

# TDPAC study of $^{181}\text{Ta}$ in $\text{HfSe}_2$

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**Abstract** Time differential perturbation correlation study of hexagonal hafnium selenide structure were performed with tantalum as probe. Efforts were made to find electric field gradient and interaction frequency with different parameters. For this the experimental data were least square fitted to investigate the results.

**Keywords** —TDPAC, EFG, Interaction Frequency,  $\text{HfSe}_2$ ,  $^{181}\text{Ta}$ ,

## I. INTRODUCTION

$^{181}\text{Ta}$  has been used extensively to study the electric field gradient and nuclear quadrupole interaction (NQI) in a variety of compounds, particularly in Hf compounds, Darriba et al 2018, Butz et al, 2009.

Electric field gradient (EFG) measured at a probe nucleus is very sensitive to the surrounding charge distributions. TDPAC technique is an ideal one, to investigate EFG, induced by defects in cubic host lattices and in various other hosts, especially, hcp lattice (Horst et al, 1978, Pleiter et al, 1978, Heubes et al, 1978, Echt et al, 1978, Deicher et al, 1981, and Bohn et al, 1978).

In the present study the  $\gamma$ - $\gamma$  angular correlation and its perturbation due to hyperfine interactions of nuclear quadrupole moments with the extra-nuclear fields has been studied. Important nuclear properties like energy, total angular momentum, parity, magnetic moment and electric quadrupole moment are associated with ground state and/or excited state of the nucleus. Mössbauer effect and Time Differential Perturbed Angular Correlation Measurements (TDPAC) are very popular to measure hyperfine interaction.

In practice, the quadrupole interaction frequency  $\nu_Q$  is defined as  $\nu_Q = \frac{eQV_{zz}}{h}$  and usual parameterization of the field gradient at the nucleus is given in the form as  $V_{zz} = (1 - \gamma_\infty)V_{zz}^{latt} + (1 - R)V_{zz}^{local}$ . For ions at other lattice points, the Sternheimer factor is denoted by  $\gamma(\infty)$ . But in case the field sources are the probe shells this factor will be less than 1.0. Thus a distance dependent Sternheimer factor  $\gamma(r)$  has to be used.

This radial dependence of Sternheimer factor is discussed in detail by Foley et al (1954) and Hygh et al (1966). A complete tabulation of Sternheimer Antishielding values has been given by Feiock and Johnson (1969).

## II. EXPERIMENTAL SETUP

In the present study we used time differential perturbation angular correlation system with two counter setup with XP2020Q photomultiplier tubes coupled with

$\text{BaF}_2$  scintillator crystals. The time resolution of the system was 0.6ns. The asymmetry parameters were found using usual asymmetry ratio function defined as  $R(t) = \frac{2[C(180^\circ, t) - C(90^\circ, t)]}{C(180^\circ, t) + 2C(90^\circ, t)}$  as given by Arends et al, 1980. The experimental data for coincidence at  $90^\circ$  and  $180^\circ$  was directly captured in computer and from that  $R(t)$  was found and then least square curve fitted to theoretical function  $Y = A_0 + A_1(t) + A_{22}G_{22}(t)$ .

## III. RESULT AND DISCUSSION

Hafnium Selenide were procured from CERAC, USA. This material was then sent to Bhabha Atomic Research Centre for irradiation. Neutron capture reaction takes place in Hf and it decays to  $^{181}\text{Ta}$  via  $\beta^-$  decay.

133 – 482 keV  $\gamma$ - $\gamma$  cascade from this source was used for the experiment. Se is group VIA elements with most probable oxidation state of 4. Electronegativity of Selenium (2.4) is much higher than hafnium (1.3), and so, we can assume that in a molecule or bonding with Hafnium, which has a strong tendency of giving its four electrons, Selenium will accept two electrons from Hafnium and will have a valency of -2.

$\text{HfSe}_2$  has a hexagonal structure with  $a = 3.748\text{\AA}$  and  $c = 6.159\text{\AA}$  and structure resembling  $\text{CdI}_2$ . Structure of the  $\text{HfSe}_2$  has selenium at  $1/3, 2/3, 1/4$  and at  $1/3, 2/3, -1/4$  positions with reference to Hf at  $0,0,0$ . Figure 1 shows the structure of this type of symmetries.

