Dirty van Hove singularity quasiparticles studied by magnetotransport

Y. Xu, F. Herman, V. Granata, D. Destraz, L. Das, J. Vonka, S. Gerber, J. Spring, M. Gibert, A. Schilling, X. F. Zhang, S. Y. Li, R. Fittipaldi, M. H. Fischer, A. Vecchione, and J. Chang

1 Physik-Institut, Universität Zürich, Winterthurerstrasse 190, CH-8057 Zürich, Switzerland
2 Institute for Theoretical Physics, ETH Zürich, Hönggerberg, CH-8093 Zürich, Switzerland
3 CNR-SPIN, I-84084 Fisciano, Salerno, Italy
4 Dipartimento di Fisica “E.R. Caianiello”, Università di Salerno, I-84084 Fisciano, Salerno, Italy
5 Laboratory for Micro and Nanotechnology, Paul Scherrer Institut, Forschungsstrasse 111, CH-5232 Villigen PSI, Switzerland
6 Laboratory for Neutron and Muon Instrumentation, Paul Scherrer Institut, Forschungsstrasse 111, CH-5232 Villigen PSI, Switzerland
7 State Key Laboratory of Surface Physics, Department of Physics, and Laboratory of Advanced Materials, Fudan University, Shanghai 200438, China

(Dated: May 5, 2020)

We present a magnetotransport study of the metamagnetic system Ca$_{1.8}$Sr$_{0.2}$RuO$_4$. The metamagnetic transition is reached by tuning a van Hove singularity to the Fermi level at a critical magnetic field $H_m$. In Ca$_{1.8}$Sr$_{0.2}$RuO$_4$, we report across $H_m$ a strong decoupling of inelastic electron scattering, measured by resistivity, and electronic mass, inferred from density-of-state probes. As a result, we observe – in contrast to conventional correlated Fermi liquids – a strong variation of the Kadowaki-Woods ratio. Fermi-liquid and non-Fermi-liquid properties associated with van Hove singularities are discussed in terms of disorder and dimensionality. We argue that Ca$_{1.8}$Sr$_{0.2}$RuO$_4$ belongs to the dirty/disordered two-dimensional limit. Disorder effects play a central role in the understanding of the (non-) Fermi liquid onset. We therefore propose that in quasi-two-dimensional systems disorder has a significant impact on the electronic properties associated with van Hove singularities in the vicinity of the Fermi level.

In two-dimensional systems, saddle points in the electronic band structure generate a diverging density of states (DOS), a so-called van Hove singularity (VHS) [1]. A divergent DOS at the Fermi level renders a system susceptible to instabilities like charge/spin density wave order or unconventional superconductivity. Gate-tuned superconductivity in magic angle bilayer graphene has, for example, been proposed to be connected to VHS physics [2, 3]. A VHS is also found in high-temperature cuprate superconductors, and recently it has been associated with the onset of the mysterious pseudogap phase [4, 5]. It is debated whether the surrounding non-Fermi liquid behavior is originating from a quantum criticality or a VHS scenario [6]. In the ruthenates, the VHS governs many interesting electronic properties. For example, the VHS can be tuned to the Fermi level by application of a magnetic field [7–10] or uniaxial [11], biaxial [12] and chemical pressure [13, 14]. In Sr$_3$Ru$_2$O$_7$, a magnetic field of 8 T along the c axis triggers a spin density phase around which non-Fermi liquid transport behavior is observed [7–10]. Similar non-Fermi liquid behavior is found in (Sr, Ba)$_3$Ru$_2$O$_7$ upon application of pressure or strain [11, 12]. Finally, metamagnetic transitions in systems such as Sr$_3$Ru$_2$O$_7$, CeRu$_2$Si$_2$ and Ca$_{1.8}$Sr$_{0.2}$RuO$_4$ have been assigned to DOS anomalies near the Fermi level [15–17].

Despite the expected connection between an ideal VHS and unconventional electronic properties observed in a wide range of materials, the effect of disorder and dimensionality has received little attention. Quasiparticles in layered materials are neither constrained perfectly in two dimensions nor are their lifetime infinite. Both effects, dimensionality and disorder, broaden the DOS anomaly [18] and hence, potentially change the ideal VHS physics substantially.

