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We report infrared spectroscopic properties of the strongly correlated layered cobalt oxide $[BiBa_{0.66}K_{0.36}O_2]CoO_2$. These measurements, performed on single crystals, allow us to determine the optical conductivity as a function of the temperature. In addition to a large temperature-dependent transfer of spectral weight, an unconventional low-energy mode is found. We show that both its frequency and its damping scale as the temperature itself. In fact, a basic analysis demonstrates that this mode fully scales onto a function of ω/T up to room temperature. This behavior suggests low-energy excitations of non-Fermi liquid type originating from quantum criticality.

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Beyond the successful Fermi liquid paradigm, non-Fermi liquid states of matter are now the subject of intense theoretical[1–3] and experimental[4,5] investigations. These states can, in particular, be reached in the vicinity of a so-called quantum critical point (QCP) which is located at zero temperature and results from competing interactions tuned by an appropriate nonthermal control parameter, such as pressure, doping, or magnetic field.[6] As quite recently raised by Kopp and Chakravarty, for instance,[7] "a fundamental question is how high in temperature can the effects of quantum criticality persist" and whether "physical observables could be described in terms of universal scaling functions originating from the QCPs" over an extended temperature range. While they demonstrate that the temperature can be surprisingly high, experimental evidence remains to be given to support these theoretical predictions. Also, famous examples of non-Fermi liquid behaviors near QCPs are provided by heavy fermion metals,[5,8,9] which early on emerged as prototypical materials for investigattion of such unconventional properties, and also by some transition metal oxides,[10,11] including the layered cobalt oxide $[BiBa_{0.66}K_{0.36}O_2]CoO_2$.[12] More specifically, the latter compound belongs to a family of doped Mott insulators[13–15] which has revealed striking properties[16–18] such as an enhanced room-temperature thermopower[19,20] and a large negative magnetoresistance.[21] Interestingly, the so-called giant electron-electron scattering inferred in $Na_{0.7}CoO_2$[22] has already led to conjecture of a possible influence of a magnetic QCP in qualitative agreement with density functional calculations predicting a weak itinerant ferromagnetic state competing with a weak itinerant antiferromagnetic state.[23]

In this context, recent susceptibility[12] and specific heat measurements[24] have given good evidence that most of the electronic properties observed in the metallic cobalt oxide $[BiBa_{0.66}K_{0.36}O_2]CoO_2$ display unconventional behaviors which seem to originate from a magnetic-field-induced QCP. In fact, the investigated susceptibility χ has revealed a scaling behavior,[12] with both the temperature T and the magnetic field B ranging from a high-T non-Fermi liquid down to a low-T Fermi liquid. In the latter Fermi liquid regime, the divergent behavior of the Pauli susceptibility follows a power-law dependence $\chi \propto b^{-0.6}$. Here $b = B - B_C$ measures the distance from the QCP and the critical magnetic field $B_C \approx 0.176T$. Furthermore, specific heat measurements allowed the interpretation of the enhancement of the Pauli susceptibility as a result of efficient ferromagnetic fluctuations by analyzing the Wilson ratio.[24] The purpose of this article is to acquire more insight into the nature of the electronic excitations of cobalt oxide $[BiBa_{0.66}K_{0.36}O_2]CoO_2$ by investigating its infrared spectroscopic properties and, especially, its scaling behavior. Therefore, reflectivity measurements have been performed with a Fourier-transformed infrared spectrometer Bruker IFS 66v/S spanning the frequency interval from 40 up to 8000 cm^{-1} and, as a function of temperature, down to 10 K. It is worth mentioning that normal incidence radiation has been used in order to measure the in-plane single-crystal reflectivity. After the initial measurement, the sample was coated *in situ* with a gold film and remeasured at each temperature. These additional data were used as reference mirrors to calculate the reflectivity in order to take into account light scattering on the surface of the sample. Let us also note that the reported results have been measured with several single crystals, thereby ensuring the reproducibility of these investigations. We emphasize that our results also agree with previous reflectivity measurements which have not led to the determination of the optical conductivity,[25] in contrast to the analysis reported in this paper. Figure 1 displays the frequency dependence of the reflectivity within the experimental range as a function of the temperature from 300 down to 10 K. The latter already illustrates an unusual temperature dependence by showing a huge transfer of spectral weight from high to low energies in agreement with previous reflectivity measurements. Here, we propose the analysis of these data by determining the optical conductivity and by investigating its unconventional temperature dependence. Indeed, the reflectivity may be related to the complex dielectric constant $\epsilon(\omega)$ following the Fresnel relation $R(\omega) = |(1 - \sqrt{\epsilon(\omega)})/(1 + \sqrt{\epsilon(\omega)}|^2$. Then the real part of the optical conductivity $\sigma(\omega)$ follows from the imaginary part of the dielectric constant as $\sigma(\omega) = 2\pi c\omega\epsilon''(\omega)$, ω being here the frequency in units of the wavelength inverse, and c the speed of light. Though the complex dielectric constant can be obtained through the standard Kramers-Kronig transformation, we rather proceed with the usual fitting routine with Drude-Lorentz oscillators as $\epsilon(\omega) = \epsilon_\infty + \epsilon_0 \sum_i \frac{\omega_{p,i}^2}{\omega_{0,i}^2 - \omega^2 - i\gamma_i\omega}$.

