Thermoelectric response near a quantum critical point of $\beta$-YbAlB$_4$ and YbRh$_2$Si$_2$ : A comparative study

Y. Machida, K. Tomokuni, C. Ogura, K. Izawa, K. Kuga, S. Nakatsuji, G. Lapertot, G. Knebel, J.-P. Brison, and J. Flouquet

1Department of Physics, Tokyo Institute of Technology, Meguro 152-8551, Japan
2Institute for Solid State Physics, University of Tokyo, Kashiwa 277-8581, Japan
3INAC, SPSMS, CEA Grenoble, 38054 Grenoble, France

(Dated: February 14, 2012)

The thermoelectric coefficients have been measured on the Yb-based heavy fermion compounds $\beta$-YbAlB$_4$ and YbRh$_2$Si$_2$ down to a very low temperature. We observe a striking difference in the behavior of the Seebeck coefficient, $S$ in the vicinity of the Quantum Critical Point (QCP) in the two systems. As the critical field is approached, $S/T$ enhances in $\beta$-YbAlB$_4$ but is drastically reduced in YbRh$_2$Si$_2$. While in the former system, the ratio of thermopower-to-specific heat remains constant, it drastically drops near the QCP in YbRh$_2$Si$_2$. In both systems, on the other hand, the Nernst coefficient shows a diverging behavior near the QCP. The results provide a new window to the way various energy scales of the system behave and eventually vanish near a QCP.

PACS numbers: 71.27.+a, 72.15.Jf, 74.40.Kb

Heavy fermion compounds have been intensively studied for their ability to use pressure or magnetic field to access a quantum critical point (QCP) from a magnetic phase or a Fermi liquid (FL) state. One consequence of the QCP is an emergence of non-Fermi liquid (NFL) behavior revealing strong deviations from the FL. The behavior has been characterized by a set of exponents of the temperature dependence of the physical properties, such as the specific heat, the resistivity, and the susceptibility in the framework of the spin fluctuation theories. So far, these theories have been successful to describe the NFL properties of a number of materials. Recently, however, there has been found a few heavy fermion materials, e.g., CeCu$_{1−x}$Au$_x$, YbRh$_2$Si$_2$, and $\beta$-YbAlB$_4$, exhibiting the NFL behavior in the neighborhood of the QCP for which the standard theory is insufficient. In spite of the considerable efforts, both experimental and theoretical, up to date nature of their unconventional criticality has still been debated. A nuclear contribution to the specific heat, for example, tends to mask an intrinsic behavior at low temperatures, and necessarily leads to ambiguity.

Recently, new insight on the QCP via the thermoelectric coefficients, e.g., the Seebeck and Nernst coefficients, has been examined. An advantage of these quantities is that only itinerant quasiparticle (QP) excitations are measured, so thus they would be complementary to the specific heat: a measure of both itinerant and localized excitations. Moreover, in contrast to the specific heat, the thermoelectric response is not determined by the number of itinerant electrons present in the system and its magnitude is directly set by their relevant energy scale. In this paper, we report on a comparative study of the thermoelectric response near the QCP of the two representative Yb-based unconventional quantum critical materials, $\beta$-YbAlB$_4$ and YbRh$_2$Si$_2$. We find that while the Nernst response behaves in a similar manner in the two systems, the Seebeck coefficient and its relation to the specific heat are quite distinct in the two cases, allowing discrimination of the nature of their quantum criticality.

$\beta$-YbAlB$_4$ is the first Yb-based heavy fermion superconductor ($T_c = 80$ mK). The striking feature of this system is an emergence of the NFL behaviors under zero field at ambient pressure. This implies that the system is located near the QCP without the external tuning parameter. Coexistence of valence fluctuation and the NFL behavior is another feature of this material. YbRh$_2$Si$_2$ is a heavy fermion compound showing the NFL properties above the antiferromagnetic (AF) transition at $T_N = 70$ mK. Significantly, $T_N$ is found to be continuously suppressed with a small magnetic field ($B_c = 0.66$ T || c-axis) to zero accessing the field-induced QCP. An existence of additional energy scale $T^*$ discussed as the crossover line of the Kondo destruction is a puzzling feature of YbRh$_2$Si$_2$. A key characteristics of both systems, which goes beyond the standard theory, is a diverging enhancement of the effective mass and the uniform susceptibility towards the QCP. Stimulated by these striking similarity, the theoretical efforts have been devoted to understand the unconventional quantum criticality of the two systems in a unified way, but no consensus has been yet reached.

