Thermal conductivity in the vicinity of the quantum critical endpoint in Sr$_3$Ru$_2$O$_7$

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(Dated: April 18, 2018)

Thermal conductivity of Sr$_3$Ru$_2$O$_7$ was measured down to 40 mK and at magnetic fields through the quantum critical endpoint at $H_c = 7.85$ T. A peak in the electrical resistivity as a function of field was mimicked by the thermal resistivity. In the limit as $T \to 0$ K, we find that the Wiedemann-Franz law is satisfied to within 5% at all fields, implying that there is no breakdown of the electron despite the destruction of the Fermi liquid state at quantum criticality. A significant change in disorder (from $\rho_0(H=0T) = 2.1 \mu\Omega$ cm to 0.5 $\mu\Omega$ cm) does not influence our conclusions. At finite temperatures, the temperature dependence of the Lorenz number is consistent with a Fermi liquid, fluctuations causing the non-Fermi liquid behavior as one would expect at a metamagnetic quantum critical endpoint.

PACS numbers:

While classical phase transitions are theoretically well understood, quantum phase transitions are in defiance of theoretical understanding. At a quantum critical point (QCP), the Fermi liquid ground state is destroyed by the diverging quantum fluctuations associated with a particular phase transition. Given that the physics up to very high temperatures can be dominated by the presence of a QCP, it is essential to try and understand the nature of the fluctuations and excitations which exist at a QCP. Part of the problem in understanding quantum phase transitions is that, given nearly a hundred different systems which show non-Fermi liquid behavior presumably due to proximity to a QCP, there is very little commonality between various observables, such as resistivity, susceptibility, and specific heat. What is desperately needed are very fundamental measurements of physical properties in the vicinity of a QCP.

The Wiedemann-Franz law (WFL), which states that thermal ($\kappa$) and charge ($\sigma$) conductivities are simply related through the expression $\kappa/\sigma T = L_0$, where $L_0$ is the Sommerfeld value of the Lorenz number ($2.44 \times 10^{-8} \text{W} \Omega / \text{K}^2$), is precisely such a fundamental probe of strongly correlated physics. At $T = 0$ the law is a consequence of the fact that all fermionic excitations carry charge $e$, while all of the possible bosonic excitations have zero charge. Should a violation be expected at a QCP? Experiments on established quantum critical systems, such as specific heat and resistivity on YbRh$_2$Si$_2$, have been interpreted as observing the breakup of the electron at a QCP, which would naively imply a violation. In addition, theories of quantum criticality are also suggesting that it may be possible to violate the WFL at a QCP.

Experimentally, a verification of the WFL was observed in CeNi$_2$Ge$_2$. While this study was a singular measurement at zero magnetic field and ambient pressure, the belief is that this point in phase space lies in close proximity to an antiferromagnetic QCP. Recently, the WFL was also confirmed in the field tuned quantum critical system CeCoIn$_5$. Another possibly relevant system in which the WFL has been measured are the high $T_c$ cuprates. In this case a violation has been observed in the field-induced normal state, which may be related to an underlying QCP in the phase diagram.

In this letter, we have chosen to study the WFL in Sr$_3$Ru$_2$O$_7$, a bilayer perovskite material. The well-known single layer compound Sr$_2$RuO$_4$ is believed to be a spin triplet superconductor, while the infinite layer compound SrRuO$_3$ is an itinerant electron ferromagnet. When the magnetic field is applied in the RuO$_2$ planes of Sr$_3$Ru$_2$O$_7$, a first order metamagnetic transition is observed. The critical endpoint of this line of first order metamagnetic transitions is systematically driven to zero temperature at $H = 7.85$ T by rotating the magnetic field out of the plane. Thus, the term quantum critical endpoint (QCEP) is used when discussing Sr$_3$Ru$_2$O$_7$. The use of magnetic field as a tuning parameter in this stoichiometric system allows for a very sensitive test of the Wiedemann-Franz law to be made on a clean system. We present thermal and charge conductivity data on Sr$_3$Ru$_2$O$_7$, which demonstrate that the integrity of the electron does survive in the vicinity of a QCEP as the WFL is satisfied at all fields.

A second aspect of this study is that finite temperature thermal and charge conductivity data allow us to comment on the nature of the fluctuations present in this system. While the above circumstantial evidence would suggest that ferromagnetic fluctuations are the most relevant magnetic excitations here, neutron scatter-
The heat current was applied in the \( c \)-axis including the critical field. The intercept represents the fermionic contribution to the thermal conductivity resulting from a constant density of states in the limit that scattering is dominated by elastic scattering. From the resistivity data (plotted as \( L_0/\rho \) for comparison with the WFL) we can see in figure 2(a) that we have clearly reached that limit below 1 K. As there is almost no temperature dependence we can reliably extrapolate our results to \( T = 0 \), where we find that the WFL (which states that \( \kappa/\sigma T = L_0 \)) is satisfied to within 5% at all temperatures. These quantities extrapolate to the same value at \( T = 0 \) if the Wiedemann-Franz law is to be satisfied. Solid lines are fits to \( \rho = \rho_0 + AT^2 \). The dashed line for \( H_c \) is a linear fit below 0.8 K. (b) Comparing two samples with different amounts of disorder. Sample #2 has been rotated by 20° with respect to the \( c \)-axis which mainly changes the value of the critical field.

