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Gaplike behavior of the *c*-axis dynamic conductivity in pure and Ti-doped Sr_2RuO_4

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We report on infrared spectroscopy of the interplane reflectivity of pure and Ti-doped Sr_2RuO_4 single crystals. The electronic part of the dynamic conductivity can be well described by a two component model, which consists of a coherent part, using the standard Drude model, and a mid-infrared peak. The scattering rate in Sr_2RuO_4 exhibits a gaplike behavior with a gap energy of 6.3 meV. On Ti doping we observe a reduction of the gap, which is fully closed for $x=0.05$. We discuss the origin of the gap and the suppression by Ti doping on the basis of magnetic origin as well as of a dimensional crossover in the charge transport.

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I. INTRODUCTION

The layered perovskite Sr_2RuO_4 has attracted considerable attention since the discovery of superconductivity at $T_c \simeq 1.4$ K.[1] Early theoretical suggestions about *p*-wave superconductivity[2,3] have been corroborated by a number of experimental observations.[4–6] But despite some similarities with superfluid ^{3}He and the search for ferromagnetic (FM) spin fluctuations, the pairing mechanism could not be established.[7] NMR measurements in the normal conducting state established the existence of antiferromagnetic (AFM) spin fluctuations, which are strongly anisotropic and related to the Fermi-surface nesting of the quasi-one-dimensional d_{xz} bands and d_{yz} bands.[8] These observations are consistent with quasi-elastic neutron-scattering results demonstrating the existence of incommensurate spin fluctuations.[9]

In addition to the superconducting properties, also the normal-conducting state received equal attention. At low temperatures Sr_2RuO_4 reveals a metallic behavior along all directions, but the in- and interplane resistivities (ρ_{ab} and ρ_c) are anisotropic with $\rho_c/\rho_{ab} \approx 2000$. Both, the in- and interplane resistivities follow $\rho = AT^2$, however with $A_c/A_{ab} \approx 10^3$ yielding a strongly anisotropic three-dimensional (3D) or quasi-2D Fermi liquid (FL).[10] The interplane resistivity exhibits a crossover to a semiconducting T dependence at $T^* \approx 100$ K, while ρ_{ab} continuously increases up to $T=1300$ K, far beyond the Ioffe-Regel limit.[11]

A "bad metallic" in-plane transport,[12] together with a nonmetallic T dependence of the interplane transport was observed in many layered transition metal oxides, e.g., high-T_c cuprates. In addition, the description of the dynamic conductivity $\sigma(\omega)$ in these systems failed using a standard Drude model (SDM), due to additional spectral weight in the mid-infrared (MIR) range. These anomalous transport properties can be reproduced either by assuming a strongly anisotropic in- and interplane scattering rate[13] (indeed detailed studies of the Fermi surface of Sr_2RuO_4 reveal a strongly *k*-dependent interplane transport, which is dominated by the β sheet, while the in-plane transport mainly arises from the γ sheet[14]), or by dynamic mean-field theory,[15] where the charge dynamics consist mainly of two components: (i) a sharp Drude peak (formed by coherent quasi particle excitation) and (ii) a broad, almost T-independent contribution (from incoherent excitations) at higher frequencies.

First optical studies on Sr_2RuO_4 reflected the strong anisotropic charge dynamic in the system.[16] Later on the far-infrared (FIR) *c*-axis reflectance on "3-K phase" samples, where inclusion of lamellar microdomains of ruthenium metal leads to an enhancement of T_c up to 3 K,[17] were investigated by Hildebrand *et al.*[18] using an extended Drude model (EDM), which assumes frequency dependencies of effective mass m^* and scattering rate γ. They observed a suppression of the scattering rate around 8 meV at low temperatures. Preliminary results on the *c*-axis charge dynamics of highly pure Sr_2RuO_4 single crystals were also given in Ref. 19.

