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Magnetic hyperfine field in the Heusler alloys Co_2YZ (Y = V, Nb, Ta, Cr; Z = Al, Ga)

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Abstract

The magnetic hyperfine field (mhf) acting on a ^{181}Ta probe dilutely substituted at the non-magnetic transition element site has been investigated in the Heusler alloys Co_2VGa, Co_2NbGa, Co_2TaAl and Co_2CrAl by TDPAC measurements utilizing the 133–482 keV gamma–gamma cascade in ^{181}Ta following the β^- decay of ^{181}Hf. The results are discussed in terms of the mhf systematics in the cobalt-based Heusler alloys, and are compared with theoretical predictions based on the Blandin and Campbell model.

Keywords: Heusler alloys; Magnetic hyperfine field; Perturbed angular correlation; BaF_2 detectors; Radioactive probe

1. Introduction

The Heusler alloys with stoichiometric composition X_2YZ have $L2_1$ cubic structure and generally exhibit magnetic behavior [1]. The X element is usually a transition or noble metal such as Cu, Pd, Ni; Y is a transition element such as Ti, Zr, Hf, V, Nb, etc.; and Z is an sp element belonging to groups IIIA–VA. The large variety of constituent elements that can be used in these alloys, together with the fact that they occupy well defined positions in the cubic structure, make Heusler alloys an excellent environment where the hyperfine magnetic field acting on different atomic sites can be studied. In particular, the influence of conduction electrons (s- or d-like electrons) on the hyperfine field may be studied by varying independently the constituent elements X, Y or Z.

The cobalt-based Heusler alloys Co_2YZ are of particular interest because of the fact that the magnetic moment which is localized on Co atom shows large variation and is known to have values ranging from 0.3 to 1.0 μ_B, in contrast with the alloys of the form X_2MnZ, where a magnetic moment of about 4 μ_B is localized on Mn atoms. This fact is most probably related to the numbers of localized or itinerant d-electrons in each type of alloy which effectively contribute to the formation of local magnetic moment.

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There are additional differences between these two types of alloys. For example, in the alloys X_2MnZ the sp element at Z site is the second nearest neighbor (2nn) of the magnetic atom Mn, whereas in the alloys Co_2YZ, both an sp element at Z site as well as the transition element at Y site are the nearest neighbors (1nn) of the magnetic atom Co. A further difference is due to the smaller distance between the magnetic atoms in Co_2YZ alloys as compared to the X_2MnZ alloys. The nearest-neighbor Co–Co distance is only slightly larger than in pure cobalt metal.

A good deal of experimental data is available for the magnetic hyperfine field in Heusler alloys of the form X_2MnZ [2,3], measured with different probes in the same host alloy, as well as with the same probe in different alloys. However, the majority of these measurements were made for the Z-site atom occupied by only few elements from groups IIIA and IVA. For the Co_2YZ type of alloys, until a few years ago, the only studies performed were the measurements at the Z-site atom with Mössbauer spectroscopy using ^{119}Sn probes [4–7]. However, since the mhf do not change appreciably on changing the sp elements in these alloys, it would be more interesting to investigate the behavior of the hyperfine field as a function of the non-magnetic transition element at Y site by measuring the interaction at the Y site itself. This has been done recently by using the ^{181}Ta probe in the perturbed angular correlation measurements [8–11]. The measurements were carried out on a large number of Heusler alloys, with Y element varying from group IIIB to VB. This forms a unique set of experimental data for the hyperfine fields at Y site atom in Co_2YZ alloys measured with the same probe. The results of previous investigations have already shown some interesting features of Y site fields in these alloys [10].

In order to investigate further the systematic behavior of the Y site field in the cobalt-based alloys as a function of the chemical nature of the non-magnetic transition element, we have carried out time differential perturbed angular correlation (TDPAC) measurements of the mhf acting on ^{181}Ta at the V, Nb, Ta and Cr sites in the alloys Co_2(V, Nb)Ga and Co_2(Ta, Cr)Al. The results are discussed and compared with the known systematics in similar alloys. The behavior of the Y site hyperfine fields in the cobalt-based Heusler alloys is also discussed in terms of theoretical predictions.

2. Experimental

The samples were prepared by arc melting the constituent elements under an argon atmosphere with 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 annealed at 800°C for 24–72 h in an argon atmosphere and quenched into water. All the samples were analyzed by X-ray diffraction, which verified that they had the $L2_1$ structure.

