On the measurement of quadrupole moments of radioactive nuclei

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Abstract. Electric quadrupole moments provide a direct insight on the single-particle structure or the collective nature of a nuclear state. This article presents a short review on some of the experimental methods available to measure quadrupole moments with emphasis on the reorientation technique in Coulomb excitation and its use in combination with radioactive ion beams.

1. Introduction
The description of a nucleus is a hard to accomplish task. We are dealing with a finite many-body system, formed by fermions, governed by quantum-mechanics and interacting via a complicated force. Ideally, one would like to determine the wave function for each energy eigenstate of this system. However, apart from nuclear properties which are very intuitive such as its mass, radius, and nucleon separation energies, the experimental information that one usually gets from the nucleus, consist on the measurement of expectation values of one- and two-body operators. The challenge is then; try to model the complex dynamics of a nucleus so to reproduce the experimental results in a consistent way. The electric quadrupole moment of the first $2^+$ state in even-even nuclei represents a precise comparison point for testing the validity of nuclear models. The combination of well established experimental techniques with new technologies in detection systems and important improvements in the radioactive ion beam production, have allowed to obtain new experimental information on the electromagnetic matrix elements of exotic nuclei that can help to understand the evolution of the nuclear structure towards nuclei far from stability.

The first experimental information that one would like to know about the nucleus are its energy eigenvalues. Unfortunately there are many theoretical Hamiltonians that are able to correctly reproduce an energy spectrum using interactions that strongly differ. To select among these interactions one requires more sensitive experimental information. In principle, electromagnetic transition probabilities that connect two energy states in a single nucleus constitute a more stringent test for theoretical models. However, in order to gain a better understanding on the single-particle structure or the collective nature of a nuclear state, quantities that depend on the properties of two eigenstates $\langle f | H_{EM} | i \rangle$ are not the best alternative, since they may carry compensatory errors in $|i\rangle$ and $|f\rangle$ but still agree with the experimental value. Quantities that involve the one state that is been investigated, and
depend on a known interaction, \( \langle f | H_{EM} | f \rangle \), are evidently a more suitable probe in this respect. Examples of these quantities are the magnetic dipole and electric quadrupole moments.

The determination of the electric quadrupole moment of an excited nuclear state is based, directly or indirectly, on the measurement of its interaction with an electric field gradient (EFG). Very different experimental techniques have been developed over the years in order to achieve its measurement (see, for example, [1]-[5]). This paper gives a short overview on some of those techniques with emphasis on their applicability to the study of exotic nuclei. Recently, the availability of Radioactive Ion Beams (RIBs) [6]-[12] has widened up the boundaries in nuclear research. For example, the study of light nuclei that are located near the drip-lines, has resulted in the observation of phenomena that traditional nuclear models are not able to predict [13]-[16], challenging our understanding of the nucleus. The experimental information on radioactive nuclei is providing extremely valuable insight about the relevant interactions that dictate the behavior of the nucleus when reaching the limits of nuclear stability.

2. Nuclear moments

The interaction energy of an external potential \( V(r) \) with a nuclear charge distribution \( \rho(r) \) can be written as a product:

\[
E_Q = \sum_{\lambda, \mu} (-)^\mu \langle \mathbf{T}^{(\lambda)}_\mu \rangle V_{\mu}^{(\lambda)},
\]

where \( \mathbf{T}^{(\lambda)}_\mu \) are the multipole tensors of \( \rho(r) \) and \( V_{\mu}^{(\lambda)} \) are the tensor components of the external field. The multipole tensors \( \mathbf{T}^{(\lambda)}_\mu \) are classified as electric if its parity is \( \pi = (-)^\lambda \) and as magnetic if \( \pi = (-)^{\lambda+1} \). The well-defined parity and time-reversal properties of the nuclear states, \( |\Psi\rangle \), implies that the diagonal matrix elements \( \langle \Psi | \mathbf{T}^{(\lambda)}_\mu | \Psi \rangle \), vanish unless the total angular momentum of the state \( J \leq \lambda/2 \) and exists only for \( \pi = +1 \). Non-vanishing multipoles are: \( E0 \), electric monopole (a number); \( M1 \), the magnetic dipole (a vector); \( E2 \), electric quadrupole (a rank-2 tensor); etc. Note that this selection rules do not allow to measure the quadrupole moment of nuclear states having \( J = 0 \) or \( 1/2 \).

