Flux Line Lattice Melting and the Formation of a Coherent Quasiparticle Bloch State in the Ultraclean URu_2Si_2 Superconductor

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We find that in the ultraclean heavy-fermion superconductor URu_2Si_2 ($T_{c0} = 1.45$ K) a distinct flux line lattice melting transition with outstanding characters occurs well below the mean-field upper critical fields. We show that a very small number of carriers with heavy mass in this system results in exceptionally large thermal fluctuations even at sub-Kelvin temperatures, which are witnessed by a sizable region of the flux line liquid phase. The uniqueness is further highlighted by an enhancement of the quasiparticle mean free path below the melting transition, implying a possible formation of a quasiparticle Bloch state in the periodic flux line lattice.

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URu_2Si_2 is a heavy-fermion superconductor [1,2], which has attracted much attention because of the so-called "hidden order" transition at 17.5 K, whose order parameter is still a mystery. In addition, recent studies using extremely clean single crystals having huge $\omega_c\tau$ values (where ω_c is the cyclotron frequency and τ is the scattering time) [3,4] reveal that the superconducting state with low transition temperature ($T_{c0} = 1.45$ K) embedded in the hidden order state is also uniquely unusual. According to several experiments [5], most of the carriers disappear below the hidden order transition, resulting in a very low carrier density (~ 0.02 electrons/U atom), reminiscent of semimetals such as bismuth and graphite. The electronic structure with such heavy low-density carriers hosts the exotic superconducting state, including a first-order transition at the upper critical fields H_{c2}, a possible chiral d-wave symmetry [4], and quasiparticle (QP) transport in the quantum limit [6].

Here we report that the small number of carriers with heavy mass and with extreme cleanness of URu_2Si_2 also give rise to an extraordinary vortex state, i.e., giant thermal fluctuations even at sub-Kelvin temperatures and a possible formation of a coherent QP Bloch state. Owing to the exceptionally few disorders we are able to observe the thermally driven melting transition from flux line (FL) lattice into the FL liquid well below the mean-field transition temperature $T_c(H)$. It is manifest by a sharp resistivity drop under a magnetic field, which is in clear contrast to the case in conventional low-T_c superconductors, where the resistivity drops at T_c. Despite the low T_c, it is analogous to the first-order melting transition observed at much higher temperatures in clean high-T_c cuprates [7–10]. It is quite remarkable that the melting transition continues to very high fields ($\sim 0.7H_{c2}$), which is in contrast to the case of cuprates where quenched disorder terminates the melting transition at low fields ($\lesssim 0.3H_{c2}$) and causes the disorder-driven glass transition from an ordered (Bragg glass) state to a highly disordered glass state [11,12]. Furthermore, an unusual change in the QP mean free path below the melting transition is observed, indicating that the quasiparticles are scattered less by the FL lattice than liquid, which has never been observed in any other type-II superconductors. This points to a possible formation of a novel Bloch-like state by the periodic FL lattice.

The ac resistivity is measured in a ^{3}He cryostat by the standard four-probe method with a current density J of 10^4 A/m^2 along the a axis at 17 Hz. The electric field E vs J characteristics are analyzed from ac-resistance data with three different excitations. No evidence for heating of the crystal is obtained down to ~ 0.7 K. The thermal conductivity κ is measured in a dilution refrigerator by a steady state method along the a axis.

In this study, we use a crystal having an exceptionally low residual resistivity $\rho_0 = 0.5$ $\mu\Omega$ cm and a large residual resistivity ratio RRR $= 670$ [inset of Fig. 1(a)], which attest the highest crystal quality currently achievable. The resistivity ρ in magnetic fields shown in Fig. 1 exhibits a "kink" or very sharp drop, which is even sharper than the transition width $\Delta T_{c0}(10\%–90\%) \approx 0.1$ K in zero field. The kink temperatures determined by the peak positions of $d\rho/dT$ [inset of Fig. 1(b)] are denoted by T_m (solid arrows). As shown in Fig. 2, the exponent α in the E-J characteristics, $E \propto J^\alpha$, exhibits a steep increase near T_m within a narrow temperature range.

