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MAGNETIC HYPERFINE FIELDS IN HEUSLER ALLOYS* Co_2YZ (Y = Ti,Zr; Z = Al,Ga,Sn)

S.D. DE SOUZA† and R.N. SAXENA

Instituto de Pesquisas Energéticas e Nucleares, IPEN-CNEN/SP, 01000 São Paulo, Brasil

W. SHREINER and F.C. ZAWISLAK

Instituto de Física UFRGS, 90049 Porto Alegre, Brasil

Magnetic hyperfine fields (mhf) acting on Ta at the Ti and Zr sites have been measured in Heusler alloys $Co_2TiAl(Ga,Sn)$ and $Co_2ZrAl(Sn)$ by the TDPAC technique utilizing the 133-482 keV gamma cascade in ^{181}Hf. Curie temperatures of all the alloys have also been measured using a vibrating sample magnetometer. Present data together with the existing results on the $Co_2HfAl(Ga,Sn)$ are discussed and compared with the mhf systematics in Heusler alloys.

INTRODUCTION

The $L2_1$ Heusler alloys are ternary intermetallic compounds formed at the stoichiometric composition X_2YZ. Typically the X atoms are transition or noble metals; Y atoms are transition elements such as Mn, Ti, Zr, Hf, V or Nb and Z atoms are sp elements of group III to V. Alloys X_2MnZ where X is Cu, Pd or Ni and Z an sp element are generally ferromagnetic with saturation magnetic moment on Mn atom of about $4\mu_B$. In the alloys of the type Co_2MnZ, besides a local moment on Mn ($\sim 3.5\mu_B$) Co atoms also carry a moment ranging from 0.5 to $0.75\mu_B$. In yet another type of alloys Co_2YZ where Y is Ti, Zr, Hf, V or Nb an Z an sp element only Co atoms have local magnetic moment varying from 0.3 to $1.0\mu_B$.

The magnetic properties of Heusler alloys X_2MnZ where only Mn atoms have moments are of particular interest to the understanding of magnetism. With relatively large Mn-Mn distances they constitute a class of magnetic materials halfway between truly dilute magnetic systems like CuMn and concentrated systems such as Fe, Co, Ni. These alloys have been studied extensively.

Somewhat lesser studied Co_2YZ where only Co atoms have local moments and where the nearest neighbour Co-Co distance is only slightly larger than in pure Co are also of interest because under these circumstances it is possible that a direct exchange interaction plays an important role. Long range magnetic coupling of localised moments via conduction electrons is however widely accepted as being the dominant exchange mechanism in Heusler alloys. Measurements of magnetic hyperfine fields (mhf) acting at the magnetic and especially at non magnetic atoms in these alloys should provide important information about the conduction electron spin polarization (CEP) induced by the magnetic atom on the surrounding sites. Such information is crucial to the understanding of the mechanisms which lead to the magnetic ordering in Heusler alloys. In this work we present the time differential perturbed angular correlation (TDPAC) measurement of the mhf acting on Ta at the Ti and Zr sites in $Co_2Ti(Al,Ga,Sn)$ and $Co_2Zr(Al,Sn)$ alloys. Curie temperatures of these alloys were also remeasured using a Foner vibrating sample magnetometer. The magnetic hyperfine field results are compared with the previous measurements on similar alloys $Co_2Hf(Al,Ga,Sn)$ /1,2/.

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† Permanent address: Departamento de Física - UFSCAR - São Carlos - SP.

EXPERIMENTAL

All the samples of Heusler alloys were prepared by mixing the constituent metal powders in stoichiometric composition (except that ~ 1% of the Ti(Zr) atoms were substituted by the radioactive ^{181}Hf atoms), compressing them and melting in an arc furnace under argon atmosphere. For alloys containing Ga and Sn the compressed metal powders were preheated at lower temperatures (100-300°C) for several hours in argon atmosphere before finally melting to prevent partial loss of more volalite metals. After one or two melting cycles the alloys were homogenized at 900°C for 24 hours and cooled slowly. The samples were then crushed and annealed during 24 hours at 800°C in argon atmosphere and quenched in to water. The purity of metals used was better than 99.99% for Al,Sn and Ga and better than 99.9% for Co and Zr.