Single crystals of $\beta$-YbAlB$_4$ and YbRh$_2$Si$_2$ were prepared using the aluminum and indium flux, respectively. We used two crystals with different qualities for $\beta$-YbAlB$_4$ (the residual resistivity ratio (RRR) is 130 and 270 for sample $\equiv 1$ and $\equiv 2$, respectively), and one crystal (RRR ~ 90) for YbRh$_2$Si$_2$. The thermoelectric
coefficients were measured by employing a steady-state method in a dilution refrigerator. The heat current \( q \) was injected parallel to the \( ab \)-plane, and the magnetic field was applied parallel to the \( c \)-axis on the sample with a size of typically \( 0.5 \times 0.1 \times 0.005 \text{ mm}^3 \) for \( \beta \)-YbAlB\(_4\) and \( 1.6 \times 2.2 \times 0.05 \text{ mm}^3 \) for YbRh\(_2\)Si\(_2\), respectively. The thermal contacts with resistance of \( \leq 10 \text{ m}\Omega \) at room temperature were made by using a spot welding technique. The same contacts were used to measure the resistivity by a standard four-contact method.

Figure 1(a) shows the temperature dependence of the Seebeck coefficient divided by temperature \(-S(T)/T\) under zero field and 5 T for the two different samples of \( \beta \)-YbAlB\(_4\) (sample \#1 and \#2). What is remarkable is that the \(-S(T)/T\) curves for sample \#1 and \#2 converge at \( T < 0.15 \text{ K} \) under each field, even though the residual resistivity is different by a factor of 2.5 (the inset of Fig. 1(a)). This indicates that the Seebeck coefficient takes an identical value at sufficiently low temperature, irrespective of the sample quality. Using the Boltzmann equation, the Seebeck coefficient is expressed as \( S/T = -(\pi^2/3)(k_B^2/e)(\partial \ln \sigma(e)/\partial e)_{e=\epsilon_F} \) (\( \sigma \): the electrical conductivity). In the simple case of a free electron gas, the term \( (\partial \ln \sigma(e)/\partial e)_{e=\epsilon_F} \) is approximated by the energy derivative of the scattering time, \( (\partial \ln \tau(e)/\partial e)_{e=\epsilon_F} \). Moreover, in the zero-temperature limit, it can be further simplified as \( 20 \)

\[
\frac{S}{T} \sim -\frac{\pi^2}{3} k_B^2 e N(\epsilon_F) \left( 1 + \frac{2\xi}{3} \right),
\]

by expressing \( \tau(e) = \tau_0 e^{\xi} \), where \( N, \epsilon_F, \) and \( n \) are the density of states, the Fermi energy, and the carrier concentration, respectively. Therefore, the Seebeck coefficient is proportional to the density of states per electron as \( T \to 0 \). This fact gives rise to an intriguing aspect of \( S/T \) that it is interrelated with the electronic specific heat coefficient \( C/T = \gamma_0 \) via the dimensionless ratio \( q = (S/T)\gamma_0 N_{\text{AV}} \approx \pm 1 \) (\( N_{\text{AV}} \): the Avogadro number), since both quantities are proportional to the density of states. It has been shown that this semi-universality is held within logarithmic accuracy in a wide variety of metals including the heavy fermions \( 21 \). However, an insight into this ratio in the vicinity of the QCP remains an issue of debate.

With the above considerations in mind, let us look at variations of \( S(T)/T \) under fields. Figure 1 shows the \(-S(T)/T\) vs \( T \) plots under several magnetic fields for (b) \( \beta \)-YbAlB\(_4\) (sample \#2) and (c), (d) YbRh\(_2\)Si\(_2\), respectively. In both systems, the Seebeck coefficient takes a negative sign as observed in other Yb-based heavy fermion compounds \( 24 \). First, we focus on the results of \( \beta \)-YbAlB\(_4\). Under zero field, \(-S(T)/T\) exhibits a logarithmic increase down to 0.2 K and reaches a large value \(-S/T \sim 6 \mu\text{V/K}^2\), being two order of magnitude larger than the values for simple metals such as Cu \((S/T \sim -30 \text{ nV/K}^2)\) \( 20 \). The large \( S/T \) was also found in other heavy-fermion compounds \( 8, 10, 20 \) and attributed to a large effective mass. On further decreasing the temperature, \(-S(T)/T\) shows a steep increase followed by a sudden drop at 80 mK due to the superconducting transition. By contrast, under the field of 25 mT which is larger than the upper critical field \( 21 \) but very close to the QCP, the drop disappears and \(-S(T)/T\) is found to continuously increase down to the lowest temperature. By increasing the field \( B \geq 0.5 \text{ T} \), the low-temperature increase is suppressed and \(-S(T)/T\) approaches constant, indicating that the system becomes the FL.

