

Development of a time-differential perturbed angular correlation spectrometer using LaBr₃(Ce) and BaF₂ detectors

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Introduction

The LaBr₃(Ce), a new generation scintillation detector, has the best energy resolution ($\sim 3\%$ at 662 keV γ -rays) among all inorganic scintillation detectors as well as very good timing properties. The BaF₂, on the other hand, has ultrafast timing properties but, has a poor energy resolution ($\sim 10\%$ at 662 keV γ -rays). We have developed a hybrid LaBr₃(Ce)-BaF₂ four detector time-differential perturbed angular correlation (TDPAC) spectrometer for material studies using the ¹⁸¹Hf radioactive probe. The low energy γ -rays have been selected in LaBr₃(Ce) to minimize the Compton background. The performance of this LaBr₃(Ce)-BaF₂ set up in TDPAC measurement of Zr₂Pd intermetallic compound is reported here.

Experimental

In TDPAC technique, perturbations of angular correlation of a suitable γ - γ cascade of the probe nucleus are measured. The probe ¹⁸¹Hf emits 133-482 keV coincident gamma rays passing through the 482 keV intermediate level with values of $T_{1/2} = 10.8$ ns, spin $I = 5/2^+ \hbar$, electric quadrupole moment $Q = 2.36$ b and magnetic dipole moment $\mu = 3.24 \mu_N$. The charge distribution of the surrounding probe environment with a non-cubic symmetry produces an electric field gradient (EFG) which interacts with the electric quadrupole moment of the probe nucleus. In a magnetic material, the angular correlation can be perturbed by the interaction of internal magnetic field with the magnetic dipole moment of the probe nucleus. For nuclear quadrupole interaction (NQI), the angular correlation of 133-

482 keV cascade of ¹⁸¹Ta after the β^- decay ($T_{1/2} = 42.4$ d) of ¹⁸¹Hf is perturbed in a material by the interaction of the extranuclear EFG with the electric quadrupole moment of the 482 keV level.

The schematic diagram of the experimental set up is shown in FIG 1. The crystal sizes used are 38.1×25.4 mm² and 50.8×50.8 mm² for LaBr₃(Ce) and BaF₂, respectively. The LaBr₃(Ce) scintillator was coupled with XP2020 PMT and the BaF₂ scintillator was coupled with Philips XP2020/Q PMT. The 133 keV γ -ray was selected in LaBr₃(Ce) detector and the 482 keV γ -ray was selected in BaF₂ detector. Four slow-fast coincidence assemblies, viz. 1/3, 1/4, 2/3 and 2/4 have been made where, 1/4 and 2/3 combinations are at 90° and 1/3 and 2/4 combinations are at 180°. Signal from the last dynode was used for energy measurements and the anode pulse was used for timing measurements. From the four coincidence spectra at 180° and 90°, the perturbation function $G_2(t)$ was found. The function $G_2(t)$ is defined as [1],

$$G_2(t) = S_{20}(\eta) + \sum_{i=1}^3 S_{2i}(\eta) \cos(\omega_i t) \exp(-\delta \omega_i t). \quad (1)$$

The ω_i are the transition frequencies between the m -sublevels of the intermediate state arising due to hyperfine splitting, δ is the frequency distribution width (Lorentzian damping) and η is the asymmetry parameter of the EFG. By fitting the TDPAC spectrum with Eq. 1, we can determine ω_i and therefrom the electric quadrupole frequency (ω_Q). EFG (V_{zz}) is related to ω_Q by the relation,

$$\omega_Q = \frac{eQV_{zz}}{4I(2I-1)\hbar}. \quad (2)$$

Typical prompt time resolution obtained with

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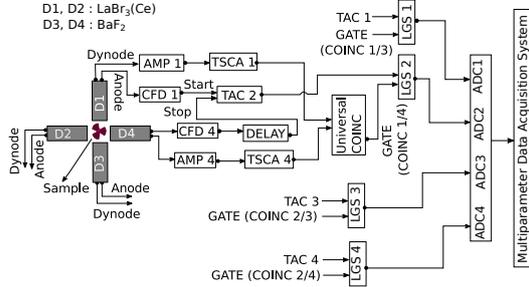


