Low temperature NMR relaxation and rattling phonons in type-I $Ba_8Ga_{16}Sn_{30}$ clathrates

Xiang Zheng[a, b], Sergio Y. Rodriguez[a], and Joseph H. Ross, Jr.[a, b]

[a]Department of Physics and Astronomy, Texas A&M University, College Station, TX 77843-4242, USA
[b]Material Science and Engineering Program, Texas A&M University, College Station, TX 77843-3003, USA

The atomic motion of guest atoms inside the cage of semiconductor clathrates is considered as one of the important sources responsible for the very low thermal conductivity. ^{69}Ga and ^{71}Ga Nuclear Magnetic Resonance (NMR) studies on type-I $Ba_8Ga_{16}Sn_{30}$ clathrates show a clear low temperature spin-lattice relaxation peak attributed to the influence of Ba rattling dynamics on the framework-atom resonance. Analysis indicates that an electric quadrupolar relaxation mechanism due to atomic motion is the leading contribution. The data are analyzed using a two-phonon Raman process, according to a recent theory involving a localized one dimensional anharmonic oscillator model potential. Excellent agreement is obtained using this model, with the parameters corresponding to a double well potential with very large anharmonicity. We have extended the theory to include a simplified two dimensional anharmonic well. In both models the best fit parameters correspond to very similar average displacement for the cage-center Ba atoms. We also examine the heat capacity behavior using this model, and compare to previously reported results obtained using these and other models for the anharmonic oscillator potential, and to low-temperature two-level system behavior of disordered materials.