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Improvements in the PIGE technique via gamma-ray angular distribution☆

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ABSTRACT

Particle induced gamma-ray emission (PIGE) is an ion beam technique used for isotopic elemental analysis of materials, based on gamma-ray spectroscopy. In the traditional PIGE setup, the gamma-ray detector is positioned at the neutral angles 55° or 125° with respect to the beam direction in order to measure gamma-rays with different angular distributions. Although these angles permit measurements without an a-priori knowledge of the specific angular distributions, this choice is not efficient if one wants to identify a specific element since a longer acquisition time is needed.

In this work, we propose to optimize the PIGE measurements by choosing the detector angle position due to the previous knowledge of the gamma-ray multipolarity and the nuclear reaction. We present the angular distribution calculations for the isotopes most frequently identified by PIGE analysis, specially in Cultural Heritage objects. Calculations demonstrate that a gain in the intensity up to 70% can be achieved allowing the elemental identification in shorter times. This result is promising and represents a more efficient alternative to perform PIGE measurements.

1. Introduction

Particle induced gamma-ray emission (PIGE) [1,2] is an ion beam technique based on gamma-ray spectroscopy largely employed to determine elemental composition in a wide variety of materials. For example, it is currently applied to study aerosol samples [3], teeth dentin [4], medicinal plants [5], welded steel joints [6], minerals [7] and Cultural Heritage objects, such as ceramic artifacts [8], glasses [9] and paintings [10]. In most cases, PIGE is commonly used to identify a specific element in a study matrix like fluorine distribution in teeth, oxygen and carbon in steel joints, and sodium in blue pigments made of lapis-lazuli.

Ion beam analysis techniques are based on accelerated charged particles colliding with atoms and nuclei and several reactions can take place simultaneously during the beam-target interaction. The elemental identification depends on several factors such as the beam intensity, beam energy, reaction cross-section, and number of atoms in the sample. In particular, in PIGE technique inelastic nuclear reactions with light nucleus beams (protons, deuterons and alpha particles) are used to excite the nuclei in the sample, but fusion reactions occur as well in the process, resulting in the production of new excited nuclei. In general, the excited nuclei decay emitting characteristic gamma-rays with well known energies which can be used to determine the elemental composition of the sample. In some fusion reactions, it is possible to produce a new isotope which β^- decays to an excited state of another nucleus with atomic number Z-1 that will also decay via gamma-ray emission. The gamma-ray intensities can be used to determine the elemental concentration at a larger depth inside a sample [11], since high energy gamma-rays (100 keV to 10 MeV) are slightly attenuated inside the material compared to the totally absorbed low energy X-rays (from hundreds of eV to tens of keV).

PIGE is usually performed in small accelerators with protons, alpha particles and/or deuteron beams [12–14]. The use of low energies to accelerate the beam projectiles (typically 2–3 MeV for protons) restricts the quantity of reactions that can be performed due to the Coulombian repulsion between the beam and target nuclei. This fact restricts the elements that can be analyzed by this technique, consequently, PIGE is mostly used to identify and to quantify light elements such as Li, Be, B, C, O, F, Na, Mg, Al, Si and P, not easily detected by other ion beam techniques, such as particle induced X-ray emission (PIXE). However, PIGE can be used to detect heavier elements with appropriate nuclear reactions.

PIGE detection limits vary greatly from isotope to isotope because it is based upon specific nuclear reactions, and typical values are between 10 and 100 ppm. To achieve those detection limits, it is necessary to take into account that nuclear reaction cross-sections are small (ranging from few mbarns to hundreds of mbarns) and also to consider the particularities of the gamma-ray emission and its detection efficiency (typically 10^{-3}–10^{-5} full-energy peak absolute efficiency for a HPGe

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gamma-ray detector). These factors contribute to the increasing of the measurement time and the beam exposure required to have enough data to perform a quantitative analysis. In some cases, however, the increase of the beam exposure raises the chance of radiation damage in the sample even when using an external beam, a specially unwanted effect in the study of Cultural Heritage objects such as paintings [15–17] and ceramics [18].

The gamma-ray emission properties play an important role in the position of the gamma-ray detector and in the beam time exposure. We know from gamma-ray spectroscopy that excited states produced in nuclear reactions generally have an oriented angular distribution which depends on the nuclear reaction type to populate the excited states and on the emitted gamma-ray multipolarities [19]. To minimize the contribution of the angular distribution in the quantitative results, a traditional PIGE setup places the gamma-ray detector at 55° or 125° with respect to the beam direction [20]. Although these positions permit to measure gamma-rays with different angular distributions, they do not detect the maximum of the emission, increasing the beam time exposure. This setup, however, is the best option when the electromagnetic properties of the transition are unknown or when the measurement of simultaneous gamma-rays with different multipolarities is required.

With all this in mind, we propose to optimize the PIGE technique in the cases where a specific element is under study by changing the detector angle position based on the previous knowledge of the gamma-ray angular distributions. With this approach, it is possible to detect the maximum of the gamma emission and consequently minimize the measurement time and the radiation dose applied to the sample, which is very important in the case of Cultural Heritage objects. In this work we present the angular distribution calculations for the isotopes most frequently identified by PIGE technique based on the previous knowledge of the isotope nuclear structure provided by gamma-ray spectroscopy studies.

