INSTYTUT FIZYKI JĄDROWEJ
INSTITUTE OF NUCLEAR PHYSICS
ИНСТИТУТ ЯДЕРНОЙ ФИЗИКИ


RAPORT No 981/PL



# HYPERFINE MAGNETIC FIELDS AT $^{111}Cd$ IN HEUSLER ALLOYS

B. STYCZEŃ, A. SZYTUŁA, W. WALUŚ

KRAKÓW 1977

# HYPERFINE MAGNETIC FIELDS AT $^{111}Cd$ IN HEUSLER ALLOYS

# NADSUBTELNE POLA MAGNETYCZNE NA JĄDRACH $^{111}Cd$ W STOPACH HEUSLERA

# ЭФФЕКТИВНЫЕ МАГНИТНЫЕ ПОЛЯ НА ЯДРАХ $^{111}Cd$ В ГОЙСЛЕРОВСКИХ СОЕДИНЕНИЯХ

B. STYCZEŃ, A. SZYTUŁA, W. WALUŚ
Instytute of Physics, Jagiellonian University, Cracow

Submitted for publication in Contributed Papers of IV International Conference on Hyperfine Interactions, 1977 June, Madison, New Jersey

C R A C O W, october 1977

*NAKŁADEM INSTYTUTU FIZYKI JĄDROWEJ W KRAKOWIE*
*UL. RADZIKOWSKIEGO 152*

*Kopię kserograficzną, druk i oprawę wykonano w IFJ Kraków*

---

*Wydanie I* *Zam. 251/77* *Nakład 70 egz.*

The magnitudes and signs of the hyperfine magnetic fields on $^{111}Cd$ nuclei at Z sites in the ordered ferromagnetic Heusler alloys $X_2MnZ$ and XMnZ / where X is Cu, Ni, Pd while Z is In, Sn and Sb / have been investigated at liquid nitrogen and room temperatures using TDPAC method. Their signs have been found to be negative. The results have been compared with the predictions of Caroli-Blandin and Campbell-Blandin models and RKKY theory. (author)

Stosując metodę TDPAC ustalono znaki i zmierzono wartości nadsubtelnych pól magnetycznych działających na jądra kadmu umieszczone w pozycji Z ferromagnetycznych stopów Heuslera typu $X_2MnZ$ i XMnZ / gdzie X - Cu, Ni, Pd a Z - In, Sn, Sb /. Pomiery wykonano w temperaturze ciekłego azotu i pokojowej. Dla wszystkich mierzonych pól uzyskano znak ujemny. Eksperymentalne rezultaty zostały porównane z wynikami obliczeń na bazie modeli Caroli-Blandina, Campbella-Blandina i teorii RKKY.

С помощью метода возмущенных угловых корреляций измерены величины и знаки эффективных магнитных полей на ядрах кадмия в местах Z ферромагнитных Гойслеровских соединений типа $X_2MnZ$ и XMnZ (где X - Ni , Cu , Pd , а Z - In, Sn, Sb).

Эксперименты проводились в температуре жидкого азота и в комнатной температуре. Все измеренные поля имели отрицательный знак. Экспериментальные реаультаты были сравнены с теоретическими вычислениями по моделям Кароли-Бландэна, теории РККЫ и Камбела-Бландэна.

## 1. Introduction

The investigated ferromagnetic Heusler alloys containing Mn atoms were magnetically and chemically ordered with $L2_1$ structure, having the typical composition $X_2MnZ$, or with the $C1_b$ structure in the composition XMnZ [1] .

Various magnetic studies have suggested that the magnetic moments of about 4 $\mu_B$ are confined only to the Mn atoms.

The hyperfine fields at non-magnetic element sites in Heusler alloys are especially interested in the relation to (i) the conduction electron spin polarisation, which originates from the Mn magnetic moment and (ii) effective exchange interactions between the magnetic atoms.

The hyperfine fields on sp elements at Z sites in such alloys have been extensively investigated in many work. The exception is the cadmium element for which it is not possible to use the Mössbauer effect and NMR methods in the hyperfine field studies. Therefore, in the present work the TDPAC method was applied.

