The Electric Field Gradient and its Temperature Dependence at ^{111}Cd in α – Uranium

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The magnitude and temperature dependence of the quadrupole interaction at the ^{111}Cd site in orthorhombic α – Uranium was investigated between 293 and 17 K. The parent activity ^{111}In was implanted into Uranium metal with an energy of 80 keV and the γ–γ TDPAC technique, applied to the 245 keV state in ^{111}Cd, was used to measure the quadrupole interaction frequency. The derived electric field gradient for Cd in Uranium was found to be highly asymmetric ($\eta \approx 1$) and led to a quadrupole interaction frequency of ν_Q = 7.10(7) MHz at 293 K. The temperature dependence of the quadrupole interaction is very strong, ν_Q increases to 14.3(2) MHz at 17 K and shows a linear dependence on the temperature.

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1. Introduction

Our experimental knowledge of electric field gradients (EFG) in non-cubic metals is quite complete by now /1/. Considering the temperature dependence of the EFG it has been found that it varied in the majority of cases according to the so called $T^{3/2}$ rule /2/. In substitutional systems the major exception to this rule occurs in the rare earth hosts where the EFG e.g. at the site of a substitutional In impurity varies linearly with temperature /3/. It is therefore interesting to study the EFG and its temperature dependence for In in the similar Actinide hosts to see if this anomalous temperature dependence can also be observed there. The only Actinide element which seems suitable at present for such an experiment is α – Uranium since it is sufficiently stable and has a non-cubic crystal structure. To the knowledge of the authors the only other measurement of an EFG in an Actinide host has been carried out by Friedt et al. /4/ for ^{231}Pa in a Protactinium host at 4.2 K.

2. Experimental Details

The nuclear quadrupole interaction (QI) of ^{111}In was observed by applying the γ–γ–time differential perturbed angular correlation technique (TDPAC) to the 173 – 247 keV γ–γ cascade populated in the decay of ^{111}In to ^{111}Cd. The intermediate 247 keV, 5/2 state of ^{111}Cd has a half-life of 84 nsec and a quadrupole moment of Q = 0.83 b /5/. The perturbation pattern described by the QI-frequency $\nu_Q = eQV_{zz}/h$ and the asymmetry parameter $\eta = (V_{xx}-V_{yy})/V_{zz}$ was observed with a conventional 2 CsF detector set-up where time spectra were recorded successively for interdetector angles of 90° and 180°.

The ^{111}In probe atoms were implanted with an energy of 80 keV and a dose of typically 10^{14} at/cm^2 into a 200 μm thick Uranium foil kept at room temperature. The mean range of ^{111}In ions of this energy in Uranium was estimated to be 130 Å.

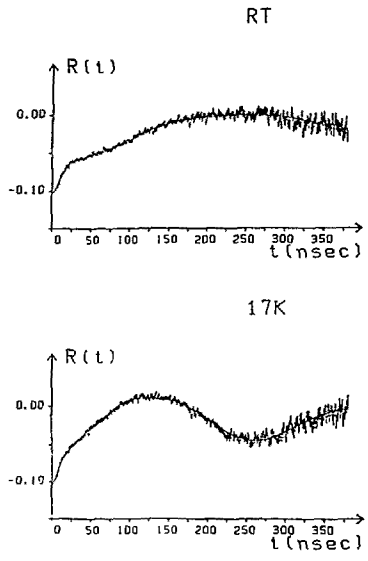

Fig. 1: TDPAC spectra of ^{111}Cd in α-Uranium at 293 and 17 K. The solid lines represent fits to the experimental data.

Prior to the implantation the Uranium foil was electrochemically polished and was kept during the measurements under a vacuum of better than 10^{-6} kPa in order to avoid surface oxidation. For measurements below room temperature the sample was cooled by means of a closed cycle helium refrigerator to various temperatures between 17 and 273 K.

3. Results and Discussion

From the time spectra $N(\Theta,t)$ recorded under $\Theta = 90°$ and $180°$ the usual asymmetry ratios $R(t) = 2(N(180°,t) - N(90°,t))/(N(180°,t) + 2\cdot N(90°,t))$ were formed. The results obtained at 293 K immediately after the implantation is shown in the upper part of Fig. 1. After a fast drop of the anisotropy within the first 25 nsec due to large nonunique EFGs produced by lattice damage caused by the heavy ion implantation one observes only a slow variation of the anisotropy making a straightforward interpretation of this result difficult. When the same sample was cooled to 17 K a dramatic increase in the interaction frequency occurs and the anisotropy of the slowly varying part recovers completely after 260 nsec, i.e. a full period of the perturbation pattern can be observed (Fig. 1). The shape of this curve is typical for an axially asymmetric EFG with an $\eta = 1$ in a polycrystalline sample. However, a similar pattern could be caused by a preferential orientation of the crystallites (texture), which is well known to occur in Uranium foils produced by a rolling process, together with a geometrical misalignment of the sample during the measurement.

