Low-energy electronic properties of clean CaRuO$_3$: elusive Landau quasiparticles

M. Schneider,1 D. Geiger,2,∗ S. Esser,1 U. S. Pracht,2 C. Stingl,1 Y. Tokiwa,1
V. Moshnyaga,1 I. Sheikin,3 J. Mravlje,4 M. Scheffler,2 and P. Gegenwart1

1. Physikalisches Institut, Georg-August-Universität Göttingen, D-37077 Göttingen
2. Physikalisches Institut, Universität Stuttgart, D-70550 Stuttgart, Germany
3. Laboratoire Nationale des Champs Magnétiques Intenses, 25, Rue des Martyrs, 38042 Grenoble, France
4. Josef Stefan Institute, SI-1000, Ljubljana, Slovenia
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We have prepared high-quality epitaxial thin films of CaRuO$_3$ with residual resistivity ratios up to 55. Shubnikov-de Haas oscillations in the magnetoresistance and a $T^2$ temperature dependence in the electrical resistivity only below 1.5 K, whose coefficient is substantially suppressed in large magnetic fields, establish CaRuO$_3$ as a Fermi liquid (FL) with anomalously low coherence scale. Non-Fermi liquid (NFL) $T^{3/2}$ dependence is found between 2 and 25 K. The high sample quality allows access to the intrinsic electronic properties via THz spectroscopy. For frequencies below 0.6 THz, the conductivity is Drude-like and can be modeled by FL concepts, while for higher frequencies non-Drude behavior, inconsistent with FL predictions, is found. This establishes CaRuO$_3$ as a prime example of optical NFL behavior in the THz range.

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Landau Fermi liquid (FL) theory assumes a one-to-one correspondence of electronic excitations in metals (so-called quasiparticles, QPs) to those of the non-interacting Fermi gas. Remarkably, FL behavior persists in materials where the electronic interactions are strong, often, however, only below a low FL temperature $T_{FL}$. FL theory is the established framework that describes metallic behavior in a broad class of materials ranging from elemental metals to correlated oxides and heavy-fermion systems. Therefore, metals that behave inconsistently with FL predictions attract a lot of attention. A quasi-linear instead of FL-predicted quadratic temperature dependence of the electrical resistivity is found in heavy-fermion metals near a magnetic quantum critical point (QCP) and in high-temperature superconductors at optimal doping [1–3]. It is often controversially discussed whether such non-Fermi-Liquid (NFL) behavior arises due to the interaction of the QPs with low-energy magnetic excitations near QCPs or the QP picture needs to be abandoned completely [4].

NFL behavior has also been discussed in certain ruthenates, i.e. 4d oxides that are, unlike the heavy-fermion compounds and cuprates, characterized by broad electronic bands (with bandwidth $W \sim 3$ eV) and small interaction strength $U \lesssim W$. The optical conductivity of SrRuO$_3$ and CaRuO$_3$ in the infrared exhibits an unusual frequency dependence $\sigma_1 \propto \omega^{-0.5}$ [5, 14], while time-domain THz measurements found signs of an unconventional metallic response at lower frequency [15]. However, disorder strongly influences the properties of ruthenates [8] and FL behavior was observed in sufficiently clean samples of SrRuO$_3$. CaRuO$_3$ was argued to be positioned right at a magnetic QCP, as signified by a logarithmic term in the specific heat coefficient and an electrical resistivity $\rho(T) - \rho_0 \propto T^{3/2}$ ($\rho_0$: residual resistivity) below 30 K [8, 9]. Since paramagnetic CaRuO$_3$ crystallizes in the same orthorhombically distorted perovskite structure as the isoelectronic itinerant ferromagnet SrRuO$_3$ [10, 11], proximity to a ferromagnetic instability indeed seems plausible. The substitution of Sr by Ca in Sr$_{1-x}$Ca$_x$RuO$_3$ results in a suppression of the ordering beyond $x \approx 0.7$ [3, 10, 12, 13]. This is associated with a drastic softening of magnetic exchange [6, 16]. First-principles band-structure calculations indicate that CaRuO$_3$ is located close to a ferromagnetic instability and paramagnon-like spin excitations are very soft [6]. Ferromagnetic dynamical scaling has been found in NMR experiments although the bulk susceptibility reveals a strongly negative Curie-Weiss temperature in CaRuO$_3$ [13].

