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1. Introduction

The interaction of the nuclear magnetic moments with the magnetic field existing at the nuclei /hyperfine interaction/ offers many possibilities for the study of the phenomenon of magnetism. The methods for the investigation of the hyperfine interaction can be divided into two groups according to the excitation of the nuclear states involved. The magnetic moments of the ground states of nuclei are of interest in nuclear magnetic resonance and neutron scattering methods /via the magnetic moment of the neutron/, while those of the excited states, decaying by γ-emission, in Mössbauer-effect as well as perturbed angular correlation and distribution methods. The latter techniques are rather new and although they are not so general, they have some features which make them preferable in some cases. These new methods are now developing very rapidly and already a lot of new information on magnetism has been collected by their use.

In this report only the perturbed angular correlation and distribution method will be dealt with.

2. Method of perturbed angular correlation and distribution.

2.1. Angular correlation and distribution

The γ-radiations emitted by the nuclei have characteristic anisotropic angular distribution relative to the direction of the spin of the nuclei in excited state. The distribution depends on the spins of

the initial excited and final states and on the multipolarity of the γ-radiation. This angular distribution cannot be observed generally because of the random distribution of the directions of nuclear spins in radioactive material. In the case of nuclear polarization the spin and multipolarity values can be determined from the angular distributions. The polarization of nuclei requires a rather difficult technique, fortunately the measurement of angular correlation gives results of similar value. Two γ-radiations emitted from a nucleus in cascade /see fig.1./

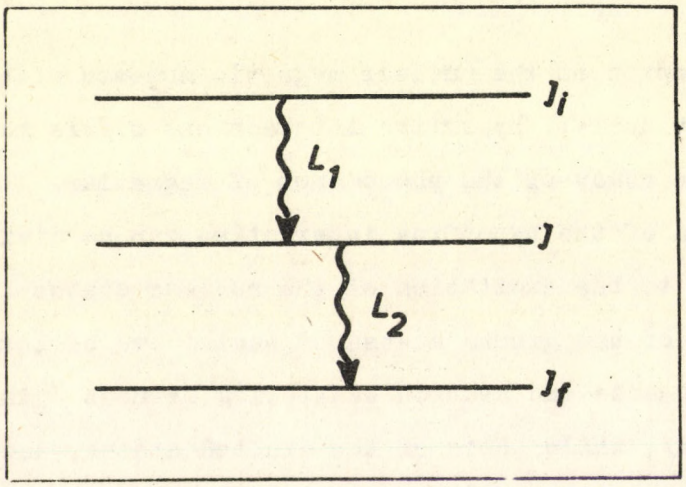

Fig.1.
γ-rays emitted in cascade

are measured with two counters /fig.2./

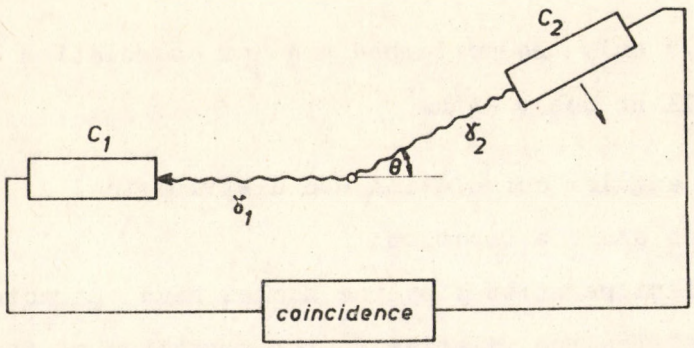

Fig.2.
Scheme of apparatus to measure angular correlations

connected in coincidence with resolving time T. The number of coincidences depends on the angle ϑ between the two counters since after the observation of the first radiation the spin distribution is already not random, some degree of polarization is obtained and the second radiation is emitted from "polarized" nuclei with their characteristic asymmetry. The angular correlation function $W(\vartheta)$ depends on the spins of the three nuclear states J_i, J, J_f and on the multipolarity L_1, L_2 of the two γ-radiations. It can be written in the form[1]

$$W(\vartheta) = \sum_{k=0}^{k_{max}} A_k P_k(\cos\vartheta),$$

where $\vartheta_{max} = \text{Min } /2J, 2L_1, 2L_2/$, the $P_k(\cos\vartheta)$-s are the Legendre-polynomials and the A_k-s are the coefficients of the angular correlation function which can be calculated from tables[2] if J_i, J, J_f, L_1 and L_2 are known. The exprerimental results on angular correlations of γ-rays from different nuclei can be found in ref[3].

