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Colossal thermomagnetic response in the exotic superconductor URu$_2$Si$_2$

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When a superconductor is heated above its critical temperature $T_c$, macroscopic coherence vanishes, leaving behind droplets of thermally fluctuating Cooper pair. This superconducting fluctuation effect above $T_c$ has been investigated for many decades and its influence on the transport, thermoelectric and thermodynamic quantities in most superconductors is well understood by the standard Gaussian fluctuation theories [1]. The transverse thermoelectric (Nernst) effect is particularly sensitive to the fluctuations, and the large Nernst signal found in the pseudogap regime of the underdoped high-$T_c$ cuprates[2, 3] has raised much debate on its connection to the origin of superconductivity. Here we report on the observation of a colossal Nernst signal due to the superconducting fluctuations in the heavy-fermion superconductor URu$_2$Si$_2$. The Nernst coefficient is enhanced by as large as one million times over the theoretically expected value within the standard framework of superconducting fluctuations. This, for the first time in any known material, results in a sizeable thermomagnetic figure of merit approaching unity. Moreover, contrary to the conventional wisdom, the enhancement in the Nernst signal is more significant with the reduction of the impurity scattering rate. This anomalous Nernst effect intimately reflects the highly unusual superconducting state embedded in the so-called hidden-order phase of URu$_2$Si$_2$. The results invoke possible chiral or Berry-phase fluctuations originated from the topological aspect of this superconductor, which are associated with the effective magnetic field intrinsically induced by broken time-reversal symmetry [4-7] of the superconducting order parameter.

The measurements of the Nernst effect provide a unique opportunity to study the superconducting fluctuations deep inside the normal state above $T_c$ [2, 3, 8-21]. The Nernst signal $N$ is the electric field $E_y (\| y)$ response to transverse temperature gradient $\nabla_x T (\| x)$ in the presence of magnetic field $H (\| z)$, $N \equiv E_y / (-\nabla_x T)$. The Nernst coefficient defined as $\nu \equiv N/\mu_0 H$ above $T_c$ consists of two contributions generated by different mechanisms: $\nu = \nu^S + \nu^N$. The first term $\nu^S$ represents the contribution of superconducting fluctuations of either amplitude or phase of the order parameter, which is always positive. The second term $\nu^N$ represents the contribution from the normal quasiparticles, which can be either positive or negative. The second contribution is usually small in conventional metals. In almost all superconductors the superconducting fluctuation contribution to the Nernst effect can be accounted for by the Gaussian-type fluctuations [14, 15]. Recently, a large Nernst signal has been reported in the pseudogap state of the underdoped high-$T_c$ cuprates, which has been discussed in terms of possible vortex-like excitations of phase disordered superconductors [2, 3, 8]. Although its origin is still controversial, these results imply that the fluctuation induced Nernst signal above $T_c$ is intimately related to the exotic superconducting state below $T_c$. The same conclusion is supported by the observation of an anomalous Nernst effect in the unconventional superconductor CeCoIn$_5$ [22].

The heavy-fermion compound URu$_2$Si$_2$ exhibits unconventional superconductivity ($T_c \approx 1.5$ K). This compound is distinguished from the other heavy fermion compounds, by the fact that the mysterious hidden-order transition takes place at $T_{HO} = 17.5$ K and no evidence of magnetic order has been found below $T_{HO}$ [23]. This system has been suggested to be a candidate of a chiral $d$-wave superconductor that spontaneously breaks time-reversal symmetry (TRS) in the superconducting state [4-7]. Indeed, angular variation of the thermal conductivity and specific heat in magnetic fields indicate the presence of point nodes in the order parameter and a chiral $d$-wave pairing symmetry in a complex form of $k_z (k_x \pm ik_y)$ has been proposed [4, 5]. Very recently, the brokenTRS has been also reported by polar Kerr effect measurements (Kapitulnik, A. private communications). Based on these results, possible Wyle-type topological superconducting states have been discussed [24]. It is therefore highly intriguing to examine the superconducting fluctuations in URu$_2$Si$_2$.