The TDPAC measurements were performed with a conventional fast–slow coincidence setup using BaF_2 detectors. The detector system had a time resolution of the order of 0.9 ns. The well known 133–482 keV gamma cascade in ^{181}Ta populated by the β^- decay of ^{181}Hf was used to measure the TDPAC spectra. The perturbation factor for an unpolarized ferromagnetic sample can be written (neglecting the A_{44} term) as

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

where $\omega_L = \mu_N \cdot g \cdot H_{hf}/\hbar$ is the Larmor frequency. The g-factor of the 482 keV $(5/2^+)$ state of ^{181}Ta is known to be $g_{5/2} = 1.3(1)$ [12]. It is therefore possible to determine the ^{181}Ta hyperfine field (H_{hf}) from the measured Larmor frequency. The $A_{22}G_{22}(t)$ measurements were performed at 77 K. The sign of the field was determined in each case by the usual method of applying an external magnetic field of the order of 0.5 T perpendicular to the plane of detectors and measuring the ratio $R(t)$ at a fixed angle. The ratio $R(t)$ may be expressed in terms of Larmor frequency (neglecting the A_{44} term) as

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

where $N_\uparrow$ and $N_\downarrow$ are the number of $\gamma\gamma$ coincidences with the applied magnetic field direction up ($\uparrow$) and down ($\downarrow$), respectively.

3. Results and discussion

A few examples of the TDPAC spectra measured for some of the alloys studied are shown in Fig. 1. The solid curves represent the least-squares fits of the experimental data to the expressions (1) and (2), respectively. The hyperfine fields H_{Ta} for the alloys determined in the present study are given in Table 1. Additional information such as the lattice parameter (a_0), Curie temperature (T_C) and the local magnetic moment on cobalt atom (μ_{Co}) taken from published data [13,14] has been included in this table.

Table 2 presents the existing experimental data on relevant magnetic and structural properties of Co_2YZ Heusler alloys taken from literature. Previous results on a series of Heusler alloys of the form Co_2YZ have shown that while the reduced field at Sn (H_{Sn}/μ_{Co}) in the alloys Co_2YSn depends mainly on the chemical nature of the transition element present at the Y site [4–7], the reduced field on Ta (H_{Ta}/μ_{Co}) at the Y site is relatively insensitive to the chemical nature of the 2nn sp element at the Z site [8–11]. The results indicated that the reduced fields both on Sn in Co_2YSn alloys and on Ta at the Y site element in the Co_2YZ alloys depend strongly on the outer electron configuration of the Y atom. This can be seen from Fig. 2, where the reduced fields H_{Sn}/μ_{Co} and H_{Ta}/μ_{Co} are plotted as a function of the outer electron configuration of the Y atom.

As shown in Fig. 2, the reduced fields H_{Sn}/μ_{Co} and H_{Ta}/μ_{Co} follow the generally observed trend of

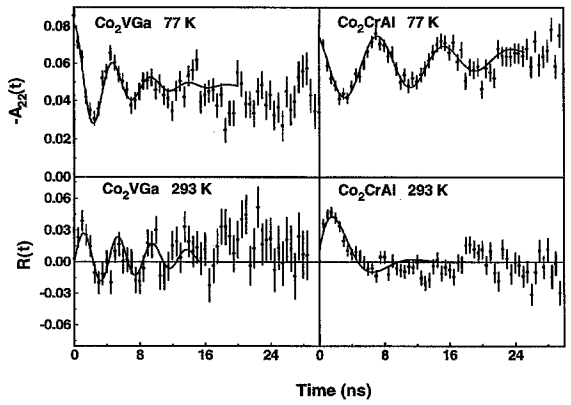

Fig. 1. TDPAC results obtained from the decay of ^{181}Hf at the V and Cr site of the Heusler alloys Co_2VGa and Co_2CrAl.

