Development of a γ-γ-perturbed angular correlation one-detector method (1-PAC) for investigation of physico-chemical properties of matter

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Abstract

A new method of the perturbed angular γ–γ-correlation (PAC) measurements was developed using one HPGe detector only (1-PAC). For quantifying the perturbation factor, the ratio $R_{\gamma_1+\gamma_2}$ of the summing peak composed of two cascade γ-rays (the perturbation-affected parameter) to the summing peak composed of one γ-ray and one X-ray (the perturbation non-affected parameter) was used. This method was demonstrated for the radionuclides ^{111}In and [111m]Cd, identifying the chemical status of the radionuclides in aqueous solutions. © 2002 Elsevier Science Ltd. All rights reserved.

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1. Introduction

Radioactive nuclides are widely used in fundamental and applied studies. While the decay characteristics of the radioisotopes studied can be exactly measured, defining and characterizing their physico-chemical states in the explored systems is difficult if not impossible. These problems may be solved in part with a method of perturbed angular correlations (PAC) of cascade γ-rays. The PAC method has been developed as a sensitive tool in condensed matter science, with applications to a wide range of tasks involving nuclear hyperfine interactions (HFI). The PAC method enables to explore properties of material in solid, fluid and gaseous states. On the one hand, the PAC method is based on the phenomenon of an angular correlation between outgoing directions of cascade γ-rays emitted from the same nucleus as it is a nuclear physics constant and a characteristic of the actual nucleus. On the other hand, the PAC method is based on the perturbation of this correlation by extranuclear fields, due to the interactions of a nuclear quadrupole moment Q and/or a magnetic dipole moment with an electric field gradient and/or magnetic fields which arise from all surrounding extranuclear charge distributions. The physical basis and the theory of the PAC method, as well as experimental techniques are described in detail in many papers and monographs, (cf. Frauenfelder and Steffen, 1965; Lerf and Butz, 1987 for example).

[111]In and [111m]Cd are the radionuclides most frequently used in γ–γ-PAC measurements. Their decay schemes are shown in Fig. 1. The typical block diagram of a two-detector traditional PAC coincidence spectrometer for measuring the angular dependencies of the relative probability $\overline{W(\theta, T)}$ of cascade γ-rays is given in Fig. 2.

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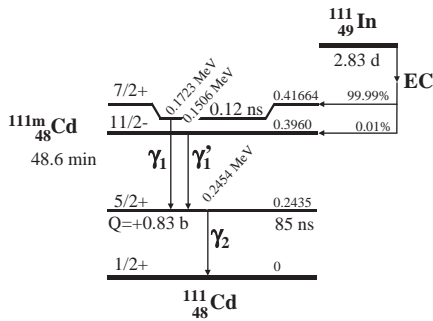*E-mail address:* frank.roesch@uni-mainz.de (F. Rösch).

Fig. 1. Decay scheme of ^{111}In and ^{111m}Cd (Lederer and Shirley, 1978).
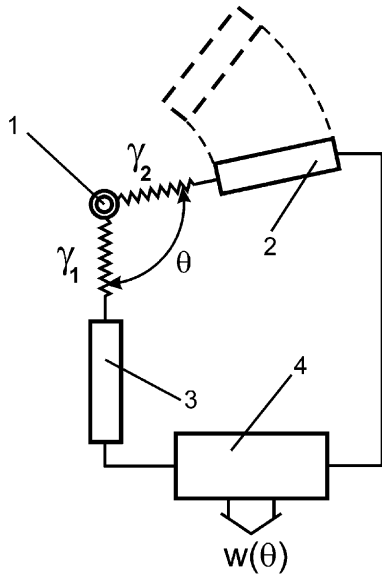

Fig. 2. A schematic view of a typical PAC experimental block diagram: 1—source; 2,3 —detectors; 4—coincidence unit.