Figure 1a shows $\nu(T)$ in the zero-field limit (see also
in the two crystals, defined by the point of zero resistivity is 1.45 K. Upper inset for #1 and #2, respectively, below 6 K. In both crystals, dependence of values are determined from RRR crystals #1 (in the zero-field limit (H//c) and the in-plane resistivity of ultraclean URu2Si2 single crystals [25] (Tc=1.45 K) with residual-resistivity-ratio (RRR) of 1080 (#1) and 620 (#2). Above T_HO, ν(T) is negligibly small and exhibits a dramatic increase on entering the hidden-order state. Below T* ~ 5 K, ν(T) shows a further enhancement and increases divergently with approaching Tc (Figs. 1a,b). The inset of Fig. 1b shows the T-dependence of the ratios of Nernst coefficient and conductivity σ = 1/ρxx in the two crystals, r_ν = ν(#1)/ν(#2) and r_σ = ρxx(#2)/ρxx(#1).

Fig. 1. Transverse thermoelectric response in URu2Si2. a, The T-dependence of Nernst coefficient ν in the zero-field limit (H//c) and resistivity ρxx for single crystals #1 (RRR = 1080) and #2 (RRR = 620). The RRR values are determined from ρ(300 K)/ρ0 by assuming the T-dependence of ρxx as ρxx(T) = ρ0 + ATn with n=1.5 and 1.7 for #1 and #2, respectively, below 6 K. In both crystals, Tc defined by the point of zero resistivity is 1.45 K. Upper inset illustrates the crystal structure of URu2Si2 and lower inset depicts the schematic measurement setup. b, Low temperature data of ν(T) and ρxx(T) for crystals #1 and #2. Below T*, ν is largely enhanced from the T-linear dependence extrapolated from higher temperatures (dashed lines). Inset shows the T-dependence of the ratios of Nernst coefficient and conductivity of the two crystals, r_ν = ν(#1)/ν(#2) and r_σ = ρxx(#2)/ρxx(#1).

We discuss the Nernst signal in the T-range free from the superconducting fluctuations (T* ≲ T ≲ T_HO). As shown in Fig. 2b, the increase of RRR or the scattering time τ leads to an enhancement of ν^S. Within the Boltzmann theory, when τ is weakly energy dependent, ν^S can be expressed as ν^S = e^2/k_B^2 T^2/τ [9, 27], where k_B is the Boltzmann constant, m* is the effective mass and ε_F is the Fermi energy.

The striking enhancement of ν below T_HO is attributed to the strong reduction of ε_F associated with the disappearance of carriers and concomitant enhancement of τ, both of which have been reported previously [4]. The fact that r_σ above T* coincides well with r_σ (inset of Fig. 1b) provides quantitative support of ν^S ∝ τ.

At lower temperatures below T*, ν of clean crystals becomes huge especially in the vicinity of Tc. Indeed, ν of the cleanest crystal #1 is comparable to that of pure semimetal Bi with the largest Nernst coefficient reported so far [27]. Moreover, the combination of the large Nernst signal and high conductivity in this system leads to a sizeable thermo-magnetic figure of merit ZT = N^2σT/κ (κ is the thermal conductivity), which quantifies the adequacy of a given material for thermoelectric refrigeration. As shown in Fig. 2c, this number exceeds by far the values of previously studied materials and approaches unity at 1.5 K and 1 T, which opens a possible route toward thermomagnetic cooling for a cryogenic Ettingshausen refrigerator [28]. Interestingly, for the Nernst effect based engine there exists universal bound for the ratio between the maximum efficiency and the Carnot efficiency [29].

Now we discuss the fluctuation induced Nernst coefficient ν^S. The enhancement of r_ν(T) below T* (inset of Fig. 1b) and no discernible enhancement of ν(T) near Tc for RRR ~ 30 (inset of Fig. 2b) indicate that ν^S is dramatically enhanced with τ. We stress that this τ-dependence of ν^S is opposite to that expected in the conventional Gaussian fluctuation theories, which predict ν^S ∝ ρxx ∝ 1/τ [15, 18–20]. It has also been reported that in underdoped cuprates the introduction of impurities by irradiation enhances ν^S [8], which is again opposite to URu2Si2. Thus these results highlight an essential difference in the superconducting fluctuations between URu2Si2 and the other superconductors.

