Measuring precise nuclear moments by studying isolated molecules: new experimental data and accurate electronic theory meet

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Nuclear quadrupole moments (Q), a measure of charge asymmetry in nuclei, are essential as benchmarks for nuclear structure models and to interpret experimentally determined nuclear quadrupole interactions in terms of electronic and molecular structure. Experimentally accessible via the quadrupole interaction frequency $\nu_Q = eQV_{zz}/h$ one needs to know V_{zz}, the electric-field gradient (EFG) at the nuclear site, whose value must come from theory, to date only precise enough for free atoms or small molecules. Experimental ν_Q data in such environments are, however, only available for nuclear ground states. On the other hand, studies of short-lived nuclear excited states had up to now only been possible in solids, where theory is yet not sufficiently accurate.

Using the pure radioactive isotope beams available at the ISOLDE-CERN (Isotope mass Separator On-Line facility) and chemically combining into halide molecules, the solid-state technique of γ-γ perturbed angular correlation was for the first time applied to measure ν_Q for nuclear excited states in isolated small linear molecules (Cd and Hg halides).

The technical challenge was the production of a highly diluted gas sample of the radioactive molecules such that no intermolecular collisions occur during the measurement time, typically below 100 ns, depending on the half-lives of the intermediate nuclear state of the radioactive isotope used.

Combined with state-of-the-art ab initio electronic structure calculations of V_{zz} for these small molecules, the nuclear quadrupole moment Q has been obtained from the measured ν_Q for two excited states, ^{111}Cd(5/2+) and ^{199}Hg(5/2-), and its value confirmed with different compounds. In the past, numerous solids were investigated by PAC and other solid-state methods and the Q value now obtained allows quoting more precise values for the EFG. One might hope that the presently not very accurate solid-state theory calculations can be improved by comparison with reliable experimental data.

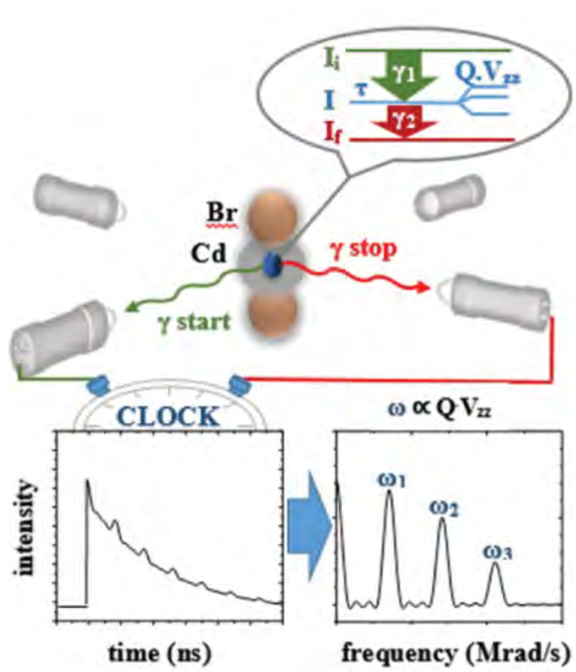

FIGURE 1
Schematic explanation of PAC measurement on a free molecule. The Cd nucleus (dark blue) with quadrupole moment Q interacts with the electric-field gradient of the electrostatic potential at the nucleus $V_{xy}=\partial^2 \Phi/\partial x \partial y$ (EFG) created by the Cd electron distribution (grey) modified by bonding to the bromine atoms (brown). This is observed by the modulation of the time-distribution of the two rays emitted in decay of radioactive ^{111m}Cd. The energy splitting of the nuclear intermediate state (due to quadrupolar coupling) is directly seen as transition frequencies (ω) in the Fourier transform.

Reference

H. Haas et al., Free molecule studies by perturbed γ-γ angular correlation: A new path to accurate nuclear quadrupole moments. Phys. Rev. Letters 125, 253001 (2021)