In practice we measure a single number for each moment, which is called spectroscopic quadrupole moment, and is given by the expectation value of the zero component of the rank-2 tensor

\[
Q_s(J) = \langle J, m = J | \mathbf{T}^{(2)}_\mu | J, m = J \rangle = \left[ \frac{16\pi}{5} \right]^{1/2} \langle J, J | r^2 Y_{20}(\theta, \phi) | J, J \rangle
= - \left[ \frac{16\pi}{5} \right]^{1/2} \frac{J(2J + 1)}{(2J + 1)(2J + 3)(J + 1)} \left( \frac{2J + 1}{2J + 3} \right)^{1/2} M_{JJ}^{(2)}
\]

where, in general for the \( \lambda \)-moment one has

\[
\langle J_s M_s | i^\lambda \lambda Y_{\lambda\mu} | J_r M_r \rangle = \langle J_s M_s | \mathcal{M}(E\lambda, \mu) | J_r M_r \rangle
= (2J_s + 1)^{-1/2} \langle J_s \lambda, M_r \mu | J_s M_s \rangle M_{rs}^{(\lambda)}
\]

with the reduced matrix element

\[
M_{rs}^{(\lambda)} = \langle J_s || i^\lambda \lambda Y_{\lambda} || J_r \rangle \quad \text{and} \quad M_{rs}^{(\lambda)} = (-)^{J_r - J_s - \lambda} M_{rs}^{(\lambda)}.
\]

The expectation value of the four remaining components of \( T^{(2)} \) can be obtained using the Wigner-Eckart theorem.
In the case that the nuclear charge distribution has an axial symmetry, the spectroscopic quadrupole moment can be directly related to the intrinsic quadrupole moment $Q_0$ and to the nuclear deformation by the expression

$$Q_s(J) = 3K^2 - J(J + 1) \frac{2J + 3}{(2J + 1)(2J + 3)} Q_0$$

where $K$ is the projection of the total angular momentum of the nucleus along the deformation axis, and $Q_0$ is related to the deformation parameter $\beta$ through the relation

$$Q_0 = \frac{3}{5\pi} ZR^2 \beta (1 + 0.36\beta), \quad \text{for} \quad R = R_0 \ A^{1/3}.\quad (6)$$

3. Experimental methods

A practical classification of the methods that are available in the laboratory to measure electric quadrupole moments can be obtained by looking at the way the EFG is produced at the nuclear site. For many years, the values of EFG produced by external electrodes in the laboratory were too small to yield a measurable effect when interacting with a charge distribution of nuclear dimensions. As an alternative, the EFG produced by electrons in atomic or molecular environments, or by other charged particles (such as muons or kaons in bound orbits), or produced during the bombardment of a target nucleus with energetic ions were used. This section presents a brief outline of these experimental methods.

3.1. Atomic or molecular environments

In a solid, electric field gradients of up to a few $10^{18}$ V/cm$^2$ can be obtained from atomic or molecular charges. The hyperfine interaction between this kind of EFG and the quadrupole moment of an excited nuclear state can be measured from: (i) the magnitude of the hyperfine splitting in atomic transitions (energy differences), (ii) the Mössbauer effect (in case of low-energy gamma transitions $E_\gamma \leq 130$ keV); (iii) the attenuation of the angular distribution of deexcitation gamma rays.

3.1.1. Mössbauer effect

When a nucleus is in a solid, fixed within a lattice, there is considerable probability that it can absorb and emit $\gamma$-rays without energy loss going into the recoiling nucleus (Mössbauer effect [17], [2]). The recoil energy and Doppler broadening of the $\gamma$-ray are characterized by the mass and random velocity of the whole crystal and are therefore completely negligible. This high precision resonant absorption effect can be used to investigate very small changes in the nuclear energy levels that result from the hyperfine interactions between the nucleus and its electronic environment.

The probability of such a recoilless event depends on the energy of the gamma-ray, the temperature and vibrational properties of the solid. In general, the Mössbauer effect is optimized for low-energy gamma-rays and low temperature lattices.

The finite energy spread of the Mössbauer gamma ray can be related to the lifetime of the nuclear excited state. Whereas the interaction of the nuclear quadrupole moment with the EFG gives rise to a splitting of the nuclear energy levels that correspond to different alignments of the quadrupole moment with respect to the principal axis of the EFG.

