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AND  $\text{Co}_2\text{HfGa}$  HEUSLER ALLOYS**

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# MAGNETIC HYPERFINE FIELD ON Ta IN THE Co<sub>2</sub>HfAl AND Co<sub>2</sub>HfGa HEUSLER ALLOYS\*

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## ABSTRACT

The measurements of the magnetic hyperfine fields (mhf) in Co<sub>2</sub>HfAl and Co<sub>2</sub>HfGa Heusler alloys have been carried out by the TDPAC technique utilizing the 133-482 KeV  $\gamma$ - $\gamma$  cascade in <sup>181</sup>Ta obtained from the  $\beta^-$  decay of <sup>181</sup>Hf. The experiments were performed at 77°K with and without an external polarizing magnetic field. The results are  $-189 \pm 4$  kOe and  $-213 \pm 4$  kOe for the fields on Ta occupying the Hf sites in Co<sub>2</sub>HfAl and Co<sub>2</sub>HfGa respectively, with a distribution of mhf of the order of 12%. The present data together with the previous existing results on the three alloys Co<sub>2</sub>HfAl(Ga,Sn) are discussed and compared with the mhf systematics in Heusler alloys.

## 1 – INTRODUCTION

The magnetic properties of the ternary intermetallic compounds with stoichiometric composition X<sub>2</sub>YZ 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<sub>1</sub> can present three different magnetically ordered situations. i) The Mn based alloys X<sub>2</sub>MnZ where in general X is Cu, Pd, and Ni and Z an sp element and only Mn carries a magnetic moment of  $\sim 4 \mu_B$  ii) Alloys of the type Co<sub>2</sub>MnZ where Z is an sp element, and besides the moment on Mn ( $\sim 3.5 \mu_B$ ), Co atoms also carry a local moment ranging from 0.3 to  $1.0 \mu_B$  iii) In the type of alloys Co<sub>2</sub>YZ where Y is Ti, Zr, Hf, V or Nb and Z an sp element and only the Co atoms have local moment (0.3 to  $1.0 \mu_B$ ).

Because these alloys show simple crystallographic and magnetic structures and also because at stoichiometric compositions 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<sub>2</sub>YZ alloys where the distance between the Co atoms is only 1.17 times larger than in pure Co, are of special 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<sub>2</sub>YZ alloys should behave similarly as the normal concentrated magnetic materials Fe, Co and Ni.

The measurements of magnetic hyperfine fields (mhf) acting at the magnetic and specially at the nonmagnetic atoms give 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<sub>2</sub>HfAl(Ga) alloys. Our results are compared with the previous measurement<sup>(1)</sup> in Co<sub>2</sub>HfSn and in other Co based alloys.

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## 2 – EXPERIMENTAL

The  $\text{Co}_2\text{HfAl}$  sample was prepared by melting in an evacuated quartz tube the mixed and compressed powders of the component metals together with  $^{181}\text{Hf}$  activity produced by  $^{180}\text{Hf}(n,\gamma)$  reaction. After melting the alloy was crushed, annealed during 24 hours at  $800^\circ\text{C}$  in argon atmosphere and quenched in to water. The non radioactive sample  $\text{Co}_2\text{HfGa}$  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 produce  $^{181}\text{Hf}$  activity and was also annealed and quenched.

The purity of the metals used was better than 99.99% with the exception of Hf which was 99.9% (not considering Zr  $\sim 2.5$  at%). After the first series of TDPAC measurements at room temperature both the samples were additionally annealed at  $300^\circ\text{C}$  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  $^{181}\text{Ta}$ . As the curie temperature ( $T_c$ ) of the alloys are of the order of  $190^\circ\text{K}$ , the measurements were performed at  $77^\circ\text{K}$ . The TDPAC perturbation factor for an unpolarised ferromagnetic material consisting of randomly oriented domains, can be written as (assuming  $A_{4,4} = 0$ ).

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

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),  $\omega_L$  can be obtained from the ratio,

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

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  $R(t, \theta = 135^\circ)$  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 1.4 nsec. In the case of  $\text{Co}_2\text{HfGa}$  sample this ratio was measured at  $165^\circ\text{K}$  rather than at  $77^\circ\text{K}$  in order to obtain a lower Larmor frequency there by making the measurements somewhat easier. Due to rather large Larmor precession frequencies in both alloys ( $\omega_L \approx 1200$  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. 6.

