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Low-temperature NMR studies of SrB$_6$

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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ferromagnetically with an onset temperature $T_C$ of the order of 900 K and involving very small magnetic moments (unpublished results).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 $\frac{1}{2}$ 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 $T < T_C$ 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...
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.

The temperature dependence of $T_1^{-1}$ for SrB$_6$ is not compatible at all with that expected 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 $T_1$'s [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.

References