Thermal conductivity was measured using a two thermometer one heater setup described elsewhere. The absolute accuracy is limited by the uncertainty in the geometric factor of the sample (\( \sim 10\% \)), but the relative changes between different fields is limited by the accuracy of the thermometer calibration which is \( \sim 1\% \) for the temperature sweeps, and due to the magnetoresistance of the thermometers can drift to as much as 5% for the field sweeps. The \( \text{Sr}_3\text{Ru}_2\text{O}_7 \) single crystals studied were grown in a floating zone furnace. With the exception of the data in figure 2(b) (which has a residual resistivity of 0.5 \( \mu\Omega \) cm and was aligned \( \sim 20° \) off the \( c \)-axis) data is presented for a sample with a residual resistivity of 2.1 \( \mu\Omega \) cm, and was aligned with the field parallel to within 5° of the \( c \)-axis. The transport was in the \( ab \)-plane.

Figure 1 presents the raw thermal conductivity data at, above, and below the critical field of \( H_c = 7.85 \) T. In zero field, the thermal conductivity has a peak at approximately 5 K which vanishes as one reaches the critical field. At still higher fields, the peak begins to return, albeit much more slowly. Comparison with the limited specific heat data on single crystals shows a qualitatively similar behavior. \( C/T \) has a peak at zero field, which is heavily suppressed close to the critical field. At this point the specific heat begins to diverge, indicative of non-Fermi liquid behavior. Theoretical work is needed to evaluate how the changing electronic structure affects both the number of carriers and the scattering rate, both of which enter into the thermal conductivity.

We now turn our attention to the low temperature end of the data. By plotting \( \kappa/ T \) vs \( T \) the intercept represents the fermionic contribution to the thermal conductivity. The specific heat data on single crystals shows a qualitatively similar behavior.

\[ \kappa (\text{mW/K/cm}) = \begin{cases} 20 & \text{at } 0 \text{T}, \\ 10 & \text{at } 5 \text{T}, \\ 5 & \text{at } 13 \text{T}, \\ 1 & \text{at } 15 \text{T} \end{cases} \]

\[ T (\text{K}) = \begin{cases} 0 & \text{at } 0 \text{T}, \\ 5 & \text{at } 5 \text{T}, \\ 10 & \text{at } 13 \text{T}, \\ 15 & \text{at } 15 \text{T} \end{cases} \]

\[ \sigma T (\text{mW/K/cm}) = \begin{cases} 20 & \text{at } 0 \text{T}, \\ 10 & \text{at } 5 \text{T}, \\ 5 & \text{at } 13 \text{T}, \\ 1 & \text{at } 15 \text{T} \end{cases} \]
fields. The phonon conductivity does not have a zero temperature intercept in $\kappa/T$ and hence does not enter into the discussion at this time.

The lack of temperature dependence at all fields also allows us to comment on the elastic scattering from finite temperature field sweeps. As a result, in the inset to figure 3 we plot $\kappa/T$ and $L_0/\rho$ as a function of field at low temperatures. The lowest temperature curves are clearly a good approximation to the $T=0$ value for the residual elastic scattering. The resistivity shows a pronounced peak (dip in $L_0/\rho$) at the critical field, the origin of which is unknown. Clearly, however, the thermal conductivity data tracks this dip. To see how precisely the thermal conductivity tracks the charge conductivity we plot the Lorenz ratio $L/L_0=\kappa/\sigma T L_0$ in the main panel of figure 3. The fact that the WFL is satisfied is again demonstrated by the fact that this quantity equals 1 at all fields. The slow drift in the value of the Lorenz number is due to calibrating the magnetoresistance of the thermometers used to measure the temperature gradient in the sample. The sharper bump at the critical field likely results from the new phase which emerges to "protect" the QCEP. The slight enhancement observed in $L$ at finite temperature in this protected phase may result from either a reduction in inelastic scattering or a shift to large angle scattering upon entering this new phase.

As disorder can dramatically modify the behavior at a QCP, we investigate its effects by also measuring a higher purity crystal shown in figure 2(b). This is particularly true of the phase which protects the QCEP. In zero field one can observe the factor of 4 increase in purity relative to the sample presented in figure 2(a), although at the critical field the change in elastic scattering is not nearly as great. The field is also applied slightly off the c-axis which has the added advantage that the resistive anomaly associated with the phase which protects the QCEP is enhanced. This anomaly is mimicked in the thermal data and at $T=0$ we find that this phase also obeys the WFL to within 5%.

The presence of exotic fermionic excitations which do not carry charge $\pm e$ (such as charge 0 spinons) or charged bosonic excitations would result in a violation of the WFL. The verification of the WFL in the $T \to 0$ K limit proves that such excitations do not exist here.