Table 1
Magnetic and structural 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)
Co_2VGa	5.78	352	1.05	−218(6)	−208(20)
Co_2NbGa	5.95	–	0.70	−135(4)	−193(28)
Co_2TaAl	5.93	260	0.75	−156(7)	−208(30)
Co_2CrAl	5.89	334	0.78	−124(5)	−159(23)

increasing field with increasing conduction electron density. For example, when a group IIIB element Sc at the Y site is substituted by a group IVB element Ti, Zr or Hf with higher number of d-electrons, the fields increase. However, the behavior of V and Nb is anomalous in this respect since the reduced fields decrease when a group IVB element (Ti, Zr or Hf) is replaced in the alloy by a group VB element (V or Nb) with still higher number of d electrons. Results of the present measurements are consistent with these earlier observations. The reduced fields H_{Ta}/μ_{Co} for the alloys Co_2VGa, Co_2NbGa and Co_2TaAl, all of which containing a group VB element, are very similar (about -200 kOe/μ_B) and are in approximately the same range observed earlier for other alloys containing V and Nb [10]. The value of H_{Ta}/μ_{Co} for the alloy Co_2CrAl is somewhat lower (about -160 kOe/μ_B), suggesting that the earlier observed anomalous behavior for group VB elements persists, although in a less pronounced manner, when a group VB element is substituted by a group VIB element.

Theoretical calculations of the magnetic hyperfine field at the non-magnetic atom site in the Heusler alloys have been made in the past using models of Jena and Geldart (JG) [15–17] and Blandin and Campbell (BC) [18,19]. In particular, these models were used to explain the systematic behavior of Sn fields, measured at Z site in the alloys of the type X_2MnZ. The theoretical predictions have been used to investigate the hyperfine fields in two situations: (1) in a series of alloys of the form X_2MnZ (X = Ni, Pd, Cu) where the Sn field increases with increasing conduction electron density, and (2) in cobalt-based Heusler alloys of the form Co_2MnZ and Co_2YZ, where the Sn field has been found to decrease with increasing conduction electron density. The behavior of the Sn field in first series of alloys has been

Table 2
Magnetic properties of the Co_2YZ 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_{Sn}/μ_{Co} (kOe/μ_B)
Co_2ScGa	6.17		0.25	(−)90(2)	(−)360	
Co_2ScGe	5.78		0.55	−209(2)	−380(4)	
Co_2ScSn	6.19	268	0.55	−187(3)	−341	+73(13)
Co_2TiAl	5.85	138	0.35	−143	−409	
Co_2TiGa	5.85	130	0.40	(−)159	(−)398	
Co_2TiSi	5.74	375(4)	0.55	−287(6)	−522	
Co_2TiGe	5.83	386(4)	0.89	−312(6)	−351	
Co_2TiSn	6.07	371	1.03	(−)480	(−)466	+81(8)
Co_2ZrAl	6.08	178	0.30	−184	−613	
Co_2ZrSn	6.25	448	0.80	(−)380	(−)475	+110(14)
Co_2HfAl	6.02	193	0.40	−189(4)	−473	
Co_2HfGa	6.03	186	0.30	−213	−710	
Co_2HfSn	6.22	396	0.77	−421(12)	−547	+138(18)
Co_2VAl	5.80	310(4)	0.92	−116(4)	−126	
Co_2VSn	5.98	105	0.60	(−)108(2)	(−)180	+12(3)
Co_2NbAl	5.94	383	0.67	−138(4)	−206	
Co_2NbSn	6.15	105	0.26			+58(15)

explained well by the conduction electron polarization model of JG [20–22]. While the BC theory does not provide a definite relationship between the hyperfine field and the conduction electron density, the calculated field values show similar behavior as predicted by Jena and Geldart [17]. In the case of cobalt-based Heusler alloys, on the other hand, the JG model predictions disagree completely with the experimental values of the hyperfine fields [7,23–25], whereas the BC theory provided a reasonable agreement with the experimental results in some cases [23]. No attempts were made in the past to predict theoretically the behavior of Y site hyperfine fields in the alloys of the form Co_2YZ largely because of the lack of experimental results.

