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Strong coupling between localized 5f moments and itinerant quasiparticles in the ferromagnetic superconductor UGe_2*

Wen Zhang(张文), Yi Liu(刘毅)†, Xiaoying Wang(王小英), Yun Zhang(张云), and Donghua Xie(谢东华)

Science and Technology on Surface Physics and Chemistry Laboratory, Jiangyou 621908, *China*

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The heavy fermion physics arises from the complex interplay of nearly localized 4f/5f electrons and itinerant band-like ones, yielding heavy quasiparticles with an effective mass about 100 times (or more) of the bare electrons. Recently, experimental and theoretical investigations point out a localized and delocalized dual nature in actinide compounds, where itinerant quasiparticles account for the unconventional superconductivity in the vicinity of a magnetic instability. Here we report the strong coupling between localized 5f moments and itinerant quasiparticles in the ferromagnetic superconductor UGe_2. The coupling is nearly antiferromagnetic. As embedded in the ferromagnetic matrix of localized 5f moments below $T_C \approx 52$ K, this coupling leads to short-range dynamic correlations of heavy quasiparticles, characterized by fluctuations of magnetic clusters. Those cluster-like spins of itinerant quasiparticles show a broad hump of magnetization at $T_X \approx 28$ K, which is typical for the spin-glass freezing. Thus, our results present the direct observation of itinerant quasiparticles coexisting with localized 5f moments by conventional magnetic measurements, providing a new route into the coexistence between ferromagnetism and superconductivity in heavy fermion systems.

Keywords: UGe_2, ferromagnetic superconductor

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1. Introduction

In heavy fermion materials, Kondo lattice is a prototypical model to describe interacting localized 4f/5f moments coupled antiferromagnetically to the conduction electron sea.[1] As a consequence of the collective hybridization, the system can approximately be described by two renormalized components, i.e., itinerant heavy quasiparticles (so-called the Kondo liquid) and unhybridized localized moments.[2] At low temperatures, itinerant quasiparticles due to the hybridization are believed to lead to a rich variety of emergent quantum phenomena such as unconventional superconductivity.

The ferromagnetic superconductor UGe_2 is an exotic example displaying both local-moment and heavy-mass itinerant behavior due to the peculiar dual character of 5f electrons.[3–9] This compound is unique, since pronounced localized 5f electrons are mainly responsible for bulk properties such as the giant magnetocrystalline anisotropy with the easy axis aligning along the a direction and the relatively large ferromagnetic moment, $M_U \sim 1.5\mu_B$, whereas superconductivity guaranteed by itinerant quasiparticles is known to occur within the ferromagnetic (FM) state, in a limited pressure range between 0.9 GPa and 1.5 GPa.[8,10–12] As deduced from the electrical transport properties above the Curie point $T_C \approx 52$ K, UGe_2 is a case with combined crystal-field and Kondo-lattice effects, supporting the idea of partial localization of 5f electrons.[8] On the other hand, the hybridized (itinerant) part of 5f states gives distinct signatures for the anomalies at around the characteristic temperature $T_X \approx 28$ K. Although it has been theoretically suggested that T_X corresponds to the formation of a simultaneous charge- and spin-density wave,[13] no evidence of such a wave has ever been detected. As the pressure increases, T_X decreases monotonically and finally disappears at $P_X \approx 1.2$ GPa, where T_{SC} of the superconductivity is highest, ~ 0.7 K.[14] In this manner, superconductivity is closely related to the vanishing of T_X. Despite intense investigations, the internal connections between T_X and itinerant quasiparticles remain unresolved.

