

# Spin-polaron band in the ferromagnetic heavy-fermion superconductor $\text{UGe}_2$

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**Abstract.** It has long been believed that coexistence among ferromagnetic ordering, superconductivity or heavy-fermion behaviour is impossible, as the former supports parallel spin alignment while the latter two phenomena assume a spin-singlet configuration. This understanding has recently been challenged by a number of observations in uranium intermetallic systems where superconductivity (SC) is found within a ferromagnetic state and both ordering phenomena are facilitated by the same set of comparatively heavy quasiparticles which bind into spin-triplet pairs in the SC state. Within the heavy-fermion scenario, this mechanism necessarily assumes that the magnetism has a band character. This band is expected to be responsible for all three phenomena — heavy-fermion behaviour, ferromagnetism and superconductivity — although its nature and the nature of the heavy quasiparticles have so far remained unclear. Our high-field muon spin rotation measurements are indicative of spin polarons of subnanometer size in  $\text{UGe}_2$ . These spin polarons behave as heavy quasiparticles made of  $5f$  electrons. Once coherence is established, they may form a narrow spin-polaron band which thus may provide a natural reconciliation of itinerant ferromagnetism with spin-triplet superconductivity and heavy-fermion behaviour.

## 1. Introduction

Within the BCS theory of superconductivity (SC), it became clear long ago [1] that pairing of electrons in the spin-singlet state is effectively destroyed by an exchange mechanism arising from strong Coulomb interactions between the valence electrons. In a ferromagnetically (FM) ordered state, this exchange interaction tends to align the spins of electrons within a Cooper pair in parallel, thereby effectively preventing the pairing. Likewise, within the standard heavy-fermion (HF) approach, the Kondo effect quenches the on-site magnetic moment by spin fluctuations, thereby destroying pairs.

Although these three phenomena — magnetism, superconductivity and HF behaviour — have been considered in the past to be mutually antagonistic, the following findings clearly establish their possible coexistence (for a recent review, see [2]). A distinctive class of  $f$ -electron Ce- or U-based systems [3] convincingly shows that HF behaviour may coexist with SC. Most importantly, the SC pairing occurs among the heavy quasiparticles rather than within a band

of light electrons: the SC energy gap opens up within the band of heavy quasiparticles. The attractive interaction between the quasiparticles is probably not provided by the electron-phonon interaction as in ordinary BCS superconductors, but rather calls for an alternative mechanism which is offered by various spin-fluctuation models [4] of magnetically mediated SC.

In these HF materials SC may coexist and couple with magnetism: in fact, many f-electron HF systems exhibit SC deep within magnetically ordered states, suggesting that magnetism may promote rather than destroy the superconductivity. It is remarkable that the same set of heavy quasiparticles apparently supports both the magnetism and superconductivity [3].

In the theory of magnetically mediated superconductivity, it is important that the ferromagnetism itself is of itinerant character, similar to that in the canonical d-electron ferromagnets Fe, Co or Ni. Conventionally, electrons in solids are classified as either itinerant or localized. In strongly correlated electron systems, specifically HF systems, such a clear distinction is often obscured, since signatures of both pictures appear. The possibility that the same electrons might simultaneously exhibit both localized and itinerant characteristics due to strong Coulomb interactions has developed into a duality problem — are they localized, itinerant or of a dual nature (partially localized and partially itinerant)? The electronic duality proposed for many different HF systems exhibiting simultaneous SC and magnetism would require the same f-electron to display both localized and itinerant nature *simultaneously*. The 5f electron duality problem is still debated in UGe<sub>2</sub>, which is proposed to be viewed as a two-subset electronic system, where some of the 5f electrons are localized and responsible for the ferromagnetic moment and huge magnetocrystalline anisotropy, while the remaining 5f electrons are itinerant and responsible for unconventional SC [5].

Here we propose a specific concept that may supply the necessary requirement of simultaneously itinerant and localized electrons: formation of a spin-polaron band in which quasiparticle excitations of a low energy scale (several meV) around the Fermi energy are responsible for HF behaviour, SC and magnetism.