In the BC model the Heusler alloy is considered

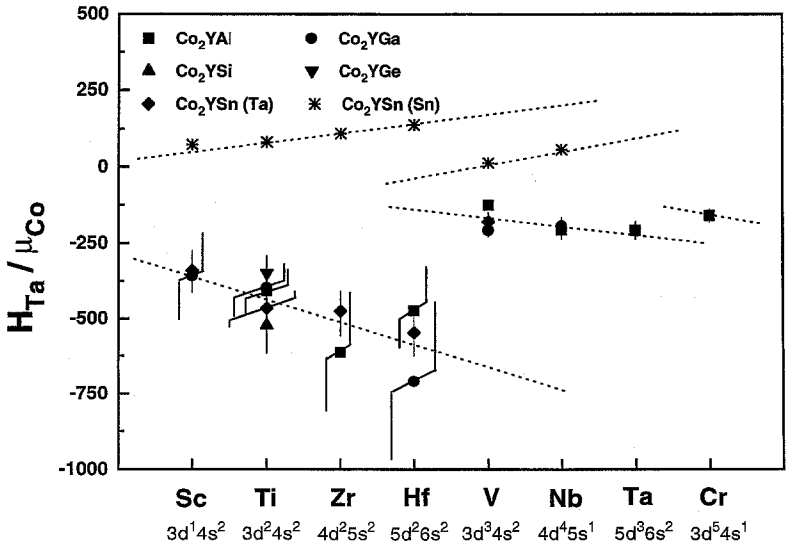

Fig. 2. Reduced mhf at Ta and Sn as a function of the outer electron configuration of the Y site transition element in the Co_2YZ alloys.

to be a non-magnetic host with magnetic as well as non-magnetic impurities. The interaction between the electrons from the conduction band of the host and the electrons from the magnetic ions induces an oscillation on the conduction electron density which is scattered by the non-magnetic impurity potential producing a net spin density on the non-magnetic site. The polarization $p(r_i)$ of the conduction band at a particular probe site due to a unit magnetic moment located at r_i is calculated using an extension [18] of the Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction which takes into account perturbations in the conduction electron density resulting from localized charge at the probe atom as

$$p(r_i) = \left(1/r_i^3\right)\cos(2k_F r_i + 2\delta_0 + \eta), \tag{3}$$

where k_F is the free-electron Fermi vector given in terms of the average number of conduction electrons per atom n_0 as

$$k_F = 1/a\left(48\pi^2 n_0\right)^{1/3}, \tag{4}$$

where a is the lattice parameter. The term δ_0 is a phase shift which accounts for the perturbation of the conduction electron density due to the effective charge of the probe atom and is given by

$$\delta_0 = \pi/4(Z - n_0), \tag{5}$$

where Z is the probe valence. The term η is a pre-asymptotic correction factor. The hyperfine field at the non-magnetic impurity site is expressed as a sum of the partial contributions from the neighboring magnetic moments [26]:

$$H = A\sum_i \mu(r_i)\, p(r_i), \tag{6}$$

where A is the hyperfine coupling constant and μ is the magnetic moment of an atom located at r_i. The sum is taken over all the magnetic neighbors.

In the following we describe some of the salient features of the theoretical calculations of the hyperfine fields at ^{181}Ta probe on the Y site in the Heusler alloys Co_2YZ. The calculated results will be compared with experimentally observed behavior of the Y site fields in these alloys. We will focus our attention on the general trends observed in a series of alloys with Y elements belonging to different chemical groups rather than comparing the precise values of mhf for single systems.

The average number of conduction electrons per atom for Co_2YZ type of Heusler alloys can be calculated from the expression [23]

$$n_0 = 1/4\left[2\left(L_{Co} - 2N_{\downarrow Co} + \mu_{Co}\right) + N_Y + N_Z\right], \tag{7}$$

where L_{Co} is the number of outer shell Co electrons, $N_{\downarrow Co}$ is the number of spin-down Co electrons, μ_{Co} is the magnetic moment localized on Co atom, and N_Y and N_Z are the numbers of electrons contributed to the conduction band by the Y and Z atoms, respectively. Assigning values to N_Y and N_Z has been some what empirical in the past. It is believed however that the value of N_Z should be assumed to be the same as its chemical valence. Dunlap and Stroink [23,27] assumed $N_{\downarrow Co} = 4.7$, and the values of N_Y for group IV element Ti and group V element V were taken as 2 and 4, respectively. With these values the authors [23] obtained a reasonably good correlation between the reduced field at Sn (H_{Sn}/μ_{Co}) in a series of alloys of type $Co_2Ti_xV_{1-x}Sn$ where increasing the conduction electron density in Co_2TiSn was found to reduce the Sn hyperfine field.

