

**Low-energy excitations around  $(\pi/2, \pi/2)$  points in the pseudogap phase of  $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_4$** A. Koitzsch,<sup>1,\*</sup> G. Blumberg,<sup>1,†</sup> A. Gozar,<sup>1</sup> B. S. Dennis,<sup>1</sup> P. Fournier,<sup>2,3</sup> and R. L. Greene<sup>2</sup><sup>1</sup>*Bell Laboratories, Lucent Technologies, Murray Hill, New Jersey 07974*<sup>2</sup>*Center for Superconductivity Research and Department of Physics, University of Maryland, College Park, Maryland 20742*<sup>3</sup>*Canadian Institute of Advanced Research and Department of Physique, University of Sherbrooke, Sherbrooke, Quebec, Canada J1K 2R1*

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Polarized electronic Raman scattering from the  $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_4$ , superconductor at optimal electron doping ( $T_c = 22$  K) reveals the formation of an anisotropic pseudogap below a characteristic temperature  $T^* \approx 220$  K and energy  $E_g^* = 850$   $\text{cm}^{-1}$ . Below  $T^*$  a pronounced suppression of the incoherent spectral weight below  $E_g^* = 850$   $\text{cm}^{-1}$  in the nodal directions of  $\mathbf{k}$  space is observed. This is concomitant with the emergence of long-lived excitations in the vicinity of the  $(\pm\pi/2, \pm\pi/2)$  points that do not contribute to the optical conductivity.

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The origin of the pseudogap (PG) in cuprate superconductors is believed to lie at the heart of a general understanding of superconductivity in this material class. A large body of experimental results have been collected for hole doped cuprates and many theoretical approaches have been developed.<sup>1</sup> A consensus on the underlying mechanism is still far from being reached. Recently studies of electron doped cuprates have shifted under focus, first, with the reexamination of the symmetry of the order parameter<sup>2-8</sup> whereby a  $d_{x^2-y^2}$  symmetry was established for under and optimal doping, and second, with the investigation of the PG.<sup>9-13</sup> It was also found that electron doped cuprates violate the Wiedemann-Franz law, indicating the breakdown of Fermi liquid theory.<sup>14</sup> These materials offer the unique possibility of comparison with their hole doped counterparts which can serve as a touchstone for theory. Furthermore, electron doped materials such as  $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_4$ , (NCCO) show their own peculiarities. The doping range where the superconducting (SC) transition occurs is much narrower than for hole doped cuprates and the maximum achievable SC transition temperatures ( $T_c$ ) are much lower. Also, the normal state in-plane-resistivity shows a tendency to a quadratic temperature dependence<sup>15</sup> in contrast to the optimally hole doped cuprates where the dependence is linear.<sup>16</sup>

While PG phenomena for hole doped cuprates were investigated by a large number of experimental probes the evidence for a PG in electron doped cuprates is still limited. A recent optics study has found a large PG for underdoped NCCO.<sup>12</sup> For optimally doped NCCO it has been shown that the scattering rate extracted from optical conductivity  $\sigma(\omega)$  is suppressed below certain energies and temperatures.<sup>9,10</sup> An ARPES experiment showed regions of suppressed intensity at the Fermi surface (FS) with a broad peak at about 100 meV ( $\approx 800$   $\text{cm}^{-1}$ ) at low temperatures.<sup>11</sup> These regions are shifted closer to the nodal directions with respect to the hole doped cuprates. On the other hand,  $c$ -axis tunneling studies did not indicate PG suppression above  $T_c$ .<sup>17,18</sup>

Here we report a Raman study of NCCO in the PG phase. In contrast to the observations for hole doped cuprates, for NCCO, a substantial suppression of the Raman response below an onset temperature  $T^* \approx 220$  K and a threshold energy

$E_g^* = 850$   $\text{cm}^{-1}$  was observed for the symmetry that probes excitations in the proximity of the nodal directions of  $\mathbf{k}$  space. Furthermore, we found long lived low energy excitations that reside in the vicinity of  $(\pm\pi/2, \pm\pi/2)$  regions in the PG phase but do not show an observable contribution to the optical conductivity. The excitations in the proximity of the Brillouin zone (BZ) boundaries observed by Raman scale to the optical conductivity data.

