ELECTRIC FIELD GRADIENT: BRIDGING EXPERIMENT AND COMPUTATIONS

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Atomic structures are often treated as known quantities, obtained either from experiment or from computation. In some systems, however, this assumption breaks down. Th-doped CaF_2, relevant for nuclear clock applications, is one such case: the local environment of Th remains poorly characterized despite significant experimental and theoretical effort. Total energy considerations alone are insufficient to resolve competing structural models, particularly in the presence of charged defects and ambiguous chemical compositions.

In this work, we explore the use of electric field gradients (EFG) as an alternative, highly sensitive probe of local structure. Combining embedding cluster models with high-level wavefunction methods (CASSCF/CASPT2), implemented in Molcas [1], we show how EFG can provide direct insight into local symmetry and serve as a stringent test of computational models. **We also discuss key mathematical properties of the EFG tensor—such as its traceless property, symmetry, and sensitivity to small perturbations—which underlie its usefulness as a structural probe.** We further highlight a less obvious issue: different implementations of EFG may differ by sign due to convention, leading to systematic discrepancies across codes. Understanding and resolving such subtleties is essential when bridging experiment and computation. Finally, the EFG values can be used for quality control for various embedding models [3].

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