Phonon dispersion curves in iron-based fcc alloys using microscopic force constants

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Phonon frequencies of $Pd_{1-x}Fe_x$ homogeneous alloys along three symmetry directions have been calculated by using the microscopic force constants derived from the force constants of the constituent atoms. The force constants of Pd and Fe atoms are obtained through a first-principles approach and are found to reproduce the experimental phonon frequencies and elastic constants of the corresponding metal. The theoretical and experimental results of $Pd_{1-x}Fe_x$ alloys for the concentrations $x=0.04$, 0.10, and 0.28 agree to within 11%. The elastic constants of these alloys are also calculated and compared with experimental results.

I. INTRODUCTION

There exist several phenomenological and first-principles theories to study the lattice dynamics of binary alloys. A few of them[1-5] are based on the coherent-potential approximation. Jaccuci *et al.*[6] made use of molecular dynamics to study the local- and band-mode behavior of RbK alloys. However, the molecular-dynamics simulation method requires a lot of computer time, perhaps beyond the reach of many workers. It is to be noted that the local modes arise when impurities of lighter mass are present in the host of heavier atoms. The other theoretical models to study the lattice dynamics of mixed crystals are the Green-function technique due to Kutty[7] and its phenomenologically modified form due to Garg *et al.*[8]

All the above studies are based on the fact that the two constituent metals of binary (type-1) alloys have the same structure, such as chromium-tungsten,[9] palladium-platinum,[10] nickel-platinum,[11] potassium-rubidium,[12], etc. However, there are some binary (type-2) alloys where the structure for the two constituent metals is different at room temperature: for example, iron-nickel,[13] iron-platinum,[14] and iron-palladium.[15,16] Experimental results of phonon frequencies suggest that the local mode in a type-1 system is present due to the impurity atom, whereas in a type-2 system the lattice vibrations are related closely to those of the host lattice, i.e., no local-mode frequency is present. This suggests that a single unique lattice is formed in type-2 systems. Garg *et al.*[17] have calculated the phonon frequencies of $Ni_{1-x}Fe_x$ and $Pd_{0.72}Fe_{0.28}$ alloys using the usual secular equation for cubic crystals and employing an angular force model and have found good agreement between calculated and experimental results. However, these authors have ignored the effect of conduction electrons in the calculation of the phonon frequencies of the alloys, as has been pointed out by Imaizumi *et al.*[18] The electron-ion interaction in metals, particularly in transition metals, plays an important role. Prakash and Joshi[19] and Hanke[20] have emphasized the need for electron-ion interactions and included them in lattice-dynamical calculations. Recently, we[21,22] have studied the lattice dynamics of a number of transition metals, including those of Pd and Fe in the fcc phase, by including the contribution of *s* and *d* conduction electrons explicitly. We feel that the linear combination of the force constants of the constituent metal atoms, which

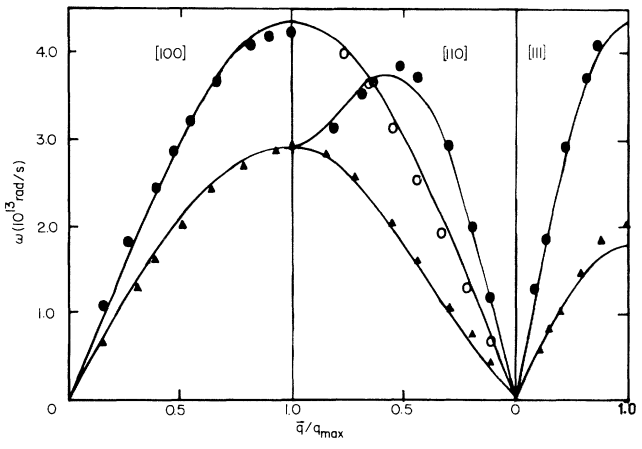

FIG. 1. Dispersion curves of $Pd_{0.96}Fe_{0.04}$ at 300 K. The solid circles are the experimental data by Maliszewski *et al.* (Ref. 16) at room temperature.