The Raman experiments were performed from a natural  $ab$  surface of platelike single crystals grown as described in Ref. 19. After growth, the crystals were annealed in an oxygen-reduced atmosphere to induce the doping level for an optimal  $T_c$ . The SC transition measured by SQUID was about 22 K with a width of about 2 K. The sample was mounted in an optical continuous helium flow cryostat. Spectra were measured in a backscattering geometry using the 6471 Å excitation of a  $\text{Kr}^+$  laser. An incident laser power less than 10 mW was focused to a 50  $\mu\text{m}$  spot on the sample surface. The referred temperatures were corrected for estimated laser heating. The spectra were analyzed by a custom triple grating spectrometer and were corrected for the instrumental response.

The presented data were taken in  $(xy)$  and  $(x'y')$  scattering geometries, with  $x = [100]$ ,  $y = [010]$ ,  $x' = [110]$ , and  $y' = [\bar{1}10]$ . For tetragonal  $D_{4h}$  symmetry the  $(xy)$  and  $(x'y')$  geometries correspond to spectra of  $B_{2g} + A_{2g}$  and  $B_{1g} + A_{2g}$  representations. Measurements in an external magnetic field have been performed with circularly polarized light where a right-left ( $RL$ ) scattering geometry corresponds to spectra of  $B_{1g} + B_{2g}$  representation. In addition, by using right-right ( $RR$ ) geometry, we checked the intensity of the  $A_{2g}$  component and found it to be negligibly weak.

The electronic Raman response function for a given scattering geometry is proportional to the sum over the density of states at the FS weighted by a momentum  $\mathbf{k}$  dependent Raman form factor. By choosing the scattering geometry one can selectively probe different regions of the FS. In the insets of Fig. 1 we sketch the first BZ for NCCO as suggested by ARPES data.<sup>4,5,11</sup> The shaded area corresponds to occupied electron states. For the  $B_{2g}$  channel the Raman form factor vanishes along  $(0,0) \rightarrow (\pi,0)$  and the equivalent lines of  $\mathbf{k}$

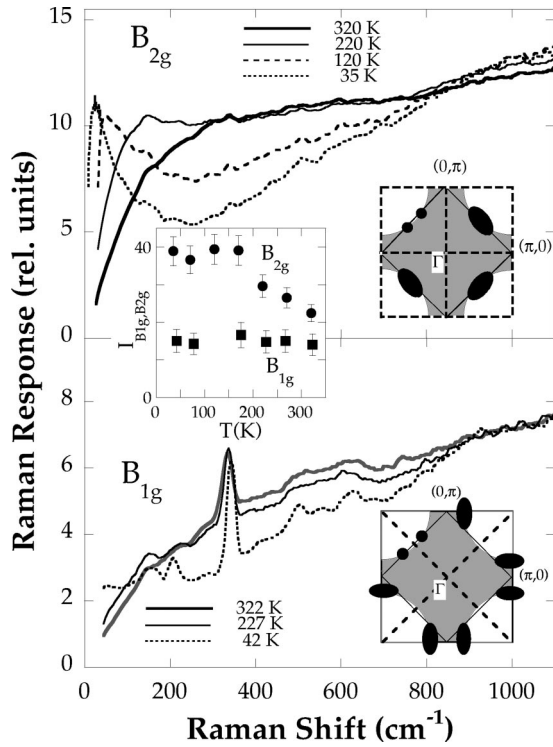


FIG. 1. Raman response function for  $B_{2g}$  and  $B_{1g}$  symmetry channels (1.9 eV excitation) at temperatures between 35 and 320 K. Sketches for the Fermi surface are based on ARPES data (Refs. 4,5,11). Regions where the Raman form factor is maximal are denoted by black ellipses and the nodal regions are shown by dashed lines. The antiferromagnetic Brillouin zone is indicated as a square rotated by  $45^\circ$ . AF fluctuations enhance interactions between fermions around the hot spots (filled circles). The sharp modes for  $B_{1g}$  are phonons (Ref. 20). The inset shows  $I_\mu = \int_0^{850} \chi''_\mu / \omega d\omega$  for  $\mu = B_{1g}$  (squares) and  $B_{2g}$  (circles).