The time-integrated angular correlation of cascade (TIAC) γ-rays is given by

$$\overline{W(\theta, \infty)} = 1 + A_{22}\overline{G_2(\infty)}\, Q_2 P_2(\cos(\theta))$$
$$+ A_{44}\overline{G_{44}(\infty)}\, Q_4 P_4(\cos(\theta)) + \cdots , \qquad (1)$$

where A_{ii} are the angular correlation coefficients depending on spins and multipolarities 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 $\overline{W(\theta, \infty)}$ for the ^{111}In (172–245 keV) and ^{111m}Cd (150–245 keV) cascade γ-ray coincidences are shown in Fig. 3. As for these isotopes $A_{44} \ll A_{22}$, 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 $(\overline{G_2(\infty)} = 0.2)$. These cases are realized experimentally in individual physico-chemical cases: $\overline{G_2(\infty)} = 1$ corresponds to solutions of these radionuclides in inorganic acids ("liquid" sample), while $\overline{G_2(\infty)} = 0.2$ corresponds to their hydroxides ("solid" sample) as formed in alkaline solutions (Demille et al., 1976; Akselrod et al., 2000). This leads to a considerable difference of 15% in the TIAC for ^{111}In at $\theta = 0°$ for solid and liquid states.

The γ-spectrum of ^{111}In (Fig. 4) has been obtained using a high-purity germanium (HPGe) detector with a beryllium window. The spectrum consists of mono peaks (MP) corresponding to γ- and X-ray full-energy peaks and summing peaks (SP) corresponding to the summing energy of these radiations in different combinations. The SP is named the true summing peak (TSP) in the case of the genetically correlated radiations (corresponding to one decay event) and it is named an accidental summing peak (ASP) in the case of the genetically un-correlated
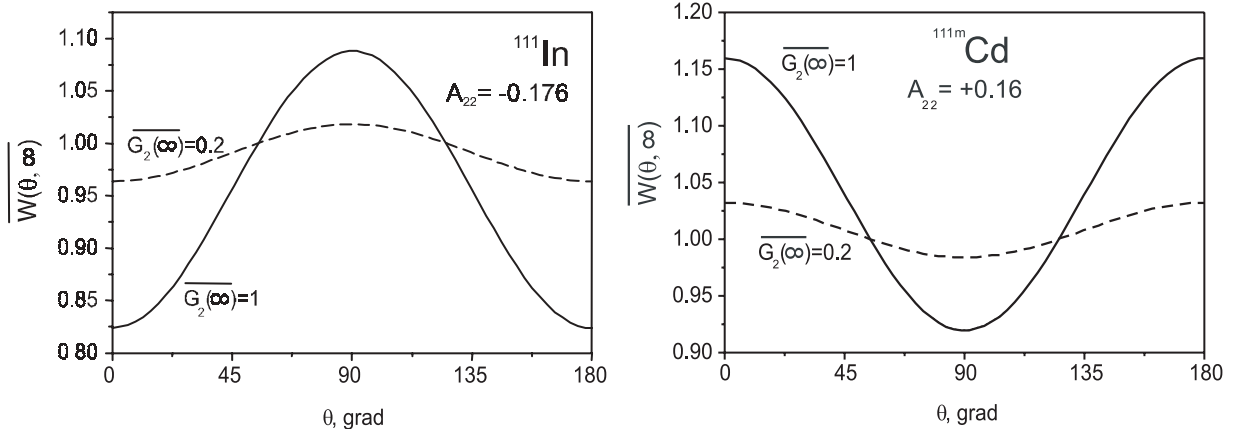