## 2. Experimental procedure

In all Heusler alloy samples investigated in the present work 1.5 at% of the Z site atoms were replaced by indium atoms containing radioactive $^{111}In$ which decays to $^{111}Cd$.

The appropriate quantities of spectrographically pure elements were pulverized, mixed together, pressed into the form of a pellet and melted in an alumina crucible in argon atmosphere, and then allowed to cool from the molten state to the room temperature. The ingots were pulverized again, mixed and pressed, and then annealed in an appropriate temperature in argon atmosphere for 2-3 days. After annealing the speacimens were either quenched into iced water or slowly cooled-down.

The X-ray powder diffraction analysis that all samples had the cubic structure expected for the Heusler alloys with a slight amount of other phases. The lattice parameters of the alloys were in agreement with the previously reported values.

The hyperfine field measurements were performed by the time-differential perturbed angular correlation TDPAC technique utilizing the 173 - 247 keV $\gamma$-$\gamma$ cascade in $^{111}Cd$. A typical electronic set-up with time resolution $2\nu_0 = 2.6$ ns was used.

The experimental values of the perturbed angular correlation coefficient $A_2G_2(t)$ were fitted with the theoretical function containing the $G_2(t)$ factor appropriate for the magnetic interaction with the assumption of the Lorentzian distribution of the hyperfine magnetic field acting on cadmium nuclei.

The measurements were performed in the liquid nitrogen and room temperatures. The fig.1 shows the example of the experimental curves for the $Ni_2MnSn$ alloy.

The signs of the hyperfine fields were established in additional experiments in which the samples were polarized by an external magnetic field $\sim 5$ kOe . Such experiments were done only for three alloys see fig.2 and for each of them the negative sign was obtained.

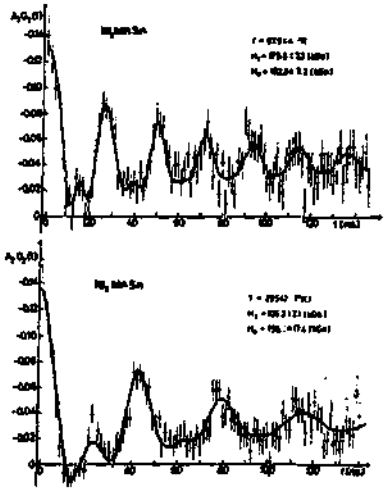

Fig.1. Experimental $A_2G_2(t)$ data for the $Ni_2MnSn$ alloy obtained for the liquid nitrogen (upper half) and the room (lower half) temperatures. The full lines represent the least squares fits.

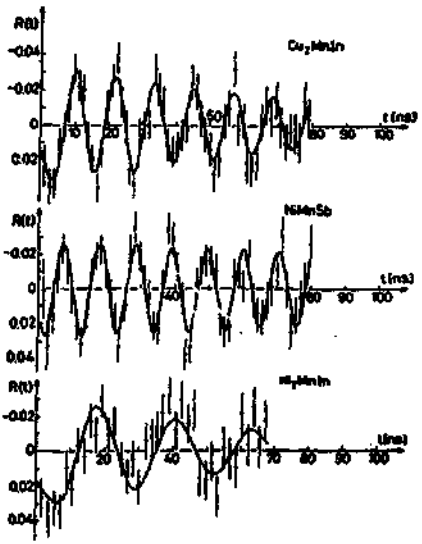

Fig.2. Experimental R(t) curves for polarized Heusler alloy hosts / T = 296 °K /.

The summary of the experimental results is given in table.