If the excited intermediate state is obtained in nuclear reaction then instead of the observation of the first γ-radiation "polarization" can be produced by the bombarding particle. In such cases the angular distribution of the γ-radiation to the direction of the incoming particles is to be measured only to obtain the same data as from the angular correlation. The most important nuclear reactions from this point of view are the $(p, \alpha), (\alpha, \gamma)$ reactions and the Coulomb excitations.

2.2. Perturbed angular correlation and distribution

The magnetic dipole and electric quadrupole moments of the excited state interact with magnetic fields and electric field gradients existing at the nuclei. As a result of this the $W(\vartheta)$ will be changed generally in a very complicated manner[1]. In the cases of static magnetic and electric fields the functions are rather simple and have obvious meaning. If the static magnetic field is perpendicular to the plane of the angular correlation and distribution, then we have

$$W(\vartheta, H, t) = \sum_{k=0}^{k_{max}} A_k P_k(\cos(\vartheta\,\omega t)),$$

where $\omega = g\mu_N H/\hbar$ is the frequency of Larmor precession, $/g\mu_N J$ is the magnetic moment of the intermediate state, μ_N is the nuclear magneton, H the magnetic field/, t is the time elapsed from the emission of the first to the emission of the second γ -radiation. The effect of the field H is that the nuclear magnetic moment, i.e. the spin of the nucleus in the excited state rotates around the field direction with frequency, ω the second γ -radiation is emitted in time t from a nucleus the direction of which has changed with an angle of ω t. Therefore the angular correlation and distribution patter rotates.

The rotation can be observed in two different ways depending on the lifetime of the intermediate state: by the differential method, ω is measured directly. The number of coincidences as a function of t is counted at fixed angle. The rotation of the angular correlation pattern appears on the exponential decay curve[4] /fig.3./.

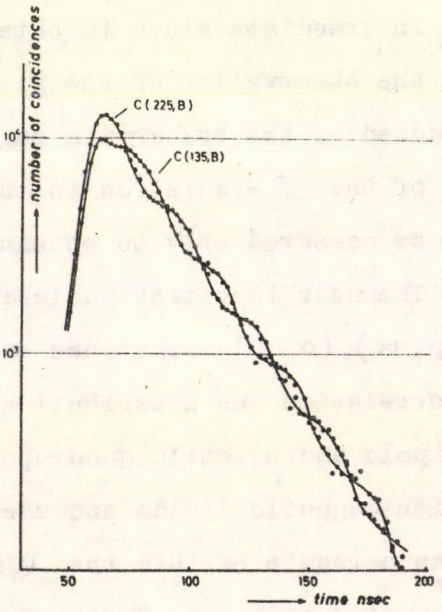

Fig.3.
The effect of the rotation of angular correlation pattern on the decay curve /differential/

From this curve the value of ω can be easily determined. The method is limited by the technique of time measurement to the states of lifetime between $10^{-9} < \tau < 10^{-7}$ sec.

The integral method is used for excited states of $\tau < 10^{-9}$ sec. In this case the integrated number of coincidences is measured. A simple function can be obtained for the description of this case, if /1/ is integrated in time. It is useful, however to write the correlation function in the form

$$W(\vartheta, H, t) = \sum_{k=0}^{k_{max}} B_k \cos k(\vartheta - \omega t)$$

and to integrate this form, then

$$W(\vartheta, H) = \frac{1}{\tau} \int_0^{\infty} \sum_{k=0}^{k_{max}} B_k \cos k(\vartheta - \omega t) =$$

$$= \sum_{k=0}^{k_{max}} \frac{B_k}{\sqrt{1+(k\omega\tau)^2}} \cos k(\vartheta - \omega\tau).$$

The angular correlation or distribution pattern rotates with an angle of $\omega\tau$ under the influence of the magnetic field. A rotated angular correlation pattern[5] is to be seen in fig.4.