For charge $\pm e$ quasiparticles, the WFL is valid only when heat and charge transport are affected equally, as is the case for elastic processes dominant in the $T \to 0$ K limit. As temperature is increased, inelastic scattering dominates the elastic scattering of quasiparticles. Thus, a finite temperature study of the Lorenz number may also shed light on to the nature of the excitations relevant to the non-Fermi liquid behavior observed at the critical field. The strength of such a study was exemplified in CeRhIn$_5$ where thermal measurements found quantitative agreement with the antiferromagnetic spin fluctuation spectrum measured by inelastic neutron scattering.

Figure 4(a) shows the temperature dependence at a few fields including the critical field. A phonon conductivity term of $\kappa_{ph} = 0.03(mW/K^3 cm) T^2$ independent of field for phonons being scattered by electrons was subtracted from the thermal conductivity data, to give purely the
electronic contribution to $L$. This choice of phonon subtraction is the minimum necessary to place $L_{el}$ in the physically allowable parameter range $0 \leq L_{el} \leq L_0$, but the conclusions drawn from these data are insensitive to the amount of phonon conductivity assumed. Interestingly, the effect of small angle inelastic scattering becomes stronger as one approaches the critical field as signified by the increasingly rapid suppression of $L$ at low temperatures. This is consistent with the divergence of the quasiparticle-quasiparticle scattering at $H_c$, as deduced from the $T^2$ coefficient of resistivity.\[1,17\] As a result, the minimum in $L(T)$ naturally also moves to lower temperature. The puzzling observation is that the magnitude of the dip in $L(T)$ is gradually suppressed as the critical field is approached. This is partially a result of the increase in elastic scattering about the critical field. However, the effect is still present if the elastic scattering is removed $L_{inel} = (\kappa/T - \kappa/T(T=0)) (\rho - \rho(T=0)$ (not shown). In CeRu$_2$Si$_2$, a heavy fermion metamagnetic system, the $T \rightarrow 0$ limit was not explored, but at finite temperature the behavior in $L(T)$ was similar to that observed here.\[20\]

Theoretically, heat and charge transport properties have been calculated for a nearly ferromagnetic metal.\[21\] It was found that as the Stoner parameter $\alpha$ is enhanced the deviation of the Lorenz number from the Sommerfeld value $L_0$ is reduced. Thus, in this picture our data would indicate that the Stoner parameter increases as the critical field is approached. This is precisely what happens in the standard picture for itinerant electron metamagnetism.\[22\] By applying a magnetic field, the spin-up and spin-down bands are pushed in opposite directions. When a peak in the density of states exists near the Fermi level, then a metamagnetic transition will occur when the peak in one of the spin-split bands is pushed through the chemical potential. The Stoner parameter $\alpha$, which is equal to the density of states at the chemical potential times the interaction term $U$, is thus maximal at the metamagnetic transition, which is in agreement with our field dependence of $L(T)$. Furthermore, the thermal resistance due to spin fluctuations $W_{SF} = 1/\kappa_{el} - \rho_0/L_0T$ shown in figure 4(b) strongly resembles that of the calculations done by Ueda and Moriya.\[21\] Further emphasizing the point that the behavior in $L(T)$ is not solely due to a change in the elastic scattering.

This simple model of metamagnetism remarkably reproduces many aspects of the observations seen in Sr$_3$Ru$_2$O$_7$ in addition to the behavior in the Lorenz number. It is natural to anticipate a peak in the density of states from a Van-Hove singularity in this twodimensional system. Fine tuning of $U$ and the initial form of the density of states in a Stoner model as described can provide a fair explanation of the magnetization and specific heat data.\[23\] In addition, it has been conjectured that the unidentified phase protecting the QCEP may be a result of a type of Pomeranchuk instability of the Fermi surface, or alternatively from nanoscale charge inhomogeneity,\[24,26\] as the van-Hove singularity passes through the chemical potential. In fact, the van-Hove singularity is one possible microscopic origin for the effective action used by Millis et al.\[28\] It should be pointed out that the renormalization group treatment of this action has proven quite successful in explaining the thermal expansion data among other experimental observations.

In conclusion we have observed the verification of the Wiedemann-Franz law in Sr$_3$Ru$_2$O$_7$ in the limit as $T \to 0$ at all fields including the field at which a quantum critical end-point occurs, which implies that there is no breakdown of the electron at the QCEP. More precisely, there are no additional fermionic carriers of heat (such as spinons) other than those which carry charge $e$. This further supports the notion that the QCEP in Sr$_3$Ru$_2$O$_7$ can be described in the Hertz-Millis formalism for quantum criticality. It will be interesting to see if the WFL is still satisfied in other quantum critical systems in which the Hertz-Millis theory fails. Finally, the finite temperature data are consistent with the standard picture for itinerant electron metamagnetism, and, as a result, one should expect that the ferromagnetic fluctuations are responsible for the observed non-Fermi liquid behavior at the quantum critical endpoint.

We thank Y.B. Kim, A. Schofield, and I. Vekhter for useful discussions. Work at Los Alamos was performed under the auspices of the US DOE.
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