The unusual nature of the superconducting fluctuations in URu2Si2 is further revealed by the off-diagonal component of the thermo-electric tensor (Peltier coeff-
Anomalously large Nernst signal and thermo-magnetic figure of merit. a, The $T$-dependence of $\nu$ (left scale) and $\rho_{xx}$ (right scale) measured at $\mu_0H=1$ T near the superconducting transition. Both $\nu$ and $\rho_{xx}$ vanishes at the vortex lattice melting transition temperature $T_{\text{melt}}$. b, Comparison of the $\nu(T)$ data at $\mu_0H=1$ T between samples with different scattering rates ($\text{RRR} = 1080$, $620$ and $30$). The data for $\text{RRR} \sim 30$ (expanded in the inset) is taken from Ref. 26. c, Thermo-magnetic figure of merit $ZT = N^2\sigma T/\kappa$ at $1.5$ K as a function of field in crystal #1 of URu$_2$Si$_2$ (red diamonds), which is compared with the previous data in the semimetals PrFe$_4$P$_{12}$ (blue line) and Bi (black line) at $1.2$ K taken from Ref. 28.
shows the peak field plotted as a function of $\ln(T/T_c)$. The solid line, which represents $H^* = \frac{\mu_0}{\pi} \ln(T/T_c)$ calculated by using $\xi_{xy}(0) = 10 \text{ nm}$, gives a quantitative consistency with the peak field.

We discuss several possible origins for the observed colossal Nernst signal. First, Eq. (1) assumes the diffusive limit, $k_B T \ll h/\tau$, while the present URu$_2$Si$_2$ appears to be in the ballistic limit, $k_B T \gg h/\tau$. However, a ballistic theory cannot explain the observed $\alpha_{xy}^S$, which is nearly one million times greater than than $\alpha_{xy}^{\text{ML}}$ (Supplementary Information). Moreover, although such a theory shows the enhancement of $\alpha_{xy}^S$ with $\tau$, this enhancement is slower than the reduction of $\rho_{xx}$ so that $\nu^S(\alpha_{xy}^S/\rho_{xx})$ is still suppressed for larger $\tau$, which is inconsistent with Fig. 2b. Second, in the multiband system, each band with different effective coherence lengths contributes differently to the total $\alpha_{xy}^S$. To explain the observed $\alpha_{xy}^S$, however, small bands with extremely large effective coherence lengths, whose effective $H_{c2}$ corresponds to less than 1 mOe, are required. The multiband effect is, therefore, highly unlikely to explain the observed $\alpha_{xy}^S$. Third, the characteristic temperature scale of phase fluctuations is given as, $T_\phi = \frac{\hbar^2}{\mu_0 \chi_{xy}(0)}$, where $\chi_{xy}(0)$, $\lambda_{ab}$ is the in-plane penetration depth, and $A$ is a dimensionless number of the order of unity [30]. Using $A = 2$ and $\lambda_{ab} = 0.8 \mu m$ [6], we obtain $T_\phi/T_c \sim 100$, suggesting that the phase fluctuations are not important.

The unprecedented colossal thermomagnetic response in URu$_2$Si$_2$ appears to point to a new type of superconducting fluctuations generated by a degree of freedom which has not been hitherto taken into account. We note that it has been reported very recently that the chirality or Berry phase associated with the superconducting state with broken TRS gives rise to a new type of fluctuations [21]. In fact, according to Ref. 21, $\alpha_{xy}^S(T)$ is strikingly enhanced with $\tau$ and its $T$-dependence is different from that predicted by the Gaussian fluctuation theories, which are consistent with the present results at least at the qualitative level. The present results suggest that superconducting fluctuations contain a key ingredient for the topological nature of superconductors, which is a new frontier of condensed matter physics.

**Methods Summary** The ultraclean single crystals of URu$_2$Si$_2$ were grown by the Czochralski pilling method in a tetra-arc furnace [25]. The well defined superconducting transition was confirmed by the specific heat measurements. The Nernst and Seebeck coefficients were measured by the standard dc method with one resistive heater, two Cernox thermometers and two lateral contacts (lower inset of Fig. 1a).

**References**

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Supplementary Information is available in the online version of the paper.

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Supplementary Information

SEEBECK COEFFICIENT

Figure S1 shows the longitudinal thermoelectric (Seebeck) coefficient $S$ for crystals with $RRR = 1080$ (#1), $RRR = 620$ (#2), and $RRR \sim 30$ measured at zero field. The data of $RRR \sim 30$ crystal is taken from Ref. 26 in the main text. In stark contrast to Nernst coefficient shown in Fig. 2b, the $RRR$- or $\tau$-dependence of the magnitude of $S$ is very small. This is consistent with the results of Boltzmann equation, in which weak energy dependence of $\tau(\varepsilon)$ is assumed.

