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SIGN OF THE ELECTRIC-FIELD GRADIENT AT ^{111}CD IN ZR AND SB

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The β-γ TDPAC technique was applied to ^{111}Ag implanted in Zr and Sb metal single crystals in order to determine sign and magnitude of the quadrupole interaction at the site of ^{111}Cd in these metals. An analysis of the data taken at 293K yielded $\nu_Q = +15.4(6)$ MHz for ^{111}Cd in Zr and $\nu_Q = -107.5(20)$ MHz for ^{111}Cd in Sb. From these values electric field gradients of $+7.3(8)\times10^{16}$ V/cm^2 and $-5.56(62)\times10^{17}$ V/cm^2 for Cd in Zr and Sb are derived respectively.

1. Introduction

Since Raghavan et al. /1/ discovered the "universal" correlation between the electronic and the ionic contribution to the electric field gradient (EFG) at impurity sites in metals a large number of EFG sign measurements were aimed at a check of this correlation. Recently it was found not to hold for a number of pure systems which are members of the group IIIb and IVb transition metals and a modified correlation was suggested by Ernst et al. /2/ Further measurements of the sign of the EFG, which is the most con- convincing test of these correlations, seemed therefore appropriate especially in transition metal hosts. Similarly Haas and Mennigen /3/ developed a model to explain the probe atom dependence of the EFG in the group V s-p metals. Predictions were made concerning the sign of the EFG expected for a number of impurities e.g. in Sb and an experimental test is certainly interesting.

2. Experimental Details

A suitable method to determine the sign of the electric quadrupole interaction (QI) is the β-γ TDPAC technique. It exploits the parity non-conservation of the β decay to obtain polarized nuclear states and the sign of the QI can then be determined from the time dependent anisotropy of the γ-radiation depopulating this nuclear state. A necessary condition is a single crystalline environment of the probe nuclei in order to ensure a unique direction of the principal component of the EFG tensor interacting with the quadrupole moment Q of the nuclear state.

As a probe we used the well known 247 keV, 5/2+ state of ^{111}Cd populated by the β-decay of ^{111}Ag. The parent radioactivity was produced by thermal neutron irradiation of isotopically enriched ^{110}Pd over a $^{110}\mathrm{Pd}(n,\gamma)^{111}\mathrm{Pd} + {}^{111m}\mathrm{Pd} \rightarrow {}^{111}\mathrm{Ag}$ reaction. Subsequently ^{111}Ag was implanted at room temperature into single crystals of Zr and Sb. The implantation energy was 80 keV and the dose typically

+ Present adress : KWU , Offenbach , FRG

1×10^{14} atoms/cm².

For the measurement we used the same geometry as described by Raghavan et al. /4/ with the $\hat{c}$-axis of Zr and the three-fold rotational symmetry axis of Sb in the plane of the movable γ-detector at 45° with respect to the two positions of the γ-detector. A plastic scintillator was used to detect the β-particles populating the 84 nsec isomeric state in ^{111}Cd and the depopulating 247 keV γ-rays were detected by a NaJ(Tl) scintillator. Conventional fast-slow electronics setup was used.

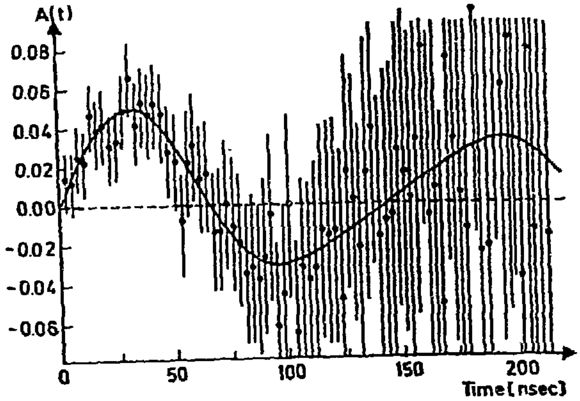

Fig. 1. β-γ asymmetry for ^{111}Ag in Zr at 293 K.

3. Results

The time spectra $N(\theta,t)$ measured for the two detector positions $\theta = +45°$ and $-45°$ were corrected for random coincidences and then used for the formation of conventional asymmetry ratios $R(t)$. The result of such a measurement for ^{111}Cd in Zr at 293K is shown in fig 1. Due to the weak population of the isomeric level in ^{111}Cd by the β-decay of ^{111}Ag (app.1%) the obtainable statistical accuracy was only limited. A least quares fit of the function

$$A(t) = a\cdot\sin\frac{6\pi}{10}\,\nu_Q t + b\cdot\sin\frac{9\pi}{10}\,\nu_Q t \qquad (1)$$

with a, b and ν_Q as free parameters led to the result shown by the solid line in fig. 1.a and 1.b in eq. 1 are amplitude factors whose signs are known from the β-γ TDPAC theory and for ν_Q, the quadrupole interaction frequency a value of +15.4(6) MHz was obtained. The magnitude of this result is in reasonable agreement with that of a γ-γ TDPAC measurement of Kaufmann /5/ yielding $|\nu_Q| = 14.1(2)$ MHz. Since $\nu_Q = e^2Qq/h$ we can now use the known quadrupole moment of the 247 keV state $Q = +0.80(9)$b /6/ to calculate the EFG for Cd in Zr to be $eq^{exp} = +7.9(15)\times10^{16}$ V/cm². To allow a comparison to the above mentioned correlation, this value has to be decomposed in the ionic and the electronic contribution to the total EFG. The ionic EFG is known to be $eq^{ion} = +11.5\times10^{16}$ V/cm² /4/. Thus one obtains

