Probing the Magnetic Forces in fcc-Fe(001) Films by Means of Surface Phonon Spectroscopy

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Helium atom scattering experiments on Fe films of a few monolayers grown on a Cu(001) substrate demonstrate that (a) in a magnetic film the interatomic magnetic forces can be directly probed from the temperature dependence of the Rayleigh wave frequency across the Curie temperature, and (b) the Rayleigh wave velocity and the topmost interlayer spacing, measured as a function of the film thickness, show a simultaneous 10% reduction at 3 to 4 monolayers—an indication of large magnetostrictive effects.

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The mechanical effects of magnetic interactions in solids are generally very small as compared to the effects of ordinary Coulomb and Pauli repulsive forces. However, in low-dimensional systems, especially in systems featuring large internal stress, quite appreciable changes in the lattice structure and dynamics can be induced by the magnetic order. Recent studies on fcc-Fe(001) films grown on nonmagnetic substrates such as Cu(001) have provided evidence for a connection between the magnetic order and a structural phase transition found in this system [1–3]. Moreover, it has been shown that the complex magnetic order of such films in the thickness range of a few monolayers (>3 ML), consisting of alternating pairs of ferromagnetically (FM) and antiferromagnetically (AFM) coupled layers [4–8], is accompanied by a varying interlayer [8] and intralayer [9] magnetostriction in the range of a few percent of the topmost interlayer distance d_{12}.

In this paper we show that the dependence of the exchange potential on the interatomic distances is sufficiently strong to give a measurable renormalization of the force constants and phonon frequencies. The influence of magnetization on the temperature dependence of the Rayleigh wave (RW) frequencies has been previously demonstrated for the (110) surface of an iron crystal by means of inelastic He atom scattering (HAS) [10]. In that case the magnetic effects were found to be similar to those observed in bulk iron with neutron scattering [11] and in general comparable in size to the ordinary anharmonic effects, although T_c was too large to allow for measurements to explore the magnetic transition [10].

Consistent with the analysis of these earlier observations, recent first principle calculations by Sabiryanov and Jaswal [3] for bcc-Fe(001) allow one to extract a magnetic contribution $J^{(2)}$ (for unitary spin) to the nearest neighbor radial force constant of about 0.92 N/m, while a 5 to 9 times larger value (for a Wigner-Seitz radius, $r_s = 2.71$ and 2.69 a.u., respectively) is obtained for $J^{(2)}$ in bulk fcc-Fe. $J^{(2)}$ is the second order derivative of the Heisenberg exchange constant with respect to the interatomic distance and accounts for phonon-magnon interaction [3].

In the present work the RW dispersion curves for fcc-Fe(001) films, stabilized on a Cu(001) substrate, were measured along the $\langle 110 \rangle$ direction at room temperature as a function of thickness for Fe coverages between 2 and 15 ML, and, for a 6 ML film, as a function of temperature below and above T_c. Significant changes in the RW velocity and frequencies were observed which are associated with corresponding changes in the magnetic structure.

The films were prepared under UHV conditions (base pressure 4×10^{-11} mbar) using molecular beam epitaxy. Following a careful decontamination of the Cu(001) surface Fe atoms from an evaporation source [12] were deposited at a rate of 0.005 ML/s while the substrate was kept at 293 K. The pressure in the UHV chamber never exceeded 2×10^{-10} mbar during film growth. Thickness and structural order of the film were controlled *in situ* by monitoring the specular HAS intensity [13].

The environmental parameters during the deposition significantly influence the structure of the film as well as the critical thickness above which the crystalline structure of the film changes from fcc to bcc. In the absence of carbon contamination the critical thickness is reported to be 11 ML [14]. In order to investigate the influence of magnetization on the surface phonons, the temperature of the sample was varied between 100 and 450 K. It is known that copper atoms tend to diffuse from the interface to the film surface at elevated temperatures exceeding 500 K [15,16]. The chemical composition of the topmost surface layer cannot be determined accurately, since Auger electron spectroscopy averages over several layers of the selvage. However, for the temperatures of the present experiments and for a film of 6 ML thickness the concentration of copper atoms at the surface is known to be smaller than 10% [16].

The temperature of the sample was measured with an accuracy of better than 2 K in the range of interest. The morphological and structural information on the film surface was gained by analyzing the specular intensity of the helium atom beam as a function of the perpendicular momentum transfer [17]. In this way the step height d_n, defined by the difference in thickness of a $(n-1)$-ML and a n-ML film [18], was measured on films of different thickness. Inelastic scattering was investigated using time-of-flight (TOF) spectroscopy. From such data one can determine the surface phonon dispersion curves in the film surfaces and, in particular, the velocity of the Rayleigh wave.

