Experimental report

14/10/2015

Proposal: 7-01-374 **Council:** 10/2012

Title: Lattice dynamics in MxV2Al20 compounds

Research area: Physics

This proposal is a continuation of 7-01-342

Main proposer: **Hannu MUTKA**

Experimental team: Hannu MUTKA
Z. HIROI

Local contacts: Michael Marek KOZA
Hannu MUTKA

Samples: MxV2Al20 M=Al, Ga, Sc, Y, La, Ce

Instrument	Requested days	Allocated days	From	To
IN4	4	4	02/05/2013	06/05/2013
IN5	2	2	20/02/2013	22/02/2013

Abstract:

We wish to continue the investigations on the lattice dynamics of the MxV2Al20 compounds started on IN4 and IN6. We have found in Al0.3V2Al20 a low energy (2meV at T=2K) optical phonon band that exhibits a strong anharmonicity evidenced by threefold increase of characteristic energy on heating up to 300K. In other compounds M=Y, La, Ce, Sc the (x=1) the low energy mode is absent and quasi-harmonic lattice dynamics prevails. The continuation aims to complete the data and to improve the data quality for optimal comparison with ab-initio calculated phonon response.

Experimental report 7-01-374, 7-01-396

Two papers concerning the dynamics have already been published , one more on the diffraction part is coming later.

Cite this: *Phys. Chem. Chem. Phys.*, 2014, **16**, 27119

Effect of the electropositive elements A = Sc, La, and Ce on the microscopic dynamics of AV_2Al_{20}

Michael Marek Koza,*[a] Andreas Leithe-Jasper,[b] Erik Sischka,[b] Walter Schnelle,[b] Horst Borrmann,[b] Hannu Mutka[a] and Yuri Grin[b]

We report on the inelastic response of AV_2Al_{20} (with A = Sc, La and Ce) probed by high-resolution inelastic neutron scattering experiments. Intense signals associated with the dynamics of Sc, La and Ce are identified in the low-energy range at 6–14 meV in ScV_2Al_{20} and at 8–16 meV in LaV_2Al_{20} and CeV_2Al_{20}. Their response to temperature changes between 2 and 300 K reveals a very weak softening of the modes upon heating in LaV_2Al_{20} and CeV_2Al_{20} and a distinguished blue shift by about 2 meV in ScV_2Al_{20}. By means of density functional theory (DFT) and lattice dynamics calculations (LDC) we show that the unusual anharmonicity of the Sc-dominated modes is due to the local potential of Sc featured by a strong quartic term. The vibrational dynamics of ScV_2Al_{20} as well as of LaV_2Al_{20} and CeV_2Al_{20} is reproduced by a set of eigenmodes. To screen the validity of the DFT and LDC results they are confronted with data from X-ray diffraction measurements. The effect of the strong phonon renormalization in ScV_2Al_{20} on thermodynamic observables is computed on grounds of the LDC derived inelastic response. To set the data in a general context of AV_2Al_{20} compounds and their physical properties we report in addition computer and experimental results of the binary V_2Al_{20} compound.

Received 11th September 2014,
Accepted 14th October 2014

DOI: 10.1039/c4cp04097j

www.rsc.org/pccp

Cite this: DOI: 10.1039/c5cp04005a

On the microscopic dynamics of the 'Einstein solids' AlV_2Al_{20} and GaV_2Al_{20}, and of YV_2Al_{20}: a benchmark system for 'rattling' excitations

Michael Marek Koza,*[a] Hannu Mutka,[a] Yoshihiko Okamoto,[b] Jun-ichi Yamaura[c] and Zenji Hiroi[d]

The inelastic response of AV_2Al_{20} (with A = Al, Ga and Y) was probed by high-resolution inelastic neutron scattering experiments and density functional theory (DFT) based lattice dynamics calculations (LDC). Features characteristic of the dynamics of Al, Ga and Y are established experimentally in the low-energy range of the compounds. In the stereotype 'Einstein-solid' compound AlV_2Al_{20} we identify a unique spectral density extending up to 10 meV at 1.6 K. Its dominating feature is a peak centred at 2 meV at the base temperature. A very similar spectral distribution is established in GaV_2Al_{20} albeit the strong peak is located at 1 meV at 1.6 K. In YV_2Al_{20} signals characteristic of Y dynamics are located above 8 meV. The spectral distributions are reproduced by the DFT-based LDC and identified as a set of phonons. The response to temperature changes between 1.6 and ~300 K is studied experimentally and the exceptionally vivid renormalization of the A characteristic modes in AlV_2Al_{20} and GaV_2Al_{20} is quantified by following the energy of the strong peak. At about 300 K it is shifted to higher energies by 300% for A = Al and 450% for A = Ga. The dynamics of A = Y in YV_2Al_{20} show a minor temperature effect. This holds in general for modes located above 10 meV in any of the compounds. They are associated with vibrations of the V_2Al_{20} matrix. Atomic potentials derived through DFT calculations indicate the propensity of A = Al and Ga to a strong positive energy shift upon temperature increase by a high quartic component. The effect of the strong phonon renormalization on thermodynamic observables is computed on grounds of the LDC results. It is shown that through the hybridization of A = Al and Ga with the V_2Al_{20} dynamics the matrix vibrations in the low-energy range follow this renormalization.

Received 9th July 2015,
Accepted 17th August 2015

DOI: 10.1039/c5cp04005a

www.rsc.org/pccp