Magnetic hyperfine fields in the Heusler alloys Co_2YZ (Y = Sc, Ti, Hf, V, Nb; Z = Al, Ga, Si, Ge, Sn)

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The magnetic hyperfine field acting on Ta at the non-magnetic transition element site has been investigated in the Heusler alloys Co_2(Sc, Hf, V)Sn, Co_2ScGa, Co_2(V, Nb)Al and Co_2Ti(Si, Ge) by TDPAC measurements utilizing the 133–482 keV gamma–gamma cascade in ^{181}Ta following the β^- decay of ^{181}Hf. An important conclusion is that the reduced mhf either on the non-magnetic transition element site or on the s–p element site depends mainly on the chemical nature of the non-magnetic transition element rather than any other factor, e.g. the local moment μ_{Co} of the alloy.

1. Introduction

The $L2_1$ Heusler alloys with stoichiometric composition X_2YZ are in general ferromagnetic when they are of the form Co_2YZ of X_2MnZ. Here X is usually a transition or noble metal such as Cu, Pd, Ni; Y is a transition element such as Ti, Zr, Hf, V, Nb, and Z is an sp element belonging to groups IIIA–VA. In the alloys Co_2YZ the local magnetic moment is carried by the Co atoms and values ranging from 0.3 to 1.0 μ_B are commonly found. In the alloys of the form X_2MnZ the magnetic moment of about 4 μ_B is localized on the Mn atoms. The study of these alloys has made a unique contribution towards the understanding of the hyperfine fields present at the impurity sites in ferromagnetic metals. The two groups of Heusler alloys are, however, distinctly different from each other. In the alloys of the form X_2MnZ the sp element at Z site is the second nearest neighbour of the magnetic atom Mn, where as in the alloys Co_2YZ, both an sp element at Z site as well as the transition element at Y site are the nearest neighbours of the magnetic atom Co. Another significant difference is due to the smaller distance between the magnetic atoms in the Co-based alloys. Heusler alloys Co_2YZ where only Co atoms have local moment and where the nearest neighbour Co–Co distance is only slightly larger than in pure Co have received more attention recently because under these cir-

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cumstances it is quite possible that direct exchange interactions play an important role in determining the magnetic properties. Long range magnetic coupling of localized moments via conduction electrons is, however, widely accepted as being the dominant exchange mechanism in Heusler alloys. In terms of magnetic properties Co_2YZ alloys are expected to behave similarly as the normal concentrated materials such as Fe, Co, Ni.

An interesting aspect of the hyperfine fields at non-magnetic atoms in the cobalt based alloys has been reported recently [1] in which it is shown that while the reduced magnetic hyperfine field (mhf) at Sn in the alloys Co_2YSn depends strongly on the chemical nature of the transition element present at Y site, the reduced mhf at Y site transition element is relatively insensitive to the chemical nature of the second neighbour sp element. In order to further investigate this behaviour as well study the Y site hyperfine field as a function of the chemical nature of the transition element, we have carried out time differential perturbed angular correlation (TDPAC) measurements of mhf acting on Ta at the Sc, Ti, Hf, V and Nb sites in the alloys: Co_2(Sc, Hf, V)Sn, Co_2ScGa, Co_2(V, Nb)Al and Co_2Ti(Si, Ge). The hyperfine field results are discussed and compared with observed systematics in Co-based Heusler alloys.

2. Experimental

The Heusler alloy samples were prepared by arc melting the constituent elements under argon atmosphere along with the radioactive ^{181}Hf substituting approximately 0.1% of the atoms of the Y site transition element in each case. The resulting alloys were homogenized at 900°C for 24 h and cooled slowly. The samples were then crushed and annealed at 800°C during 24–72 h in argon atmosphere and quenched in water.

The TDPAC measurements were carried out with a conventional fast–slow coincidence set-up using BaF_2 detectors having a time resolution of about 0.9 ns. The well known 133–482 keV gamma cascade in ^{181}Ta was used to measure the TDPAC spectra. The perturbation factor for an unpolarized ferromagnetic sample consisting of randomly oriented domains can be written (neglecting the A_{44} terms) as

$$A_{22}G_{22}(t) = A_{22}(0.2 + 0.4\cos\omega_L t + 0.4\cos 2\omega_L t)\,, \qquad (1)$$

where $\omega_L = \mu_N g H_{hf}/\hbar$ is the Larmor frequency. The g-factor of the 482 keV state of ^{181}Ta is known as $g_{5/2} = 1.3(1)$ [2]. It is then possible to determine the ^{181}Ta hyperfine field from the measured Larmor frequency. The $A_{22}G_{22}(t)$ measurements were performed at 77 K in all cases except for the alloy Co_2HfSn, where a relatively large field is expected these measurements were performed at 300 K.