Complementary insights into ruthenates have come from the dynamical mean-field theory (for a review, see Ref. [17]) which pointed out how the Hund’s rule coupling suppresses the coherence scale and induces strong electronic correlations even in cases where $U \lesssim W$. Interestingly, even in such a local picture, NFL effects are found, with ongoing debate about how much can they persist into the zero-temperature limit [17–19]. The material that might realize such a local NFL is CaRuO$_3$. Precise data on CaRuO$_3$ is thus strongly desired but difficult to obtain, as in CaRuO$_3$ the crystal growth is found to be sophisticated [10, 20] and does not lead to pure-enough samples to address whether the $T^{3/2}$ behavior is a consequence of disorder or intrinsic and whether at low-enough temperatures a FL ground state is established. Answering these questions demands improved quality of the samples. This challenging task is worth undertaking also because other perovskite ruthenates like Sr$_2$RuO$_4$ [21] or Sr$_3$Ru$_2$O$_7$ [22] have shown fascinating low-temperature states in extremely clean samples.

Previously, thin films of CaRuO$_3$ have been synthesized e.g. by sputtering and pulsed-laser deposition tech-
niques [12, 23–25]. However, residual resistivity ratio (RRR) values of order 5-10 only have been obtained. Below, we report the synthesis of high-purity CaRuO$_3$ thin films, first observation of quantum oscillations, and a study of the mass enhancement in this material. Our results demonstrate a fragile FL ground state and robust NFL behavior in its vicinity at elevated temperatures. The coherence scale $T_{FL}$ increases rapidly under applied magnetic field, signaling a strong influence of spin fluctuations on the electronic properties. Low-temperature THz conductivity data indicate that at frequencies above 0.6 THz the optical response is inconsistent with simple Drude behavior and FL predictions. Instead, we find a very strong frequency dependence of the optical scattering rate, which highlights the role of electronic interactions also for the optical properties of CaRuO$_3$.

Details of the CaRuO$_3$ thin film synthesis by the molecular-beam deposition technique, as well as their characterization are provided in Supplementary Material (SM) [26]. An important improvement of the sample quality has been obtained by using (110) oriented NdGaO$_3$ substrates with 3° miscut angle, yielding RRR values up to 55 which is comparable to the best SrRuO$_3$ thin films [27]. Fig. 1a displays the temperature dependence of the electrical resistivity of a CaRuO$_3$ thin film. The surface morphology shown by a scanning tunnelling microscopy (STM) image (Fig. 1b) clearly indicates a step bunching growth mode with steps of about 10 nm height and 220 nm lateral extension, resulting from the 3° miscut angle of the NGO substrate (cf. the profile in Fig. 1c). This evidences a perfect crystal growth. The preferential orientation of the steps is compatible with in-plane epitaxial growth, while out-of-plane epitaxy is also evidenced by XRD (see SM). High-resolution transmission electron microscopy measurements (see SM) indicate homogeneous growth of CaRuO$_3$ without any indication for secondary phases or grain boundaries.

The excellent quality of our thin films is corroborated by the observation of Shubnikov-de Haas (SdH) oscillations as obtained by subtraction of a second-order polynomial from the isothermal magnetoresistance of CaRuO$_3$ at 50 mK for two different thin films at two different field ranges for $B \parallel (110)$. (b) Angular dependence of the oscillation frequencies (symbols) compared to the LDA prediction (lines). (c) Temperature dependence of the SdH amplitude for sample D12. Solid line indicates fit to the Lifshitz-Kosevich formula yielding an effective mass of $4.4m_e$.
tions does not allow their precise determination. In addition, rotating the field out of the direction perpendicular to the thin film plane results in a decrease of the SdH signal and disappearance beyond about 40°. Comparison with LDA band structure suggests that the observed two frequency branches belong to extremal orbits of Fermi surface sheets δ2 and β3 (see SM). The LDA masses of both orbits are about 1.0 m*. The observed mass enhancements due to correlations, ~ 4.4 and ~ 4.2 for the former and latter orbit, respectively, are thus a bit smaller than the average enhancement ~ 7.0 at zero field estimated from the experimental specific heat coefficient of 73 mJ/mol K² [28]. Interestingly, assuming a Kadowaki-Woods relation m* ∝ \gamma ∝ A^{1/2}, the reduction of A by a factor 2 between 0 and 16 T (see below) accounts for most of the difference between the "specific heat mass" and the effective mass observed by SdH oscillations. The field dependence of the oscillation amplitude (not shown) yields an electronic mean free path of 40 nm characteristic for a clean metal, and a quasiparticle scattering time of \tau_{qp} ∼ 1.3 ps. Using the transport life time \tau_{\text{transp}} = 2\tau_{qp} (taking for Z the inverse mass enhancement), the conductivity evaluated from the band theory is \sim 4 \mu \Omega cm, consistent with the residual resistivity of our samples.