The term η in Eq. (3) is an additional phase shift which accounts for the pre-asymptotic behavior of the RKKY oscillations and has a radial dependence given by [17]

$$\eta(r_i) = \pi a/4r_i. \tag{8}$$

The pre-asymptotic factor η has usually been taken as $\pi/2$ for Z site atoms which are 2nn of the magnetic ion in the Mn-based Heusler alloys [19,24]. It was suggested by Yehia et al. [28] that for alloys of the type Co_2YZ (Y ≠ Mn) the phase shift should be taken as π for 2nn in order to explain the observed trends in the Z site fields measured with ^{119}Sn and ^{111}Cd probes. We feel however that the choice of η in Heusler alloys X_2YZ should be made so that one gets the correct sign of the field at a given probe site. Furthermore, one is not necessarily justified in assuming a priori that the pre-asymptotic correction will be identical for fields at different sites in the same alloy [29].

Using only n_0 and η as parameters, we have calculated the polarization $p(r_{1nn})$ for ^{181}Ta probe $(Z = 5)$ at Y site in the Heusler alloys Co_2YZ. The

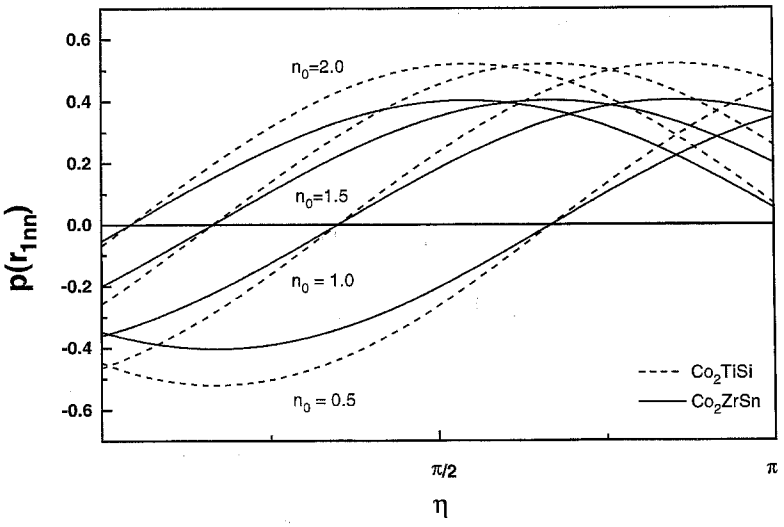

Fig. 3. Conduction electron polarization per unit moment at 1nn for Co_2TiSi and Co_2ZrSn plotted against the pre-asymptotic phase shift for different values of n_0.

values of n_0 relevant to the Heusler alloys of this type were taken in the range 0.5–2.0 [4,23,28]. For a given n_0, η is initialized in the range 0 to π, and the polarization is calculated at 1nn distance from the Co atom. Fig. 3 shows the calculated polarization as a function of η for four values of n_0 and two 1nn distances corresponding approximately to the largest (Co_2ZrSn) and smallest (Co_2TiSi) lattice parameters of the alloys under investigation (see the lattice parameters of the different alloys listed in Tables 1 and 2). From Fig. 3 we find that in order to be able to cover simultaneously the expected range of n_0 values, the polarization can be negative only for a rather limited narrow range of η values.

In order to narrow down further the choice of η we have plotted in Fig. 4 the polarization $p(r_{1nn})$ as

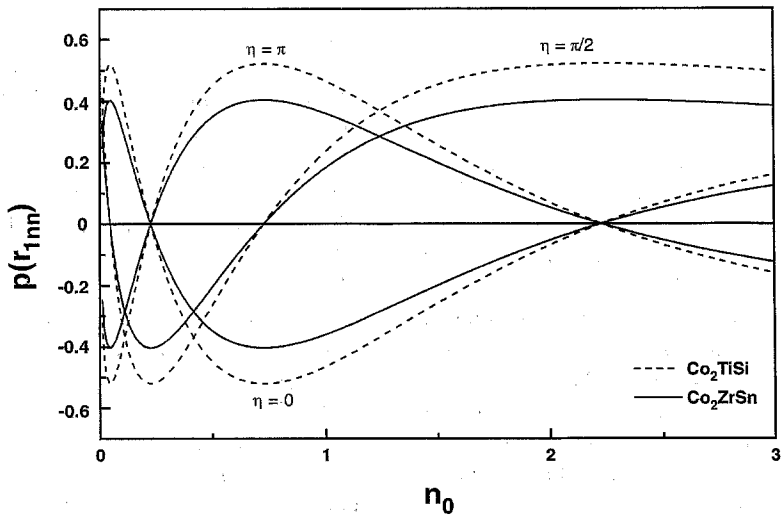

Fig. 4. Conduction electron polarization per unit moment at 1nn for Co_2TiSi and Co_2ZrSn as a function of n_0 for different values of η.