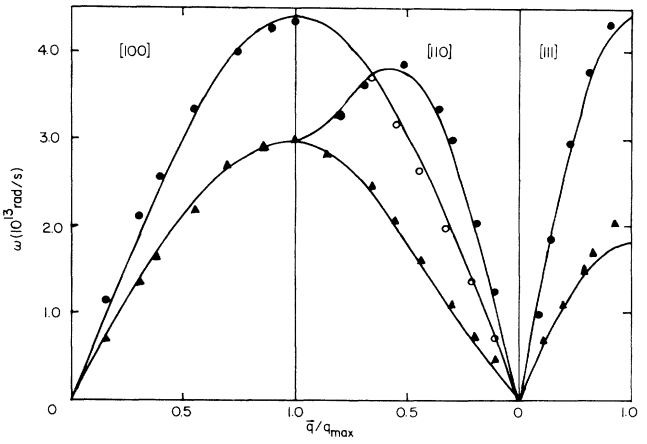

FIG. 2. Dispersion curves of $Pd_{0.90}Fe_{0.10}$ at 300 K. The rest of the description is the same as that of Fig. 1.

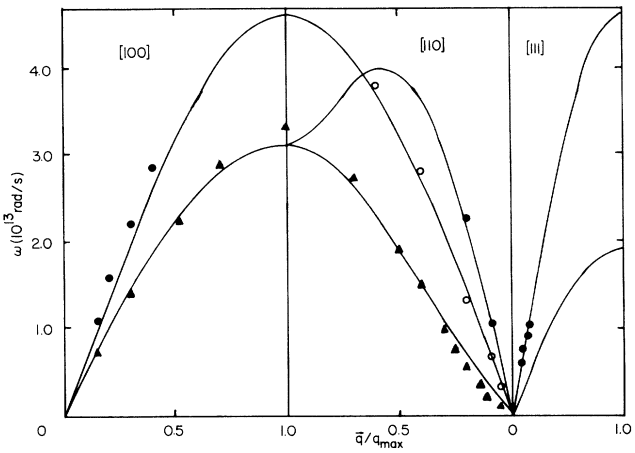

FIG. 3. Dispersion curves of $Pd_{0.72}Fe_{0.28}$ at 300 K. The solid circles are the experimental data by Sato *et al.* (Ref. 15) at room temperature.

reproduce the experimental phonon frequencies and elastic constants, will represent adequately the force constants of their alloys. In this presentation, we use these microscopic, derived force constants to calculate the phonon spectra and elastic constants of $Pd_{1-x}Fe_x$ alloys.

II. THEORY

The iron-based alloys are homogeneous and crystallize in the fcc structure. Thus the phonon frequencies corresponding to a wave vector $\mathbf{q}$ can be obtained just as for cubic crystals, i.e., by solving the secular equation

$$|D_{\alpha\beta}(\mathbf{q})-M\omega^2(\mathbf{q})\delta_{\alpha\beta}|=0 , \quad (1)$$

where $D_{\alpha\beta}(\mathbf{q})$ are the elements of the dynamical matrix and α and β are Cartesian components (x,y,z). The average atomic mass M and valence Z of the mixed system are obtained using the expressions

$$M=(1-x)M_{\mathrm{Pd}}+xM_{\mathrm{Fe}} \quad (2)$$

and

$$Z=(1-x)Z_{\mathrm{Pd}}+xZ_{\mathrm{Fe}} , \quad (3)$$

where M_{Pd} and M_{Fe} are the atomic masses and Z_{Pd} and Z_{Fe} are the valences of crystalline Pd and Fe, respectively. x is the concentration of Fe in the Pd host. In order to evaluate the force constants of the mixed systems, we use the following relation:

$$K=(1-x)K_{\mathrm{Pd}}+xK_{\mathrm{Fe}} , \quad (4)$$

where K_{Pd} and K_{Fe} are the force constants of the crystalline Pd and Fe, respectively. From the interionic force constants, we obtain the elastic constants for the cubic Pd-Fe alloy by using the dynamical-long-wavelength phonon method.[21]