Recently, the change at T_X is demonstrated to be a crossover at ambient pressure, instead of a sharp thermodynamic phase transition, and no discontinuity has been detected in heat-capacity, thermal-expansion, neutron scattering, nuclear magnetic resonance, and nuclear quadrupole resonance measurements.[14–17] Moreover, T_X at 1.2 GPa is established to be a first-order phase transition, which is apparently in contrast with the theoretical prediction that quantum phase transitions should be of second order.[18] Driven by quantum fluctuations, such a first-order phase transition is usually pre-empted by the formation of an inhomogeneous magnetic phase.[19] In this paper, we show the strong coupling between localized 5f moments and itinerant quasiparticles in UGe_2 by conventional magnetic measurements. Our results clearly indicate the emergence of short-range dynamic correlations of itinerant quasiparticles embedded in the FM matrix of localized 5f moments. Further, these itinerant quasiparticles form a cluster-glass state

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†Corresponding author. E-mail: liuyifat@163.com

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below T_X, shedding new light on the coexistence between ferromagnetism and superconductivity.

2. Experiment

High-quality single crystals of UGe_2 were grown by Czochralski pulling method in a tetra-arc furnace under protective argon atmosphere. A small oriented cubic lump was cut from the cleaved ingot, and subsequently annealed at 1073 K under ultrahigh vacuum for a week. The quality of the specimen was verified by means of x-ray diffractions. The residual resistivity ratio $\rho(300\ \mathrm{K})/\rho(2\ \mathrm{K})$ amounts to 435, confirming the high quality of the single crystals. Magnetic measurements were performed using a quantum design magnetic property measurement system (MPMS), in the temperature range from 2 K to 300 K and in magnetic fields up to 50 kOe. In the measurements of field-cooling (FC) magnetization, data were recorded on cooling in a magnetic field of 5 kOe. In the measurements of thermal remanent magnetization (TRM), data were recorded on warming in zero field, after an FC process in 5 kOe. In the measurements of ac susceptibility, a dc bias field of 1 kOe was always applied, and an ac excitation of 10 Oe was used, operating at 10 Hz, 100 Hz, 1 kHz, and 10 kHz, respectively. Both dc and ac magnetic fields in measurements were applied along the *a* direction, the easy axis of magnetization.

3. Results and discussion

Figure 1 shows the FC magnetization and the subsequent TRM of UGe_2 as a function of temperature. FC magnetization was measured on cooling in 5 kOe applied along the *a* axis, which is far above the saturation field, ~ 0.5 kOe, ensuring the monodomain of the sample. After this FC process, the sample was slowly heated in zero field and TRM was synchronously recorded on warming. As the temperature increases, TRM drops rapidly and then shows an upturn at $T_X \approx 28$ K. Followed by the turn point, TRM shows a small negative magnetic moment, similar to $-0.05\mu_B$, which varies slowly with temperature and finally disappears above $T_C \approx 52$ K. Herein, the small negative value of TRM below T_C is primarily attributed to the magnetic moment of itinerant quasiparticles, as suggested in the dualism of 5f electrons. In comparison with the FC magnetization, $\sim 1.5\mu_B$, itinerant quasiparticles carry a really small magnetic moment, indicating that bulk magnetic properties are mainly governed by localized 5f moments. It is noted that our TRM results provide the first detection of itinerant quasiparticles by conventional magnetic measurements. The negative sign of TRM just below T_C justifies an antiferromagnetic (AFM) coupling between localized 5f moments and itinerant quasiparticles. In a Kondo lattice, the AFM coupling is easily understood, which stems from the fact that localized moments are completely or partly screened by conduction electrons with antiparallel spins. Surprisingly, these results are in full agreement with muon spin relaxation and neutron scattering measurements.[11,20,21] As the authors of these papers claim, comparison among bulk magnetization, neutron form factor data, and positive muon spin rotation/relaxation results provides a really small magnetic moment of itinerant electrons, $\sim 0.02\mu_B$, relatively isotropic and probably in antiparallel alignment with those localized moments.

