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Title: μ^+SR Studies of Borocarbides

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μSR STUDIES OF BOROCARBIDES

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Muon-spin-relaxation technique has been utilized to characterize the magnetic properties of the coexisting (superconductivity and antiferromagnetism) borocarbide compounds $R\mathrm{Ni_2B_2C}$, where R = Tm, Er, Ho, and Dy. Some general features of their magnetic ground-state and unusual results obtained in the Ho, Tm and Dy systems are discussed.

1. Introduction

From earlier studies of the rhodium borides and the Chevrel compounds, it is known that ferromagnets generally destroy superconductivity, while antiferromagnets coexist with superconductivity [1]. The recently discovered borocarbides $R\mathrm{Ni_2B_2C}$ [2], which exhibit coexistence of superconductivity and antiferromagnetism for the rare-earth ions R = Tm, Er, Ho and Dy, provide new opportunities for investigating the interplay between magnetism and superconductivity. This is particularly the case because some unusual new features are observed in these systems. For instance, in R = Ho ($T_c = 7.5$ K) an incommensurate magnetic structure has been observed between 5–6 K, where a unique reentrance to the normal state is found for this antiferromagnetic superconductors [3,4]. More recently, $\mathrm{DyNi_2B_2C}$ was found to be superconducting, with $T_c = 6.2$ K, well below $T_N = 10$ K [5]. The latter example provides a chance for studying the influence of superconductivity on magnetism. In this paper, we report our up-to-date μSR studies on all four coexisting borocarbide systems.

2. Experiment

The μSR measurements were carried out at the Paul Scherrer Institute (PSI) in Villigen, Switzerland. Polycrystalline samples were prepared as described in [2], and then powered and pressed into pellets. Single-crystalline samples were prepared via the Ni_2B flux growth method [6], with their crystallographic c-axis perpendicular to the largest sample surface. All the specimens described here were obtained from Ames Laboratory.

3. Results and Discussions

Figure 1 shows the spontaneous muon precession frequency ν as a function of temperature. Except for R = Tm, we find that the zero-temperature frequency is well defined ($\delta\nu/\nu \approx 0.03$) and roughly scales with the free-ion moments. Our Knight-shift measurements in the single-crystal Tm, Ho and Dy systems show that the hyperfine coupling constants are nearly identical for all these systems. Thus, we conclude that for the Er, Ho and Dy systems the ordered moments are about the same as their free-ion moments, and aligned in the ab plane for Ho and Dy, and along the c-axis for Tm. These spin alignments are consistent with the crystal-field splitting. As also evidenced in Fig. 1, the temperature dependence of the order parameter in Er, Ho and Dy shows an abrupt phase transition at T_N, followed by a weak temperature variation below about $0.7T_N$. This is inconsistent with a simple 2-dimensional XY model expected for these systems [4]. A possible explanation for the flat $\nu(T)$ is that the magnon excitations are weakened due to the effects of a long-range magnetic coupling, such as the RKKY or magnetic dipolar interactions [7].

In addition to these general characterizations of the magnetic behavior in the ordered antiferromagnetic state, we also find other interesting features in these systems, which are discussed below.

3.1. $HoNi_2B_2C$

As shown in Fig. 1, a change in $\nu(T)$ is seen near 5 K in $HoNi_2BC$, where a commensurate-to-incommensurate magnetic transition has been observed by neutron studies [3,4]. Figure 2 shows a ZF spectrum in the incommensurate state at T = 5.4 K. The solid line denotes a fit to the relaxation function given by

$$G(t) = J_0(2\pi\nu t)\exp(-\lambda t)\,, \qquad (1)$$

where J_0 denotes the Bessel function and λ is comparable with the dynamic relaxation rate obtained from $\mathbf{S} \perp \mathbf{c}$. This indicates that the muon internal field has a sinusoidal distribution [7].