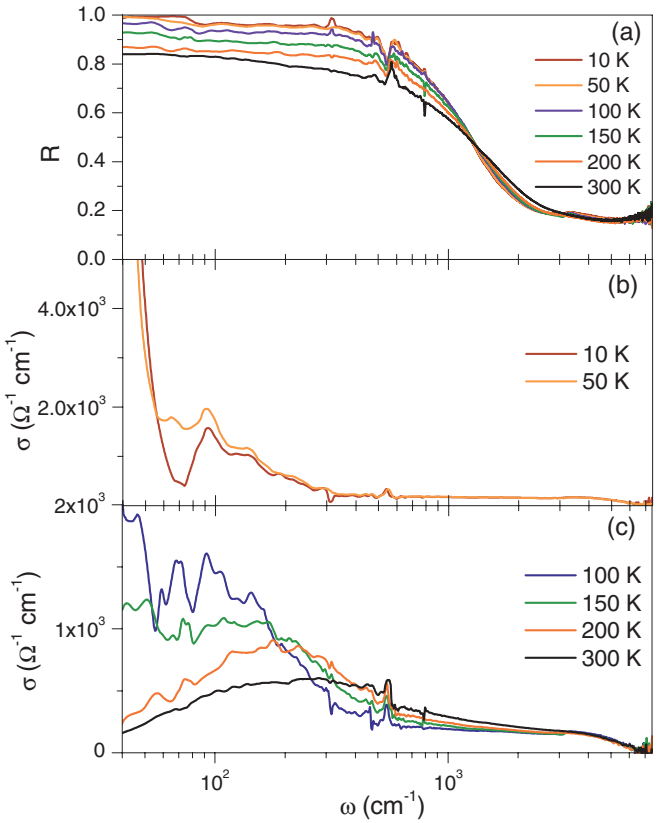

FIG. 1. (Color online) Frequency dependence of (a) the single-crystal reflectivity and (b), (c) the optical conductivity as functions of temperature from 10 K up to 300 K. $\sigma(\omega)$ was obtained with $\epsilon_\infty \approx 7$.

Here, ϵ_∞ is related to an effective contribution of all the high-frequency oscillators out of the experimental range, while $\omega_{p,i}$, $\omega_{0,i}$, and γ_i are the effective plasma frequency, the frequency, and the damping of the ith mode, respectively. Note that a Drude term may be considered to take free charge carriers into account by vanishing the frequency $\omega_{0,i}$.

In order to determine the optical conductivity, we used a variational routine with a causal dielectric function, namely, Kramers-Kronig consistent, which provides the best match to the experimental reflectivity by using typically 500 oscillators. It must be emphasized that according to this procedure, these oscillators do not necessarily have a physical microscopic origin, which makes this routine closer to a fitting procedure than to a modeling one. Yet, such a procedure is equivalent to the standard Kramers-Kronig analysis.[26] Figure 1 shows the variations of the optical conductivity determined using the previous routine within the frequency range from 40 up to 8000 cm^{-1}. In particular, the low-frequency optical conductivity determined at 300 K is of the order of 200 Ω^{-1} cm^{-1}, in agreement with the previously reported dc value 250 Ω^{-1} cm^{-1}.[25] A strong temperature dependence is also observed, with spectral weight redistribution over an anomalously large frequency interval, of the order of the experimental range. We note that a transfer of spectral weight from high energies down to low energies occurs when the temperature is decreased, in order to form a Drude-like peak at a low frequency, below 50 K.