In TRM measurements, it is necessary to check the real applied magnetic field in the magnetometer, since a residual field of typically ± 5 Oe usually remains in the superconducting magnet of MPMS. Hence, we repeat TRM measurements in ± 10 Oe to eliminate the confusion from the residual field, as shown in Fig. 1. As expected, the value of TRM is slightly different in those small magnetic fields, but the shape of all TRM curves is almost the same, invariably manifesting the small negative magnetic moment from itinerant quasiparticles. A subsequent re-cooling process always follows closely after each TRM measurement, but no such negative signals can be observed. Therefore, the negative signal is not caused by the trapped field in the coil of the superconducting magnet.

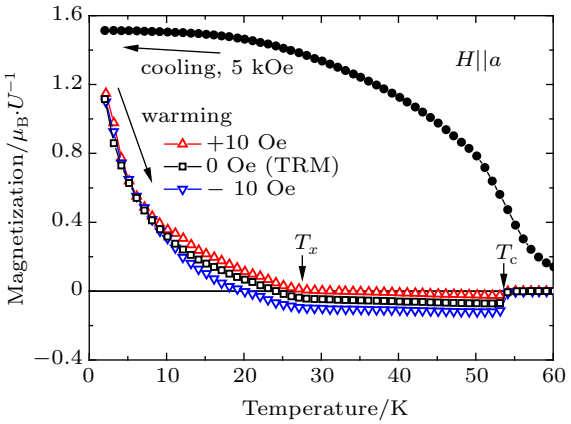


Fig. 1. (color online) Temperature dependences of the FC magnetization and the subsequent TRM of a UGe_2 single crystal. TRMs were measured on warming in zero field and ± 10 Oe, after an FC process in 5 kOe. All magnetic fields were applied along the easy *a* axis.

We now emphasize the importance of TRM measurements. In the dualism of 5f electrons, UGe_2 has to be viewed schematically as a two-subset electronic system, where localized 5f electrons coexist with itinerant quasiparticles. Since the contribution of itinerant quasiparticles is weak in bulk properties, it is commonly difficult to exactly tell apart their behavior by magnetic measurements. However, in a different way, TRM tries to approach the zero-field limit of the FM monodomain of localized 5f electrons, and then the AFM coupling in some senses induces a detectable component from itinerant quasiparticles. Both components of localized and itinerant 5f electrons were apparently identified, namely $\sim$

$1.5\mu_B$ for localized 5f electrons and similar to $-0.05\mu_B$ for itinerant quasiparticles, respectively. Most importantly, the negative sign of itinerant quasiparticles indicates antiferromagnetic correlations with localized 5f moments rather than the external magnetic fields, although the ferromagnetic order is usually fixed by the prior magnetic field. As a matter of fact, because spins of both localized and itinerant electrons tend to align themselves along the magnetic field, the AFM coupling makes the system intrinsically frustrated. As a consequence, no long-range order of itinerant quasiparticles has been established and short-range dynamic correlations of a spin-glass type may occur in such a magnetic frustrated system instead.

To further demonstrate the novel features of itinerant quasiparticles, we have measured the temperature dependences of FC magnetization in rotating magnetic fields within the ac plane, as shown in Fig. 2. The total magnetization can be expressed as $M(\theta) = M_{\rm loc}\cos\theta + M_{\rm itin}$, where $M_{\rm loc}$ indicates the Ising-like magnetic moment of localized 5f electrons, θ the angle between the direction of magnetic fields and the easy a axis, and $M_{\rm itin}$ relatively isotropic from itinerant quasiparticles. Apparently, the angle-dependent $M(\theta)$ is pronounced with a maximum at $\theta = 0^\circ (H\|a)$, and as θ is rotated towards 90°, $M(\theta)$ decreases continuously. By the fits of experimental data, we have plotted the deduced $M_{\rm itin}$ as a function of temperature, as shown in Fig. 2. At low temperatures, $M_{\rm itin}$ is small and negative, and its magnitude shows a hump-like maximum near T_X in the temperature dependence. The maximum value of the hump, approximately $-0.03\mu_B$, gives the best evaluation of itinerant quasiparticles in all hitherto existing experiments, which is precisely in accordance with our previous TRM measurements. In the spin-glass scenario, the broad hump at T_X indicates the collective freezing of itinerant quasiparticles, which is thermally activated, characterized by short-range dynamic correlations of cluster-like spins.[22]

