Band dependent normal-state coherence in Sr$_2$RuO$_4$: Evidence from Nernst effect and thermopower measurements

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We present the first measurement on Nernst effect in the normal state of odd-parity, spin-triplet superconductor Sr$_2$RuO$_4$. Below 100 K, the Nernst signal was found to be negative, large, and, as a function of magnetic field, nonlinear. Its magnitude increases with the decreasing temperature until reaching a maximum around $T^* \approx 20 - 25$ K, below which it starts to decrease linearly as a function of temperature. The large value of the Nernst signal appears to be related to the multiband nature of the normal state and the nonlinearity to band-dependent magnetic fluctuation in Sr$_2$RuO$_4$. We argue that the sharp decrease in Nernst signal below $T^*$ is due to the suppression of quasiparticle scattering and the emergence of band-dependent coherence in the normal state. The observation of a sharp kink in the temperature dependent thermopower around $T^*$ and a sharp drop of Hall angle at low temperatures provide additional support to this picture.

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Support for an odd-parity, spin-triplet pairing in Sr$_2$RuO$_4$ by increasing number of experiments$^{[1, 2, 3, 4]}$, including in particular the first phase-sensitive measurements$^{[2]}$, has provided strong motivation to pursue a detailed understanding of the mechanism of superconductivity in Sr$_2$RuO$_4$. Rice and Sigrist$^{[5]}$ suggested that ferromagnetic (FM) fluctuation might be responsible for spin-triplet superconductivity in Sr$_2$RuO$_4$. Inelastic neutron scattering (INS) measurements$^{[6]}$ revealed magnetic fluctuation with peaks in the dynamic susceptibility only around $k$-vectors $(\pm 0.6, \pm 0.6, 0)\pi/a$, where $a$ is the lattice constant, suggesting that incommensurate magnetic fluctuation (IMF) dominates in Sr$_2$RuO$_4$. Theoretical calculations$^{[7]}$ suggest that the IMF originates from the one-dimensional (1D) $d_{x^2-y^2}$ bands in Sr$_2$RuO$_4$, and spin-singlet rather than spin-triplet superconductivity is favored in these 1D bands. These observations have raised questions on the mechanism on superconductivity in Sr$_2$RuO$_4$. So far, models based on FM$^{[8]}$, antiferromagnetic (AF) fluctuations$^{[9]}$, spin-orbital coupling$^{[10]}$, or Hund’s rule coupling$^{[11]}$ for superconductivity in Sr$_2$RuO$_4$ have been proposed. But none has gained universal acceptance.

The Fermi surface of Sr$_2$RuO$_4$ consists of three cylindrical sheets$^{[12, 13]}$: one hole (the $\alpha$) and two electronlike (the $\beta$ and the $\gamma$) bands. It was suggested that superconductivity in Sr$_2$RuO$_4$ is band-dependent with its energy gap large on the $\gamma$ band but tiny on the $\alpha$ and the $\beta$ bands$^{[14]}$. The interesting question is whether the orbital-dependent superconductivity comes from orbital-dependent normal-state properties to begin with. This idea can actually be traced back to the original paper of Baskaran$^{[11]}$, who was among the first to suggest that Sr$_2$RuO$_4$ is a $p$-wave superconductor. Thermoelectric measurements provide a natural probe for the orbital-dependent physical properties, as demonstrated previously$^{[15]}$. In this Letter, we present the first comprehensive study of the Nernst effect and thermopower in Sr$_2$RuO$_4$. Our results show clearly that quasiparticle scattering is strongly suppressed and band-dependent coherence emerges at low temperatures.

Single crystals of Sr$_2$RuO$_4$ were grown by the floating-zone method as reported previously$^{[16]}$. Three single crystals, labelled as 214-A, B and C, were measured in this experiment. The values of the in-plane residual resistivity were found to be less than 0.5 $\mu\Omega$cm for all three crystals, confirming the ultra-high quality of the crystals$^{[16]}$. In addition, we measured each of these crystals in a Quantum Design MPMS-5 system to ensure the absence of the intergrowth of other phases in the Ruddlesden-Popper series of Sr$_{n+1}$Ru$_n$O$_{3n+1}$. The thermoelectric properties were measured by the steady-state technique. The magnetic field was applied along the $c$ axis. The temperature gradient, around 0.5 K/mm, was applied in the $ab$ plane and determined by a pair of differential Type E thermocouples. All measurements were performed in a Quantum Design PPMS-9 system.