FIELD DEPENDENCE OF NERNST SIGNAL

Figure S2a shows the $H$-dependence of the Nernst signal $N(H)$ above $T_c$ for crystal #2. At high temperatures, $N(H)$ increase nearly linearly with $H$. With approaching $T_c$, $N(H)$ becomes nonlinear as a function of $H$. We determined the Nernst coefficient $\nu = N/\mu_0 H$ in the zero field limit by fitting the $H$ dependence of $N(H)$ using the polynomial functions, and by taking a derivative $dN(H)/d(\mu_0 H)$ at $H = 0$.

The Nernst signal $N = \nu \mu_0 H$ is written as,

$$N = \alpha_{xy} \rho_{xx} - S \tan \theta_H. \quad (S1)$$

Figure S2b depicts the $H$-dependencies of $N$ and $S \tan \theta_H$ for #2 crystal above $T_c$. Here $\tan \theta_H \equiv \rho_{xy}/\rho_{xx}$ is the Hall angle, where $\rho_{xx}$ and $\rho_{xy}$ are the in-plane diagonal and Hall resistivities, respectively. Figure S2b shows that $\alpha_{xy} \rho_{xx}$ well dominates over $S \tan \theta_H$, indicating that $\nu$ can be approximated as $\nu \approx \alpha_{xy} \rho_{xx}/\mu_0 H$.

REMARKS ON THE PELTIER COEFFICIENT IN THE BALLISTIC LIMIT

The validity of Eq. (1) from the main part of the paper has been microscopically established only for superconductors with the short mean free path $\ell \ll \xi(0)$, which is equivalent to the condition of diffusive scattering $T \tau \ll 1$. It is important to realize that the opposite limit consists of two regimes: the ballistic limit, $\xi(0) \ll \ell \ll \xi(T)$, or equivalently $1 \ll T \tau \ll \sqrt{T_c/(T - T_c)}$, and ultra-ballistic limit, $\ell \gg \xi(T)$, which is equivalent to $T \tau \gg \sqrt{T_c/(T - T_c)}$ near $T_c$. The latter case was rarely discussed in the literature despite the fact that it becomes of primary importance for the description of most experiments. It is feasible to expect that at $T \tau \gtrsim 1$ temperature dependence of the Peltier coefficient should crossover to a different law in a parameter $T \tau$, which requires separate theoretical investigation.

We follow here the calculation of Ref. S1 in order to trace the $T \tau$ dependence of the transverse thermoelectric
coefficient $\alpha_{xy}$. The latter is defined as

$$\alpha_{xy} = -\frac{j_y^Q}{E_x T} + \frac{c M_z}{T} = \alpha_{xy} + \frac{c M_z}{T}, \quad (S2)$$

where the last term $\propto M_z$ corresponds to the magnetization contribution, while the first term is the thermal response to electric field $E_x$ in the presence of the magnetic field $H$. Thermal current $j_y^Q$ can be found through the Kubo formula (hereafter $h = 1$)

$$\frac{j_y^Q}{E_x} = - \lim_{\Omega, Q \to 0} \frac{H}{i\Delta Q} \text{Re}[\mathcal{K}_{xy}(Q, i\Omega_m)]_{i\Omega_m \to \Omega + i0}, \quad (S3)$$

where we assumed low field limit $H \ll H_c$. The response kernel is defined as follows

$$K_{xy}(Q, i\Omega_m)$$

becomes nonlocal, which gives us

$$\lim_{\Omega, Q \to 0} \frac{H}{i\Delta Q} \text{Re}[\mathcal{K}_{xy}(Q, i\Omega_m)]_{i\Omega_m \to \Omega + i0},$$

where we assumed low field limit $H \ll H_c$. The response kernel is defined as follows

$$K_{xy}(Q, i\Omega_m)$$

and $j^c$ and $j^Q$ are the electrical and heat current vertices, respectively, and summation goes over the Matsubara frequencies $\omega_n = 2\pi n T$. Superconducting fluctuations in the clean limit are essentially nonlocal, which may strongly influence their electromagnetic response. The propagator (pair susceptibility) of the preformed Cooper pairs is of the form [S2]

$$(S4)$$

$$(S5)$$

$$(S6)$$

$$(S7)$$

where $D$ is the density of states in the normal state. Calculation of $K_{xy}$ with this form of the propagator is extremely involved, however Eq. (S5) can be simplified by expanding it over $v_F q / \max(T_c, \tau^{-1}) \lessapprox 1$ which is justified as long as $T \ll \sqrt{T_c / (T - T_c)}$. Assuming this limit and generalizing Eq. (S5) for the anisotropic three-dimensional case one finds

$$(S8)$$

$$(S9)$$

$$(S10)$$

with $\psi(z)$ being the Euler digamma function. The coherence length $\xi$ along the $c$-axis has the same functional form but different Fermi velocity $v_{Fe}$ (for simplicity we took scattering rate to be independent of the direction). In this ballistic limit both current and heat vertices are still local

$$J^c_y(q, i\omega) = 2e B_z(q, i\omega), \quad J^Q_y(q, i\omega) = -i\omega B_z(q, i\omega),$$

