PHONON AND MAGNON DISPERSION OF BCC IRON TO 10 GPA

S. Klotz[1] and M. Braden[2]

[1]Physique des Milieux Condensés, CNRS UMR 7602, Université P&M Curie, B77, 75252 Paris

[2]Laboratoire Léon Brillouin (CEA-CNRS) and Forschungszentrum Karlsruhe, IFP, 76021 Karslruhe (Germany)

The properties of iron under extreme conditions are intensively studied [1] due to its relevance for geophysical models of the earth's interior. From a more general point of view, the behaviour of bcc iron under compression is interesting since almost half of the elements contain the bcc structure in their phase diagram. The phonon dispersion of bcc elements was studied in the past in search of dynamical precursor effects of lattice instabilities. Phonon dispersion curves exist for example for alkaline metals (Li, Na and K [2]) at low temperature as well as for transition metals (Zr, Ti, Hf and Sc) at high temperatures [3]. The lattice dynamics of bcc metals under reduced volume, however, is almost completely unknown. Similarly, the behaviour of the magnon dispersion under pressure is unknown for any of the ferromagnetic transition metals.

We have measured both the phonon and magnon dispersion of Fe at room temperature up to 10 GPa, i.e. over almost the entire stability range of the bcc-phase (which transforms to hcp iron at ~11 GPa) [4]. The measurements were carried out at the 1T1 spectrometer similar to previous experiments on Ge and Zn [5]. Typical scans through phonon and magnon branches are shown in Fig. 1. The position of the frequencies could be determined with a statistical error of < 1% in almost all cases and the ambient pressure frequencies agree within ~ 2% with those of published values.

Figure 2 shows the phonon dispersion along high-symmetry directions, 55 phonon mode frequencies have been determined at 0 and 9.8 GPa. All measured frequencies increase by 5-10% for a volume reduction corresponding to 5%. Of particular interest is the T_1-mode with wave vector along [110] and [1-10] polarisation which increases completely regularly at the zone boundary (N-point). This observation is important for the transition mechanism to the hcp-phase at ~10 GPa. The latter can be obtained from the bcc-phase by displacements which involve the $T_1(N)$ phonon (Burgers-mechanism). *Ab initio* calculations on the bcc-hcp transition in Ba predict a substantial softening of the $T_1(N)$ phonon on approaching the transition pressure [6]. In iron, such dynamical precursor effects due to the Burgers mechanism seem to be absent. Our results are in agreement with spin-polarised full-potential total energy calculations [7] which predict a slight increase of the $T_1(N)$ phonon frequency in the 0-10 GPa range. This scenario of the bcc-hcp transition is therefore different from that in Ba under pressure [6], and different from the situation found in all group 3 and 4 transition elements where there is a pronounced softening of the $T_1(N)$ frequency at ambient pressure and elevated temperature as the hcp phase is approached [3]. Iron seems to be an exceptional case in the sense that no dynamical precursor effects are visible on the $T_1(N)$ mode. The critical ingredient for this behaviour is the presence of ferromagnetism which stabilises the bcc-phase with respect to the non-magnetic fcc-phase. The effect of pressure is essentially to broaden the flat d-bands and thereby to decrease the density of states at the Fermi level $D(E_F)$ below the stability limit for ferromagnetism [7]. The bcc-hcp instability is hence not primarily due to phonon softening but due to the effect of pressure on the magnetism of iron.

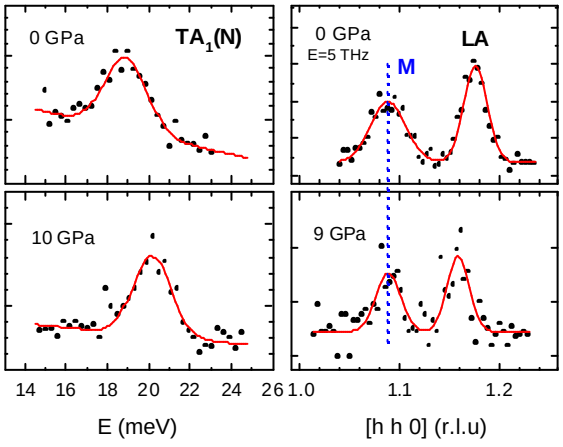

Figure 1 Constant-Q scans through the $TA_1(N)$ phonon branch (left) and constant-E scans through the magnon (M) and LA-phonon branches (right), at

ambient and maximal pressure. Note the insensitivity of the magnon energy to high pressure.

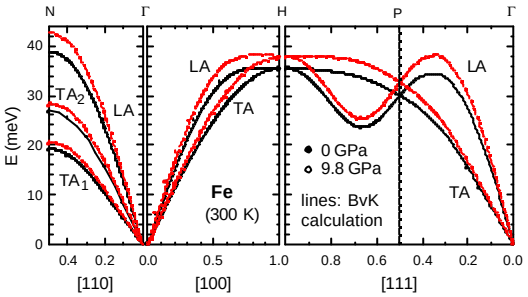

Figure 2. Phonon dispersion along high-symmetry directions at 0 and 9.8 GPa.

We performed lattice dynamical calculations using a general force constant model similar to previous ambient pressure investigations on iron and other bcc elements (Fig. 2). This model contains 13 force constants, seven of which were allowed to vary with pressure. It enables us to calculate the pressure dependence of the elastic constants to high accuracy. It also allows to extract the density of phonon states (Fig. 3) which gives thermodynamic parameters such as the specific heat at constant volume or the equivalent Debye temperature θ_D. Such results are difficult or impossible to obtain by other techniques.

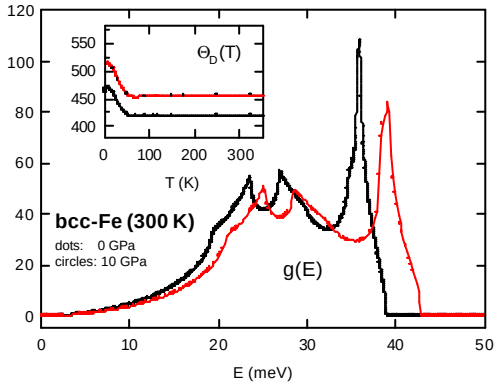

Figure 3. Phonon density of states and Debye temperature $\theta_D(T)$ (inset) at 0 and 9.8 GPa.

In view of the important role of Fe in stabilising the bcc-phase we decided to study also the magnetic excitations as a function of pressure. As for the spin-wave dispersion of bcc-iron and other ferromagnetic transition elements, it is known to be isotropic and follows for small $\mathbf{q}$ the dispersion law $E = D|\mathbf{q}|^2$, where $D = 2 J S a^2$ is the spin wave stiffness constant, S the spin, a the lattice parameter and J the coupling constant. Consequently, a measurement of the magnon dispersion under pressure allows us to determine the dependence of the magnetic interaction on the interatomic distance, providing $\partial S/\partial P$ is known. Fortunately, $\partial S/\partial P$ is well constrained by both experiments and theory to be ~ -3% between 0 and 10 GPa. Most surprisingly, no variation in the magnon energies is observed in our measurements up to 10 GPa (see Fig. 1). This unusual weak pressure dependence of the magnetic excitation spectrum is unexpected and hitherto unexplained by first-principle calculations. Hower, it appears to be in excellent quantitative agreement with Wohlfarth's itinerant electron model for ferromagnetism [8]. Within this theory, the weak pressure coefficient of D can be related to the pressure *in*dependence of the Curie temperature ($\partial T_C/\partial P=0\pm0.3$ K/GPa) and the elevated bulk modulus of Fe (164 GPa).

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