A Nuclear Resonant Inelastic X-Ray Scattering Study of the Stabilization of bcc Fe by Magnon-Phonon Interactions

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Most of the entropy of materials comes from the vibrations of atoms, and my group has been studying how this vibrational entropy originates with the electrons and atomic structures of materials. At high temperatures, the vibrational entropy is more complicated than expected from harmonic models -- there is a hierarchy of non-harmonic behaviors from quasiharmonic, to anharmonic, to electron-phonon interactions in metals.

It is known that the spin-polarized electronic structure favors bcc Fe at low temperatures, but this bcc phase persists to surprisingly high temperatures. Phonon densities of states (DOS) curves were measured for bcc $^{57}$Fe between 30 K and 1184 K using nuclear resonant inelastic x-ray scattering (four are shown in panel a). All phonons shifted to lower frequencies with temperature, but the low transverse modes shifted especially rapidly above 700 K. Fits to the DOS curves using a Born - von Karman model showed that the second-nearest neighbor force constants had the most anomalous thermal trend (panel b). Panel c shows that the nonharmonic behavior followed the thermal trend of the magnetic entropy. This excess phonon shift makes a contribution of 35 meV/atom to the free energy at 1100 K [1]! A new ab-initio approach for quantitatively assessing effects of magnon-phonon interactions and lattice expansion showed that for some phonon branches, the magnon-phonon interaction is an order of magnitude larger than the phonon shift due to thermal expansion or anharmonicity [2]. The magnon-phonon interaction makes a surprisingly large contribution to the thermodynamic stability of bcc Fe at high temperatures.

(a) Four phonon DOS curves out of 38 measured by NRIXS at different temperatures. (b) Interatomic force constants for longitudinal forces between first and second-nearest neighbor Fe atoms. (c) Thermal trends of the magnetic entropy, nonharmonic part of the phonon entropy, and the magnetization curve of bcc Fe.