space. The Raman response projects out excitations in the vicinity of  $(\pm\pi/2, \pm\pi/2)$  points of the BZ. In contrast, the  $B_{1g}$  intensity vanishes along the diagonals and the intensity is collected from the regions of the BZ distant from the diagonals. The  $45^\circ$  rotated square indicates the antiferromagnetic Brillouin zone (AFBZ). Intersections of the FS and the AFBZ are marked in the upper left quadrant with black circles. These points are connected by the AF unit vector  $(\pi, \pi)$ .

In Fig. 1 we show the Raman response as a function of temperature for the  $B_{1g}$  and  $B_{2g}$  channels. For  $B_{2g}$  symmetry two distinct features develop in the spectra with cooling:

(i) A “knee-like” intensity loss of the Raman response below  $850 \text{ cm}^{-1}$ . This suppression of spectral weight starts at  $T_{B_{2g}}^* \approx 220 \text{ K}$ . It deepens with cooling and decreasing energy yielding an almost linear slope of the spectra between 300 and  $850 \text{ cm}^{-1}$ . We identify this suppression as the opening of a PG with a temperature independent energy scale.

(ii) An overdamped peak is seen at about  $350 \text{ cm}^{-1}$  at room temperature. This peak sharpens with cooling and shifts to lower frequencies from  $150 \text{ cm}^{-1}$  at 220 K to  $25 \text{ cm}^{-1}$  at 35 K. We refer to this feature as a quasielastic scattering peak (QEP).

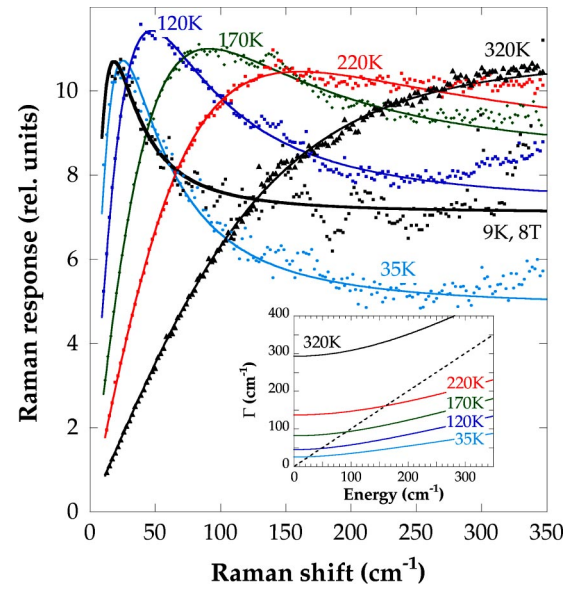


FIG. 2. Low frequency Raman response for the  $B_{2g}$  channel between 35 and 320 K. The quasielastic peak develops with cooling. The 9 K spectra was measured in an external magnetic field applied parallel to the  $c$  axis in RL polarization ( $B_{1g} + B_{2g}$  channel). The solid lines are fits to the extended Drude model (1). Inset: Frequency dependent scattering rate  $\Gamma^{B_{2g}}(\omega, T)$  resulting from the fits to the Drude model (1) for temperatures between 35 and 320 K. The dashed  $\Gamma = \omega$  line represents a crossover from an overdamped regime to longer lived excitations.

The opening of the PG with a similar energy scale is also present but not as pronounced in the  $B_{1g}$  channel.

In contrast, for hole doped cuprates the PG suppression is most pronounced for the  $B_{1g}$  channel.<sup>21–26</sup> For hole underdoped cuprates a destruction of the FS near its intersection with AFBZ boundary has been observed.<sup>27,28</sup> Therefore Raman scattering for the  $B_{1g}$  symmetry is suppressed leaving only small PG effects for the  $B_{2g}$  channel.<sup>22,29,30</sup>

A conjecture that the PG opens up at the intersections of the AFBZ and FS, the “hot spots,”<sup>31–34</sup> reconciles the observed differences for the electron and hole doped materials. Strong antiferromagnetic interactions suppress the spectral weight at the chemical potential in the hot spots leading to a “destruction” of the FS in this region of  $\mathbf{k}$  space and opening of the PG. In hole doped cuprates hot spots are located closer to the  $(\pi, 0)$  points while for electron doping the FS shrinks shifting hot spots closer to the nodal directions.<sup>6,11,27</sup> Hence, for the hole underdoped systems the PG is more pronounced in  $B_{1g}$  while for electron doped systems in the  $B_{2g}$  scattering channel.

