-124 was produced at ambient pressure using an aqueous solution technique with citric acid as complexant. The details of the synthesis are described in a separate publication.[40] Resistivity measurements on Pr-124 were performed using a standard four-probe ac technique with liquid He as cryogen. Contact pads were painted on the pellet with silver paste and the spring-loaded probes were simply pressed against the pads. Magnetization measurements on Pr-124 were performed using a standard superconduting quantum interference device technique.

The Raman-scattering experiments were carried out with a Dilor LabRam single-grating notchfilter-based micro-Raman setup using the 633 nm line of a He-Ne laser for excitation in a backscattering geometry. The spectrometer is equipped with a Peltier-cooled charge-coupled device detector. The laser spot size was approximately 4 μm in diameter and the laser power incident on the sample was about 2 mW. The samples were mounted in a CryoVac He continuous flow cold finger cryostat capable of reaching temperatures down to 10 K with a stability better than ±0.5 K. All Raman data presented are corrected for the background contribution from the spectrometer and for the thermal Bose factor. We use the Porto notation to indicate polarization geometries, $d_1(p_1p_2)d_2$, where d_1 and d_2 represent the direction of incident and collected scattered light, and p_1 and p_2 represent the polarization of incident and collected light, respectively. All directions are taken with respect to the crystal axes. Primed coordinates represent a coordinate system rotated by 45° from the a/b axes of the crystal.

III. RESULTS AND DISCUSSION

The temperature dependence of the resistance of Pr-124, shown in Fig. 1(a), reveals a peculiar nonmonotonic behavior in the investigated temperature range, with a maximum around 160 K. The overall resistivity in Pr-124 is much lower than in the single-chain compound Pr-123, indicating that the double-chain structure is capable of good electric conductivity. The data indicate a change from a high-temperature semiconductor-like behavior to a low-temperature metallic behavior at about 150–180 K. This observation is well in line with those previously reported.[29,30,33,34] Also magnetization data shown in Fig. 1(b) on Pr-124 show an anomaly occurring at around 150 K, where the susceptibility deviates from a simple Curie-Weiss law, in agreement with the results of Adachi *et al.*[32]

Figure 2 shows Raman spectra taken at 10 K for Y-124 and Pr-124. The spectra contain only modes of the 124 phase, indicating that the impurity content is low (<1%). The Raman-active phonon modes in Y-124 have been as-

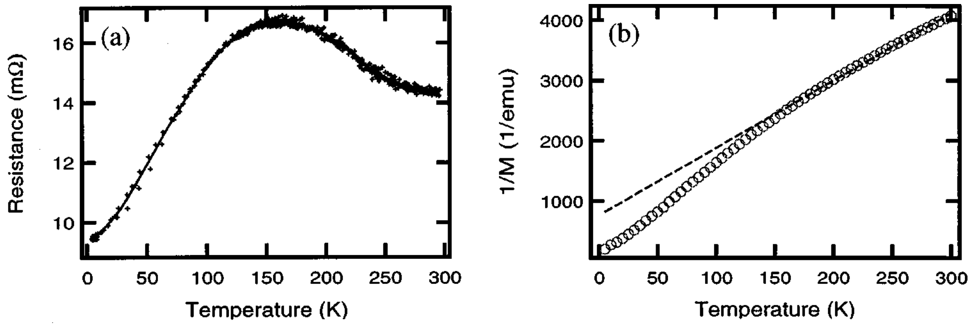

FIG. 1. (a) Resistance as a function of temperature for Pr-124. The line is a guide to the eye. Several thermal cyclings are included in the data. (b) Inverse magnetization vs temperature for Pr-124. The straight line shows an ideal Curie-Weiss behavior.

