

# 1-PAC: A one-detector gamma-gamma perturbed angular correlation technique for the determination of physical-chemical properties of radioindium

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$^{111}\text{In}$  is widely used in fundamental and applied studies. The defining and measuring of its physico-chemical status in the explored systems, however, is still a crucial issue due to its ultra-micro concentration. The method of analysing perturbed angular correlations (PAC) of cascade  $\gamma$ -emissions has been developed as a sensitive tool in condensed matter science, with applications to a wide range of tasks involving nuclear hyperfine interactions (HFI) [1,2].

For a radionuclide decaying by two  $\gamma$ -rays in a cascade such as  $^{111}\text{In}$ , the detection of  $\gamma_1$  in a given direction selects an aligned subensemble of nuclear spins with non-isotropic orientation. The emission of a subsequent  $\gamma_2$  with  $\gamma_1$  already emitted general depends on the angle between  $\gamma_1$  and  $\gamma_2$ , i.e. the coincidence count rate for these  $\gamma$ -rays can be anisotropic. Both single count rates for  $\gamma_1$  and  $\gamma_2$  do not depend on angles for randomly oriented spins, but their coincidence count rate does. During the time between the emission of these two  $\gamma$ -rays this angular correlation can be "perturbed" by the interaction of nuclear moments of this intermediate state with extranuclear fields such as, for example, the nuclear quadrupole moment  $Q$  and the electric field gradient (EFG) tensor at the nuclear site which arises from all extranuclear charges. Quantum mechanically, this HFI causes an energy splitting of the intermediate energy level of the nucleus, provided the intermediate state life time  $\tau_n$  is long enough.

The time-integrated angular correlation of cascade (TIAC)  $\gamma$ -rays is given by  $\overline{W(\theta, \infty)} = 1 + A_{22} \overline{G_2(\infty)} Q_2 P_2(\cos(\theta)) + \dots$ , where  $A_{ii}$  is the angular correlation coefficients depending on spins and multiplicities of transitions;  $P_i(\cos\theta)$  are the Legendre polynomials;  $Q_i$  are the solid angle correction factors taking into account the detector and source finite sizes and  $\overline{G_i(\infty)}$  is the TIAC perturbation factor which yields information on HFI. The TIAC  $\overline{W(\theta, \infty)}$  of the  $^{111}\text{In}$  cascade  $\gamma$ -ray coincidences are given in Fig. 1. As for this isotope  $A_{44} \ll A_{22}$ , only  $A_{22}$  has been considered in further calculations. The solid line represents the unperturbed angular correlation ( $\overline{G_2(\infty)} = 1$ ); the dotted line represents the perturbed angular correlation ( $\overline{G_2(\infty)} = 0.2$ ). Experimentally,  $\overline{G_2(\infty)} = 1$  corresponds to solutions of  $^{111}\text{In}$  in inorganic acids ("liquid"), while  $\overline{G_2(\infty)} = 0.2$  corresponds to the hydroxide ("solid" sample) as formed in alkaline solutions [3,4]. This leads to a considerable difference of 15% in the TIAC for  $^{111}\text{In}$  at  $\theta = 0^\circ$  for "solid" and "liquid" states.

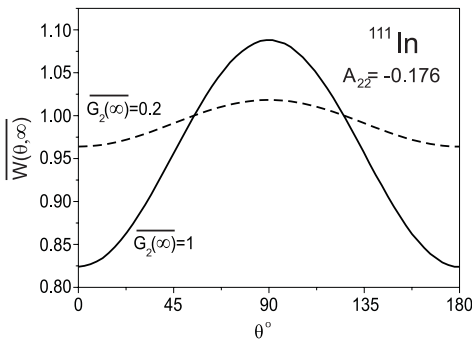


Fig. 1.  $^{111}\text{In}$  time integrated angular correlation dependencies for "solid" and "liquid" states.

The first observations of the sum peak intensity change due to chemical environment was reported for  $^{181}\text{Hf}$  compounds by De Bruin et al [5] and for  $^{111}\text{In}$  compounds by Yoshihara et al [6,7]. However, proposed methods had not received practical applications.

