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Authors
Gavilano, JL
Ambrosini, B
Ott, HR
et al.

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Low-temperature NMR studies of SrB_6

J.L. Gavilano[a,]*, B. Ambrosini[a], H.R. Ott[a], D.P. Young[b], Z. Fisk[b]

[a]*Laboratorium für Festkörperphysik, ETH-Hönggerberg, CH-8093 Zürich, Switzerland*
[b]*National High Magnetic Field Laboratory, Tallahassee, FL 32306-4005, USA*

Abstract

We report the results of a ^{11}B nuclear magnetic resonance (NMR) study of SrB_6 at temperatures between 0.1 and 30 K and in a magnetic field of 4.74 T. Below 30 K the NMR spectrum is temperature independent but the spin–lattice relaxation rate T_1^{-1} exhibits different features in two different temperature regimes. At high temperatures, between 30 K and a field-dependent crossover temperature T_B between 0.5 and 2 K, T_1^{-1} is almost temperature independent. We point out that for T in the crossover temperature range the magnitude of T_1^{-1} of SrB_6 is distinctly larger than for LaB_6, a metal with a charge carrier concentration at least two orders of magnitude higher than that of SrB_6. A possible cause for this behavior maybe the very weak itinerant ferromagnetism that has subsequently been established to occur in nominally pure SrB_6. At low temperatures, below T_B, T_1^{-1} decreases substantially with decreasing temperature confirming a cross-over or phase transition phenomenon as observed by measurements of thermal and transport properties.

Keywords: SrB_6; Low carrier system; Magnetism; NMR

SrB_6 is a semimetal with a very small itinerant charge carrier density, of at least two orders of magnitude smaller than that of LaB_6 [1]. Its low-temperature properties seem to depend critically on details of the electronic structure [2]. From the results of magnetization measurements, it has recently been inferred that SrB_6 orders ferromagnetically with an onset temperature T_C of the order of 900 K and involving very small magnetic moments (unpublished results).

Our NMR studies were performed at the B sites which form a network of octahedra joined by covalent bonds. The symmetry of the B sites is 4 mm, which allows for a nonzero field gradient, with axial symmetry. In Fig. 1 we display an example of the ^{11}B-NMR spectrum for SrB_6 measured at a frequency of 64.81 MHz and at a temperature of 1.31 K. The shape of the spectrum is that of a characteristic powder pattern for spin $\frac{3}{2}$ nuclei, where a small quadrupolar perturbation splits the Zeeman lines. The Knight shift is very small and temperature independent.

The quadrupolar parameters and the width of the NMR lines are very similar to those of LaB_6 [3]. However, one expects that for a powdered SrB_6 sample at $T \ll T_C$ the ferromagnetic order would result in a distribution of demagnetization fields of approximately $4\pi m$ (where m is the magnetization) and a corresponding broadening of the NMR lines. From this consideration and a comparison of our data with that of LaB_6 we infer that the ordering below T_C involves very small magnetic moments of less than $10^{-2}\mu_B$ per unit cell, in agreement with the results of the magnetization measurements on the same sample (unpublished results).

The T_1 measurements were performed on the narrow central line of the $+\frac{1}{2} \leftrightarrow -\frac{1}{2}$ ^{11}B nuclear transition, in an applied field of 4.74 T (see Fig. 1). In Fig. 2 we display $T_1^{-1}(T)$ for SrB_6. Two temperature regimes with qualitatively different T-dependencies for the spin–lattice relaxation may be distinguished. At high temperature, above $T_B \approx 2$ K, T_1^{-1} is approximately T-independent. At temperatures below T_B, T_1^{-1} decreases substantially with decreasing temperatures, suggesting a cross-over

* Corresponding author. Tel.: + 41-1-633-1077; fax: + 41-1-633-2245.

E-mail address: gavilano@solid.phys.ethz.ch (J.L. Gavilano)

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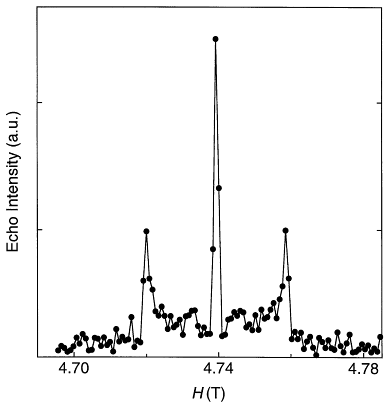

Fig. 1. ^{11}B-NMR spectrum of SrB_6 measured at 64.81 MHz and 1.13 K.

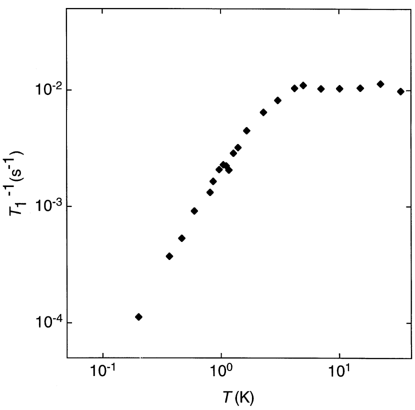

Fig. 2. $T_1^{-1}(T)$ for SrB_6 measured in an applied magnetic field of 4.74 T.

phenomenon previously indicated by specific heat and resistivity data.

The temperature dependence of T_1^{-1} for SrB_6 is not compatible at all with that expectated for a paramagnetic metal or a semiconductor. Furthermore the magnitude of T_1 at $T \approx T_B$ is surprisingly large, even larger than that for LaB_6. This is not expected because LaB_6 has a much larger charge carrier concentration. We rule out magnetic impurities as a possible source for the anomalous relaxation because these result in a characteristic temperature and field dependencies for T_1 [4], not observed in our experiments. In addition, in the absence of spin diffusion, which seems to be the case here, the relaxation of paramagnetic impurities also implies a distribution of T_1s [4], again not observed here.

In view of the above one is tempted to associate the relaxation with excitations related to the 'small-moment ordering'. However, in the temperature range of our experiments ($T \ll T_C$) this yields a $T_1^{-1} \propto T$ [5], instead of the observed T-independent relaxation. The crossover phenomenon at T_B only adds another puzzle to the unexpected features of this seemingly simple compound.

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