The main drawback of this kind of measurement is that the electric quadrupole moment is a function of both nuclear and electronic properties combined in such a way that independent quantitative information on both properties cannot be obtained. The determination of the nuclear quadrupole moment should then rely on a calculation of the EFG at the nuclear site.
3.1.2. Perturbed angular correlations  The angular correlation of a cascade \( I_i \rightarrow I \rightarrow I_f \) is altered as soon as the nucleus in the intermediate level \( I \) is subject to a torque, due to the interaction of its electric quadrupole moment with the EFG that surrounds the decaying nucleus \([4],[18]\). If the quantization axis is chosen to coincide with the direction of emission of the first radiation, the interaction will change the relative angle between these two axis (quantization-emission) inducing transitions among the \( m \)-states. The second radiation is therefore emitted from a level with an altered \( m \)-population distribution and this will cause an attenuation of the angular correlation.

When the extra-nuclear field is static a semiclassical picture is suitable, the population of the \( m \)-states quantized along the direction of the first emission remains constant and its change in time is due to the precession of the nucleus around the symmetry axis. However, when the interaction is time-dependent, the direction of the EFG at the position of each nucleus changes continuously in a random manner, no quantization axis exists for which the population of the \( m \)-states remains constant. Consequently all the \( m \)-states are equally populated for any choice of the quantization axis and the directional correlation becomes isotropic.

The magnitude of a static perturbation can be measured by the precession frequency:

\[
\omega_Q = -\frac{e Q V_{zz}}{4I(2I-1) \hbar}
\]  

(7)

As a raw criterion \([4]\), if \( \tau \) represents the mean life of the intermediate level \( I \), the cascade will be perturbed if \( \omega\tau \geq 1 \). For time-dependent perturbations one has that the \( m \)-states approach to an uniform population exponentially, with a relaxation constant \( \lambda \), so the criterion to have perturbation in the angular correlation becomes \( \lambda\tau \leq 1 \).

3.1.3. \( \beta \)-NMR on polarized beams  In this method, a polarized beam of radioactive ions is implanted into a crystal, characterized by a well known electric field gradient and oriented along the direction of a static magnetic field \([5],[19]-[21]\). A variable radio frequency magnetic field, applied perpendicular to the static field, is used to scan around the Larmor frequency, giving rise to equidistant magnetic resonances in the \( \beta \)-decay. The distance between resonances gives a measurement of the product of the nuclear quadrupole moment and the electric field gradient at the nucleus site.

Several techniques are used to produce spin-oriented radioactive ion beams. One of them is the optical pumping using collinear polarized laser light. Another method is the use of nuclear reactions such as: fusion-evaporation, projectile fragmentation, Coulomb excitation and transfer reactions.

Most these experiments determine the interaction energy, \( i.e. \) the product of the quadrupole moment and the EFG, and therefore to obtain meaningful information about the nuclear quadrupole moment an independent knowledge of the EFG is required.

3.2. Muonic X-rays  Negative muons are usually obtained from the decay of negative pions in flight \([22]\). To stand a chance of been captured into an atomic orbit, muons must be slowed down to energies \( \leq 2 \) keV so that their velocity becomes lower than the velocity of the valence electrons. The initial distribution of states is not well known, however, a muon orbit of the same size as the electron \( K \)-orbit \((n_e = 1)\) will have a principal quantum number

\[
n_{\mu} \sim \left( \frac{m_\mu}{m_e} \right)^{1/2} \sim 14,
\]  

(8)

As all muonic states are unoccupied, the muon will cascade down from \( n_{\mu} = 14 \) towards lower energies, until reaching the most deeply bound states: 1s \((K\)-shell\) and 2p \((L\)-shell\). The close
proximity of these states (muon K-shell is $\sim$ 207 times closer to the nucleus than an electron K-shell) allow muons to probe the nuclear volume sensitively. The interaction strength of the muon with the electric quadrupole moment of the nucleus varies proportional to $r^{-3}$, and therefore gets enhanced for low energy states.

The influence of the nuclear electric quadrupole moment in a muonic atom is manifested as an energy splitting of its muonic X-ray lines. To extract the nuclear quadrupole moment from this energy difference one needs to use a model for both, the Hamiltonian that describes the muonic-atom and the nuclear charge distribution [3].