signed by Heyen *et al.*[41] From left to right, the peaks are the Ba mode at 100 cm^{-1}, the plane Cu(2) mode at 150 cm^{-1}, the chain Cu(1) mode at 254 cm^{-1}, the plane O(2)–O(3) out-of-phase vibration at 337 cm^{-1}, the O(2)+O(3) in-phase at 430 cm^{-1}, the apical O(4) mode at 497 cm^{-1}, and finally the chain O(1) mode at 603 cm^{-1}. Two striking features in the $z(x'x')\bar{z}$ spectrum of Y-124 are the remarkably asymmetric modes, Cu(1) at 254 cm^{-1} and O(2)+O(3) at 430 cm^{-1}, which are seen as almost steplike features in the spectrum. The Pr-124 spectrum shows the corresponding modes at almost the same positions, with the exception of the O(2)–O(3) mode that shifts by about 35 cm^{-1}. The shifts of the phonon energies when replacing Y with Pr can mostly be explained by the size difference between the two ions.[42,43]

We now turn to the temperature dependence of the Y-124 spectra. Around T^*, we find some interesting effects on phonon parameters when altering the temperature. For example, the O(2)+O(3) mode in $z(x'x')\bar{z}$ geometry and the O(2)–O(3) mode in $z(x'y')\bar{z}$ geometry soften and their line profiles become more symmetric below T^*. The energy of the Cu(1) mode in $z(x'x')\bar{z}$ geometry stays almost constant below T^*, in contrast to the expected further hardening. These observations suggest that the interplay between electronic and vibrational degrees of freedom is affected at T^*. The strongest effects occurring around T^* is shown by the Ba mode at 100 cm^{-1}. We will hereby focus our attention to this mode in $z(x'x')\bar{z}$ configuration. The temperature dependence of the Y-124 $z(x'x')\bar{z}$ spectrum in the energy region of the Ba mode is shown in Fig. 3(a). It is clearly seen that the phonon first hardens when lowering the temperature down to 90 K and then softens at still lower temperatures. A Fano line shape [Eq. (1)] was fitted to the phonon profiles, and the extracted fitting parameters for the asymmetry (Q), half-width (Γ), and the position (ω_0) as functions of temperature are shown in Figs. 3(b), 3(c), and 3(d), respectively. The energy of the mode exhibits a clear softening below T_c. Furthermore, the half-width of the phonon decreases rapidly below 150 K and stays almost constant between 90 K and 10 K. The negative Fano asymmetry parameter shows a decrease below 150 K, showing that the line shape becomes more symmetric, indicating that the electron-phonon interaction constant or the strength of electronic continuum decreases. The sharpening occurs at about the same temperature T^* as earlier experiments report anomalies that can be attributed to the opening of a pseudogap.[21–24,44] These

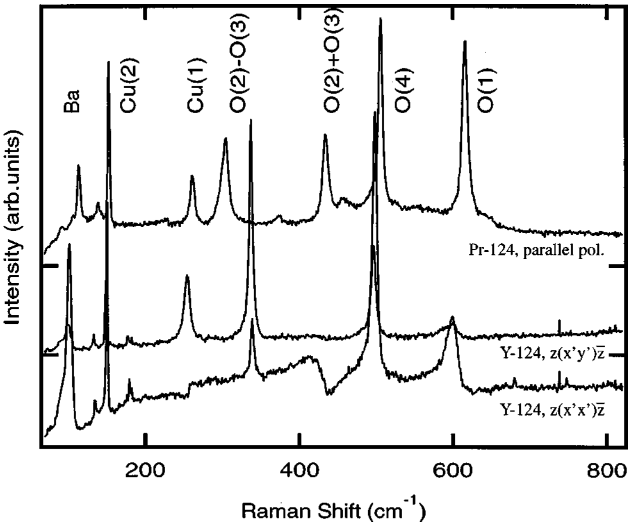

FIG. 2. Raman spectra of Y-124 and Pr-124 taken at 10 K. Baselines are indicated.