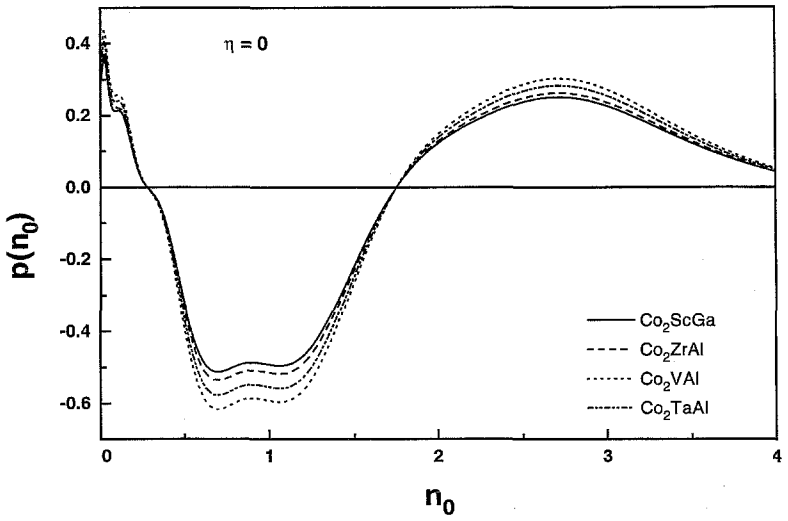

Fig. 5. Conduction electron polarization per unit moment from Eq. (3) summed up to 8nn Co atoms plotted against n_0 using $\eta = 0$.

a function of n_0 for $\eta = 0$, $\pi/2$ and π. It can be seen that for the n_0 values limited to the range 0.5–2.0 the most appropriate choice is $\eta = 0$ where the polarization is negative. Fig. 5 shows the conduction electron polarization as a function of n_0 for some of the Heusler alloys. We have considered magnetic neighbors out to 8nn in the summation in Eq. (6).

The choice of n_0 for an individual alloy was not based on any established criteria. However, since the aim was not to reproduce the individual hyperfine field values but to analyze the general behavior of the field as a function of the chemical nature of the Y element, it was possible to obtain a set of n_0 values for these alloys which would give a reasonable correlation between the calculated hyperfine field values and those observed experimentally. It is expected that this set would be consistent with the generally accepted idea that the conduction electron density increases gradually as the Y element changes progressively from group IIIB to group VIB.

Since the variation of the Z site elements do not appreciably modify the hyperfine fields, the experimental values of the reduced fields as well as the calculated polarizations were averaged for alloys containing the same Y but different Z elements. The results are presented in Table 3, along with the average values of n_0, k_F and δ_0.

Table 3
Values of average reduced mhf, polarization and the parameters used in the calculations

Alloy	H_{Ta}/μ_{Co} (av.) (kOe/μ_B)	n_0 (av.)	δ_0 (av.)	k_F (av.) (Å^{-1})	Σp (av.) (Å^{-3})
Co_2ScZ	−361(78)	0.49	1.77	1.02	−0.319
Co_2TiZ	−429(42)	0.51	1.77	1.06	−0.384
Co_2ZrZ	−544(106)	0.61	1.72	1.07	−0.485
Co_2HfZ	−577(111)	0.63	1.72	1.10	−0.517
Co_2VZ	−171(13)	1.59	1.34	1.56	−0.153
Co_2NbZ	−200(21)	1.55	1.35	1.52	−0.188
Co_2TaAl	−208(47)	1.55	1.35	1.52	−0.190
Co_2CrAl	−156(42)	1.60	1.34	1.55	−0.139