Table. Hyperfine fields at cadmium sites

| Alloy | $T_{exp}$ [°K] | $H_T$ [kOe] | $H_o$ [kOe] |
|---|---|---|---|
| $Ni_2MnIn$ | 107 (4)<br>117 (4)<br>296 (2) | - 150.0 (21)<br>- 151.2 (17)<br>- 70.3 (16) | - 155.8 (25)<br>- 158.4 (22)<br>- 171.5 (308) |
| $Ni_2MnSn$ | 88 (4)<br>295 (4) | ± 179.8 (32)<br>± 106.2 (21) | ± 182.8 (33)<br>± 198.2 (174) |
| $Ni_2MnSb$ | 85 (4) | ± 222.8 (30) | ± 225.1 (31) |
| NiMnSb | 95 (4)<br>293 (2) | - 210.8 (41)<br>- 194.3 (38) | - 211.3 (41)<br>- 207.9 (41) |
| $Cu_2MnIn$ | 106 (4)<br>124 (4)<br>295 (2) | - 213.3 (50)<br>- 210.4 (35)<br>- 183.0 (14) | - 214.9 (51)<br>- 213.1 (36)<br>- 216.4 (17) |
| $Cu_2MnSn$ | 295 (4) | ± 153.2 (40) | ± 179.5 (50) |
| $Pd_2MnSb$ | 86 (4)<br>89 (4)<br>91 (4) | ± 225.4 (45)<br>± 227.8 (47)<br>± 234.2 (52) | ± 235.1 (51)<br>± 239.2 (54)<br>± 246.5 (58) |
| PdMnSb | 90 (4)<br>295 (3) | ± 260.9 (45)<br>± 209.7 (40) | ± 262.4 (45)<br>± 252.2 (51) |

The values of the hyperfine fields in 0°K - $H_o$ were obtained by the extrapolation of the $H_T$ values according to the Brillouin curves for $J = 5/2$.

## 3. Theoretical calculations

The hyperfine fields calculations were carried out on the basis of the three models: Caroli-Blandin, RKKY and Campbell-Blandin.

Considering a weak convergency of the summations in the formulae describing hyperfine fields, the contributions of the Mn atoms confined within a sphere of $15a_0$ ( $\approx 90\text{Å}$) were included / this corresponded to aproximately 50 thousands of Mn atoms / then the convergency was complete to one percent.

In C a r o l i - B l a n d i n model [3] the hyperfine magnetic field at the non-magnetic atom site is described by the formula

$$H_{eff} = \frac{\xi |a(s)|}{2|g_I|} \Omega_0 \sum_i \delta(r_i)$$

with $\delta(r_i)$ being the spin polarisation due to all surrounding Mn atoms given by

$$\sum_i \delta(r_i) = \frac{5}{4\pi^2} \sum_i \frac{\cos(2k_F r_i - \delta^{\downarrow})}{r_i^3} \sin\delta^{\downarrow}$$

where $r_i$ is the distance to the i-th Mn atom, $a(s)$ is the hyperfine coupling contant for an s electron in the free atom ( for $^{111}$Cd $a(s) = 0.085$ cm$^{-1}$[3]), $g_I$ is the nuclear ground state gyromagnetic ratio ( for $^{111}$Cd $g_I = -5.9815 \times 10^{-24}$ erg . Oe$^{-1}$[4]), $\xi$ is the ratio of the probability densities of the conduction electrons in the metal and free atom (for $^{111}$Cd $\xi = 1.6$[3]), $\Omega_0$ is volume per atom ($\Omega_0 = a_0^3/16$ and $a_0^3/12$ for $L2_1$ and $C1_b$ structures, respectively), $n_0$ is number of the conduction electrons per atom ( it is normaly only estimated very crudely from valency considerations assuming the conductivity band to be isotropic ), $k_F = (3\pi^2 n_0/\Omega_0)^{1/3}$ is the Fermi wave vector, $\delta^{\downarrow} = \pi(5-m)/5$

is phase shift, m is the magnetic moment on the Mn atom in units of $\mu_B$.

The Curie temperature can be written as

$$T_c = -\frac{50 k_F^2 k^2}{3\pi k_B m_e^*} \sin\delta^4 \sum_{\text{(over Mn atom pairs)}} \frac{\cos(x_i + 2\delta^4)}{x_i^3}$$

where $x_i = 2k_F r_i$, $k_B$ is the Boltzmann constant, $m_e^*$ is the effective electron mass / it was assumed that $m_e^* = 1.5\ m_e$ [5]/.

Fig. 3 shows the results of the calculations of $H_{eff}$/ at Cd/ and of $T_c$ as a function $n_o$ for the $L2_1$ and $C1_b$ structures.

