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* MAGNETIC HYPERFINE FIELD ON Ta IN THE Co_2HfAl AND Co_2HfGa HEUSLER ALLOYS

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INTRODUCTION

The magnetic properties of the ternary intermetallic compounds with stoichiometric composition X_2YZ called Heusler alloys have been extensively investigated. The neutron diffraction and X-ray studies have shown that depending on the component elements,the same chemically ordered superstructure Heusler $L2_1$ can present three different magnetically ordered situations i) The Mn based alloys X_2MnZ where in general X is Cu, Pd, and Ni and Z an sp element and only Mn carries a magnetic moment of ~4μ_B. ii) Alloys of the type Co_2MnZ where Z is an sp element and besides the moment on Mn(~3.5μ_B), Co atoms also carry a local moment ranging from 0.3 to 1.0μ_B. iii) In the type of alloys Co_2YZ where Y is Ti, Zr, Hf, V, or Nb and Z an sp element only the Co atoms have local moment (0.3 to 1.0μ_B).

Because these alloys show simple crystallographic and magnetic structures and also because at stoichiometric composition all the atoms are in identical cubic environment, they are very attractive systems for the study of magnetism. The most investigated are the Mn based alloys. Somewhat lesser studied Co_2YZ alloys where the distance between the Co atoms is only 1.17 times larger than in pure Co, are of especial interest from the point of view of the various models proposed to explain the magnetic properties of Heusler alloys. In terms of magnetic properties the Co_2YZ alloys should behave similarly as the normal concentrated magnetic materials Fe,Co and Ni.

The measurement of magnetic hyperfine fields(mhf) acting at the magnetic and especially at the nonmagnetic atoms gives information about the conduction electron spin polarisation(CEP) induced by a magnetic atom on surrounding sites and is important for a better understanding of the mechanisms which lead to the magnetic order. In this contribution we present the time differential perturbed angular correlation(TDPAC) measurement of the mhf acting on Ta at the Hf site in Co_2HfAl (Ga) alloys. Our results are compared with the previous measurement[1] in Co_2HfSn and in other Co based alloys.

EXPERIMENTAL

The Co_2HfAl sample was prepared by melting in an evacuated quartz tube the mixed and compressed powders of the component metals together with ^{181}Hf activity produced by $^{180}Hf(n,\gamma)$ reaction. After melting the alloy was crushed, annealed during 24 hours at 800°C in argon atmosphere and quenched in to water. The non radioactive sample Co_2HfGa was supplied to us kindly by Dr. P.J. Webster(University of Salford England) and was prepared by using the same procedure. This sample was later neutron irradiated to procedure ^{181}Hf activity and was also annealed and quenched.

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The purity of metals used was better than 99.99% with the exception of Hf which was 99.9% (not considering Zr ~2.5 at%). After the first series of TDPAC measurements at room temperature both the samples were additionaly annealed at 300°C for 48 hours. The results showed that this low temperature annealing improved considerably the homogeneity of the alloys.

The mhf on Ta have been measured through the well known 133-482 KeV gamma-gamma cascade in ^{181}Ta. As the curie temperatures (Tc) of the alloys are of the order of 190°K, the measurements were performed at 77°K. The TDPAC perturbation factor for an unpolarised ferromagnetic material consisting of randomly oriented domains, can be written as (assuming $A_{44} = 0$)

$$A_{22}G_{22}(t) = A_{22}(\frac{1}{5} + \frac{2}{5}\cos\omega_L t + \frac{2}{5}\cos 2\omega_L t) \qquad (1)$$

where $\omega_L = \mu_N g H_{hf}/h$ is the Larmor precession frequency. When an external polarising field is applied perpendicular to the plane of the detectors alternatively up (+H) and down (-H), ω_L can be obtained from the ratio,

$$R(t,\theta = 135^o) = \frac{W(t,135^o, +H) - W(t,135^o, -H)}{W(t,135^o, +H) + W(t,135^o, -H)} = -b_2 \sin(2\omega_L t) \qquad (2)$$

The measurements of $A_{22}G_{22}(t)$ were carried out by using a three detector system and the conventional electronic set up. The ratios $R(t,\theta = 135^o)$ were measured using a fast lead loaded plastic scintillator for the detection of 133-KeV gamma ray, this improved the time resolution from 2.2 nsec (for NaI(Tl) detectors) to 1.4nsec. In the case of Co_2HfGa sample this ratio was measured at 165°K rather than at 77°K in order to obtain a lower Larmor frequency thereby making the measurements somewhat easier. Due to rather large Larmor precession frequencies in both alloys ($\omega_L \simeq 1200$ MHz) a more accurate magnitude of the mhf is obtained from the $A_{22}G_{22}(t)$, but the measurement with an external field yields the sign of the mhf. A detailed description of the TDPAC technique is presented in ref. 2.