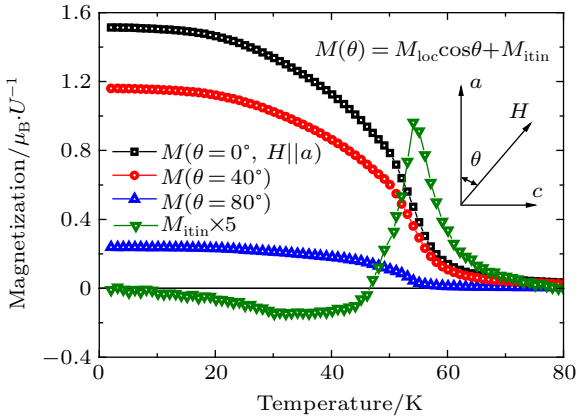


Fig. 2. (color online) Temperature dependences of the FC magnetization, and the itinerant-quasiparticle magnetization of UGe_2. The data for itinerant quasiparticles exaggerated for clarity were deduced from $M(\theta) = M_{\rm loc}\cos\theta + M_{\rm itin}$, where $M_{\rm loc}$ is the Ising-like magnetic moment of localized 5f electrons and $M_{\rm itin}$ relatively isotropic from itinerant quasiparticles.

The spin-glass behavior of itinerant quasiparticles is clearly reflected in the frequency-dependent magnetic ac susceptibility of UGe_2, as shown in Fig. 3. In all measurements, a dc bias field of 1 kOe was always applied, which is above the saturation field ensuring the monodomain from localized 5f moments. Both dc and ac magnetic fields were applied along the easy a axis. Besides the sharp peak at T_C, ac susceptibility shows at T_X another hump-like peak whose position varies with frequency and approaches 28 K in the static limit, as denoted by the anomaly of a shoulder-like maximum in zero-field-cooling magnetization at 0.1 kOe. In the inset, we show the frequency dependence of T_X exhibiting the Arrhenius relation $f = f_0\exp(-E_a/k_BT)$ with the activation energy $E_a \approx 725$ K and $f_0 \approx 4\times10^{11}$ Hz. This result signals the collective freezes of short-range magnetic correlations. In the frequency dependence of T_X, the parameter of $\Delta T_X/[T_X\Delta(\log f)] \approx 0.1$ can be evaluated. This value is slightly larger than those found for canonical spin glasses (0.005—0.06), yet it is below typical values of superparamagnets (0.3).[23] Furthermore, it is noteworthy that the broad hump at T_X points out a distribution of cluster-like spins for the unusual short-range order. The wide distribution in the size and/or the coupling strength of magnetic clusters is supported by the frequency dispersion of ac susceptibility. Due to the wide distribution of the energy of fluctuating moments from itinerant quasiparticles, formation of such a cluster-glass state is expected when the thermal energy is comparable with the energy of fluctuating moments.

The short-range order of itinerant quasiparticles can be understood in the framework of quantum phase transitions (QPTs) in metallic ferromagnets. When the control parameter