The magnetic structure of the incommensurate state derived from two neutron studies [3,4] are different: the measurements on the single-crystal

Figure 1 goes here

Fig. 1. Temperature dependence of muon precession frequency observed in $DyNi_2B_2C$, $HoNi_2B_2C$, $ErNi_2B_2C$ and $TmNi_2B_2C$ in zero applied field.

sample indicate two spiral modulations, while the measurements on the polycrystal sample suggest a single spiral. To differentiate these two scenarios, we note that a single spiral modulation yields a nearly uniform field distribution at the muon site since the magnitude of the moments are constant. By contrast, two spirals are possible to give rise to a sinusoidal field distribution if these two modulations have about the same amplitude and their wave vectors are perpendicular to each other [8]. Thus, our μSR results are consistent with two incommensurate spiral modulations [3].

3.2. $TmNi_2B_2C$

As shown in Fig. 1, the frequency in Tm is very small, which corresponds to a frozen moment about 1/30 of the Tm free-ion moment. This small frequency in Tm can not be due to a strongly canted magnetic arrangement, which has been suggested by Cooke *et al.* [9]. As temperature increases, ν persists above T_N and displays a broad maximum near T_N. Between 3 and 25 K, ν follows a Curie-like temperature dependence, $\nu = C/T$ with $C = 4.3$ MHzK (the solid line in Fig. 1). When a sufficiently large longitudinal field is applied, the oscillation disappears, but significant relaxation remains, indicating a quasi-static moments coexisting with other sources of spin fluctuations. Finally, there is no observable change in $\nu(T)$ near the onset of superconductivity ($T_c = 10.5$ K).

Figure 2 goes here

Fig. 2. (a) Crystal and magnetic structure in the commensurate state of $HoNi_2B_2C$. (b) ZF spectrum at 5.4 K for $\mathbf{S} \parallel \mathbf{c}$.

Figure 3 shows the relaxation rate λ as a function of longitudinal field H_L at 4 and 15 K. For $H_{\mathrm{L}} \geq 300$ G the relaxation rate is enhanced with increasing H_{L}. Since $\lambda \sim \Delta\omega^2/(\omega_{\mathrm{L}}^2 + \nu_{\mathrm{f}}^2)$ (where $\Delta\omega$ and ν_{f} correspond to the fluctuating field and rate, respectively) generally decreases with increasing H_{L}, the observed λ suggests that the magnetic state in Tm is very sensitive to a moderate external field ($\sim$ 1 kG). Possible causes for this unusual behavior are (1) a gradual spin flop to the *ab* plane as H_{L} is increased, yielding an enhanced fluctuation field $\Delta\omega$, (2) very small ordered moments corresponding to a nearly non-magnetic ground state. If there is a magnetic excited state right above the ground-state, then an applied field may mix these two states, and lead to a larger relaxation rate.

3.3. $DyNi_2B_2C$

Figure 4 shows the temperature dependence of λ in both the paramagnetic and antiferromagnetic phase of $DyNi_2B_2C$. Above about 20 K, we found no field dependence of λ, indicating sufficiently fast spin fluctuations. However, λ does depend on the initial muon spin polarization, with $\lambda_\perp/\lambda_\parallel \approx 3/4$. The temperature dependence of λ follows the Korringa relaxation process, $\lambda = C/T$, namely the Dy spin are relaxed by scattering via the s-f exchange coupling I to the conduction electrons. Using $C_\parallel = 300$ μsK, we estimate $I \approx 23$ meV, comparable to 13 meV obtained by scaling ΔT_c to the de Gennes factor [10].

Figure 3 goes here

Fig. 3. Dynamic relaxation rate λ as a function of longitudinal field observed in $TmNi_2B_2C$ at 4 K and 15 K.

One of the interesting observation in $DyNi_2B_2C$ is the change of $\lambda(T)$ in zero field near $T_c = 6.2$ K, as shown in Fig. 4. When an external field of 6 kOe is applied along the *c*-axis, T_N is depressed by nearly 1 K and the transition becomes broader. More importantly, the change in $\lambda(T)$ near 6.2 K disappears as superconductivity is destroyed by the external field ($H_L > H_{c2} \approx 5$ kG). This reveals that the onset of superconductivity may modify the Dy spin fluctuations. The exact mechanism is yet to be investigated.