In the first set of experiments the step height d_n as well as the surface phonon dispersion curves for films of different thicknesses were studied at room temperature (RT). At this temperature, films which are less than 4 ML thick are known to be ferromagnetic with a Curie temperature $T_c \approx 370$ K (Fig. 1a [19]) and a unit cell that is tetragonally elongated in the normal direction with respect to the ideal fcc cell [1]. At coverages of about 4 ML the film undergoes a structural phase transformation from the tetragonal to the cubic phase for which T_c is depressed to about 270 K [1]. For coverages above 5 ML the films are paramagnetic (PM) at RT with an ideal fcc cell. T_c remains fairly constant for increasing coverage [19] until the film transforms into the bcc bulk structure with ferromagnetic order.

The step height d_n, measured as a function of the film thickness (Fig. 1b), shows for FM films up to 3 ML (phase I) an expansion with respect to the ideal value of 1.78 Å for the bulk fcc phase, in agreement with the values found in LEED for the tetragonally expanded FM phase (1.87 Å) [20–22]. For $n > 4$ ML (phase II), the films are PM at room temperature (Fig. 1a) and d_n approaches the limiting bulk value (1.78 Å). For a coverage of 16 ML d_n suddenly jumps to 1.96 Å due to the transformation to the FM bcc phase (phase III) [19]. In the transition region between phase I and phase II (3 to 4 ML), a contraction of d_n down to $\approx$1.6 Å is observed which is due to the fact that steps occur between phase I and phase II domains. If one domain contains three FM layers (1.87 Å interlayer spacing) and the other one three AFM layers (1.78 Å interlayer spacing) plus a topmost FM layer spaced 1.87 Å [21,23], the resulting step height must be $d_3 = 1.6$ Å.

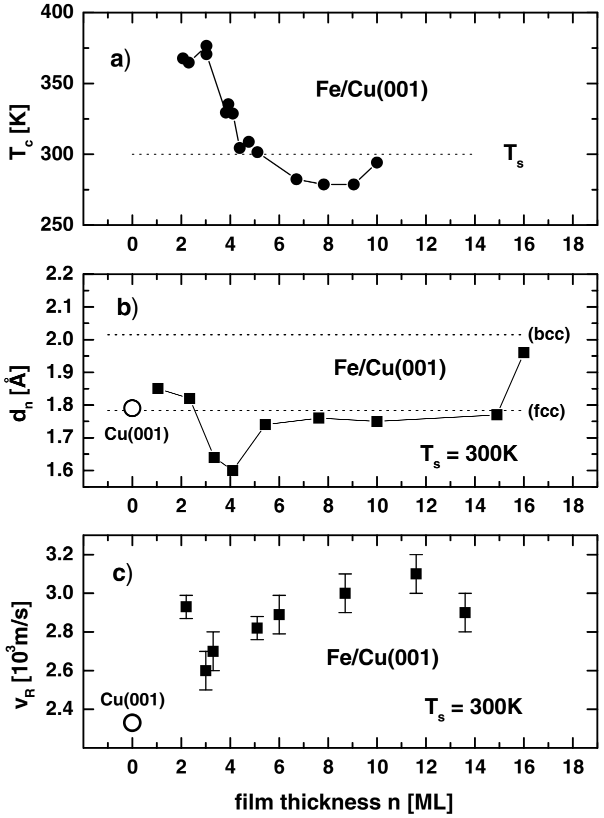

FIG. 1. (a) Curie temperature T_c in fcc-Fe films on Cu(001) as a function of film thickness (after Ref. [19]). (b) Step height d_n at the surface of fcc-Fe films vs film thickness from present HAS experiments. (c) Dependence of the Rayleigh wave (RW) phase velocity v_R on the film thickness from HAS measurements for fcc-Fe films.

In addition to the structural aspects discussed so far, a similarly complex behavior was found in the surface lattice dynamics of these films on changing their thickness. Figure 1c shows the phase velocity v_R of the RW in the $\langle 110 \rangle$ direction as a function of the film thickness. v_R was extracted from the measured RW dispersion curves in a region of phonon wave vectors between 0.1 and 0.3 Å^{-1} where the dispersion is still linear and the penetration length comparable to the film thickness. In the critical thickness regime v_R is also strongly affected and shows a minimum also located somewhere between 3 and 5 ML.

