MAGNETIC HYPERFINE FIELD IN THE HEUSLER ALLOYS $Co_2Y^1_{1-x}Y^2_xZ$ (Y = Ti,V,Nb,Cr; Z = Al, Sn)*

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Magnetic hyperfine field (mhf) acting on ^{181}Ta probe dilutely substituted at the non magnetic transition element site has been investigated in the quaternary Heusler alloys $Co_2Ti_{1-x}Nb_xAl$, $Co_2Ti_{1-x}V_xAl$, $Co_2Ti_{1-x}V_xSn$, $Co_2V_{1-x}Cr_xAl$ with $0 \leq x \leq 1$ by TDPAC measurements utilizing the 133-482 keV gamma-gamma cascade in ^{181}Ta following the β^- decay of ^{181}Hf. The results of mhf on Ta occupying the non-magnetic transition element site are discussed and compared with the mhf systematics in the cobalt based Heusler alloys.

1. Introduction

The Heusler alloys are ternary intermetallic compounds and provide an excellent environment where a systematic behaviour of the magnetic hyperfine field can be studied by varying the constituent elements of the alloy, as well as by measuring the hyperfine field at different atomic sites of the alloy. The $L2_1$ Heusler alloys have stoichiometric composition X_2YZ, where 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 group IIIA to VA. The Co-based Heusler alloys Co_2YZ are of particular interest because the local magnetic moments, carried by the Co atoms in these alloys, are known to have values ranging from 0.3 to 1.0 μ_B, while in the alloys of the form X_2MnZ, with X $\neq$ Co, the magnetic moment of about 4 μ_B is localized on the Mn atoms. This fact is most probably related to the number of localized or itinerant d-electrons in each type of alloy which contribute to the local magnetic moment. Furthermore, in the Heusler alloys Co_2YZ the nearest neighbour Co-Co distance is only slightly larger than in pure Co and it is quite possible that direct exchange interactions play an important role in determining their 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 previous work[1, 2] some interesting features of the hyperfine fields at non-magnetic atoms in the cobalt based alloys have been reported. The reduced mhf either on the non-magnetic transition element site or on the sp element Sn in the Co_2YZ Heusler alloy has been shown to depend mainly on the chemical nature of the non-magnetic transition element at the Y atom site. Another important observation is that the reduced mhf on Sn as well as on the non-magnetic transition element site follows 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, Nb, Ta and Cr is anomalous in this respect since the reduced fields decrease when a IVB group element is replaced by a VB group element or VIB group

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element with presumably still higher number of d-electrons.

In order to further investigate this behaviour as well as to study the Y site hyperfine field as a function of the chemical nature of the transition element, specially when that element is switched from IVB to VB or VIB group element, we have carried out the time differential perturbed angular correlation (TDPAC) measurements of mhf acting on Ta at the Y sites in quaternary Heusler alloys $Co_2Ti_{1-x}Nb_xAl$, $Co_2Ti_{1-x}V_xAl$, $Co_2Ti_{1-x}V_xSn$, $Co_2V_{1-x}Cr_xAl$ with $0 \leq x \leq 1$. The hyperfine field results are discussed and compared with observed systematics in Co-based Heusler alloys.

2. Experimental

The quaternary Heusler alloy samples were prepared by arc melting the constituent elements with the required stoichiometric composition, 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 ^oC for 24 hours and cooled slowly. The samples were then crushed and annealed at 800 ^oC during 24 to 72 hours in argon atmosphere and quenched in to water. All the samples were analyzed by x-ray diffraction to ensure that they had the correct $L2_1$ structure.

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 :

$$A_{22}G_{22}(t) = A_{22}[0.2 + 0.4cos\omega_L t + 0.4cos2\omega_L t] \quad (1)$$

where $\omega_L = \mu_N g H_{hf}/\hbar$ is the Larmor frequency. The g-factor of the 482 kev $(5/2)^+$ state of ^{181}Ta is known $g_{5/2} = 1.3(1)$ [3]. 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. The sign of the field was measured by the usual method using an externally applied magnetic field of the order of 0.5 T.

