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Hyperfine field at Mn in the intermetallic compound $LaMnSi_2$ measured by PAC using ^{111}Cd nuclear probe

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Abstract Magnetic hyperfine field at Mn site has been measured in the orthorhombic intermetallic compound $LaMnSi_2$ with PAC spectroscopy using radioactive ^{111}In-^{111}Cd nuclear probe. Samples of $LaMnSi_2$ were prepared by melting pure metallic components in stoichiometric proportion in an arc furnace under argon atmosphere. The samples were sealed in a quartz tube under helium atmosphere, annealed at 1000 °C for 60 h and quenched in water. Samples were analyzed with X-ray diffraction method. ^{111}In was introduced in the samples by thermal diffusion at 1000 °C for 60 h. PAC measurements were carried out with a six BaF_2 detector spectrometer at several temperatures between 50 K and 410 K. Results show well defined quadrupole and magnetic interactions at all temperatures. The magnetic hyperfine field (B_{hf}) measured at 50 K is 7.1(1) T. The temperature dependence of B_{hf} follows the normal Brillouin-like behavior expected for a simple ferromagnetic ordering. The ferromagnetic transition temperature (T_c) was determined to be 401(1) K.

Keywords Intermetallic compounds · Hyperfine field · Perturbed γ-γ angular correlation

1 Introduction

Intermetallic compounds of the series $RMnSi_2$, where R is a rare-earth element, crystallize in an orthorhombic $TbFeSi_2$-type structure belonging to space group Cmcm. This structure can be described as built from the alternating (010) planes containing R, Mn and Si atoms in a sequence R-Si-Mn-Si-R-Si-Si-R-Mn-Si... atomic planes. This structure is closely related

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to the tetragonal $ThCr_2Si_2$-type structure with space group I4/mmm where the same atoms lie in alternate layers along the c-axis in a sequence Th-Si-Cr-Si-Th-Si-Cr-Si. . . An example of this type of structure is the compound $LaMn_2Si_2$. In both $RMnSi_2$ and RMn_2Si_2 the silicon atoms form tetrahedra around the transition metal atom Mn. During the last several years the magnetic properties of $RMn_2(Si,Ge)_2$ compounds have been studied by magnetometric and neutron diffraction methods as well as hyperfine interaction techniques such as PAC and Mössbauer spectroscopy [1–5]. These compounds exhibit strong antiferromagnetic behavior with a transition to ferromagnetic ordering near room temperature. The main characteristic is the presence of long range ordering, of intra- and inter- planar Mn moments which, persists even at temperatures above room temperature.

The much less studied $RMnSi_2$ compounds have been investigated in the past by X-ray and neutron diffraction and magnetization measurements [6–8]. The Mn sub-lattice orders ferromagnetically up to high temperatures with T_c values well above room temperature. At lower temperatures, additional magnetic transitions due to the ordering of rare earth sublattices appear in some cases. The only magnetic atom in this compound is Mn, which carries a local magnetic moment of 2.6 μ_B with the moments aligned along the stacking axis (c-axis) of the structure [6]. The ferromagnetic behavior of this compound is mainly due to long range Mn-Mn interactions. Despite some of the earlier studies on the structural and magnetic properties of $LaMnSi_2$ there is no report on the investigation of this compound using microscopic techniques. In this work we report on the results of temperature dependence of magnetic hyperfine field (B_{hf}) in $LaMnSi_2$ measured by Perturbed Angular Correlation (PAC) spectroscopy using ^{111}In-^{111}Cd nuclear probe. PAC measurements in this compound using ^{140}La-^{140}Ce nuclear probe are in presently in progress in our laboratory.