The standard theory for the formation of the extremely narrow high mass bands characteristic of heavy fermion metals starts from a set of strongly localized f-electrons. The appearance of a new (low) energy scale in this approach results from hybridization with the delocalized conduction states and strong correlations within the f-shells. A different approach, that we wish to develop here, starts with a delocalized band carrier whose transport depends upon the strength of its coupling with excitations of the medium. This is similar to the case of a lattice polaron (LP), where an electron accompanied by lattice modes forms a quasiparticle in which a local distortion of the crystal structure follows the charge carrier adiabatically and whose bandwidth is reduced by up to 4 orders of magnitude relative to that of ordinary electrons in conventional metals [6]. At low temperature, as long as the polaron is still much lighter than the atoms composing the medium, the charge is then delocalized within the LP band. A remarkable collapse of LP band at higher temperature marks a crossover from coherent band dynamics to incoherent hopping of localized states, analogous to the so-called dynamic destruction of the band for the tunnelling dynamics of heavy particles, such as protons, isotopic defects, or muons and muonium [7].

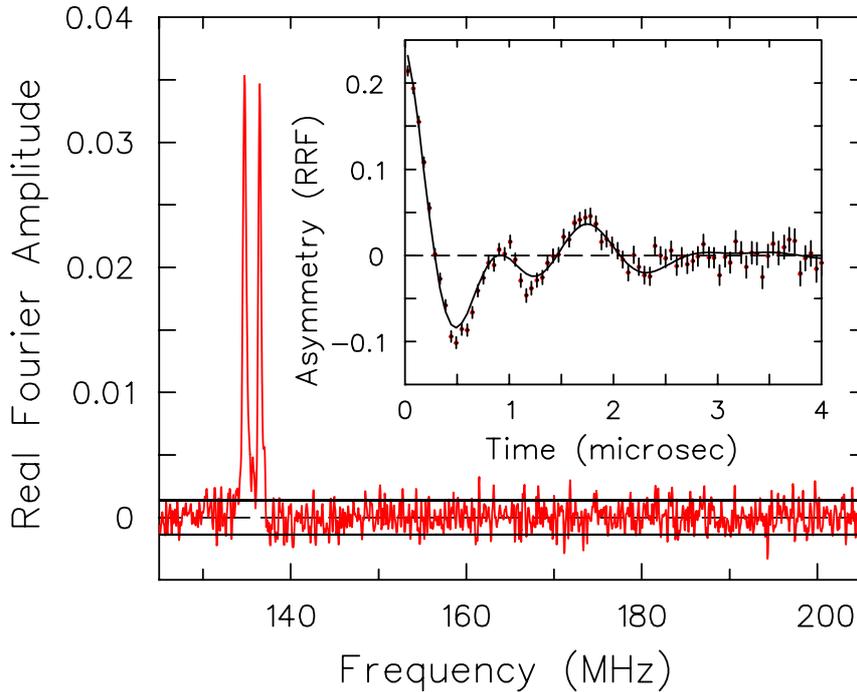
In close analogy, the exchange interaction (J) between a free carrier and local spins can cause electron localization into a FM droplet on the scale of the lattice spacing in a paramagnetic (PM) or AFM sea [8]. This charge carrier, accompanied by reorientations of local spins, forms a spin polaron [6, 8] (SP) with a large composite spin. As in the case of the LP, formation of a SP profoundly renormalizes the bare electron band into an extremely narrow ( $\sim 0.001 - 0.1$  eV) spin-polaron band, which will favour coherent SP band dynamics at low temperature as long as spin fluctuations are suppressed. A remarkable result is that heavy f-like quasiparticles (SP) become part of the Fermi surface. We specifically note that within this framework the heavy quasiparticle is not an f-electron but the composite quasiparticle, a SP, formed as a result of

s(p)-f interaction. Here we present spectroscopic indication, obtained by muon spin rotation, for spin polarons in  $\text{UGe}_2$ , confined within  $R = 0.25(1)$  nm, with a high spin of  $S = 4.3 \pm 0.3$ . At low temperature, SP tend to form a narrow spin-polaron band in the vicinity of  $E_F$ , profoundly modifying the magnetic, transport, optical and thermodynamic properties of the host.

