Magnons and Magnon-Phonon Interactions in Iron

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First-principle studies of magnons and magnon-phonon interactions are carried out in bcc and fcc iron in the adiabatic approximation. It is shown that the phonons have a minor effect on magnons in bcc Fe and thus the lattice vibrations make a small contribution to the Curie temperature. fcc Fe is unstable against magnon excitations but the phonons seem to reduce this instability. The magnon-phonon interactions are analyzed in terms of the pair-exchange interaction variations as functions of the interatomic distances. The fcc results suggest that metastable fcc Fe in thin-film or nanostructure form should have interesting magnetic properties.

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Generally, it is expected that phonons do not have an appreciable effect on the magnetic properties of most materials. However, some iron-rich systems have low Curie temperature (T_c) which increases significantly with a small lattice expansion. For example, R_2Fe_{17} (R = rare earth) compounds have T_c's around 350 K which increases to about 750 K with a volume expansion of $\sim$6% [1]. Such systems, which are close to magnetic and/or structural instability, should have reasonable magnon-phonon interactions. Classic examples of such systems are metastable fcc and amorphous Fe. fcc Fe has been stabilized recently in thin films and as a precipitate in Cu matrix [2]. Theoretical studies show that fcc Fe is a weak magnet exhibiting magnetovolume instability and noncollinear magnetic structure under compression [3]. On the other hand, bcc Fe is a strong magnet with a high T_c and stable magnetic structure. The very different magnetic properties of bcc and fcc iron should be reflected in their magnon dispersion curves without and with phonon excitations.

Because of the complexity of the magnon-phonon interactions, only a few simple model studies have been reported so far [4–6]. *Ab initio* calculations of the magnon spectra in the absence of phonons in some transition metals have been reported only recently using a frozen-magnon [7] and linear-response scheme [8]. The former calculations are based on linear muffin-tin orbitals (LMTO) method in the atomic sphere approximation, while the latter use the full-potential LMTO method. The results for Fe by the two methods are similar for small wave vectors. We report here a first-principles study of the influence of lattice vibrations on the magnon excitations in bcc and fcc Fe using a frozen-magnon frozen-phonon scheme. The results show significant differences in the magnon spectra of magnetically stable (bcc) and metastable (fcc) structures.

Starting with the Hamiltonian, H, a brief outline of the magnon-phonon theory is as follows:

$$H = \frac{1}{2}\sum V^{(2)}_{\alpha\beta}(ij)u_\alpha(i)u_\beta(j) + \sum_{ij} J(\vec{R}_i,\vec{R}_j)\vec{S}(i)\cdot\vec{S}(j) + \text{anharmonic terms}, \tag{1}$$

where the first term is the vibrational term in the harmonic approximation with $u_\alpha(i)$ being the α component of the vector displacement of atom i from its equilibrium position and the second term is the Heisenberg exchange term with $\vec{S}(i)$ as the unit vector. The distance dependent exchange parameter $j(\vec{R}_i,\vec{R}_j)$ is expanded in terms of the atomic displacements as follows:

$$\begin{aligned} J(\vec{R}_i,\vec{R}_j) &\equiv J(|\vec{R}_i - \vec{R}_j|) \\ &= J^{(0)}(ij) + J^{(1)}_\alpha \Delta R_\alpha(ij) \\ &\quad + \tfrac{1}{2}\Delta R_\alpha(ij) J^{(2)}_{\alpha\beta}\Delta R_\beta(ij), \end{aligned} \tag{2}$$

where $\Delta\vec{R}(ij) = \vec{u}(i) - \vec{u}(j)$, $J^{(1)}_\alpha = [\partial J_{ij}/\partial R_\alpha(ij)]|_0$, and $J^{(2)}_{\alpha\beta} = [\partial^2 J_{ij}/\partial R_\alpha(ij)\partial R_\beta(ij)]|_0$. With this Hamiltonian one gets the following coupled equations in the reciprocal q space for one atom per unit cell systems:

$$\sum_\beta D_{\alpha\beta}(q)e_\beta(q) = \omega_q^2 e_\alpha(q), \tag{3}$$

$$\sum_\beta J_{\alpha\beta}(q)\theta_\beta(q) = E_q\theta_\alpha(q), \tag{4}$$

where $D_{\alpha\beta}(q) = \sum e^{-i\vec{q}\cdot\vec{R}_{ij}}[V^{(2)}_{\alpha\beta}(ij) + J^{(2)}_{\alpha\beta}(ij)\langle\vec{S}(i)\cdot\vec{S}(j)\rangle + V^{(4)}_{\alpha\beta}(ij)\langle u^2\rangle]$ with $V^{(4)}_{\alpha\beta}(ij)\langle u^2\rangle$ being the anharmonic term in random phase approximation, $J_{\alpha\beta}(q) = \sum e^{-i\vec{q}\cdot\vec{R}}[J^{(0)}(ij) + J^{(2)}_{\alpha\beta}(ij)\langle u^2\rangle]$, $e_\beta(q)$ (ω_q^2) and $\theta_\beta(q)$ (E_q) are the eigenvectors (eigenvalues) of phonons and magnons, respectively, and $\langle u^2\rangle$ is the mean-square displacement.