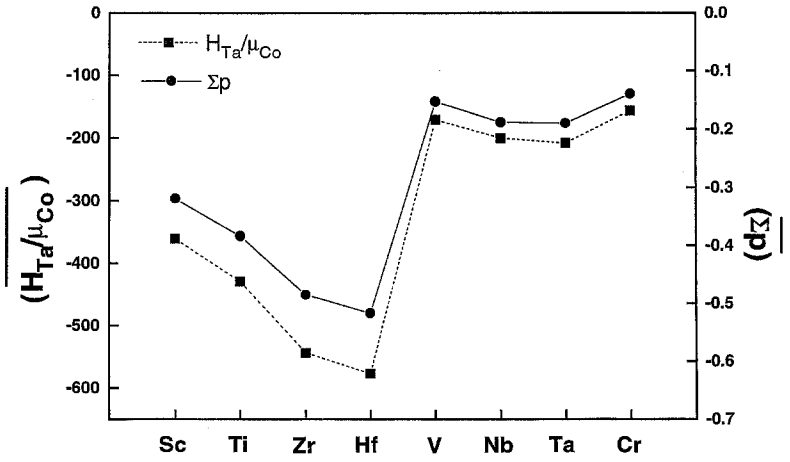

Fig. 6. Comparison of experimental values of reduced hyperfine fields with the calculated polarization as a function of Y element in Co_2YZ Heusler alloys. Experimental as well as theoretical values were averaged for alloys containing the same Y but different Z elements.

The hyperfine coupling constant A in Eq. (6), which represents the strength of the coupling between the conduction electrons and nuclei, is difficult to calculate [30,31]. Usually the atomic hyperfine coupling parameters are used where they exist. However, for a given impurity probe the coupling parameter A is constant and should not be relevant in the present context. The plot of reduced fields and calculated polarizations as a function of Y element is shown in Fig. 6. It can be seen that the correlation between the experimental and theoretical results is reasonably good. It should be emphasized at this point that the general form of theoretically predicted behavior is sensitive only to the range of n_0 values, which are different for each chemical group to which Y element belongs, rather than the precise values for each individual alloy.

Although the RKKY mechanism is based on the assumption that the spin coupling is made essentially between the localized d-electrons (d_l) of the magnetic ion and the s-like electrons of the conduction band, it was pointed out by Stearns [32–34] that a similar coupling may be possible with itinerant d-like conduction electrons also at 1nn distance and beyond. This could explain the observed fact that the mhf at ^{181}Ta increases gradually within each chemical group, when the outer electron configuration of the Y site element changes from 3d to 4d to 5d (see Fig. 2). It reflects an atomic size effect which leads to a higher d-contribution to the conduction electron density for larger atoms in the same group presumably due to a better delocalization of the outer shell electrons.

An examination of the data presented in Table 2 shows that, in every series of alloys containing a given Z element, the value of the local magnetic moment decreases somewhat as the outer electron configuration of Y element changes from 3d to 4d to 5d, and this being observed in both group IVB and VB elements. Although this effect is somewhat obscured by the large experimental errors in the value of the local magnetic moment (μ_{Co}) for most of the alloys, it nevertheless indicates that number of electrons which contribute to form the magnetic moment decreases and, as a consequence, the contribution to the conduction electron band increases in going down the group.

4. Conclusions

The results presented here for the ^{181}Ta field systematics indicate that the observed trends of the hyperfine fields on non-magnetic atoms at the Y site of the Co_2YZ Heusler alloys can be explained by incorporating the pre-asymptotic corrections appropriately in the RKKY like BC model [18] and treating the average number of electrons contributed to the conduction band per atom, n_0 as a parameter to be determined phenomenologically. It is shown that

the most appropriate value of the pre-asymptotic correction factor is $\eta = 0$ for the studied alloys. The only free parameter of the calculation is therefore n_0 which was taken in the range 0.5–2.0 [4,23,28] without being concerned with any other contributing terms for n_0 such as those given in Eq. (7). One of the limitations of the BC model which makes use of the RKKY interaction is that it does not provide any plausible means of calculating the number of conduction electrons per atom, n_0 in the individual systems. This difficulty has been overcome by analyzing the general behavior of hyperfine fields for a series of Heusler alloys as a function of the chemical nature of the Y element rather than attempting to calculate the values for individual systems. It has been further shown that the general form of the theoretically predicted behavior of the polarization is sensitive only to the range of n_0 values assumed for each chemical group to which Y element belongs rather than to the precise n_0 values for each individual alloy. Additional measurements of the Y site hyperfine fields in quaternary Heusler alloys like $Co_2Y^1_{1-x}Y^2_xZ$, where Y^1 and Y^2 belong to different chemical groups, are now in progress. They offer the possibility of observing in more detail the correlation between the hyperfine field and conduction electron polarization.

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