III. CALCULATIONS AND RESULTS

Constituent metal atoms Pd and Fe of the alloy Pd-Fe crystallize in fcc and bcc structure, respectively, at room temperature. However, the neutron inelastic-scattering data[15,16] of phonon frequencies of the Pd-Fe alloy suggest that a single unique fcc lattice is formed by this alloy, i.e., impurity Fe atoms in the Pd host are distributed homogeneously in the fcc phase. Therefore, in order to obtain the force constants of Pd-Fe alloys, we use the force constants of Pd and Fe in the fcc phase, as we[22,23] obtained earlier. For ready reference, the force constants of Pd and Fe up to the seventh neighbor are given in Table I. It is to be noted that the contributions up to the seventh neighbor shall have been found sufficient to achieve a convergence of phonon frequencies of Pd and Fe metals. The lattice parameters for the Pd-Fe alloys are taken from Refs. 16 and 15. The calculated phonon frequencies of the Pd-Fe alloys for the concentrations $x=0.04$, 0.10, and 0.28, obtained by using Eq. (1) along three symmetry directions [100], [110], and [111], are compared with the experimental[15,16] values in Figs. 1, 2, and 3, respectively. It is found that the calculated and experimental[16] results of phonon frequencies for $Pd_{0.96}Fe_{0.04}$ and $Pd_{0.90}Fe_{0.10}$ alloys are in good agreement in all the symmetry directions except the [111] T mode near the Brillouin-zone (BZ) boundary. The maximum discrepancies between theoretical and experimental results at the BZ boundary along the [111] T mode are found to be 11% and 10% for the

TABLE I. Radial (K_r) and tangential (K_t) force constants (in dyn/cm) of Pd (Ref. 22) and Fe (Ref. 23).

		Pd		Fe	
Shell	Type of atom	K_r	K_t	K_r	K_t
1	$\frac{a}{2}$ (110)	43 169.622	−2240.944	42 707.477	−4250.876
2	$\frac{a}{2}$ (220)	−2632.109	269.486	−886.216	149.980
3	$\frac{a}{2}$ (211)	162.136	52.417	3.990	45.301
4	$\frac{a}{2}$ (220)	−222.056	48.922	−196.647	27.538
5	$\frac{a}{2}$ (310	−225.808	14.462	−89.486	7.321
6	$\frac{a}{2}$ (222)	−8.395	3.811	3.749	4.061
7	$\frac{a}{2}$ (321)	20.117	4.982	−13.127	3.658

TABLE II. The calculated values of the elastic constants (in Mbar) of $Pd_{1-x}Fe_x$ alloys at $T=300$ K. The experimental values are taken from Refs. 15 and 16. The numbers in parentheses represent the error (%) in the theoretical values.

	$x=0.04$		$x=0.10$		$x=0.28$	
	Theor.	Expt.	Theor.	Expt.	Theor.	Expt.
C_{11}	1.716(−25)	2.3	1.724(−25)	2.29	1.791(+28)	1.40±0.20
C_{12}	1.273(−17)	1.53	1.292(−22)	1.65	1.380(+3)	1.34±0.18
C_{44}	1.015(+30)	0.78	1.004(+14)	0.86	0.992(+24)	0.80±0.02

concentration $x=0.04$ and 0.10, respectively. The calculated phonon frequencies of the $Pd_{0.72}Fe_{0.28}$ alloy also agree reasonably well with experimental[15] values. The theoretical values of elastic constants as calculated in the long-wavelength limit are given in Table II. The maximum discrepancy between calculated and experimental[15,16] data of elastic constants for Pd-Fe alloys are found to be 30%, 25%, and 28% for $x=0.04$, 0.10, and 0.28, respectively.

IV. CONCLUSIONS

In summary, the force constants of the iron-based alloy are derived from the microscopic force constants of its constituent atoms where the contributions of *s* and *d* electrons are included explicitly. These derived force constants are then used to calculate the phonon spectra and elastic constants of alloys for various concentrations without including the clustering or disorder effects. The good agreement between calculated and experimental results shows that in iron-based alloys the local environment effects are not present. The same conclusion also has been drawn by Garg *et al.*[17] from the calculated phonon spectra of Ni-Fe alloys.

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