4. Conclusions

We have performed μSR measurements to characterize the magnetic ground-states of the borocarbide systems. We found that the ordered moments for the Ho, Dy and Er systems are about the same as their free-ion moments and aligned according to the crystal-field splitting. For the Tm system the ordered moments are much smaller. This magnetic state also persists up to at least $10T_N$, and is easily modified by a moderate external field, suggesting a weakly ordered magnetic ground-state, possibly due to the crystal-field splitting. For the Ho system our analysis of the ZF spectrum allows us to discuss the magnetic structure of this incommensurate state, which distinguishes between different results obtained by two neu-

Figure 4 goes here

Fig. 4. Dynamic relaxation rate vs. temperature in (a) paramagnetic phase and (b) antiferromagnetic phase of single-crystal $DyNi_2B_2C$.

tron scattering studies [3, 4]. For the Dy system we find a change in the temperature dependence of the dynamical relaxation rate at T_c, indicating the possible influence of superconductivity on magnetism.

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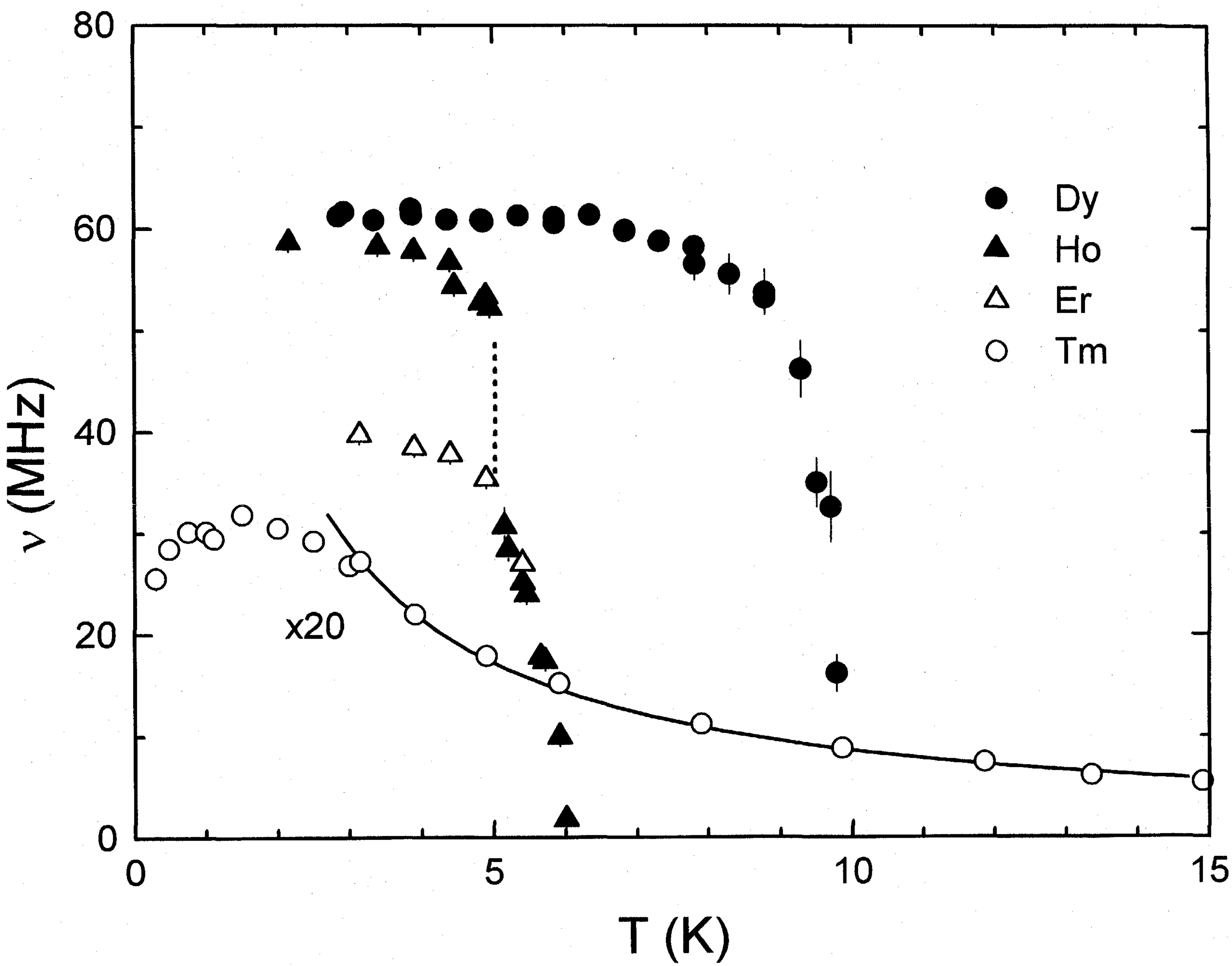
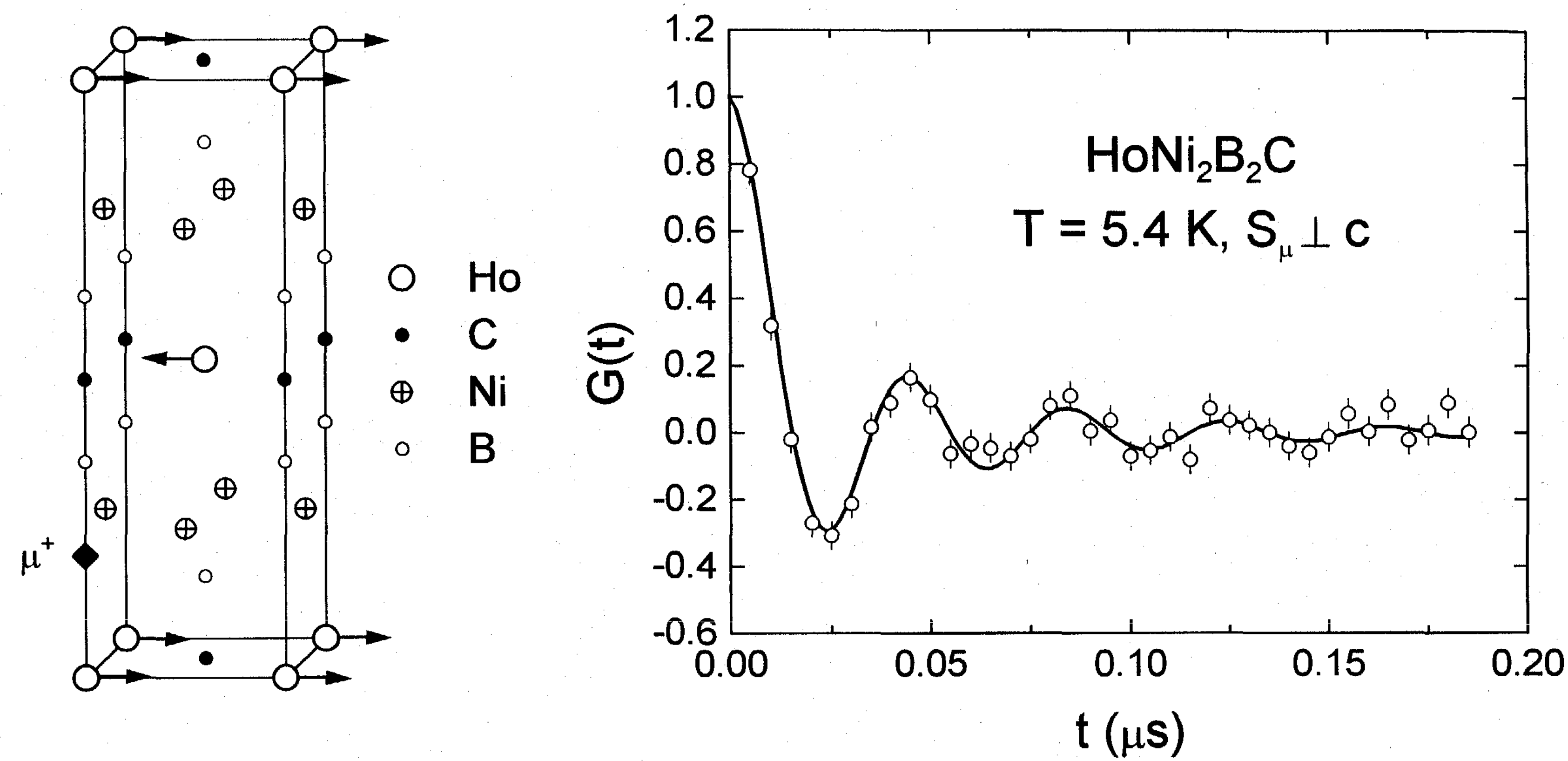


