

Investigation of Electron Capture after- Effects and Environmental Changes in some Barium Compounds with ^{133}Ba Nuclear Probe

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Perturbed angular correlation parameters in various compounds of barium are measured to investigate influences of chemical and physio-chemical environments in the K-capture decay of ^{133}Ba to ^{133}Cs . It is found that the correlation coefficients of 356-81 keV cascade are attenuated purely as a matter of half-life ($T_{1/2} = 6.36$ ns) of 81 keV intermediate state and the electric field gradients available at the probe site. The results are a step forward to establish insignificance of after-affects preceding K-capture and K-conversion.

Key Words: (K-Capture, K-Conversion, Electron Capture Decay After-Affects, Correlation Time, Electric Field Gradients)

INTRODUCTION

The perturbed angular correlation spectroscopy is sensitive to the charge distribution around a probe atom on an atomic scale. If such a probe atom is part of an electrically active complex, the charge transition can be observed directly. The gamma-gamma Perturbed Angular Correlation (PAC) technique, described in detail in the review by Frauenfelder and Steffen¹ and with special emphasis on biological applications by Bauer², has been useful for the study of gyromagnetic-ratios, electric quadruple moments of the excited nuclear states and electric field gradients at the sites of probe nuclei in some solid crystalline materials. The technique has also been applied to determine the binding constants of some biomolecules and relaxation constants of some liquids. A difficulty is however encountered in the interpretation of PAC results, where gamma-gamma cascade is preceded by electron capture. The different workers³⁻⁷ differ over the question of EC-after-affects.

The present measurements are made to observe changes in PAC parameters in different environments such as ethylene diamine tetra acetic acid (EDTA), Ba-1,2,3-trihydroxypropanol (Glycerol) and Ba- oxyquinoline (Oxine). To study the problem of electron capture after-effects, in addition to 356-81 keV cascade prompt 276-161 keV cascade of ^{133}Cs following the EC-decay of ^{133}Ba ($T_{1/2} = 10.7$ y) has also been observed in various chemical environments. The binding characteristics of radioactive probe with the host atoms are also explored.

EXPERIMENTAL

The radioactive isotope ^{133}Ba was procured from Bhabha Atomic Research Centre (BARC), Trombay, Bombay, India in the form of BaCl_2 in dilute HCl solution. For each compound, the radioactive solution was taken in sufficient quantities to produce 500-800 counts per second of 356 keV singles gamma ray of ^{133}Ba . The count rate was measured twice, first radioactive BaCl_2 solution before synthesis and second resulted compound of barium separated from its by-products after synthesis. This was done to make sure about the radioactive contamination of the by-products during synthesis and the right execution of the synthesis process. About 100,000 coincidence counts were taken at each angle for each sample, while each radioactive sample was placed in cylindrical Perspex vial of dimensions $3\text{mm}^2 \times 10\text{mm}$.

The time integral perturbed angular correlation measurements of the 356-81 and 276-161 keV gamma-gamma cascades were made with a combination of coaxial 120c.c. HpGe and $2'' \times 2''$ NaI (TI) detectors coupled with fast-slow coincidence circuitry with resolving time $2\tau = 50$ ns. Anisotropy for each of the samples was measured at two angles of 180° and 90° . The angular correlation coefficients (A_{22}) measured in different chemical environments were corrected for chance coincidences and finite source-detector solid angles.

Determination of time integral perturbed angular correlation coefficients:

The $G_{22}(\infty)$ coefficients were determined from $A_{22} \pm \Delta A_{22}$, the measured time integral angular correlation coefficients for different compounds of Barium for 356-81keV and 276-161keV cascades. The G_{22} corresponds to the interaction between the nucleus and its environment. Angular frequency W_o and quadrupole interaction frequency W_Q are related to each other by the following relation,

$$\begin{aligned} W_o &= 3 W_Q \quad \text{for even } l, \\ &= 6W_Q \quad \text{for odd } l \end{aligned} \quad (1)$$

$$\text{also } W_Q = \frac{eQV_{zz}}{4I(2I-1)\hbar} \quad (2)$$

where Q is the electric quadrupole moment of 81keV state of ^{133}Cs . The attenuation factor G_{22} in terms of correlation time, given by Marsden *et al.*⁸ is

$$G_{22} = \frac{1}{1 + \lambda\tau_N} \quad (3)$$

where, $\lambda = 3/5 \tau_c \omega_Q^2 K(K+1) \times [4I(I+1) - K(K+1) - 1]$.

Here, τ_N is the mean life time of the 81 keV level. The variation of correlation time (τ_c) with varying concentrations of the environment was investigated by the author in one of earlier works⁹. Correlation time was found to be directly proportional to the concentration of the ligand and very large sized molecules were required to have much correlation times.