The  $\gamma$ -spectrum of  $^{111}\text{In}$  (Fig. 2) has been obtained using an HPGe-detector with a beryllic window. The spectrum consists of "mono peaks" (MP) corresponding to  $\gamma$ - and X-rays full-energy peaks and "summing peaks" (SP) corresponding to the summing energy of these radiations in different combinations. The "summing peak" is named the "true summing peak" (TSP) in the case of the genetically related radiation (corresponding to the decay of one nucleus). It is assumed that the relative probability of recording of cascade  $\gamma$ -rays  $R_{\gamma_1+\gamma_2}$  in TSP at the individual values of  $A_{ii}$ ,  $\overline{G_i(\infty)}$  and  $Q_i$  is equal to

$$\text{TIAC } \overline{W(\theta, \infty)} \text{ with } R_{\gamma_1+\gamma_2} = \overline{W_{A_{ii}, G_i, Q_i}^{\gamma_1, \gamma_2}}(\theta = 0^\circ, \infty)$$

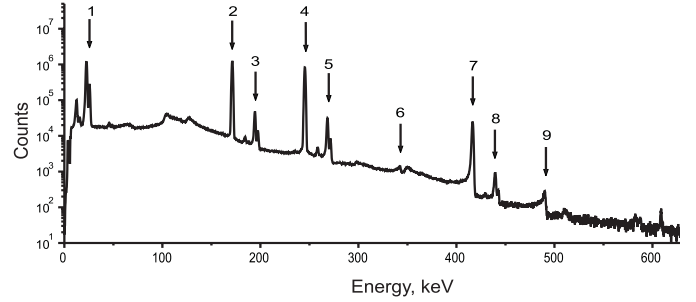


Fig. 2. The  $\gamma$ -spectrum of  $^{111}\text{In}$  obtained using an HPGe-detector.

1 - X-ray peaks ( $K_{\alpha\beta}$ ); 2,4- cascade  $\gamma$ -ray full-energy peaks (171.3 keV and 245.4 keV); 3,5- summing of a X-ray and a cascade  $\gamma$ -rays full-energy peak; 6,9 - accidental coincidences peaks (171.3 keV + 171.3 keV and 245.4 keV + 245.4 keV); 7- summing of cascade  $\gamma$ -rays peaks (171.3 keV + 245.4 keV); 8- triple summing peaks (171.3 keV + 245.4 keV +  $K_{\alpha\beta}$ ).

Probabilities of the SP and MP registration for the case of small count rates and small full-energy efficiencies of  $\gamma$ -rays in the case of a point source ( $S_{\text{MP}} \gg S_{\text{TSP}} \gg S_{\text{ASP}}$ ) are  $S_{\gamma_1} = \varepsilon_{\gamma_1} \cdot y_{\gamma_1} \cdot A \cdot t$  and  $S_{\gamma_2} = \varepsilon_{\gamma_2} \cdot y_{\gamma_2} \cdot A \cdot t$  or

$S_{\gamma_1+\gamma_2} = \varepsilon_{\gamma_1} \cdot y_{\gamma_1} \cdot \varepsilon_{\gamma_2} \cdot (1 - \alpha_{\text{int}}(\gamma_2)) \cdot A \cdot t \cdot R_{\gamma_1+\gamma_2}$  where  $S_{\gamma_n}$  - MP area;  $S_{\gamma_1+\gamma_2}$  - TSP area;  $A$  - radioactivity of the source;  $t$  - time of measuring;  $\varepsilon_{\gamma_n}$  - detector efficiency of  $\gamma_n$  of a full-energy peak at the given source-detector geometry;  $y_{\gamma_n}$  - abundance of  $\gamma_n$  per decay;  $\alpha_{\text{int}}(\gamma_n)$  - internal conversion coefficient of the  $\gamma_n$  emission. As  $y_{\gamma_2} = (1 - \alpha_{\text{int}}(\gamma_2))$  for  $^{111}\text{In}$  yields  $\frac{S_{\gamma_1+\gamma_2}}{S_{\gamma_1} \cdot S_{\gamma_2}} = \frac{R_{\gamma_1+\gamma_2}}{A \cdot t}$

