Dependence of diffusion on composition in pseudo-binary $La(In_{1-x}Sn_x)_3$ phases

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Motivation

Measurements were made of jump frequencies of tracer atoms in a series of phases $La(In_{1-x}Sn_x)_3$ having the $L1_2$ crystal structure. The goal was to determine how the average jump frequency of ^{111}In/Cd probe atoms is modified when indium atoms are replaced by tin solute atoms. Jump frequencies were determined from nuclear relaxation in quadrupole interaction signals caused by reorientation of local electric-field gradients, using perturbed angular correlation spectroscopy (PAC).

Background

PAC spectroscopy

Perturbed angular correlation spectroscopy is used to measure nuclear hyperfine interactions. It detects the angular correlation of probe nuclei (radioactive ^{111}In) as it is perturbed by extranuclear fields, specifically the electric-field gradient due to the local crystal structure.

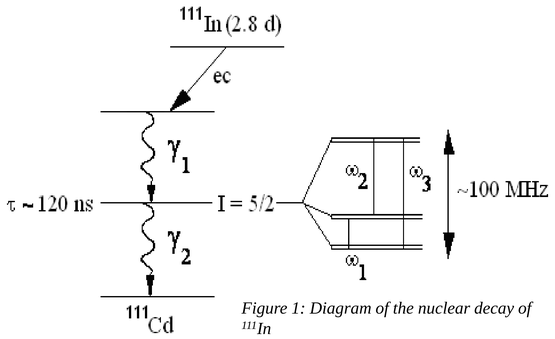


Figure 1: Diagram of the nuclear decay of ^{111}In

^{111}In radioactive probe nuclei decay and emit a cascade of gamma rays that exhibit an angular correlation.

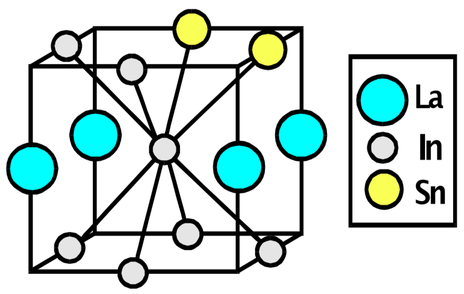
Stationary atoms are subject to a static nuclear quadrupole interaction; however, jumps of the probe atoms among different sites in the crystals lead to motional averaging of the quadrupolar precession. The relaxation appears as damping of the quadrupole interaction signals.

$L1_2$ Structure

$La(In_{1-x}Sn_x)_3$ forms a pseudo-binary compound with $L1_2$ crystal lattice structure. The atoms of the compound are all located on face centered cubic lattice sites.

The tin and indium atoms are assumed to be distributed at random, with figure 2 illustrating the local geometry for jumps from an indium site to any of eight neighboring sites. This configuration exhibits two randomly located tin solute atoms.

Figure 2: Example of the $L1_2$ structure with randomly distributed atoms



Experimental

Samples were made by arc-melting highly pure metals (>99.9%) and ^{111}In activity under argon. $La(In_{1-x}Sn_x)_3$ samples with compositions x = 0.13, 0.25, and 0.75 were created at the lanthanum-poor boundary composition. Measurements were made over the range 20 to 700°C.

It can be seen that the composition affects the amount of damping of the quadrupole interaction signals. The jump frequency λ is equal to the inverse of the relaxation time.

Figure 3: Quadrupole interaction perturbation functions measured at approximately 550 ℃