## 2. Results and discussion

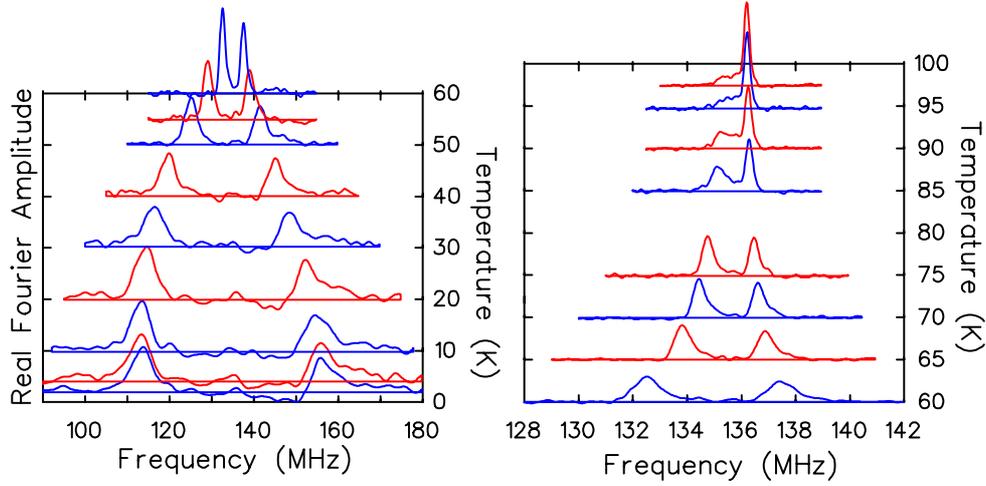
Single crystals of  $\text{UGe}_2$  for the current studies were grown by the Czochralski technique under purified Ar atmosphere with a radio-frequency heating. Single crystals of a disk shape were oriented using a white beam X-ray backscattering Laue method, sparkcut and etched to remove the oxidised surface. The electrical resistivity shows a residual resistivity ratio RRR=62 and FM transition at  $T_c = 52.6$  K.

Time-differential  $\mu^+$ SR experiments, using 100% spin-polarized positive muons implanted into these samples, were carried out on the M15 surface muon channel at TRIUMF using the HiTime spectrometer. At high temperature, Fourier transforms of the time spectra in a magnetic field ( $B$ ) transverse to the initial muon spin polarization direction and parallel to the easy magnetization direction ( $a$  axis) exhibit a single peak 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). However, below  $T = 100$  K the  $\mu^+$ SR spectra change abruptly to reveal two peaks (Fig. 1). The evolution of these signals with temperature is presented in Fig. 2. These two peaks are also shifted to lower frequencies relative to the single peak (not shown in Fig. 1) detected in a reference sample ( $\text{CaCO}_3$ ), which occurs at the bare muon frequency.



**Figure 1.** Frequency spectrum of muon spin precession in  $\text{UGe}_2$  in a transverse magnetic field of  $H=1$  T at  $T=75$  K. Inset: same spectrum in the time domain in a rotating reference frame at 135.53333 MHz. The two-frequency precession pattern characteristic of a localized electron hyperfine-coupled to a muon is clearly apparent in both domains.

Observation of two peaks in the Fourier spectra prompted the authors of [9] to suggest two magnetically inequivalent sites occupied by the positive muon in  $\text{UGe}_2$ . Although such



**Figure 2.** Fourier transforms of the muon spin precession signal in UGe<sub>2</sub> in a transverse external magnetic field of 1T at different temperatures. The characteristic SP lines appear below about 100 K and persist through the FM transition ( $T_c = 52.6$  K) down to the lowest measured temperature.

an approach constitutes the conventional assignment, the two-peak signal may alternatively be interpreted as indication of a muon-electron bound state.