### 3.3. Reorientation in Coulomb excitation

Coulomb excitation occurs when a nucleus undergoes transitions to its excited states due exclusively to the time dependent electromagnetic interaction acting between target and projectile as they approach to each other in a collision. This mechanism is extremely useful because its cross section can be accurately related to the electromagnetic properties of the nucleus, such as its reduced electromagnetic transition probabilities and moments [23, 24].

A simplified theoretical evaluation of the Coulomb excitation process can be obtained by assuming that the projectile follows closely a classical hyperbolic trajectory (semi-classical theory). The differential cross section for finding the nucleus in a state $f$ after the collision can be written as the product of the Rutherford differential cross section in the center-of-mass system, $d\sigma_R/d\Omega$, and the excitation probability, $P_{i\rightarrow f}$

\[
\frac{d\sigma_{i\rightarrow f}}{d\Omega} = \frac{d\sigma_R}{d\Omega} P_{i\rightarrow f},
\]

This probability can be evaluated using a perturbation approach. To estimate the perturbation order that is suitable in each case it is useful to calculate the parameter $\chi^{(\lambda)}_{i\rightarrow f}$, that measures the strength of the interaction with which the state $J_i$ is coupled to state $J_f$ by multipole order $\lambda$:

\[
\chi^{(\lambda)}_{i\rightarrow f} = \frac{(16\pi)^{1/2}(\lambda + 1)!}{(2\lambda + 1)!!} \frac{Z_2 e}{\hbar v_i v_f} \langle J_f|M(E\lambda)||J_i \rangle a_{ij}^{\lambda} (2J_i + 1)^{1/2},
\]

where $2a_{ij}$ is the symmetrized distance of closest approach in a head-on collision. To first-order in perturbation theory, the transition amplitudes are proportional to $B(E\lambda; J_i \rightarrow J_f) = (2J_i + 1)^{-1} |\langle J_i|M(E\lambda)||J_f \rangle|^2$, the matrix elements of the electric quadrupole operator between the initial and the final excited state. To second-order, the excitation of the state $|f\rangle$ will also be influenced by the static quadrupole moment of $|f\rangle$, which is proportional to the diagonal element $\langle J_f|M(E\lambda)||J_f \rangle$. The change on the excitation observables introduced by the static quadrupole moment of the excited state $f$ is called “reorientation effect” [1, 25].

There are several experimental possibilities for measuring reorientation, that can be divided in: (i) absolute measurements, which require the independent determination of both the $B(E2)$ values and the absolute cross section, and (ii) relative measurements, that consist on the comparison of the excitation probabilities for two different bombarding conditions, such as: different reaction partners, different bombarding energies or different scattering angles. To choose the best conditions for a reorientation experiment one has to consider the way the quadrupole moment affect the measured quantity as well as the accuracy and ease with which such an experiment can be performed.

For the particular case of an even-even nucleus the probability of a $0^+ \rightarrow 2^+$ transition can be written as:

\[
P_{02} = P_{02}^{(11)} + P_{02}^{(12)} + P_{02}^{(22)},
\]
where the first order term is given by
\[ P_{02}^{(11)} = \frac{1}{5} \left[ \chi_{02}^{(2)} \right]^2 \sum_{\mu} \left[ \mathcal{K}_{2\mu}(\theta, \xi_{02}) \right]^2, \tag{12} \]
the second term in (11) is an interference term
\[ P_{02}^{(12)} = \frac{1}{\sqrt{5}} \left[ \chi_{02}^{(2)} \right]^2 \chi_{22}^{(2)} \sum_{\mu} \left[ \mathcal{K}_{2\mu}(\theta, \xi_{02}) B_{2\mu}^{(22)}(\xi_{02}, 0, \theta) \right]^2, \tag{13} \]
and term in (11) can be neglected as it becomes important only if the interference between first- and third-order is also important. Functions \( \mathcal{K}_{\lambda \mu} \) and \( B_{\lambda \mu}^{(\lambda_1 \lambda_2)} \) are defined in equations 15 and 27 of Ref. [1]. In all the previous equations \( \theta \) is the scattering angle of the projectile in the centre of mass, and \( \xi_{if} \) is the adiabaticity parameter that gives the ratio of the collision time \( \tau_{\text{coll}} = a/v \) relative to the period of the nuclear transition \( \tau_{\text{nuc}} = \hbar/\Delta E \) and is given by
\[ \xi_{if} = \frac{Z_1 Z_2 e^2}{\hbar} \left( \frac{1}{v_i} - \frac{1}{v_f} \right). \tag{14} \]
In terms of the reorientation coefficient
\[ r \equiv P_{02}^{(12)} / P_{02}^{(11)} \approx \frac{A_1}{Z_2} \frac{\Delta E}{1 + A_1/A_2} \langle 2^+ || M(E2) || 2^+ \rangle K(\theta, \xi_{if}) \tag{15} \]
the excitation probability can be written as
\[ P_{02} \approx P_{02}^{(11)} (1 + r), \tag{16} \]
where
\[ K(\theta, \xi) = 1.135 \frac{\sum_{\mu} \mathcal{K}_{2\mu}(\theta, \xi_{02}) B_{2\mu}^{(22)}(\xi_{02}, 0, \theta)}{\xi \sum_{\mu} \left[ \mathcal{K}_{2\mu}(\theta, \xi_{02}) \right]^2}. \tag{17} \]
It is important to notice that the reorientation coefficient, \( r \), is linear on both the excitation energy \( \Delta E \) and the mass of the projectile, whereas the probability (16) depends on both the \( B(E2) \) and the \( Q \) values. From this last observation follows that, a challenge bound to any reorientation experiment is to try to single out a small interference term in the presence of a large first-order term, and this requires an understanding of all the processes that may influence the measurement.