The sign of the field was determined by placing the sample in an external polarizing magnetic field of about 5 kG applied perpendicular to the plane of the detectors

and measuring the ratio $R(t)$ at a fixed angle. The quantity $R(t)$ is expressed in terms of Larmor frequency as follows (neglecting the A_{44} term):

$$R(t, \theta = 3\pi/4) = \frac{N\uparrow - N\downarrow}{N\uparrow + N\downarrow} \approx -3/4 A_{22} \sin 2\omega_L t , \quad (2)$$

where $N\uparrow$ and $N\downarrow$ are the numbers of coincidences with the applied magnetic field direction up ($\uparrow$) and down ($\downarrow$) respectively. The measurements of $R(t)$ were made at $\sim$ 120 K using a cold finger except in the case of Co_2HfSn where the measurement was made at 300 K. The external magnetic field was supplied by a compact design oil cooled electromagnet.

3. Results and discussion

Results of the TDPAC measurements for some of the alloys are shown in fig. 1. Solid curves are the least squares fit of the experimental data to expressions (1) and (2) respectively. Results of the fitting indicated that most of the alloys have unique fields with small distribution. However, the alloys Co_2VAl and Co_2VSn showed two distinct fields with approximately 40% of the Ta nuclei probing somewhat lower fields. This could be attributed to Ta nuclei occupying other sites within the $L2_1$ structure since the X-ray diffraction analysis did not reveal the presence of any other phase. The magnetic hyperfine fields H_{Ta} for a number of Heusler alloys determined in the present study are given in table 1. Additional results like the lattice parameter (a_0), Curie temperature (T_c) and the local magnetic moment on cobalt atom (μ_{Co}) taken from published data are included in this table. For the alloy Co_2ScGa the μ_{Co} has not been measured experimentally and the result given in table 1 is based on an empirical correlation [3] between the local moment (μ_{Co}) and the corresponding difference in the electronegativity of Co and Y component in the Co_2YGa and Co_2YSn alloys.

Previous results [1] indicated that the Ta hyperfine field at Y site in the alloys Co_2YZ where Y is a group IVB element Ti, Zr or Hf and Z an sp element Al, Ga or Sn behave similarly as the field on non-magnetic impurities in magnetic materials such as Fe, Co and Ni. Additional results obtained in the present study reveal a similar behaviour also in the alloys where Y is a group IIIB element Sc or a group VB element V or Nb. The fields are all negative in cases where the signs were measured and they are roughly proportional to Curie temperature T_c and the local moment (μ_{Co}) of the alloy within each group.

It is now well recognized that the reduced mhf on Sn in the alloys Co_2YSn is strongly dependent on the outer electron configuration of the Y site atom. For example the H_{Sn}/μ_{Co} value increases from about +70 kOe/μ_B for Y = Sc [4] to $\sim$ +100 kOe/μ_B for Y = Ti, Zr, Hf [5] and then drops rather unexpectedly to $\sim$ +30 kOe/μ_B for Y = V, Nb [6]. This is in apparent contradiction with the generally observed trends where the field increases with the conduction electron den-

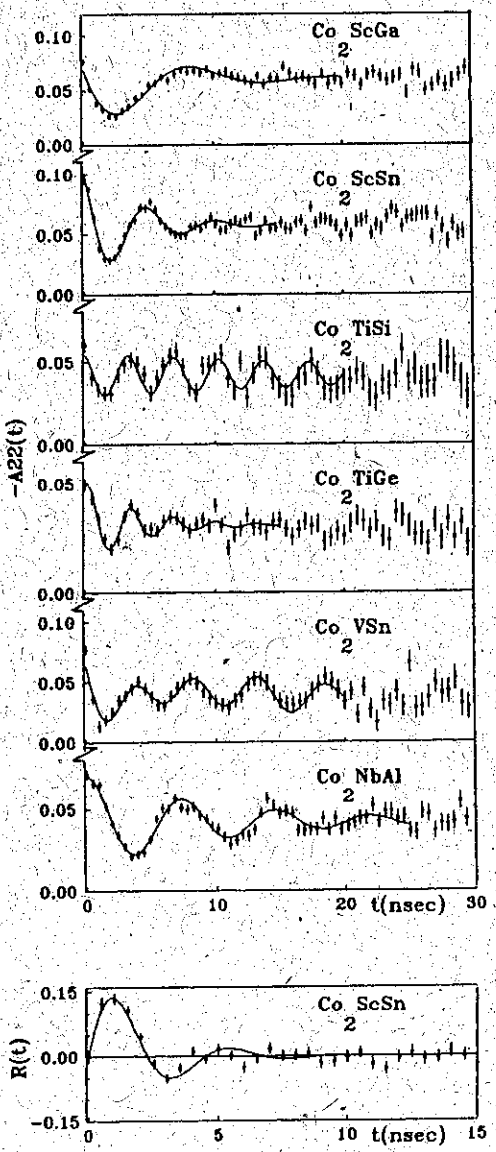

Fig. 1. TDPAC results for the alloys Co_2YZ (Y = Sc, Ti, Hf, V and Nb; Z = Al, Ga, Si, Ge and Sn).

sity. While there is no satisfactory explanation as yet for the sudden drop in the H_{Sn}/μ_{Co} value in the case of Y = V, Nb it is quite clear from the present as well as earlier results shown in fig. 2 that an almost similar behaviour is observed for the H_{Ta}/μ_{Co} at Y site atom in the Co_2YZ alloys. For example the H_{Ta}/μ_{Co} values are ~ -350 kOe/μ_B for Y = Sc, ~ -510 kOe/μ_B for Y = Ti, Zr, Hf and ~ -165 kOe/μ_B for V, Nb respectively.