2. Angular distribution theory overview

The angular distribution can be classified by the shape of the orientation with respect to the quantization axis as non-orientation, polarization and alignment [19]. The non-orientation is defined when there is no preferential orientation, an example is the gamma-ray emission of a radioactive source. Polarization is defined when there is a preferential direction of the gamma-ray emission, like the thermal equilibrium orientation realized by Zeeman effect at low temperatures. Alignment occurs when there is an orientation regarding to a quantization axis. The degree of orientation depends on the state formation process and is closely related with the reaction mechanisms. Nuclear reactions or gamma–gamma angular correlations are processes conserving the parity of the states, consequently, only alignment is realized in such processes [19].

Several reaction mechanisms can occur with different probabilities during the interaction of the beam projectiles with the sample depending on the beam's energy and on the type of accelerated particle. As a result, a state with angular momentum j and a projection m along the quantization axis (the direction of the beam projectiles in a nuclear reaction) can be populated with a probability depending on the nuclear reaction type. To solve this problem, we are going to deal with an assembly of systems composed of pure states with probability $P(m)$ to be oriented with respect to a suitable symmetry axis as the quantization axis, which can be described by the statistical tensor $\rho_k(j)$ (1):

$$\rho_k(j) = \sqrt{2j+1}\sum_m (-1)^{j-m}\langle \mathrm{jmj}-m|k0\rangle P(m) \qquad k = 0, 1, \ldots 2j. \tag{1}$$

The angular distribution is also dependent on the gamma-ray multipole radiation and the mixture between them. This multipole radiation is closely connected to electromagnetic properties of nuclear states and can be classified as 2^{λ}-pole charge ($E\lambda$) or magnetization ($M\lambda$) distribution. The degree of mixture between electric and magnetic radiation is given by the mixing ratio δ, defined as $\delta = \langle j_f \| \lambda' \| j_i\rangle / \langle j_f \| \lambda \| j_i\rangle$. In this work, we follow the δ signal convention of Krane and Steffen [21], in accordance with the used in the nuclear data sheet tables [22].

The gamma-ray angular distribution $W(\theta)$ [19] from an oriented state is expressed as follows,

$$W(\theta) = \sum_k^{k_{\max}} A_k\left(j_i \lambda \lambda' j_f\right) P_k(\cos\theta) \qquad k_{\max} = \min\{2j_i,\ \max\{2\lambda, 2\lambda'\}\} \tag{2}$$

$$A_k\left(j_i \lambda \lambda' j_f\right) = \rho_k(j_i)\frac{1}{1+\delta^2}\left[F_k\left(j_f \lambda \lambda j_i\right) + 2\delta F_k\left(j_f \lambda \lambda' j_i\right) + \delta^2 F_k\left(j_f \lambda' \lambda' j_i\right)\right] \tag{3}$$

$$F_k\left(j_f \lambda \lambda' j_i\right) = (-1)^{1+j_i-j_f}\sqrt{(2j_i+1)(2\lambda+1)(2\lambda'+1)} \times \langle \lambda 1 \lambda' -1|k0\rangle W\left(j_i j_i \lambda \lambda'; k j_f\right) \tag{4}$$

where $P_k(\cos\theta)$ is the Legendre polynomial; j_i and j_f are the spins of the initial and final states; λ and λ' are the multipole order of the radiation; δ is the mixing ratio of the electromagnetic transition; $W(j_i j_i \lambda\lambda'; k j_f)$ are the six-symbol Racah coefficients and $\rho_k(j_i)$ is the statistical tensor given by Eq. (1).

Eq. (2) is normalized to take into account an isotropic angular distribution ($k = 0$), so the quantities $A_0(j_i \lambda\lambda' j_f)$ and $P_0(\cos\theta)$ are equal to 1. $W(\theta)$ describes the gamma-ray intensities dependency with the observation angle and usually varies between 0 and 2.

An isotropic angular distribution happens in the special cases of $j = 0$ and $j = 1/2$ as can be seen from Eq. (2), since for $j = 0$ no orientation exists and a $j = 1/2$ state has only two substates $m = \pm 1/2$. In general, an aligned oriented state with population parameters $P(m)$ has the same statistical tensor $\rho_k(j)$ for even k as the inversely oriented state with population parameters $P(m) = P(-m)$. Therefore the angular distribution for both states is equal due to the invariance under space inversion.

It is possible to have a total or partial alignment of the nuclear substates m. In total oblate alignment, $\rho_2(j) < 0$ and the population of the substates are $P(m = 0) = 1$ or $P(m = \pm 1/2) = 1$ while in the total prolate alignment, $\rho_2(j) > 0$ and $P(m = \pm j) = 1$. In the case of a partial alignment, the population of the substates m can be approximated by a Gaussian [23] given by:

$$P(m) = \frac{\exp(-c^2/2\sigma^2)}{\sum_{m'=-j}^{j}\exp\left(-m'^2/2\sigma^2\right)} \tag{5}$$

where $c = m$ for oblate alignment and $c = j - |m|$ for prolate alignment; σ is the Gaussian width, usually described as a $\sigma = constant^* j$.

The case $\sigma \to 0$ corresponds to complete alignment while the value for σ in the partial alignment depends mainly on the nuclear reaction type and on the incident beam energy. To determine if a state is totally aligned in indirect reactions, it is necessary to position a particle detector near 180° and perform a coincidence method and correct the angular distribution due to correlations [24]. If a particle is detected in this angle, in coincidence with the gamma-ray, the substate m populated is the one from the total alignment.

3. Results and discussion

A routine to calculate $W(\theta)$ was developed using MATLAB® [25] to consider different nuclei input parameters such as the initial (j_i) and final (j_f) state spins, the transition multipolarities and subsequently the transition mixing ratio (δ). In the mixed transitions, the experimental δ was used when known and the experimental error of this value was