A change in the RW velocity on the order of 10%, as found in this data, is remarkably large. It is difficult to attribute such a phonon softening to magnetic effects alone, but it may well be associated with the structural phase transition reported by Zharnikov *et al.* [1]. However, the softening of the interatomic forces, which leads to a decrease of the RW velocity, is accompanied by a reduction of the average interlayer distance. Such a behavior is not at all expected for ordinary Coulomb forces or Pauli repulsion but indicates the presence of magnetic interactions. In fact, a FM order between neighboring atoms (i, j), contributing with the positive term $J^{(2)}\langle \vec{S}_i \cdot \vec{S}_j \rangle$ to the total interatomic force constant (S_i is the atomic spin), is favored at larger interatomic separations, whereas the AFM order, giving a negative contribution to the interatomic force constant, is favored at shorter interatomic distances. Thus the magnetic effects on the RW velocity and frequencies depend on the balance between the magnetic forces and the associated elastic relaxation.

For larger thicknesses, where the film at room temperature is paramagnetic, the RW velocity monotonically increases to values which are larger than the average RW velocity for the bcc (110) surface (2.85×10^3 m/s [10]). At coverages above 13 ML, however, v_R drops to about this value as a result of the transition to the FM bcc phase.

The contribution of magnetic order to the interactions between adjacent layers was directly probed by measuring the RW frequency for temperatures above and below T_c at a fixed wave vector $\mathbf{Q} = 0.53$ Å^{-1} along the $\bar{\Gamma}\bar{X}$ direction. This is roughly halfway to the zone boundary where the exponential decay of the RW displacement field covers only a few layers. At this wave vector, according to the slab calculation described below, the RW eigenvectors $e_l(\mathbf{Q})$ for the first three layers ($l = 1, 2, 3$) account already for 90%

of the squared RW frequency $\omega^2(\mathbf{Q}) = \sum_{l,l'} e_l^* D_{l,l'} e_{l'}$, where $D_{l,l'}$ are the elements of the interlayer dynamical matrix. Therefore $\mathbf{Q} = 0.53$ Å^{-1} seems to be an optimal choice. At larger wave vectors the HAS intensities become weak, whereas at smaller wave vectors the RW field starts penetrating into the Cu substrate resulting in a reduction of the magnetic effect [24].

TOF spectra were collected for different surface temperatures between 100 and 420 K while keeping the incident angle Θ_i and momentum $\hbar k_i$ constant. Rayleigh wave energies at $\mathbf{Q} = 0.53$ Å^{-1} as a function of T are shown in Fig. 2 for a 6 ML film. A sharp increase of the RW frequency, amounting to 0.51 ± 0.10 meV at low temperature, is found for temperatures below $T_c \approx 255$ K which clearly bears the signature of a magnetization effect and largely exceeds the slow, uniform changes due to anharmonicity. The T dependence of $\hbar\omega(\mathbf{Q})$ at $\mathbf{Q} = 0.53$ Å^{-1} for the clean Cu(001) substrate exhibits no abrupt change at 270 K.

The observed temperature dependence of the squared frequency can be represented by [10]

$$\omega^2(\mathbf{Q},T) = \omega_0^2(\mathbf{Q}) - A\omega_0^2(\mathbf{Q})\mathrm{cth}[\hbar\omega_0(\mathbf{Q})/2kT] + BM^2(T)/M^2(0), \tag{1}$$

where $\omega_0(\mathbf{Q})$ is the harmonic frequency for the normal (paramagnetic) phase and $A = 0.011$ is chosen to fit the T dependence in the normal phase due to anharmonicity (Fig. 2, dotted line). The last term is the change due to the magnetic ordering, with B the magnetic shift and $M(T)$ the surface magnetization. A good fit of $\omega^2(\mathbf{Q},T)$ (solid line in Fig. 2) can be obtained with $\omega_0(\mathbf{Q}) = 9.39$ meV, $B = 13$ (meV$/\hbar)^2$, and an expression $M(T)/M(0)$ which fits the magneto-optical data for these films reported by Li *et al.* [25].