In particular, the two lines shown in Fig. 1 and Fig. 2 may indicate the characteristic signature of a coupled muon-electron spin system in high magnetic field [10]. We suggest that the observed bound state is a spin polaron. In a PM or metallic environment, the strong pair exchange interaction of the bound electron with itinerant spins would result in rapid spin fluctuations of this electron, averaging the hyperfine interaction to zero which, in turn, would result in a collapse of the doublet into a single line at  $\nu_\mu$  [11], if the local FM ordering mediated by this electron did not hold the electron's spin orientation locked. In metals, however, even the protective local FM environment of a SP does not ensure observation of the doublet unless the SP spin ( $S$ ) is decoupled from its magnetic environment [12, 13]. Such decoupling is possible in high  $B$  when the Zeeman energy of  $S$  exceeds an exchange interaction ( $I$ ) between local spins [10]. This is the case in magnetic insulators where the SP doublet is detected up to very high temperature. In metals, RKKY interactions make  $I$  much stronger, so that decoupling would require a very high magnetic field that is inaccessible in the current experiment. In UGe<sub>2</sub>, above about 100 K, spin exchange with the magnetic environment effectively averages the muon-electron hyperfine interaction, causing the collapse of the doublet into a single peak as presented in Fig. 2. Such a collapse does not necessarily mean that the SP does not form in UGe<sub>2</sub> above 100 K; we just may not see its fingerprint, which is a characteristic doublet [11]. The abrupt appearance of a SP doublet below about 100 K is possible due to another effective decoupling mechanism the opening of a spin gap due to crystal field splitting of the U ions spin excitation, characteristic of U compounds [5]. This is consistent with optical conductivity measurements which exhibit a dramatic reduction of the spin scattering rate below 120 K [14].

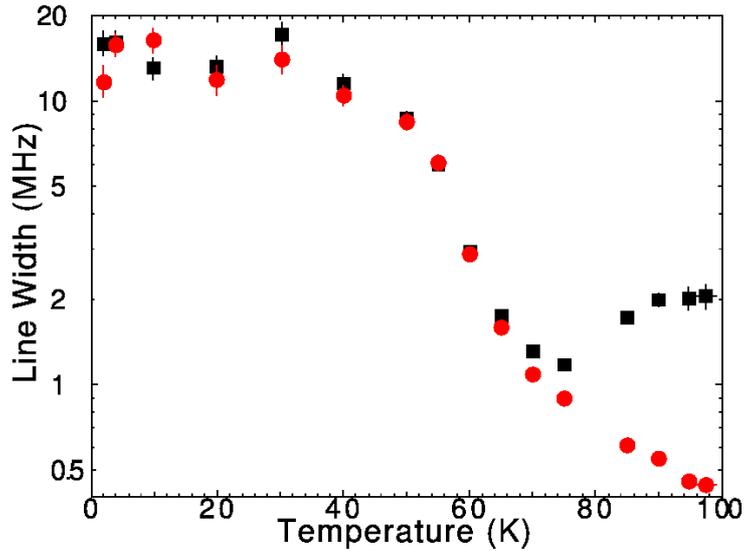
Temperature and magnetic field dependencies of the signal frequency splitting,  $\Delta\nu$ , provide information on the characteristic size through the hyperfine coupling ( $A$ ) and determine the composite spin of the SP [10, 11]. Within a mean field approximation,  $\Delta\nu$  is proportional to a Brillouin function. For  $g\mu_B B \ll k_B T$ ,  $\Delta\nu$  is a linear function of both  $B$  and  $1/(T - T_c)$  [10]:

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

At low  $T$  and high  $B$ , however, this equation is no longer valid, as the composite spin  $S$  is fully

polarized. Therefore, in a magnetic field high enough that the muon Zeeman energy exceeds the hyperfine coupling,  $\Delta\nu$  saturates at the value of  $A$  [10, 11]. In  $\text{UGe}_2$ ,  $\Delta\nu$  saturates as a function of both inverse temperature (in a magnetic field  $H = 1$  T) and  $H$  (at  $T = 5$  K) at the same value,  $A = 41 \pm 2$  MHz.

This hyperfine coupling in the SP is about 100 times less than that for Mu in vacuum, implying that the SP radius is  $0.25(1)$  nm. This is consistent with the muon being centred between two U atoms ( $x=0.5, y=0.5, z=0.5$ ) giving a muon-U distance of  $0.214$  nm. Fitting Eq. (1) to the data yields  $S = 4.3(3)$  and  $T_c = 52(1)$  K, consistent with our estimate of  $S$  from the SP radius and  $T_c$  of our sample, respectively.



