Diffusion in Al_4Sr and Ga_4Sr studied using PAC Spectroscopy

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Introduction

We studied atomic diffusion in highly ordered crystals using perturbed angular correlation spectroscopy (PAC). Measurements were made on Al_4Sr and Ga_4Sr phases, which have the Al_4Ba crystal structure, to complement earlier studies on Al_4Ba and In_4Ba. It was found that the new Al_4Ba phases exhibit large changes in signals attributed to diffusion, as in the earlier studies.

The Al_4Ba Structure

The Al_4Ba crystal has tetragonal symmetry with ten atoms per unit cell. There are two inequivalent Al-sites, labeled Al1 and Al2, and a Ba-site. Tracer atoms (^{111}In) inhabit the two Al-type sublattices. The two aluminum sites are arranged such that atomic diffusion within the one sublattice is energetically unfavorable. Instead, atomic jumps of atoms on an Al-site will be to the other Al-site. All local electric field gradients are collinear with the major V_{zz} component along the c-axis.

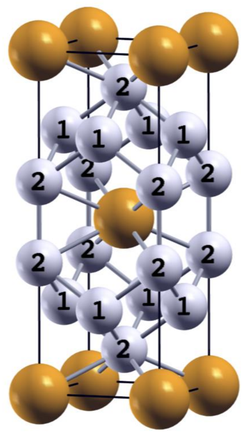

Figure 2: Prototype of Al4Ba crystal structure. The Ba-type sites sit on the unit cell corners and at the center. The Al-type sites are arranged in alternating layers with a body-centered structure.

Experimental method

Hyperfine interactions can be studied using PAC spectroscopy. This method detects the angular correlation of probe nuclei (here ^{111}In) as it is perturbed by extranuclear fields, specifically the electric field gradient (EFG) due to the local crystal structure.

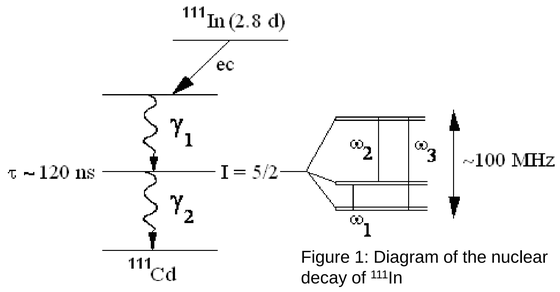

Figure 1: Diagram of the nuclear decay of ^{111}In

^{111}In radioactive probe nuclei decay and emit a cascade of gamma rays that exhibits an angular correlation. Stationary atoms are subject to a static nuclear quadrupole interaction; however, jumps of the probe atoms among different sites in the crystals leads to motional averaging of the quadrupolar precession.

For the Al_4Ba structure, NN atomic jumps among different sites result in changes in magnitude of the EFGs but not reorientation of EFGs due to the collinear nature of EFGs in this crystal structure.

Further information and acknowledgments

For previous research in which diffusion leads to changes in orientation of the EFG instead of magnitude, see Phys. Rev. Lett. **92**, 225901 (2004) and Phys. Rev. Lett. **102**, 155901 (2009).

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Experiments and Results

Samples of Al_4Sr and Ga_4Sr were made by arc melting high purity metals (>99.9%) and ^{111}In activity under argon gas and annealing at 400C for a day to equilibrate the samples. Fourier PAC spectra and temperature dependences of fitted fundamental quadrupole interaction frequencies are shown in the four figures below. Note that each quadrupole interaction signal has three frequency harmonics with 1:2:3 proportions.

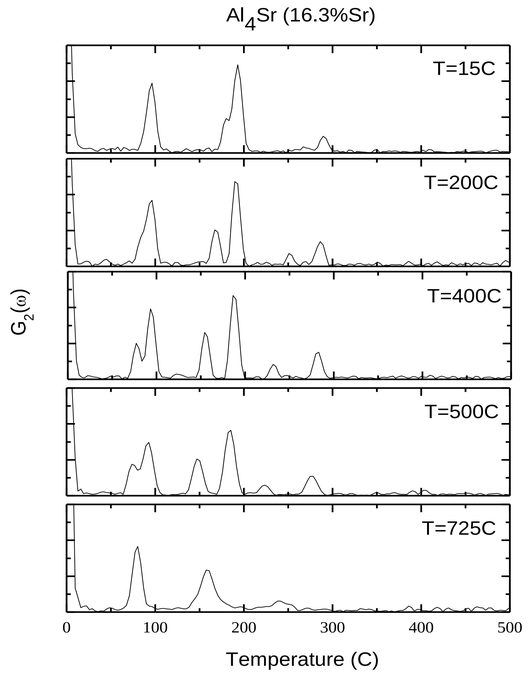

Figure 3: Frequency spectra of Al_4Sr at the indicated temperatures. The splitting and re-merging of the samples can be seen as the temperature is raised.