Figures 2 and 3 shows the TDPAC spectrum of  $^{181}\text{Ta}$  in  $\text{HfSe}_2$ . The solid line shows the computer fit of the experimental data. Data were least square fitted to the function of the form

$$Y_{fit} = A_0 + A_1(t) + A_{22}G_{22}(t)$$

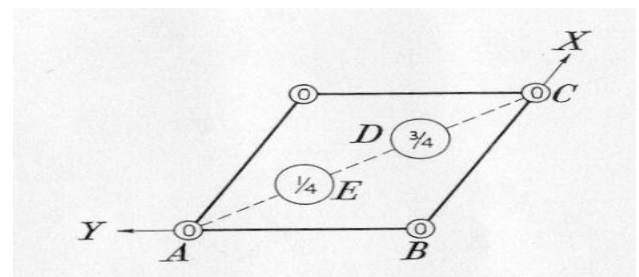


Figure 1:  $\text{CdI}_2$  type structure

To get the theoretical values of the interaction frequency and electric field gradient in this hexagonal material we calculated the ionic character of the  $HfSe_2$ , by calculating the difference in the electronegativities of Hf and Se ions using periodic table of Sargent- Welch. The percent ionic character then can be obtained by the relation given by Pauling (1960) or from the table given therein.

For this particular case of  $HfSe_2$ , the electronegativity of Hf is 1.3 and that of Se is 2.4 and thus, the difference of electronegativity equals 1.1. At this difference the ionic character of  $HfSe_2$  would be 26 percent. So the  $Z_{eff}$  in  $HfSe_2$  lattice at the site of Hf can be given by

$$\left(\frac{26}{100}\right) \times 4 = 1.04$$

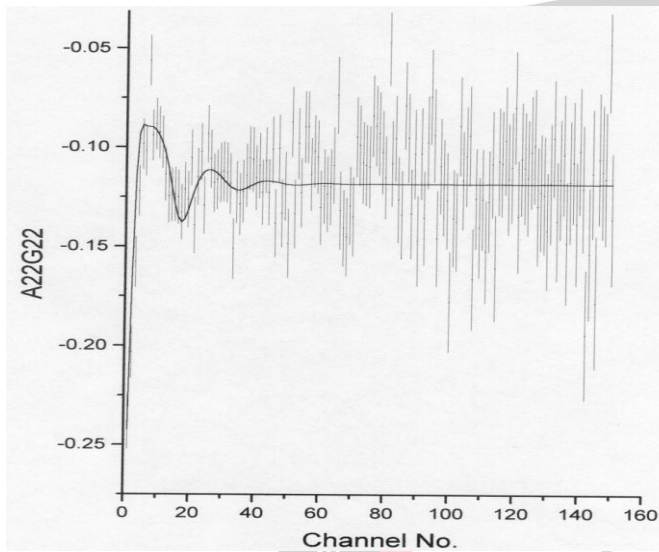


Figure 2: Fitted spectrum of  $^{181}Ta$  in  $HfSe_2$  ( $\eta=0, \chi^2=1.69$ )

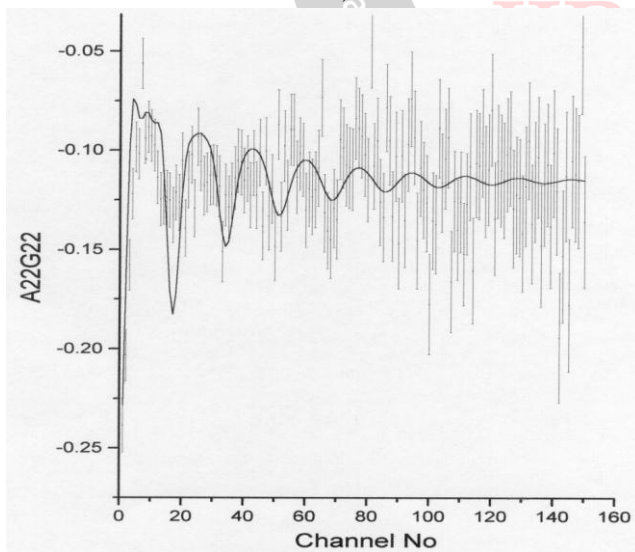


Figure 3: TDPAC spectrum of  $^{181}Ta$  in  $HfSe_2$  ( $\eta=0, \chi^2=1.06$ )