Curie temperatures (T_c) of the studied Heusler alloy samples were remeasured using a vibrating sample magnetometer because there are discrepancies in the reported values of T_c in some cases. Our results for T_c are presented in Table 1. The TDPAC measurements were carried out using the well known 133-482 keV gamma cascade in ^{181}Ta populated in the β^- decay of ^{181}Hf. A series of measurements at temperatures above T_c showed only small quadrupole interaction in most of the alloys indicating that they are essentially cubic. The mhf measurements were carried out at 77K in the case of Co_2TiAl, Co_2TiGa and Co_2ZrAl and at 339K and 420K respectively in the case of Co_2TiSn and Co_2ZrSn, where larger fields were expected, rendering the observation of Larmor frequency from the TDPAC data easier. The TDPAC perturbation factor for an unpolarized ferromagnetic sample consisting of randomly orientated domains is given by/3/ (assuming $A_{44} \simeq 0$).

$$A_{22}G_{22}(t) = A_{22}\left[0.2 + 0.4\ \mathrm{Cos}\omega_L t + 0.4\ \mathrm{Cos}2\omega_L t\right] \qquad (1)$$

Where $\omega_L = \mu_N g H_{hf}/\hbar$ is the Larmor precession frequency. With the known g-factor/4/ of the 482 keV state of ^{181}Ta $g(5/2^-) = 1.30(1)$ it is then possible to determine the ^{181}Ta hyperfine field from the measured ω_L. The sign of the field is determined by applying an external polarizing magnetic field normal to the plane of the detectors alternatively up (↑) and down (↓) and measuring the ratio R(t) at a fixed angle.

$$R(t, \theta = 135^o) = \frac{N\uparrow - N\downarrow}{N\uparrow + N\downarrow} = -\ 0.75\ A_{22}\mathrm{Sin}2\omega_L t \qquad (2)$$

Where N↑ and N↓ are the number of coincidences with field up and field down. The measurements of $A_{22}G_{22}(t)$ and R(t) were carried out by using two NaI(Tl) detectors and a conventional fast-slow coincidence system. A more accurate magnitude of the mhf was obtained from the $A_{22}G_{22}(t)$ data with better counting statistics however the measurement with the external field (H_{ext} = 4 kG) gave the sign of the mhf.

RESULTS AND DISCUSSION

Results of the TDPAC measurements are presented in fig.1. Solid curves are the least square fit of the experimental data to the expression (1). Results of the fitting indicated that alloys Co_2TiAl, Co_2ZrAl and Co_2ZrSn have unique fields with approximately 8% of distribution. In the case of Co_2TiGa and Co_2TiSn however it was necessary to fit the data with two distinct fields with ~ 30% of the Ta nuclei experiencing somewhat lower fields. This would be explained if there was a small amount of different magnetically ordered phase present or if some Ta nuclei occupied other sites within the $L2_1$ structure. Since previous X-rays studies have shown no evidence for a second phase, the latter explanation seems to be more reasonable.

It can be observed from the results in Table 1 that the Ta fields are all negative where they have been measured. This is similar to that observed for non-magnetic transition element impurities in other magnetic invironments like Fe,Co, Ni. In order to further illustrate this point we have presented in Table 1 the ratios of Ta hyperfine field H_{Ta} to T_c and to the magnetic moment on μ_{Co}. With

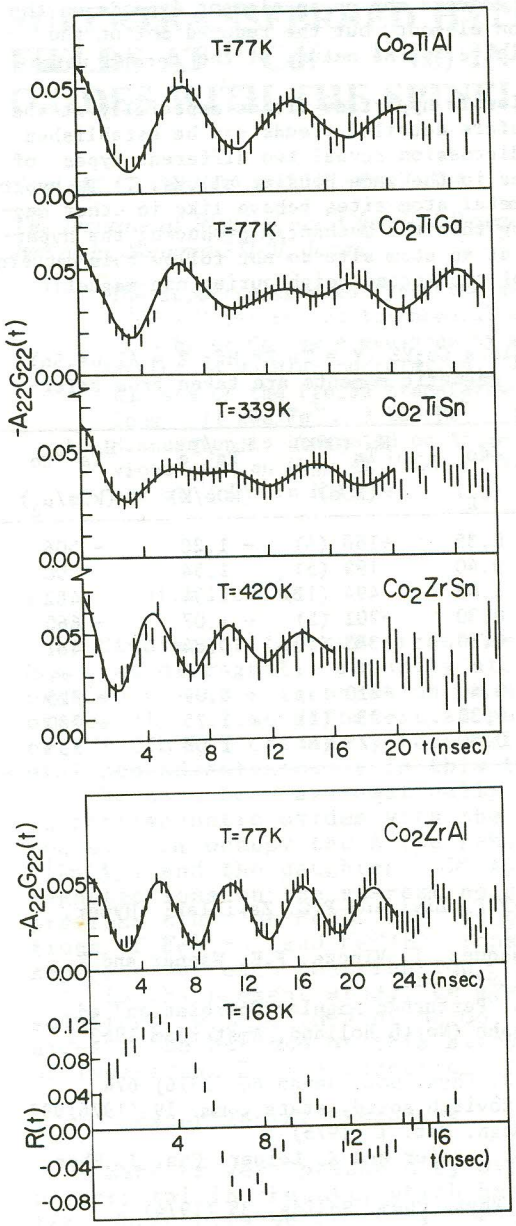

Fig. 1 - TDPAC results for ^{181}Ta at the Y site of Heusler alloys Co_2YZ(Y = Ti,Zr; Z = Al,Ga,Sn)