**Figure 3.** Temperature dependences of the SP line widths of the signals with higher (red circles) and lower (black squares) frequencies in  $\text{UGe}_2$  in a magnetic field of  $H = 1$  T.

The exchange interaction governs SP formation and dynamics in  $\text{UGe}_2$ , since the Coulomb interaction is effectively screened. Therefore the role of the muon, which may be important for SP formation in magnetic semiconductors and insulators [10], is reduced to that of an innocent bystander in metals. The appearance of the mid-infrared feature in optical conductivity experiment in  $\text{UGe}_2$  is attributed to a polaron response [14]. We argue that once the host lattice is populated by free spin polarons, one of them may be captured by the muon to reveal the fingerprint of a bound muon-electron state the characteristic doublet.

In  $\text{UGe}_2$ , the dynamics of such SP clearly shows a qualitative change around 80 K. At high temperature, the asymmetric doublet shape (Fig. 2, right panel) indicates the nearly static character of the SP [12]. A remarkable crossover occurs at about 80 K, where not only do the line widths become equal (Fig. 3), but also the spectral weights (corresponding state populations) of the two lines within the doublet effectively equilibrate, clearly indicating the onset of the effective spin-exchange mechanism within the SP system, as other spin-exchange channels are rather ineffective [15]. This marks a crossover from the static SP to itinerant polaron behaviour. At high temperature, the characteristic length (in Angstroms) of the ferromagnetic fluctuations determined from neutron measurements,  $\xi_{FM} = 3.45/(T/T_c - 1)^{1/2}$ , is much less than the SP size,  $2R$ , which causes strong SP localization due to the significant energy shift of the nearest equivalent SP positions within the lattice (so-called static destruction of the band [7]). At lower temperature, once  $\xi_{FM}$  exceeds the SP size, energy levels equilibrate within the length scale of  $\xi_{FM}$ , which causes SP delocalization. A simple estimate shows that  $\xi_{FM}$  becomes equal to  $2R$  at

77 K, which agrees well with the experiment. The amplitudes or spectral weights of both lines are temperature independent below about 80 K (Fig. 2), which clearly indicates temperature independent populations of the spin-up and spin-down states which in turn is inconsistent with a localized SP but rather signifies its itinerant nature. Thus, our model suggests that the SP, captured by the muon, stays localized and is protected from spin exchange above 80 K; however, below 80 K it remains localized but undergoes active spin exchange with free itinerant SP.

However, the remarkable itinerancy acquired by SP below 80 K does not mean they form a band at 80 K. Instead, SP transport in UGe<sub>2</sub> just below 80 K is determined by hopping (diffusive) dynamics similar to the hopping dynamics of muon or Mu states in the regime dominated by static destruction of the band when dynamic fluctuations of the environment (phonons in the case of Mu) makes such hopping dynamics possible [7]. In fact, coherent SP transport is only possible in a spin-polarized band where spin-flip scattering is suppressed due to the absence of SP with the opposite spin state. In a FM state, such coherent transport of SP becomes possible due to splitting of the majority and minority spin subbands. This splitting may be viewed as Zeeman splitting due to spontaneous magnetization. In UGe<sub>2</sub>, such splitting of spin subbands occurs at  $T^*$ , which separates the high-temperature weakly polarized phase (FM1) from the low-temperature strongly polarized phase (FM2) and shows up as anomalies in resistivity, heat capacity, magnetization, magnetoresistivity and de Haas-van Alphen measurements [5].

Formation of the spin polaron band may provide an explanation for a list of mysteries of UGe<sub>2</sub> as well as other HF and strongly correlated materials, such as itinerant versus localized carriers in a duality problem when local moments acquire itinerancy, small Fermi surface versus large Fermi surface including both conduction electrons and local spins, huge anisotropic magnetoresistance, Fermi surface reconstruction at  $T^*$  and the nature of the mysterious FM transition within the FM phase.

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