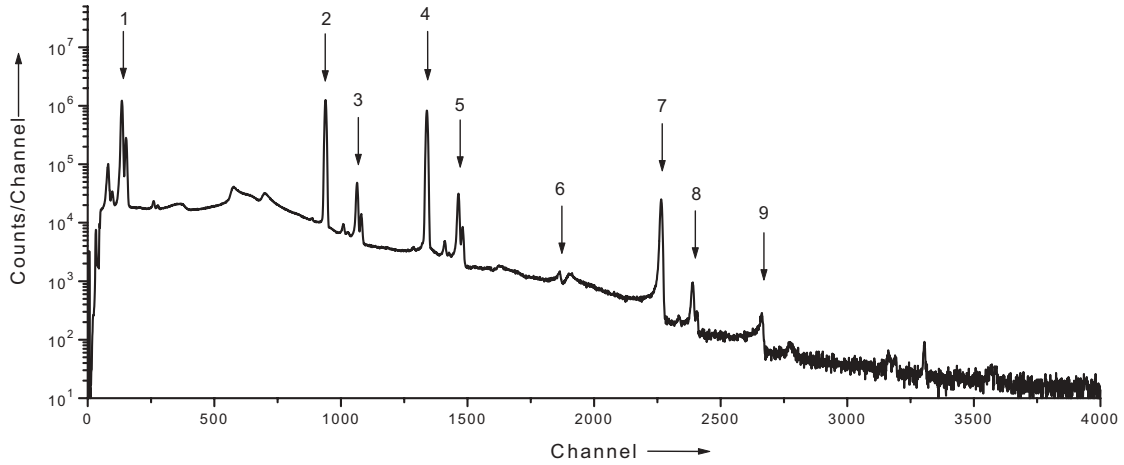Fig. 3. Angular dependencies of relative probability of cascade γ-ray coincidences for ^{111}In and ^{111m}Cd.

Fig. 4. The γ-spectrum of ^{111}In: 1—X-ray peaks ($K_{\alpha,\beta}$); 2,4—the cascade γ-ray full-energy peaks (171.3 and 245.4 keV); 3,5—summing of a X-ray and cascade γ-ray full-energy peaks; 6,9—accidental coincidences peaks (171.3 + 171.3 keV and 245.4 + 245.4 keV); 7—summing of cascade γ-ray peaks (171.3 + 245.4 keV); 8—triple summing peaks (171.3 + 245.4 keV + $K_{\alpha,\beta}$).

radiation (corresponding to the different decay events). The concept to register SPs from the nuclides with one detector is in many respects similar to their registration with a coincidence unit in two- or multi-detector facilities of the PAC method.

It is assumed that the relative probability of recording the cascade γ-rays γ_n, γ_m in TSP $R_{\gamma_n+\gamma_m}$ at the respective A_{ii}, $\overline{G_i(\infty)}$ and Q_i values is equal to $\overline{W(\theta, \infty)}$:

$$R_{\gamma_n+\gamma_m} = \overline{W^{\gamma_n,\gamma_m}_{A_{ii},G_i,Q_i}(\theta = 0^0, \infty)}. \quad (2)$$

The first observations of the sum peak intensity variations due to changes of the chemical environment was reported for ^{181}Hf compounds by de Bruin and Korthoven (1975) and for ^{111}In compounds by Yoshihara et al. (1983a, b). However, the proposed methods have not found significant application.

2. A quantitative analysis of the 1-PAC method

For the quantitative analysis of the 1-PAC method the radionuclides ^{111}In and ^{111m}Cd shall be considered. Areas of the SP and MP in the case of small count rates and small full-energy efficiencies of the γ-ray registration in the case of a point source ($S_{MP} \gg S_{TSP} \gg S_{ASP}$) are

$$S_{\gamma_1} = \varepsilon_{\gamma_1} y_{\gamma_1} At, \quad (3)$$

$$S_{\gamma_2} = \varepsilon_{\gamma_2} y_{\gamma_2} At, \quad (4)$$

$$S_{\gamma_1+\gamma_2} = \varepsilon_{\gamma_1} y_{\gamma_1} \varepsilon_{\gamma_2} (1 - \alpha_{\text{tot}}(\gamma_2)) At R_{\gamma_1+\gamma_2}, \quad (5)$$

where S_{γ_n} is the MP area, $S_{\gamma_n+\gamma_m}$ the TSP area, A the radioactivity of the source, t the time of measuring, ε_{γ_n}

the detector registration efficiency of γ_n in a full-energy peak at the given source-detector geometry, y_{γ_n} the abundance of γ_n per decay, $\alpha_{tot}(\gamma_n)$ the internal conversion coefficient of the γ_n emission.