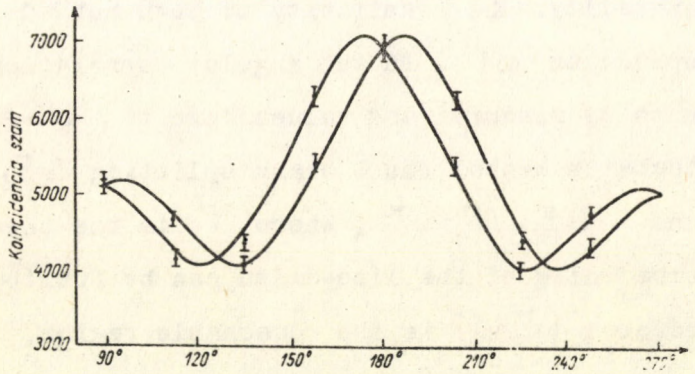

Fig.4.
Rotation of the angular correlation pattern
/integral/

3. Measurement of the internal magnetic field

Angular correlation measurements perturbed by internal magnetic field were performed first with the aim to obtain data on nuclear g-factors. In the earliest experiments of this kind[6,7] the field at Fe nuclei in Fe was used to produce angular rotation. The first experiment to observe the effect of the magnetic field at impurity nuclei in Fe was performed by Keszthelyi et al.[8] . Following these first steps many other groups started to use the internal magnetic field at impurity nuclei to measure g-factors. The main advantage of this method is the rather high magnetic field which allows to measure g-factor of states which are of very short lifetime, 10^{-11}-10^{-12} sec. In these cases the integrated method is used, the internal magnetic field is oriented with a small external electromagnet perpendicularly upwards and downwards to the plane of correlation.

The other feature of interest in this type of investigations is the study of the internal field, its systematics according to the impurity and host atoms, its dependence on the concentration of impurities, on the temperature etc.

These studies are similar to those made with the Mössbauer effect. The angular correlation method surpasses, however, the Mössbauer method in sensitivity and generality. The sensitivity of both methods in the measurement of H depends on $\omega\tau$. In the angular correlation method it is the value which is to be measured and values down to $\omega\tau \gtrsim 0{,}01$ can be covered. In the Mössbauer method the Zeemann splitting ΔE_z is the measurable quantity and $\Delta E_z/\Gamma = \omega\tau$, where Γ is the theoretical line-width. A small broadening of the line-width can be attributed to very different causes therefore $\omega\tau \approx 1$ is the reasonable region, where the magnetic field is measurable with Mössbauer effect.

Much more nuclei are suitable for perturbed angular correlation experiments than for Mössbauer-effect, and with the new methods such as to excite nuclei with nuclear reaction or with Coulomb-excitation[9,10] or to implant the nuclei to be investigated into ferromagnetic lattices with electromagnetic ion separator[11] practically all of the isotopes can be investigated.

This method is very useful for the study of the temperature dependence of the internal field, since the angular correlation itself doesnot depend on the temperature in contrast with other methods suitable for the study of internal magnetic field /e.g.nuclear polarization[12] , measurement of specific heat[13] and Mössbauer effect/.

We think that the perturbed angular correlation and distribution method is just at the initial stage of its application in solid state physics. In the following we should like to present some interesting new results obtained with this method and to outline some further possibilities of its usefulness.

4. Temperature dependence of the internal magnetic field

The first study on magnetism with the angular correlation method was performed by Caspari et al.[14] . They have measured the temperature dependence of angular correlations in rare earth iron garnets and observed large effects on the asymmetry of the correlation at the Néel point without external orienting field and a temperature dependence of $\omega\tau$ with external orienting field. Their results on ^{152}Sm nuclei from the decay of ^{152}Eu embedded in $3EU_2O_3 \cdot 5Fe_2O_3$ garnet are reproduced in fig.5. and 6.