Now the  $V_{zz}^{latt}$  can be obtained as (Das and Pomerantz, 1961),

$$V_{zz}^{latt} = \frac{1}{4\pi\epsilon_o} \left[ 0.0065 - 4.3584 \left( \frac{c}{a} - 1.6333 \right) \right] \frac{eZ_{eff}}{a^3}$$

$$V_{zz}^{latt} = -1.0517 \times 10^{18} V/m^2$$

and taking sternheimer antishielding factor = -61, the effective field gradient is calculated to be

$$V_{zz}^{latt\ eff} = (1 - \gamma_\infty) V_{zz}^{latt} = -6.52 \times 10^{19} V/m^2$$

The least square fitted parameters gave the value of field gradient as  $3.35 \times 10^{22} V/m^2$  and quadrupole interaction frequency  $\nu_Q = 2033MHz$  with asymmetry parameter taken as  $\eta=0$ . We tried to fit the data with various values of  $\eta$  varying from 0 to 1 but could not get a proper fit.

The experimental data shows an almost undamped perturbation, after initial damping at the first cycle, shows that there may be a relaxation parameter involved in the frequency. However, our efforts to fit the data with relaxation parameter  $\lambda$ , could not get a proper fit. Other method to fit the data may be a combination of the fast and slow frequencies.

Our results shows reasonable fit with these two frequencies. Figures 4 and 5 shows the two frequency fit of the data. Different parameters fitted are given in the following table 1. Two frequencies were fitted with a function of the type

$$Y_{fit} = cons + A_{21}G_{21}(t) + A_{22}G_{22}(t)$$

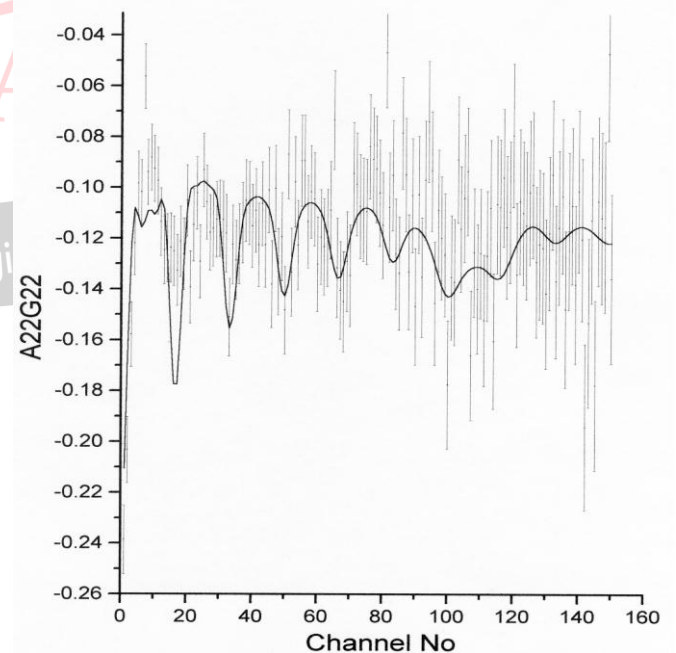


Figure 4: Two frequency fit of the spectrum of  $^{181}Ta$  in  $HfSe_2$  ( $\eta=0, \chi^2=1.54$ )

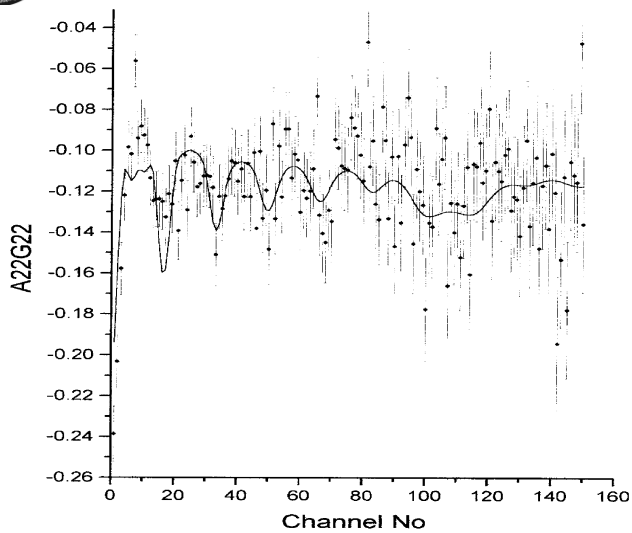


Figure 5: Two frequency fit of the spectrum of  $^{181}\text{Ta}$  in  $\text{HfSe}_2$  ( $\eta=0$ ,  $\chi^2 = 1.40$ ) (1ch = 0.2ns)