As $y_{\gamma_2} = (1 - \alpha_{tot}(\gamma_2))$ for ^{111}In, Eqs. (3)–(5) yield

$$\frac{S_{\gamma_1+\gamma_2}}{S_{\gamma_1} S_{\gamma_2}} = \frac{R_{\gamma_1+\gamma_2}}{At}. \quad (6)$$

Thus, if the absolute activity of the source is known it is possible to determine the $R_{\gamma_1+\gamma_2}$ value with one detector only. In practice, with identical geometry it is possible to use ratios of type

$$\frac{S_{\gamma_1+\gamma_2}}{S_{\gamma_2}} \quad \text{or} \quad \frac{S_{\gamma_1+\gamma_2}}{S_{\gamma_1}}$$

as an estimation of the $R_{\gamma_1+\gamma_2}$. These ratios are proportional to the registration efficiencies ε_{γ_1} and ε_{γ_2} accordingly. In this case, the relative error of the results obtained involves a relatively large individual error of the determination of ε_{γ_1} and ε_{γ_2} due to the error of source-detector geometry and the source self-absorption. This approach was used by Yoshihara et al. (1983a, b). We propose to minimize the influence of these factors by involving X-rays ($K_{\alpha,\beta}$). This radiation occurs with a probability $y_{K_{\alpha,\beta}}$ each time, when the electron hole had formed on the K-shell. Areas of TSP of γ_1 or γ_2 and $K_{\alpha,\beta}$ are

$$S_{K_{\alpha,\beta}} = \varepsilon_{K_{\alpha,\beta}} y_{K_{\alpha,\beta}} (EC_K + y_{e(K)}(\gamma_1) + y_{e(K)}(\gamma_2)) At, \quad (7)$$

$$S_{K_{\alpha,\beta}+\gamma_1} = \varepsilon_{\gamma_1} y_{\gamma_1} \varepsilon_{K_{\alpha,\beta}} y_{K_{\alpha,\beta}} (EC_K + y_{e(K)}(\gamma_2)) At R_{K_{\alpha,\beta}+\gamma_1}, \quad (8)$$

$$\frac{S_{K_{\alpha,\beta}+\gamma_1}}{S_{K_{\alpha,\beta}}S_{\gamma_1}} = N_{K_{\alpha,\beta}+\gamma_1}\frac{R_{K_{\alpha,\beta}+\gamma_1}}{At}, \tag{9}$$

where

$$N_{K_{\alpha,\beta}+\gamma_1} = \frac{EC_K + y_{e(K)}(\gamma_2)}{EC_K + y_{e(K)}(\gamma_1) + y_{e(K)}(\gamma_2)} \tag{10}$$

where EC_K is the fraction of the electron capture (EC) from the K-shell within the electron capture decay, and $y_{e(K)}(\gamma_n)$ is the yield of electrons in the conversion process on the K-shell, corresponding to γ_n.

As there is no angular correlation between γ- and X-rays, $R_{K_{\alpha,\beta}+\gamma_1}$ is equal to 1. After dividing expression (6) by (9), we obtain

$$R_{\gamma_1+\gamma_2} = N_{K_{\alpha,\beta}+\gamma_1}\frac{S_{\gamma_1+\gamma_2}S_{K_{\alpha,\beta}}}{S_{\gamma_2}S_{\gamma_1+K_{\alpha,\beta}}} \tag{11}$$

or, analogously,

$$R_{\gamma_1+\gamma_2} = N_{K_{\alpha,\beta}+\gamma_2}\frac{S_{\gamma_1+\gamma_2}S_{K_{\alpha,\beta}}}{S_{\gamma_1}S_{\gamma_2+K_{\alpha,\beta}}} \tag{12}$$

with

$$N_{K_{\alpha,\beta}+\gamma_2} = \frac{EC_K + y_{e(K)}(\gamma_1)}{EC_K + y_{e(K)}(\gamma_1) + y_{e(K)}(\gamma_2)}. \tag{13}$$

