Muon spin rotation and relaxation study of the ferromagnet β-UB_2C

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Magnetic ordering and spin dynamics of $5f$ electrons in ferromagnetic β-UB_2C with $T_C = 74.5$ K have been investigated by muon spin rotation and relaxation (μSR). The experimental data indicate a slowing down of the spin fluctuations of the U moments on approaching T_C from the paramagnetic regime. In the ferromagnetic state, a spontaneous muon spin precession with a single frequency is observed, characteristic of a quasi-static mean magnetic field at a single muon site. The damping rate of the precessing signal is essentially of static origin, and it shows a dip at around 40 K, close to the temperature where the magnetic specific heat and the temperature derivative of the resistivity reveal an anomaly. Therefore, these anomalies are mainly related to the static component of $5f$ electrons. The spin-lattice relaxation rate measured in the ferromagnetic phase seems to probe the spin dynamics of the itinerant electronic rate. This component remains finite down to the lowest measured temperature; therefore, it is characterized by an appreciable density of magnetic fluctuations at extremely low energy. Hence, as for UGe_2, the $5f$ electrons in β-UB_2C are suggested to exist in two different substates of localized and itinerant nature.

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I. INTRODUCTION

Recent interest in strongly correlated electron systems has focused on $5f$-electron ferromagnets, in particular on the compounds UGe_2,[1–4] URhGe,[5] UIr,[6] and UCoGe,[7] in which coexistence of superconductivity and ferromagnetism has been observed. These compounds together with a few others like UPd_2Al_3 and UNi_2Al_3 form a new class of so-called unconventional uranium superconductors, in which the formation of Cooper pairs is probably mediated through magnetic fluctuations.[8,9] Another fascinating feature of the U-based superconductors is the possibility that the same $5f$ electrons possess both itinerant and localized properties, and therefore are responsible for both magnetism and superconductivity.[10,11] However, this property is not obvious, since in some systems like UGe_2, it was shown by the muon spin rotation and relaxation (μSR) technique that the $5f$ electrons exist in two different substates.[12,13] Because only a few such ferromagnetic superconductors are known to date, the understanding of the nature of the $5f$-electron behavior in these compounds is still scarce. Thus, any comparative study of magnetic properties on further uranium-based ferromagnets is highly desired.

In this context, we recently studied fundamental properties of β-UB_2C (see Refs. 14,15) and found that this compound enters a ferromagnetic state below the Curie temperature $T_C = 74.5$ K. In addition to the ferromagnetic transition, a characteristic temperature $T^* \simeq 37$ K was found, at which both the electrical resistivity and specific heat show anomalies. Interestingly, T_C and T^* decrease with increasing applied pressure, and both are expected to reach 0 K at a critical pressure above 20 kbars.[15,16] The thermoelectric power is positive and displays a maximum at 12 K. The observed features are comparable to those of ferromagnetic UGe_2 and UIr, known as superconductors under pressure. The itinerant character of ferromagnetism in β-UB_2C was deduced from specific heat measurements, which indicated an enhanced electronic specific heat coefficient [34.7 mJ/(mol K^2)]. Note that this does not exclude the possibility of localized magnetic electrons in the system. The analysis of magnetization, specific heat, and resistivity data suggests the existence of an energy gap Δ in the electronic excitation spectrum with $\Delta/k_B = 11\,(1)$ K. This is rather small in view of the large magnetocrystalline anisotropy of the order of 100 K that is typically found in U-based systems. Neutron powder diffraction experiments revealed that the uranium moments are close to the *ab* plane forming ferromagnetic chains parallel to the hexagonal *c* axis. At 1.5 K the ordered magnetic moments of the uranium ions at the two uranium sites are approximately equal with a value of 1.06 μ_B/U. This low value could support the itinerant nature of $5f$ electrons in β-UB_2C.