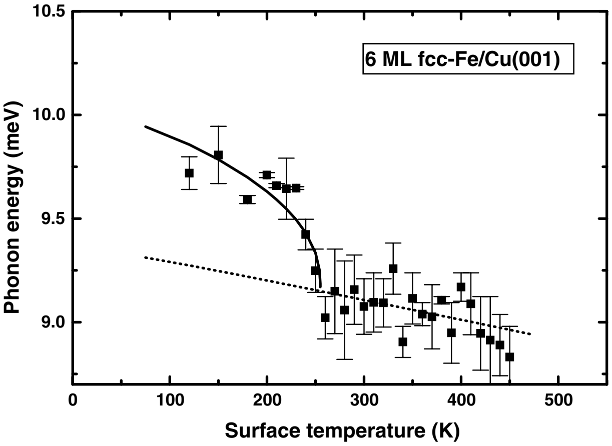

FIG. 2. Changes in the RW frequency with surface temperature for a 6 ML fcc-Fe film measured at $\mathbf{Q}_0 = 0.53$ Å^{-1} along the $\langle 110 \rangle$ direction. The kink in the curve occurs at the Curie temperature of fcc-Fe (263 K). The dotted base line, fitted to the points above T_c, shows the anharmonic frequency shift. Below T_c the frequency shifts away from the anharmonic base line by an amount proportional to the squared magnetization (solid line). The data were extracted from helium TOF spectra using the fitting code PEAKFIT 4.06 (SPSS Inc.) and corrected for constant $\mathbf{Q} = \mathbf{Q}_0$. The plotted points are averages of two different fitting runs with different boundary conditions. The deviation of the two fits from the average is indicated by bars. The average error bar (not shown) is ±0.14 meV.

The constant $J^{(2)}$ can be extracted from the experimental magnetic shift by means of a lattice dynamical calculation. The phonon dispersion curves of a 6 ML slab of fcc-Fe(001) on a Cu(001) substrate have been calculated with a force constant model including, in the normal phase, only the nearest neighbor central force constant f_0. This model is known to work well for the (fcc) substrate Cu(001) with a force constant $f_S = 28.0$ N/m [26] and gives a good fit of the experimental RW branch of the PM fcc-Fe films with $f_0 = 38$ N/m. The interface force constant has been taken as $(f_0 + f_S)/2$. In the PM phase the interatomic distances and interplanar spacings of the fcc-Fe slab were assumed to be the same as in the substrate. In the FM phase the additional force constants $J^{(2)}\langle \vec{S}_i \cdot \vec{S}_j \rangle$ between nearest neighbor atom pairs of spins $\vec{S}_i$ and $\vec{S}_j$ [3] are switched on to account for the magnetic effects. Consistent with Sabiryanov's and Jaswal's definition, the $\vec{S}_i$ are taken as unit vectors for the fcc-Fe crystal where the magnetic moments μ_i are all the same (2.2 Bohr magnetons [27]). In the real film the spins are corrected by the factor $\mu_i/2.2$ with the actual values μ_i of the magnetic moments taken from the recent first principle calculations by Spišák and Hafner [27]. This allows one to work with the single adjustable constant $J^{(2)}$. The calculation for the magnetic phase of the 6 ML film in the ground-state spin configuration (*uudduu*) [27] yields an upward shift $\Delta\omega(\mathbf{Q})$ of the RW frequency for positive values of $J^{(2)}$. Here *u* and *d* denote spin-up and spin-down layer configurations when a FM order is assumed within each layer. At $\mathbf{Q} = 0.53$ Å^{-1} the experimental shift of 0.51 meV is obtained for $J^{(2)} = 7$ N/m. This value agrees with the calculations by Sabiryanov and Jaswal for $r_s = 2.69$ a.u. [3] and confirms the magnetic origin of the observed RW frequency shift.

In summary we have shown that the temperature dependence of Rayleigh wave frequency in thin magnetic films is considerably affected by the ferromagnetic transition and can be used as a direct probe of the interplanar magnetic forces. These forces appear to be more effective in thin fcc-Fe films than in the ordinary bcc-Fe(110) surface in agreement with the first principle calculations of the magnon-phonon coupling constants by Sabiryanov and Jaswal [3]. This fact is likely to be related to large magnetostriction effects in the surface bilayer and assisted by the internal stress which is present in the substrate-stabilized fcc-Fe structure. Electron-energy-loss spectroscopy measurements by Daum *et al.* [28] on 1 to 5 ML thick films revealed an anomalous softening of the zone boundary RW frequency which has been associated with a negative (compressive) surface stress. Although in this work the surface stress has been associated with the epitaxial strain, the present HAS results suggest a magnetic contribution to such surface stress.

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