RESULTS AND DISCUSSION

The results of the measurements without and with the external field $H_{ext} = 5$ kOe are presented in figs. 1 and 2. The fittings of the curves have been performed using the expressions (1) and (2) and by further assuming a gaussian distribution (~12%) of the magnetic hyperfine fields. The extracted hyperfine fields at 77°K are in table 1. Also shown in this table are the lattice parameters and some magnetic properties of the three alloys $Co_2HfAl(Ga,Sn)$[3,4]. It can be observed that the amplitude of the oscillations in both $A_{22}G_{22}(t)$ and $R(\theta=135^o,t)$ curves are attenuated as a function of time. This can have two different origins: a distribution of magnetic frequencies or the effect of combined magnetic dipole and electric quadrupole interactions. In fact the TDPAC measurement of the samples at 300°K show the presence of a distributed low frequency quadrupole interaction, which could be due to a small disorder (~2%) and/or due to the defects(there is nearly 2.5 at.% of Zr in Hf) in the cubic alloy. The statistics of the data however are not sufficient to distinguish between the two effects. As mentioned earlier the experimental data were fitted assuming a distribution of the magnetic frequencies. The presence of small quadrupole interactions do not affect the extracted values of mhf.

Several comments can be made concerning the results of the present experiment combined with the previous results in Co based Heusler alloys[5,6,7] (see also ref. 8 for a summary of the experimental data).

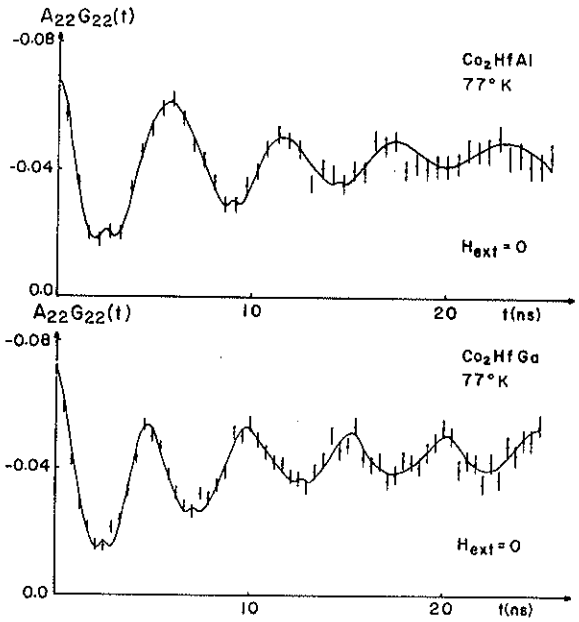

Fig. 1- TDPAC coefficients $A_{22}G_{22}(t)$ for Co_2HfAl and Co_2HfGa.

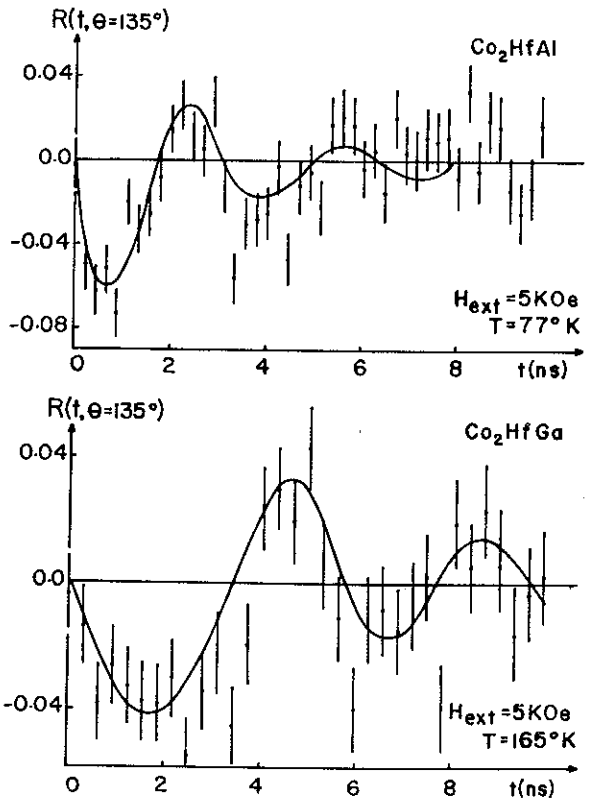

Fig. 2-The ratio $R(t,\theta=135^{o})$ for Co_2HfAl and Co_2HfGa.