## 3 – RESULTS AND DISCUSSION

The results of the measurements without and with the external field  $H_{\text{ext}} = 5$  kOe are presented in Figures 1 and 2. The fittings of the curves have been performed using the expressions (1) and (2) and by further assuming a gaussian distribution ( $\sim 12\%$ ) of the magnetic hyperfine fields. The extracted hyperfine fields at  $77^\circ\text{K}$  are in Table I. Also shown in this table are the lattice parameters and some magnetic properties of the three alloys  $\text{Co}_2\text{HfAl}(\text{Ga}, \text{Sn})$ <sup>(8,7)</sup>. It can be observed that the amplitude of the oscillations in both  $A_{22}G_{22}(t)$  and  $R(\theta = 135^\circ, t)$  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  $300^\circ\text{K}$  shows the presence of a distributed low frequency quadrupole interaction, which could be due to a small disorder ( $\sim 2\%$ ) 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 is 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 does not effect 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<sup>(3,4,5)</sup> (see also ref. 2 for a summary of the experimental data).

**Table I**  
Structure and Magnetic Properties of the  $\text{Co}_2\text{HfAl}$  (Ga, Sn) Alloys

|                          | $a_0^a$ | $T_c^a$ | $\mu_{\text{Co}}^a$ | $H(T_a)$ (77°K)          | $H(T_a)$ (0°K) | $H(T_a)$ (0°K)     |
|--------------------------|---------|---------|---------------------|--------------------------|----------------|--------------------|
|                          | (Å)     | (K)     | ( $\mu\beta$ )      | (kOe)                    | $T_c$          | $\mu_{\text{Co}}$  |
|                          |         |         |                     |                          | (kOe/K)        | (kOe/ $\mu\beta$ ) |
| $\text{Co}_2\text{HfAl}$ | 6.019   | 193     | 0.4                 | -189 (4)                 | -1.09          | -525 (130)         |
| $\text{Co}_2\text{HfGa}$ | 6.032   | 186     | 0.3                 | -213 (4)                 | -1.25          | -780 (250)         |
| $\text{Co}_2\text{HfSn}$ | 6.218   | 394     | 0.8                 | (-) 347 (8) <sup>b</sup> | -1.09          | -535 ( 60)         |

a) from ref. 8; b) from ref. 1

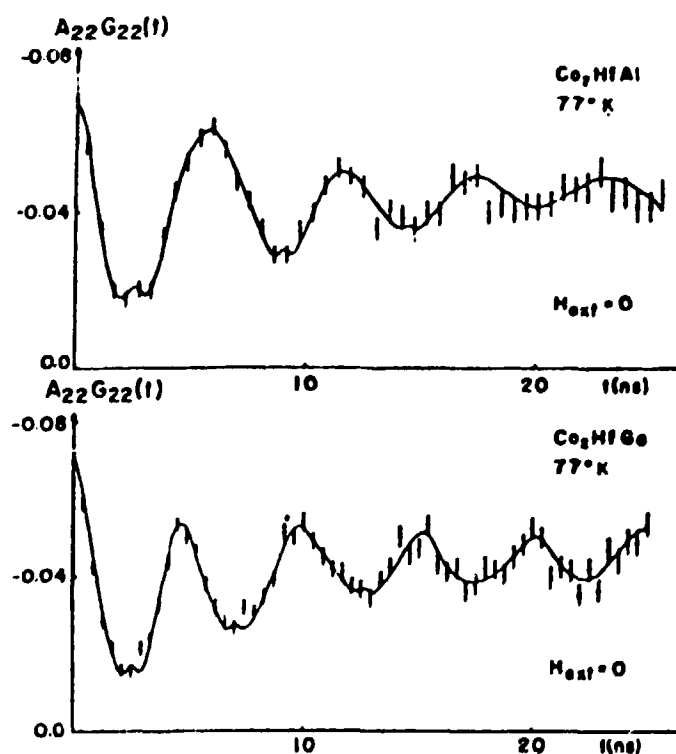


Figure 1 - TDPAC coefficients  $A_{22}G_{22}(t)$  for  $\text{Co}_2\text{HfAl}$  and  $\text{Co}_2\text{HfGa}$ .