In Fig. 3b, we show the temperature-dependent part of the electrical resistivity \Delta \rho(T) = \rho(T,B) - \rho_0(B) down to 50 mK. At low T, the data follow \Delta \rho(T) = A(B)T^2. However, this FL behavior is limited to temperatures below a tiny T_{FL} = 1.5 K. At zero magnetic field, T_{FL} (cf. Fig. 3d) is not only small compared to other oxides (e.g., vanadates and molybdates with T_{FL} > 100 K) but is also substantially smaller than T_{FL} = 7 K found in Sr3Ru2O7 [8]. Taking into account that the strength of correlations as suggested by the specific heat enhancement in Sr3Ru2O7 is actually larger (about 10, at zero field), the FL behavior in CaRuO3 is surprisingly fragile.

The low coherence scale T_{FL} increases strongly in the magnetic field, while the coefficient A is reduced by a factor 2 between 0 and 16 T (Fig. 3c). This behavior is opposite to what is found in Sr3Ru2O7 where T_{FL} decreases in the magnetic field in response to the field-enhanced spin fluctuations driven by the proximity to a metamagnetic critical point. In CaRuO3, the increase of T_{FL} in magnetic field rather suggests a suppression of the magnetic fluctuations and the associated spin entropy. This may hint at the proximity to a zero-field QCP.

Figure 3: (color online) (a) Electrical resistivity of CaRuO3 vs. T^{3/2} at zero field and a field of 9 T applied transverse to the film plane. Dotted straight line indicates T^{3/2} dependence. (b) Low-temperature (between 50 mK and 4 K) electrical resistivity as \rho - \rho_0(B) vs T^2 for various fields. Straight dotted lines indicate Fermi liquid behavior. (c) Coefficient A from \rho - \rho_0 = AT^2 behavior vs. magnetic field. (d) Phase diagram indicating the existence range of Fermi liquid T^2 behavior in the electrical resistivity.

We now address the data beyond the FL regime. Above T_{FL}, the resistivity crosses over from a quadratic to a milder T^{3/2} dependence, as evidenced in Fig. 3a where \rho plotted vs. T^{3/2} appears as a straight line in a broad range up to \sim 25 K. The NFL contribution to the electrical resistivity in this temperature regime is large compared to the residual resistivity of our high-quality thin film (at 25 K, \Delta \rho \sim \gamma \rho_0, cf. Fig. 3a). Such behavior is characteristic for clean NFL metals [3, 29–34]. Importantly, it supports the idea that the NFL behavior is intrinsic to CaRuO3 and not dominated by disorder, e.g. magnetic clusters in a paramagnetic environment [35]. Interestingly, the exponent of 1.5 is similar as found in the NFL state of MnSi [33] but different to the expectation from the itinerant Hertz-Millis-Moriya theory for 3D ferromagnetic quantum criticality [4]. Unlike the lower boundary of the T^{3/2} regime, T_{FL}, its upper boundary (\sim 25 K) is found to be field independent.

We now turn to the optical properties. We have evaluated the complex optical conductivity \sigma(\omega) = \sigma_1(\omega) + i\sigma_2(\omega) from transmission and phase shift measurements at frequencies 0.2 – 1.4 THz and temperatures 2 – 300 K [11] (see also SM). Comparable measurements on single crystals are impossible due to reflectivity very close to unity.

The improved sample quality has striking consequences on the THz response. In Fig. 4(a) we show \sigma_1 at T = 15 K. At low frequencies, \sigma_{1,2} are fit well by a simple Drude response \bar{\sigma} = \sigma_{dc}/(1-i\omega\tau_D) with Drude relaxation time \tau_D. The crossing of the \sigma_1 and \sigma_2 spectra, at (2\pi\tau_D)^{-1} = 0.4 THz, signals the transition from the low-frequency dissipative (transport) regime to the inductive (relaxation) regime. For our lowest temperatures, we obtain \tau_D = 1 ps, in agreement with the SdH and the dc resistivity values discussed above. At frequencies above 0.6 THz, \sigma_1 clearly deviates from the Drude prediction. On the high frequency side, \sigma_1 and \sigma_2 meet again, indicating that the scattering has increased strongly. Comparison with data from Ref. [15], also shown in Fig. 4(a), where such dynamic effects can be resolved only with great difficulty, highlights the role of sample quality for these studies.