In order to clarify the origin of the scattering mechanism in Sr_2RuO_4 we performed a series of infrared (IR) reflectivity measurements on $Sr_2Ru_{1-x}Ti_xO_4$ with $0 \leqslant x \leqslant 0.05$. The substitution of Ru^{4+} with nonmagnetic Ti^{4+} results in a strong enhancement of ρ_{ab}, which increases towards low temperatures for higher x.[20,21] The *c*-axis transport reveals a purely semiconducting behavior for $x > 0.05$ below room temperature. Already at low doping levels the magnetic susceptibility χ_m exhibits a Curie-Weiss behavior, with a strong Ising-like anisotropy and undergoes a spin-glass transition at $T_m \approx 10$ K.[20,21] In neutron-scattering experiments incommensurate fluctuations were detected evolving into a spin-density wave (SDW) at low temperatures.[22]

II. EXPERIMENTAL DETAILS

Single crystals of $Sr_2Ru_{1-x}Ti_xO_4$ with $x=0$, 0.001, and 0.05 were grown by the floating-zone melting technique in a IR image furnace.[23,20] The pure Sr_2RuO_4 sample reveals superconductivity below $T_c=1.43$ K, the 0.1% sample below 0.62 K, while superconductivity is totally suppressed for $x \gtrsim 0.0015$.[24] There is no indication of the inclusion of the 3-*K* phase in the samples used in this study. The samples examined in this work were rods with typical dimensions of 3–7 mm in ab direction and 2–3 mm in c direction. The

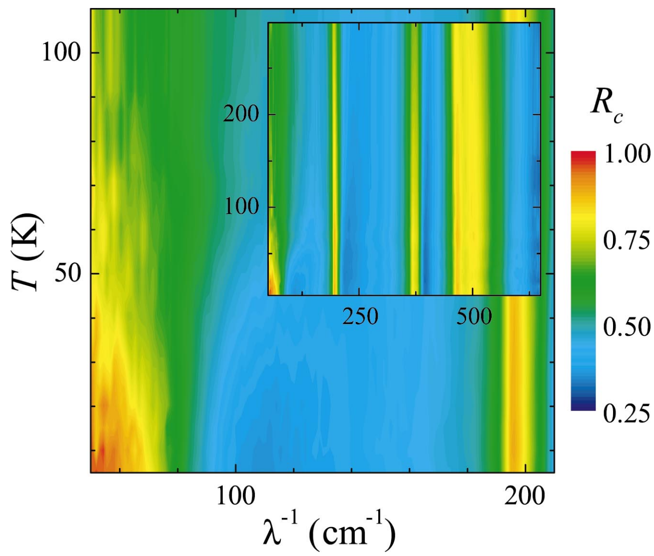

FIG. 1. (Color) Contour plot of the low-energy c-axis reflectivity R_c of pure Sr_2RuO_4 ($T_c = 1.43$ K) vs temperature and wave number (derived from interpolation of 15 measured spectra). The inset shows R_c within the whole FIR range.

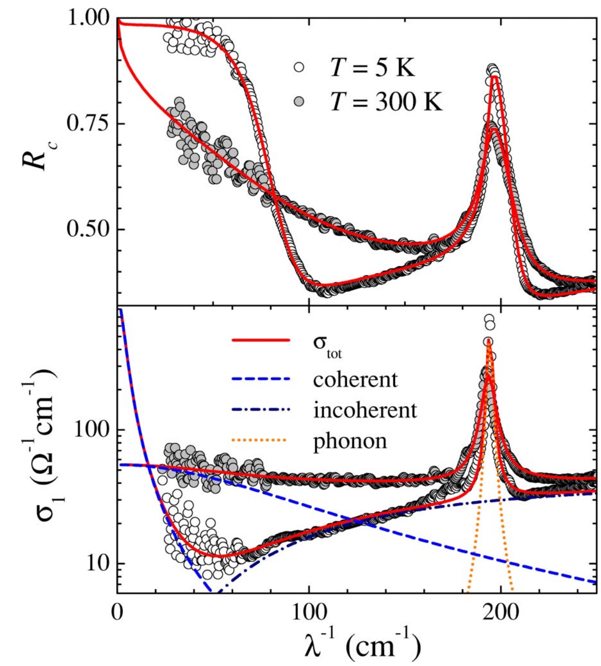

FIG. 2. (Color online) Low frequency c-axis reflectivity (upper frame) and conductivity (lower frame, semilogarithmic representation) vs wave number Sr_2RuO_4 at room temperature and 5 K. The solid lines are the results of fits as described in the text. In the lower frame the different contributions (coherent Drude-like contributions—dashed lines, incoherent contribution—dash-dotted line, and phonon contribution—dotted line) are indicated separately.

measurements of the optical reflectivity were carried out on polished single crystals using a Fourier-transformation IR spectrometer with a full bandwidth of 10–8000 cm^{-1} (Bruker IFS 113v) together with a ^{4}He cryostat (Oxford Optistat) in the range of 5 K$<T<$300 K. For the orientation of the samples we use a IR microscope (Bruker IRscope II).