Here, we address electronic transport properties of a quasi-two-dimensional disordered system for which the VHS is aligned with the Fermi level by an external magnetic field. Magnetotransport anomalies in Ca$_{1.8}$Sr$_{0.2}$RuO$_4$ are directly linked to the metamagnetic transition. Although Fermi liquid properties are preserved across the metamagnetic transition, the electronic scattering processes are highly unusual. In particular, we report a decoupling of the inelastic electron scattering from the electronic mass. This results in a five-fold variation of the Kadowaki-Woods ratio – an empirical ratio linking the quasiparticle lifetime and mass – across the metamagnetic transition. Our observations are presented in a broader context of Fermi liquid/non-Fermi liquid properties across metamagnetic transitions in strongly correlated electron systems with DOS anomalies. Specifically, the role of dimensionality and disorder in the context of VHS physics is discussed along with possible scenarios for the strong variation of the Kadowaki-Woods ratio.

Single crystals of Ca$_{1.8}$Sr$_{0.2}$RuO$_4$ were grown by the flux-feeding floating-zone technique [19, 20]. Our experimental results were reproduced on several crystals that were cut and polished into a rectangular shape, with the largest natural plane being the ab plane. Magnetic fields...
Figure 1. (a) The resistivity $\rho$ of Ca$_{1.8}$Sr$_{0.2}$RuO$_4$ as a function of temperature $T$ and magnetic field $H$. (b) The temperature dependence of $\rho$ for selected fields. (c) The exponent $\alpha$ in the $H$-$T$ space with the resistivity of Ca$_{1.8}$Sr$_{0.2}$RuO$_4$ fitted to $\rho = \rho_0 + C T^\alpha$. (d) Magnetoresistance $\left(\frac{\rho(H) - \rho(0 \text{ T})}{\rho(0 \text{ T})}\right)$ isotherms for selected temperatures. Shaded area indicates the maximum around $H_m$.

$\mu_0 H$ ($\mu_0$ being the vacuum permeability) up to 9 T were applied along the $c$ axis and silver paste electrical contacts were made on the $ab$ plane. Resistivity measurements were performed in a physical property measurement system (PPMS, Quantum Design) with a Helium-3 option.

The temperature dependence of the resistivity $\rho$ measured on Ca$_{1.8}$Sr$_{0.2}$RuO$_4$ at various magnetic fields, is shown in Figs. 1(a) and 1(b). A region of enhanced resistivity fans out around the metamagnetic transition at $\mu_0 H_m \sim 5.5$ T (see Supplemental Material [21]) in the $\rho(H, T)$ plot [Fig. 1(a)]. Insights into the scattering mechanisms are commonly gained by analyzing $\rho = \rho_0 + C T^\alpha$ with $C$ being a constant. The residual resistivity $\rho_0$ is temperature independent, but allowed to vary with field. Figure 1(c) shows the $H$-$T$ plot of the exponent $\alpha$ for Ca$_{1.8}$Sr$_{0.2}$RuO$_4$ obtained from this procedure. The low-temperature yellow region demonstrates that Fermi liquid behavior ($\alpha \sim 2$) is found at all fields across $H_m$. The Fermi liquid cutoff temperature $T_{FL}$ remains constant below $H_m$ and increases above the transition. Magnetoresistance (MR) isotherms, defined by $\left(\frac{\rho(H) - \rho(0 \text{ T})}{\rho(0 \text{ T})}\right)$, all exhibit a maximum around $H_m$ that broadens with increasing $T$ [Fig. 1(d)].

Since Fermi liquid behavior is observed at low temperature for all fields, we fix $\alpha = 2$ and fit with $\rho = \rho_0 + A T^2$ [see Fig. 2(a)], where $A$ is the inelastic electron-electron scattering coefficient. The resulting $\rho_0$ and $A$ are plotted versus magnetic field in Figs. 2(c) and 2(d), respectively. While the field dependence of $\rho_0$ closely tracks the MR isotherms, $A$ decreases by a factor of three across $H_m$. Two key observations are revealed by our magnetotransport experiment: Across the metamagnetic transition, (1) the Fermi liquid state persists at low temperatures and (2) the inelastic scattering coefficient $A$ undergoes a dramatic drop. In addition to the Fermi liquid cutoff temperature $T_{FL}$ indicated by arrows in Fig. 2(a), we identify another temperature scale $T_{SM}$, above which a strange metal behavior $\rho \sim T$ is observed for all fields, as shown in Fig. 2(b).