μSR'96 Nikko, LPLe-2: Figure 1



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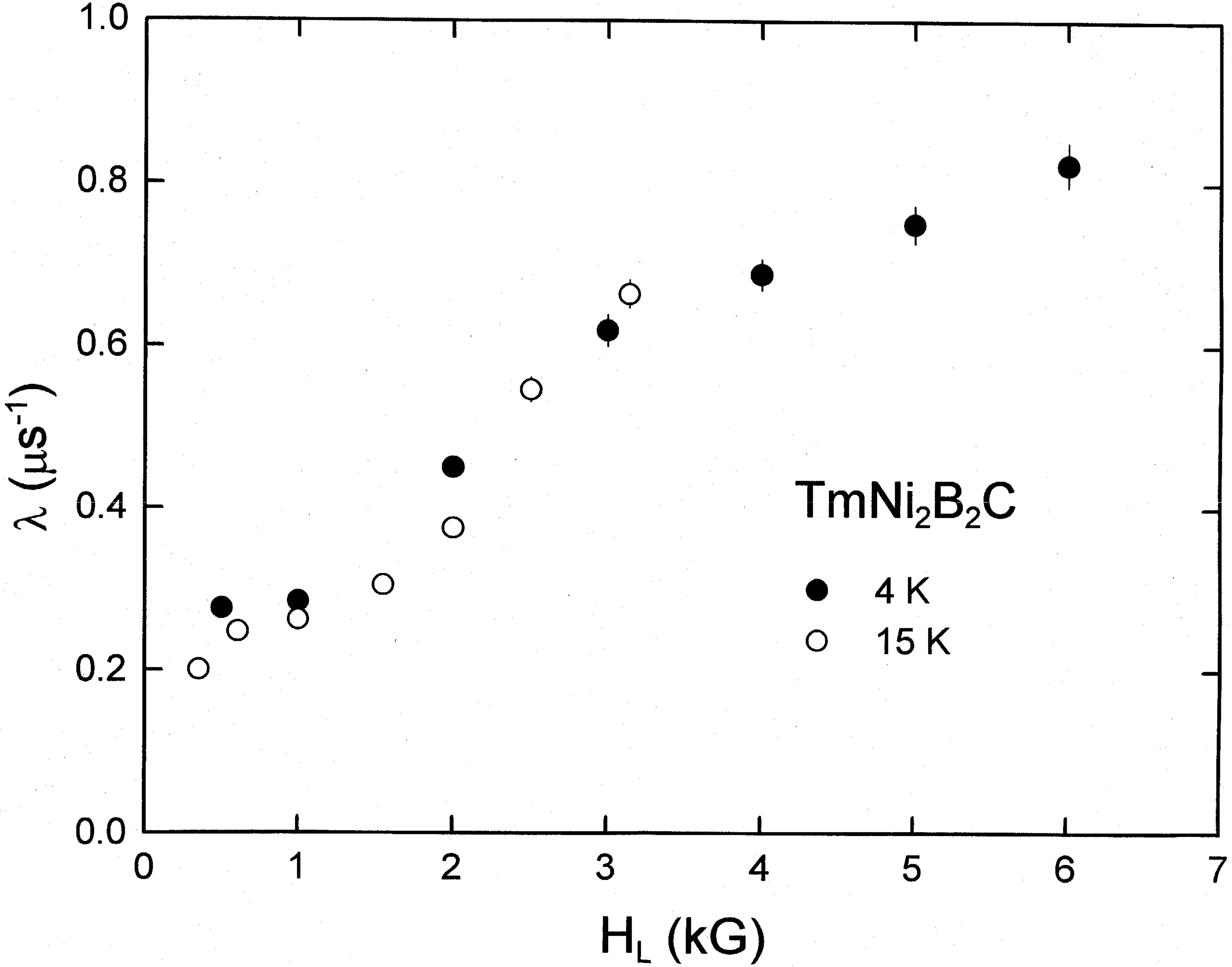