2 Experimental procedure

The $LaMnSi_2$ samples were prepared by melting the high purity elements (La = 99.9 %, Mn = 99.99 % and Si = 99.999 %) in stoichiometric proportions in an arc furnace under argon atmosphere. After melting, the samples were sealed in a quartz tube under helium atmosphere and annealed for 60 hours at 1000°C and then quenched in water. The radioactive probe^{111} In was introduced in the samples by thermal diffusion. Approximately 50 μCi of carrier free ^{111}In in the form of dilute $^{111}InCl_3$ solution was deposited on the sample, in the form of a small disc, cut out from the spherical sample ingot and dried under infrared lamp. The sample was then sealed in a quartz tube under helium atmosphere and heated at 1000°C for 60 hours in a tubular resistance furnace. PAC measurements were performed by using a six BaF_2 detectors spectrometer. The well-known gamma cascade of 172-245 keV in ^{111}Cd populated from the electron capture (EC) decay of ^{111}In was used for the PAC measurements. Thirty simultaneous delayed $\gamma\gamma$-coincidence spectra (six with detectors at 180° and 24 with detectors at 90°) are obtained with the six detectors spectrometer. Further details about the PAC technique and experimental method can be found in references [9, 10]. PAC spectra were measured at different temperatures between 50 K and 410 K. For low temperature measurements a closed cycle helium loop cryogenic device was used. The measurements at temperatures higher than room temperature were carried out using a small graphite furnace.

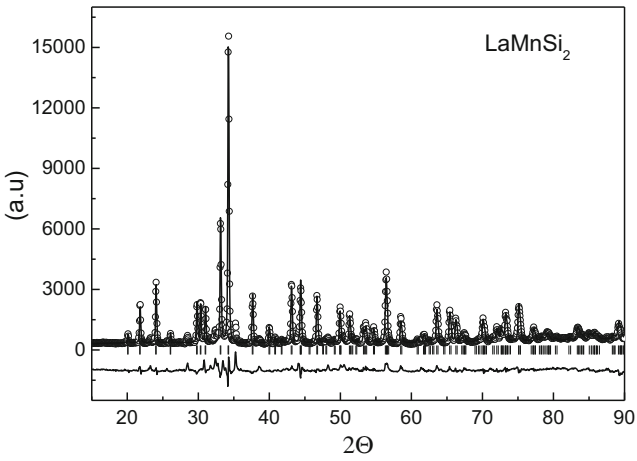

Fig. 1 X-ray diffraction pattern for $LaMnSi_2$ sample and the fitted data

3 Results and discussions

$LaMnSi_2$ samples were analyzed by powder X-ray diffraction and their spectra were refined with Rietveld method using Rietica software [1]. The orthorhombic crystal structure was confirmed for $RMnSi_2$. The present X-ray diffraction results are in excellent agreement with the previous measurements reported in [7]. The X-ray diffractogram is shown in Fig. 1.

The results of PAC measurements were analyzed by using a model including combined electric quadrupole and magnetic dipole interactions. At all temperatures unique magnetic and quadrupole interactions were observed. Some of the PAC spectra for the compound $LaMnSi_2$ measured at different temperatures are shown in Fig. 2.

In the analysis of PAC spectra it was assumed that the nuclear probe ^{111}Cd substitutes Mn atoms. This assumption is justified from the following arguments: a) earlier PAC results for $LaMn_2(Si,Ge)_2$ and $CeMn_2(Si,Ge)_2$ [2–4] show that the major fraction of ^{111}Cd probes are substituting the Mn atoms b) results of *ab initio* calculations in the case of $CeMn_2(Si,Ge)_2$ compounds confirm this assignment [11]. Since the crystal structure of $LaMnSi_2$ is very similar to $LaMn_2Si_2$, it is reasonable to assume that ^{111}Cd probe replaces Mn atoms in $LaMnSi_2$ rather than La or Si. The temperature dependence of the magnetic hyperfine field B_{hf} is shown in Fig. 3. The experimental data are quite well fitted by the Brillouin function for the host magnetization with spin 5/2. In order to determine a precise value of Curie temperature (T_c) the experimental hyperfine field values near the transition temperature were fitted by the well-known power law $B_{hf}(T) = B_0(1\text{-}T/T_c)^{\beta}$ for magnetic materials. Here β is the exponent and B(0) is the hyperfine field value at 0 K. The resulting values of β and T_c are 0.43(1) and 401(1) K respectively. The present value of T_c is considerably higher than the previously reported value of 386 K from the magnetization measurement [7]. The measured exponent β is found to be very similar to those for compounds $LaMn_2(Si,Ge)_2$ and $CeMn_2(Si,Ge)_2$ and therefore indicates that the nature of magnetic order is similar in two cases. Since ^{111}Cd probe is nonmagnetic (sp-element) it only probes the magnetic interaction from the neighboring Mn atoms through long range Mn-Mn interactions.