One way to get around the strong dependency on the \( B(E2) \) value is by measuring two excitation probabilities for different bombarding conditions \( \alpha \) and \( \beta \), and taking the ratio
\[ \frac{P_{\alpha}}{P_{\beta}} \approx \frac{P_{\alpha}^{(11)}}{P_{\beta}^{(11)}} \frac{1 + r_\alpha}{1 + r_\beta} \approx \frac{P_{\alpha}^{(11)}}{P_{\beta}^{(11)}} (1 + r_\alpha - r_\beta), \tag{18} \]
that cancels out large uncertainties that may arise form an absolute calibration. The deviation of the experimental ratio \( P_{\alpha}/P_{\beta} \) from its calculated value is proportional to \( (r_\alpha - r_\beta) \), which is proportional to the quadrupole moment of the excited state.
4. Reorientation experiments with radioactive ion beams
In recent years we have been interested in using the reorientation effect to measure the electric quadrupole moment of the neutron-rich nucleus $^{78}$Ge. The last systematic study for the stable $Z = 32$ isotopes were performed by a JAERI group using Multiple Coulomb Excitation [26]. Their results confirm the observation that the values of the diagonal matrix element $\langle 2 \frac{1}{2}^+ | |M(E2)| | 2 \frac{1}{2}^+ \rangle$ evolve from a very small but positive value in $^{70}$Ge towards small but negative values in $^{72,74,76}$Ge [27]. The question we would like to answer with a measurement in $^{78}$Ge is how does the quadrupole moment evolves as we move towards neutron-rich nuclei?

We chose bombarding energies below the Coulomb barrier to guarantee that the observed nuclear excitation was due to a pure Coulomb interaction. The RIBs of $^{78}$Ge were produced using the Isotope Separation On-Line (ISOL) technique at the HRIBF Facility in Oak Ridge National Laboratory (ORNL) [6]. To single out the reorientation effect from the presence of the large first-order term we performed a relative measurement using two different target materials: $^{12}$C and $^{24}$Mg mounted on the same holder.

The Coulomb excitation cross section of the projectile-like nuclei was obtained from the coincidence yield of a de-excitation $\gamma$-ray detected in the HPGe segmented array CLARION [28] and a target-like nucleus identified by a large angular coverage charged-particle array, Bare-Ball [29], based on CsI(Tl) crystals coupled to Si-photodiodes. The use of the powerful analysis codes GOSIA and GOSIA2 [30] was extremely important in this analysis, in order to achieve a careful balance a large number of effects that came into play in the measurement. A detailed discussion on the analysis procedure and final results of this analysis will be reported in a forthcoming publication [31].

5. Conclusions
The experimental techniques mentioned here are examples of well established methods to determine electric quadrupole moments of nuclear excited states. In the last years some of these techniques have been combined with new detection systems and applied to the study of exotic nuclei using radioactive ion beams.

Quadrupole moments are a very sensitive probe of the interplay between macroscopic and microscopic effects present in a nuclear wave function and can help to understand changes in the nuclear shell structure when approaching to large values of the proton-to-neutron ratio.

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