Expressions (11) and (12) are extremely 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. The deduction of the formulas for ^{111m}Cd and their analysis is identical to the previous one:

$$R_{\gamma_1'+\gamma_2} = N_{K_{\alpha,\beta}+\gamma_2}'\frac{S_{\gamma_1'+\gamma_2}S_{K_{\alpha,\beta}}}{S_{\gamma_1'}S_{\gamma_2+K_{\alpha,\beta}}}, \tag{14}$$

$$N_{K_{\alpha,\beta}+\gamma_2}' = \frac{y_{e(K)}(\gamma_1')}{y_{e(K)}(\gamma_1') + y_{e(K)}(\gamma_2)}. \tag{15}$$

For the further calculations the decay data of the radionuclides given in Lederer and Shirley (1978) and Sowby (1983) were used and resulted in $N_{K_{\alpha,\beta}+\gamma_1} = 0.9176(10)$, $N_{K_{\alpha,\beta}+\gamma_2} = 0.9507(10)$, and $N_{K_{\alpha,\beta}+\gamma_2}' = 0.897(1)$.

3. Experimental

Non-carrier-added ^{111}In was purchased from Amersham. ^{111m}Cd was produced via the ^{110}Cd$(n,\gamma)^{111m}$Cd nuclear reaction at the TRIGA research reactor of the University Mainz.

The 1-PAC measurements for ^{111}In and ^{111m}Cd were carried out with two coaxial HPGe detectors of volumes of 200 and 65 cm^3. The conditions of measurements were optimized in order to obtain 10^5–10^6 counts in TSP during 0.5–2 h. For the 200 cm^3 detector, for example, the radioactivity of the samples amounted to ≈ 40 kBq corresponding to a count rate of ≈ 8000 counts/s (cps) at a selected distance of the sample of 39 mm from the detector surface. All measurements have been performed at room temperature.

For all the measurements the samples were measured repeatedly for several times ($n = 5-7$). From these individual measurements, a mean value was calculated, the statistical error being $< 0.5\%$.

3.1. Sample volume and geometry effects

A first series of measurements with no-carrier-added ^{111}In in 0.1 N HCl solution (pH = 1) in a volume of 0.4 ml at 39 and 23 mm sample–detector distances was followed by subsequently increasing the volume of the same sample with 0.1 N HCl up to 0.8 ml at constant sample–detector geometry. The $R_{\gamma_1+\gamma_2}$ values were calculated according to Eqs. (11) and (12). The obtained data are summarized in Table 1.

For a given sample–detector distance the two-fold increased sample volume did not influence the $R_{\gamma_1+\gamma_2}$ values. The deviations observed between the two sample volumes were $\approx 0.25\%$ (at 39 mm distance) and $\approx 0.05\%$ (at 23 mm distance) and are within the statistical error.

A decrease of the distance between sample and detector surface from 39 mm to almost half of this distance (23 mm) resulted in a still negligible change of the corresponding $R_{\gamma_1+\gamma_2}$ values of about 3%. The small effect is mainly due to the change of the geometry factor Q.

Table 1
$R_{\gamma_1+\gamma_2}$ values of no-carrier-added ^{111}In in 0.1 N HCl solution depending on sample volume and sample-detector distances compared to $S_{\gamma_1+\gamma_2}/S_{\gamma_1(2)}$ values (200 cm^3 HPGe-detector)

Volume of sample (ml)	Source–detector distance (mm)	Number of individual measurements	$R_{\gamma_1+\gamma_2}$ (average results with statistical error)		
			$R_{\gamma_1+\gamma_2}$	$S_{\gamma_1+\gamma_2}/S_{\gamma_1}$	$S_{\gamma_1+\gamma_2}/S_{\gamma_2}$
0.4	39	6	0.8867 (5)	0.03736 (4)	0.04714 (4)
0.8	39	6	0.8850 (14)	0.03618 (3)	0.04562 (4)
0.4	23	8	0.9134 (5)	0.06773 (5)	0.08706 (6)
0.8	23	6	0.9138 (5)	0.06611 (5)	0.08484 (5)