Next, we turn to the results of YbRh\(_2\)Si\(_2\). The \(-S(T)/T\) curves taken under the fields below and above the critical field \( (B_c = 0.66 \text{ T}) \) are shown in Fig. 1(c) and 1(d), respectively. Under zero field, \(-S(T)/T\) shows a logarithmic increase at high temperature \( T > 0.2 \text{ K} \), the same as \( \beta \)-YbAlB\(_4\). However, upon cooling, \(-S(T)/T\) shows logarithmic increase at high temperature \( T > 0.2 \text{ K} \). The spin-density-wave (SDW) \( 22 \) and the Kondo breakdown scenarios \( 23 \). Although the experimental results agree with these predictions at high temperature, they contradict at low temperature, indicating that the loga-
The arithmetic divergence of $S/T$ does not necessarily capture the intrinsic quantum criticality. Rather, the contrasting behavior found at sufficiently low temperature would be crucial for understanding the QCP. As further increasing the fields, $-S(T)/T$ turns to increase below 60 mK at 1.5 T and shows a continuous increase down to the lowest temperature at 2.0 T. Above 3 T, the saturation behavior eventually appears after taking broad maxima, which roughly coincide with the crossover temperature to the Fermi liquid state [7]. These observations are qualitatively consistent with the results of S. Hartmann [10] in which the magnetic field is applied perpendicular to the c-axis.

To clearly demonstrate the contrasting behavior of $S/T$ near the QCP, we depict the contour plots of $-S(T)/T$ in the $B$-$T$ plane for (a) $\beta$-YbAlB$_4$ and (b) YbRh$_2$Si$_2$ in Fig. 2, respectively. One immediately notices that in the vicinity of the zero-field QCP ($B_c = 0$ T), $-S(T)/T$ for $\beta$-YbAlB$_4$ is very large, while the one for YbRh$_2$Si$_2$ is strongly suppressed around the field-induced QCP ($B_c = 0.66$ T). In addition, we find two enhanced regions in the plot of YbRh$_2$Si$_2$; one is a zero-field region above $T_N$ and the other is a zero-temperature region around 2 T. Moreover, the $T^*$ line runs through between two enhanced regions. It seems that the evolution of $-S/T$ towards the QCP due to increasing QP mass is unexpectedly suppressed around the QCP. An interpretation of this suppression will be discussed later. On the other hand, the fact that the large $-S/T$ survives up to as high as 3 T, away from the QCP, implies an additional source for the enhancement. The field-induced FM fluctuations detected by the NMR measurements [24] could be a candidate. We also mention that the persistent evolution of $-S/T$ in the low-field side of $T^*$ line, which amounts to as much as the one in the FL state, seems to contradict with the Kondo destruction scenario predicting the presence of the small Fermi surface [25, 26].

Let us further discuss the contrasting behavior of $S/T$ by examining the dimensionless ratio $q = (S/T \gamma_0) N_{AV}/e$. Figure 3 displays the field dependence of $-S(B)/T$ at 50 mK and $\gamma_0(B)$ [8, 27] for (a) $\beta$-YbAlB$_4$ and (b) YbRh$_2$Si$_2$, respectively. It is clearly seen that $\gamma_0(B)$ diverges as $B \rightarrow B_c$ in both systems, indicating a divergence of the QP mass on approaching the QCP [7, 8]. As expected from Eq. (1), $-S(B)/T$ for $\beta$-YbAlB$_4$ rapidly grows in parallel with $\gamma_0(B)$. On the contrary, although $-S(B)/T$ tracks $\gamma_0(B)$ at high field in YbRh$_2$Si$_2$, it turns to decrease as $B \rightarrow B_c$. Consequently, $|q|$ appears to be field-independent $|q| \sim 4$ in $\beta$-YbAlB$_4$, while the one of YbRh$_2$Si$_2$ shows a considerable decrease towards the QCP from a nearly constant value ($|q| \sim 1$) at high field (Fig. 3(c)). Therefore, the semi-universality of $q$ is held in $\beta$-YbAlB$_4$ even at the QCP, while it becomes invalid for YbRh$_2$Si$_2$ on approaching its QCP. This striking difference of $q$ seems to point to distinct nature of the quantum criticality in these systems.

