Two-Level States in Substitutional Mixed Crystal: Ab Initio Predictions for Glass Transitions

J. Lee and T. A. Arias

Laboratory of Atomic and Solid State Physics, Cornell University, Ithaca, NY, USA.

The nature of glass transitions has been a subject of great interest in solid state community due to their universal characteristics occurring for a large range of materials. Among many explanations for the origin of this universality, the two-level states (TLS) theory [1, 2] stands as the simplest but most successful one. However, the identities of the two-level states have been confirmed for only few systems, and even so, only at the level of molecular dynamic simulation, due to a large computational cost of ab initio calculations.

Here we present an ab initio investigation for the nature of the two-level states in one of the substitutional mixed crystals observed experimentally to be a glassy material [3], Ba$_{1-x}$Ca$_x$F$_2$. We determine the underlying mechanism of the formation of the two-level states, which is the symmetry breaking in crystal structure driven by chemically induced local strains. A modulated Landau theory reproduces all the ab initio simulation results, and leads us to a prediction for the spectral density of states in very good agreement with the experiment.


Contact: jl572@cornell.edu