$B_z$ is obtained by replacing $\xi_{ab} \to \xi_c$. Having these expressions we perform summation over the Matsubara frequency in Eq. (S4) followed by an analytical continuation which gives for the thermal part of the Peltier coefficient

$$\alpha_{xy} = \frac{4e^2 H}{\pi c T} \sum_q B^2_z(q) B^2_y(q) \int \frac{d\omega \coth \frac{\omega}{2T} ((\Re L^R(q, \omega))^3$$

where $L^R$ is the retarded component of Eq. (S6). In the immediate vicinity of superconducting transition ($\xi^2_e \sim \omega / T \sim \ln(T / T_c) \ll 1$ so that one can safely approximate $\coth(\omega / 2T) \approx 2T / \omega$). Next, it is convenient to rescale all the momenta in units of coherence length $\xi_{ab} q_x = \kappa_x, \xi_{ab} q_y = \kappa_y, \xi_{ab} q_z = \kappa_z$ and pass to the spherical coordinates in the new momentum variable $\kappa$, namely $\kappa_x = \kappa \sin \theta \sin \phi, \kappa_y = \kappa \sin \theta \cos \phi, \kappa_z = \kappa \cos \theta$, which gives us

$$\alpha_{xy} = \frac{2}{\pi \ell_H^2 \xi_{ab}^2 c_e} \int \frac{d\omega}{\omega} \frac{\kappa^2 \sin \theta \sin \phi \cos \phi \sin^2 \theta \cos^2 \phi}{8 \pi^3} 16 D^4 \xi_{ab}^4 \kappa^4 \sin^4 \theta \sin^2 \phi \cos^2 \phi$$

$$(S11)$$

where magnetic length is $\ell_H = \sqrt{\frac{c}{eH}}$. As a final step we introduce dimensionless variables $\lambda = \ln(T / T_c), x =$...
\( \kappa^2, \ y = \pi \omega / 8T, \) which implies for the propagator
\[
\Re L^R = - \frac{1}{D} \frac{\lambda + x}{(\lambda + x)^2 + y^2}, \quad \Im L^R = - \frac{1}{D} \frac{y}{(\lambda + x)^2 + y^2}. \tag{S11}
\]
and use integrals
\[
\int_0^{2\pi} \cos^2 \phi \sin^2 \phi d\phi = \frac{\pi}{4}, \quad \int_0^\pi \sin^5 \theta d\theta = \frac{16}{15},
\int_{-\infty}^{+\infty} \frac{dy}{(1 + y^2)^3} = \frac{3\pi}{8}, \quad \int_0^\infty \frac{x^{5/2} dx}{(1 + x)^4} = \frac{5\pi}{16}
\]
to obtain Peltier coefficient for the range of scattering satisfying \( 1 \ll T \tau \ll \sqrt{T_c/(T - T_c)} \)
\[
\tilde{\alpha}_{xy} = \frac{k_B \epsilon}{4\pi \hbar} \frac{\xi_{ab}(0) T_c}{\ell_H^2} \frac{f(T\tau)}{\sqrt{\ln(T/T_c)}} \tag{S12}
\]
here \( \epsilon = \xi_{ab}/\xi_c \) is the anisotropy parameter and dimensionless function is
\[
f(z) = \frac{z}{\sqrt{3}} \left[ \psi\left(\frac{1}{2}\right) + \frac{1}{4\pi z} \psi'\left(\frac{1}{2}\right) - \psi\left(\frac{1}{2} + \frac{1}{4\pi z}\right) \right]. \tag{S13}
\]
Magnetization contribution \( cM_z/T \) is of the same form as \( \alpha_{xy} \) but opposite in sign and comes with the coefficient \( 1/6\pi \) so that an overall coefficient in \( \alpha_{xy} \) of Eq. (S2) is \( 1/12\pi \). Function \( f(T\tau) \) governs dependence of \( \alpha_{xy} \) on a scattering time which displays sharp growth by a factor of six followed by a rapid saturation at \( T\tau \gtrsim 1 \), see Fig. S3 for the illustration.