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Abstract
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Compositional variation of the phonon dispersion curves of bcc Fe-Ga alloys

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Inelastic neutron scattering techniques have been used to measure the phonon dispersion curves of bcc $Fe_{1-x}Ga_x$ ($x=10.8$, 13.3, 16.0, 22.5) alloys as a function of Ga concentration. The phonon frequencies of every branch were found to decrease significantly with increasing Ga concentration. The softening was most pronounced for the $T_2[\xi\xi 0]$ branch and, to a lesser extent, the $L[\xi\xi\xi]$ branch in the vicinity of $\xi=2/3$. The concentration dependence of the shear elastic constant $C'=1/2(C_{11}-C_{12})$, calculated from the slope of the $T_2[\xi\xi 0]$ branch, was found to agree with the results of sound velocity measurements. For the higher concentration sample measured, 22.5 at. % Ga, new branches appeared, an effect associated with the increase in the number of atoms per unit cell.

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Because of the practical applications of magnetostrictive (MS) materials, considerable effort is being spent in the development of simple alloys with large MS coefficients.[1] Fe-Ga alloys are particularly promising, since, in spite of the complexity of their phase diagram, they can be obtained as practically single-phase bcc alloys.[2,3] As a result, their physical properties have been extensively studied by a variety of experimental techniques.[4] The results of sound velocity measurements[5,6] showed that the shear elastic constant $C'=1/2(C_{11}-C_{12})$ decreases linearly with increasing Ga concentration and extrapolates to zero at approximately 26 at. % Ga. In addition, as the authors[5] pointed out, the concentration dependence of C' bears a striking resemblance to the temperature dependence of C' in martensitic alloys and therefore may be indicative of a cubic to tetragonal structural transition. These interesting results motivated us to undertake a neutron scattering study of the phonon dispersion curves of bcc Fe-Ga alloys as a function of Ga concentration. In this paper we present the results of this study.

Experimental details. The inelastic neutron scattering experiments were performed on single crystals of bcc Fe-Ga with concentrations of 10.8, 13.3, 16.0, and 22.5 at. % Ga. A sample of 28.8 at. % Ga, in which approximately 50% of the sample was in the ordered DO_3 phase, was also studied. The single crystal samples were prepared at the Materials Preparation Center of the Ames Laboratory by the Bridgman technique. Stoichiometric quantities of gallium and electrolytic iron of 99.999% and 99.99% purity, respectively, were cleaned and repeatedly arc melted together under an argon atmosphere. The resulting material was remelted and drop cast into a copper mold to obtain compositional homogeneity. This ingot was heated in an alumina crucible under vacuum to 600 °C for degassing, and then, under an argon atmosphere, the temperature was raised to 1650 °C, approximately 200 °C above the melting point of the alloys. After approximately 1 h at this temperature, the crystals were grown using a withdrawal rate of 5 mm/h. The samples were annealed at reduced temperatures appropriate for their composition and then water quenched to room temperature. The compositions were determined to within ±0.3% by energy dispersive spectroscopy in a scanning electron microscope (SEM). Relatively large single crystals of 2–3 cm^3 of varying overall dimensions and shapes were extracted from the resulting crystal boules. Further details of the single crystal preparation are given elsewhere.[3]

The crystals used in the experiments were checked and oriented by standard neutron diffraction techniques. Although of sufficient volume (2–3 cm^3), they were not of the quality usually required for inelastic neutron experiments, probably because of the stresses developed during quenching from high temperature; the measured mosaic spread [full width at half maximum] for the crystals used in the experiments was between 0.75° and 1.0°. Contamination by the DO_3 phase was checked by comparing the intensities of several bcc Bragg reflections with those allowed only in the DO_3 and B_2 structures. No measurable DO_3 phase was found in the 10.8, 13.3, and 16.0 at. % Ga crystals. On the other hand, approximately 24% of the 22.5 at. % Ga crystal was in the DO_3 phase. In the 28.8 at. % Ga sample studied, the DO_3 phase reached 50%.

The inelastic neutron scattering experiments were performed using the HB1A and HB1 triple-axis spectrometers at the High Flux Isotope Reactor (HFIR) of the Oak Ridge National Laboratory (ORNL). The monochromators and analyzers of both instruments used the (002) reflection of pyrolitic graphite (PG). Highly oriented PG filters (HOPG) were used to minimize higher-order contaminations of the beam. The HB1A spectrometer operates at a fixed incident energy of 14.6 meV, requiring most scans to be performed with neutron-energy gain. HB1, a variable incident energy instrument, was used in the fixed final energy mode (13.5 and 14.7 meV) and all scans were performed with neutron energy loss. Nominal collimations of $48'-40'-40'-240'$ were used and all scans were performed in the constant-Q mode, while counting against neutron monitor counts.

To assess if there is any soft phonon behavior, the dispersion curves of the 22.5 at. % Ga sample were measured as a function of temperature. These measurements were performed with the sample mounted in a 10^{-5} Torr vacuum furnace.