The thermal conductivity κ also provides important information on the vortex states [Fig. 3]. In zero field,

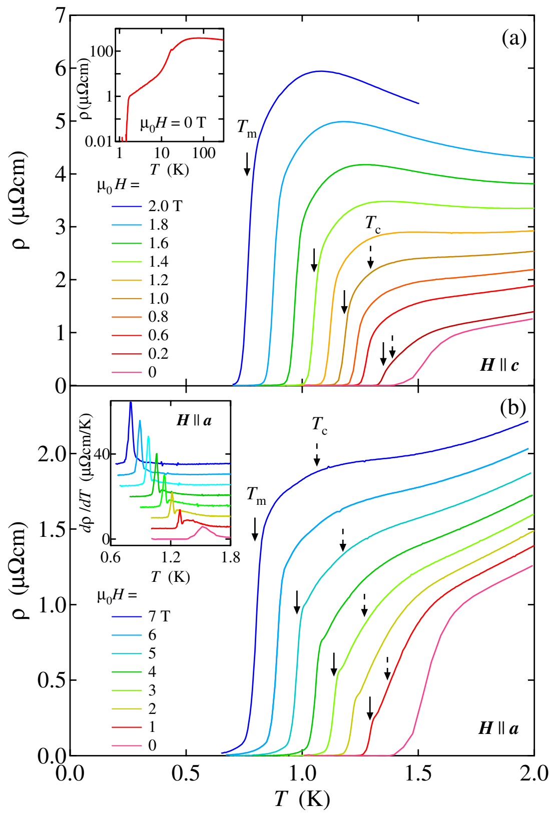

FIG. 1 (color online). Temperature dependence of the resistivity for (a) $\boldsymbol{H} \parallel c$ and (b) $\boldsymbol{H} \parallel a$. The very large magnetoresistance in the normal state stems from the compensation, i.e., essentially equal number of electrons and holes, $n_e = n_h$ [4]. The solid arrows indicate the melting transition T_m, which is defined as a peak of $d\rho/dT$ [inset of (b)]. The dashed arrows indicate the mean-field transition temperature T_c determined by the cusp of the thermal conductivity [see Fig. 3]. The inset of (a) shows the resistivity in zero field. The inset of (b) shows $d\rho/dT$ vs T. The data are vertically shifted.

κ/T increases with decreasing T and more rapidly below T_{c0}. The Wiedemann-Franz ratio L in the normal state near T_c is very close to the Sommerfeld value L_0 expected for the electronic contribution. Moreover, the phonon contribution κ_{ph}/T is reported to be less than 0.3 W/K^2 m around 0.8 K [13], which is much smaller than the observed κ/T. These indicate that in this temperature range, the electron contribution well dominates over the phonon contribution. The electronic heat conduction is described by $\kappa/T \sim N(0)v_F\ell$, where $N(0)$, v_F, and ℓ are the QP density of states, Fermi velocity, and QP mean free path, respectively. The enhancement of κ/T below T_{c0} is caused by a striking enhancement of ℓ due to the gap formation, which overcomes the reduction of $N(0)$ in the superconducting state, as observed in several strongly correlated electron systems [14,15]. This is a natural consequence of the more rapid reduction of the QP scattering rate than the $N(0)$ reduction, since the number of QPs and the number of QP