In Fig. 4(b) we show the frequency dependence of \sigma_1/\sigma_{dc} for a set of low temperatures. Upon cooling, the
electron-electron contribution to the low-frequency scattering is diminished and the low-frequency Drude roll-off becomes increasingly sharp. At lowest temperatures measured, the narrowing saturates as the scattering due to electron-electron interactions becomes comparable to the impurity contribution. In the NFL temperature range (2 – 25 K), the THz data at frequencies below 0.6 THz can be modeled well by a simple Drude response with an ansatz \( 1/\tau_D = a T^{3/2} + 1/\tau_0 \), with \( \tau_0 = 1.3 \) ps, whereas at higher frequencies all spectra show pronounced deviations from Drude behavior (see SM).

Such deviations were recently discussed by Berthod et al. who have, assuming FL self-energy, derived a universal parameter scaling function \([16]\). In particular, a non-Drude foot in \( \sigma_1 \) and two crossings between \( \sigma_1 \) and \( \sigma_2 \) were discussed there, in qualitative resemblance to our data. In the low-frequency regime, Drude behavior is recovered. As shown in Fig. 4(b), our overall data can be fit remarkably well with this theory, yielding a coherence scale \( T_0 = 150 \) K and an impurity scattering \( Z T = 0.3 \) meV. However, the agreement is only in the Drude part. Substantial deviations from Drude dynamics, predicted by that theory, occur only at frequencies too large to directly account for the unusual behavior we find here \([26]\).

To describe these deviations, an extended Drude analysis \( \hat{\sigma} = D(\omega)/(\Gamma(\omega) - i\omega) \) can be used, where a frequency-dependent scattering rate \( \Gamma(\omega) \) replaces the constant \( 1/\tau_D \) \([37]\). In Fig. 4(c) and (d) we evaluate \( \Gamma(\omega) \) via \( \rho_1(\omega) = \text{Re}(1/\hat{\sigma}(\omega)) \propto \Gamma(\omega) \). For 5 K and frequencies below 0.6 THz, \( \rho_1 \) is almost constant, but for higher frequencies there is a pronounced increase of \( \rho_1 \) due to electronic interactions. This abrupt increase is incompatible with the quadratic FL prediction, calculated from the dc transport prefactor below \( T_{FL} \), as indicated by the dotted line in Fig. 4(c), while the data below 0.6 THz is in perfect agreement with the FL increase. A weaker power-law exponent 3/2, that describes well the temperature dependence in this regime, is even more incompatible with the data above 0.6 THz. For higher temperatures the strong frequency dependence of \( \Gamma(\omega) \) is lost and \( \rho_1 \) basically coincides with the dc resistivity throughout our frequency range, as evident from Fig. 4(d).

To conclude, we have grown high-quality thin films of CaRuO\(_3\) which display for the first time for this ruthenate quantum oscillations. Electrical resistivity indicates an extended NFL regime below 25 K that crosses over to FL behavior below 1.5 K. The optical data reveal a Drude response that at frequencies below 0.6 THz can be modeled with FL concepts. Pronounced deviations from FL behavior are found above 0.6 THz where an abrupt increase of the scattering might signal the coupling to soft spin-fluctuations, a scenario that could be tested by looking at the optical properties as a function of magnetic field.

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\* present address: Institut für Festkörperphysik, Technische Universität Wien

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Thin films of CaRuO$_3$ (CRO) have been prepared by utilizing a metalorganic aerosol deposition technique [1]. Here, a solution of commercial acetylacetonates of Ca$^{2+}$ and Ru$^{3+}$, dissolved in dimethylformamide, is sprayed with a pneumatic nozzle with dried air onto a heated substrate [2]. In the hot region close to the substrate, the acetylacetonates decompose (above 250°C), the metal ions oxidize and the oxide film grows on the substrate. In our previous work on the system Sr$_{1-x}$Ca$_x$RuO$_3$ we have used high-quality SrTiO$_3$ (STO) as substrate and obtained after optimization of the growth conditions (110) oriented thin films of high quality with RRR=29 (x = 0) and 16 (x = 1) [2, 3]. Careful x-ray diffraction (XRD) analysis has revealed that these epitaxial CRO films are fully strained. While STO is well suited for films with composition between x=0 and 0.5, the larger lattice mismatch of $-2\%$ for x = 1 presents higher RRR values for pure CRO. Thus, we have chosen NdGaO$_3$ (NGO) as substrate for the growth of high-quality CRO thin films for which the mismatch is considerably smaller ($-0.5\%$). Very similar growth parameters as reported earlier [2] with a substrate temperature slightly exceeding 1000°C have enhanced the RRR values up to 35. In order to further improve the thin film quality and RRR value, we have chosen vicinal NGO substrates with a 3° miscut angle, which optimizes regular step bunching growth. The surface morphology studied by scanning tunneling microscopy (see Fig. 1b of main paper) has proven a step bunching growing corresponding to the steps of the NGO substrate.