III. RESULTS AND DISCUSSION

Figure 1 shows the interplane reflectivity R_c as obtained for pure Sr_2RuO_4 in a 2D-contour plot vs wave numbers λ^{-1} and T. The inset gives an overview of the full FIR range and the phonons can easily be observed as bright vertical lines, associated with the three A_u modes at 196, 358, and 465 cm^{-1}.[25] All phonon modes are only weakly T dependent. Between 150 K$<T<$300 K the reflectivity increases below 150 cm^{-1}, which is characteristic for lightly doped semiconductors or bad metals. But at the low temperatures R_c increases towards 1 and becomes minimal close to 100 cm^{-1} indicating a well defined plasma edge. The results immediately signal a very small interplane Drude weight, but also a very low γ, prerequisites for the clear experimental signature of a plasma edge.

For a quantitative analysis we directly fitted R_c. We were unable to describe R_c utilizing a SDM plus Lorentzian for the phonons, due to missing spectral weight in the MIR. As mentioned before, earlier analysis on Sr_2RuO_4 used a EDM to solve this problem.[16,18] Here we use a two-component model, consisting of a T-dependent coherent (SDM) and a T-independent incoherent (broad Lorentzian) contribution (details given in Ref. 26). Our investigations reveal that the incoherent part remains unchanged for $T<100$ K. Although we cannot exclude a small shift from the coherent to incoherent contribution for $T>100$ K, we feel that a two-component analysis is straightforward and provides excellent fits.

The upper frame of Fig. 2 shows the remarkable agreement between these fits (solid lines) and the measured R_c at 5 and 300 K for $\lambda^{-1} \leqslant 250$ cm^{-1}. In the lower frame the conductivity is shown as function of wave number in a semilogarithmic representation. In this case the optical conductivity was calculated from a Kramers-Kronig analysis of R_c assuming a Hagen-Rubens extrapolation towards zero wave number. Again the results of the fits are shown as solid lines. The different contributions for $T=5$ and 300 K, namely the SDM (dashed lines), the incoherent (dash-dotted line), and the phonon (dotted line) contribution are shown separately. The lower frame of Fig. 2 provides clear experimental evidence that an incoherent part, not included in the previous reports, is absolutely necessary to describe the dynamic conductivity in Sr_2RuO_4 agreeing well with theoretical predictions (e.g., Ref. 15) and confirming the two-component model.

As mentioned above, the incoherent contribution was assumed T independent using a resonance frequency of 2000 cm$^{-1}\approx$248 meV, a damping of 26 500 cm^{-1} $\approx$3.3 eV, and an optical dielectric permittivity $\varepsilon_\infty = 5.5$. The Drude conductivity $\sigma_1(\omega) = \sigma_{dc}/(1+\omega^2\tau^2)$ was fitted with a T-dependent dc conductivity $\sigma_{dc} = e^2N\tau/m^*$ and a T dependent, but ω-independent scattering rate $\gamma = 1/\tau$. From these fits the T dependence of γ, σ_{dc}, and $\omega_p = [e^2N/(\varepsilon_0 m^*)]^{1/2}$ can be unambiguously derived and the results are shown in Fig. 3. The plasma frequency (upper frame, left scale) is only weakly T dependent and decreases from 570 cm$^{-1}\approx$71 meV at room temperature to

