Study of hyperfine fields in Co$_2$ScSn Heusler alloy

N Lakshmi*, Anil Pandey & K Venugopalan
Department of Physics, Mohanlal Sukhadia University, Udaipur, India
*E-mail : nambakkal@yahoo.com

Received 23 July 2004; revised 23 December 2004; accepted 4 January 2005

Heusler alloy Co$_2$ScSn has been prepared and studied. X-ray diffraction studies showed the structure to be L2$_1$ type. Using Sn-119, Mössbauer studies were carried out to investigate the magnetic hyperfine fields present at the Sn site in this alloy. Applicability of the Blandin and Campbell model for this alloy is tested by calculating the hyperfine fields at Sn site. The results of these calculations are compared with the experimental results.

Keywords: Hyperfine field distribution; Heusler alloys; Sn-119 Mössbauer spectroscopy
IPC code: G01J3/28

1 Introduction
Recently, there has been a lot of interest in Heusler alloys because theoretical band structure calculations predict that some of the Heusler alloys are half-metallic ferromagnets. Although Groot et al. had predicted the Heusler alloys NiMnSb and PtMnSb to be half-metallic as early as 1983; this property has given rise to a new surge of interest in these systems due to the possibility of use of Heusler alloys as material for spin electronic devices. This has prompted a search for stable, room temperature ferromagnetic Heusler systems. These systems are attractive because of the considerable change in their magnetic properties that can be made by altering the chemical ordering, substituting for one or more components by other atoms, mechanical treatment, etc.

Ternary alloys of stoichiometric composition bearing the general formula, X$_2$YZ are generally termed as Heusler alloys. In this class of alloys, X and Y are transition elements like Ni, Co, Pd, Pt, Fe, etc and Z is an sp-element like Si, Al, Ge, etc. Magnetic properties of X$_2$YZ alloys arise from magnetic moments on transition metal atoms located on either the X or Y sites and so these alloys offer excellent systems for the study of the mechanism of magnetic interactions. Extensive studies have been carried out on Heusler alloys using hyperfine methods, to understand the coupling mechanism responsible for magnetic hyperfine fields at non-magnetic sites.

The hyperfine field systematics in cobalt based Heusler alloys are quite different from systematics in Heusler alloys which contain only Mn as the magnetic atom. In cobalt rich Heusler alloys of the form Co$_2$MnZ, which contain 25% of Mn, magnetic moment exists at two inequivalent lattice sites. In these alloys, Mn carries about 4 $\mu_B$ magnetic moment, while Co carries moments ranging between 0.3 and 1 $\mu_B$, depending on the Z element. The Blandin-Campbell model has been used fairly successfully to describe hyperfine fields at the Y site in Co$_2$YZ (Y$\neq$Mn) type of alloys by a proper choice of sp valency. On the other hand, the volume overlap model proposed by Stearns would not be suitable for these alloys since the sp impurities are only second nearest neighbour to the magnetic atom. As Sn is one of the components in Co$_2$ScSn, this alloy offers an excellent possibility of testing the applicability of the Campbell-Blandin model by calculating the hyperfine field at the Z site.

2 Experimental Details
The starting materials for the preparation of the alloys were obtained from M/s Spex Industries Inc., USA. These were of at least 99.99% purity. Sc and Sn were in the form of granules while Co was in the form of sheets. The constituent materials were weighed out in the desired atomic ratio and melted together in an arc furnace in a flowing argon atmosphere. Weight loss was observed to be less than 1%. The alloy was
seen to be hard and brittle. The samples were crushed and ground to fine powder. They were then sealed into quartz ampoules evacuated to $10^{-5}$ torr and kept in a furnace at 800°C for 7 days. They were then allowed to cool down to room temperature in the furnace itself.

Approximately 300 mg each of the powdered sample (400 mesh size) was used for X-ray characterization of the prepared alloys. X-ray diffraction studies of the samples were done at room temperature with a Philips X-ray diffractometer using Cu-Kα radiation.