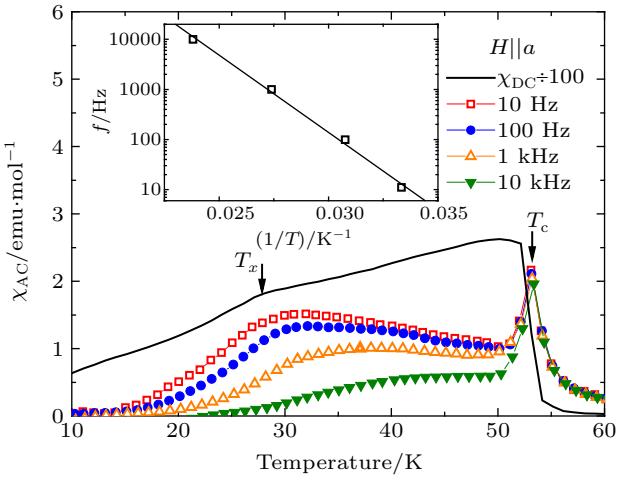


Fig. 3. (color online) The frequency-dependent ac susceptibility of UGe_2 measured as a function of temperature. The amplitude of ac excitation is 10 Oe, operating at 10 Hz, 100 Hz, 1 kHz, and 10 kHz, respectively. In ac susceptibility measurements, a dc bias field of 1 kOe was always applied. Both dc and ac magnetic fields were parallel with the easy a axis. For comparison, zero-field-cooling magnetization in 100 Oe is also plotted, divided by a factor of 100. The inset shows the frequency dependence of T_X in an Arrhenius plot fitted by a full line.

is tuned towards a putative quantum critical point (QCP), QPTs may be discontinuous or continuous, and even a state with glass-like spin dynamics is possible.[24] For example, in a clean system CeFePO, the ground state is a short-range ordered state, although it is very close to an FM instability.[25] In view of recent advances on QPTs, we conclude that there are two different FM QCPs in UGe_2. As the pressure increases, the disappearance of T_C at 1.5 GPa corresponds to the FM QCP of localized 5f electrons, whereas T_X is likely to vanish at 1.2 GPa, a putative FM QCP associated with the glass-like short-range order from itinerant quasiparticles. Additionally, T_X (~ 3 K) is still observable at 1.33 GPa in muon spin relaxation measurements,[21] providing evidence for the tail of the quantum criticality with spin-glass freezings.

4. Conclusion

We have studied the strong coupling between localized 5f moments and itinerant quasiparticles by conventional magnetic measurements in the ferromagnetic superconductor UGe_2. In both the thermal remanent magnetization and the field-cooling magnetization in rotating magnetic fields, this antiferromagnetic coupling leads to the emergence of short-range dynamic correlations, characterized by magnetic fluctuations of cluster-like spins. Since itinerant quasiparticles are immersed in the ferromagnetic matrix of localized 5f moments below T_C, they show a detectable small negative magnetic moment, about $-0.03\mu_B$. Moreover, the collective behavior of cluster-like spins has been observed below T_X, signifying that the unusual short-range order is indeed a cluster-glass state of itinerant quasiparticles. The spin-glass scenario consequently explicitly rules out the possibility of a thermodynamic phase transition at T_X at ambient pressure. As the pressure increases, T_X gradually decreases, nevertheless, the glass-like state persists at pressures at least up to 0.95 GPa, where itinerant quasiparticles are still characterized by a small magnetic moment, $\leq 0.03\mu_B$.[21] Close to the putative quantum critical point, ~ 1.2 GPa, itinerant quasiparticles show a magnetic moment of about $-0.2\mu_B$ at 1.4 GPa, calculated from the difference between the bulk magnetization of $0.9\mu_B$ and the neutron data of $1.1\mu_B$.[11,20,21] Such a value is much more prominent than that at ambient pressure, favoring an appreciable increase in the density of charge carriers and a sudden reconstruction of the Fermi surface, where a sharp first-order phase transition is likely to happen. The conjecture is verified by the detected first-order $T_X(P)$ boundary line approaching the critical point of $P_X \approx 1.2$ GPa.[26]