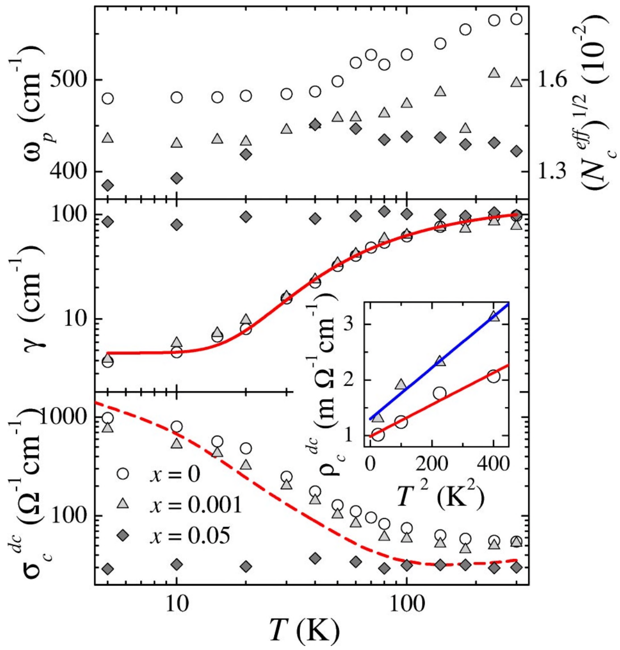

FIG. 3. (Color online) Temperature dependence of the fit parameter ω_p, γ, and σ_c^{dc} as obtained from the standard Drude model for $Sr_2Ru_{1-x}Ti_xO_4$. The solid line in the middle panel is given by fitting the data using Eq. (1). The interplane dc conductivity for $x=0$ measured by four-probe technique[20] is indicated as dashed line in the lower panel. The inset shows $\rho_c = 1/\sigma_c^{dc}$ vs T^2 at low temperatures including fit results (solid lines).

$480\ \mathrm{cm}^{-1} \approx 60$ meV at 5 K. The interplane dc conductivity (Fig. 3, lower frame) is almost constant above 100 K, and strongly increases below 100 K. It closely follows σ_c^{dc} as measured via four-probe techniques (dashed line). The effective number of charge carriers participating in the coherent transport along *c*, N_c^{eff}, (upper frame, right scale) can directly be derived from ω_p and is of the order of 3×10^{-4}, 15 times lower compared to the value determined by band-structure calculations.[27] However, at low temperatures the anisotropy $N_{ab}^{eff}/N_c^{eff} \approx 2\times10^3$ (where N_{ab}^{eff} is taken from Ref. 16) is in agreement with the obtained dc ratio.[24] We estimate the mean free path of the charge carriers $l_c = v_F/\gamma$ using the Fermi velocity v_F from Ref. 27. We find $l_c = 1198$ Å (48 Å) at $T=5$ K (300 K), enhanced by one order of magnitude when compared to the results from Mackenzie *et al.*[28] This enhancement can be partly attributed to the improvement of the crystal quality. But it is also well known that band-structure calculations often overestimate the interplane Fermi velocity giving too large mean-free-path values l_c.

However, the most fascinating *T* dependence is observed in the scattering rate γ, which can be described by an exponential decrease towards low temperatures (middle frame of Fig. 3):

$$\gamma(T) = \gamma_0 + \gamma_1 e^{-\Delta/T}. \tag{1}$$

Here a thermally activated behavior with a gap energy of $\Delta = 74\ \mathrm{K} \approx 51\ \mathrm{cm}^{-1} \approx 6.3$ meV, $\gamma_0 = 4.7\ \mathrm{cm}^{-1}$ and $\gamma_1 = 121\ \mathrm{cm}^{-1}$ reproduces almost perfectly the observations

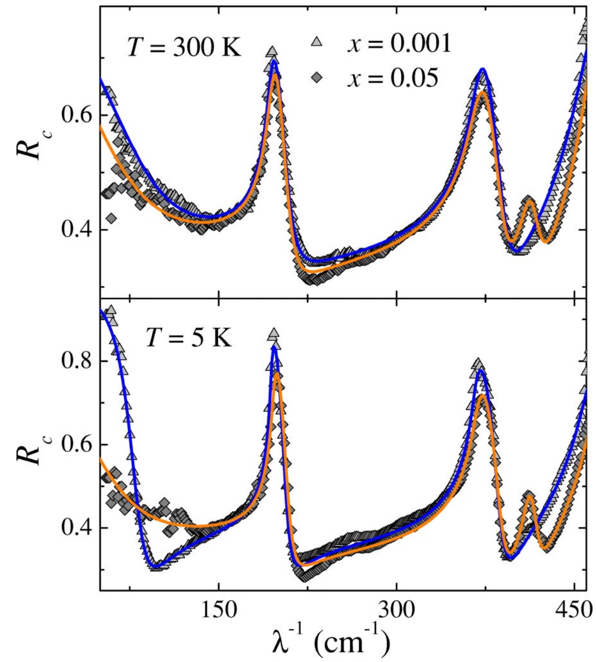

FIG. 4. (Color online) FIR *c*-axis reflectivity of $Sr_2Ru_{1-x}Ti_xO_4$ with $x=0.001$ and 0.05 at $T=300$ K (upper panel) and 5 K (lower panel). The solid lines are results of the fits.

in the complete temperature range investigated. Figures 1–3 suggest a gaplike behavior of the relaxation rate that dominates ρ_c at least up to room temperature and the incoherent transport plays a minor role. But certainly at low temperatures, ρ_c can also be described using the FL ansatz $\rho_c = \rho_0 + A_c T^2$ (solid lines in inset of Fig. 3) with an interplane conductivity coefficient $A_c = 2.9\ \mu\Omega\ \mathrm{cm/K}^2$ ($4.6\ \mu\Omega\ \mathrm{cm/K}^2$) for $x=0$ (0.001), both values in agreement with dc measurements.[10]