Although the metamagnetic transition has been well established in Ca$_{1.8}$Sr$_{0.2}$RuO$_4$, its impact on magnetotransport has not been addressed by previous studies [17, 23, 24]. Our results demonstrate a direct connection between the metamagnetic transition and transport properties. As such, Ca$_{1.8}$Sr$_{0.2}$RuO$_4$ can now be directly compared to other metamagnetic systems. As shown in Table 1, Ca$_{1.8}$Sr$_{0.2}$RuO$_4$, CeRu$_2$Si$_2$ and Sr$_3$Ru$_2$O$_7$ all display a peak in the residual resistivity $\rho_0$ and the Sommerfeld coefficient $\gamma$ across the metamagnetic transition. Both $\rho_0$ and $\gamma$ are proportional to the DOS at the Fermi
level. Therefore, these compounds share a field-induced traversal of a DOS peak through the Fermi level. The DOS peak in Ca$_{3}$Sr$_{0.2}$RuO$_{4}$ is likely associated with a VHS [25–27]. Interestingly, the inelastic electron-electron scattering process varies dramatically across these compounds. Non-Fermi liquid behavior is reported down to the lowest measured temperatures in Sr$_{3}$Ru$_{2}$O$_{7}$ at $H_m$. As in CeRu$_2$Si$_2$ [16], we report Fermi liquid behavior across $H_m$ in Ca$_{3}$Sr$_{0.2}$RuO$_{4}$. However, in CeRu$_2$Si$_2$ the scattering coefficient $A$ peaks together with the Sommerfeld coefficient, whereas in Ca$_{3}$Sr$_{0.2}$RuO$_{4}$, $A$ undergoes a step-like drop across $H_m$. In the following, we discuss the Fermi liquid versus non-Fermi liquid aspect before turning to the unusual behavior of the Kadowaki-Woods ratio in Ca$_{3}$Sr$_{0.2}$RuO$_{4}$.

In strongly correlated electron systems, $\rho$ is generally dominated by impurity and electron-electron scattering at low temperatures. States contributing to the transport properties lie within the scattering phase space defined by $f_T(\varepsilon)[1 - f_T(\varepsilon)]$, where $f_T(\varepsilon)$ is the Fermi-Dirac distribution at temperature $T$. The associated energy scale, full-width half maximum $W_{\text{SPS}} \sim 3.5k_B T$, can be compared with that of the DOS peak $W_{\text{DOS}}$. In the low-temperature limit $T \lesssim T_{\text{FL}} \sim \kappa W_{\text{DOS}} / (3.5k_B)$ with $\kappa \ll 1$, Fermi liquid behavior ($\rho \sim T^2$) is anticipated, since the DOS is almost flat within the scattering phase space. By contrast, for $T \gtrsim T_{\text{SM}} \sim \beta W_{\text{DOS}} / (3.5k_B)$ with $\beta \sim 1$, strange metal behavior, such as $\rho \sim T$, $\sim T^{3/2}$, or $\sim T^2 \log T$, is expected, once the DOS peak is fully covered by the scattering phase space [38–43]. These two limits, together with the intermediate region $T_{\text{FL}} < T < T_{\text{SM}}$ are schematically shown in Figs. 2(f)-(h). Whereas the scattering phase space $W_{\text{SPS}}$ is set by temperature, $W_{\text{DOS}}$ is controlled by dimensionality and disorder. Utilizing $\rho_{ab}/\rho_{c}$ and $\rho_{c}$ as effective gauges for the dimensionality and disorder, respectively, we place different systems with large DOS at the Fermi level in a dimensionality-disorder-temperature diagram (Fig. 3). For clean two-dimensional systems, such as Sr$_2$RuO$_4$ and Sr$_3$Ru$_2$O$_7$, the sharp DOS peak (small $W_{\text{DOS}}$) makes it difficult to experimentally access the temperature scales $T_{\text{FL}}$ and $T_{\text{SM}}$. In both systems, when the Fermi level and VHS are tuned to match, strange metal behavior is observed down to lowest temperatures before being cut-off by instabilities (superconductivity and spin density wave order) [7, 11]. In clean three-dimensional systems,
a larger $T_{\text{FL}}$ is expected, and indeed Fermi liquid behavior was found across $H_m$ in CeRu$_2$Si$_2$ [16]. To our knowledge, in the two-dimensional dirty limit, Fermi liquid properties have not been explored/discussed in the context of a van Hove singularity. Notably, this limit is represented by Ca$_{1.8}$Sr$_{0.2}$RuO$_4$, where $T_{\text{FL}} \sim 2$ K [Figs. 1(c) and 2(a)] and $T_{\text{SM}} \sim 20$ K [Fig. 2(b)] are identified. Angle-resolved photoemission (ARPES) suggests that $W_{\text{DOS}} \sim 20$ meV [44] and hence, we extract reasonable values for $\kappa \sim 0.03$ and $\beta \sim 0.3$. These values of $\kappa$ and $\beta$ are weakly material dependent as they stem from the ratio of the widths of the DOS and scattering phase space. Hence this information can be applied to, for example, the pseudogap problem [4–6] found in La-based cuprates. Assuming $\beta \approx 0.3$ for La$_{1.36}$Nd$_{0.4}$Sr$_{0.24}$CuO$_{4}$, where $W_{\text{DOS}} \sim 15$ meV [45], yields $T_{\text{FL}} \sim 15$ K. However, since $\rho \sim T$ ($C/T \sim \log(1/T)$) is observed down to $1(0.5)$ K [46–49], we conclude that quantum criticality must be taken into account. Our results thus have direct implications for the interpretation of the strange metal properties in cuprates.