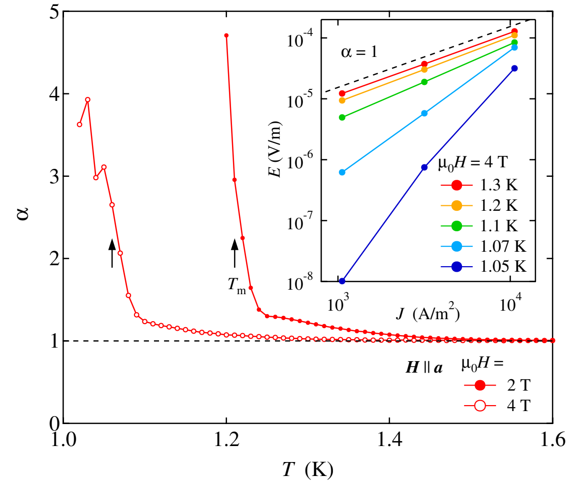

FIG. 2 (color online). Exponent α in $E \propto J^{\alpha}$, as a function of temperature for $\boldsymbol{H} \parallel a$ at 2 T and 4 T. The inset shows the E-J characteristics around T_m. The Ohmic resistivity $\alpha = 1$ is observed at high temperatures, and α increases gradually with approaching T_m and shows a sharp increase at T_m.

scatters are both reduced below T_{c0} in the electron-electron scattering. Under a magnetic field, however, κ/T begins to decrease below a distinct cusp (dashed arrows) as the temperature is lowered. This is an indication that $N(0)$ decreases below this cusp temperature. Indeed, according to recent theories [16], thermal conductivity has no fluctuation correction, in contrast to the resistivity, magnetic susceptibility, and specific heat which are subject to the

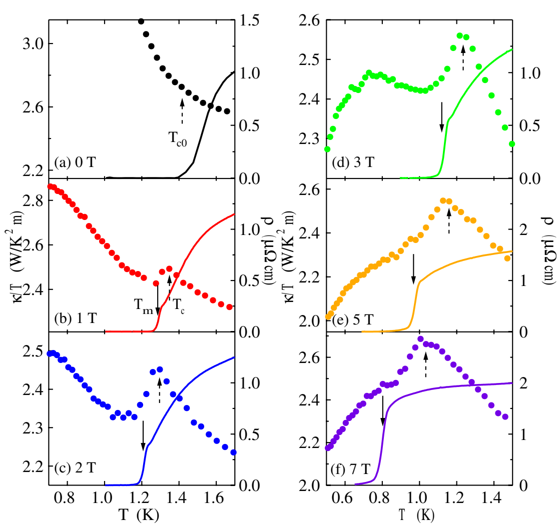

FIG. 3 (color online). Temperature dependence of the thermal conductivity divided by temperature κ/T (circles) in zero field (a) and in magnetic fields $\boldsymbol{H} \parallel a$ (b)–(f). The resistivity data at corresponding fields are also shown (solid lines). The dashed arrows indicate the temperature at which κ/T shows cusps, which correspond to the mean field $T_c(H)$. The solid arrows mark the melting temperature T_m [inset of Fig. 1(b)]. Below $\sim T_m$, κ/T becomes bigger than extrapolated values.

fluctuations. Therefore, it is natural to consider that the cusp temperature of κ/T corresponds to the mean-field transition temperature $T_c(H)$. The decrease of κ/T below $T_c(H)$ indicates that ℓ remains short under magnetic fields, which shows a clear contrast to the enhanced κ/T below $T_c(H)$ in, e.g., $CeCoIn_5$ [14]. Further lowering the temperature brings a second anomaly below which κ/T becomes bigger than that extrapolated from high temperatures. This second anomaly is located close to T_m (solid arrows) but far from $T_c(H)$, indicating that the QP scattering is dramatically changed at $\sim T_m$, which will be discussed later.

In Fig. 1, $T_c(H)$ determined by the thermal conductivity is shown by dashed arrows. It is obvious that $\rho(T)$ shows only a gradual decrease near $T_c(H)$, while a sudden drop occurs at T_m well below $T_c(H)$. We note that the difference between T_m and $T_c(H)$ becomes more pronounced at higher fields and exceeds 20% of T_c at 7 T [see Fig. 3(f)]. The features of the resistive transition of URu_2Si_2 bear striking resemblance to that of clean $YBa_2Cu_3O_7$, in which the sharp drop of the resistivity is observed in a linear scale at the melting transition without a sharp anomaly at $T_c(H)$ and the E-J characteristics become strongly non-Ohmic below T_m [7]. Based on these results, we conclude that the melting transition takes place at T_m [17].