the exception of Co_2TiGa the ratio H_{Ta}/T_c is nearly constant(~ 1.1 kOe/K) within 10-15% for all the alloys.Similarly the average ratio H_{Ta}/μ_B is also approximately constant (~ 560 kOe/μ_B), although there are rather large experimental uncertainties in the individual values of this ratio mainly resulting from the errors in μ_{Co}. This proportionality between the local moment on Co, curie temperature T_c and mhf on Ta shows that mhf on Ta in these alloys follow similar trends as the field on non-magnetic dilute impurities in magnetic materials such as Fe,Co,Ni. It may therfore be rasonable to assume that the mechanism producing the fields on Ta in the alloys Co_2YZ (Y = Ti,Zr,Hf) may also be similar. In fact the Co-Co distance in these alloys is nearly the same as in pure Co and the impurity atom Ta is first near neighbour of the magnetic atom in both systems.

There is experimental evidence that the mhf on a given sp element at Z site in Co_2YZ alloys is strongly dependent on the nature of element Y, for example the Sn fields in the alloys Co_2YSn where Y is a group IVB element Ti,Zr or Hf are +82, +88 and +106 kOe respectively/5/. In the alloys where Y is a group VB element V or Nb the fields have been found to be approximately +10 and +15 kOe respectively/6/. From the data given in Table 1 one sees that a similar dependence on the nature of Z site sp element is observed in relation to the Y site mhf. For example in the Co_2YZ alloys when Z is a group IIIA element Al or Ga the Ta fields are approximately - 200 kOe, whereas when Z is a group IVA element Sn the fields are about - 400 kOe (assuming the fields to be negative for these alloys also). The discussion on these dramatic changes in the mhf is somewhat complicated by the different values of the magnetic moment on Co in these alloys. To eleminate the effect of differences in the value of the μ_{Co} one should compare the reduced mhf i.e H_{Sn}/μ_{Co} and H_{Ta}/μ_{Co}. The H_{Sn}/μ_{Co} values are ~ 100 kOe/μ_B in Co_2(Ti,Zr,Hf)Sn and ~ 40 kOe/μ_B in Co_2(V,Nb)Sn respectively and as seen before the H_{Ta}/μ_{Co} is approximately 560 kOe/μ_B and constant for all the alloys Co_2YZ(Y = Ti,

Zr,Hf; Z = Al,Ga,Sn). It seems that the reduced mhf on sp element depends on the nature of the second neighbour transition element, but the reduced mhf on the transition element is relatively insensitive to the nature of the second neighbour sp element.

Obviously more data are necessary for Ta hyperfine fields especially at the Y site of the Co_2VZ and Co_2NbZ alloys before detailed trends can be established. However the present data and the above discussion reveal two different types of behaviour for the non-magnetic impurities in the same Heusler alloys. 1) Ta hyperfine fields on non-magnetic transition metal atom sites behave like in other magnetic invironment e.g Fe,Co,Ni suggesting that the mechanism producing the hyperfine field is also the same. 2) The mhf at sp atom site do not follow this pattern and are rather sensitive to the nature of the second neighbouring non-magnetic transition metal atom.

Table 1 - Magnetic properties of the alloys Co_2YZ (Y = Ti,Zr,Hf; Z = Al,Ga,Sn) lattice parameters and Cobalt magnetic moments are taken from reference/9/.

	a_o (Å)	T_c (K)	μ_{Co} (μ_B)	$H_{Ta}(0^oK)$ (kOe)	H_{Ta}/T_c (kOe/K)	H_{Ta}/μ_{Co} (kOe/μ_B)
Co_2TiAl	5.85	148 (2)	0.35	-168 (4)	- 1.20	- 506
Co_2TiGa	5.85	130 (2)	0.40	199 (5)	1.54	500
Co_2TiSn	6.07	370 (2)	1.03	494 (12)	1.34	482
Co_2ZrAl	6.08	185 (2)	0.30	-201 (5)	- 1.07	- 660
Co_2ZrSn	6.25	460 (2)	0.80	385 (10)	0.84	481
Co_2HfAl [a]	6.02	193	0.40	-210 (5)	- 1.09	- 525
Co_2HfGa [a]	6.03	186	0.30	-234 (5)	- 1.25	- 780
Co_2HfSn [b]	6.22	423	0.80	428 (8)	1.08	535

a) from ref./1/ b) from ref./2/

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