Cooling the sample below  $T_c = 22 \text{ K}$  leads to opening of the SC gap and formation of a pair breaking  $2\Delta$  peak.<sup>6</sup> The QEP is suppressed in this regime. An applied magnetic field up to 9 T did not influence either the PG or the QEP above  $T_c$ . In contrast, below  $T_c$ , an external magnetic field of this magnitude strongly suppresses SC correlations<sup>35</sup> and reveals the underlying QEP (see Fig. 2).

In Fig. 2 we show the low frequency part of the Raman response in the  $B_{2g}$  channel. The QEP develops with de-

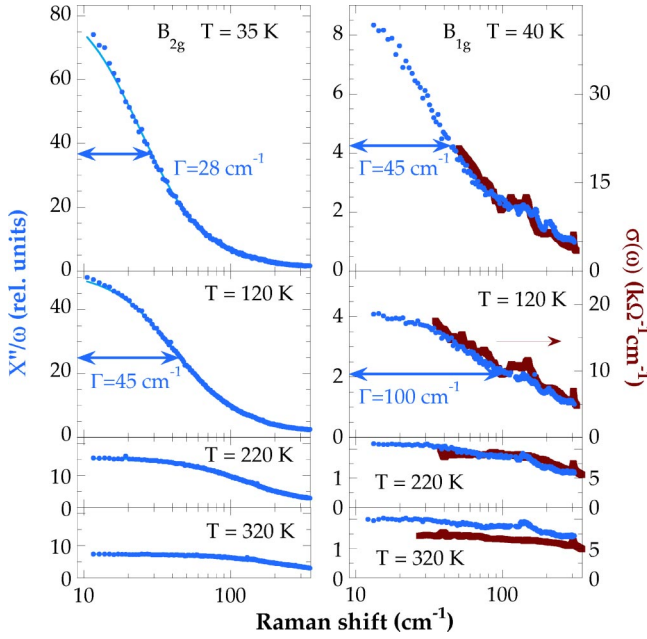


FIG. 3.  $\chi''_{\mu}/\omega$  response for  $\mu=B_{2g}$  and  $B_{1g}$  channels (dots in left and right panels) between 35 and 320 K. The optical conductivity [thick dark (red) solid line] is shown on the right scale (Ref. 9) and is consistent with  $\chi''_{B_{1g}}/\omega$ . The thin solid lines on the left represent the fit to the extended Drude model (1). Phonons have been removed from the  $B_{1g}$  spectra.

creasing temperature. It is also present at temperatures below  $T_c$ , if superconductivity is suppressed by a magnetic field.<sup>36</sup> We fit the low energy  $B_{2g}$  response by an extended Drude model:

$$\chi''(\omega, T) \propto N_F^{B_{2g}} \frac{\omega \Gamma^{B_{2g}}(\omega, T)}{\omega^2 + \Gamma^{B_{2g}}(\omega, T)^2}, \quad (1)$$

where  $N_F^{B_{2g}}$  is the density of states at the Fermi level weighted by a symmetry dependent Raman form factor and  $\Gamma^{B_{2g}}(\omega, T)$  is a frequency and energy dependent scattering rate. The scattering rate  $\Gamma^{B_{2g}}(\omega, T)$  for different temperatures extracted from the extended Drude fits is shown in the inset of Fig. 2.  $\Gamma^{B_{2g}}$  is about  $300 \text{ cm}^{-1}$  at room temperature and decreases rapidly with cooling. The small variation with frequency suggests that a simple Drude model is a good description of the data. However, qualitative differences appear when the momentum dependence of the scattering rate is analyzed.