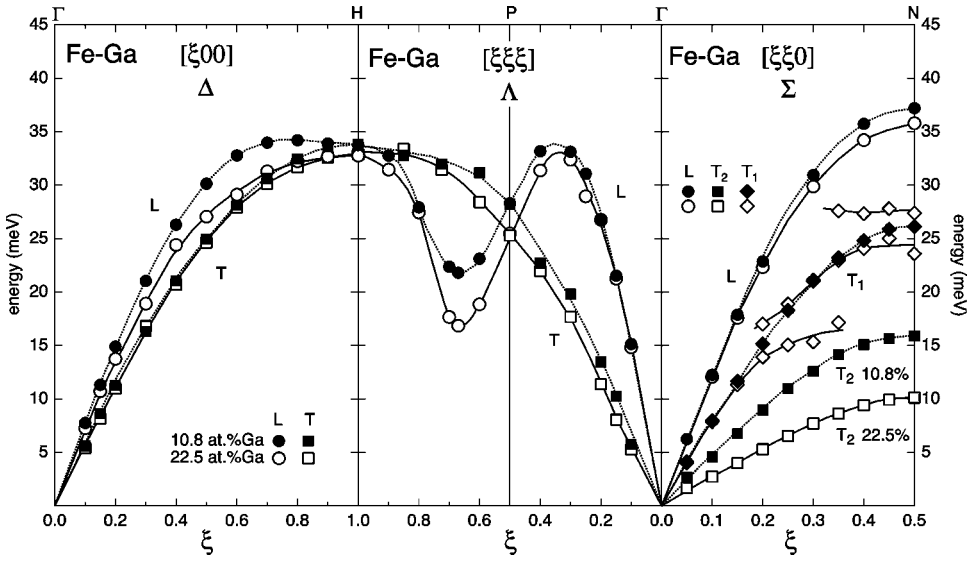

FIG. 1. Comparison of the dispersion curves of Fe-10.8 and 22.5 at. % Ga samples. (The solid symbols are Fe-10.8 at. % Ga and the open symbols are Fe-22.5 at. % Ga.)

Experimental results and discussion. The experimental results are summarized in Figs. 1–3. The dispersion curves of the 10.8 at. % Ga sample are compared in Fig. 1, with those of the 22.5 at. % sample and in Fig. 2 with those of α-Fe.[7] It is seen (Figs. 1 and 2) that the phonon frequencies of every branch decrease significantly with increasing Ga concentration. Most striking is the extremely large softening of the $T_2[\xi\xi 0]$ branch, transverse phonons propagating in the [110] direction with $[1\bar{1}0]$ polarization (Fig. 3). There is also a large softening of the longitudinal [111] branch, $L[\xi\xi\xi]$, in the region of $\xi=\frac{2}{3}$. The shear elastic constant C', estimated from the slope of the $T_2[\xi\xi 0]$ branch at small phonon wave vectors, decreases linearly, to a good approximation, with increasing Ga concentration and extrapolates to zero at approximately 30 at. % Ga concentration, in quite good agreement with the results of sound velocity measurements by Wuttig *et al.*;[5] the elastic constant C_{44}, determined from the slopes at small phonon wave vectors of the $T_1[\xi\xi 0]$ and $T[\xi 00]$ branches, is also in good agreement with the sound velocity measurements[5] (Fig. 4). On the other hand, no pronounced phonon anomalies indicative of a lattice instability were observed. Furthermore, no soft phonon behavior was observed, to within experimental precision, in the measured temperature dependence of the dispersion curves of the 22.5 at. % Ga crystal. Comparison of the measured dispersion curves at 650 °C and room temperature shows only the expected increase of all phonon frequencies with decreasing temperature. At low Ga concentrations (10.8, 13.3, and 16.0 at. % Ga), no new branches were observed, to within the resolution of the present experiments. In the 22.5 at. % Ga sample, new transverse branches in the $[\xi\xi 0]$ direction were observed, characteristic of the ordered (diatomic) DO_3 structure.

There is currently little experimental information on the compositional dependence of the phonon dispersion curves of Fe-based alloys with nonmagnetic atoms. For the purpose of comparison with the results of the present experiment, however, the results obtained in the studies of the Fe-Al,[8] Fe-Si,[9] and, more recently, the Fe-Be[10] alloys are very useful. The interesting features exhibited by the dispersion curves of these alloys are similar to but much less pro-

