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^{209}Bi NMR in the heavy-electron system YbBiPt

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Abstract

Bismuth NMR Knight shift and spin lattice relaxation rate $1/T_1$ data are reported between 35 and 325 K in the low-carrier heavy fermion system YbBiPt. The Knight shift is strongly temperature dependent and negative. Its temperature dependence tracks the bulk susceptibility with a hyperfine coupling constant $A_{\text{hf}} = -7.88 \text{ kOe}/\mu_B$. At low temperatures $1/T_1$ exhibits a dramatic increase, such that the average 4f spin correlation time τ_r shows a crossover behavior at about 75 K. The rate $1/\tau_r$ is proportional to temperature above 75 K, consistent with non-interacting 4f local moments which are relaxed via Korringa-type scattering with the conduction electrons. We discuss the behavior below 75 K in terms of crystal-field effects or a strongly temperature dependent contribution from non-zero q regime of the dynamical susceptibility.

1. Introduction

The discovery of a large γ in the heavy-electron compound YbBiPt has attracted a number of recent investigations [1–3]. YbBiPt has a very large Sommerfeld constant ($\gamma \approx 8 \text{ J/mol K}^2$) and possesses an apparent small-moment [4], spin-density wave [5] transition at 0.4 K which gaps part of the Fermi surface and which arises from a low-carrier density heavy electron state. Although many interesting aspects of this compound occur at low temperatures ($< 1 \text{ K}$), it is important to investigate other relevant properties at high temperatures. For example, neutron scattering data reveal an inelastic peak at $\sim 6 \text{ meV}$ which was attributed to a crystal-field splitting of a $J = 7/2$ ground state multiplet [6]. Also, muon spin relaxation data exhibit an anomalous cusp-like behavior in the vicinity of 20 K [7]. The work reported here provides microscopic information to help clarify the physics of this compound using ^{209}Bi nuclear magnetic resonance. Because the measurements are

restricted to temperatures much larger than the Kondo temperature, the data are expected to exhibit properties of a system where the 4f-electrons are well localized.

YbBiPt crystallizes in a cubic MgAgAs type (half-Heusler structure) with $F43m$ symmetry. The sample was grown in a bismuth flux as described in Ref. [2]. One gram of polycrystalline material was crushed and quickly mounted in a helium cryostat to avoid sample degradation, which had been observed previously when the material came in contact with air [8]. The quality of our sample was verified by measuring the temperature dependence of the susceptibility after the NMR data were taken. No appreciable change was observed compared to a reference measurement. Extra care was also taken to avoid eddy current heating of the sample due to high voltage RF pulses from the conventional pulsed NMR spectrometer. In this regard, we observed a rather large temperature rise (as much as $\sim 40 \text{ K}$) for duty cycles exceeding 0.1% at 77 K, possibly due to low thermal conductivity associated with low carrier density. This situation could also have been aggravated by oxide layers that might have formed on the surface of the grains. Although the oxides would prohibit efficient thermal

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conduction throughout the sample and container, their microscopic effects are not detectable by NMR.

The NMR spectra exhibit a single Gaussian line at all temperatures measured. All atomic sites have cubic local symmetry so that there are no quadrupolar effects. The single line observed in the NMR spectrum is associated with the ^{209}Bi nuclei ($I=9/2$). The platinum and ytterbium signals are expected to be weaker by more than an order of magnitude because of their smaller spins ($I=1/2$ for ^{195}Pt and ^{171}Yb , $I=5/2$ for ^{173}Yb). In addition, Yb would have been visible as two lines corresponding to the two isotopes having relatively low natural abundance. If the observed line were due to Pt, the calculated Knight shifts would be $\approx 26\%$ at room temperature, which is unphysically large. We note that the demagnetization corrections were negligible (0.04%) at fields where the Knight shifts were measured (6–9 T).

2. Results and discussion

Fig. 1 shows the temperature dependence of the ^{209}Bi Knight shift K in YbBiPt. The Knight shift is large, isotropic, negative and follows the Curie–Weiss behavior of the bulk susceptibility. A large shift is not unexpected for bismuth, and the negative sign is typical of the behavior observed in other rare earth intermetallics with more than half-filled f-shells, i.e. $K > 0$ for $J=L-S$ and $K < 0$ for $J=L+S$ [9]. These facts suggest that the Knight shift is mainly due to a transferred hyperfine field from Yb 4f-moments.