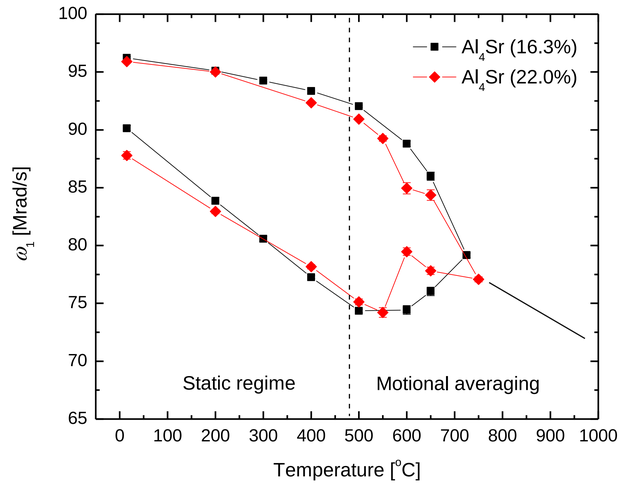

Figure 4: Fundamental quadrupole interaction frequencies as a function of temperature. The split between two temperature regimes is clearly visible; the Al_4Sr samples had diverging static signals at low temperature and motional averaging leading to merging of the signals at high temperature.

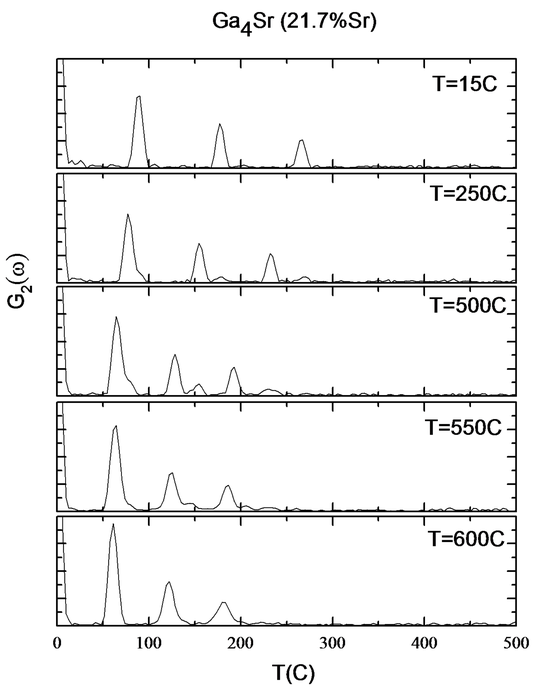

Figure 5: Fourier spectra of Ga_4Sr. The frequencies split statically below 500C and then merge dynamically above 500C. Amplitude of the high-frequency signal increases with temperature due to a temperature dependent site-preference.

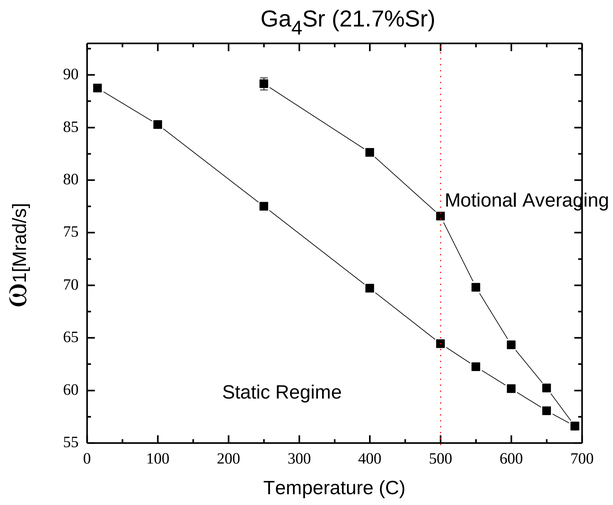

Figure 6: Fitted fundamental quadrupole interaction frequencies as a function of temperature. Static quadrupole interaction frequencies below 500 C give way to motionally averaged signals above 500 C.

^{111}In probe atoms occupy both Al-type sites in Al_4Ba-type structures. Spectra for Al_4Sr and Ga_4Sr were found to exhibit two signals at low temperature that appear be static , but which rapidly converge at temperatures above 500C and merge at ~700C. This is attributed to motional averaging of quadrupole interactions due to atoms jumping rapidly between the two sublattices. In Al4Ba and In4Ba, in contrast, a static regime could not be distinguished experimentally below much lower merging temperatures of about 400 C.