The samples for the Mössbauer measurements were prepared by weighing out the required amount of the powdered material and packing into copper rings after thoroughly mixing with boron nitride. The quantity of material used was such as to have a thickness of 9 mg/cm² of Sn. The source used was a 5 milli Curie Sn¹¹⁹ in CaSnO₃ matrix. Data were accumulated for an average of 72 h. Low temperature studies were performed using a closed cycle refrigerator.

### 3 Results and Discussion

The X-ray diffraction spectra were analyzed and the peaks were indexed using the PDP program. It was observed that the alloy was homogeneous and well ordered with a lattice parameter 6.19±0.01 Å. This matches with previously reported values for this alloy.

Sn-¹¹⁹ Mössbauer spectra recorded at 50 K for Co₂ScSn is given in Fig. 1. The Mössbauer spectrum was fitted using the Window’s method which fits for a distribution of magnetic hyperfine fields. The width of the lines of the sextet was constrained to be equal to the line width for tin. The hyperfine field distribution curve obtained for the spectrum at 50 K is shown in Fig. 2. In the distribution, the predominant peak is centered at a field of 38 kOe. The average hyperfine field obtained is equal to 42 kOe. This value is consistent with the value of 40 kOe obtained by Malik et al.

The hyperfine magnetic fields at non-magnetic probes, for example, at Sn in the present study, or at substitutional sites in Heusler alloys are understood in terms of conduction electron polarization via Fermi contact term. Conduction electron polarization varies sinusoidally with distance from the transition element where magnetic moment is assumed to be localized. The value of the average number of conduction electron $n_0$ is given by:

$$n_0 = \frac{1}{4} [2(L_x - 2D_x + \mu_x) + (L_y - 2D_y + \mu_y) + N_z]$$  \hspace{1cm} (1)

Here $L_i$ is the number of outer shell electrons and $D_i$ the number of spin down outer electrons in $X_2YZ$ alloys. $N_z$ is the electrons contributed to the conduction band by the Z element. The Fermi vector $k_F$ is given by:

$$k_F = \frac{1}{a_0} \left[ \frac{4 \pi}{\alpha} n_0 \right]^{1/3}$$  \hspace{1cm} (2)

Here $a_0$ is the lattice parameter.

The contribution to the hyperfine field arising from polarization of the conduction band at a particular probe site due to a magnetic moment located at $r_i$ is given according to Blandin and Campbell (BC) model by:

$$p(r_i) \propto \left( \frac{1}{r_i^3} \right) \cos (2k_F r_i + 2\delta_0 + \eta)$$  \hspace{1cm} (3)

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**Fig. 1**—Sn-¹¹⁹ Mössbauer spectra of Co₂ScSn at 50 K

**Fig. 2**—Hyperfine field distribution of Co₂ScSn at 50 K
where, $2\delta_0$ accounts for the perturbations to the conduction-electron density from the effective charge of the impurity atom and $\eta$, the preasymptotic factor.

From the value of magnetic moment of an atom located at $r_i$ and $p(r_i)$, the hyperfine field can be calculated as:

$$H = \sum_i \mu_i p(r_i) \quad \ldots \quad (4)$$

The summation is performed over all the magnetic atoms. In practice, however, it is sufficient to sum over the first, second and third nearest neighbour magnetic atoms. The unit cell for $X_2YZ$ Heusler alloys is given in Fig. 3. It can be described in terms of four interpenetrating fcc sublattices A, B, C and D. A and C are equivalent sites. In a well ordered $L_{21}$ structure of this sort, $X$ atoms are located on the A and C sites, $Y$ atoms at the B and $Z$ atoms at the D sites, respectively. The nearest neighbour configuration in terms of lattice parameter from $Z$ atom is given in Table 1.