FIG. 1: Schematic diagram of the four detector TDPAC spectrometer

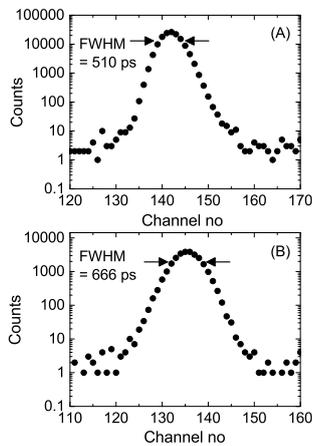


FIG. 2: Prompt time resolution using a $\text{LaBr}_3(\text{Ce})\text{-BaF}_2$ set up (A) for 511 keV annihilation γ -rays of ^{22}Na (B) 133-482 keV γ -ray energy selection with ^{22}Na as a prompt source. TAC calibration is 103 ps/ch.

$\text{LaBr}_3(\text{Ce})\text{-BaF}_2$ detector set up is shown in FIG 2.

The sample Zr_2Pd was prepared in argon arc furnace using the stoichiometric amounts of constituent elements. The metal Zr was procured from M/S Alfa Aesar (purity 99.2% excluding Hf) and metal Pd was procured from SIGMA-ALDRICH (purity 99.9%).

Results and discussion

The TDPAC measurement in the inter-metallic Zr_2Pd has been carried out to determine the crystalline EFG in the sample. The Zr_2Pd has tetragonal MoSi_2 type crystal

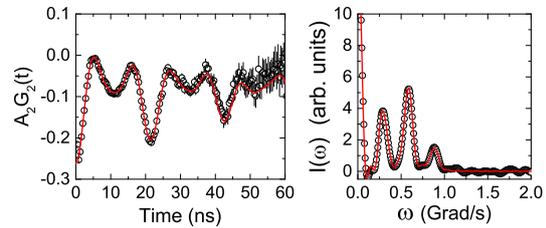


FIG. 3: Figure in the left shows the TDPAC time spectrum in Zr_2Pd at room temperature and its fourier cosine transform is shown in the right.

structure. The TDPAC spectrum obtained at room temperature in Zr_2Pd is shown in FIG 3. The major component ($\sim 80\%$) produces $\omega_Q = 49.1(1)$ Mrad/s, $\eta = 0$ and $\delta = 2.4(3)\%$. The results can be compared with the earlier reported results [2]. A single quadrupole frequency component with values of $\omega_Q = 45.7(6)$ Mrad/s and $\eta = 0$ was observed in Zr_2Pd by Wodniecka et al. [2] where TDPAC measurements were performed using a $\text{BaF}_2\text{-BaF}_2$ set up. The present result of ω_Q for the predominant component is found to be slightly larger than the earlier reported result. This component can, therefore, be assigned to Zr_2Pd . Along with the predominant component, two minor components have been observed from present measurement. The second minor component ($\sim 8\%$) produces values of $\omega_Q = 45.8(8)$ Mrad/s, $\eta = 0.59(5)$ and the third minor component ($\sim 9\%$) with values of $\omega_Q = 43.0(9)$ Mrad/s, $\eta = 0$ is found to be present. These minor components probably arise due to the presence of Hf impurity atoms (3-5%) in the sample. The Hf atom can be present in the Zr_2Pd unit cell as one nearest neighbor. It can also occupy the interstitial site in the lattice or its presence at second nearest neighbor can influence the EFG in Zr_2Pd .

References

- [1] Matthew Zacate et al., Defect and Diffusion Forum **311**, 3-38 (2011).
- [2] B. Wodniecka et al., J Alloys Compd. **219**, 132-134 (1995).