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Study of Tantalum in $HfSi_2$

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Abstract Time differential perturbed angular correlation technique is very popular to investigate electric field gradient that is induced by defects in different hosts and is especially very effective for cubic and hcp lattice. Electric field gradient induced by defects can be resolved by their unique interactions measured by TDPAC. In pure cubic crystal the field gradient vanishes at each lattice point due to the spherical symmetry of charge distribution. When impurity is introduced in this system then the symmetry is disturbed and field gradient arises. In the present study an attempt has been made to investigate effect of radioactive tantalum in $HfSi_2$ using TDPAC technique. The observed electric field gradient was $4.34X10^{21}$ V/m^2 and quadrupole interaction frequency was ω_Q= 263.33 MHz with 10 percent spread and asymmetry parameter η=0.

***Keywords* — Angular correlations, $HfSi_2$, Hyperfine Interactions, Perturbation, ^{181}Ta, TDPAC,**

I. INTRODUCTION

The γ-γ angular correlation study employs a cascade of two gamma radiations emitted in succession. In this type of cascade a nucleus in its excited state say with spin I_i decays by first gamma radiation γ_1 to the intermediate state of spin I. After certain time it decays to final state I_f by emission of the second γ-radiation γ_2.

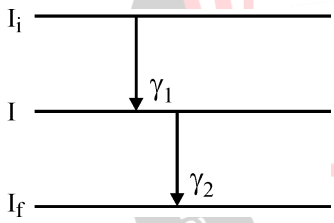

Figure 1 : Gamma-gamma cascade

The probability of emission of radiation by a radioactive nucleus depends in general on the angle between the nuclear spin axis and the direction of emission. Since the nuclei are randomly oriented, the total radiation emitted by radioactive nuclei is isotropic in nature. The emitted radiation will show anisotropic behaviour if they are not randomly oriented and can be observed by placing the radioactive nuclei sample at very low temperature in a strong magnetic or electric field gradient, thereby polarizing or aligning the nuclei and then measuring the angular distribution of the emitted radiation with respect to the direction of the applied field [1-3].

An anisotropic emission can in general be obtained by selecting only those nuclei whose spin happen to lie in a preferred direction. If the nucleus decays through successive emission of two radiations γ_1 and γ_2 then the observation of γ_1 in a fixed direction selects a number of nuclei that are having a nonisotropic distribution of spin orientations [3]. The succeeding second radiation γ_2 then shows a definite angular correlation with respect to that fixed direction. For a nucleus emitting two gamma rays γ_1 & γ_2 in the two directions, the angular correlation is defined as the relative probability $W(\theta)d\Omega$ and γ_2 is emitted into the solid angle $d\Omega$ at an angle θ with respect to the first fixed direction. It was found that the angular correlation function is sensitive to extranuclear field, if the lifetime of the intermediate state is longer than 10^{-10} secs. [3-5].

Unique field gradient are observed because of specific defect configurations and as TDPAC can resolve these unique interactions, one can study the defect distribution around a probe in cubic lattice. In a pure cubic crystal, the EFG vanishes at each lattice point due to the spherical symmetry of charge distribution. In such a lattice, when an impurity atom differing in charge or size or both is introduced, the cubic symmetry of charge distribution is destroyed and an EFG arises due to Spherical valence effect [6-8] and due to indirect and direct size effect [9-12].

In TDPAC studies, the EFG is measured at a radioactive probe-isotope, usually differing from the alloy constituents. In such cases, the impurity potential, modified by different charge transfer mechanism and conduction electron screening, interacts with the charge excess of the probe, leading to preferential distribution of impurities around the probe [13]. This distribution of the impurities around the probe atom depends on the interaction between the probe and impurity, in the particular host lattice. According to Blandin & Deplante if the probe and the impurity have excess opposite charges in comparison to the host lattice, the system will show an attractive interaction.

Jena (1976) and Nishiyama et al (1976) demonstrated that irrespective of the nature of the system, whether pure metals or compounds, the temperature dependence of EFG could be explained by the thermal vibrations of the crystal lattice [14-15].

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II. EXPERIMENTAL SETUP

Detection of coincidence of γ_2 with respect to γ_1 as a function of angle between them is termed as angular correlation. A simple angular correlation measurement involves two detectors detecting γ_1 and γ_2 respectively. Out of these two detectors, the one detecting γ_1, is kept fixed and the other one detecting γ_2, is given an angular sweep to obtain the angular correlation. In this type of experiment it is important to find out if the two gamma rays are from the same nuclear event or from different ones. Energy and time coincidence circuits are used to find out whether γ_1 & γ_2 are correlated or not. In this case, the mean lifetime τ of the intermediate nuclear state is comparable to the 'interaction time' $\hbar/\Delta E$, where ΔE is the energy of interaction between the nucleus and the extranuclear fields. This interaction causes a change in the intermediate state during the time interval between the arrival of the first γ-ray and the emission of the second γ-ray. This is called perturbation and the correlation between two gamma rays in such a case is known as perturbed angular correlation. If the life time is sufficiently high, we can observe its exponential decay as a function of time, then the experiment is termed Time Differential Perturbed Angular Correlation (TDPAC). TDPAC technique relies on perturbed angular correlation of cascade gamma rays emitted by the probe nucleus and is based on Hyperfine interactions technique [16].