The evolution of the Kadowaki-Woods ratio (KWR) $A/\gamma^2$ across the metamagnetic transition in Ca$_{1.8}$Sr$_{0.2}$RuO$_4$ is rather unusual. In the simplest case, the ratio $A/\gamma^2$ is invariant to electron correlations [60–63]. This implies that both $A$ and $\gamma^2$ are expected to increase with enhanced electron interaction. In practice, even in systems where $A/\gamma^2$ is not constant, $A$ and $\gamma^2$ still correlate positively, as seen in Sr$_3$Ru$_2$O$_7$ across its metamagnetic transition and YbRh$_2$(Si$_{0.95}$Ge$_{0.05}$)$_2$ across its quantum critical point [38, 64]. This is in stark contrast to Ca$_{1.8}$Sr$_{0.2}$RuO$_4$ where $A$ and $\gamma^2$ anticorrelate on approaching the metamagnetic transition on the low-field side. A factor-of-five variation [Fig. 2(e)] of the KWR is the consequence of this decoupling of $A$ and $\gamma^2$. We stress that the bare band structure is not expected to change significantly by the application of magnetic field and hence is not the source [62, 63] for the strong variation of the KWR. Worth noticing is also that elastic scattering – probed by $\rho_0$ – is linked to the DOS at the Fermi energy. The field evolution of the KWR is therefore also reflected in the magnetic-field dependencies of $A$ and $\rho_0$ [Figs. 2(c) and 2(d)]. A similar decoupling of $A$ and $\rho_0$ has also been reported in the multiband heavy fermion system CeTiGe [37].

While Ca$_{1.8}$Sr$_{0.2}$RuO$_4$ is a multiband system, we resort to Boltzmann transport theory for a single-band situation to gain qualitative insight into the KWR (see

| Compound          | Tuning         | Critical value | $\rho_0$ peak | $\gamma$ peak | $A$ peak | FL | Reference  |
|-------------------|----------------|----------------|---------------|---------------|----------|----|------------|
| Sr$_2$RuO$_4$     | Uniaxial strain| $\epsilon = 0.5\%$ | Yes           | Yes           | Yes      | No | [11, 28, 29]|
| Sr$_3$Ru$_2$O$_7$ | Magnetic field | $H = 7.8$ T     | Yes           | Yes           | Yes      | No | [7, 30–33] |
| Ca$_{1.8}$Sr$_{0.2}$RuO$_4$ | Magnetic field | $H = 5.5$ T     | Yes           | Yes           | No       | Yes| This work, 22 |
| CeRu$_2$Si$_2$    | Magnetic field | $H = 8.0$ T     | Yes           | Yes           | Yes      | Yes| [16, 34–36]|
| CeTiGe            | Magnetic field | $H = 12$ T      | Yes           | -             | No       | Yes| [37]       |