#### IV. CONCLUSION

The fraction  $f_0$  contributes only a constant background to the perturbation factor. It may be thought of as caused by those probes, which are positioned such that EFG at these sites is approximately equal to zero. The  $f_1$  fraction sees a unique interaction frequency  $\nu_{Q1} = 2103\text{MHz}$  with  $\eta_1 = 0.0$ , which may be attributed to the interaction of the nearest neighbours of the Hexagonal  $\text{HfSe}_2$ . The fraction  $f_2$  sees another frequency  $\nu_{Q2} = 323\text{MHz}$  with  $\eta = 0.0$ . From the Table 2, it is clear that about 44 percent of the probe atom are not seeing any perturbation and are contributing to the constant background only. 47 percent of the total probes are contributing to the very high frequency with 6 percent damping or spread in the frequency. Rest of the

probes, 9 percent, are such probes, which are not at the nearest neighbours distance and are producing almost undamped contribution with interaction frequency 323MHz and a spread of 1.4 percent in the frequency. A very large difference in the calculated and experimentally obtained values may be, because of the fact that, we haven't calculated the EFG because of the Se atoms and the effect it may have on the Hf atoms and Ta probe.

Table 1

$A_{21}$	$\nu_{Q1}(\text{MHz})$	$\eta_1$	$A_{22}$	$\nu_{Q2}(\text{MHz})$	$\eta_2$	$\chi^2$
-0.19(1)	2033(33)	0.0	-	-	-	1.69
-0.22(1)	1978(21)	0.0	-	-	-	1.06
-0.12(16)	2100(25)	0.0	-0.021(4)	330(14)	0.0	1.54
-0.10(15)	2103(25)	0.0	-0.019(4)	323(14)	0.0	1.40

The parameters obtained from the fitting procedure of two frequency fit shows that at least three distinct fractions are contributing to the total perturbation factor. The best fit of the data is shown in the figure 5 and its fitted parameters are given in Table 2.

Table 2

$f_0$	$f_1$	$\nu_{Q1}, \text{MHz}$	$\delta\nu_{Q1}, \text{MHz}$	$\eta_1$	$f_2$	$\nu_{Q2}, \text{MHz}$	$\delta\nu_{Q2}, \text{MHz}$	$\eta_2$
0.44(1)	0.47(1)	2103 (25)	133.6 (32)	0	0.09(1)	323(14)	4.6(17)	0

#### REFERENCES

- [1] Arends A. R., C. Hohenemser, F. Pleiter, H. De Waard, L. Chow, and R. M. Suter, 1980, *Hyperfine Interactions* **8**, 191
- [2] Bohn R., J. Christiansen, P. Heubes, R. Kiettel, W. Klinger, W. Sander & W. Witthuhn, 1978, *Hyperfine Interactions* **4**, 758
- [3] Butz T., S. K. Das and Y. Manzhur, 2009, *Z. Naturforsch.* **64a**, 103 – 111
- [4] Darriba G. N., E. L. Muñoz, A. W. Carbonari, and M Rentería, 2018, *Experimental TDPAC and Theoretical DFT Study of Structural, Electronic, and Hyperfine Properties in (111In) 111Cd-Doped SnO2 Semiconductor: Ab Initio Modeling of the Electron-Capture-Decay After-Effects Phenomenon*, *J. phys. Chem.*, **122**(30), pp 17423–17436
- [5] Das T. P. and M. Pomerantz, 1961, *Phys. Rev.* **123**, 2070
- [6] Deicher M., E. Recknagel & Th. Wichert, 1981, *Hyperfine Interactions* **10**, 675.
- [7] Echt O., E. Recknagel, G. Schatz, A. Weidinger & Th. Wichert, 1978, *Hyperfine Interactions* **4**, 585
- [8] Feiock F. D. and W. R. Johnson, 1969, *Phys. Rev.* **187**, 39.