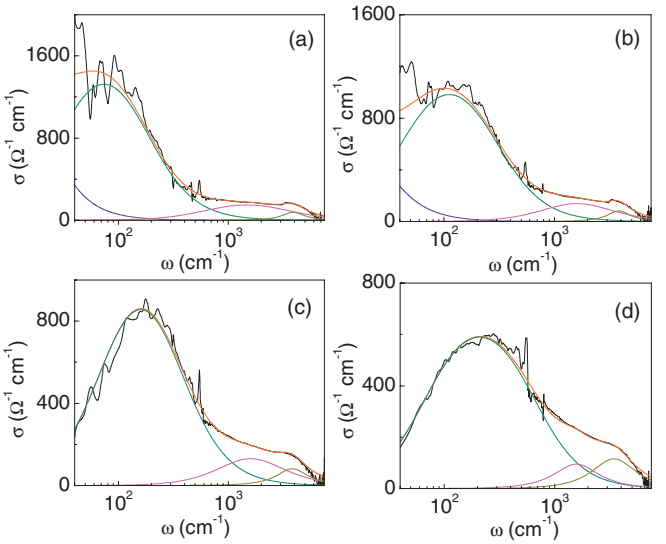

FIG. 2. (Color online) Results of the modeling of the global shape of the reflectivity with variation of the optical conductivity as a function of the frequency at (a) 100 K, (b) 150 K, (c) 200 K, and (d) 300 K. Three Drude-Lorentz oscillators were used, with the frequencies $\omega_{0,i} \approx k_B T/hc$, 1400 cm^{-1}, 4000 cm^{-1}, and a very weak Drude-like contribution at 100 and 150 K.

From a qualitative point of view, let us mention that such a spectral redistribution has already been observed in materials near the Mott metal-to-insulator transition, where low-energy electronic excitations strongly couple to the high-energy ones.[14] Therefore, the behavior of the optical conductivity is likely due to strong electronic correlations as exemplified by the narrow width of the Drude-like peak, which suggests renormalized quasiparticle energies. Interestingly, Fig. 1(c) exhibits an unusual feature above 100 K as a broad maximum with a frequency which increases with temperature and seems to amazingly coincide with $k_B T$, k_B being the Boltzmann constant.

Beyond the previous determination of the optical conductivity, a modeling procedure has been performed in order to analyze quantitatively the aforementioned unusual maximum. Thus, a minimal number of Drude-Lorentz oscillators has been used to account for the global shape of the reflectivity.[27] As shown in Fig. 2, three Drude-Lorentz oscillators have been identified to basically account for the temperature dependence of the optical conductivity as a function of the frequency, with a very weak Drude-like contribution below 150 K. These figures all reveal an important contribution, namely, with a sizable spectral weight, originating from a low-frequency oscillator. They also already demonstrate that the frequency of the latter mode ω_0 increases continuously with the temperature, in a linear fashion. This amazing behavior was indeed checked by plotting in Fig. 3 the temperature dependence of this frequency. Moreover, it appears that not only ω_0 but also the damping γ is directly proportional to the temperature as reported below, with their contribution to the conductivity σ^*:

$$\omega_0 = \frac{k_B T}{hc} = \frac{\gamma}{\pi} \quad \text{and} \quad \sigma^* = \frac{2\pi c \epsilon_0 \gamma \omega^2 \omega_p^2}{\left(\omega_0^2 - \omega^2\right)^2 + \gamma^2 \omega^2}. \qquad (1)$$

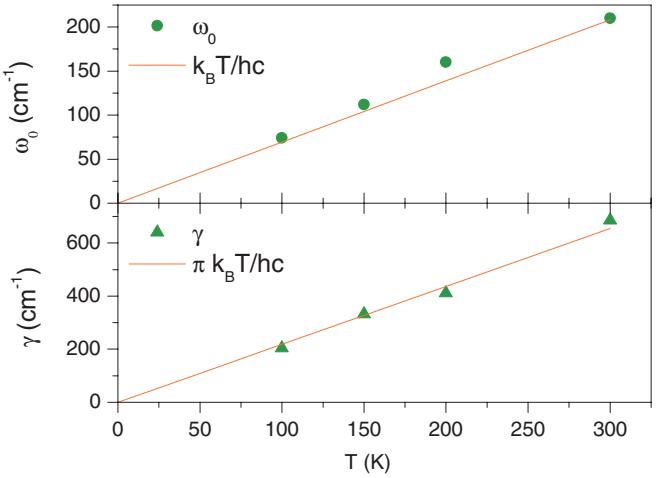

FIG. 3. (Color online) Temperature dependence of the frequency ω_0 and the damping γ for the low-frequency oscillator.