The theoretical behaviour of $G_{22}(\infty)$ against W_0 is shown in the figure. The W_0 values were noted from the figure as against the experimentally determined $G_{22}(\infty)$ values and W_Q , the quadrupole interaction frequency was obtained using Eq. (1). The Z-components of electric field gradient, V_{ZZ} were calculated using Eq. (3) for various environments.

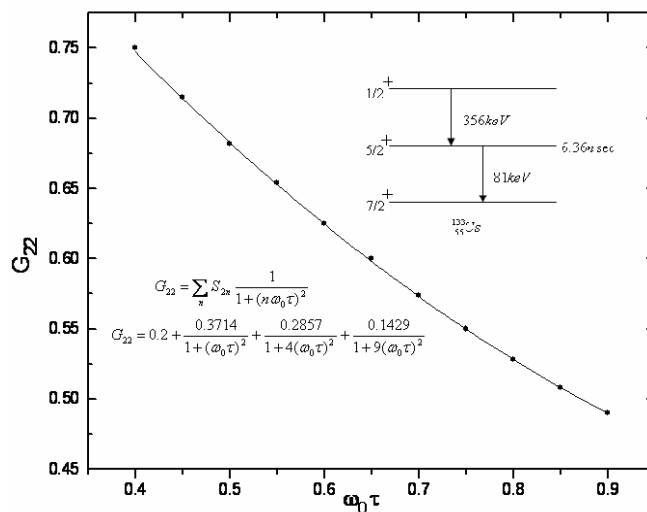
RESULTS AND DISCUSSION

The angular correlation coefficients for the 356-81 keV and 276-161 keV cascades of ^{133}Ba in different chemical environments are listed in the Table. The attenuation coefficients G_{22} and the quadrupole interaction frequencies ' W_Q ' for 356-81 keV cascade of ^{133}Ba in Oxine, Glycerol and M/8 EDTA are also shown in the last two columns of the Table.

Table: The $A_{22} \pm \Delta A_{22}$ Co-efficients for 356-81keV and 276-161keV cascade correlations alongwith the values of attenuation coefficients and quadrupole interaction frequencies obtained in some barium compounds.

Barium compounds	$A_{22} \pm \Delta A_{22}$	$A_{22} \pm \Delta A_{22}$	Attenuation coefficients (G_{22})	Interaction frequencies (W_Q) mrad/sec
	(356-81 keV cascade)	(276-161 keV cascade)		
Ba + HCl (dilute)	0.035±0.002	-0.328±0.0027	-	-
Ba + Glycerol	0.026±0.005	-0.337±0.026	0.75±0.05	7.355
Ba + Oxine	0.025±0.004	-0.334±0.023	0.75±0.04	7.409
Ba + M/8 EDTA	0.024±0.005	-0.332±0.025	0.66±0.20	9.843

The low values of interaction frequencies represent weak bonding in the compounds. Further, the electric quadrupole moment of the 81 keV state of ^{133}Cs has been theoretically estimated as 0.5b. The value of the electric field gradient, V_{ZZ} as measured from Eq. (2) comes out to be equal to $1.29 \times 10^{16} \text{ V/cm}^2$. These values are generally found to be of the order of (2 to 8) $\times 10^{17} \text{ V/cm}^2$ for compounds of comparable molecular weights¹⁰. This quite a low value of the electric field gradient at the probe site explains weak bonding. Obviously one or two of the hydroxyl groups in the glycerol molecule at least, are expected to form

Fig 1: $G_{22}(\infty)$ verses $\omega_0 \tau$ plot for $l=5/2$.

ionic bond with the probe atom. Ionic bonds are electrostatic attractions between oppositely charged species. The strength of an ionic bond is related to distance. Ionic bonds are not broken but their strength is simply diminished by distance. This justifies the weak interaction and hence a lower field gradient at the probe site.

Further, both the orienting transitions of 276γ and 356γ originate from the same level of 437 keV. The energy level at 161 keV which is the intermediate state of 276-161 keV cascade is short lived while the 81 keV level in the 356-181 keV cascade has significant life time (6.36 ns). The overlapping of the correlation coefficients of 276-161 keV cascade indicates that there is no effect of the preceding electron captures and/or internal conversions in the case of EC-decay of ^{133}Ba to ^{133}Cs .

Conclusion

The presence of electron capture after affects would have perturbed the angular correlation coefficients of 276γ - 161γ keV cascade. However the overlapping of A_{22} coefficients for 276γ - 161γ cascade for all the compounds had ruled out any perturbations. The fact excludes the presence of any decay after-affects. The attenuations of coefficients observed in different chemical compounds are caused due to exposure of the nuclear probe to the interactions of nuclear quadrupole moment and the electric field gradient generated at the probe site due to environment of the probe. It is arrived at that, if the PAC technique is a useful tool to perturbation studies, the ^{133}Ba probe is certainly a useful probe to

determine the EFG's in measurements especially for compounds having low natural abundance (0.1%).

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