ANNALES
UNIVERSITATIS MARIAE CURIE-SKŁODOWSKA
LUBLIN — POLONIA
VOL. XL/XLI, 7 SECTIO AAA 1985/1986

Instytut Fizyki UMCS

M. BUDZYŃSKI, M. SUBOTOWICZ, H. NIEZGODA,
H. SPUSTEK, W. TAŃSKA-KRUPA, R. WASIEWICZ

Influence of High Pressure on the Hyperfine Interaction Parameters in Laves Phase Compounds: $(Y_{0.9}Hf_{0.1})Fe_2$ and $(Zr_{0.9}Hf_{0.1})Fe_2$

Wpływ wysokiego ciśnienia na parametry oddziaływań nadsubtelnych w związkach Lavesa: $(Y_{0.9}Hf_{0.1})Fe_2$ i $(Zr_{0.9}Hf_{0.1})Fe_2$

Влияние высокого давления на параметры сверхтонкого взаимодействия в фазовых соединениях Лавеса $(Y_{0.9}Hf_{0.1})Fe_2$ и $(Zr_{0.9}Hf_{0.1})Fe_2$

1. INTRODUCTION

The influence of pressure on the hyperfine interaction parameters has been studied for some years but it still is little known. The high pressure investigations are difficult, mainly because of the technological problems connected with the generation, the measurement and maintaining of high pressure for a sufficiently long time. This pressure should be at least quasihydrostatic to avoid structural defects in the sample investigated.

For some years the intensive and comprehensive studies of Laves phase compounds RFe_2 (where R is the rare earth element) have been performed. Hyperfine interactions in these compounds have been investigated, including the dependence of interaction parameters on the distance between ions and so-called "lantanic squeezing".[1]

2. EXPERIMENTAL

The samples of $(Y_{0.9}Hf_{0.1})Fe_2$ and $(Zr_{0.9}Hf_{0.1})Fe_2$ were obtained by melting the stoichiometric quantities of initial components in the induction or arc furnace in the inert gas atmosphere. Only the central part of the cast from each smelt has been used to produce the samples. The annealing of samples in vacuum (10^{-6} Tr) at temperature 1250 K for 200 hours has been carried out to increase or to reconstruct their homogenity. After they had been exposed for 100 hours to a neutron flux $\Phi = 7 \times 10^{13}\, n/cm^2 \cdot s$ they were annealed once again to remove the radiation defects. The investigated compounds crystallize in $MgCu_2$ structure, shown in fig.1.

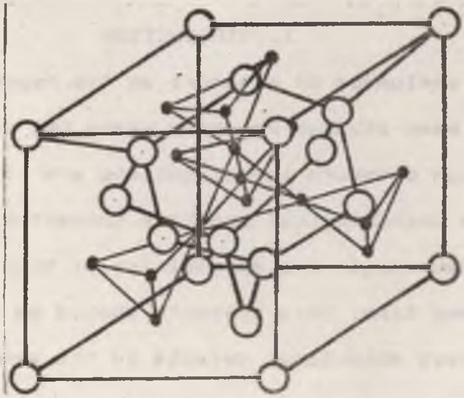

Fig.1. Laves phase compound $MgCu_2$. ● – Cu, ○ –Mg.

High pressure was produced in Bridgman's anvil pressure chamber (see fig.2), similar to that described in [2]. The anvils were made from sintered carbide alloy. The volume of samples was about 0.05 mm^3 . The greatest errors arose during the measurement of the pressure acting on the radioactive sample. The pressure was measured by means of a tensometer stuck into the interior wall of the chamber (3 in fig 2).

During the turning of cap(1) which exerted the pressure the Bridgman's anvils, the wall elongated a litlle; that led to the increase of the resistivity of the tensometer. This resistivity was measured by means of the bridge. The pressure change of the resistivity of the conductor (8) during the phase transitions in Bi, Sn and Pb was utilized to calibrate the pressure. Simultaneously the calibration of the conductor (7) made from manganine was performed. Its resistivity increases linearly with the pressure growth. After the calibration we put the tablet (d) instead of the control tablet (c). In the central part of the tablet (d) the mixture of the tested samples was placed together with the powdered substance of large coefficient of the internal friction and transferred the pressure. Usually it was a mixture of boron nitride and amorphic boron. The edges of the tablet (d) were formed of the substance transferring the pressure. Owing to this construction one could suppose that our radioactive sample was submitted to the quasihydrostatic pressure.This pressure was measured by the increase of the resistivity of the wire made from manganine. If it would be broken however, one would use the indications of the tensometer. For smaller values of pressure the anvils with hollowed

