Lattice Dynamic in Bulk and Nanostructured CrSi2

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Résumé

Chromium disilicide, CrSi2, is a promising material for thermoelectricity. Its power factor is comparable to that of PbTe and its constituting elements meet the environmental and economic demands. Nevertheless, the high lattice thermal conductivity of CrSi2 limits its thermoelectric performance, preventing any use in thermoelectric modules. A strong decrease in the lattice thermal conductivity is observed on nanostructured CrSi2 obtained by ball milling [1]. This effect can originate from an increase in the phonon scattering at the grain boundaries but also from an increase in the point-defects concentration that further scatters the propagating acoustic phonons. A better understanding of the lattice dynamics in bulk and nanostructured CrSi2 is necessary to further optimize the phonon scattering process and reduce the thermal conductivity.

In this presentation we investigate in detail the effect of the grain size reduction and residual microstrains on the lattice dynamics. The phonon dispersion curves are measured on single-crystalline CrSi2 using inelastic neutron scattering, while the generalized vibrational density of states (GVDOS) are determined on bulk and nanostructured CrSi2. These experimental results are compared with DFT calculations. We show that the optical phonons contribute from 50 to 70% of the lattice thermal conductivity. The temperature variations in the GVDOS of CrSi2 follow a quasi-harmonic behavior, which explains the rather large

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lattice thermal conductivity measured on the single-crystals. In addition, the GVDOS of nanocrystalline CrSi2 evidences a spectral weight transfer at low energy, which is related to a decrease of the Debye temperature and the sound velocities. These observations explain the decrease in the lattice thermal conductivity observed in nanostructured CrSi2 [2].

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