In this paper, we present μSR measurements on β-UB_2C with the aim of shedding additional light on its physics. Our aim is two-fold: (i) to further characterize the anomaly at T^* previously observed in the resistivity and specific heat measurements,[14,15] and (ii) to determine whether the $5f$ electrons exist in two different substates as found for UGe_2.[12,13]

II. EXPERIMENTAL

A polycrystalline sample of β-UB_2C was prepared by argon arc-melting of high-purity elements on a water-cooled copper hearth. Prior to melting, the mixture of crystallized boron (98% ^{11}B-enriched isotope) and carbon was compacted in a steel die into a tablet. After melting, weight losses

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were found to be less than 0.5 mass%. The sample phase purity was examined by powder x-ray and neutron diffraction measurements.[14] The lattice parameters are $a = 0.6530(2)$ and $c = 1.0764(7)$ nm. The zero-field (ZF) μSR experiments on the characterized polycrystalline sample of β-UB_2C were performed in the temperature range of 5–90 K using the general purpose surface-muon instrument of the Swiss Muon Source (Paul Scherrer Institute). An introduction to this technique can be found in Ref. 17.

III. RESULTS

As examples of μSR spectra of β-UB_2C measured below T_C and in the paramagnetic state, we display in Fig. 1 the data obtained at $T = 70$ and 80 K, respectively. The qualitative change in the shape of these spectra confirms that a magnetic transition occurs between these two temperatures. A μSR spectrum describes the asymmetry $a_0 P_Z^{\rm exp}(t)$ of the decay positrons emitted from initially fully polarized muons implanted into the sample. In our case, a_0 is found to be independent of temperature and close to 0.25, which is a value in the expected range at our experimental conditions. $P_Z^{\rm exp}(t)$ characterizes the evolution of the projection of the muon polarization along the Z axis. Since Z is by definition the direction of the muon initial polarization, $P_Z^{\rm exp}(t = 0) = 1$.

In the paramagnetic state, the spectra can be analyzed using the product of a static Gaussian Kubo-Toyabe function, $P_{\rm KT}(t)$, and an exponential relaxation

$$P_Z^{\rm exp}(t) = P_{\rm KT}(t)\exp(-\lambda_Z t). \qquad (1)$$

$P_{\rm KT}(t) = \frac{1}{3} + \frac{2}{3}(1 - \Delta_{\rm KT}^2 t^2)\exp(-\Delta_{\rm KT}^2 t^2/2)$ accounts for the muon depolarization induced by the boron nuclei. The exponential factor in Eq. (1) corresponds to the relaxation arising from the fast fluctuating electronic moments of the system under study. Since the fields produced at the muon site by the nuclei and the electrons are uncorrelated, it is justified to take the product of $P_{\rm KT}(t)$ and $\exp(-\lambda_Z t)$ to describe the evolution of the muon polarization. A first inspection of the paramagnetic state data showed that consistent fits to Eq. (1) could be obtained with $\Delta_{\rm KT} \simeq 0.130(10)\ \mu{\rm s}^{-1}$. In a second step, new fits were performed taking $\Delta_{\rm KT}$ fixed to $0.130\ \mu{\rm s}^{-1}$ in order to get a more accurate estimate of the spin-lattice relaxation rate λ_Z. The temperature dependence of λ_Z is shown in Fig. 2. Clearly, λ_Z sharply increases as T_C is approached from above. Such a behavior is expected in the vicinity of a second-order magnetic phase transition. It is due to the slowing down of the magnetic fluctuations and the onset of pair correlations between the uranium magnetic moments.

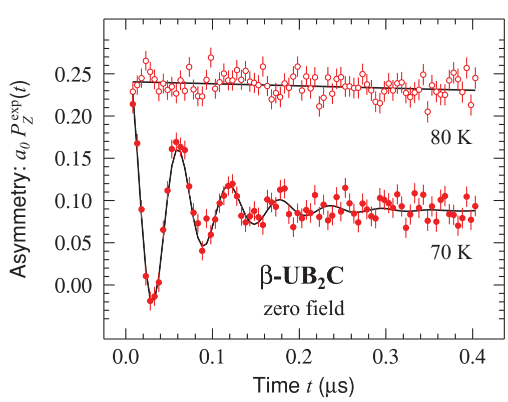

FIG. 1. (Color online) μSR spectra recorded in zero external magnetic field in β-UB_2C at 70 and 80 K. The full lines are fits to models explained in the text.