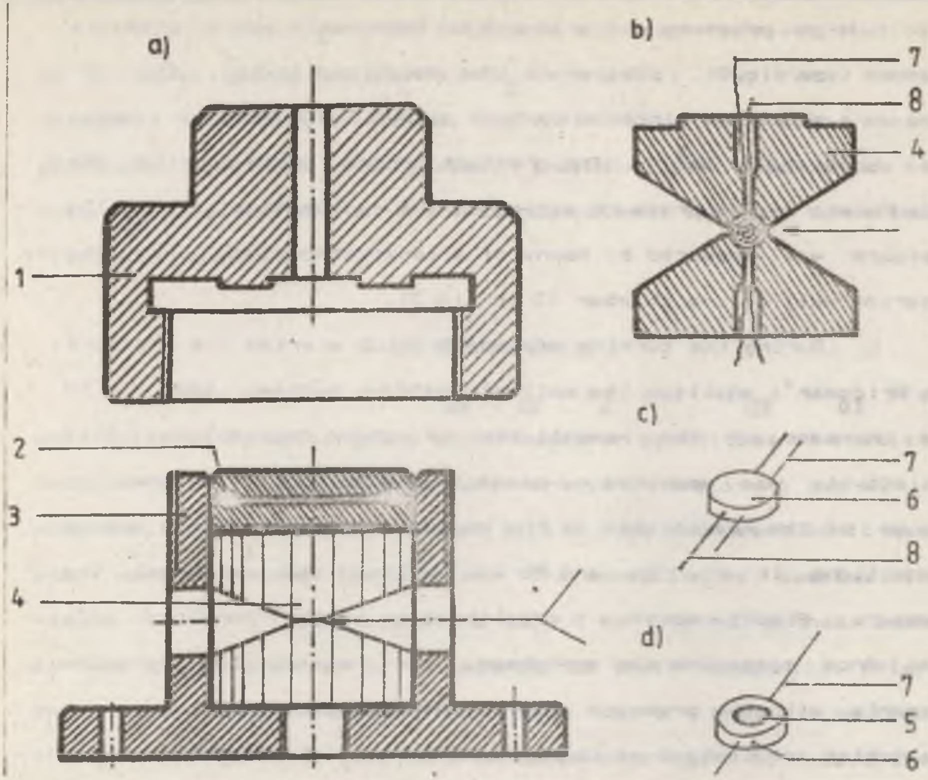

Fig.2. The chamber for the generation of the hydrostatic high pressure: it is equipped wth the anvils of Bridgman's type a)with flat anvils ; b)with hollowed anvils c) control tablet; d)the tablet with sample investigated Notation: 1) cap; 2) ball bearing; 3) frame; in its interior the tensometer is stuck; 4) anvils; 5) sample investigated; 6) mixture of nitride of boron and amorphic boron; 7) conductor made from nickeline; 8) conductor made from Bi, Sn or Pb.

hemispheres (fig 2b) assured the homogenuous distribution of pressure.Such a construction guaranteed the self-seal on the edges of the anvils.

TDPAC measurements have been performed for (133-482)keV cascade in ^{181}Ta (after the β^- decay of ^{181}Hf). A standard spectrometer has been used. with BaF$_2$ and NE 111 scintilators working with XP 2020 Q photomultiplier. The time resolution was equal to $2\tau_0 = 0.9 \div 1.9$ ns. Measurements were performed on policrystalline samples without the external magnetic field.

In the investigated materials the perturbation of the angular correlation is caused by the hyperfine magnetic interactions. For this reason the spin precession function can be written as follows:

$$R(t) = a_0 + \sum_{i=1} a_i \left(e^{-\Lambda_i \omega_i t} \cos \omega_i t + e^{-2\Lambda_i \omega_i t} \cos 2\omega_i t \right) \qquad (1)$$

a_0 parameter takes into account the influence of the nucleus which occupy the irregular positions in the crystalline lattice. The values of a_i coefficients are proportional to the number of nuclear samples in the positions influenced by the internal magnetic field B_{hf}. In this case the following relation is realized between the Larmor precession frequency ω_i and the magnetic field B_{hf}:

$$\omega_i[rad/s] = -62.26 \cdot 10^6 B_{hf}[T]$$

Λ parameter means the relative width ($\Lambda = \lambda/\omega_0$. λ - the width at half maximum, ω_0 - the frequency for the given position of the ions and the Lorentz-type frequency distribution. The values of hyperfine interaction parameters, obtained from our experiment are given in Table I.

