Spin Gap in Heavy Fermion UBe$_{13}$

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Muon spin rotation ($\mu^7$SR) experiments have been carried out in the heavy fermion compound UBe$_{13}$ in the temperature range from 0.025 K to 300 K and in magnetic fields up to 7 T, supported by resistivity and magnetization measurements. The $\mu^7$SR spectra are characteristic of formation of a spin polaron which persists to the lowest temperature. A gap in the spin excitation spectrum of f-electrons opens up at about 180 K, consistent with anomalies discovered in resistivity, heat capacity, NMR and optical conductivity measurements of UBe$_{13}$.

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In strongly correlated metallic materials, the interplay between local spins and itinerant electrons determines the spin fluctuation spectrum. In a 3d-electron system, such a spectrum emerges as a result of a strong dominance of the Fermi energy over magnetic energy. By contrast, in f-electron itinerant systems the Fermi energy is heavily renormalized down to the scale of the magnetic energy, resulting in a strong influence of the spin dynamics and spectral weight of spin fluctuations on transport, magnetic and thermodynamic properties of a system. A depletion of spectral weight upon cooling may indicate the opening of a gap (or pseudogap) in the spectrum of spin fluctuations.

The concept of a pseudogap has recently become essential for understanding strongly correlated electron systems. Particularly interesting is the opening of a gap in a system of f-electron spin excitations. At low temperature, such systems display a continuous transition from an array of uncorrelated local magnetic moments to a Fermi-liquid (FL) phase in which f electrons are strongly hybridized with conduction band electrons. As a result, f electrons not only create a sharp resonance on the Fermi level, which gives rise to a large effective mass $m^*$ of quasiparticles, but also transfer their magnetic entropy to the Fermi surface. In this case a gap may open up as in URu$_2$Si$_2$ at $T = 17.5$ K. In this Letter, we present experimental results that are explained by a spin gap opening in UBe$_{13}$ at $T = 180$ K.

UBe$_{13}$ crystallizes in the cubic NaZn$_{13}$ structure with lattice constant $a = 1.025$ nm. The U atoms form a simple cubic sublattice with a rather large U-U spacing, suggesting strong hybridization with the itinerant carriers. Unlike in other uranium-based HF compounds, there is no evidence for static magnetic order in UBe$_{13}$. In UBe$_{13}$, superconductivity (SC) arises from a paramagnetic (PM) metallic phase as a cooperative phenomenon involving heavy quasiparticles that form pairs. At high temperature, the susceptibility $\chi$ exhibits a Curie-Weiss behavior with $\mu_{\text{eff}} \approx 3.36 \mu_B$, which deviates from this law below 200 K and levels off below 20 K.

Interest in UBe$_{13}$ is reinforced by various observations of non-Fermi-liquid (NFL) behavior, often explained by the proximity of quantum critical point, which challenges the validity of the quasiparticle concept. However, various experiments indicate a dominant role of quasiparticles, although the specific quantum states that might replace the Fermi liquid remains unclear.

Quite remarkably, FL behavior is restored by application of a strong magnetic field. Another remarkable feature which requires explanation is that while at low temperature UBe$_{13}$ displays coherent quasiparticle behavior, at higher temperature it reveals an incoherent metallic state dominated by spin-flip scattering. Both NFL and loss of coherence phenomena cannot be explained by quadrupolar Kondo effect. Furthermore, energy band calculations produce an $m^*$ value at least an order of magnitude too low. Moreover, the band theory fails to explain the loss of coherence.

In order to identify a fundamental electronic state consistent with all the crucial experimental results, different polaron models have been proposed. Although both electronic and magnetic polaron models may account for NFL behavior and the loss of coherence and produce a correct $m^*$, the former fails to explain the strong influence of a magnetic field. By contrast, a spin polaron (SP) model claims to establish an electronic state which may help to reconcile theoretical treatment with the experiment. In particular, the SP model offers a straightforward explanation of a major puzzle of UBe$_{13}$—a remarkable magnetoresistivity (MR) — which can hardly find explanation within any other
FIG. 1: Frequency spectrum of muon spin precession in UBe$_{13}$ in a transverse magnetic field $B = 1$ T at $T = 150$ K. Inset: same spectrum in the time domain in a rotating reference frame. The two-frequency precession characteristic of a muon-electron hyperfine coupled state is evident in both domains.

Both MR and magnetostriction indicate that the carrier number is a strong function of magnetic field as if “the carrier is released by $B$” [11]. The remarkable sensitivity of the electron transport to the magnetization (an order of magnitude stronger than in hole-doped manganites) shows that carrier localization into SP, and its release by a magnetic field, may be a missing key ingredient of HF models. A SP model [16] predicts that the narrowness of the conduction band may cause the carriers to self-trap as SP. Recent observation of SP in magnetoresistive Lu$_2$V$_2$O$_7$ [17] supports this model. In this Letter, we present observation of SP in UBe$_{13}$. It is this observation which allows detection of a spin gap.