$$eq^{el} = eq^{exp} - eq^{ion} = -\ 3.6(7)\times10^{16}\ \text{V/cm}^2$$

or using the more precise value of eq from Kaufmann /5/

$$eq^{el} = -\ 4.2(5)\times10^{16}\ \text{V/cm}^2$$

This result places the system in a very odd position relative to the two existing systematic trends found for the relation between eq^{el} and eq^{ion} in metals /1,2/. As far as the relative sign is concerned it agrees with the "universal" correlation of Raghavan et al. /1/ since eq^{el} has an opposite sign to eq^{ion}. However according to this systematic one would expect eq^{el} to be larger by about a factor of 8.

The second rule suggested by Ernst et al. /2/ says that the sign of eq^{el} is only determined by the electronic structure of the host lattice and should be positive for all IIIb and IVb transition metal hosts. This was found to be true for Cd in the other two IVb metals Ti and Hf /1,7/ but the present result for Cd in Zr is in contradiction to this trend. However it should be pointed out that for such small values of eq^{el} small errors in eq^{ion} caused e.g. by local lattice distortion could probably account for the observed disagreement with this otherwise very successful correlation.

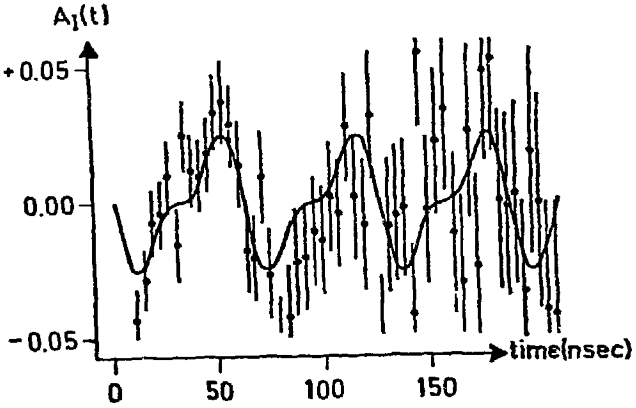

Fig. 2. β-γ asymmetry for ^{111}Ag in Sb at 293 K

The data obtained for the system ^{111}Cd in Sb are shown in fig.2. Here a least squares fit yields $\nu_Q = -107.5(20)$ MHz at roomtemperature. Thus one calculates an EFG $eq^{exp} = -5.56\times10^{17}$ V/cm^2 and using an ionic EFG of $eq^{ion} = -1.55\times10^{17}$ V/cm^2 /8/ we derived an electronic contribution to the total EFG of $eq^{el} = -4.01(62)\times 10^{17}$ V/cm^2. This result is in strong disagreement to the above mentioned "universal" correlation since both contributions to the total EFG have the same sign. Only two systems behaving similarly have been known (InIn and CdIn) till now and have been regarded as pathological exceptions due to special cancellation effects of the different contributions to the total EFG. However recently four more systems have been found to show the same behaviour /9/ and it seems now to be quite common for group III and V s-p metal hosts.

In the last years Haas and Menningen /3/ developed a model specially for the group V metal hosts to explain the variation of the EFG with increasing atomic number of probes from the fourth period. Their model, based on the assumption of purely covalent bonding in As, Sb and Bi, predicts a variation of the EFG depending on the probe atom as shown in fig. 3. Although this model reproduces well the qualitative trend indicated by signs and magnitudes of the EFG for Sn in Sb and pure Sb the present result is in striking contradiction to the model.

This two sign measurements of the EFG for Cd in Zr and Sb presented here do not fit into any of the existing systematics for the EFG. This failure illustrates again the difficulties encountered in the formidable task to develop a comprehensive theory of the EFG in noncubic metals. Additional sign measurements seem to be necessary specially in the group V s-p metals before the observed results can be explained.

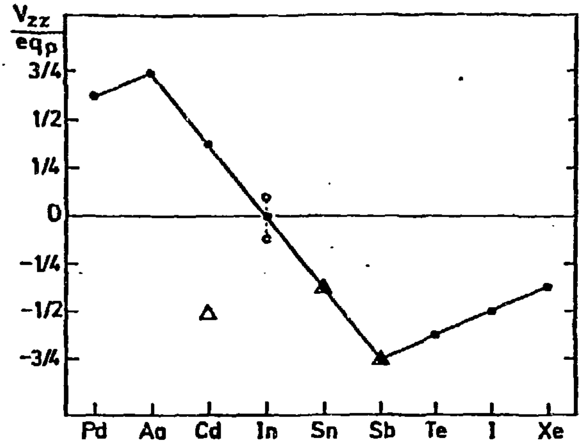

Fig. 3 Trend of the EFG at impurities in Sb in units of the valence p-electron gradient eq_p. The open triangles are experimental EFG values normalized to the EFG for Sb. For In only the EFG magnitude is known and the two possible values are indicated. Taken from Ref. /3/.

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