3. RESULTS

As Table I indicates, the amplitude of the measured hyperfine magnetic fields on ^{181}Ta nuclei occupying the position of R ions, i.e. Y or Zr, increases with the increase of pressure in both compounds. The smearing of B_{hf} values (indicated by Λ parameter) increases, too. This may be connected with the fact that the pressure acting on the sample is not quasihydrostatic because of some imperfections in the construction of our pressure chamber.

Table I
The influence of pressure on the change of hyperfine magnetic fields for ^{181}Ta nuclei in the investigated samples.

Sample	P[kbar]	B_{hf} [T]	Λ
$(Y_{0.9}Hf_{0.1})Fe_2$	0	-14.40(10)	0.05(1)
	27(1)	-14.95(15)	0.07(1)
	50(3)	-15.10(15)	0.09(1)
	74(3)	-15.45(15)	0.11(1)
	80(4)	-15.95(15)	0.12(1)
$(Zr_{0.9}Hf_{0.1})Fe_2$	0	-6.55(10)	0.03(1)
	24(1)	-7.05(15)	0.09(1)
	50(3)	-7.34(15)	0.09(1)
	75(3)	-7.65(15)	0.10(1)
	85(4)	-8.07(15)	0.12(2)

It is interesting to compare the pressure derivatives measured by us and those obtained for the same compounds by means of NMR method [3]. They are similar as far as the signs of values are considered. Our results confirm the difference between signs of $\partial \ln B_{hf}/\partial p$ for the radioactive samplers occupying the Fe positions and those of R (i.e. Ta, Y and Zr).

In [4] a supposition, based on the experimentally observed abnormal temperature dependence of hyperfine fields, has been put forward. It claimed that in R positions, occupied by nuclear samplers Ta, the localized magnetic moment was created, although the Y, Zr and Ta atoms were nonmagnetic.

It has been assumed that the measured hyperfine magnetic field is the sum of the fields originating from core polarization (B_{hf}^{CP}) and conduction electrons polarization (B_{hf}^{CEP}):

$$B_{hf} = B_{hf}^{CP} + B_{hf}^{CEP}$$

The hyperfine field B_{hf}^{CP} connected with the core polarization is of exchange interaction origin. It is antiparallel to the magnetic moment, acting on the atom. The B_{hf}^{CEP} contribution is proportional to the magnetic moment localized near the nuclear sampler. Taking into account the results of [4,5] one can estimate the contribution of both components to B_{hf} on the Ta nuclei at room temperature: $B_{hf}^{CP} \approx +1.8$ T and $B_{hf}^{CEP} \approx -16.2$ T in YFe_2 and $B_{hf}^{CP} \approx +11.4$ T and B_{hf}^{CEP} -17.9 T in $ZrFe_2$.

Recently the energy band calculations were performed for YFe_2 and $ZrFe_2$, applying the ASW method [6]. It has been revealed that in the positions occupied by Y or Zr, the magnetic moment exists equal to $-0.45\,\mu_B$ and $-0.56\,\mu_B$, respectively. It is antiparallel to the Fe magnetic moment. The last one is equal to $1.66\,\mu_B$ for YFe_2 and $1.90\,\mu_B$ for $ZrFe_2$.

Generally, the pressure changes the hyperfine interaction parameters by changing 1) the distribution of density of states, namely the distribution of the conduction electrons, 2) the distance between ions and 3) the deformation of electronic shells (mainly valence ones) . The local deformation of the lattice around Ta ions plays particular role, too.

The contribution of these factors may be different in various samples and linear approximation, presented in fig.**3** is the first approximation. For higher pressures the deviation from linearity occurs. Because of lantanic squeezing [1], experienced by atoms occupying the R positions, the external pressure leads mainly to the decrease of the value of the magnetic moment localized in this position and consequently to the decrease of

B_{hf}^{CP} . Because of opposite signs B_{hf}^{CP} and B_{hf}^{CEP} and reciprocal relation between them, it leads to the increase of the amplitude of magnetic field observed on Ta nuclei (see Table II). In the above considerations we assumed that B_{hf}^{CEP} does not change with the pressure, or its decrease with the increase of pressure is considerably smaller than in the case of B_{hf}^{CP} .