The fundamental parameter which governs the strength of the thermal fluctuations is the Ginzburg parameter, $G_i = [\epsilon k_B T_c / H_c(0)^2 \xi_a^3]^2/2$, which measures the relative size of the thermal energy $k_B T_c$ and the condensation energy within the coherence volume [19,20]. Here $H_c = \Phi_0 / 2\sqrt{2}\pi\lambda_a\xi_a$ is the thermodynamic critical field, and λ_a and ξ_a are penetration and coherence lengths in the basal plane at $T = 0$ K, respectively. In zero field, the critical region where Gaussian fluctuation breaks down is given by $|T - T_c|/T_c < G_i$. Such a region is extremely small even in high-T_c cuprates. However, in magnetic fields sufficiently strong, the superconducting fluctuations acquire an effective one-dimensional (1D) character along the field direction. This reduction of the effective dimensionality increases the importance of fluctuations, resulting in a serious broadening of the resistive transition around $T_c(H)$, particularly in superconductors with large G_i [21]. Large G_i also leads to the reduction of T_m, extending the FL liquid region. The thermodynamic melting line for the 3D system is determined by

$$T_m - T_c(H) = 2y\left(\frac{H}{\tilde{H}_{c2}(0)}\right)^{2/3} \sqrt[3]{G_i/\epsilon^2}\, T_c(H) \qquad (1)$$

with $y \approx -7$ [20]. Here $\tilde{H}_{c2}(0)$ is a linear extrapolation of the initial slope of $H_{c2}(T)$ at T_{c0} to $T \rightarrow 0$ K.

Let us now quantitatively compare URu_2Si_2 with other systems. In conventional low-T_c superconductors, G_i ranges from 10^{-11} to 10^{-7}, while in $YBa_2Cu_3O_7$ G_i is as large as $\sim 10^{-2}$ [19]. Now, the penetration depth of URu_2Si_2 is unusually long ($\lambda_a \sim 1\ \mu$m according to μSR [22]), giving rise to a large $G_i \sim 3 \times 10^{-4}$. Such a long penetration depth is a natural consequence of the combination of a small Fermi surface (i.e., a low carrier density) with a large effective mass. Both these features are directly inferred from de Haas–van Alphen measurements [3] and are confirmed by a host of converging experimental evidence [5]. Thus, G_i is roughly increased by 5 orders of magnitude, leading to a sizable separation of T_m and $T_c(H)$ over a large portion of the phase diagram (see Fig. 4), as is the case in the high-T_c cuprates.

The solid lines in Figs. 4(a) and 4(b) represent the melting curves obtained from Eq. (1) with a single fitting parameter $G_i = 3.8 \times 10^{-4}$. This value is very close to the above estimate. These results lead us to conclude that the exceptionally large thermal fluctuations play an important role in URu_2Si_2 even at sub-Kelvin temperatures.

We here point out several unique features in URu_2Si_2. First, in high-T_c cuprates, the 2D pancake vortices are