Single crystals for the current studies display lattice constants, resistivity behavior, $T_c$ and effective magnetic moments consistent with literature data. Time-differential $\mu^+\text{SR}$ experiments [18] using 100% spin-polarized positive muons implanted into these samples were carried out on the M15 muon channel at TRIUMF using the HiTime and DR spectrometers. At high temperature, in magnetic fields $B$ transverse to the initial muon polarization, Fourier transforms of the $\mu^+\text{SR}$ time spectra exhibit a single line at the muon frequency $\nu_\mu = \gamma_\mu B/2\pi$ (where $\gamma_\mu = 2\pi \times 135.53879$ MHz/T is the muon magnetogyric ratio), which coincides with that detected in a CaCO$_3$ reference sample used for independent measurements of $B$. However, below $T^* \approx 180$ K such simple spectra change abruptly to reveal a characteristic doublet (Fig. 1) which persists down to the lowest temperature (Fig. 2).

Previous observations of such two-line spectra [19] prompted a suggestion of two inequivalent sites occupied by positive muons in UBe$_{13}$. However, the two lines do not follow the temperature dependence of the magnetization, which indicates that the muon does not stay bare and does not act as a local magnetometer. Instead, while one line goes up in frequency the other goes down as temperature varies — a signature of a muon-electron bound state [19]. Similarly, if the muon stayed bare, both lines’ frequencies should stay constant as the susceptibility levels off below 20 K, which contradicts the experiment. Furthermore, a two-site assignment suggests a temperature independent strict 1:2 muon site occupation ratio [19] while experiment shows a temperature dependent amplitude ratio well below 0.5. Finally, the departure from linear magnetic field dependences of the two frequencies allows one to rule out possible Knight shifts within the bare muon scenario.

Instead, the doublet on Fig. 1 and Fig. 2 is a fingerprint of a coupled $\mu^+e^-$ spin system in high transverse field [17, 19, 20, 24]: the two lines correspond to two muon spin-flip transitions between states with electron spin orientation fixed, the splitting between them being determined by the muon-electron hyperfine interaction $A$ [20]. In a magnetic system, an electron’s energy depends strongly on the magnetization, with the minimum energy being achieved by FM ordering [25]. Then the strong exchange interaction $J$ between a carrier and local f(d) moments can cause electron localization into a ferromagnetic (FM) “droplet” over the extent of its wavefunction (typically, the first coordination sphere) in a PM (or AF) sea [26]. This charge carrier, accompanied by reorientations of local spins to form its immediate FM environment, together behave as a single quasiparticle with a giant spin $S$ — a spin polaron [25, 26]. In the process of electron localization into a SP in a metal the exchange energy gain upon transition from the PM to the FM state is opposed by the increase of the electron kinetic energy (the entropy term due to ordering within the SP becomes
significant only at very high $T$) \[20, 22, 23\].

High magnetic field destroys the SP because in high $B$ the spins are already polarized, so that the exchange coupling of the carrier with these spins offers no energy advantage to compensate the increase in kinetic energy due to localization. Application of a magnetic field thus releases the carrier from SP into the conduction band — a process which offers not only an explanation of the huge negative MR but also reveals the reason why the carrier number is a strong function of magnetic field in UBe$_{13}$.

Such SP states with the electron confined in a $R \approx 0.2 - 0.5$ nm FM “droplet” attached to a positive muon are found in strongly correlated insulators \[21, 27\], semiconductors \[20, 21, 24\] and metals \[22, 23\]. In metals, an itinerant SP is captured by the muon to exhibit the characteristic $\mu^+e^-$ hyperfine splitting \[22, 23\] through the frequency splitting $\Delta \nu$ between two SP lines (Fig. 3). Within a mean field approximation, $\Delta \nu$ follows a Brillouin function \[20, 22, 23\]. For $g \mu_B B \ll k_B T$,

$$\Delta \nu = A \left[ \frac{g \mu_B B}{3k_B (T - \Theta)} \right] (S + 1).$$

(1)

A strong deviation from the Curie-Weiss law, which lies in the heart of HF behavior, restricts UBe$_{13}$ to the small $B/T$ limit so that Eq. (1) stays valid in the entire measured $B$ range, in contrast to other systems which support SP \[20, 21, 24\]. Fitting $\Delta \nu(B)$ for $T$ between 15 K and 150 K with $\Theta$ found from magnetization measurements (see inset in Fig. 4) and taking into account the dominance of the orbital magnetic moment which causes $g = 0.8$ \[28\] gives $A = 45(5)$ MHz and $S = 8.5(0.5)$. From the value of $A \propto R^{-3}$ we get $R = 0.25(1)$ nm, which rather remarkably indicates a maximum overlap of corresponding $s$ and $f$ wavefunctions within the SP \[20, 22, 23\], as the U-U distance is 0.5124 nm and the radius of the $f$-orbital is 0.0527 nm. This is consistent with the muon sitting in between the two U ions \[19\] that captured a SP consisting of an electron whose wavefunction overlaps $f$-orbitals of said U ions, each having a magnetic moment $\mu_U = g \mu_B (S + 1/2)/2 = 3.6 \mu_B$, which is close to the $\mu_{\text{eff}}$ found from susceptibility measurements.