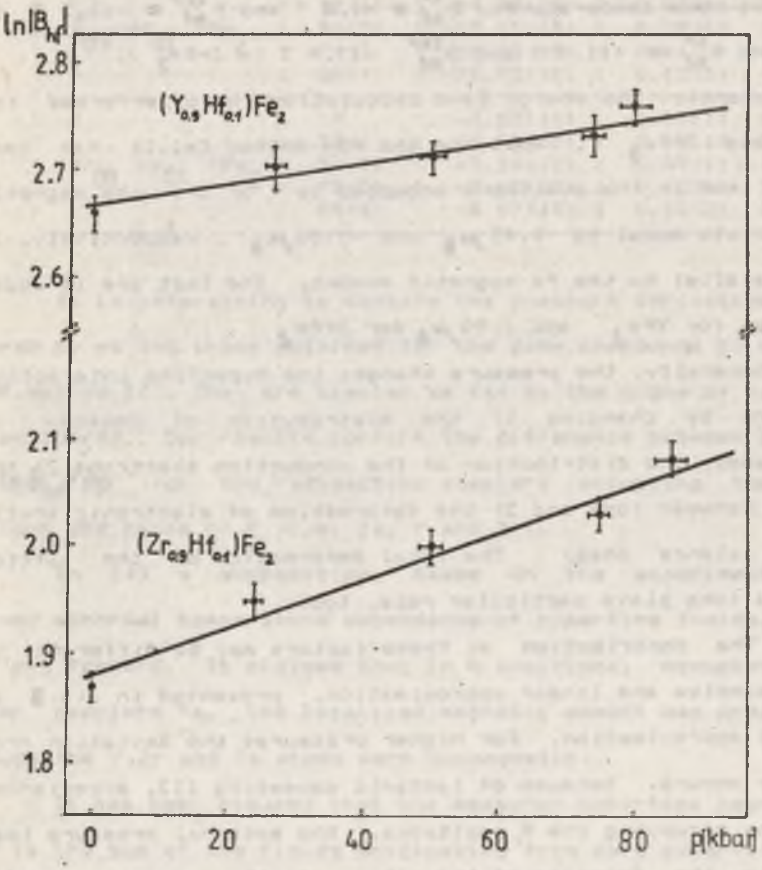

Fig.3. The pressure dependence of ln |B_{hf}| for $(Y_{0.9}Hf_{0.1})Fe_2$ and $(Zr_{0.9}Hf_{0.1})Fe_2$.

The decrease of Fe magnetic moment does not necessarily lead to the reduction of fields induced on Ta (transferred hyperfine fields) . It is true, if this field is compensated by the decrease of overlapping of the wavefunction of Fe(3d)-Ta(6s),like in the case of YFe_2[3]. Such behavior was noticed for nickel [7].

On the contrary, the decrease of the localized moments of Fe nuclei connected with the increase of the pressure, causes the magnetic field reduction ($\partial ln B_{hf}/\partial p < 0$). because the B_{hf}^{CP} contribution is very small in this case.

The difference between values $\partial ln B_{hf}/\partial p$ measured in B positions for Y(Zr) and Ta is caused probably by the fact that the contributions from the core polarization for 5d ions are the greatest in the whole periodic table (and are equal to 73T/unpaired spin/). Certainly, the induced localized magnetic moment on Ta impurity atoms will be different from the intrinsic moment of Y(Zr) atoms in YFe_2 or $ZrFe_2$ compounds, as far as its value is concerned.

Table II.
Hyperfine fields and their pressure derivatives.

