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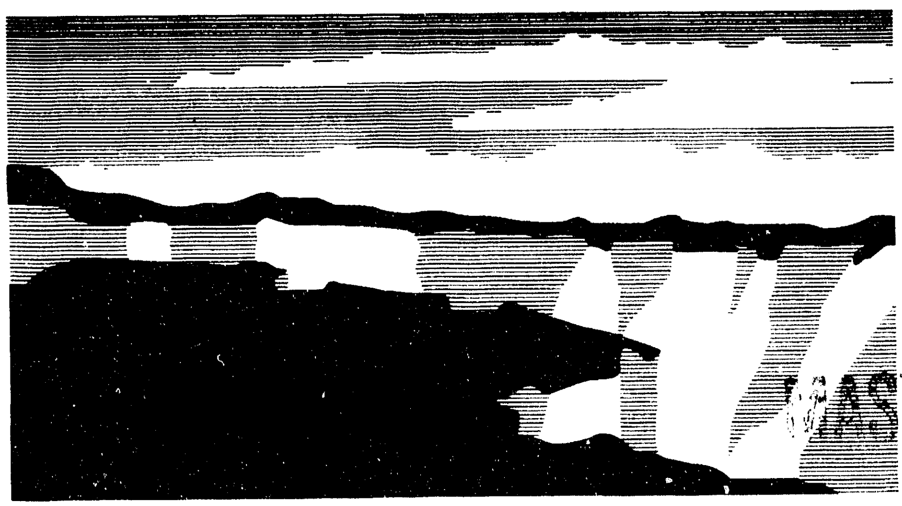

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# $\mu^+$SR Studies of Magnetic Properties of Boron Carbide Superconductors

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## Abstract

Positive-muon spin rotation ($\mu^+$SR) has been carried out in the recently-discovered rare-earth boron carbide superconductors $R\mathrm{Ni_2B_2C}$, $R$ = Ho, Er and Tm. For $R$ = Ho and Er zero-field $\mu^+$SR measurements showed a well-defined internal field below the Néel temperatures ($T_N \approx 5.5$ K) coexisting with the superconducting state down to 0.1 K. The observed temperature dependence of the order parameter in $R$ = Ho is consistent with a 2-dimensional Ising model. For $R$ = Tm a spontaneous internal field appears above 30 K, whose magnitude saturates below about 3 K at a value corresponding to a rare earth moment much smaller than for Ho and Er. Transverse-field $\mu^+$SR measurements in $R$ = Tm showed a superconducting penetration depth $\lambda = 1,200$ Å. The temperature dependence of $\lambda$ is consistent with conventional s-wave pairing.

Keywords: superconductivity, magnetism, $\mu^+$SR

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Recently, superconductivity has been discovered in several intermetallic boron carbide compounds, with the highest $T_c$ being above 20 K [1,2]. In the series $R\mathrm{Ni_2B_2C}$, superconductivity is depressed gradually with increasing rare-earth ($R$) de Gennes factor $G$, and is quenched completely for $R =$ Dy and Tb, which have the largest $G$. For the systems with middle-ranged $G$ ($R =$ Ho, Er and Tm), magnetic order is found to coexist with superconductivity [3]. These features are reminiscent of the rare-earth rhodium borides $R\mathrm{Rh_4B_4}$ and Chevrel phases $R\mathrm{Mo_6S_8}$ [4]. In order to understand their magnetic nature, as well as the interplay between superconductivity and magnetism in these systems, we performed muon spin relaxation ($\mu^+$SR) studies [5] in $R\mathrm{Ni_2B_2C}$, with $R =$ Ho, Er and Tm.

The $\mu^+$SR experiments were performed at the Paul Scherrer Institute (PSI) in Villigen, Switzerland, using the Low Temperature Facility and the General Purpose Spectrometer. Polycrystalline samples of $R\mathrm{Ni_2B_2C}$ were prepared as previously described [2], and then powdered and pressed into pellets which were attached to the cryostat cold finger.

The zero-field $\mu^+$SR spectra were well described by a two-component relaxation function given by:

$$A(t) = A\left[\frac{1}{3}\exp(-\lambda_1 t) + \frac{2}{3}\exp(-\lambda_2 t)\cos(2\pi\nu_\mu t)\right]. \tag{1}$$

The first term of Eq. (1) corresponds to the relaxation of the muon polarization parallel to the internal field and thus the relaxation rate $\lambda_1$ reflects the dynamic properties of the host moments. The second term describes muon precession transverse to the internal field and therefore $\lambda_2$ represents relaxation due both to dynamic effects and static field inhomogeneities. In a polycrystalline specimen the ratio of the amplitudes is 1:2.