1) The measurement of mhf on impurities in Heusler alloys are some times questionable because the exact site of the impurity is not known. In the present investigation the impurity atom ^{181}Ta on which the mhf is measured is at a regular Hf site, since the β^- decay (^{181}Hf → ^{181}Ta) provides very little recoil energy to displace the parent atom from its normal site.

2) The measured fields on Ta are negative in both the alloys and in what follows we will assume that the field on Ta in Co_2HfSn(sign not known) is also negative.

3) Assuming that the fields follow the magnetisation versus temperature curve from 77°K down to 0°K, the extrapolated mhf on Ta at 0°K are -210, -234 and -428 kOe for Co_2HfAl(Ga,Sn) respectively. The last two columns in table 1 show that the ratio between mhf at 0°K and Tc is within 10-15% same for the three alloys (~1.1kOe/K). Further the ratio between mhf and the magnetic moment on Co is also constant (~600kOe/μ_B) within the experimental errors(large errors come from μCo). This proportionality between the local magnetic moment on Co, the curie temperature and the mhf on Ta for the three alloys means that the fields on Ta in these alloys follow the same behaviour as the fields on nonmagnetic ditute impurities in concentrated magnetic materials(Fe,Co,Ni). It may be therefore reasonable to assume that the mechanism producing the mhf on Ta in Co_2HfZ may also be similar. In fact the distance between the Co atoms in Co_2HfZ is nearly the same as in pure magnetic materials. The impurity atom Ta is the 1nn of the magnetic atom in both systems.

4) Previous measurements have shown that the mhf on the same sp element at Z site in Co_2YZ alloy is strongly dependent on the nature of the element Y: fields on Sn in Co_2V(Nb,Hf)Sn are respectively + 9, + 15, and + 106 kOe. The present results reveal that this effect is reciprocal such that the sp element on Z site (Al,Ga,Sn) also affects the mhf on the same transition element (Ta) on Y site. In addition it is observed that the large changes in mhf and Tc take place when Y or Z belong to different groups in the periodic table: in Co_2YSn for Y = V,Nb(VB element) the fields are small ~10kOe and Tc is ~100°K, for Y = Ti, Zr,Hf(IVB element) the fields are much larger ~100kOe and Tc ~400°K. Similar behaviour is observed from our results, when the sp element at Z site changes from Al,Ga(IIIA) to Sn(IVA) the mhf on Ta and Tc of the alloy increases by neary a factor of two.

Table 1- Structure and Magnetic Properties of the Co_2HfAl(Ga,Sn) Alloys

	a_o (Å)	T_c (K)	μ_{Co} (μ_B)	H(Ta)(77°K) (kOe)	$\frac{H(Ta)(0^oK)}{Tc}$ (kOe/K)	$\frac{H(Ta)(0^oK)}{\mu Co}$ (kOe/μ_B)
Co_2HfAl	6.019	193	0.4	- 189 (4)	- 1.09	- 525 (130)
Co_2HfGa	6.032	186	0.3	- 213 (4)	- 1.25	- 780 (250)
Co_2HfSn	6.218	394	0.8	(-)347 (8)[b]	- 1.09	- 535 (60)

a) from ref3 , b) from ref1

5) It is also interesting to compare the reduced mhf, H/μCo (thereby eliminating the effect of the magnetic moment on Co) at the Y and Z sites. As shown in table 1. H(Ta)/μCo is nearly constant ~ - 600kOe/μ_B for the three alloys $Co_2HfAl(Ga,Sn)$, on the other hand H(Sn)/μCo is ~ + 100kOe/μ_B for $Co_2Ti(Zr,Hf)Sn$ whereas it is ~ + 40kOe/μ_B for $Co_2V(Nb)Sn$. It seems that the reduced mhf on sp element depends on the nature of 2nn nonmagnetic transition element, but the reduced mhf on a transition element atom is independent of the nature of 2nn nonmagnetic sp element. Obviously more data are necessary before this behaviour can be established.

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