It has been pointed out that the semi-universality of $q$ is satisfied when the whole Fermi surface (FS) equally contributes to $S/T$ and $\gamma_0$ [28]. This is a case for the QCP with $Q = 0$ fluctuations where the QP mass is enhanced on the entire region of the FS (the whole FS is “hot”), leading to the uniform enhancement of $S/T$ and $\gamma_0$. However, the semi-universality is invalid for the case of AF ($Q \neq 0$) QCP because the mass enhancement occurs on some parts of the FS, “hot regions” connected...
by the AF ordering wavevector $Q$. In this case, since the transport is dominated by the cold electrons, $S/T$ is diminished due to a reduction of the hot electrons contribution, while $\gamma_0$ is enhanced because it always has a contribution from the whole FS. This leads to imbalance of $S/T$ and $\gamma_0$, and results in the reduction of $|q|$ from order of unity. In fact, a considerable decrease of $|q|$ near the QCP was also found in CeCoIn$_5$ \cite{29}, for which the AF (SDW) scenario well accounts for the QCP. According to this interpretation, the unchanged $|q| \sim 4$ and the diminishing $|q|$ ($|q| < 1$) towards the QCP suggest dominant $Q = 0$ and finite $Q$ fluctuations in $\beta$-YbAlB$_4$ and YbRh$_2$Si$_2$, respectively. Apparently, this finding is at odds with the theoretical predictions which attempt to understand the quantum criticality of the two systems in a unified way \cite{10, 17}.

In contrast to the Seebeck coefficient, the Nernst coefficient $\nu/T$ is found to diverge in the vicinity of the QCP in both systems. Figure 4(a) and 4(b) represent $-\nu(T)/T$ as a function of temperature for $\beta$-YbAlB$_4$ and YbRh$_2$Si$_2$, respectively, under several fields. In $\beta$-YbAlB$_4$, we find a diverging increase of $-\nu(T)/T$, reaching as much as $7\mu V/K^2\cdot T$ at $0.5\ T$, while the enhancement is rapidly suppressed by the field of $B \geq 1\ T$. We note that $-\nu(T)/T$ is very large, being three order of magnitude larger than that of simple metals \cite{29}. Since the Nernst coefficient is expressed as the energy derivative of the Hall angle, $\nu/T \propto \partial \theta_H / \partial e_{\perp}$, $\nu/T$ is very small if $\tan \theta_H$ is only weakly dependent on energy, which is a good approximation in simple metals \cite{29}. On the other hand, in the case of the energy-dependent $\tan \theta_H$, $\nu/T$ can be obtained from the following simple expression, valid as $T \to 0$ \cite{29}: $\nu \approx \frac{\pi^2 k_B^2}{3} e_f$, where $\mu$ represents the carrier mobility. Therefore, the divergence of $-\nu(T)/T$ is interpreted as a consequence of vanishing of $e_f$, defined as the characteristic energy scale of the FL. A similar diverging enhancement of $-\nu(T)/T$ is also observed under the critical field ($B_c = 0.66\ T$) in YbRh$_2$Si$_2$ (Fig. 4(b)), although the behaviors of $-\nu(T)/T$ is rather complicated including a sign change. By further applying the fields, the diverging behavior is gradually suppressed and peaks are formed above $2\ T$ (downward arrows). These peak temperatures correspond to $T_{FL}$ in Ref. \cite{3}, indicating that the FL is restored below the peak temperatures. The sigh change of $-\nu(T)/T$ suggests an existence of two FSs with dominant carriers of opposite signs in accordance with the band calculations \cite{30}, which may result in the relatively small magnitude of $-\nu(T)/T$ compared with $\beta$-YbAlB$_4$ due to canceling of opposite sign contributions to the Nernst signal.