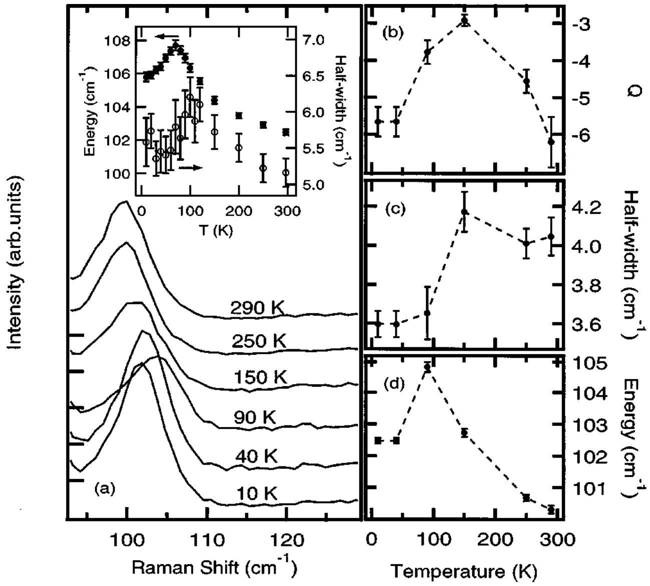

FIG. 3. The Ba phonon mode in Y-124, $z(x'x')\bar{z}$ scattering geometry. (a) shows the phonon line profile as measured, (b) shows the Fano asymmetry parameter Q, (c) shows the half-width, and (d) the energy of the phonon vs temperature. The inset shows the half-width and the energy for the Ba phonon in polycrystalline Y-124 from Ref. 46. Baselines in (a) are indicated. Dashed lines in (b), (c), and (d) are guides to the eye.

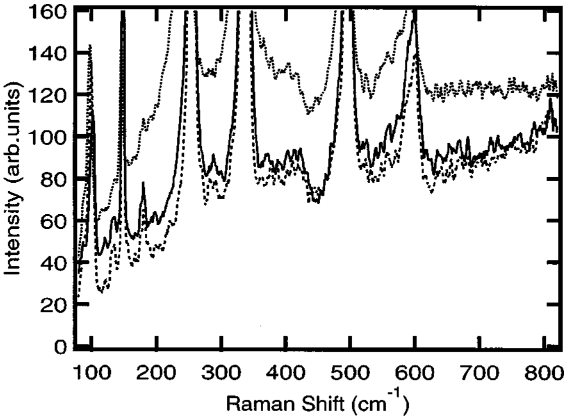

FIG. 4. The electronic continuum in Y-124, $z(x'y')\bar{z}$ geometry. The dotted spectrum is taken at 290 K, the solid line spectrum at 90 K, and the broken line spectrum at 10 K. The curves have been smoothed for clarity.

changes of phonon parameters agree qualitatively well with earlier single crystal[45] and polycrystal[46] Raman-scattering studies on Y-124 (see inset of Fig. 3). Especially the softening of the mode below T_c and the sharpening of the profile below T^* are well reproduced.

We now turn to the changes of the low-energy electronic Raman scattering, which is present as an almost energy-independent continuum in the normal state of cuprate superconductors.[47] Figure 4 shows the 10 K, 90 K, and 290 K Raman spectra of Y-124 in $z(x'y')\bar{z}$ geometry. The spectra taken at 150 K and 250 K overlap essentially with the 290 K spectrum, whereas the 40 K spectrum is almost indistinguishable from the 10 K spectrum. We note a large loss of spectral weight in the whole region below 800 cm^{-1} occurring between 150 K and 90 K. When cooling below T_c, there appears to be a slight further loss of scattering intensity, mainly below 400 cm^{-1}. This indicates a further depression of the low-energy electronic density of states when entering the superconducting state. This would suggest that there is a partial opening of a gap, a pseudogap, occurring between 150 K and 90 K, i.e., well above $T_c = 82$ K, followed by a further, but smaller opening below T_c. Heyen *et al.*[45] have investigated the superconducting gap in Y-124 using Raman scattering. In their study, no sign of a gap opening above T_c was reported. However, their electronic Raman data were collected in $z(xx)\bar{z}$ and $z(yy)\bar{z}$ geometries, which weighs the whole Fermi surface almost equally in contrast to the $z(x'y')\bar{z}$ geometry used by us that is emphasizing along the reciprocal principal axes $\mathbf{k}_x$ and $\mathbf{k}_y$. However, the CuO$_2$ plane related superconducting gap shown in their data to open below 325 cm^{-1} corresponds well to the extra loss of spectral weight we observe below the superconducting transition temperature.