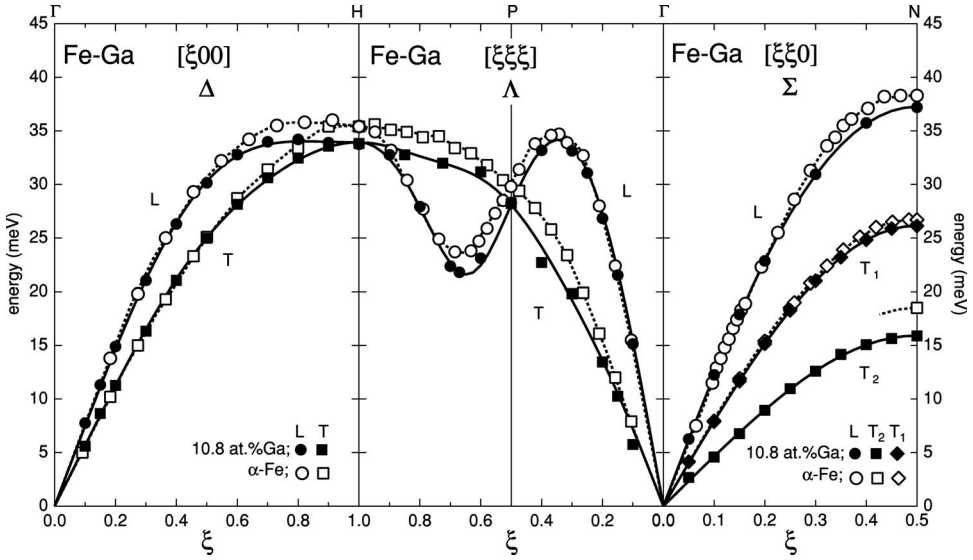

FIG. 2. Comparison of the dispersion curves of Fe-10.8 at. % Ga and α-Fe (Ref. 6). (The solid symbols are Fe-10.8 at. % Ga and the open symbols are α-Fe.)

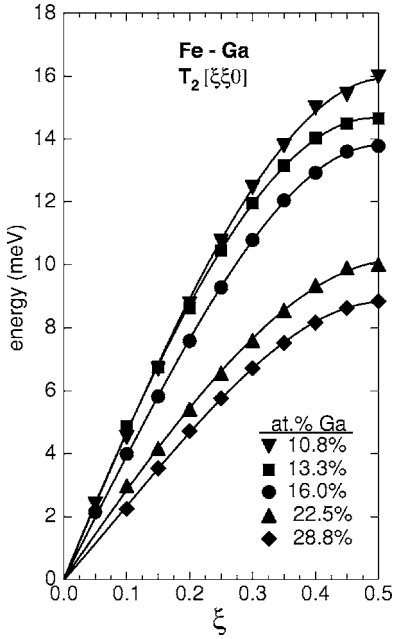

FIG. 3. $T_2[\xi\xi 0]$ phonon dispersion curves of Fe-Ga for five Ga concentrations.

nounced than those observed in the present experiments (see previous paragraph). It is particularly important to notice that this comparison reveals that the softening of the $T_2[\xi\xi 0]$ branch increases with increasing atomic number of the nonmagnetic atoms, a trend that is also followed by the measured magnetostriction coefficients of these alloys. Thus, for two reasons, the overall softening of the $T_2[\xi\xi 0]$ branch could be of fundamental importance in our understanding of the large magnetostriction coefficients observed in Fe-based alloys. First, as pointed out by Wuttig *et al.*,[4] the dramatic decrease of the shear elastic constant C' with increasing Ga concentration can explain the increase in magnetostriction observed in the low Ga concentration region (disordered A_2 phase). Second, the low migration barrier presented by the low-lying (in energy) $T_2[\xi\xi 0]$ branch may explain the "anomalously" large magnetostriction observed as one approaches the Fe_3Ga composition,[11,12] as well as the diffusivity in these alloys.[11–13]

Only a detailed comparison of the results of the present experiment with first-principles calculations of the dispersion

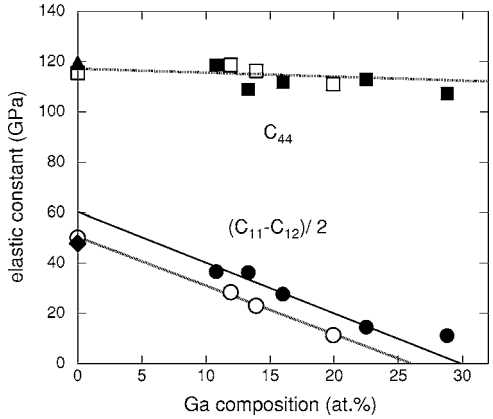

FIG. 4. Fe-Ga elastic constants estimated from the slopes of the phonon dispersion curves for the five Ga concentrations studied and from the Born-von Kármán analysis of Minkiewicz *et al.* (Ref. 6) for α-Fe (solid symbols). The open symbols are from the ultrasonic measurements of Wuttig *et al.* (Ref. 4).

curves as a function of composition can provide a fundamental understanding of the electronic origin of the dramatic softening observed for the $T_2[\xi\xi 0]$ branch with increasing Ga concentration. In recent years, many first-principles calculations of the physical properties of systems containing 3*d* transition-metal atoms including the magnetoelastic energy and magnetostriction, have been performed using the density functional theory in both the local spin density and generalized-gradient approximations.[12,14] To the best of our knowledge, however, first-principles calculations of the phonon dispersion curves were performed only for (fcc) γ-Fe,[15] Ni_3Al,[16] and FeAl.[17] Such first-principles calculations[18] of the phonon dispersion curves of Fe-Ga alloys or calculations based on the Varma-Weber method[19] could provide invaluable information regarding the electronic origin of the observed softening.

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