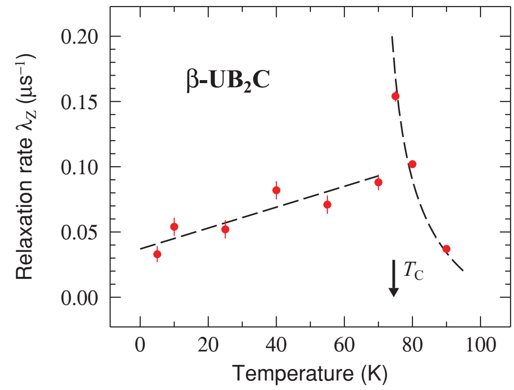

FIG. 2. (Color online) Temperature dependence of the spin-lattice relaxation rate λ_Z measured in β-UB_2C. The dashed lines are guides to the eye.

At temperatures below T_C, the asymmetry displays oscillations due to the precession of the muon spins in the spontaneous static field present in β-UB_2C. Here the time dependence of the asymmetry is fitted to

$$P_Z^{\rm exp}(t) = f_\perp \exp(-\lambda_X t)\cos(\gamma_\mu B_0 t + \varphi) + f_\parallel \exp(-\lambda_Z t), \qquad (2)$$

with $f_\perp + f_\parallel = 1$. The first term in the right-hand side describes the evolution of the polarization of the muons which experience a field transverse to their initial polarization, whereas the second term accounts for the spin-lattice relaxation of the muons submitted to a field parallel to their initial polarization. In a way similar to the paramagnetic case, a first fit of the spectra was performed with the relative weight of these two terms left as a free parameter. The ratio $f_\perp/f_\parallel$ was found to be close to 2, as expected for a polycrystalline sample with no texture. In a subsequent stage, the spectra were fitted to Eq. (2) with $f_\perp = 2/3$ and $f_\parallel = 1/3$.

In the oscillating term of Eq. (2), $\gamma_\mu = 851.6$ Mrad s^{-1} T^{-1} is the muon gyromagnetic ratio, B_0 refers to the spontaneous field at the muon site, φ models an instrumental phase which is close to 0 degree and λ_X is the spin-spin relaxation rate characterizing the damping of the wiggles. A Gaussian form for this damping was also tested but turned out to give slightly poorer fits than the exponential in terms of confidence parameter. The temperature dependencies of B_0, λ_X, and λ_Z are shown in Figs. 3 and 2.

In a metallic magnet such as β-UB_2C, $\mathbf{B}_0$ is the sum of the dipolar field arising from the localized uranium magnetic moments and of the hyperfine field which is due to the polarization of the conduction electrons at the muon site, essentially through the Ruderman-Kittel-Kasuya-Yosida interaction with the localized uranium moments. We denote these two fields as $\mathbf{B}_{\rm dip}$ and $\mathbf{B}_{\rm hyp}$, respectively. While the temperature dependence of $B_{\rm dip}$ is expected to follow that of the order parameter, this is not necessarily the case for $B_{\rm hyp}$. As