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References

[1] Stewart G R 1984 *Rev. Mod. Phys.* **56** 755
[2] Yang Y F 2016 *Rep. Prog. Phys.* **79** 074501
[3] Pfleiderer C 2009 *Rev. Mod. Phys.* **81** 1551
[4] Moore K T and Laan G V D 2009 *Rev. Mod. Phys.* **81** 235
[5] Sato N K, Aso N, Miyake K, Shiina R, Thalmeier P, Varelogiannis G, Geibel C, Steglich F, Fulde P and Komatsubara T 2001 *Nature* **410** 340
[6] Zwicknagl G and Fulde P 2003 *J. Phys. -Condens. Matter* **15** S1911
[7] Durakiewicz T, Joyce J J, Wills J M and Batista C D 2006 *J. Phys. Soc. Jpn.* **75** 39
[8] Troć R, Gajek Z and Pikul A 2012 *Phys. Rev. B* **86** 224403
[9] Fujimori S I, Takeda Y, Okane T, Saitoh Y, Fujimori A, Yamagami H, Haga Y, Yamamoto E and Ōnuki Y 2016 *J. Phys. Soc. Jpn.* **85** 062001
[10] Saxena S S, Agarwal P, Ahilan K, Grosche F M, Haselwimmer R KW, Steiner M J, Pugh E, Walker I R, Julian S R and Monthoux P 2000 *Nature* **406** 587
[11] Kernavanois N, Grenier B, Huxley A, Ressouche E, Sanchez J P and Flouquet J 2001 *Phys. Rev. B* **64** 174509
[12] Ōnuki Y, Ukon I, Yun S W, Umehara I, Satoh K, Fukuhara T, Sato H, Takayanagi S, Shikama M and Ochiai A 1992 *J. Phys. Soc. Jpn.* **61** 293
[13] Watanabe S and Miyake K 2002 *J. Phys. Soc. Jpn.* **71** 2489
[14] Pfleiderer C and Huxley A D 2002 *Phys. Rev. Lett.* **89** 207201
[15] Hardy F, Meingast C, Taufour V, Flouquet J, Löhneysen H V, Fisher R A, Phillips N E, Huxley A and Lashley J C 2009 *Phys. Rev. B* **80** 174521
[16] Huxley A, Sheikin I, Ressouche E, Kernavanois N, Braithwaite D, Calemczuk R and Flouquet J 2001 *Phys. Rev. B* **63** 144519
[17] Kotegawa H, Kawasaki S, Harada A, Kawasaki Y, Okamoto K, Zheng G Q, Kitaoka Y, Yamamoto E, Haga Y and Onuki Y 2002 *J. Phys. -Condens. Matter* **15** S2043
[18] Löhneysen H V, Rosch A, Vojta M and Wölfle P 2007 *Rev. Mod. Phys.* **79** 1015
[19] Conduit G J, Green A G and Simons B D 2009 *Phys. Rev. Lett.* **103** 207201
[20] Kuwahara K, Sagayama H, Iwasa K, Kohgi M, Haga Y, Ōnuki, Kakurai K, Nishi M, Nakajima K, Aso N and Uwatoko Y 2002 *Physica B* **312–313** 106
[21] Sakarya S, Gubbens P C M, Yaouanc A, Réotier P D D, Andreica D, Amato A, Zimmermann U, Dijk N H V, Brück E and Huang Y 2010 *Phys. Rev. B* **81** 024429
[22] Binder K and Young A P 1986 *Rev. Mod. Phys.* **58** 801
[23] Mydosh J A 1993 *Spin Glasses: An Experimental Introduction* (London: Taylor & Francis)
[24] Brando M, Belitz D, Grosche F M and Kirkpatrick T R 2016 *Rev. Mod. Phys.* **88** 025006
[25] Lausberg S, Spehling J, Steppke A, Jesche A, Luetkens H, Amato A, Baines C, Krellner C, Brando M and Geibel C 2012 *Phys. Rev. Lett.* **109** 216402
[26] Taufour V, Aoki D, Knebel G and Flouquet J 2010 *Phys. Rev. Lett.* **105** 217201