**STRUCTURAL PROPERTIES**

Fig. 1 displays XRD scans of CRO on a vicinal NGO substrate. Here, the top panel shows the intensity spectrum of the plain NGO film while the two lower panels display CRO thin films grown on NGO. The black arrow marks the (110) reflexes from CRO in the orthorhombic Pbnm perovskite structure. No secondary phases are visible. For the thinnest film of thickness 9 nm, Laue fringes are visible which reflect...
Figure 5: $\Theta - 2\Theta$ XRD scans around the (110) reflex of plain vicinal NdGaO$_3$ (NGO) substrate (upper panel), CaRuO$_3$ (CRO) thin film (thickness 78 nm) on vicinal NGO (middle panel) and CRO (9 nm) on vicinal NGO (lower panel). The black arrow indicates the CRO 110 reflex while the grey arrows mark Laue fringes in the very thin film.

Figure 6: Dependence of the (110) lattice constant for CRO thin films on vicinal NGO as function of film thickness, determined by small angle XRD.

Figure 7: (color online) Upper part: transmission electron microscopy (TEM) image on $\mu$m scale of the cross-section of a lamella with NdGaO$_3$ (NGO) substrate, CaRuO$_3$ (CRO) thin film and Pt protection layer (from bottom to top). The 3° saw-tooth surface of the thin film is marked by red lines. The red arrows indicate crystallographic orientations as determined by Fourier transforms of atomic-resolution TEM images of the substrate (lower left) and thin film (lower right).

Figure 8: Residual resistivity of CRO thin films on NGO substrates vs. film thickness. The room temperature resistivity amounts to 230 $\mu\Omega$cm.

A very low surface roughness.

Fig. 2 displays the (110) lattice constant as function of thin film thickness. NGO exhibits a slightly bigger lattice constant compared to CRO which leads to a small in-plane tensile strain acting on the CRO thin films. This results in a decrease of the out-of-plane lattice constant, observed particularly for CRO films of low thickness (cf. Fig. 2). A relaxation of the in-plane tensile strain caused by the negative lattice mismatch of $\sim -0.5\%$ is reached for film thicknesses larger than 20 nm.

High-resolution transmission electron microscopy (TEM) measurements, displayed in Fig. 3, indicate homogeneous growth of CRO without any indication for secondary phases or grain boundaries. The same holds also true for studies on larger lateral dimension. The calculated fast Fourier transform of the cross-sectional TEM images indicate well ordered pattern and epitaxial growth with equal orientation of the film and substrate.

As shown in Fig. 4, the residual resistivity increases with reduced film thickness. This may be attributed to surface scattering and/or the effect of in-plane tensile strain, which may act slightly inhomogeneously.
SHUBNIKOV-DE HAAS EXPERIMENTS

For studying the Shubnikov-de Haas oscillations in the electrical resistance of CaRuO$_3$, two different thin films were micro-structured [3]. The bar width of sample D12 is 50µm, that of sample D41 is 100µm. Dilution refrigerators were utilized for the four-probe resistivity measurements. The first set of data has been obtained on sample D10 in a superconducting laboratory magnet with maximum field of 18 Tesla, applied perpendicular to the thin film plane along the [110] direction. Subsequently, we have performed high-field measurements at the LNCMI Grenoble in an electromagnet with maximum field of 35 Tesla. There, we have measured both thin films simultaneously. Sample D41 has been mounted such that we could study the variation of the quantum oscillations upon rotating the field from the [110] towards the [001] direction. However, as the field is rotated into the thin film plane for an angle larger than 40°, the SdH oscillations are suppressed because the cyclotron orbits interfere with the sample edges.

FERMI SURFACES IN BAND-THEORY

We have calculated the electronic structure of CaRuO$_3$ within the density-functional theory using the local-density approximation (LDA) as implemented in the Wien2k package [4]. The structural information was taken from Table I in Ref. [5]. The electronic structure of CaRuO$_3$ was previously calculated and was in detail discussed [6], however, the detailed information on its Fermi surfaces was not given. Using the SKEAF program [7] we have calculated also the extremal orbits and the corresponding frequencies of quantum oscillations and their effective masses. We visualized the surfaces and the extremal orbits using python scripts that used Mayavi libraries [8]. We checked our visualizations against XCrysdan [9] for consistency. To verify that the residual resistivity is compatible with the band theory we calculated the conductivity using the Boltzmann transport approach described in Ref. [10] and obtained $\sigma_{dc}/\tau_{tr} = 0.15 \times 10^{22}/\Omega\text{m}\text{s}$.