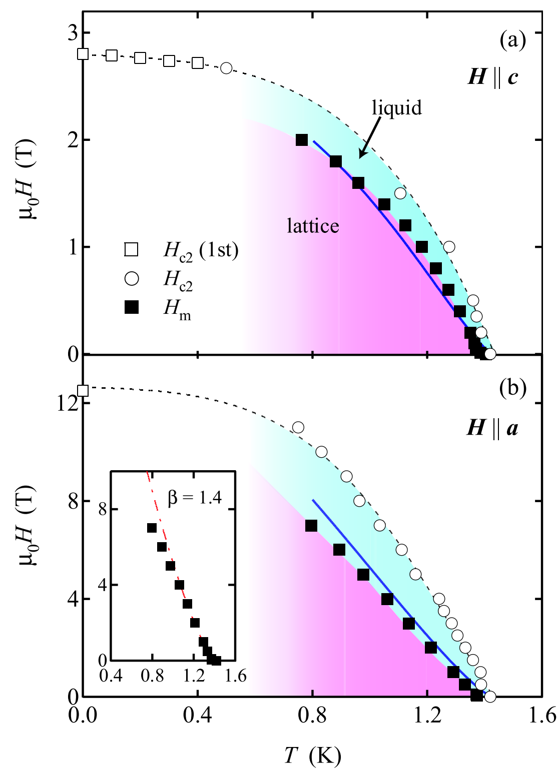

FIG. 4 (color online). H-T phase diagram of URu_2Si_2 determined by the present study for (a) $\boldsymbol{H} \parallel c$ and (b) $\boldsymbol{H} \parallel a$. Open symbols represent the mean-field H_{c2} lines. At low temperatures, this line becomes first order (open squares) [4]. The dashed lines are guides for the eyes. The solid squares represent the melting transition which is fitted by Eq. (1) (solid line). The FL liquid phase occupies a large portion in the H-T diagram for both field directions. The inset of (b) shows an expanded view of $H_m(T)$ near T_{c0} for $\boldsymbol{H} \parallel a$. The dash-dotted line is a fit to $H_m \propto (T_c - T)^\beta$ with $\beta = 1.4$. This β-value is consistent with that in $YBa_2Cu_3O_7$ [7].

weakly connected by the Josephson strings for $\boldsymbol{H} \parallel c$ [23], and the melting transition is accompanied by the strong decomposition of the FLs or "decoupling" along the c axis [24,25]. On the other hand, in URu_2Si_2 where the c axis coherence length is much longer than the lattice constant, a 3D melting transition to a line liquid is expected to occur. Second, the transition persists at least up to $\sim 0.7 H_{c2}(0)$, which is much higher than other systems; even in very clean $YBa_2Cu_3O_7$, $H_m(T)$ ceases to increase at $\sim 0.3 H_{c2}(0)$, above which the glass transition without sharp resistive drop is observed [12]. This implies that the present ultraclean URu_2Si_2 has remarkably few quenched disorders. Finally, according to the recent experiments [4], the first-order transition takes place at H_{c2} below ~ 0.4 K, owing to the strong Pauli paramagnetism, as shown in Fig. 4. Consequently, two first-order transition lines with different origins appear to coexist, making the H-T phase diagrams of URu_2Si_2 unprecedented. Exploring the detailed H-T diagram at lower temperatures is therefore intriguing.

The present ultraclean system may also provide important information of the QP transport in the vortex state, which has been a controversial issue [26,27]. The QP scattering is caused by Andreev scattering on the velocity field associated with the vortices, and a single FL acts as a strong scattering center. In our URu_2Si_2, the QP mean free path ℓ well exceeds 1 μm, 2 orders of magnitude longer than the intervortex distance at $\mu_0 H = 1$ T. In a naive picture, such a long ℓ would not be influenced by the melting transition. So the observed enhancement of κ/T below T_m is highly unusual. Although the full understanding of the unusual change of ℓ requires further studies, a possible explanation for this is that below T_m the nearly perfect FL lattice is formed, in which low energy QPs are described by Bloch wave function and are less scattered [28]. It should be noted that the enhancement of κ/T in the FL solid state has never been observed even in very clean $YBa_2Cu_3O_7$, implying that an ultraclean system is required for the formation of the QP Bloch state.

In summary, we provide strong evidence for thermal melting of a FL lattice to a FL liquid at sub-Kelvin temperatures in URu_2Si_2. The periodic FL lattice is suggested to form the QP Bloch state with long mean free path. The present results demonstrate that heavy-fermion superconductors may provide a new playground to study novel vortex matter physics as well as quasiparticle dynamics in the vortex state of type-II superconductors.