For $R =$ Ho and Er a well-defined muon-spin precession frequency $\nu_\mu$ is observed below the Néel temperatures $T_N \approx 5.5$ K. ($\nu_\mu/\lambda_2 \approx 6$ at low temperatures.) The observation of $\nu_\mu$ in zero-external field indicates the onset of magnetic order, where the spontaneous magnetization is proportional to $\nu_\mu$. The frequency is 59 MHz for Ho and 40 MHz for Er at the lowest measured temperature of 0.1 K and 3 K, respectively. The large values of $\nu_\mu$ (about 10 times larger than observed in $\mathrm{LaCu_2O_4}$ [6]) indicate a frozen moment of several $\mu_B$, consistent with that expected from Ho and Er. The temperature dependence of $\nu_\mu$ is similar in both the Ho and Er compounds (Fig. 1).

For $R =$ Ho, $\nu_\mu$ remains almost constant from 0.1 K up to 5 K and then drops rapidly. The weak temperature dependence of $\nu_\mu$ below 5 K indicates a strong suppression of low-energy excitations, such as spin-waves, and the rapid change of $\nu_\mu$ around $T_N$ indicates an abrupt and possibly first-order transition. However, neither our ZF-$\mu^+$SR nor susceptibility measurements show signs of thermal hysteresis near $T_N$.

We are able to fit the temperature dependence of $\nu_\mu$ quite well using a 2-dimensional (2D) Ising model. This is demonstrated by the solid line in Fig. 1, for an intra-plane ferromagnetic exchange interaction $J/k_B = 2.4$ K. The temperature dependence of the sublattice magnetization is not unreasonable considering the crystal structure of $HoNi_2B_2C$ and the possible effects of the crystal field splitting. These compounds have a 2D crystal structure, where the HoC layers alternate with the $Ni_2B_2$ layers [7], with an intra-plane Ho-Ho distance of 3.53 Å and interplane Ho-Ho distance of 6.32 Å. A strong anisotropy has been observed in the normal-state susceptibility, which leads to the frozen Ho moments in the basal *ab*-plane below $T_N$ [9]. Crystal field parameters have not been determined for this system. We note, however, that the Ho-ion ground state in $HoRh_4B_4$, also possessing a tetragonal structure, exhibits an Ising behavior due to crystal field splitting [8].

Figure 1 also shows the temperature dependence of $\nu_\mu$ in $TmNi_2B_2C$. A muon precession frequency appears at the rather high temperature of 30 K, indicating the existence of a spontaneous internal field. Between 20 and 3 K, $\nu_\mu$ is inversely proportional to temperature (dashed line), $\nu_\mu = CT^{-1}$, where C=4.3 MHz·K, consistent with previous ZF-$\mu^+$SR studies by Cooke *et al.* [10]. Below 3 K, $\nu_\mu$ starts to saturate, reaching a maximum of 1.6 MHz near 1.5 K. In comparison with $R$ = Ho and Er, the temperature dependence of $\nu_\mu$ for $R$ = Tm is distinctively different. No abrupt onset of magnetic order is observed. Furthermore, the local field below $T_N$ is 25–35 times smaller for $R$ = Tm than for Ho and Er.

The magnitude of $\nu_\mu$ found below 3 K in $TmNi_2B_2C$ corresponds to a local field of about 120 G. Assuming dipolar $\mu^+$-Tm coupling, this corresponds to a frozen moment of order 0.1 $\mu_B$, much smaller than the free-ion value for Tm (7.7 $\mu_B$) deduced from the susceptibility [3]. If one associates the internal field seen by the muon with Tm ordering, the reduced frozen-moment could be due to crystal-field effects and/or rapid, limited-amplitude fluctuations. Longitudinal field measurements at 0.83 K were performed to elucidate the spin dynamics. At $H_L = 1$ kG, the precession signal disappears, but significant $\mu^+$ relaxation is still observed. This relaxation rate was changed only slightly in applied field up to 10 kG, indicating fluctuation rates at least as large as $\gamma_\mu H_L \sim 10^9$ s$^{-1}$. The simultaneous occurrence of precession in a local field of 120 G and fluctuation rates of order $10^9$ s$^{-1}$ can only occur if the fluctuations are of limited amplitude (giving rise to a small frozen moment), or if there are two independent sources for the local field sensed by the muon: one producing precession and the other relaxation. Further experiments and analysis will be undertaken to explore these possibilities.