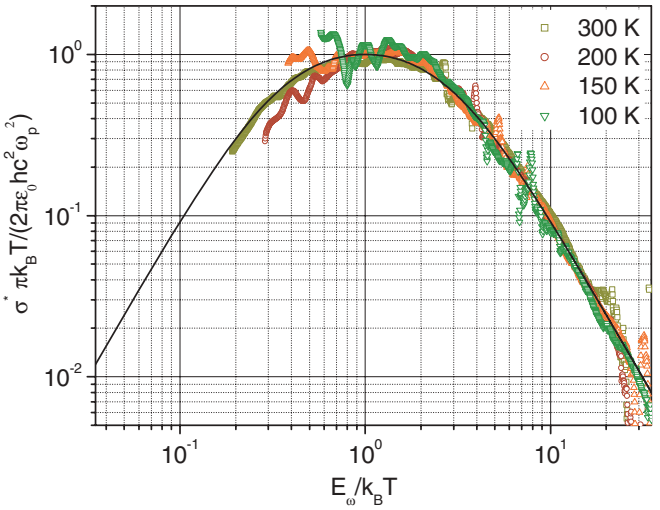

FIG. 4. (Color online) Scaling of the unconventional contribution to the optical conductivity as a function of the dimensionless ratio E_ω/k_BT for $T = 100$, 150, 200, and 300 K as explained in the text. Note that this contribution is plotted here as the dimensionless quantity $\sigma^*\pi k_BT/(2\pi\epsilon_0 hc^2\omega_p^2)$. The solid line represents the right-hand side of Eq. (2).

In addition, it is worth mentioning that the temperature dependence of γ is very close to $\pi k_BT/hc$, as demonstrated in Fig. 3.

While not shown here, it should be noted that the effective plasma frequency of this oscillator seems to exhibit a moderate and rather linear increase with temperature as $\omega_p \approx 3653 + 4.5T$. Nevertheless, one cannot firmly ascribe a linear power-law dependence because of the weakness of the latter variation. On the other hand, according to the linear temperature dependencies of both ω_0 and γ in Eq. (1), the contribution σ^* actually appears to depend only on the dimensionless ratio between the frequency ω and the temperature following Eq. (2), with the photon energy $E_\omega = hc\omega$ and $x = E_\omega/k_BT$:

$$\frac{k_BT}{2hc^2\epsilon_0\omega_p^2}\sigma^* = \frac{\pi^2x^2}{\pi^2x^2 + (1-x^2)^2}. \tag{2}$$

As a result, one should be able to highlight such a contribution by subtracting those of the other two oscillators displayed in Fig. 2 and corresponding to the frequencies $\omega_{0,i}$ around 1400 and 4000 cm^{-1}. By using the values of the effective plasma frequency as given by the aforementioned approximate linear relation and the parameters of the two high-energy Drude-Lorentz oscillators, including the small Drude part at 100 and 150 K shown in Fig. 2, the residual low-energy contribution can thus be selected and plotted as a dimensionless quantity as a function of the dimensionless ratio E_ω/k_BT. As a strong check of the overall analysis, Fig. 4 shows that the data related to σ^* nicely collapse onto a single curve over nearly two decades, in full agreement with Eq. (2). The normalized maximum is, in particular, recovered for $E_\omega/k_BT = 1$, which firmly checks the unconventional linear temperature dependencies of both ω_0 and γ as displayed in Fig. 3.