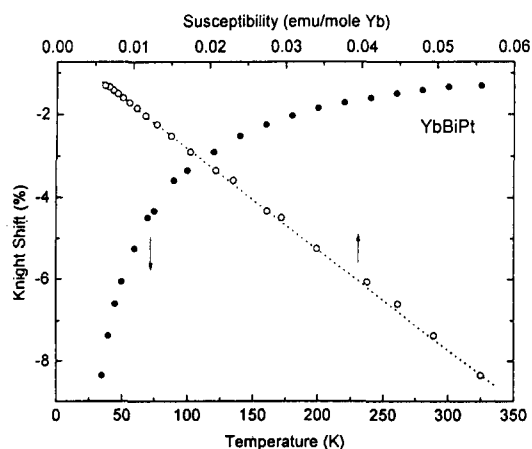


Fig. 1. ^{209}Bi Knight shift in YbBiPt plotted against temperature (bottom scale), and against susceptibility with temperature as implicit parameter (upper scale).

The Knight shift plotted against the bulk susceptibility yields a hyperfine coupling constant of $A_{\text{hf}} = -7.88(4)$ kOe/ μ_{B} (Fig. 1). A high temperature extrapolation of these data results in a temperature independent Knight shift of $-0.38(2)\%$, which, in the tight-binding limit, can be separated into orbital and conduction electron contributions. Because the former is always positive, we speculate that the measured shift originates mainly from the conduction electrons near the Fermi level, presumably due to core polarization by the unfilled Bi 6p orbitals.

The spin dynamics of YbBiPt show interesting behavior at high temperatures. In Fig. 2 we show the temperature dependence of the spin-lattice relaxation time T_1 and its inverse $1/T_1$. The relaxation rate $1/T_1$ increases monotonically as the temperature is lowered, but below ~ 75 K increases by almost two orders of magnitude. By about 35 K the T_1 value has become so short (15 μs), that the NMR echo is no longer visible. The spin-spin relaxation time T_2 (not shown) behaves similarly and approaches the value of T_1 at low temperatures.

In general, the spin-lattice relaxation in paramagnetic metals is related to the imaginary part of the complex dynamical susceptibility $\chi(\mathbf{q}, \omega)$ by the relation

$$\frac{1}{T_1} = \gamma_N^2 k_B T \sum_q [A_{\text{hf}}(\mathbf{q})]^2 \frac{\chi''(\mathbf{q}, \omega_0)}{\omega_0}, \quad (1)$$

where $A_{\text{hf}}(\mathbf{q})$ is the spatial Fourier transform of the transferred hyperfine field from Yb^{3+} ions and ω_0 is the Larmor frequency. Neglecting the wave number dependence of the susceptibility and assuming a Lor-

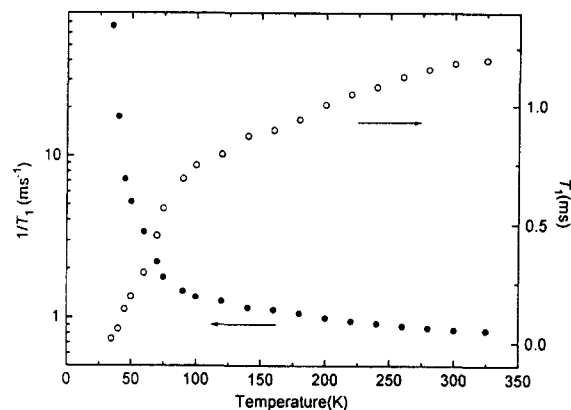


Fig. 2. Temperature dependence of spin-lattice relaxation time T_1 and rate $1/T_1$.

entzian energy spectrum for spin–spin correlation and an isotropic hyperfine field, $1/T_1$ can be expressed in terms of the spin correlation time τ_f of the 4f local moments as

$$\frac{1}{T_1} = \gamma_N^2 k_B A_{\text{hf}}^2 T \chi_f(T) \tau_f, \quad (2)$$

where static f-susceptibility $\chi_f(T)$ is derived from the bulk measurements. The hyperfine field is taken from the Knight shift data of Fig. 1.