The CaRuO$_3$ DOS is shown on Fig. 9. The DOS at the Fermi energy is 4.41/f.u. eV, which corresponds to a linear coefficient in specific heat $\gamma = 10.4\text{mJ/molK}^2$. The low energy (“$t_{2g}$”) manifold is gapped from the higher energy (“$e_g$”) manifold due to orthorhombic splittings. CaRuO$_3$ electronic dispersions are shown on Fig. 10. As there are 4 formula units in the orthorhombic P$_{bnm}$ unit cell, there are 12 “$t_{2g}$” bands that have also some $e_g$ character due to orthorhombic distortions. In the algorithm, the bands are numbered according to their energy at each k-point separately; thus the band-character is not necessarily followed properly along the k-path. All the crossings between bands are mistaken for anticrossings, as can be seen by following the orbital character that is indicated by the symbol size on the plot. This has no consequences for the identification of Fermi surfaces.

Several bands cross the Fermi energy and several Fermi surface sheets are found. The Fermi surfaces of CaRuO$_3$ are presented on Fig. 11. As the naming convention, we use Greek letters with Latin indices to identify different Fermi surface sheets where $\alpha$ is used for the sheet that encloses the smallest and $\zeta$ the largest volume. Several sheets of the Fermi surface fully enclose another sheet of the Fermi surface of a smaller volume. This is the case for instance for sheets $\beta$. In such cases we label all the corresponding sheets with the same Greek letter, but use Latin number indices to distinguish the sheets among themselves, e.g. $\beta_1$, $\beta_2$, $\beta_3$, $\beta_4$ for the innermost until the outermost sheet, respectively.

On Fig. 11(a) one can see a small hole pocket, that one encounters in the direction $\Gamma \rightarrow U$, and on Fig. 11(e,f) two small electron pockets $\beta_1$ and $\beta_2$, that are at the edge of the Brillouin zone, touching the surface perpendicular to 100 direction. Figs. 11(c,d) show more complex surfaces, where each sheet displayed in (d) has its companion with a larger volume displayed in (c). The sheets $\epsilon_1$ and $\epsilon_1'$ that appear to be separate on (d), merge in their larger volume realization $\epsilon_2$ shown on (c), which is also why we used $\epsilon_1'$, to label one of the two $\epsilon$-sheets presented in (c). Sheet $\gamma_1$ has a peculiar pea-pod (containing three ‘peas’) shape and is enclosed by a more irregular $\gamma_2$ sheet. Center of the face 010 is enclosed in two flat-pebble-like sheets, $\delta_{1,2}$. Fig 11(b) displays $\zeta$ sheet with a complicated shape and topology, see also Fig. 12 for a view from the top. It can vaguely be described as consisting of a central Greek cross shape with additional wings close to the Brillouin zone boundaries perpendicular to the 010 direction. The central cross furthermore contains a spaceous cave around the $\Gamma$ point in its interior, with small “windows” that are visible when looked at from the 001 direction.

Some of the extremal orbits for a few orientations of

![Figure 9: Calculated density of states (DOS) for CaRuO$_3$. The energy is measured with respect to the Fermi energy.](image-url)
Figure 10: LDA band structure close to Fermi energy in CaRuO$_3$. The $d_{xz}$ weight is indicated by the thickness of the symbols. The points along the path are (in units of reciprocal unit wave-vectors) $\Gamma=(0,0,0)$, $X=(0,1\frac{1}{2},0)$, $S=(1\frac{1}{2},1\frac{1}{2},0)$, $R=(1\frac{1}{2},1\frac{1}{2},1\frac{1}{2})$, $U=(0,1\frac{1}{2},1\frac{1}{2})$, $Z=(0,0,1\frac{1}{2})$, $T=(1\frac{1}{2},0,1\frac{1}{2})$. Fermi level (dashed) and its possible spin-dependent shifts in the magnetic field of 10 T, taking Stoner enhancements into account (dotted) are indicated.

Figure 11: Fermi surfaces in CaRuO$_3$. The extremal orbits for magnetic field in 110, 1-10, and 001 directions are also shown. In (c) the experimentally observed orbit is indicated in red. In (d), the experimentally observed orbit in the tilted magnetic field is indicated in cyan.

On Fig. 13 the angular dependence of the SdH frequencies is shown. The window from about 0.1 to about 1kT is quite densely covered. The smallest pockets have frequencies about 80T.