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[1] T. T. M. Palstra *et al.*, Phys. Rev. Lett. **55**, 2727 (1985).
[2] H. Amitsuka *et al.*, J. Magn. Magn. Mater. **310**, 214 (2007).
[3] H. Ohkuni *et al.*, Philos. Mag. B **79**, 1045 (1999).
[4] Y. Kasahara *et al.*, Phys. Rev. Lett. **99**, 116402 (2007).
[5] K. Behnia *et al.*, Phys. Rev. Lett. **94**, 156405 (2005).
[6] H. Adachi and M. Sigrist, arXiv:0710.3110.
[7] W. K. Kwok *et al.*, Phys. Rev. Lett. **69**, 3370 (1992).
[8] E. Zeldov *et al.*, Nature (London) **375**, 373 (1995).
[9] A. Schilling *et al.*, Nature (London) **382**, 791 (1996).
[10] U. Welp, J. A. Fendrich, W. K. Kwok, G. W. Crabtree, and B. W. Veal, Phys. Rev. Lett. **76**, 4809 (1996).
[11] In some low-T_c superconductors, a disorder-driven transition from ordered to disordered glass has been observed. See, e.g., Y. Paltiel *et al.*, Nature (London) **403**, 398 (2000).
[12] T. Nishizaki, T. Naito, S. Okayasu, A. Iwase, and N. Kobayashi, Phys. Rev. B **61**, 3649 (2000).
[13] K. Behnia *et al.*, Physica (Amsterdam) **196C**, 57 (1992).
[14] Y. Kasahara *et al.*, Phys. Rev. B **72**, 214515 (2005).
[15] Y. Kasahara *et al.*, Phys. Rev. Lett. **96**, 247004 (2006).
[16] D. R. Niven and R. A. Smith, Phys. Rev. B **66**, 214505 (2002); S. Vishveshwara and M. P. A. Fisher, Phys. Rev. B **64**, 134507 (2001).
[17] To obtain thermodynamic evidence of the first-order transition, we have also tried the local magnetization measurements by using a small Hall sensor of HgCdTe with an active area of 100×100 μm^2, which is placed on the surface of URu_2Si_2. However, the jump of the magnetization is not resolved within experimental resolution. The jump ΔB associated with the first-order melting transition is given as, $\Delta B[G] \approx (1.5 \times 10^{-6} \epsilon) T_m[K] \times (B_m[G])^{1/2}$, where $\epsilon = H_{c2}^a / H_{c2}^c$ is the anisotropy parameter [18]. Using $\epsilon \approx 4$ for URu_2Si_2, ΔB is estimated to be 1.8×10^{-4} G at $T_m = 1.4$ K and $B_m = 500$ G, which is indeed found to be much smaller than our resolution $\sim 10^{-2}$ G.
[18] M. J. W. Dodgson, V. B. Geshkenbein, H. Nordborg, and G. Blatter, Phys. Rev. Lett. **80**, 837 (1998).
[19] G. Blatter, M. V. Feigel'man, V. B. Geshkenbein, A. I. Larkin, and V. M. Vinokur, Rev. Mod. Phys. **66**, 1125 (1994).
[20] A. Larkin and A. Varlamov, *Theory of Fluctuations in Superconductors* (Oxford Univversity Press, Oxford, 2005).
[21] R. Ikeda, T. Ohmi, and T. Tsuneto, J. Phys. Soc. Jpn. **58**, 1377 (1989).
[22] A. Amato, Rev. Mod. Phys. **69**, 1119 (1997).
[23] J. R. Clem, Phys. Rev. B **43**, 7837 (1991).
[24] T. Shibauchi *et al.*, Phys. Rev. Lett. **83**, 1010 (1999).
[25] M. B. Gaifullin, Y. Matsuda, N. Chikumoto, J. Shimoyama, and K. Kishio, Phys. Rev. Lett. **84**, 2945 (2000).
[26] Y. Matsuda, K. Izawa, and I. Vekhter, J. Phys. Condens. Matter **18**, R705 (2006).
[27] I. Vekhter and A. Houghton, Phys. Rev. Lett. **83**, 4626 (1999).
[28] M. Franz and Z. Tesanovic, Phys. Rev. Lett. **84**, 554 (2000); K. Yasui and T. Kita, Phys. Rev. Lett. **83**, 4168 (1999); M. R. Norman, A. H. MacDonald, and H. Akera, Phys. Rev. B **51**, 5927 (1995).