In contrast, the values obtained from the calculation of $S_{\gamma_1+\gamma_2}/S_{\gamma_1(2)}$ are very sensitive to changes both of the sample volume and the distance between detector and source, as expressed by changes of 3–4% for the two sample volumes measured at the same sample–detector distance and an almost two-fold increase of $S_{\gamma_1+\gamma_2}/S_{\gamma_1(2)}$ for identical samples at increasing sample–detector distance. This is explained by the significant dependence of these values on the registration efficiencies ε_{γ_n}. Probably this could be the reason, why the de Bruin–Yoshihara treatments have failed to reach routine application in the studies of physico-chemical states of matter.

In addition, several experiments were performed for ^{111}In with much larger (100 ml) and much smaller (100 μl) sample volumes using the 65 cm^3 detector. In the presence of a 10^{-5} M In carrier two different physico-chemical forms of In were considered: In^{3+} in acidic solution (HNO$_3$/NaNO$_3$, pH ≈ 1.5) and In(OH)$_3$ in alkaline solutions (NH$_4$NO$_3$, NH$_3$, pH ≈ 12), $\mu = 0.1$, cf. also the experiments described below. The results obtained are presented in Table 2.

From Table 2 it is evident, that the absolute values of $R_{\gamma_1+\gamma_2}$ for the very large samples (their volumes are comparable to the volume of the detector) strongly differ from the $R_{\gamma_1+\gamma_2}$ values obtained for the very small (point-like) samples with the same chemical form of In (17% for In^{3+}, 14% for In(OH)$_3$). However, the relative changes in $R_{\gamma_1+\gamma_2}$ between the two different physico-chemical species, i.e. from In^{3+} to In(OH)$_3$ are comparable (9.1% for the large and 12.7% for the small sample).

3.2. Solid and liquid states

In a next series of measurements using the proposed 1-PAC method for the determination of $R_{\gamma_1+\gamma_2}$ the pH of the ^{111}In and ^{111m}Cd solutions was varied. A carrier was added in concentrations of 10^{-5} M for In and 10^{-3} M for Cd. The results obtained using the 200 cm^3 volume HPGe-detector at a constant sample–detector distance of 39 mm in acidic and alkaline aqueous solutions of HNO$_3$, NaNO$_3$ and NaOH with a sample volume of 500 μl and an overall ionic strength of $\mu = 0.1$ are summarized in Table 3.

It is known that at pH ≈ 1–2 In and Cd exist in the form of hydrated cations, whereas at pH ≈ 12 hydroxides are formed (Bas and Mesmer, 1986). From Table 3 it can be seen that the value of $R_{171+245}$ for ^{111}In and $R_{150+245}$ for ^{111m}Cd strongly differ for the two states of these elements (solution and precipitate). Table 3 also gives the values of R_{calc} by Eq. (2) for the values $\overline{G_2(\infty)} = 1$ (liquid) and $\overline{G_2(\infty)} = 0.2$ (solid) with $Q_2 = 1$. It is important to note that both the values of R_{exp} for the solution and precipitate states are in agreement with the values of R_{calc} for liquid and solid. Furthermore, the relative changes are comparable: 11.4% as experimentally measured vs. 14.6% theoretically calculated.

Similar experiments have been carried out with ^{111}In on the 65 cm^3 volume HPGe-detector. The volume of the samples was 100 μl and the distance between sample and detector was 25 mm. The values of $R_{171+245}$ for the acidic and alkaline media are represented in Table 4. It can be seen that the results of the measurements obtained with different detectors are in good agreement.