Finally Fig. 4 shows R_c as measured at $T=300$ K (upper frame) and 5 K (lower frame) for two Ti-doped samples with $x=0.001$ (bold triangles) and $x=0.05$ (full rhombus). At first sight the spectra for $x=0$ ($T_c=1.43$ K) and $x=0.001$ ($T_c=0.62$ K) look rather similar. A closer inspection at 5 K reveals that ω_p is slightly reduced for $x=0.001$, indicating that N_{eff}^c becomes reduced. For $x=0.05$ the *T* dependence of R_c is rather weak and the evolution of a plasma edge is fully suppressed. The scattering rate remains large for all temperatures.

The strength of all phonon becomes slightly reduced on increasing *x* and a new phonon close to $410\ \mathrm{cm}^{-1}$ becomes apparent. The strength of this impurity mode increases with Ti doping and can probably be assigned to the Ti-O bending mode. The *T* dependencies of R_c for $x=0.001$ and 0.05 have been fitted as outlined above and the resulting parameters have been included in Fig. 3. The plasma frequency and concomitantly N_c^{eff} are significantly reduced on Ti doping, which either is caused by a shift of coherent towards incoherent transport or by an enhancement of the effective mass m^*. For $x=0.001$ the scattering rate closely follows the results observed for $x=0$, but always being slightly enhanced, resulting in a decrease of σ_c^{dc} in good agreement with four-probe dc results.[20] Using Eq. (1) the *T* dependence of γ for

$x = 0.001$ was fitted, yielding a slightly reduced gap of $\Delta = 4.8$ meV. For $x = 0.05$ the scattering rate remains large and almost T independent, indicating that the gap now has been finally closed. As a matter of fact, at low temperatures, the c-axis conductivity becomes significantly reduced on Ti doping. For $x = 0.05$ the dc values are only weakly T dependent and at 5 K reduced by almost a factor 50 when compared to $x = 0$, again in reasonable agreement with the dc measurements.[20]

IV. CONCLUSION

In conclusion our data provide clear experimental evidence that an interpretation of the dynamic interplane conductivity in terms of two components is adequate. In particular the separation into a narrow strongly T-dependent coherent and a broad T-independent incoherent contribution is straightforward. We want to outline two possible scenarios to explain the coherent transport properties.

The strong increase of coherent c-axis conductivity below 100 K results from an abrupt and strong decrease of γ. What is the microscopic origin of this unusual T dependence? It could be the coupling to gapped magnetic excitations, reminiscent to the pseudogap behavior in the high-T_c cuprates. However, neither by NMR nor by neutron scattering a gaplike behavior of spin fluctuations has been observed. The spectrum is dominated by ungapped AFM spin fluctuations arising from Fermi-surface nesting. A correlation between the freezing of spin fluctuations and the suppression of γ can be excluded on the basis of our experiments. A spin-density wave evolves on Ti doping, e.g., $T_m \approx 6$ K for $x = 0.05$, but in this case the scattering rate is T independent as documented in Fig. 3. Of course, the anomalous electronic scattering rate could result from the coupling to so far unobserved FM fluctuations peaked about $q = 0$, which of course is a mere speculation. Very recently, a gaplike structure in the density of states with a width of 5 meV has been observed by tunneling microscopy in pure and lightly ($x = 0.001\,25$) doped $Sr_2Ru_{1-x}Ti_xO_4$.[29] The size of the gap seems to be in the order of our observation, however, the nature of the gap remains unexplained.

A more intriguing explanation could be that at low temperatures Sr_2RuO_4 behaves like a strongly anisotropic but essentially 3D Fermi liquid, where the 2D Ru-O planes are coupled via the coherent c-axis transport to a 3D whole. This change in the effective dimensionality correlates with the presence of coherent quasiparticles in all three dimensions as has been observed recently by Valla *et al.* in layered cobaltates.[30] On increasing temperature the scattering rate increases Fermi-liquid-like proportional to T^2, but when the mean free path decreases to the order of the interplane separation, in-plane scattering processes start to dominate the c-axis transport. That implies several in-plane scattering events of the charge carriers between two successive interplane tunneling transitions and therefore the loss of phase coherence which explain the almost T-independent scattering rate along c at higher temperatures.

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