Several remarks can be made from the data presented in fig. 2. (1) Whereas the reduced mhf on Ta depends strongly on the chemical nature of the Y element in the Co_2YZ alloy they are rather insensitive to the chemical nature of the sp element on Z site at least within the experimental errors which come mainly from the measured values of μ_{Co}. (2) The ratio $(H_{Ta}/\mu_{Co})/(H_{Sn}/\mu_{Co})$ is approximately 5 for

Table 1
Magnetic properties of the studied Heusler alloys.

Alloy	a_0 (Å)	T_c (K)	μ_{Co} (μ_B)	H_{Ta}(77 K) (kOe)	H_{Ta}/μ_{Co}) (kOe/μ_B)	H_{Ta}/T_c (kOe/K)
Co_2ScGa	6.17	–	0.25	(–)90(2)	(–)360	–
Co_2ScSn	6.17	268	0.55	–187(3)	–341	–0.70
Co_2TiSi	5.74	375(4)	0.55	–287(6)	–522	–0.76
Co_2TiGe	5.83	386(4)	0.89	–312(6)	–351	–0.81
Co_2HfSn	6.24	396	0.77	–421(12)	–547	–1.07
Co_2VAl	5.85	310(4)	0.92	–116(4)	–126	–0.37
Co_2VSn	5.98	105	0.60	(–)108(1)	(–)180	(–)0.98
Co_2NbAl	5.94	383	0.67	–138(4)	–206	–0.36

every pair of alloy containing the same transition element Y and this ratio is independent of the chemical group to which the element Y belongs. This suggests that the mechanisms producing the Ta and Sn fields are very similar despite a difference in the sign of the fields. The trend from negative to positive fields at the sp elements with increasing number of valence electrons found to exist in Fe, Co, Ni as well as in Heusler alloys has been explained by Blandin and Campbell [7] in terms of charge screening at the site of the sp element. (3) Both H_{Ta}/μ_{Co} and H_{Sn}/μ_{Co} values seem to follow the generally observed trend of increasing field with increasing conduction electron density when for example a group IIIB element Sc at Y site is substituted by a group IVB element Ti, Zr, or Hf with higher number of d-electrons. The behaviour of V and Nb is anomalous in this respect since the reduced fields decrease when a group IVB element is replaced by a group VB element with still

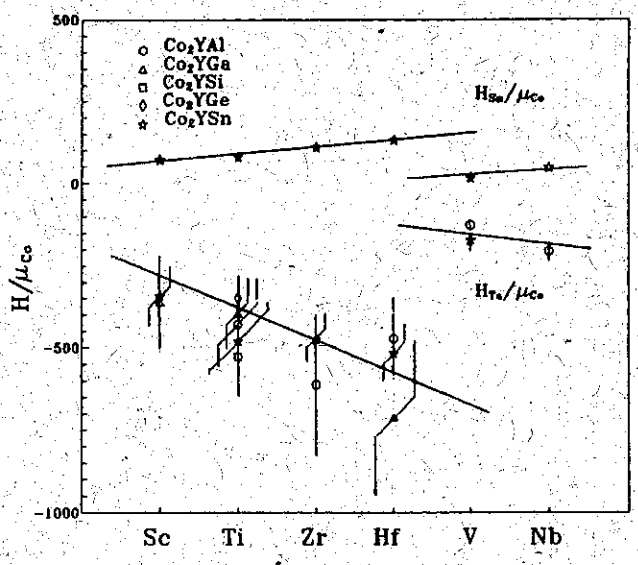

Fig. 2. Reduced mhf at Ta and Sn as a function of the Y site transition element in the alloys Co_2YZ.

higher number of d-electrons. (4) The reduced mhf at Ta as well as at Sn increases, although slowly, within each chemical group, when the outer electron configuration of the Y site element changes from 3d to 4d to 5d. This may be understood in terms of the atomic size effect which leads to higher conduction electron density for the larger atom in each group probably due to a better delocalization of the outer shell electrons.

The main conclusion of the present work is that the reduced mhf either on the non-magnetic transition element site or on the sp element Sn in the Co_2YZ Heusler alloy depends mainly of the chemical nature of the non-magnetic transition element rather than any other factor e.g. local moment μ_{Co} of the alloy, lattice parameter etc. The behaviour of fields in alloys containing V or Nb at Y atom site is anomalous and cannot be understood easily in terms of net conduction electron spins polarization at the probe nucleus.

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