Table 2
$R_{\gamma_1+\gamma_2}$ values of ^{111}In for "very small" and "very large" sample volumes ([In] = 10^{-5} M, overall ionic strength $\mu = 0.1$, 65 cm^3 HPGe-detector)

	Volume of sample	In^{3+} pH ≈ 1.5	In(OH)$_3$ (colloid) pH ≈ 1.5	Relative change (%)
$R_{\gamma_1+\gamma_2(exp)}$	100 μl	0.893(5)	0.982(6)	9.1
	100 ml	0.738(4)	0.845(5)	12.7

Table 3
$R_{\gamma_1+\gamma_2}$ values of ^{111}In and ^{111m}Cd in acidic and alkaline nitrate solutions ([In] = 10^{-5} M, [Cd] = 10^{-3} M, overall ionic strength $\mu = 0.1$, 200 cm^3 HPGe-detector)

Isotope	pH	$R_{\gamma_1+\gamma_2(exp)}$				$R_{\gamma_1+\gamma_2(calc)}$ by Eq. (2) with $Q_2 = 1$
		Eq. (11)	Eq. (12)	Eq. (14)	Average	
^{111}In	1.04	0.869(5)	0.863(5)	—	0.866(5)	0.824 (liquid)
	11.9	0.982(6)	0.971(6)	—	0.977(6)	0.965 (solid)
^{111m}Cd	2.0	—	—	1.095(7)	1.095(7)	1.160 (liquid)
	12.0	—	—	0.972(6)	0.972(6)	1.032 (solid)

Table 4
$R_{\gamma_1+\gamma_2}$ values of ^{111}In in acidic and alkaline nitrate solutions ([In] $= 10^{-5}$ M, overall ionic strength $\mu = 0.1$, 65 cm^3 HPGe-detector)

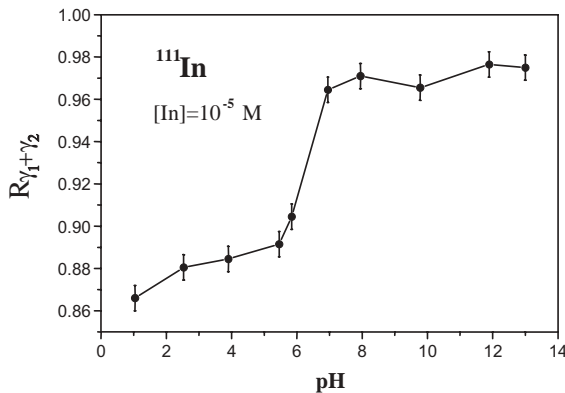| Isotope	pH	$R_{\gamma_1+\gamma_2(exp)}$			$R_{\gamma_1+\gamma_2(calc)}$ by Eq. (2) with $Q_2 = 1$
		Eq. (11)	Eq. (12)	Average	
^{111}In	1.25	0.896(5)	0.891(5)	0.893(5)	0.824 (liquid)
	12.4	0.985(6)	0.979(6)	0.982(6)	0.965 (solid)

Fig. 5. The $R_{\gamma_1+\gamma_2}$ dependencies on pH of inert aqueous nitrate electrolytes for ^{111}In in aqueous solutions, $\mu = 0.1$.

3.3. Hydrolysis of ^{111}In

In an extension of these experiments, the pH of the aqueous solutions was varied systematically over a broad range of pH for ^{111}In, i.e. experiments on the hydrolysis of In have been carried out by means of the 1-PAC method. The pH was changed in solutions of HNO$_3$, NaNO$_3$ and NaOH with an overall ionic strength of $\mu = 0.1$. An In(III) carrier was added in concentrations of 10^{-5} M. The sample–detector distance (39 mm) and the sample volume of 500 µl were kept constant. The 200 cm^3 volume HPGe-detector was used. The obtained data are illustrated in Fig. 5.