Extremal orbits at around 35 degrees inclination

The Fourier transformed SdH data at 35 degrees inclination from 110 to 001 reveals two well identifiable peaks: one with frequency 541±20T and mass 4.9±0.3$m_e$ and one with frequency 472±20T and mass 4.3 ± 0.4$m_e$. We associate the origin of the signal to the 460 T extremal orbit on sheet $\beta_3$ with mass $1m_e$. Other extremal orbits have frequencies within experimental errorbars, but not only their angular dependence is incompatible with our observations but also their masses are too high (1.3$m_e$ for a 460T orbit associated with $\gamma_2$ sheet which is the lightest among them).
Table I: The data about SdH extremal orbits when the magnetic field is in the 110 direction.

| name | sheet | “band” index | frequency [T] | mass [el.mass] | comment |
|------|-------|---------------|---------------|----------------|---------|
| $\alpha_1$ | $\alpha$ | 87 | 86 | 0.32 | hole pocket |
| $\alpha_2$ | $\alpha$ | 87 | 96 | 0.36 | same hole pocket, from different side |
| $\zeta_1$ | $\zeta$ | 88 | 300 | 1.44 | hole-like (wing, next-most-far from body) |
| $\zeta_2$ | $\zeta$ | 88 | 335 | 1.18 | hole-like (wing, most-far from body) |
| $\zeta_3$ | $\zeta$ | 88 | 338 | 2.41 | hole-like (wing, most-close to body) |
| $\zeta_4$ | $\zeta$ | 88 | 396 | 1.48 | hole-like (goes through “stomach”) |
| $\zeta_5$ | $\zeta$ | 88 | 434 | 1.17 | hole-like (wing, next most-close to body) |
| $\zeta_6$ | $\zeta$ | 88 | 1089 | 1.45 | electron-like (follows necks that penetrate out of Brillouin zone) |
| $\zeta_7$ | $\zeta$ | 88 | 1512 | 4.72 | hole-like (goes around the wing in the long direction) |

THz STUDY

THz experiment and determination of optical conductivity

We have studied the THz properties of a 40 nm thick CRO film grown on a non-vicinal (110) NGO substrate. The residual resistivity ratio of this film is 31. At this stage, we cannot employ thicker films with even higher RRR for the THz experiments because the transmitted signal would be too small to obtain sufficient signal-to-noise; the transmission of the present sample is of order 0.001 at low temperatures and frequencies. This small transmission also sets the low-frequency limit of the spectral range that we can employ reliably for this study: at low frequencies, a small fraction of the THz radiation reaches the detector without interacting with the sample. This ‘parasitic radiation’, which is not understood well, is probably caused by diffraction at e.g. apertures of cryostat windows. Its strength increases with decreasing frequency, and in our case of a low-transmission sample we can rule out significant errors due to the parasitic radiation only for frequencies above 0.2 THz.

We performed transmission and phase measurements using a set of backward-wave-oscillators (BWOs) as tunable monochromatic and coherent THz sources. Phase measure-
Figure 13: The angular dependence of SdH frequencies as the magnetic field is turned from the 110 to 001 direction. Broad (top) and narrower frequency range (bottom) are shown. Different symbols are used for extremal orbits for the Fermi surface sheets presented in Fig. 11(a-f), for (i-vi), respectively.

Figure 14: (a) Real part $\sigma_1$ and (b) imaginary part $\sigma_2$ of optical conductivity for our sample #D5 at temperatures between 5 K and 300 K.

Fig. 14 shows conductivity data for a set of temperatures from 5 K up to 300 K. The evolution of the Drude-type roll-off in the real part $\sigma_1$ can be tracked smoothly, see Fig. 14(a): at the highest temperatures, the scattering rate is higher than our spectral range, and as a result the $\sigma_1$ spectra are basically flat and coincide with the dc conductivity. Upon cooling, the scattering rate decreases and a Drude roll-off in $\sigma_1$ develops. For temperatures lower than 40 K, the scattering rate is clearly visible in our frequency range. In the case of a simple temperature-dependent Drude response, the relaxation rate as a function of temperature can be evaluated by determining the temperature with the highest $\sigma_1$ at a given frequency. Also in this aspect we find a smooth evolution (0.6 THz $\approx$ 30 K, 0.45 THz $\approx$ 20 K, 0.35 THz $\approx$ 15 K, 0.2 THz $\approx$ 10 K) within our data. For temperatures lower than 10 K, the scattering rate has decreased to a frequency lower than what we can access with the present THz experiment. In the imaginary part $\sigma_2$, shown in Fig. 14(b), we see the corresponding behavior: With decreasing temperature, $\sigma_2$ continuously increases for all temperatures. Starting around 40 K, we can identify high and strongly temperature- and frequency-dependent dielectric constant combined with large dielectric losses.)
a maximum in the $\sigma_2$ spectra which moves toward lower frequencies upon further cooling. In the simple Drude picture, this maximum indicates the relaxation rate, and we find consistent frequencies compared to the behavior in $\sigma_1$. For temperatures below 15 K, we cannot identify the position of this maximum any more because it has moved to frequencies lower than we can reliably address in this study.