Sample	Nuclear sampler	B_{hf}[T] at P=0	$\frac{\partial ln B_{hf}}{\partial P}$ [10^{-3} kbar]	Temp. [K]	Lit.
$(Y_{0.9}Hf_{0.1})Fe_2$	^{181}Ta	-14.4(1)	11.0(1)	300	*)
YFe	^{89}Y	-22.0(1)	6.7(3)	4.2	[3]
YFe	^{57}Fe	-21.3(1)	-4.2(2)	4.2	[3]
$(Zr_{09}Hf_{0.1})Fe_2$	^{181}Ta	-6.5(1)	22.3(3)	300	*)
ZrFe	^{91}Zr	-12.6(1)	11.1(3)	4.2	[3]
ZrFe	^{57}Fe	-22.2(1)	-7.3(1)	4.2	[3]

*) our experiment

Acknowledgements

The authors acknowledge the financial support of the Goverment's Central Programm of the Fundamental Research CPBP-01.09 in the realization of the present paper.

References:

1. E.Burzo, Z.Angew.Phys. 32(1971)127.
2. Z.Z.Akselrod, B.A.Komissarova, L.N.Kryukova, A.A.Opalenko, G.K.Ryasnyi and A.A.Sorokin, phys.stat.sol. (a) 73(1982) K229.
3. T.Dumelov, P.C.Riedi, P.Mohn, K.Schwarz and Y.Yamada J.Magn.Mat. 54-57 (1986) 1081.
4. Z.Z.Akselrod, M.Budzynski, T.Khazratov, B.A.Komissarova, L.N.Kryukova, S.J.Reiman, G.K.Ryasnyi and A.A.Sorokin Hyp. Int. 14 (1983) 7.
5. Z.Z.Akselrod, M.Budzynski, B.A.Komissarova, L.N.Kryukova, G.K.Ryasnyi and A.A.Sorokin phys. stat. sol. (b) 119 (1983) 667.
6. P.Mohn and K.Schwarz, Physica B, 130 (1985) 26.
7. J.F.Janak. Phys.Rev.B. 20 (1979) 2206.

Streszczenie

Przeprowadzono pomiary TDPAC w temperaturze pokojowej w celu wyznaczenia pól nadsubtelnych na jądrach ^{181}Ta w związkach $(Y_{0.9}Hf_{0.1})Fe_2$ i $(Zr_{0.9}Hf_{0.1})Fe_2$. Wyznaczono wielkości tych pól w funkcji ciśnienia do 80 kbar, oraz ich pochodne ciśnieniowe $\frac{\partial \ln B_{hf}}{\partial p}$. Uzyskane wyniki potwierdzają różnicę znaków $\frac{\partial \ln B_{hf}}{\partial p}$ dla próbników jądrowych obsadzających położenia Fe i położenia R /tzn. Ta, Y i Zr/. Jest to zgodne z przypuszczeniem,

że w położeniach R, zajmowanych przez próbniki Ta indukowany jest moment magnetyczny, mimo to, że atomy Ta, Y i Zr są niemagnetyczne.

РЕЗЮМЕ

Для определения сверхтонких полей на ядрах ^{181}Ta в соединениях $(Y_{0.9}Hf_{0.1})Fe_2$ и $(Zr_{0.9}Hf_{0.1})Fe_2$ проводились измерения ДВУК при комнатной температуре. Определены значения этих полей как функции давления, а также их производные относительно давления, $\frac{\partial \ln B_{hf}}{\partial p}$. Полученные результаты подтверждают тот факт, что величина $\frac{\partial \ln B_{hf}}{\partial p}$ имеет разные знаки для ядерных зондов в положении Fe и в положении R (т.е. Ta, Y, Zr). Это согласуется с предположением, что в положениях R занимаемых зондами Ta индуцируется магнитный момент - несмотря на то, что атомы Ta, Y и Zr - немагнитные.

ne w położeniach B, [illegible]
[illegible] magnetyczny, gdyż atomy Ti, Y i Zr, są niemagnetyczne.

Р е з ю м е

Для определения сверхтонких полей на ядрах ^{119}Sn в соединениях $(Ti_{0.9}Sn_{0.1})Fe_2$ и $(Zr_{0.9}Sn_{0.1})Fe_2$ проводились измерения ЯГР при комнатной температуре. Определены значения этих полей как функция давления, а также их производные относительно давления, $\frac{d\ln H}{dP}$. Полученные результаты подтверждают тот факт, что величина $\frac{d\ln H}{dP}$ имеет равный знак для ядерных полей в положении Fe и в положении B (т.е. Ti, Y, Zr). Это согласуется с предположением, что в положениях B ядерные поля на Sn индуцируются магнитными моментами Fe, в то время как атомы Ti, Y и Zr – немагнитные.