In acidic solution, $R_{\gamma_1+\gamma_2}$ represents the values already measured in similar solution (Tables 3 and 4). In the region of pH $= 5$–7 the values of $R_{\gamma_1+\gamma_2}$ strongly increase. This is due to the transition of the radionuclide from the ^{111}In-[In(H$_2$O)$_6$]$^{3+}$ form to that of the insoluble hydroxide ^{111}In-In(OH)$_3$. This is in agreement with stoichiometric equilibrium constants of the In hydrolysis (Bas and Mesmer, 1986). At pH > 7, $R_{\gamma_1+\gamma_2}$ values similar to those determined in the above-mentioned experiments were obtained. The difference of $R_{\gamma_1+\gamma_2}$ in acidic and alkaline media is explained again by the difference of the perturbation factors $\overline{G_2(\infty)}$ for In^{3+} in the case of [In(H$_2$O)$_6$]$^{3+}$ (liquid, $\overline{G_2(\infty)} = 1$) and In(OH)$_3$ (solid, $\overline{G_2(\infty)} = 0.2$), cf. also Tables 3 and 4.

4. Discussion

The optimum conditions for the measurements on the HPGe-detectors with beryllium windows are achieved at radioactivities of a sample of ≈ 30–60 kBq and at a distance between detector and sample of about 2–4 cm. Under these conditions the count rate is several thousand cps, providing a sufficient statistics. The total efficiency of the registration of the photons of 245.3 keV energy is ≈ 3–5%. The relative statistical error of $R_{\gamma_1+\gamma_2}$ determined by Eqs. (11), (12) and (15) is very low for each of the approaches ($< 0.5\%$) and does not exceed an average error between the three approaches of 1% at a measurement time of about 1 h.

For all ^{111}In measurements with volume samples of < 1 ml the values of $R_{\gamma_1+\gamma_2(exp)}$ computed by Eq. (11) are only slightly different from $R_{\gamma_1+\gamma_2(exp)}$ computed by Eq. (12). This fact indicates the reproducibility of results and the reliability of the proposed 1-PAC method, in particular when compared to the approaches previously described (de Bruin and Korthoven, 1975; Yoshihara et al., 1983a, b). Moreover, for both ^{111}In and ^{111m}Cd the experimentally determined values of $R_{\gamma_1+\gamma_2(exp)}$ for the states "solution" and "precipitate" are in good agreement with the theoretical values as expressed by Eq. (2), $R_{\gamma_1+\gamma_2(calc)}$, for the "liquid" and "solid states", respectively.

The dependence of the $R_{\gamma_1+\gamma_2(exp)}$ values on the physico-chemical state of the radionuclide as demonstrated for the hydrolysis of ^{111}In is obvious. Each individual chemical form, i.e. ^{111}In-[In(H$_2$O)$_6$]$^{3+}$ and ^{111}In-In(OH)$_3$, is represented by its species-specific $R_{\gamma_1+\gamma_2}$ value. Moreover, the systematic changes of $R_{\gamma_1+\gamma_2(exp)}$ values between the two species existing in a chemical equilibrium might allow for the determination of equilibrium constants.

5. Conclusions

It was shown, that reproducible and reliable values of $R_{\gamma_1+\gamma_2}$ can be obtained by the proposed 1-PAC method. The approach of γ–γ-perturbed angular correlation can now be utilized at a much higher number of institutions, having access to adequate detectors. Because of the fact

that $R_{\gamma_1+\gamma_2}$ only slightly depends on the experimental set-up such as detector characteristics, source–detector distance, volume of a sample, etc., the $R_{\gamma_1+\gamma_2}$ data can be considered as a reliable characteristic of the physico-chemical state of a radionuclide. The small changes in $R_{\gamma_1+\gamma_2}$ for large and small samples allows the application of the 1-PAC method to measurements even with large samples, e.g. for the determination of the states of radiopharmaceuticals in vivo. This non-invasive measurement of the radionuclides might directly allow for the characterization of their actual chemical speciation. Besides the above-mentioned isotopes it might be possible to use other radionuclides such as [199m]Hg, [133]Ba, [204m]Pb and [118m]Sb in the 1-PAC method as well.

Acknowledgements

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