3. Results and discussion

TDPAC measurements were analysed as usual by least square fitting the experimental data to the expression 1. Results of the fitting indicated that most of the alloys had unique fields with small distribution. Some of the alloys for example $Co_2Ti_{0.2}Nb_{0.8}Al$, $Co_2Ti_{0.2}V_{0.8}Sn$, $Co_2Ti_{0.6}V_{0.4}Sn$ and $Co_2V_{0.2}Cr_{0.8}Al$ however showed two distinct fields with approximately 30% of the Ta nuclei probing a somewhat smaller field. Since the x-ray results did not reveal the presence of a second phase we attribute this to Ta nuclei occupying other sites with in the $L2_1$ structure. The magnetic hyperfine fields H_{Ta} for a number of Heusler alloys determined in the present study are given in Table 1.

The mhf on Ta at the Y site is plotted in figure 1 as a function of the concentration of the element Y^2 in the alloys $Co_2Y^1_{1-x}Y^2_xZ$.The results show that the Ta field in the alloys $Co_2Ti_{1-x}V_xAl$ and $Co_2Ti_{1-x}Nb_xAl$ gradually increases (becoming more negative) between $0 \leq x \leq 0.8$ and then drops suddenly at $x = 1.0$. Unfortunately the values of the local magnetic moment on Co atom in theses alloys are not available at present to permit further discussion on the possible trend of the reduced hyperfine field H_{Ta}/μ_{Co}. Saturation magnetization measurements for these alloys, using a Foner type vibrating sample magnetometer

are in progress in our laboratory. The behaviour of the Ta mhf in the alloys $Co_2V_{1-x}Cr_xAl$ is broadly similar to the $Co_2Ti_{1-x}V_x(Nb_x)Al$ alloys although less dramatic in showing a maximum around 80% Y^2 concentration.

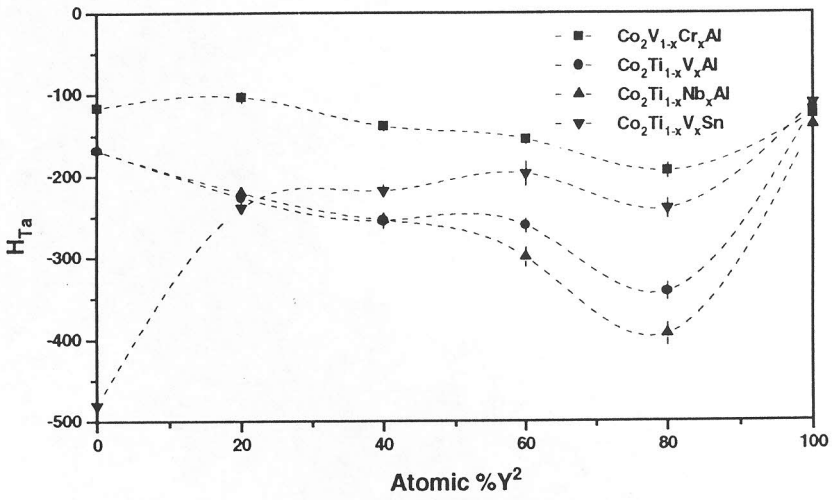

Figure 1: Mhf on Ta as a function of Y^2 concentration for the alloys $Co_2Y^1_{1-x}Y^2_x(Al,Sn)$.

For the case of the alloy $Co_2Ti_{1-x}V_xSn$ the present results indicate that the hyperfine field H_{Ta} drops suddenly in going from x = 0 to x = 0.2 and then decreases rather slowly for $0.2 \leq x \leq 1.0$. Dunlap and Stroink[4] have obtained the values of local magnetic moment on Co from their saturation magnetization data in these alloys. A peculiar aspect of the magnetization data is that the μ_{Co} values remain relatively constant for $0 \leq x \leq 0.8$ and then decreases dramatically for x = 1.0. We have used the μ_{Co} values taken from their work to calculate the reduced hyperfine field H_{Ta}/μ_{Co} in the series of alloys $Co_2Ti_{1-x}V_xSn$ studied in the present work and the results are shown in figure 2. The reduced hyperfine field decreases rather abruptly in going from x = 0 to x = 0.2 and then remains practically constant for $0.2 \leq x \leq 1.0$. This behaviour is quite different when compared with the results of the Sn fields, H_{Sn}/μ_{Co} obtained by Dunlap and Stroink from Mössbauer study where the authors found an almost linear decrease (field becoming less positive) in the reduced field at Sn for $0 \leq x \leq 0.8$ and then a slight increase at x = 1.0. The authors concluded that their results could be explained easily if it is assumed that increasing the conduction electron density in Co_2TiSn decrease the Sn hyperfine field. This was demonstrated by the authors by

Table 1: Magnetic hyperfine fields measured for the studied Heusler alloys.