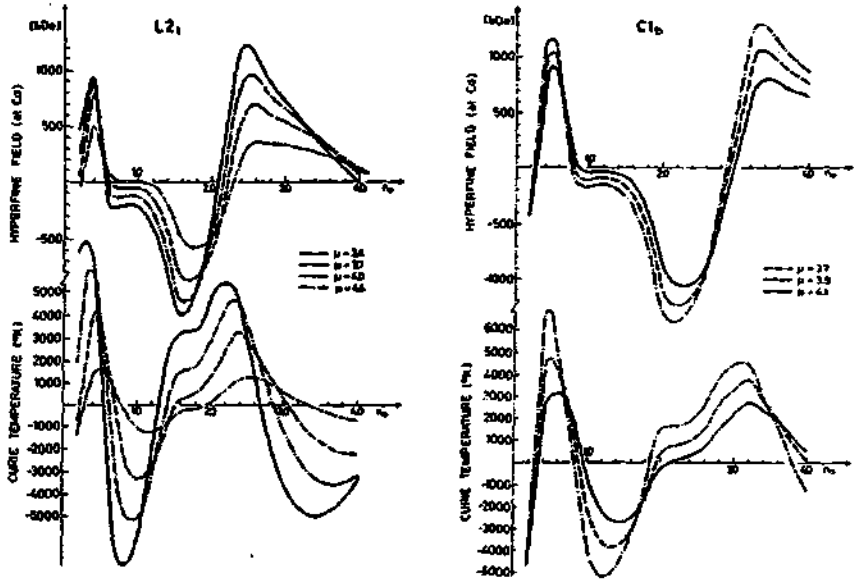


Fig.3. Calculations of $H_{eff}$ and $T_c$ for Heusler alloys according to Caroli-Blandin model. / For the ferromagnetic structure should be $T_c > 0$/.

The traditional R K K Y t h e o r y gives the following expressions for $H_{eff}$ and $T_c$ [6,7,8].

$$H_{eff} = \frac{\gamma |a(s)|}{2 |g\mu|} \Omega_0 \sum_i \delta(r_i)$$

where

$$\sum_i \delta(r_i) = - \frac{6 J(0) k_F n_0 m_e^* \langle S \rangle}{\pi k^2} \sum_i F(x_i)$$

while $F(x) = (\cos^2 x - \sin x) / x^4$, $x_i = k_F r_i$

and

$$T_c = - \frac{12 J^2(0) k_F n_0 \Omega_0 m_e^* S(S+1)}{\pi k_B k^2} \sum_i F(x_i)$$

(over the atomic pairs)

$\langle S \rangle$ is a polarized spin, localized on the Mn atom, $J(0)$ is a constant value taken for the s-d exchange integral / $J(0) = - 0.33$ eV [9] /.

In fig.4 are displayed the results of the calculations according to this model.

It is seen that the theory does not describe the experimental data; there is no such $n_0$ range where $H_{eff} < 0$ and $T_c > 0$, simultanously.

The C a m p b e l l - B l a n d i n model takes into account the influence of the local effects connected with impurity atoms [10].

The hyperfine field at a nonmagnetic site is described by the formula

$$H_{eff} \cong - \frac{J(0) \Omega_0 K}{8\pi \mu_B} \left( \sum_i \langle S \rangle \frac{\cos (2 k_F r_i + 2\delta_0)}{r_i^3} \right)$$

where K is the Knight shift, $2\delta_0 = \pi (Z_I - n_0)/4$ and $Z_I$ is the impurity charge.