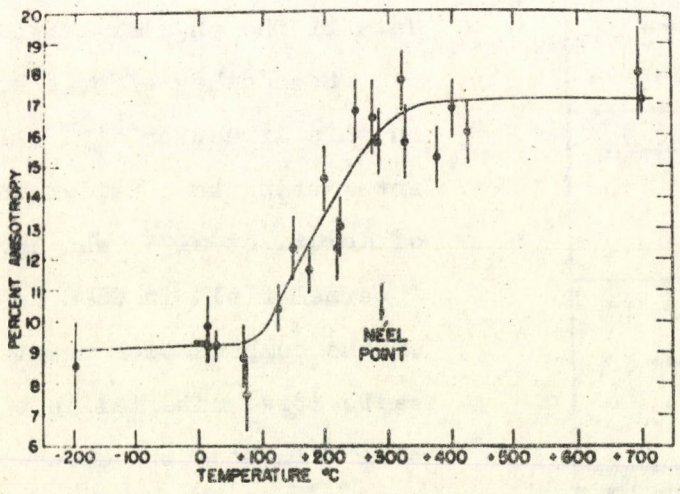

Fig.5.
Change of the anisotropy at the Néel point in Eu IG.

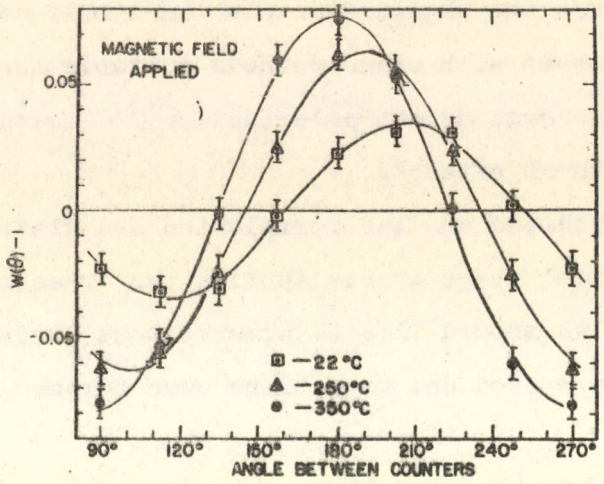

Fig.6.
Rotation of the angular correlation pattern in Eu IG in external orienting field

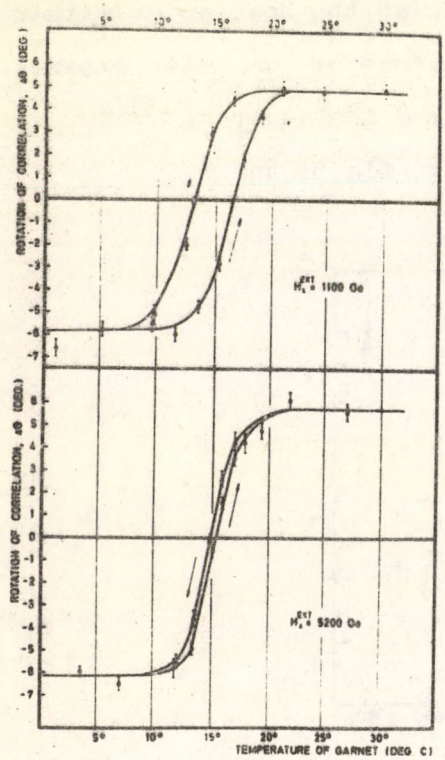

Fig.7.
Rotation angle-vs temperature in the vicinity of compensation point in SdIG.

The most important result of this investigation was the experimental confirmation of a short electronic relaxation time of $\sim 3 10^{-12}$ sec of the rare earth ions in garnets, which was extracted from the anisotropy data of the angular correlation.

Some other efforts have been made in this direction[15,16]. It is interesting to consider the measurement of Koicki et al[17] who investigated the internal field in GdIG in the vicinity of the compensation point. The radioactive material in this garnet was ^{169}Tm from the decay of ^{169}Lu, and the integral rotation of the 178-130 keV cascade was observed. The results are shown in fig.7. It can be seen that the sign of the internal

field changes rapidly at the compensation temperature, while the absolute value of the field remains unchanged. The transition takes place in an $\sim 10^{o}C$ temperature interval and has a hysteris which depends on the external orienting field.

In the case of impurity atoms in ferromagnetic lattices the temperature dependence reflects the effect of local moments observed first at Mn atoms in Fe by NMR method[18] and by neutron scattering[19] . The first, tentative experiment in this direction was performed in our laboratory[20] , in which the temperature dependence of the magnetic field at Pt nuclei in Fe was measured at three poihts /fig.8./.