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In this work we are interested in studying the effect of phonons on magnon dispersion curves. It is not necessary to solve Eqs. (3) and (4) self-consistently for this purpose. It is sufficient to perform magnon calculations for a lattice distorted by a realistic simulation of the phonons. This is done by using frozen phonons [9] whose amplitude is the room-temperature mean-square displacement ($\langle u^2 \rangle = 0.75\%$ of the lattice constant). The wave vector of the phonon is the same as that of the magnon. Thus, we neglect multiphonon-multimagnon interactions. The magnon energy E_q is related to the difference in the total energy $\Delta E(\vec{q}, \theta)$ of the phonon-distorted lattice with and without the frozen spin spiral of wave vector $\vec{q}$. In the absence of spin-orbit interactions, $\Delta E(\vec{q}, \theta) = \frac{1}{2} E_q \theta^2$, where θ is the spin-spiral angle. Thus, E_q is given by the second derivative of $\Delta E(\vec{q}, \theta)$ for small θ.

$\Delta E(\vec{q}, \theta)$ is calculated for $\vec{q}$ in the [001] direction for a supercell of $1 \times 1 \times 20$ atoms. The self-consistent spin-polarized electronic structure calculations are based on the LMTO method in the atomic sphere and local density approximations and use von Barth-Hedin exchange-correlation potential [10]. Brillouin zone mesh of $16 \times 16 \times 6$ (1536 k-points) is used for reciprocal space integration. This procedure involving noncollinear magnetic structure is similar to that in Ref. [7].

The calculated magnon dispersion curve for bcc Fe is plotted in Fig. 1. Our results for magnons in the absence of phonons are similar to those in Ref. [7] including a Kohn anomaly. For magnon-phonon interactions in bcc Fe we use a longitudinal phonon mode because its energy is similar to that of the magnon. The magnon dispersion curve with magnon-phonon interactions is similar to the one without interactions, and the calculated results are in very good agreement with the experimental data at 4 and 300 K available only at small $\vec{q}$ [11]. One can see that the lattice vibrations have very little effect on the magnon spectrum including the Kohn anomaly. This can be understood from the dependence of the exchange parameters on intratomic distance as discussed below.

In case of bcc Fe single-atom displacement towards one of the nearest neighbors in a 16-atom (15 atoms or 5 neighbor shells are stationary) supercell is used to derive $J(\vec{R}_i - \vec{R}_j)$, while a 32 atom supercell is used for fcc Fe and hcp Co. The details of the calculations of exchange parameters based on the LMTO method are given in Ref. [12]. We use $20 \times 20 \times 20$ divisions of the Brillouin zone for k-space integration in both cases. The calculated $J(|\vec{R}_i - \vec{R}_j|)$ as a function of interatomic distance is plotted in Fig. 2. The almost linear dependence for both the first- and second-neighbor exchange parameters in bcc Fe implies a very small value of $J^{(2)}$, the second derivative of exchange parameter responsible for the magnon-phonon coupling. This also implies a small phonon contribution to the total exchange parameter J_0 and hence to the Curie temperature. A direct calculation of the change in J_0 with the displacement shows that this effect is less than 4%. We also see from Fig. 2 that the exchange parameter dependence on the

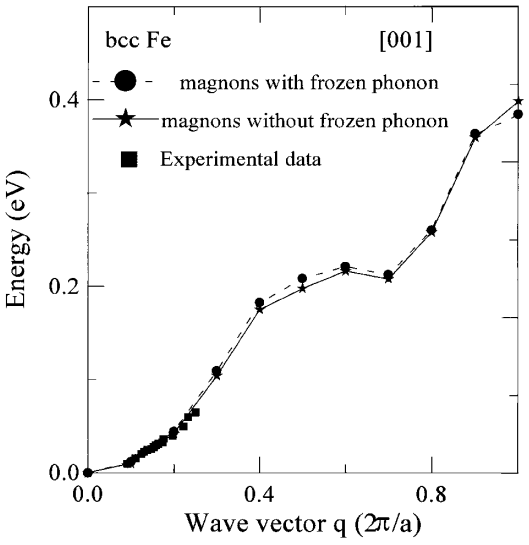

FIG. 1. Magnon energies in [001] direction for bcc Fe. Dashed curve represents the result when the frozen phonon with the same wave vector is present. The experimental data are from Ref. [11].