**Optical spectra at temperatures below 5 K**

The THz data for temperatures between 2 K and 5 K were performed in a separate experimental run using a home-built low-temperature optical cryostat [11]. The conductivity spectra are shown in Fig. 15. The trend observed at higher temperatures smoothly continues: the scattering rate reduces further, but since it is lower than our lowest accessible frequency, we observe this only by the further narrowing (toward zero frequency) of the roll-off in the $\sigma_1$ spectrum.

**Comparison with theory for a Fermi liquid**

We have compared our THz data to the theoretically expected curves for a Fermi liquid as recently discussed by Berthod et al. [16]. This theory describes the optical response of a local Fermi liquid. It assumes a Fermi-liquid self energy $\Sigma = (1 - 1/Z)\hbar \omega - i(\hbar^2 \omega^2 + \pi^2 k_B^2 T^2)/(Z\pi k_B T_0)$ that does not depend on the wave-vector (i.e. is local). The vertex corrections to optical conductivity are assumed to vanish. $Z$ is the quasiparticle residue and $T_0$ is the coherence scale.

Based on these assumptions, a scaling form for the optical conductivity has been derived. The optical conductivity reads

$$\frac{\sigma}{\sigma_{dc}} = \mathcal{S}\left(\frac{\hbar \omega}{2\pi k_B T}, \omega \tau_{qp}\right)$$

and is thus described in terms of the single parameter $\hbar/\tau_{qp} = 2\pi(k_B T)^2/k_B T_0$. The scaling function $\mathcal{S}$ can be analytically evaluated. The optical response of a local Fermi liquid shows on a log-log plot a characteristic non-Drude plateau around 4 THz for the 5 K curve. The dashed lines are simple Drude curves for frequency-independent scattering rate $1/\tau_0$ with NFL temperature dependence $1/\tau_0 = aT^{3/2} + 1/\tau_0$.

Figure 15: Real part $\sigma_1$ of optical conductivity for our sample #D5 at temperatures between 2 K and 5 K.

Figure 16: THz data for a few temperatures compared to theoretical predictions. The full lines follow a FL description [16], where a non-Drude plateau is predicted around 4 THz for the 5 K curve. The dashed lines are simple Drude curves for frequency-independent scattering rate $1/\tau_0$ with NFL temperature dependence $1/\tau_0 = aT^{3/2} + 1/\tau_0$.

As signs of a plateau were also found in our measurements, we attempted to fit the Berthod et al. theory to our $\sigma_1$ data. We found that the low frequency part can be fit well with $Z\Gamma_{imp} = 3\text{ meV}$ and $T_0 = 150$ K (see Fig. 4(b) of main paper). The onset of the plateau, on the other hand, appears at a too-high frequency to account for our data, see Fig. 16. The plateau that we observe is thus not a consequence of a FL frequency dependence of the self energy. Furthermore, its onset does not scale with temperature.

We also note that $T_0$ was for a doped Mott insulator described within the dynamical mean-field theory found to be about $10T_{FL}$. Insisting on Berthod et al. description, despite the obvious deviations, for CaRuO$_3$ we find that $T_0/10 >> T_{FL}$. $T_{FL}$ thus appears anomalously low also from this perspective.
In Fig. 16 we also plot the frequency dependence for the NFL regime if one assumes a bare Drude response, where the frequency-independent scattering rate $1/\tau_D \sim aT^{3/2} + 1/\tau_0$ reproduces the temperature dependence of the dc resistivity ($\tau_0 = 1.3$ ps). In the NFL temperature range, the data below 0.6 THz is described well by this simple model.

**Comparison with previous optical studies**

Our data obtained on sample #D5 roughly compares to the results obtained previously by Kamal *et al.* using THz time-domain spectroscopy [15]. In Fig. 17 we show our data at temperatures 10 K and 15 K, and data by Kamal *et al.* for 10 K and 16 K. The main differences are that the low-frequency conductivity of our sample is much higher due to the improved sample growth, and that the order of the curves is exchanged: for our sample and above 0.3 THz, $\sigma_1(T=15 \text{ K})$ exceeds $\sigma_1(T=10 \text{ K})$, because here the sample is already in the relaxation regime, with excitation frequency larger than the optical scattering rate. Our data are also consistent with the previous infrared results by Lee *et al.* [14], as shown with the dashed curve for 10 K in Fig. 17. The slight mismatch in absolute conductivity we again attribute to the different sample quality.

* present address: Institut für Festkörperphysik, Technische Universität Wien

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