Traces of Nernst signal as a function of magnetic field for Sample 214-A at various temperatures are displayed in Fig. 1. Above 100 K, the Nernst signal is small and linear as a function of magnetic field. As temperature decreases, the magnitude of the Nernst signal increases at a given field, reaching a large value of about -0.43 $\mu$V/K
under $B = 8$ T at 20 K. However, the growth of the magnitude of the Nernst signal, $|e_y|$, is reversed around $T^*$, below which $|e_y|$ decreases linearly. An equally striking feature is that, as the temperature is lowered, the Nernst signal is found to become nonlinear above a characteristic field. The "characteristic" field decreases with decreasing temperature, indicating that the nonlinearity becomes increasingly pronounced at low temperatures. Similar data are also obtained for the other two samples.

Figure 2 shows the temperature dependence of Nernst signal at a magnetic field of 6 T obtained from all three samples measured. The magnitude of Nernst signal starts to increase around 100 K as the temperature is lowered. After reaching a maximum around $T^*$, the magnitude of the Nernst signal is seen to decrease linearly with the decreasing temperature, approaching zero as temperature decreases to zero. The temperature dependence of the Nernst coefficient, $\nu \equiv \partial e_y / \partial B$ determined from the initial slope of the $e_y$ vs. $B$ curves, shows similar behavior (the inset of Fig. 2).

A large and strongly nonlinear Nernst signal observed in a metallic sample is highly unusual. In a single-band conventional metal, the Nernst signal is usually small because of the Sondheimer cancellation[17], and linearly dependent on magnetic field. We are aware of only few examples showing otherwise. The first is the vortex motion in the mixed state of a type II superconductor, as observed in the mixed state and a certain temperature range above $T_c$, of high-$T_c$ superconducting cuprates[17, 18, 19]; The second example is that a large and nonlinear Nernst signal was found below and even above the Curie temperature in a ferromagnet[20]; The third example was found in the Kondo lattice, heavy Fermion superconductor CeCoIn$_5$[21], even though its physical origin is not understood; The fourth example is related to the difference in scattering rates of different energy bands in a multiband metal such as NbSe$_2$[12]. The presence of two types of carriers, invalidates the Sondheimer cancellation, resulting in a large Nernst signal.

For Sr$_2$RuO$_4$, no superconducting fluctuation is expected at a temperature as large as 100 K. Furthermore, the Nernst signal is negative rather than positive as it would be expected in a type II superconductor. On the other hand, Sr$_2$RuO$_4$ is a multiband metal whose Nernst signal is enhanced as seen in NbSe$_2$[13]. In a simple two-band model, the Nernst signal can be express as

$$e_y = S\left(\frac{\alpha_{xy}^h + \alpha_{xy}^e}{\alpha_{xx}^h + \alpha_{xx}^e} - \frac{\sigma_{xy}^h + \sigma_{xy}^e}{\sigma_{xx}^h + \sigma_{xx}^e}\right),$$

where $S$ is the thermopower, $\alpha$ is the Peltier conductivity tensor and $\sigma$ is the electric conductivity tensor with the superscripts $e$ and $h$ referring to electrons and holes. The subscripts $xx$ and $xy$ refer to the diagonal and off-diagonal components of the tensors. It can be seen that the Nernst signal is sensitive to the different temperature dependence of scattering rates in different bands.

The observed nonlinearity in the Nernst signal, on the other hand, appears to be related to magnetic fluctuation. As pointed above, IMF originating from $\alpha$ and $\beta$
bands dominates in Sr$_2$RuO$_4$\cite{24,25}. If the IMF is suppressed so that its contribution to the Nernst signal decreases at high magnetic fields, or alternatively, FM fluctuation is induced by an applied field, a nonlinear Nernst signal can be understood. In other case, the maximum in the temperature dependence of the Nernst signal near $T^*$ (Fig. 2) suggests a sharp change in band-dependent quasiparticle scattering rates or the density of states at this temperature. To address the latter possibility, we measured the specific heat and found no anomaly present around $T^*$ (Inset of Fig. 3). Our results suggest that it is unlikely that the density of states is changed around $T^*$. On the other hand, below $T^*$, the Fermi liquid behavior, i.e., the $T^2$ behavior, was found in both in- and out-of-plane resistivities. This is consistent with linear temperature dependence of Nernst signal or Nernst coefficient observed in Sr$_2$RuO$_4$ below $T^*$ \cite{24}. The emergence of the Fermi liquid and the change in temperature dependence of the Nernst signal at the same temperature therefore signal an important change in the character of the normal state. We argue that a coherence state emerges at $T < T^*$, which will be further discussed below.