{1} we shall refer to a TSP composed of  $\gamma_1$  and  $\gamma_2$  each involving a  $K_{\alpha\beta}$  X-ray. This event occurs with a probability of  $y_{K_{\alpha,\beta}}$  each time when the electron hole is formed on the K-shell:

$$S_{K_{\alpha,\beta}} = \varepsilon_{K_{\alpha,\beta}} \cdot y_{K_{\alpha,\beta}} \cdot (EC_k + \alpha_k(\gamma_1) + \alpha_k(\gamma_2)) \cdot A \cdot t$$

$$S_{K_{\alpha,\beta}+\gamma_1} = \varepsilon_{\gamma_1} \cdot y_{\gamma_1} \cdot \varepsilon_{K_{\alpha,\beta}} \cdot y_{K_{\alpha,\beta}} \cdot (EC_k + \alpha_k(\gamma_2)) \cdot A \cdot t \cdot R_{K_{\alpha,\beta}+\gamma_1}$$

$$\frac{S_{K_{\alpha,\beta}+\gamma_1}}{S_{K_{\alpha,\beta}} \cdot S_{\gamma_1}} = \frac{1}{N_{K_{\alpha,\beta}+\gamma_1}} \cdot \frac{R_{K_{\alpha,\beta}+\gamma_1}}{A \cdot t} \quad \{2\} \text{ with } N_{K_{\alpha,\beta}+\gamma_1} = \frac{EC_k + \alpha_k(\gamma_2)}{EC_k + \alpha_k(\gamma_1) + \alpha_k(\gamma_2)}$$

with  $EC_k$  - fraction of the electron capture (EC) from the K-shell within the electron capture decay;  $\alpha_k(\gamma_n)$  - internal conversion coefficient  $\gamma_n$  of the radiation from the K-shell. As the angular correlation between  $\gamma$ - and X-ray is isotropic in any case,  $R_{K_{\alpha,\beta}+\gamma_1}$  is equal to 1. Dividing {1}/{2}, yields

$$R_{\gamma_1+\gamma_2} = N_{K_{\alpha,\beta}+\gamma_1} \cdot \frac{S_{\gamma_1+\gamma_2} \cdot S_{K_{\alpha,\beta}}}{S_{\gamma_2} \cdot S_{\gamma_1+K_{\alpha,\beta}}} \text{ or } R_{\gamma_1+\gamma_2} = N_{K_{\alpha,\beta}+\gamma_2} \cdot \frac{S_{\gamma_1+\gamma_2} \cdot S_{K_{\alpha,\beta}}}{S_{\gamma_1} \cdot S_{\gamma_2+K_{\alpha,\beta}}}$$

$$\text{with } N_{K_{\alpha,\beta}+\gamma_2} = \frac{EC_k + \alpha_k(\gamma_1)}{EC_k + \alpha_k(\gamma_1) + \alpha_k(\gamma_2)}$$

**Conclusion:** A method for the measurement of the  $\gamma$ - $\gamma$  perturbed angular correlation for cascade  $\gamma$ -emissions was developed using one detector only. The expressions are convenient to determine  $R_{\gamma_1+\gamma_2}$  due to their symmetry because both the numerator and the denominator involve one MP and one SP.

[1] Frauenfelder H, Steffen RM Alpha-, Beta, and Gamma-ray Spectroscopy (Siegbahn K, ed.), North-Holland, Amsterdam, 2 (1965) 97-1198. [2] Lurf A, Butz T. Angew Chem Int Ed Engl, 26 (1987) 110-126. [3] Demille GR et al. Chemical Physics Letters 44 (1976) 164-168. [4] Akselrod ZZ et al. Z Naturforsch 55a (2000) 151-154. [5] M de Briun, PJM Korthoven, Radiochem Radioanal Letters, 21 (1975), 287; [6] K Yoshihara et al. Radiochem Radioanal Letters, 58 (1983) 9. [7] K Yoshihara et al. Radiochem Radioanal Letters, 58 (1983) 17.