If the Ba mode is coupled to the electronic continuum, we would expect a linear relation between the linewidth of the phonon and the electronic density of states [see Eq. (3)]. We would furthermore expect to see the modulus of the asymmetry parameter Q increase [see Eq. (2)], i.e., the phonon line shape would become more symmetric when the electronic continuum vanishes. This is what we observe in Fig. 3(b). In Fig. 5, we show the integrated Raman intensity between 100 cm^{-1} and 800 cm^{-1} as a function of temperature, together with the half-width of the Ba-phonon profile. It is clear that the temperature dependence of the total intensity correlates well with the Ba phonon half-width for Y-124. In particular, the rapid drop of the intensity below T^*, the pseudogap opening, is well reflected by the phonon half-width.

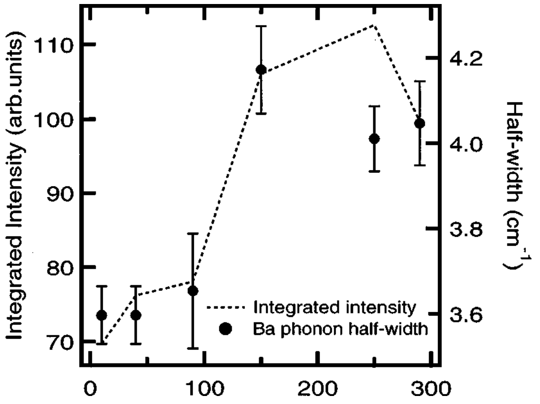

FIG. 5. The 100 cm^{-1}–800 cm^{-1} integrated Raman intensity (dashed line, left scale) and the Ba phonon half-width (circles, right scale) for Y-124 vs temperature.

We now compare the Raman-scattering results obtained for Y-124 with the phonon Raman scattering of the polycrystalline Pr-124 sample. In the Pr-124 spectra, most of the phonons show temperature dependencies typical of normal anharmonic decay. The Ba-phonon mode in Pr-124 (see Fig. 6) is an exception, showing both differences and similarities as compared to the Ba mode of Y-124. The changes with temperature on the phonon parameters are overall more dramatic in Pr-124, as seen by direct inspection of Fig. 6(a). The data were fitted with a Lorentzian[48] to extract the half-width, Fig. 6(b), and the energy, Fig. 6(c), of the phonon as functions of temperature. The phonon energy shows a temperature dependence similar to that observed for Y-124 down to the temperature where Y-124 becomes superconducting. The phonon hardens with decreasing temperature in a manner not in agreement with anharmonic decay. The total change of the Ba phonon energy in the investigated temperature region in

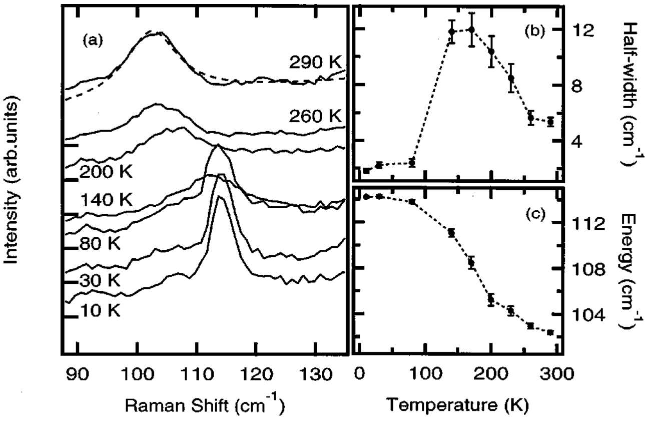

FIG. 6. The Ba phonon mode in Pr-124. (a) shows the line profile as measured; (b) and (c) show the half-width and the energy of the phonon vs temperature, respectively. Baselines in (a) are indicated; the dashed curve is the fitted profile for the 290 K spectrum. Dashed lines in (b) and (c) are guides to the eye.