Since thermopower is sensitive to the change of quasiparticle scattering rates in different bands, any change in the electronic state should also result in a sharp feature in the temperature dependence of the thermopower. As shown in the upper panel of Fig. 3, the $S(T)$ curves indeed exhibit a sharp change of slope around $T^*$. This change becomes even more striking in the plot of $dS/dT$ vs. $T$ as shown in the lower panel of Fig. 3. A feature in thermopower was found previously in Sr$_2$RuO$_4$ \cite{24}, although not as pronounced as seen in the present work.

The Hall coefficient was found previously to change its sign from hole-like (suggesting that the $\alpha$ band dominates) above $T^*$, to electron-like below this temperature\cite{24,26,27}. It is known that Hall angle, rather than Hall coefficient, is directly related to the scattering rate of the quasiparticle scattering. In Fig. 4, we show the temperature dependence of Hall angle, which shows a steep drop at low temperatures. This drop marks the increase of the scattering time, which may be taken as direct evidence of the existence of normal-state coherence. The temperature dependence of Hall coefficient can be well understood\cite{27} in a multi-band model\cite{24}. The sharp drop of $R_H$, thus tan$\theta$, below 25 K, indicates a decreasing $\ell_h/\ell_e$ ratio, where $\ell_h$ ($\ell_e$) is the mean free path in the hole-like (electron-like) band. The sharp increase in $\ell_e$ should result from the band-dependent change in the scattering time. As pointed out before, quasiparticle scattering in $\alpha$ as well as $\beta$ band is dominated by IMF. However, no change was found in IMF around $T^*$\cite{24}. Therefore it is reasonable to conclude that the change in quasiparticle scattering rate revealed by the Nernst effect and Hall effect measurements must be limited to the $\gamma$ band. The increase in $\ell_e$ is consistent with the emergence of a coherence state in the electron-like $\gamma$ band. This implies that the coherence among quasiparticles occurs in this particular band originating from $d_{xy}$ orbitals.

Nernst measurement probes both the off-diagonal Peltier current and ordinary Hall current characterized by

$$e_y = \rho \alpha_{xy} + \rho_{xy} \alpha_{xx} = S(\tan \theta_{\alpha} - \tan \theta)$$ \hspace{1cm} (2)

where the "Peltier Hall angle" $\tan \theta_{\alpha}$ is defined as $\tan \theta_{\alpha} = \alpha_{xy}/\alpha_{xx}$. Actually each angle includes the contributions from both electron and hole bands (see Eq. 1). The "Peltier Hall angle" $\tan \theta_{\alpha}$ is also shown in Fig. 4. At temperatures above 25 K, the Hall angle $\tan \theta$ shows very weak temperature dependence, and the absolute magnitude of the "Peltier Hall angle" $\tan \theta_{\alpha}$ increases gradually. It is clear that the gradual increase in $e_y$ originates from the increase of $\tan \theta_{\alpha}$. Around $T^*$, a kink in $\tan \theta_{\alpha}$ was observed. While the Hall angle probes the scattering time, the "Peltier Hall angle" is sensitive to the energy dependence of the scattering time\cite{17}. The band dependent coherence state is responsible for features seen in both $\tan \theta$ and $\tan \theta_{\alpha}$.

An interesting question is whether the quasiparticle coherence in the $\gamma$ band below $T^*$ can be viewed by...

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the opening of a pseudogap. Infrared spectroscopy measurements did find evidence for a gap of 6.3 meV opening in the normal state of Sr$_2$RuO$_4$.[20] A kink in the temperature dependence of $1/T_1$ around 70 K in the NMR measurements is consistent with the opening of a pseudogap.[22] However, the presence of Fermi liquid behavior in Sr$_2$RuO$_4$ seems to be inconsistent with the pseudogap idea.[20] The new insight provided by the present study is that the pseudogap may be band-dependent, and coexist with the Fermi liquid behavior. The emergence of normal-state coherence should be related to superconductivity in Sr$_2$RuO$_4$. It was proposed previously[14] that the $\gamma$ band is the active band that gives rise to spin-triplet superconductivity in Sr$_2$RuO$_4$. It seems natural that the coherence among quasiparticles and superconductivity in the $\gamma$ band are correlated. Alternatively, if the coherence below $T^*$ reflects a hidden order of a non-superconducting state, it may actually compete with rather than help superconductivity. In this regard, it may be useful to point out that in Sr$_3$Ru$_2$O$_7$, a paramagnetic compound closely related to Sr$_2$RuO$_4$, tendencies for both FM and AF orderings were found to coexist, and evidently compete for stability.[31] Thus our thermoelectric measurements may provide insight into the question raised originally by Baskaran[11] - why is the $T_c$ of Sr$_2$RuO$_4$ so low?

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