Since neither the spectral weight nor the scaling properties as a function of ω/T can easily be ascribed to phonons,[28] they suggest that the relevant energy scale of these likely electronic excitations is given by the temperature. Whereas the analyzed oscillator does not contribute to the zero-frequency resistivity at finite temperatures, this kind of scaling shares similarities with some properties measured in the high-temperature cuprates superconductors. Indeed, Van der Marel *et al.* gave good evidence that the optical conductivity of some cuprates follows scaling laws as $\sigma(\omega) = T^{-\mu}g(\omega/T)$.[11] In fact two different scaling functions were found: one in the far infrared for $\omega/T < 1.5$, with $\mu = 1$, and one in an intermediate frequency range (yet with $\omega/T > 3$), with $\mu = 0.5$, stressing the role of the temperature as a crossover frequency put forward by Damle and Sachdev.[29] One should also mention that the scaling form $\sigma(\omega)T = g(\omega/T)$ is automatically fulfilled if the only contribution to the optical conductivity is Drude-like, namely, with $\omega_0 = 0$ in Eq. (1), with a damping varying linearly with temperature, $\gamma \propto T$. In this context, the previous scaling form implies, by considering the zero-frequency limit, a resistivity which increases linearly with the temperature [with $g(0) \neq 0$ here] as experimentally observed. Therefore, the origin of such a scaling form appears to be a central issue since it is directly related to the enigmatic transport in the so-called normal state in cuprates. This is commonly interpreted in terms of quantum critical fluctuations originating from a QCP ($T = 0$). The distance from such a QCP being given by k_BT, the temperature is then the only relevant energy scale governing electronic excitations in the quantum critical regime. In addition, the fluctuation energy is inversely proportional to the correlation time τ and is also related to the correlation length ξ as $\omega \propto \tau^{-1} \propto \xi^{-z}$, with the dynamical exponent z. In the framework where the quantum critical behavior is accounted for by the one-parameter scaling hypothesis, Phillips and Chamon[30] have demonstrated that charge conservation implies the following general scaling form for the optical conductivity:

$$\sigma(\omega, T) = \frac{e^2}{h}\xi^{2-d}g(\omega\tau), \tag{3}$$

with a dimensionless function $g(\omega\tau)$, the elementary charge e, and the spatial dimension d. In Eq. (3) the optical conductivity

is expressed in units of the quantum of conductance e^2/h. Further, one may infer the scaling form Eq. (4) as a function of both the temperature and the frequency as

$$\sigma(\omega,T) = \frac{e^2}{h}\left(\frac{k_B T}{hc_0}\right)^{(d-2)/z} g\left(\frac{\omega}{T}\right), \qquad (4)$$

with c_0 a nonuniversal velocity for $z = 1$.[29] It results that in a three-dimensional system, even anisotropic as in the case of cuprates or in the case of this layered cobalt oxide, the scaling $\sigma(\omega,T) \propto T^{-1} g(\omega/T)$ implies an unphysical negative dynamical exponent $z = -1$ and, thus, an inconsistency between the scaling form, Eq. (2), and the generic equation, (4). Furthermore, the observation of such a scaling behavior over a vast temperature range and up to 300 K invalidates most of the other possible interpretations, in particular, in terms of a classical phase transition. Indeed, its physics is driven by thermal fluctuations which are only critical in a small temperature range, basically around the transition temperature, which vanishes in a quantum critical system. On the contrary, it has been predicted theoretically that quantum criticality can extend up to surprisingly high temperatures, which can even exceed room temperature, because they are essentially bounded by the bare energy scale.[7] Therefore, while the scaling behavior observed here over two decades can be taken as evidence for the persistence of a quantum criticality, the scaling function itself points towards its unconventional nature. This could require a description beyond the one-parameter paradigm[30] as already supported by the previously reported susceptibility measurements.[12] Let us, finally, mention that our method, based on the extraction of the quantum critical contribution, could be tested further in other strongly correlated systems as in some cuprates,[31] in order to analyze the observed peaks at temperature-dependent frequencies.

In conclusion, infrared spectroscopic measurements have been performed on single crystals of cobalt oxide $[BiBa_{0.66}K_{0.36}O_2]CoO_2$. The determined optical conductivity has thus exhibited a large temperature-dependent transfer of spectral weight and an unconventional low-energy mode. We have shown that both its frequency and its damping scale as the temperature itself. So, a basic analysis has demonstrated that this mode fully scales onto a function of ω/T up to room temperature. This behavior suggests low-energy excitations of non-Fermi liquid type originating from quantum criticality, in agreement with the susceptibility, the specific heat, and thermopower measurements.

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