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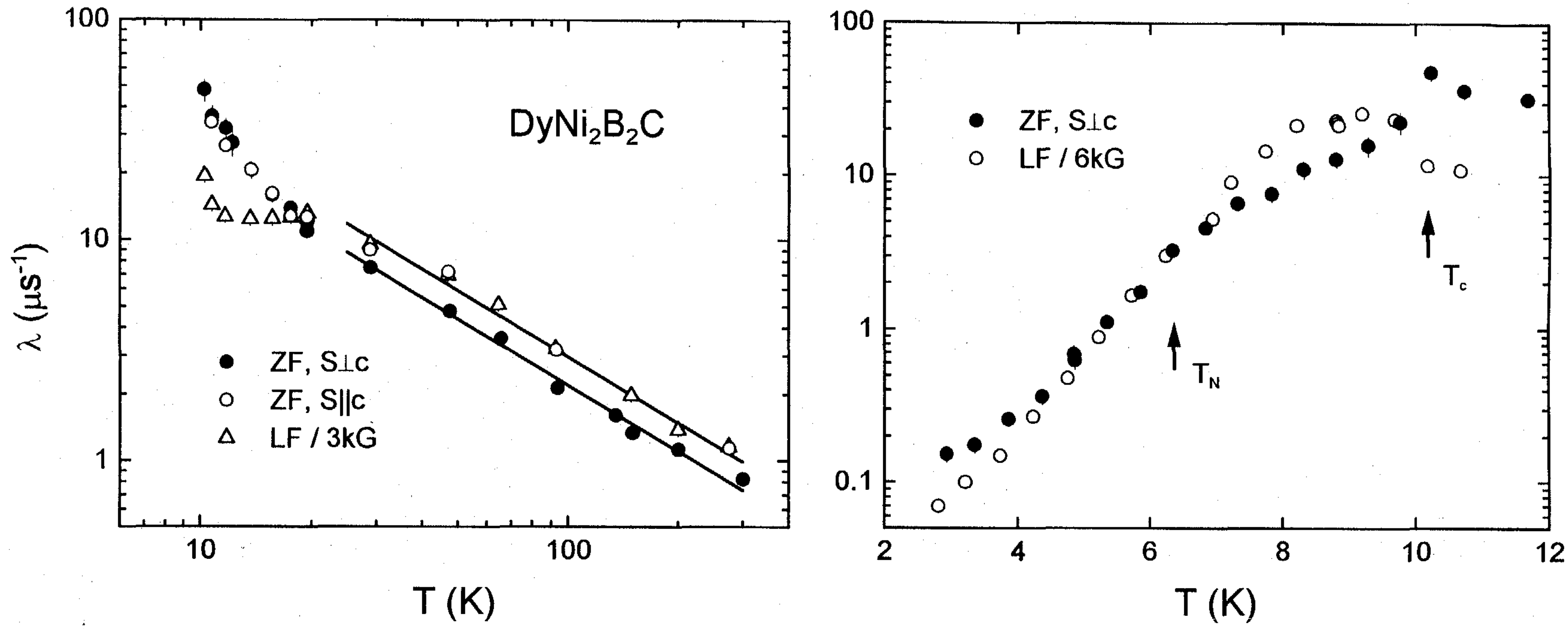

μSR'96 Nikko, LPLe-2: Figure 4