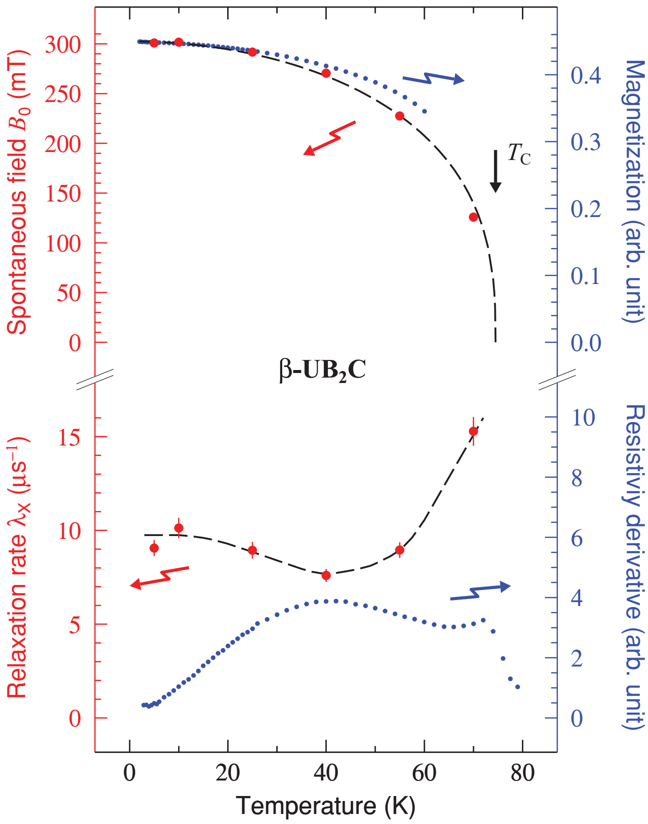

FIG. 3. (Color online) Top panel: temperature dependence of the spontaneous field B_0 and of the magnetization of β-UB_2C measured in a field of 0.5 T.[14] Bottom panel: the spin-spin relaxation rate λ_X and the temperature derivative of the resistivity taken from Ref. 14. The dashed lines are guides to the eye.

shown in Fig. 3, B_0 and the magnetization behave in a similar way at low temperature, but a significant deviation is found for temperatures above $\simeq$40 K. This could indicate the influence of the external field in the bulk measurement or the fact that B_{hyp} is not proportional to the magnetization. As an example for such a behavior in a metal, we cite metallic iron.[18,19]

The origin of the damping of the wiggles associated to the muon spin precession is two-fold: (i) a static distribution of the spontaneous field probed by the muon ensemble which reveals the defects of the magnetic structure and (ii) dynamical effects associated with spin-lattice relaxation processes. It is known that the dynamical contribution to λ_X is equal to λ_Z in the limit of isotropic spin dynamics; see, e.g. Ref. 17. Since λ_X is found to be at least two orders of magnitude larger than λ_Z, it is reasonable to conclude that the damping of the wiggles has an essentially static origin. In contrast, the muon spin relaxation rate λ_Z only probes the dynamical properties of the $5f$ electrons.

Generally, $\lambda_X(T)$ is expected to be temperature independent at low temperature and to rise as the temperature is increased toward the magnetic transition. This rise manifests the shortening of the correlation length of the magnetic structure and the strengthening of *critical* fluctuations while the transition is approached. Interestingly $\lambda_X(T)$ shows a dip at $T^* \simeq 40$ K, not too far from where the magnetic specific heat and the temperature derivative of the resistivity present an anomaly;[14] see Fig. 3 for the resistivity data. Such a resistivity anomaly has previously been observed for UGe_2.[20] In the previous paragraph we argued that the behavior of $\lambda_X(T)$ is essentially controlled by the static field distribution at the muon site. Hence the minimum in $\lambda_X(T)$ should reflect a static phenomenon associated with spatial inhomogeneity of the magnetic field. Previously, the question of whether T^* is the signature of a transition has been discussed for UGe_2.[13] In the case of β-UB_2C, no additional magnetic phase transition besides the ferromagnetic transition at 74.5 K seems to take place around T^*. Therefore, the observation of the $\lambda_X(T)$ minimum at 40 K does not support the interpretation of the anomalous bump in the specific heat and derivative of the resistivity in terms of quantum fluctuations of the localized spins.[14]

