Experimental and theoretical studies of vibrational density of states in Fe_3O_4 single crystalline thin film

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Despite long lasting studies, the precise origin and the interactions responsible for the Verwey transition in magnetite Fe_3O_4 are still the subject of some dispute. Strong nearest-neighbour Coulomb repulsion is usually considered to dominate, but some other interactions are required to stabilize cationic arrangement. Here we report the results of experimental and theoretical studies of lattice vibrations in a single crystalline Fe_3O_4(001) film, MBE grown on MgO. The investigations were undertaken to see if electron-lattice interactions participate in the transition.

Vibrational densities of states (DOS) were obtained from the results of nuclear inelastic scattering of synchrotron radiation, while theoretical DOS were calculated ab initio, together with electronic structure, within density functional theory.

We have experimentally scanned the energy region –80 to +80 meV at 295K, 140K, 120K, 100K, 25K. i.e. above and below the Verwey transition temperature T_V; additionally, short energy scans (-5 to 30meV) at 80K, 95K, 105K, 110K, 115K and 130K were made to precisely trace the changes in lattice dynamics at the transition.

Experimental phononic density of states show good coincidence with calculated DOS, reproducing both the general features of main line groups as well as the groups' structure. This is also in qualitative accord with heat capacity data provided that experimental DOS is augmented with that calculated for oxygen atoms.

We have observed the gradual change in the experimental spectrum as well as the relevant DOS while lowering the temperature. In particular, the main peak in the energy region 15-25 meV shows increasing splitting on cooling. The Lamb-Mössbauer factor calculated in the course of DOS evaluation shows the pronounced drop in the vicinity of the Verwey transition that may partly be connected to the observed abrupt lowering of the experimental signal at ca. 7 meV for $T<T_V$. Since this is the indication of the lattice stiffening below T_V, we thus found the experimental evidence for lattice participation in the mechanism leading to the Verwey transition.