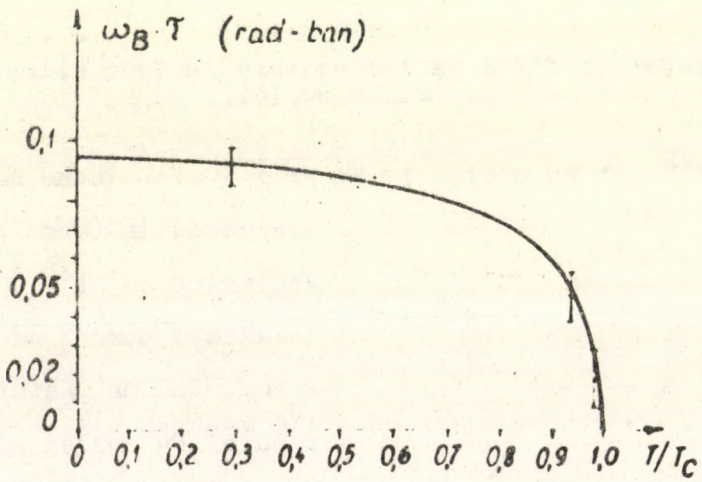

Fig.8.
$\omega\tau$ vs temperature in Fe-Pt alloy at Pt nuclei

From this it could be established only that the field coincides with the values expected from the magnetization curve at high temperatures too close to the curie point. The necessity of accurate measurement was not apparent at this time.

In the case of Pd in Fe, Johansson et al[21] found a small dip in field values at Pd nuclei /fig.9./, a much greater effect was observed

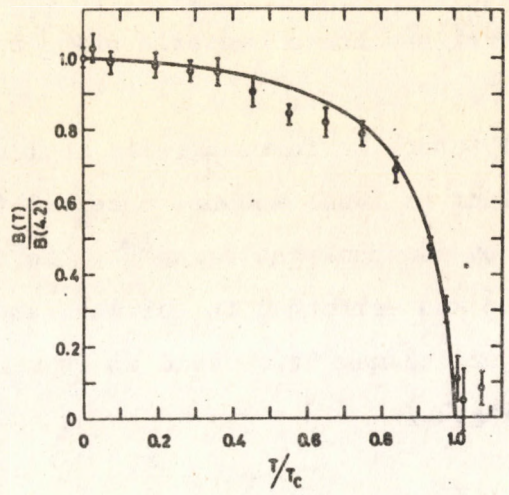

Fig.9.
Internal field vs temperature in FePd alloy at Pt nuclei

by Shirley et al[22] at Ru nuclei in Ni /fig.10./. These measurements were evaluated in terms of the theory of Jaccarino et al[18], i.e. there is localized moment at the inpurity atoms. In the interpretation of the case of Ru nuclei also the nonlocal conduction-electron polarization had to be taken into account.

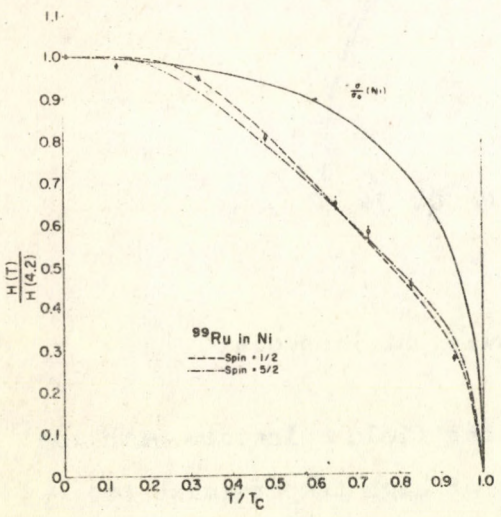

Fig.10.
Internal field vs temperature in NiRn alloy at Rn nuclei

In a recent measurement Deutch et al[23] implanted radioactive ^{169}Yb nuclei into iron foil with isotope separator producing thus very small concentrations of impurity atoms in the lattice.