In $Co_2ScSn$, the number of first nearest neighbour to $Sn$ atom is eight Co atoms. In the next two positions with Co as neighbour atoms there are 24 Co atoms at each of these positions. The corresponding distances for the three near neighbour positions for Co are $r_{11} = 2.6617\text{Å}$, $r_{22} = 5.1317\text{Å}$ and $r_{33} = 6.6852\text{Å}$. Malik et al. and Chen et al. have established that the magnetic moment in this alloy is entirely carried by Co atoms with a moment of $0.55 \mu_B$ per Co atom. This implies that the Sc atoms do not contribute to the magnetic field and so Eq. 1 can be written as:

$$n_0 = (1/4) [2(L_{Co} - 2D_{Co} + \mu_{Co}) + N_y + N_z].$$

Here $\mu_{Co}$ is the magnetic moment of the compound per Co atom, $L_{Co}$ is the number of outer shell electrons in Co and $D_{Co}$ is the number of spin down outer electrons in Co. $D_{Co}$ and $L_{Co}$ were assumed to be 4.7 and 9, respectively. $N_y$ and $N_z$ are the number of conduction electrons contributed by the atoms at $Y$ and $Z$ sites, respectively. The Sn atom has four electrons in its outer most shell with $s^2$-$p^2$ configuration. It can, therefore, have a valence 4 and can contribute these electrons to the conduction band. The Sc atom with $s^2$-$d^1$ configuration in the outer most shell could contribute one, two or all the three electrons to the conduction band. Thus, the valence can be assumed to be 1, 2 or 3. A value $n_0 = 1.58$ is obtained if Sc is assumed to contribute two electrons to the conduction band and $n_0 = 1.83$ if it contributes 3 electrons.

The hyperfine field at $Sn$ atom in $Co_2ScSn$ was calculated using Eq. (4). As all the constants in the equation were not known, a normalization factor was calculated using the data of Saad on the magnetic field at Sn in $Co_2HfSn$. Using Eq. 4 and the constants used by Saad, the field in $Co_2HfSn$ was calculated.

**Table 1**—Nearest neighbour configuration in terms of lattice parameter from $Z$ atom

<table>
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<tr>
<th>No. of shell neighbour distance in lattice parameters</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
<th>7</th>
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<tr>
<td>X</td>
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<td>6X</td>
<td>12X</td>
<td>---</td>
<td>8X</td>
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<td>24X</td>
</tr>
<tr>
<td>(Co)</td>
<td>4Y</td>
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<td>---</td>
<td>12Y</td>
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<td>---</td>
<td>12Y</td>
<td>---</td>
</tr>
<tr>
<td>Y</td>
<td>8X</td>
<td>---</td>
<td>---</td>
<td>24X</td>
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<td>24X</td>
<td>---</td>
</tr>
<tr>
<td>(Sc)</td>
<td>---</td>
<td>---</td>
<td>12Y</td>
<td>---</td>
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<td>6Y</td>
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</tr>
<tr>
<td>Z</td>
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<td>6Z</td>
<td>---</td>
<td>8Z</td>
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</tbody>
</table>

Fig. 3—Heusler $L_{21}$ structure
and normalized to the value given by Saad\(^{20}\). This normalization was employed in calculating the hyperfine field in Co\(_2\)ScSn. In this way, the constants used to calculate the hyperfine field in Co\(_2\)ScSn were
\[
n_0 = 1.58, \quad N_y = 2 \quad \text{and} \quad N_z = 4.
\]
Using these values, the hyperfine field at Sn site was obtained to be 59 kOe, which is in fairly good agreement with that of the average field of 42 kOe obtained from the present experiment.

It can therefore be concluded that for Co\(_2\)ScSn, the observed hyperfine field experienced by Sn – 119 can be understood in terms of the Blandin-Campbell model. The magnitude of the hyperfine field can be approximately reproduced from the theory through a suitable choice of parameters and normalizing the Sn field to give agreement with the experimental value.

Acknowledgement

This work has been supported by the UGC-DRS and COSIST schemes of the Department of Physics, M.L. Sukhadia University, Udaipur.

References