Table I. Fermi liquid behaviors as the DOS peak and the Fermi level are tuned to match. For each compound the tuning parameter (uniaxial strain or magnetic field) and the associated critical values are indicated. The behavior (peak or no peak) across the critical tuning of residual resistivity $\rho_0$, Sommerfeld coefficient $\gamma$ and electron scattering coefficient $A$ (see text) is indicated. Finally, the observed resistivity behaviour (Fermi liquid or non-Fermi liquid) at the critical tuning and lowest measured temperature is given.
Supplemental Material [21]). Within this framework, inelastic electron scattering is more sensitive than elastic scattering, to the detailed relation between the DOS profile and the scattering phase space. This is most significant in systems with a DOS peak around the Fermi level, as is the case here. Furthermore, momentum-dependent on-site Hubbard-$U$-type coupling produces a non-local (momentum-dependent) self-energy [65–72], which provides another source for the unusual behavior of the KWR [63]. Most likely, single-band models are too simplistic in the description of the systems discussed. It is therefore not inconceivable that the here observed variation of the KWR has its origin in the complexity brought by multiband physics. The orbital selective Mott insulating scenario for Ca$_{1.8}$Sr$_{0.2}$RuO$_4$ [73, 74] is not resonating with experimental observations. Instead, a most recent ARPES experiment suggests an orbital differentiated self-energy [44]. Yet decoupling of quasiparticle mass and lifetime going beyond a single band picture requires more exotic scenarios such as a breakup of spin and charge degrees of freedom [64]. Since we observe persistence of Fermi liquid behavior across the metamagnetic transition, however, a quasiparticle decomposition appears unlikely.

In summary, we have addressed the electronic transport properties associated with a van Hove singularity in the quasi-two-dimensional dirty limit. In Ca$_{1.8}$Sr$_{0.2}$RuO$_4$, the metamagnetic transition is interpreted as evidence for a van Hove singularity being tuned through the Fermi level by application of magnetic field. In contrast to Sr$_3$Ru$_2$O$_7$, no Fermi-liquid breakdown is found across the metamagnetic transition in Ca$_{1.8}$Sr$_{0.2}$RuO$_4$. Instead, a breakdown of the positive correlation between quasiparticle life time and mass is observed. Our study of the quasi-two-dimensional dirty limit provides (in combination with existing literature) a complete picture of the low-temperature Fermi liquid and non-Fermi liquid properties in the context of van Hove singularities and disorder.

We thank Louis Taillefer and Benot Fauqu for helpful discussions. Y. X., F. H., and J. C. were financially supported by the Swiss National Science Foundation (SNSF) (Grant No. PP00P2_179097 and 184739). L. D. was partially funded through Excellence Scholarship by the Swiss Government.