They measured the temperature dependence of the anisotropy and rotation of the angular correlation in ^{169}Tm after the β -decay and found similar results to those

obtained from the garnets mentioned above. The electronic relaxation time was found to be 2.10^{-12} sec, and the curve for internal magnetic field at Tm nuclei vs temperature showed the characteristic effects of localized moments /fig.11./.

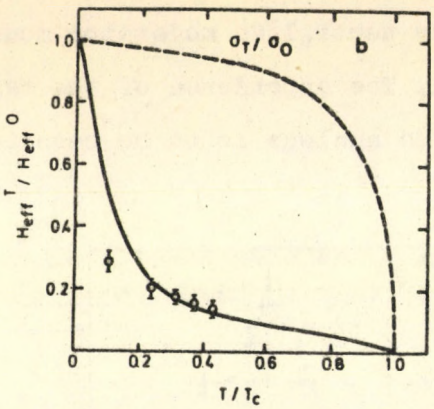

Fig.11.
Internal field vs temperature at Tm nuclei implanted into Fe

4. Time dependence of the internal magnetic field

The large scale of lifetimes for different excited states opens an extended sphere for the research of the time dependence of the hyperfine field. These studies have been made up to now with the use of Coulomb--excitation. In the experiments of Grodzins et al[24,25] different excited nuclei were recoiled with energy of ≈ 10-20 MeV into Fe lattice by the bombarding ^{16}O ions. The excited states had lifetimes of 10^{-11}-10^{-12} sec.

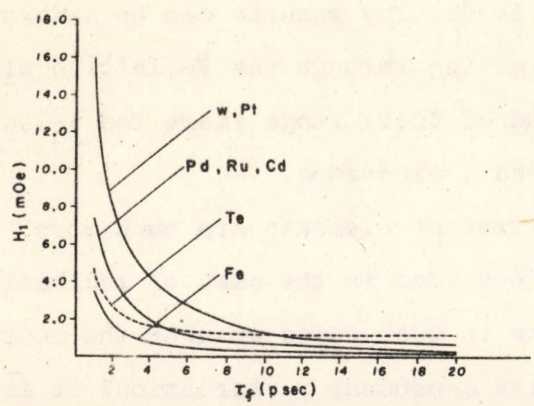

Fig.12.
Time and atomic number dependence of the internal field at nuclei recoiled into Fe lattice by high energy O ions

It turned out from the analysis of the data that the internal field depends on the lifetime of the excited states and on the atomic number of the impurity atoms /see fig.12./. The slowing down time of the recoiled ions was

≈ 1 psec, the field at the nuclei reached in this short time interval very high values as compared with the static field.

To understand the phenomena, the rotation of the angular correlation patterns was measured as a function of the velocity of the ions recoiled into the Fe lattice. The velocity of the ions was varied by changing the thickness of the magnetically neutral Cu moderator mounted between the Fe foil and the target material. The dependence of the value of $\omega\tau$ on the Cu thickness in the case of ^{116}Cd nucleus is to be seen in Fig.13.

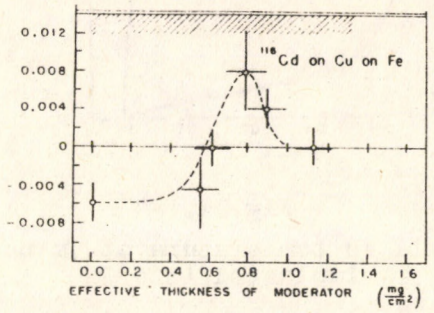

Fig.13.
Dependence of $\omega\tau$ on the thickness of Cu between Fe and target

The value of $\omega\tau$ obtained from static experiment, e.g. from the rotation of angular correlation pattern, is positive, the value got from Coulomb--excitation is at first negative, then approaches the static value and drops to zero. This shows that the time dependent internal field disappears at a definite velocity $\omega\tau$ approaches the static value and there is naturally, no rotation if the recoiled ions stop in Cu. The results can be understood if one assumes that the recoiled ions moving through the Fe lattice pick up polarized electrons close to the end of their range where the velocity is egual to the velocity of the polarized d electrons.

The possibilities to study the different elements are much more extended in the case of nuclear reactions than in the case of radioactivity. Therefore, it is very important to know in such cases whether the measured field is static, or subject to some time dependent perturbation? It is important to verify the absence of time-effects if the velocity of the recoiled ions is smaller than that of d electrons. Our group has made some

experiments in this direction[26] .