In summary, we present the low-temperature thermoelectric coefficient measurements of $\beta$-YbAlB$_4$ and YbRh$_2$Si$_2$. Both systems display the diverging enhancement of the Nernst coefficient on approaching their QCPs, indicative of the vanishing of the Fermi energy. On the other hand, the significant deviation from the semi-universal ratio $q$ of the Seebeck coefficient and the electronic specific heat is found in YbRh$_2$Si$_2$ in the vicinity of the QCP, while the universality is sustained in $\beta$-YbAlB$_4$. This striking difference suggests distinct type of fluctuations underlying the quantum criticality; $Q = 0$ and finite $Q$ fluctuations for $\beta$-YbAlB$_4$ and YbRh$_2$Si$_2$, respectively. Further experiments on other quantum critical materials in terms of $q$ would enable systematic clarification of the quantum criticality.

This work is supported by Grants-in-Aids (Nos. 23340099, 23740263, 21684019) for Scientific Research from the Japanese Society for the Promotion of Science, a Grant-in-Aid for Scientific Research on Innovative Areas “Heavy Electrons” (Nos. 20102006, 23102706) of the Ministry of Education, Culture, Sports, Science, and Technology (MEXT), and a Grant-in-Aid for the Global COE Program from the MEXT through the Nanoscience and Quantum Physics Project of the Tokyo Institute of Technology.

\begin{thebibliography}{99}
\bibitem{1} P. Gegenwart, Q. Si, and F. Steglich, Nature Phys. 4, 186 (2008).
\bibitem{2} T. Moriya and T. Takimoto, J. Phys. Soc. Jpn. 64, 960 (1995).
\bibitem{3} J. A. Hertz, Phys. Rev. B 14, 1165 (1976).
\bibitem{4} A. J. Millis, Phys. Rev. B 7183, 48 (1993).
\bibitem{5} W. Knafo, S. Raymond, P. Lejay, and J. Flouquet, Nature Phys. 5, 753 (2009).
\bibitem{6} H. v. Löhneysen et al., Phys. Rev. Lett. 72, 3262 (1994).
\bibitem{7} P. Gegenwart et al., Phys. Rev. Lett. 89, 056402 (2002).
\bibitem{8} S. Nakatsuji et al., Nature Phys. 4, 603 (2008).
\bibitem{9} K. Izawa et al., Phys. Rev. Lett. 99, 147005 (2007).
\bibitem{10} S. Hartmann et al., Phys. Rev. Lett. 104, 096401 (2010).
\bibitem{11} Y. Matsumoto et al., Science 21, 316 (2011).
\bibitem{12} M. Okawa et al., Phys. Rev. Lett. 104, 247201 (2010).
\bibitem{13} J. Custers et al., Nature 423, 524 (2003).
\bibitem{14} S. Paschen et al., Nature 432, 881 (2004).
\bibitem{15} J. Custers et al., Phys. Rev. Lett. 186402, 104 (2010).
\bibitem{16} T. Misawa, Y. Yamaji, and M. Imada, J. Phys. Soc. Jpn.
\end{thebibliography}
78, 084707 (2009).
[17] S. Watanabe and K. Miyake, Phys. Rev. Lett. 105, 186403 (2010).
[18] R. T. Macaluso et al., Chem. Mater. 19, 1918 (2007).
[19] G. Knebel et al., J. Phys. Soc. Jpn. 75, 114709 (2006).
[20] K. Behnia, D. Jaccard, and J. Flouquet, J. Phys.: Condens. Matter 16, 5187 (2004).
[21] K. Kuga et al., Phys. Rev. Lett 101, 137004 (2008).
[22] I. Paul and G. Kotliar, Phys. Rev. B 64, 184414 (2001).
[23] K.-S. Kim and C. P´epin, Phys. Rev. B 81, 205108 (2010).
[24] K. Ishida et al., Phys. Rev. Lett. 89, 107202 (2002).
[25] Q. Si, S. Rabello, K. Ingersent, and J. L. Smith, Nature 413, 804 (2001).

[26] P. Coleman, C. P´epin, Q. Si, and R. Ramazashvili, J. Phys.: Condens. Matter 13, R723 (2001).
[27] The value of $\gamma_0$ at the QCP is taken from the temperature dependence of the electronic specific heat, $C_{el}(T)/T \equiv \gamma_0$ at 50 mK [8, 13]. In addition, for $\beta$-YbAlB$_4$, $C_{el}(T)/T$ at 50 mK is estimated from the extrapolation by assuming $C_{el}(T)/T \propto -\ln T$.
[28] K. Miyake and H. Kohno, J. Phys. Soc. Jpn. 74, 254 (2005).
[29] K. Behnia, J. Phys.: Condens. Matter 21, 113101 (2009).
[30] G. A. Wigger et al., Phys. Rev. B 76, 035106 (2007).