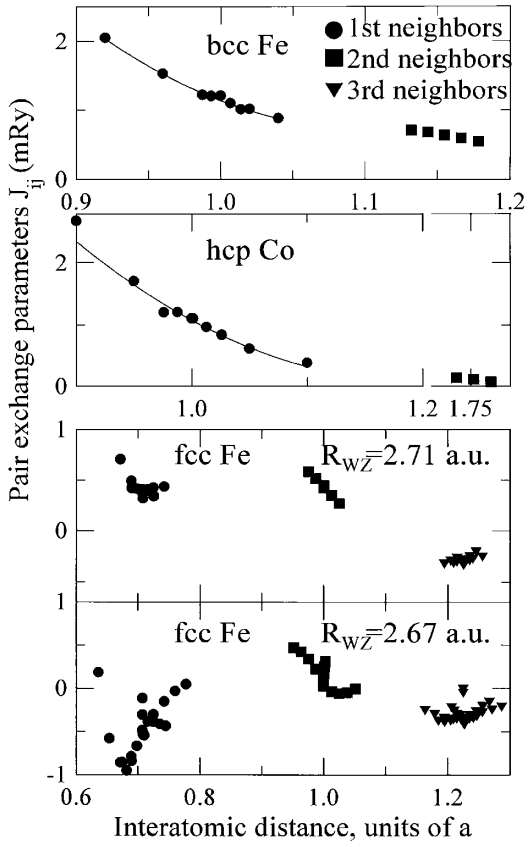

FIG. 2. Distance dependence of the exchange parameters for bcc Fe, hcp Co, and for two different volumes of fcc Fe.

interatomic distance for hcp Co is similar to that of bcc Fe. Thus the magnon-phonon interactions in hcp Co are expected to be similar to those in bcc Fe, i.e., weak.

The [001] magnon dispersion curves for fcc Fe (Wigner-Seitz radius = 2.76 a.u.) are shown in Fig. 3. We see that the ferromagnetic (FM) state of fcc Fe becomes unstable (negative magnon energies) when subjected to magnon excitations. The ground state is not collinear at this volume due to the strong negative interactions with third neighbors (24 of them) despite the positive total exchange parameter obtained using infinitesimal rotation from the FM state. The origin of this instability is related to the sharp peak in the t_{2g} majority spin states at the Fermi energy. This results in a large change in the density of states at the Fermi energy with a small change in the volume which leads to a strong volume dependence of the magnetization and exchange interactions in fcc Fe. Such an instability does not exist in bcc Fe because nearest neighbors are much closer in bcc than in fcc Fe and therefore t_{2g} states in the former have a small delocalized peak at the Fermi energy due to the strong hybridization. We refer the reader to Refs. [3] and [12] for more details.

It is interesting to note in Fig. 3 that magnon-phonon interactions are quite large in fcc Fe (a transverse phonon mode is considered because its energy is closest to the magnon energy) and have the effect of making the FM order "less unstable" against magnon excitations. This can also be understood from the exchange parameter dependence on the interatomic distance for fcc Fe given in Fig. 2. The first-neighbor exchange parameters have a dip near the equilibrium distance. This makes $J^{(2)}$ large which leads to appreciable magnon-phonon interactions in fcc Fe. J_{ij}'s have larger dips for smaller volumes which suggests an even stronger effect of the lattice vibration on the magnon spectrum. Of course, fcc Fe is antiferromagnetic at these volumes.

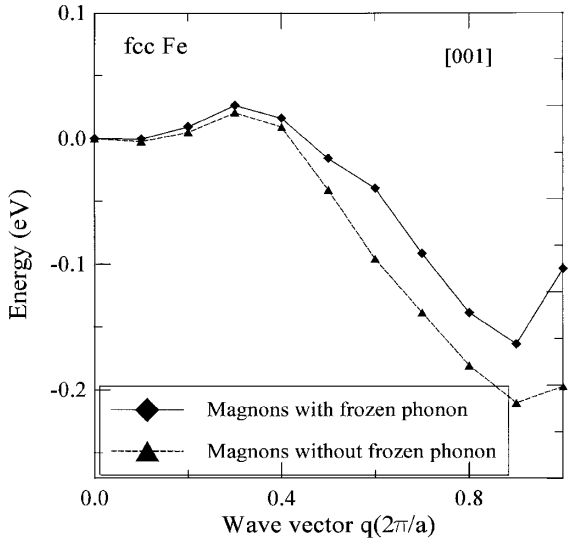

FIG. 3. Magnon energies in [001] direction for fcc Fe. The dashed curve represents the result when the frozen phonon with the same wave vector is present.

The magnetovolume effect and magnon-phonon interactions in fcc Fe suggest that once fcc Fe is formed in thin films or nanostructures, it should have very interesting magnetic properties such as spin frustration and glassiness as functions of the volume and temperature. Also magnon instability in fcc Fe is an indication as to why T_c is low in fcc-like Fe-rich compounds mentioned in the introduction.

In conclusion, we have shown from first principle calculations that the magnon-phonon interactions are negligible in the stable phase (bcc) of Fe. The fcc phase develops soft magnons implying magnetic instability. The magnon-phonon interactions are significant in fcc Fe and make the lattice less unstable against magnon excitations. These properties are related to the volume dependence of the magnetization and exchange interactions and can be understood in terms of the variations of the exchange parameters with interatomic distances.

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