The limiting energy of the recoiled ions according to the estimation of Grodzins[24] is ~ 350 keV /Fe ions moving through Fe/ and depends linearly on mass number. We have made experiments with V and Pt atoms alloyed with Fe and bombarded them with 2,5 MeV protons. The recoil energies are smaller than the limiting values / ~ 200 keV in the case of V in Fe and ~ 50 keV in the case of Pt in Fe/.

In the case of V in Fe, alloys of 1,2 and 4,5 % V content were studied, the internal field was found to be within the experimental error independent of concentration /see fig.14./,

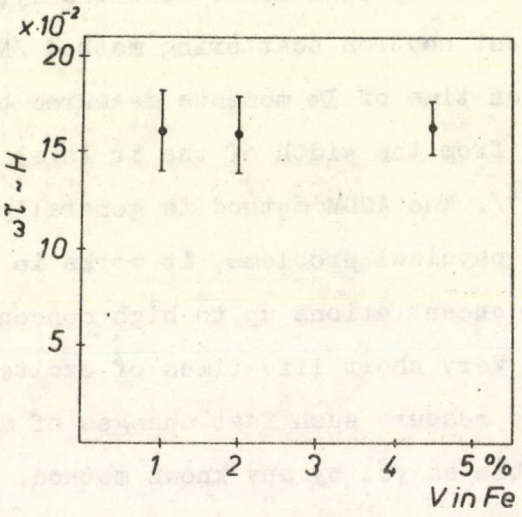

Fig.14.
Dependence of $\omega\tau$ on the V concentration in FeV alloy

and agreeing with the static value[28] .

The field values at Pt nuclei were studied in a $Fe_{0,73}$ $Pt_{0,27}$ alloy. The field was found to be about 20 % lower than the static field in dilute / < 1 % Pt content/ alloy. This decrease can be the consequence of the higher Pt content and of the structure of the material. The situation is not very clear in this alloy. Different authors obtained different field values. The value of Benczer-Koller et al. agrees with our value[29] , that of Buyrn et al is ~ 20 % higher. The magnetization curve measured on our sample showed an abrupt drop at 400 °C on heating and became again magnetic

at 80 °C on Cooling. It seems to be worth while to investigate this alloy more thoroughly.

6. Discussion

The above discussed investigations are only representative examples to show the efficiency of the method. We have seen that this technique has given - among others - new information about the localized magnetic moments and the time dependence of the hyperfine field. In this part we should like to discuss the further advantages of the method.

a/ Localized moments: the angular correlation and distribution method /ACDM/ permits to evaluated the internal field at the impurity nuclei and the relaxation time of the localized moment. Essentially, the same data are obtainable by the incoherent neutron scattering method /NSM/, /for example, the 2.10^{-12} psec relaxation time of Tm moments measured by ACDM agrees well with the value calculated from the width of the Er level determined by NSM $\tau = 1{,}9.10^{-12}$ sec[31]/. The ACDM method is generally not limited by concentration and neutron physical problems, it works in principle at any concentration from trace concentrations up to high concentrations.

b/ Time effects: the very short life-times of excited nuclear states /10^{-11}-10^{-12} sec/ allow to measure such fast changes of magnetic fields which have been inaccessible as yet by any known method. The longer lifetimes /10^{-8}-10 sec/ open a new region of relaxation measurements similar to that covered by NMR methods. This region is far from being explored by ACDM ℜ, since only a few experiments have been performed to date[32,33]. This region is well suitable for work with particle accelerators.

c/Transition - effects: with appropriately chosen nuclear reactions it would be possible to investigate the build-up of magnetic fields at the boundaries of magnetic material. If the γ -radiation originates from a sharp resonance having a width of some eV, then by increasing the energy of the bombarding particles it can be observed that the reaction takes places first at the surface, and then it is going deeper and deeper in to the material. The measured $\omega\tau$ value reveals the mode of transition from zero field to full field values. Also the effects of domain walls can be studied by this method. The resolution of the method depends on the stability of accelerators. The best resolution values can be 10^{-5}-10^{-6} cm thiknesses.

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