We turn now to the discussion of $\lambda_Z(T)$ in the ordered phase. We first recall its expected behavior in the ordered phase of a ferromagnet with only localized magnetic moments. In the case of a weak magnetic anisotropy, a Raman-type relaxation process of the muon spin is observed.[21] It roughly leads to a quadratic temperature dependence of λ_Z at least up to $T_C/2$. It has been nicely observed in the intermetallic compound $GdNi_5$ where the magnetic anisotropy arises from the dipolar interactions between the Gd^{3+} spins.[22] However, the magnetic anisotropy, and therefore the energy gap Δ, is sizeable for β-UB_2C since $\Delta/k_B = 11\,(1)$ K.[14] Because the muons probe the dynamics at very small energy (on the order of $\hbar\gamma_\mu B_0 \simeq$ 0.2 μeV in our case, i.e., $\hbar\gamma_\mu B_0/k_B = 23$ mK) relative to Δ, one expects λ_Z to rapidly vanish for temperature tending toward zero.[17] As seen in Fig. 2, this trend is not observed. Instead, λ_Z is found to weakly increase with the temperature up to T_C, and λ_Z remains finite for $T \to 0$. Such a persistent spin dynamics has also been detected for UGe_2, but the size of λ_Z is much smaller for this latter system.[12] Our analysis strongly suggests that the observed relaxation is not associated with localized magnetic moments but rather with the magnetic density of itinerant electrons. Hence, as for UGe_2,[12,13] the $5f$ electrons are viewed as existing in two different substates. The itinerant component drives the observed dynamics and contributes to a large electronic coefficient of specific heat. The localized component is responsible for the dipolar field at the muon site considered when discussing the spontaneous field.

To proceed further, we refer to the analysis performed by Moriya and Ueda of the spin-lattice relaxation rate measured by nuclear magnetic resonance for so-called weakly ferromagnetic metals;[23] see also Ref. 24. These metals are characterized by an intense quasi-elastic neutron scattering. Because of the strong spin-orbit coupling in a uranium compound, the quasi-elastic line width $\Gamma_0(q)$ is proportional to the wave vector of the excitations, q, in the neighborhood of the center of the Brillouin zone of interest here.[25] Hence, we can write $\Gamma_0(q) = \mathcal{F}q/\chi_0$, where χ_0 is the homogeneous static susceptibility and $\mathcal{F}$ a constant. The transverse wave-vector-dependent susceptibility for $T < T_C$ is given by $\chi_0^\perp(q) = \kappa^2\chi_0/q^2$, where κ is the inverse of the magnetic correlation length.[26] For an isotropic system $\lambda_Z \propto T\int_{q_\ell}^{q_u}[\chi_0^\perp(q)/\Gamma_0(q)]q^2dq$, where q_u and q_ℓ are cutoff wave vectors,[27] this leads to $\lambda_Z \propto T\kappa^2\chi_0^2\ln(q_u/q_\ell)$. Recalling that $\kappa \propto \sqrt{|T - T_C|/T_C}$ and $\chi_0 \propto 1/|T - T_C|$, we expect κ

and χ_0 to be temperature independent in the low-temperature limit. The temperature dependence of the term $\ln(q_u/q_\ell)$ should be mild because of the logarithm function. This simple analysis predicts $\lambda_Z \propto T$. Our data suggest the presence of such a linear term on top of a temperature-independent spin-lattice relaxation rate with a value $\simeq 0.04\ \mu s^{-1}$; see Fig. 2. As already noticed for MnSi, the qualitative description of the spin dynamics using the phenomenological theory for weakly ferromagnetic metals is useful, but a quantitative insight requires a more involved model. In particular, the observation of a persistent muon spin relaxation at low temperature is intriguing. This behavior, which has already been observed in numerous geometrically frustrated compounds, including magnetically ordered materials,[28–30] is still not understood.

IV. CONCLUSIONS

The present μSR investigation of β-UB_2C has confirmed that this compound has a single magnetic phase transition at $T_C = 75$ K, and that inside the ferromagnetic state a crossover occurs at $T^* \simeq 40$ K. The measured spin-lattice relaxation rate in the magnetically ordered state suggests the $5f$ electrons to exist in two different substates of localized and itinerant nature, respectively. The similarity with UGe_2 in terms of the existence of T^* and of two $5f$ electrons substates is remarkable. However, the crossover at T^* in β-UB_2C presumably does not involve itinerant $5f$ electrons. Resistivity measurements of β-UB_2C under high pressure have already been reported.[15,16] It would certainly be interesting to extend this type of work to other measurements, including μSR measurements under pressure.

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