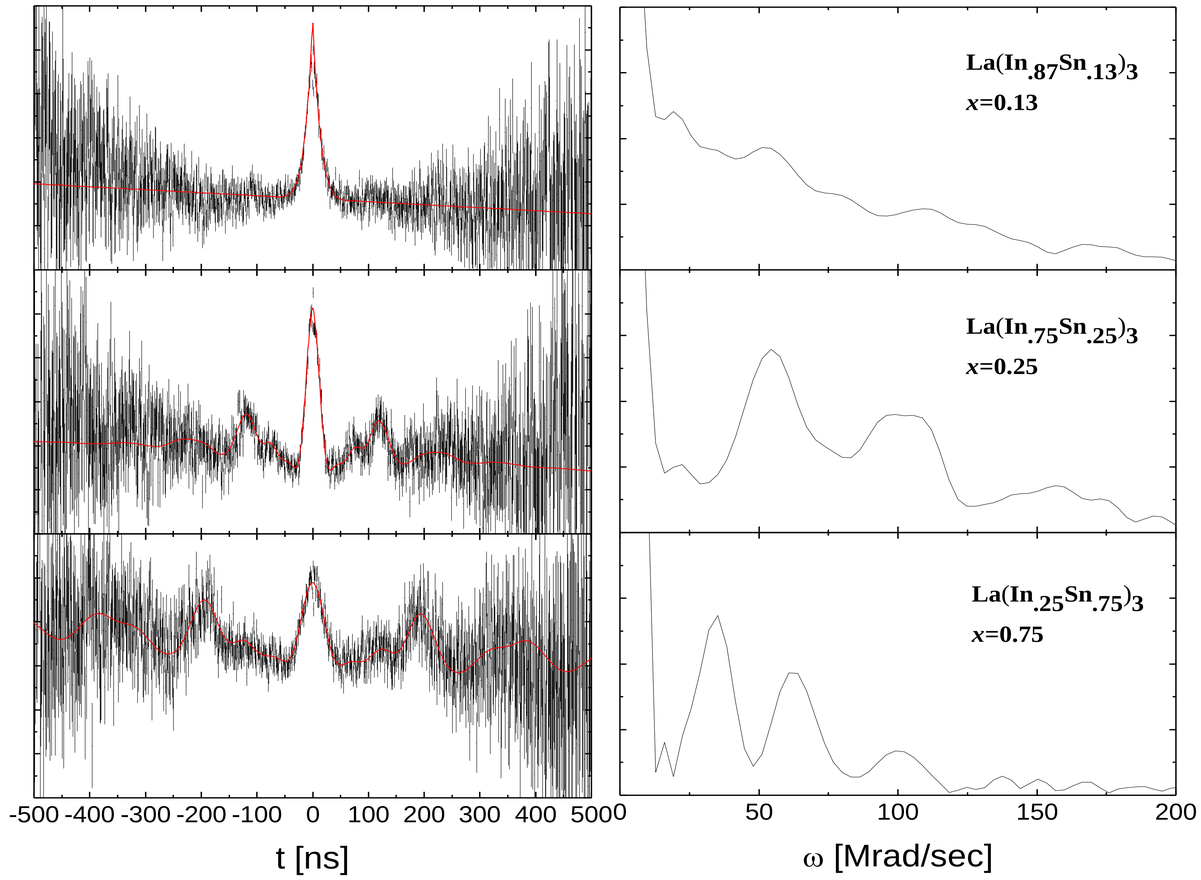
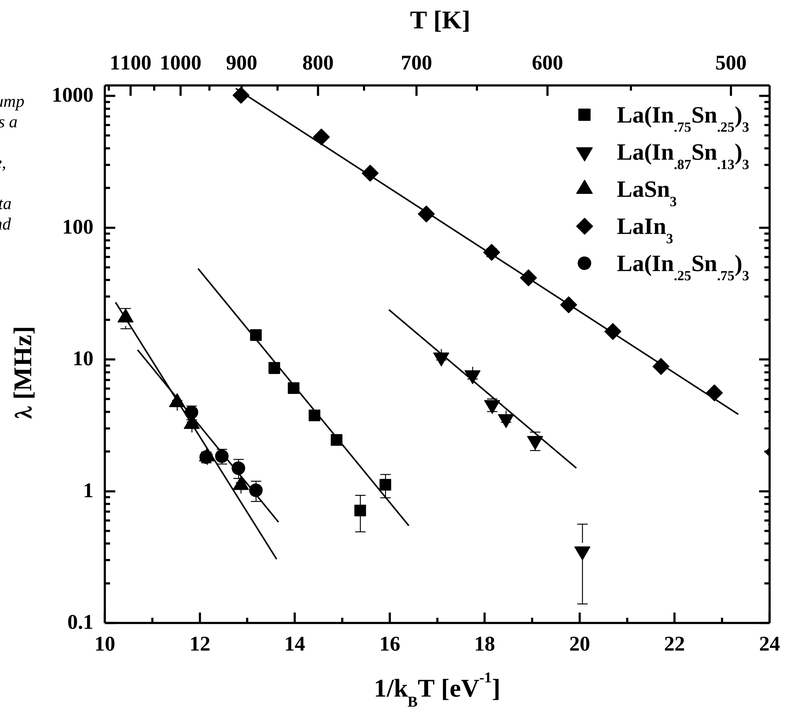


Figure 4: Fourier transforms of the perturbation functions measured at approximately 550 ℃

Conclusion

By comparing the damping of the quadrupole interaction signals, the effect of composition on the jump frequency λ is observed.

Figure 5: Jump frequency as a function of temperature, including previous data on $LaSn_3$ and $LaIn_3$



Replacement of indium atoms by even small numbers of tin atoms was found to cause drastic reduction in the jump frequency. Tin atoms appear to "block" jump paths of probe atoms on the indium-tin sublattice.

Work in Progress

This study is being continued at the lanthanum-rich boundary composition. Initial results suggest that an excess of lanthanum increases the jump frequency of the probe ^{111}In atoms.

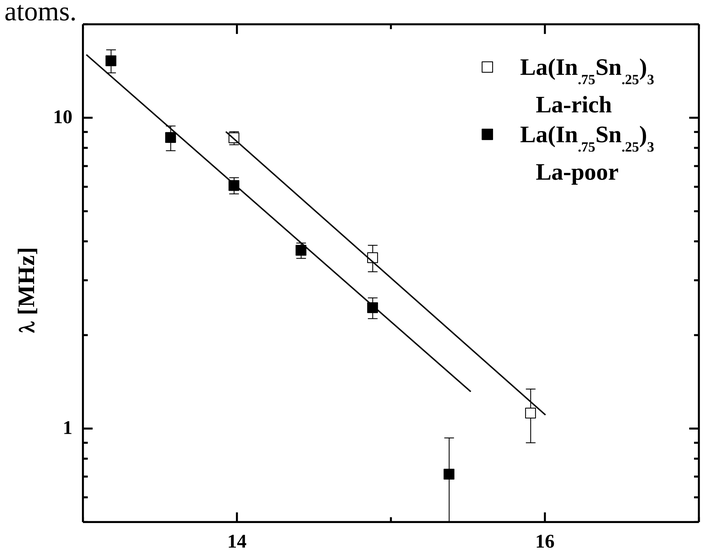


Figure 6: Jump frequency as a function of temperature, comparing lanthanum-poor and lanthanum-rich samples of otherwise similar compositions

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