We used two XP2020Q photomultiplier tubes coupled with BaF_2 scintillators in a gamma-gamma delayed coincidence system. The delayed detector is moved between 90 degrees and 180 degrees. Energy information is taken from dynode signal while anode signal is used for timing signal. The constant fraction discriminator and single channel analyser were set in such a way that they trigger at the 133keV for start and 482keV for stop gamma ray. The system was calibrated with standard Na-22 and was observed to be 0.6ns.

III. RESULTS AND DISCUSSION

Orthorhombic $HfSi_2$ [17] was procured from CERAC, USA. Lattice parameters of hafnium silicide are a=3.677Å, b=14.550Å, c=3.649Å. This material was sent to BARC for neutron irradiation. The sample was used as a source after annealing at 600^{o}C for 12 hours. The behaviour of silicon, with the transition metals, differs in one important aspect from that of hydrogen, and the elements of the second period, in that, it may play the part of a metalloid. Its solubility in some metals is known to be substitutional and may be quite high.

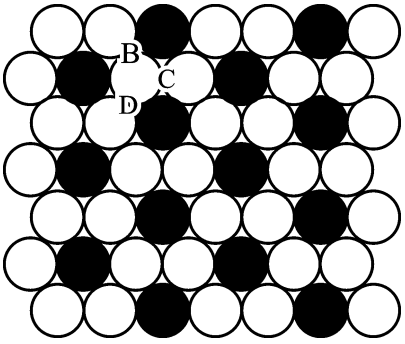

Figure 2: $TiSi_2$ type structure

Most of the disilicides except those of nickel, cobalt, osmium and a few more are related more or less to the $TiSi_2$ structure which can be visualised as contrived from a closed packed layer of composition $TiSi_2$.

These layers are so superimposed as to give triangular arrangements of atoms between layers, atoms being placed above points B, C or D as shown in figure 2. The reason for this must be much stronger bonding between silicon atoms in continuous layers (Si-Si = 2.54Å) than between those in the same layers (Si-Si = 2.75Å) [18]. When the stacking sequence of layers is ABCD, the $TiSi_2$ structure is formed. When it is ABC, the $TaSi_2$ structure results while the sequence AB yields $MoSi_2$ structure. It has been observed that the transition from one kind of stacking to the next one is a function of the valence-electron concentration (v.e.c.) in the phases [19]. Experimental verification of this postulate was made possible, because, many of the disilicides of Groups IV to VI form solid solutions [19-20] and also, it is possible to substitute aluminium for silicon. Theoretical calculation, for the field gradient in orthorhombic $HfSi_2$ could not be done here.

The following Figure 3 shows the TDPAC spectrum of ^{181}Ta in hafnium silicide. Solid line show the computer fit of the data. From the least square fitting of the data, the field gradient found is $4.34X10^{21}V/m^2$ with quadrupole interaction frequency ν_Q= 263.33 MHz with 10 percent spread and asymmetry parameter η=0. The value of A_{22} effective was observed to be 0.19.

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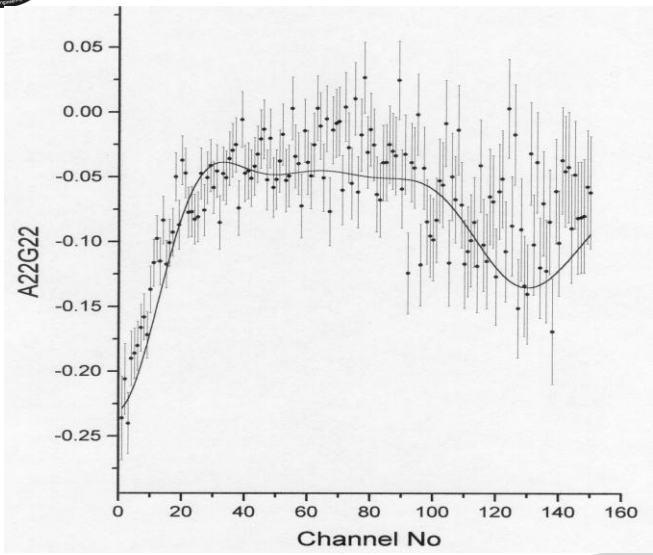

Figure 3: ^{181}Ta in $HfSi_2$

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