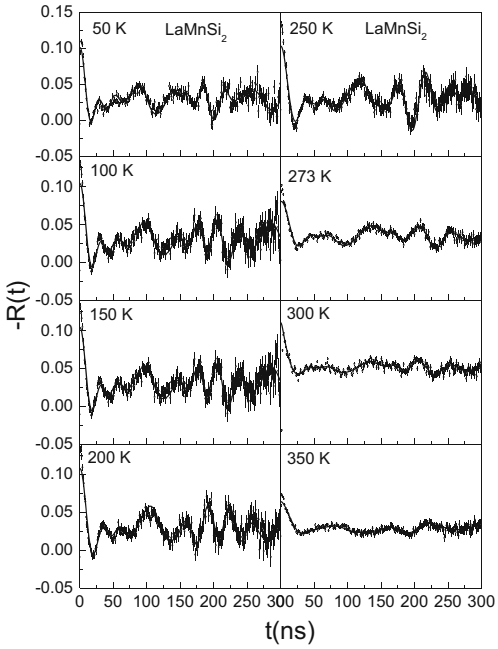

Fig. 2 Perturbation functions for $LaMnSi_2$ with ^{111}Cd probe nuclei at temperatures from 50 K to 350 K. Solid lines are least squares fit of the theoretical functions to the experimental data

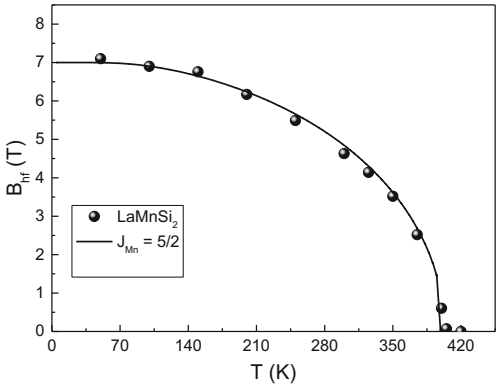

Fig. 3 Temperature dependence of the magnetic hyperfine field of ^{111}Cd in $LaMnSi_2$. The solid line corresponds to the Brillouin function for $J = 5/2$

4 Conclusions

PAC spectroscopy has been utilized to measure the hyperfine field in the orthorhombic $LaMnSi_2$ compound using ^{111}In-^{111}Cd nuclear probe in the temperature range 50–410 K. The ^{111}Cd probe nuclei are found to substitute only Mn atom sites. A well defined magnetic interaction was observed at all themperatures. The temperature dependence of magnetic hyperfine field shows the Brillouin type behavior. The Curie temperature was determined to be $T_c = 401(1)$ K. This value is considerably higher than the earlier reported value of 386 K [7]. The ferromagnetic order in $LaMnSi_2$ is attributed to the magnetic ordering of the Mn sublattice where Mn magnetic moments are aligned along the stacking axis (c-axis). The long range Mn-Mn interaction of RKKY type is believed to be the mechanism responsible

for the observed hyperfine field at Mn in this compound. The PAC measurements in this compound using the ^{140}La-^{140}Ce nuclear probe are presently in progress in our laboratory.

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