It is generally difficult to investigate the superconducting properties of these magnetic superconductors because the relaxation rate from the magnetic ions is often too large and temperature dependent to permit a clear observation of field broadening due to the superconducting vortex lattice. For $R$ = Ho we found it even impossible to determine the superconducting transition temperature $T_c$ using $\mu$SR. The strong internal magnetic fields apparently cause the same problems for other techniques, such as magnetization measurements. The superconducting penetration depth $\lambda$, for instance, has not yet been determined for the Ho or Er compounds.

Here we report TF-$\mu^+$SR measurements in $TmNi_2B_2C$, where we are able to separate the superconducting signal from large magnetic background. The spectra were fitted with a phenomenological formula

$$A(t) = A_s \exp(-\sigma_s^2 t^2/2) \exp(-\lambda_s t) \cos(2\pi\nu_s t) + A_b \exp(-\sigma_b^2 t^2/2) \cos(2\pi\nu_b t), \qquad (2)$$

where the subscript $s$ and $b$ denote signals from sample and background silver, respectively. We find $A_b/(A_s + A_b) \approx 10\%$.

Figure 3 shows the temperature dependence of $\sigma_s(T)$ (solid circles) and $\lambda_s(T)$ (open triangles) under a transverse field of 1 kG. A rather sudden enhancement of $\sigma_s$ is found below 10.5 K (which corresponds to the reported $T_c$ [2]), while $\lambda_s$ varies smoothly with temperature. We thus attribute $\sigma_s$ to superconductivity and $\lambda_s$ to magnetism. Multiplication of Gaussian and exponential relaxation functions in Eq. (2) indicates that muons see both relaxation processes simultaneously, indicating a coexistence of magnetism and superconductivity. The values of $\lambda_s$ are also comparable with the zero-field relaxation rates attributed to magnetic relaxation.

In a type-II superconductor, the formation of the vortex lattice leads to a field broadening $\Delta B$, reflecting the superconducting penetration depth $\lambda$ as $\sigma \propto \Delta B \propto \lambda^{-2}$. Since $\sigma_s$ above $T_c$ is nearly temperature independent (up to 25 K), we assume that this residual relaxation rate also remains unchanged below $T_c$. Thus $\sigma^2(T) = \sigma_s^2(T) - \sigma_s^2(T_c)$. We then find that $\sigma(T)$ can be best described by the weak-coupling BCS theory (solid line in Fig. 3). This is consistent with a conventional s-wave pairing in $TmNi_2B_2C$. The extrapolated relaxation rate $\sigma(0) = 7.5\ \mu s^{-1}$ yields a powder-averaged $\lambda = 1{,}200$ Å. This penetration depth is comparable with the in-plane $\lambda = 1{,}500$ Å obtained in the non-magnetic superconductor $YNi_2B_2C$ [12]. Using the upper critical field $H_{c2} \approx 2.5$ Tesla obtained by magnetization measurements [3], we further calculate the Ginzburg-Landau parameter $\kappa \approx 10$ and the lower critical field $H_{c1} \approx 250$ G.

In conclusion, we have investigated magnetic ordering in $RNi_2B_2C$ ($R$ = Ho, Er and Tm) using muon spin relaxation. For $R$ = Ho and Er spontaneous magnetic order was observed below $T_N = 5.5$ K; the temperature dependence of the sublattice magnetization is consistent with a 2D Ising system. For $R$ = Tm, an oscillation frequency appears below 30 K, saturating at low temperatures with a value about 30 times smaller than seen in other two compounds. Significant magnetic fluctuations were also found below the expected magnetic ordering temperature for $R$ = Tm. The penetration depth was measured in the superconducting state of the Tm compound. Further studies of magnetism in these rare-earth boron carbide systems should stress the difference between the Tm and Ho compounds.

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## FIGURES

FIG. 1. Temperature dependence of zero-field muon-spin precession frequency observed in $HoNi_2B_2C$, $ErNi_2B_2C$ and $TmNi_2B_2C$. The solid line denotes 2-dimensional Ising model, and the dashed line denotes a reciprocal temperature dependence.

FIG. 2. Temperature dependence of transverse-field Gaussian ($\sigma_s$) and exponential ($\lambda_s$) relaxation rate observed in $TmNi_2B_2C$. $\sigma_s$ and $\lambda_s$ are defined according to Eq. (2). The solid line refers to weak-coupling BCS theory.

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