* johan.chang@physik.uzh.ch

[1] G. E. Volovik, Low Temp. Phys. 43, 47 (2017).
[2] Y. Cao, V. Fatemi, S. Fang, K. Watanabe, T. Taniguchi, E. Kaxiras, and P. Jarillo-Herrero, Nature 556, 43 (2018).
[3] N. F. Q. Yuan, H. Iseobe, and L. Fu, Nat. Comm. 10, 1 (2019).
[4] W. Wu, M. S. Scheurer, S. Chatterjee, S. Sachdev, A. Georges, and M. Ferrero, Phys. Rev. X 8, 021048 (2018).
[5] N. Doiron-Leyraud, O. Cyr-Choinire, S. Badoux, A. Ataei, C. Collignon, A.ourgout, S. Dufour-Beaussier, F. F. Tafti, F. Laliberté, M.-E. Boulanger, M. Matusiak, D. Graf, M. Kim, J.-S. Zhou, N. Momono, T. Kurosawa, H. Takagi, and L. Taillefer, Nat. Comm. 8, 1 (2017).
[6] J. M. Buhmann, M. Ossadnik, T. M. Rice, and M. Sigrist, Phys. Rev. B 87, 035129 (2013).
[7] S. A. Grigera, R. S. Perry, A. J. Schofield, M. Chiao, S. R. Julian, G. G. Lonzarich, S. I. Ikeda, Y. Maeno, A. J. Millis, and A. P. Mackenzie, Science 294, 329 (2001).
[8] S. A. Grigera, P. Gegenwart, R. A. Borzi, F. Weickert, A. J. Schofield, R. S. Perry, T. Tayama, T. Sakakibara, Y. Maeno, A. G. Green, and A. P. Mackenzie, Science 306, 1154 (2004).
[9] R. A. Borzi, S. A. Grigera, J. Farrell, R. S. Perry, S. J. S. Lister, S. L. Lee, D. A. Tennant, Y. Maeno, and A. P. Mackenzie, Science 315, 214 (2007).
[10] C. Lester, S. Ramos, R. S. Perry, T. P. Croft, R. I. Bewley, T. Guidi, P. Mannel, D. D. Khalyavin, E. M. Forgan, and S. M. Hayden, Nat. Mater. 14, 373 (2015).
[11] M. E. Barber, A. S. Gibbs, Y. Maeno, A. P. Mackenzie, and C. W. Hicks, Phys. Rev. Lett. 120, 076602 (2018).
[12] B. Burganov, C. Adamo, A. Mulder, M. Uchida, P. D. C. King, J. W. Harter, D. E. Shai, A. S. Gibbs, A. P. Mackenzie, R. Uecker, M. Brueckm, M. R. Beasley, C. J. Pennie, D. G. Schlom, and K. M. Shen, Phys. Rev. Lett. 116, 197003 (2016).
[13] N. Kikugawa, C. Bergemann, A. P. Mackenzie, and Y. Maeno, Phys. Rev. B 70, 134520 (2004).
[14] K. M. Shen, N. Kikugawa, C. Bergemann, L. Balicas, F. Baumberger, W. Meevasana, N. J. C. Ingle, Y. Maeno, Z.-X. Shen, and A. P. Mackenzie, Phys. Rev. Lett. 99, 187001 (2007).
[15] A. Tamai, M. P. Allan, J. F. Mercure, W. Meevasana, R. Dunkel, D. H. Lu, R. S. Perry, A. P. Mackenzie, D. J. Singh, Z.-X. Shen, and F. Baumberger, Phys. Rev. Lett. 101, 026407 (2008).
[16] R. Daou, C. Bergemann, and S. R. Julian, Phys. Rev. Lett. 96, 026401 (2006).
[17] S. Nakatsuji, D. Hall, L. Balicas, Z. Fisk, K. Sugahara, M. Yoshioka, and Y. Maeno, Phys. Rev. Lett. 93, 137202 (2003).
[18] M. Horio, K. Hauser, Y. Sassa, Z. Mingazheva, D. Sutner, K. Kramer, A. Cook, E. Nocerino, O. K. Forslund, O. Tjernberg, M. Kobayashi, A. Chikina, N. B. M. Schröter, J. A. Krieg, T. Schmitt, V. N. Strocov, S. Pyon, T. Takayama, H. Takagi, O. J. Lipscombe, S. M. Hayden, M. Ishikado, H. Eisaki, T. Neupert, M. Månsson, C. E. Matt, and J. Chang, Phys. Rev. Lett. 121, 077004 (2018).
[19] S. Nakatsuji and Y. Maeno, J. Solid State Chem. 156, 26 (2001).
[20] H. Fukazawa, S. Nakatsuji, and Y. Maeno, Physica B Condens. Matter 281, 613 (2000).
[21] See Supplemental Material for the magnetization measurements and the Boltzmann transport theory for understanding the novel behavior of the Kadowaki-Woods ratio, which includes Refs. [17, 65, 75].
[22] J. Baier, T. Zabel, M. Kriener, P. Steffens, O. Schumann, O. Friedt, A. Freimuth, A. Revcolevschi, S. Nakatsuji, Y. Maeno, T. Lorenz, and M. Braden, Physica B Condens. Matter 281, 613 (2000).
[74] A. Koga, N. Kawakami, T. M. Rice, and M. Sigrist, Phys. Rev. Lett. 92, 216402 (2004).

[75] M. Kriener, P. Steffens, J. Baier, O. Schumann, T. Zabel, T. Lorenz, O. Friedt, R. Müller, A. Gukasov, P. G. Radaelli, P. Reutler, A. Revcolevschi, S. Nakatsuji, Y. Maeno, and M. Braden, Phys. Rev. Lett. 95, 267403 (2005).