Alloy	H_{Ta} (kOe) 77 K					
	x=0.0	x=0.2	x=0.4	x=0.6	x=0.8	x=1.0
$Co_2Ti_{1-x}Nb_xAl$	-168(8)	-220(4)	-253(6)	-299(12)	-393(14)	-138(4)
$Co_2Ti_{1-x}V_xAl$	-168(8)	-225(6)	-254(10)	-260(9)	-341(11)	-116(4)
$Co_2Ti_{1-x}V_xSn$	-480(10)	-238(5)	-217(8)	-196(15)	-239(12)	-110(2)
$Co_2V_{1-x}Cr_xAl$	-116(4)	-103(7)	-138(4)	-154(4)	-193(9)	-124(5)

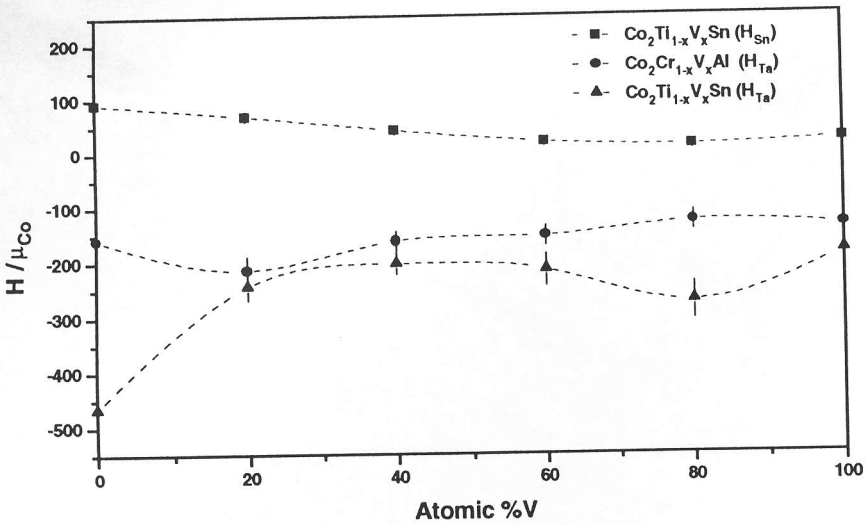

Figure 2: Reduced mhf at Ta and Sn as a function of the V concentration in the alloy $Co_2Ti_{1-x}V_xSn$, and reduced mhf at Ta as a function of V concentration in the alloy $Co_2Cr_{1-x}V_xAl$.

calculating the conduction electron density in this series of alloys for $0 \leq x \leq 1.0$, using the Blandin Campbell theory[5]. A similar conclusion however cannot be drawn from the results presented here for the H_{Ta}/μ_{Co} values in these alloys since it would require the conduction electron density to decrease suddenly in going from x = 0 to x = 0.2 and then to remain almost constant for $0.2 \leq x \leq 1.0$.

The results for the series of alloys $Co_2Cr_{1-x}V_xAl$ obtained here show (figures 1,2) that the Ta fields as well as the reduced fields H_{Ta}/μ_{Co} change only slightly over the entire range of Cr concentration ($0 \leq x \leq 1.0$). The value of local moments, μ_{Co} for these alloys were taken from the reference 6 for the purpose of calculating the H_{Ta}/μ_{Co} values. Once again the experimental data show quite a similar trend as obtained in the case of the alloys $Co_2Ti_{1-x}V_xSn$ except for x = 0 where the reduced field decreases suddenly for the Co_2TiSn alloy. It seems obvious therefore that a simple calculation of the conduction electron density in these alloys such as suggested by Dunlap and Stroink[4] is inadequate in describing the Ta hyperfine field results in these alloys. The sensitivity of the Co moment to the chemical nature of the non-magnetic atoms in the Co-based Heusler alloys is quite evident from the large variation in the μ_{Co} values (0.3 to 1.0 μ_B) in various alloys[7] and it may be necessary to make elaborate band calculations in order to explain the observed results in these alloys.

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