Local moments mutually interacting via exchange produce hyperfine fields at nuclear sites with a relaxation rate that is independent of temperature: $1/T_1 \approx \gamma_N^2 A_{\text{hf}}^2 / \omega_e$, which is valid in the limit where the fluctuation frequency of the local moments $1/\tau_f \approx \omega_e \gg \omega_0$ [10]. The observed decrease in $1/T_1$ is not consistent with this expectation and we argue that this may then be due to the temperature dependence of $1/\tau_f$. We demonstrate this in Fig. 3 where we show the temperature dependence of $1/\tau_f$ derived from T_1 data using Eq. (2). The experimental $1/T_1$ values have contributions both from f- and conduction electrons, however: $1/T_1 = 1/T_{1f} + 1/T_{1s}$. Because the contribution from the former is expected to dominate in this system, we have assumed in Fig. 3 that $1/T_{1s} \ll 1/T_{1f}$. Over the entire temperature range $1/\tau_f$ increases monotonically with temperature, but with a changeover in behavior around 75 K. A Korringa-like relation ($1/\tau_f \propto T$) is observed above this temperature, suggesting that the relaxation of the 4f moments is dominated by scattering from the conduction electrons. In the Korringa limit, the slope of this plot is a

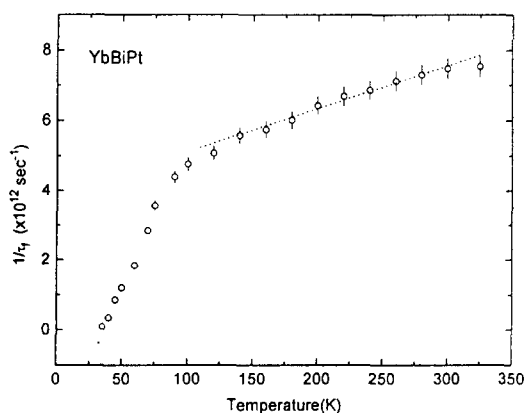


Fig. 3. Temperature dependence of spin correlation rate of Yb local moments derived from the relaxation data assuming $1/T_{1s} \ll 1/T_{1f}$ (see text).

measure of density of states ρ : $1/\tau_f = 8\pi^2 k_B T (J\rho)^2 / h$, where J is the exchange integral. We obtained $J\rho = 0.0879$, consistent with the expected low carrier density deduced from resistivity [1].

In metallic rare earths crystal-field excitations affect the relaxation of the f-moments [11]. The behavior of $1/\tau_f$ below 75 K may therefore be caused by the reduced thermal excitation of a higher crystal-field multiplet as seen in other rare earth systems [12]. In fact inelastic neutron scattering experiments [6] reveal a CEF level near the temperature at which the relaxation rate anomaly is observed. (Note that the observed crossover is independent of the assumptions made regarding the conduction band contributions to T_1 .) Crystal-field effects may not be the only means to account for the temperature dependence of $1/\tau_f$, however. It is possible that the simplifying assumptions made in the derivation of the spin-correlation time (Eq. (2)) are no longer valid below 75 K and that one must take into account the q -dependence of the dynamical susceptibility. This means that spin-correlations at finite q may form below 75 K, leading to a temperature-dependent enhancement of the spin-lattice relaxation rate. Indeed, the observed relaxation phenomena typify a critical slowing down of the electronic spin system. The spin correlation time at 35 K is somewhat long (10^{-11} s), perhaps characteristic of a frustrated spin system, similar to observations in μ SR experiments [4]. We should note that this would not be detectable by the Knight shift, which samples only the long wavelength ($q=0$) component of the spin susceptibility. Further work on this aspect is being planned in the future.

In summary, we have presented NMR data on the heavy electron compound YbBiPt. A transferred hyperfine coupling was attributed to Yb f-moments. A cross-over in the behavior of the spin-correlation rate was observed at about 75 K, from a high-temperature local moment behavior to a low-temperature regime suggestive of crystal field effects. The latter might also be explained by a strongly temperature-dependent finite q contribution to the f-electron spin susceptibility.

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