Pr-124 is around 12 cm^{-1}, which is significantly larger than the corresponding energy change of 5 cm^{-1} in Y-124. The half-width of the phonon increases significantly with decreasing temperature down to about 150 K, then decreases drastically with further cooling to 80 K. Below 80 K the width remains almost constant. This behavior is reminiscent of that observed for Y-124, i.e., for both compounds there is a rapid sharpening of the Ba phonon line below 150 K. The total change of the linewidth is, however, one order of magnitude larger in Pr-124. It can be noted that the very sharp Ba-phonon profile at low temperatures in Pr-124 speaks against a disorder induced by, for instance, any significant Pr substitution on the Ba site.

The polycrystalline nature of the Pr-124 sample made polarized spectra inaccessible, and we cannot make a comparison between the low-energy electronic scattering for Pr-124 and Y-124. It is a reasonable assumption, though, to state that the Ba-phonon anomaly in Pr-124 around 150 K has its origin in a depletion of the electronic density of states to which the phonon couples, similar to Y-124, considering the similar behavior. Resistance measurements do show that there is a crossover in both Y-124 (Refs. 21–24) and Pr-124 occurring at about 150 K. In other words, there are several similarities between the anomalies observed in Y-124 and Pr-124 around 150 K, the temperature where the pseudogap opens in Y-124. This leads to the suggestion of a common origin of the behaviors around T^* in Y-124 and Pr-124.

As a precaution, we note that in some reports it has been suggested that anomalies around 150 K in Y-124 might be related to a lattice instability. For instance, in slightly Ca-doped Y-124 (2.5% Ca), several studies indicate the presence of a second-order phase transition[49–51] attributed to such an instability. It seems likely that these effects are connected to anomalies observed in (pure) Y-124 at the same temperature, considering that even such small Ca dopings induce the effects. Our observations of the change in linewidth for the Ba mode might reflect this instability. However, the excellent correlation between the Ba mode sharpening and the suppression in the electronic excitation spectrum in Y-124 together with the agreement with the Fano formalism, shows that the Ba-phonon anomaly in Y-124 is more likely a pseudogap-induced effect. This does suggest that there is a pseudogap opening also in Pr-124, and therefore that a pseudogap opening is not necessarily followed by a transition to a superconducting state at lower temperatures.

This clearly raises questions about the nature and origin of the pseudogap. One common element connecting Y-124 and Pr-124 is the presence of antiferromagnetic correlations in both compounds. Hence, if part of the pairing interaction is related to these correlations, it is not surprising that one can observe similar effects in the superconducting and nonsuperconducting systems. The difference might arise from that the charge carriers in Pr-124 are more localized and, therefore, are unable to establish a global superconducting state. It appears as if the pseudogap state might be important for the development of the superconducting state, but is in itself insufficient to establish it.

In summary, we have presented results from Raman scattering experiments performed on a single crystal of Y-124 and a polycrystalline sample of Pr-124 at temperatures from 10 K to 290 K. In Y-124, we observe a large loss of spectral weight in the electronic continuum below 800 cm^{-1} at temperatures below $T^* = 150\,K$. We attribute the suppression of spectral weight to an opening of a pseudogap, as several other properties indicate the opening of a pseudogap at this temperature. This suppression correlates well with a sharpening of the Ba-phonon line. The data indicate that a further loss of spectral weight occurs at energies mainly below 400 cm^{-1} when cooling the sample below the superconducting transition temperature. In the phonon Raman data of Pr-124 a sharpening of the Ba mode occurs around 150 K, similar to Y-124. This leads us to suggest that a pseudogap opens also in nonsuperconducting Pr-124 at around 150 K.

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*Present address: University of Illinois at Urbana-Champaign, 1110 West Green Street, Urbana, IL 61801.

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