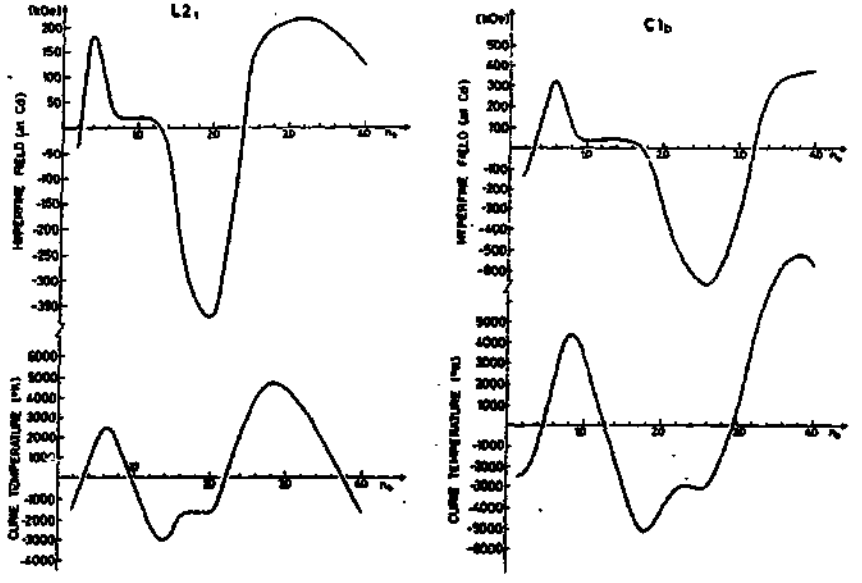


Fig.4. Calculations of $H_{eff}$ and $T_c$ for Heusler alloys according to RKKY theory

The results of the calculations are shown in fig.5. The use of the above asymptotic formula to describe the hyperfine field of the nearest neighbours may yield incorrect results. Therefore, the authors of ref. 12 suggest the preasymptotic phase correction of $\pi/2$ for distances $r_1 \leq 10k_F^{-1}$. It is seen in fig.5 that introducing of this correction worsens the agreement between the theory and the experimental data.

Instead, the Campbell-Blandin model calculations without any correction reproduce fairly well the experimental results. The noticed displacement between the theoretical curve and the measured field values for the investigated Heusler alloys may be due to $n_c$ which as pointed out is uncertain quantity, crudly estimated from electron valency considerations.

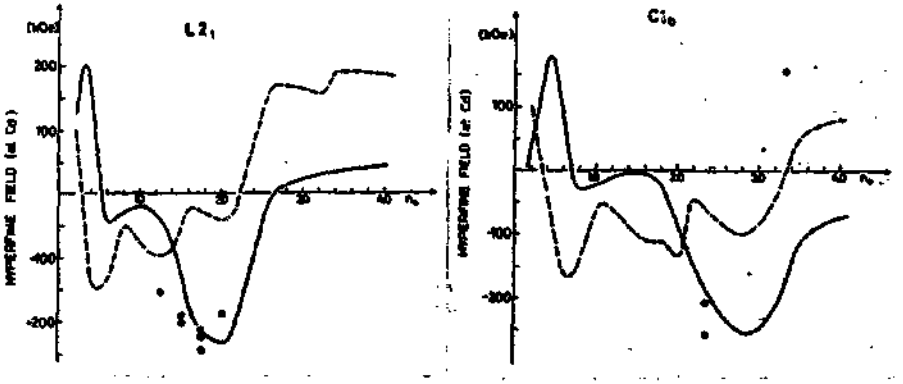


Fig.5. Calculations of $H_{eff}$(at Cd) for Heusler alloys according to Campbell-Blandin model./• - our data,o- data from ref. 11./

References


1. P.J. Webster 1969 Contemp. Phys.10, 559-77
2. B.Caroli, A. Blandin 1966 J.Phys.Chem.Solids 27,503-8
3. W.D.Knight 1956 Solid state Physics vol.2 ed.F.Seits and D.Turnbull/New York: Academic Press/ pp 93-136
4. I.Lindgren 1965 Ark.Fys. 29, 553-63
5. J.S. Brooks,J.M.Williams 1974 J.Phys.F:Metal Phys. 4,2033-49
6. M.A.Ruderman,C.Kittel 1954 Phys.Rev. 96, 99-102
7. J.Kasuya 1956 Prog.Theor.Phys.16,45-57
8. D.Yosida 1957 Phys.Rev. 106, 893-8
9. K.Endo, K. Tabushi,R.Kimura 1972 J.Phys.Soc.Japan 32, 285-8
10. A. Blandin, I.A.Campbell 1973 Phys.Rev.Lett. 31, 51-4
11. B.Lindgren, K.Pernestål, S.Bedi,E.Karlsson 1975 Annual Report of Uppsala University, 33-7
12. I.A.Campbell, A. Blandin 1974 Report of Universite Paris-Sud, Orsay