In a PM, strong spin exchange with the environment \[18\] would result in rapid spin fluctuations of the SP electron, averaging the hyperfine interaction to zero, which in turn would result in a collapse of the doublet into a single line at $\nu_{\mu}$ (see \[29\] for details) unless the SP spin $S$ is decoupled from the local spins \[20, 22, 23\]. Such decoupling is possible in high $B$ when the Zeeman energy of $S$ exceeds an exchange interaction $J$ between local spins — this is the case in magnetic insulators where the SP doublet is detected up to very high temperature \[17, 20, 21\]. In metals, RKKY interactions make $J$ much stronger, so that decoupling would require $B$ strengths inaccessible in the current experiment. That’s why we see a single line above 180 K, which by no means indicates the absence of the SP above 180 K — just active spin exchange with the environment \[20, 22, 23\]. The abrupt appearance of a SP doublet at 180 K (Fig. 3) we interpret as a result of opening of a spin gap that eliminates low-lying spin excitations, making spin exchange of $S$ with its environment ineffective. This explanation is consistent with anomalies in specific heat \[30\], NMR \[31\] and optical conductivity \[32\], which are discussed in terms of crystal field splitting of the $5f^3$ U ion by a characteristic energy of $\sim 180$ K.

Our measurements of the electric resistivity $\rho$ of the same single crystals (Fig. 4) confirm a spin gap opening in UBe$_{13}$ at about 180 K. A basic behavior of resistivity lies in the context of carrier scattering in metals with local spins \[32\], as in UBe$_{13}$ below 300 K $\rho$ is dominated by carrier scattering on spins \[33\]. At high temperature, it consists of a temperature-independent local scattering term and a $1/T$ term due to scattering on paramagnons \[32\]. At lower temperature down to $\Theta$, Kondo scattering takes over. A crossover from $\rho(T) = A/T + C$ to $\rho(T) = D \ln(E/T)$ at about 180 K signifies a characteristic change in the spin fluctuation spectrum of the system. (Fitting the data gives the following values: $A = 8.5 \times 10^{-3}$ Ohm-cm-K, $C = 94 \times 10^{-6}$ Ohm-cm, $D = 47.4 \times 10^{-6}$ Ohm-cm and $E = 3500$ K.) At high temperature, the largest contribution to $\rho$ comes from carriers with small momentum $q \sim 0$ \[32\] scattered by low-lying excitations dwarfing both $1/T$ and $C$ as the spin gap opens up, eliminating such excitations. On the other hand, an opening of the spin gap promotes resonant Kondo scattering effective at a significant $q$ and energy \[34\]. Moreover, disappearance of the low-lying spin excitations causes strong suppression of long-wavelength magnetic fluctuations accompanied by deviation of $\chi$ from the Curie-Weiss law at 180 K seen in $\Theta(T)$ dependence (inset in Fig. 4). An opening of the spin gap might
be explained by the position of an $f$-level lying 180 K below the Fermi surface.

A standard approach to HF systems considers as a starting point a set of strongly localized $f$-electrons; the appearance of a new energy scale results from hybridization of conduction electrons with local $f$ moments so that heavy quasiparticles appear on the Fermi surface. An alternative approach starts from a delocalized band carrier whose transport depends upon the strength of its coupling with excitations of the medium: in the limit of strong coupling an electron accompanied by lattice or spin excitations forms a quasiparticle — a polaron. As is the case for the well-known lattice polaron, formation of a spin polaron may profoundly renormalize the bare electron band (bandwidth $\Delta_0 \sim 1$ eV) into an extremely narrow ($\Delta_{SP} \sim 10^{-4} - 10^{-3}$ eV) SP band [23]. At low temperature, such an SP band supports coherent SP dynamics [24]. As SP have spin higher than 1/2, they do not need to follow a FL state — hence NFL behavior is possible. At still lower temperature, formation of spin bipolarons might cause SC [22, 36]. Here, the opening of a spin gap is a “must have” ingredient, as it protects paired electrons from spin exchange with the environment, which destroys pairs. At higher $T$, however, the SP dynamics occur on a background of strong coupling to spin fluctuations, which destroys coherence. A dramatic renormalization of the SP band is expected to go hand-in-hand with a significant increase of $m^*$, which may allow application of general concepts developed for coherent-to-incoherent crossover of the tunneling dynamics of heavy particles: suppression of coherence in a metal is expected at $T \sim \Delta_{SP}$ [37].

In summary, a spin gap opens up in UBe$_{13}$ at 180 K, detected by spin polarons. Formation of the SP band may explain several mysteries of UBe$_{13}$: NFL behavior, loss of coherence in a metal, and huge magnetoresistance. Emergence of such SP bands might be a general phenomenon in HF systems [38]. Within this picture, the SP is itself the celebrated heavy fermion.

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38. Recently, by using $\mu^+$SR technique we found SP in sev...
eral HF systems: CeCu₆, CeCoIn₅, Ce₂RhIn₈ etc.