In order to exclude the latter possibility an additional experiment was carried out, where the sample was rotated during the TDPAC measurement around several axes thus averaging out possible texture in the sample. Since in this experiment no significant change in the interaction pattern could be observed, its shape had to be attributed to a large asymmetry parameter of the EFG interacting with the ^{111}Cd probe. Under these conditions a least squares fit to the experimental data gave for 60 % of the ^{111}Cd probe nuclei a quadrupole interaction frequency of $\nu_Q = 7.10(7)$ MHz, $\eta = 1$ at 293 K which increased at 17 K to $\nu_Q = 14.3(2)$ MHz with η remaining 1. From this value an effective EFG of $|V_{zz}^{eff}| = 0.35 \times 10^{17}$ V/cm^2 with $\eta = 1$ can be derived.

For an interpretation of these results one has to make an assumption concerning the site of the probe nucleus in the Uranium lattice after the implantation. However, the mass ratio between the implanted and host atoms makes replacement collisions rather probable. Further it is known that ^{111}In and ^{181}Hf occupy substitutional sites in the similar rare earth metals after melting and implantation /3/ and a low solubility of In in α-Uranium is not excluded /6/. Therefore in the following discussion a substitutional site for implanted ^{111}In in α-Uranium will be assumed.

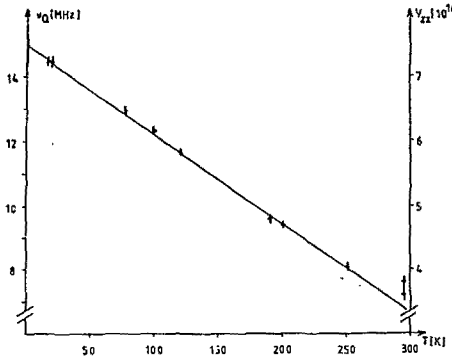

Fig. 2: Temperature dependence of the quadrupole interaction frequency for ^{111}Cd in Uranium

We can then use the point charge model /7/ to carry out a lattice sum calculation for the substitutional site in the orthorhombic α-Uranium. For the lattice constants measured at 293 K /8/ and a charge of Z = +6 for the Uranium ions one obtains V_{zz}^{ion} = -6.27 x 10^{17} V/cm^2 with η = 0.357. Here the enhancement by the Sternheimer antishielding factor for Cd $(1-\gamma_\infty)$ = 30.3 has already been taken into account.

This result is quite in disagreement with previously found systematics where e.g. the ionic EFG was usually found to be much lower than the effective EFG. Also the values one would obtain for the electronic contributions to the total EFG is too large by a factor of 5-6 according to a systematic suggested by Raghavan et al. /9/. A rule found by Forker et al. /3/ for the ^{111}Cd probe in the rare earth metals relating the effective to the ionic EFG by a so called enhancement factor $\alpha = V_{zz}^{eff}/V_{zz}^{ion}$ comes out unusually *small α =0.06 for the CdU system.*

These discrepancies and also the high value found for the asymmetry parameter η could be due to a lattice deformation caused by the impurity character of the ^{111}Cd probe in the Uranium lattice. Such a deformation or even a Jahn-Teller like permanent displacement of the probe atom from a regular lattice site could lead to much larger asymmetry parameters η and also to an increase of the EFG magnitude.

The large temperature dependence of the EFG (Fig. 2), an increase by about a factor of two is observed between 293 K and 17 K, is also in contrast to the moderate variation of the EFG in the rare earth metals /3/. The reason cannot be found in an unusual variation of the lattice parameter since a point charge calculation using measured lattice constants /8/ gives only a change of less than 10 % in V_{zz}^{ion} in this temperature range. *Possibly a local lattice deformation as* mentioned above allows large vibration amplitudes of the probe atom thus producing the strong EFG variation with temperature. Similarly in recent measurements of the EFG at ^{111}Cd in the light rare earth metals Praseodymium and Neodymium /10/ a smaller but still remarkably strong temperature dependence has been found.

The variation of the EFG with temperature can only be described by a linear function and not by the above mentioned $T^{3/2}$ rule. This is in agreement with the results found in the other rare earth hosts /3/. An explanation for this behaviour of the 4f and now also the 5f metals is still lacking.

4. References

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