In Fig. 3 we compare optical conductivity and  $\chi''_{\mu}/\omega$  for  $\mu=B_{2g}$  and  $B_{1g}$  channels between 35 and 300 K. In the lowest order approximation  $\chi''_{\mu}(\omega)/\omega$  is proportional to optical conductivity weighted by a geometrical Raman form factor.<sup>37</sup> We notice that the Drude response narrows with cooling for  $\sigma(\omega)$  as well as for  $\chi''_{\mu}/\omega$  in both  $\mu=B_{2g}$  and  $B_{1g}$  channels. For  $B_{1g}$  the width of the peak decreases from above  $400 \text{ cm}^{-1}$  around room temperature to  $45 \text{ cm}^{-1}$  above the superconducting transition, retaining however a

value about twice that of the  $B_{2g}$  channel at corresponding temperatures. This difference implies a momentum dependence of the scattering rate: a longer lifetime for excitations in the vicinity of  $(\pm \pi/2, \pm \pi/2)$  points and more incoherence for excitations around the BZ boundaries. Our observation of increased coherence in the  $B_{2g}$  channel is consistent with ARPES results, where the quasiparticle peak linewidth at the Fermi energy decreases in going from the BZ boundary to  $(\pi/2, \pi/2)$  point.<sup>11</sup>

The momentum dependence of the scattering rate is consistent with the high frequency Raman data (Fig. 1). In the inset of Fig. 1 we present  $I_{\mu} = \int_0^{850} \chi''_{\mu}/\omega d\omega$  ( $\mu=B_{1g}, B_{2g}$ ) as a function of temperature.<sup>38</sup> While  $I_{B_{1g}}$  is a constant  $I_{B_{2g}}$  increases with cooling until 170 K indicating a shift of spectral weight from regions beyond  $850 \text{ cm}^{-1}$  to lower frequencies.<sup>38</sup> The pronounced suppression of the  $B_{2g}$  Raman continuum intensity is another indication of increased coherence at low energies for excitations in the vicinity of  $(\pm \pi/2, \pm \pi/2)$  points.

On the right side of Fig. 3 we also plot optical conductivity from Ref. 9. Surprisingly,  $\sigma(\omega)$  exhibits almost perfect scaling to  $\chi''_{B_{1g}}(\omega)/\omega$  Raman data: the scaling factor for the double vertical axes was determined for  $T=120 \text{ K}$  data and then used for all temperatures. The fact that the  $B_{1g}$  response bears out the pseudoidentity  $\text{Re } \sigma \propto \chi''_{B_{1g}}(\omega)/\omega$  (Ref. 37) suggests that quasiparticle contributions to the low frequency and dc conductivities are dominated by parts of the Fermi surface away from the  $(0,0) \rightarrow (\pi, \pi)$  diagonal. The more coherent excitations in the vicinity of  $(\pm \pi/2, \pm \pi/2)$  points do not contribute significantly to the optical response. Nevertheless the latter excitations dominate the low frequency  $B_{2g}$  Raman response in the PG state. Although further studies are necessary, we suggest that these chargeless excitations may be responsible for the excessive heat transport that leads to the Wiedemann-Franz law violation.<sup>14</sup>

In summary, we study charge carrier relaxation dynamics in the PG phase of NCCO. We observe suppression of spectral weight below  $850 \text{ cm}^{-1}$  for the  $B_{2g}$  Raman response and identify it as an anisotropic PG in the vicinity of  $(\pm \pi/2, \pm \pi/2)$  points of the BZ. We propose that the PG originates from enhanced AF interactions in hot spot regions, which for electron doped cuprates, in contrast to hole doped, are located closer to  $(\pm \pi/2, \pm \pi/2)$  points than to the BZ boundary. For the Raman response in the  $B_{2g}$  channel we observe a narrow Drude-like QEP in the PG phase. This QEP reveals the emergence of long lived excitations in the vicinity of the  $(\pm \pi/2, \pm \pi/2)$  points that do not contribute to optical conductivity. In contrast, the excitations in the  $B_{1g}$  Raman response were found to be in agreement with optical conductivity data.

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