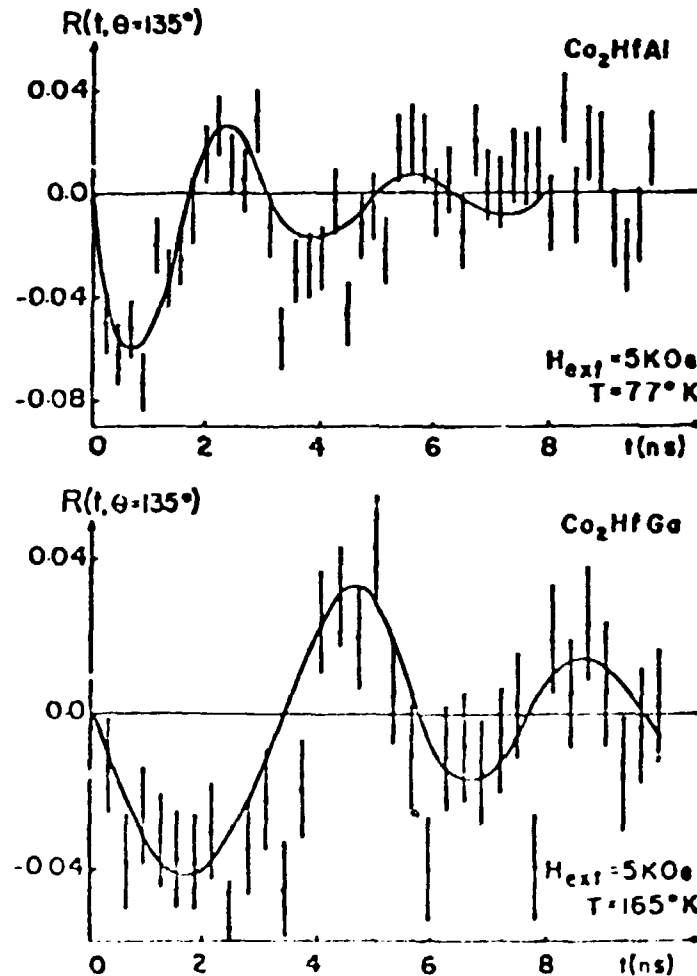


Figure 2 – The ratio  $R(t, \theta = 135^\circ)$  for  $\text{Co}_2\text{HfAl}$  and  $\text{Co}_2\text{HfGa}$ .

- 1) The measurement mhf on impurities in Heusler alloys are sometimes questionable because the exact site of the impurity is not known. In the present investigation the impurity atom  $^{181}\text{Ta}$  on which the mhf is measured is at a regular Hf site, since the  $\beta^-$  decay ( $^{181}\text{Hf} \rightarrow ^{181}\text{Ta}$ ) 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  $\text{Co}_2\text{HfSn}$  (sign not known) is also negative.
- 3) Assuming that the fields follow the magnetisation versus temperature curve from  $77^\circ\text{K}$  down to  $0^\circ\text{K}$ , the extrapolated mhf on Ta at  $0^\circ\text{K}$  are  $-210$ ,  $-234$  and  $-428\text{kOe}$  for  $\text{Co}_2\text{HfAl}(\text{Ga}, \text{Sn})$  respectively. The last two columns in Table I show that the ratio between mhf at  $0^\circ\text{K}$  and  $T_c$  is within 10-15% for the three alloys ( $\sim 1.1\text{kOe/K}$ ); further, the ratio between mhf and the magnetic moment on Co is also constant ( $\sim 600\text{kOe}/\mu_B$ ) within the experimental errors (large errors come from  $\mu_{\text{Co}}$ ). 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 dilute impurities in concentrated magnetic materials (Fe, Co, Ni). It may be therefore reasonable to assume that the mechanism producing the mhf on Ta in  $\text{Co}_2\text{HfZ}$  may also be similar. In fact the distance between the Co atoms in  $\text{Co}_2\text{HfZ}$  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<sub>2</sub>YZ alloy is strongly dependent on the nature of the element Y: fields on Sn in Co<sub>2</sub>V(Nb,Hf)Sn are respectively + 9 + 15, and + 106 kOe. The present result reveal that this effect is reciprocal such 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 T<sub>c</sub> take place when Y or Z belong to different groups in the periodic table: in Co<sub>2</sub>YSn for Y = V,Nb(VB element) the fields are small ~ 10kOe and T<sub>c</sub> is ~ 100°K, for Y = Ti, Zr, Hf(IVB element) the fields are much larger ~ 100kOe and T<sub>c</sub> ~ 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 T<sub>c</sub> of the alloy increases by approximately a factor of two.
- 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 I. H(Ta)/μCo is nearly constant ~ -600kOe/μβ for the three alloys Co<sub>2</sub>HfAl(Ga,Sn). On the other hand H(Sn)/μCo is ~+ 100kOe/μβ for Co<sub>2</sub>Ti(Zr,Hf)Sn where as it takes the value ~ + 40kOe/μβ for Co<sub>2</sub>V(Nb)Sn. It seems that the reduced mhf on the 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.

## PESUMO

Foram realizadas medidas dos campos magnéticos hiperfinos nas ligas de Heusler Co<sub>2</sub>HfAl e Co<sub>2</sub>HfGa por meio de técnica TDPAC utilizando a cascata gama-gama 133-482 keV no <sup>181</sup>Ta, obtida pelo decaimento β<sup>-</sup> do <sup>181</sup>Hg. As medidas foram realizadas a 77K com e sem um campo magnético externo de polarização. Os resultados encontrados foram 189 ± 4 kOe para os campos sobre o Ta ocupando as posições do Hf no Co<sub>2</sub>HfGa respectivamente, com uma distribuição de campo magnético hiperfino da ordem de 12%. Os dados obtidos no presente trabalho juntamente com os resultados existentes para três ligas Co<sub>2</sub>HfAl(Ga,Sn) são discutidos e comparados com a sistemática dos campos magnéticos hiperfinos em ligas de Heusler.

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