Impact of Atomic Structure on the Magnon Dispersion Relation: A Comparison Between Fe(111)/Au/W(110) and Fe(110)/W(110)

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We present a combined experimental and theoretical study of the interplay between the atomic structure and the magnon excitations in low dimensional ferromagnets. Two monolayer thick Fe films on W(110) with and without a Au buffer layer are investigated. Our experiments show that adding the Au layer leads to a significant softening of the magnons. First-principles calculations confirm the experimental results revealing a strong dependency of exchange interactions on the atomic structure. It is observed that the *intralayer* exchange interactions increase with increasing distance between Fe layers. This unusual relationship is attributed to the complexity of the electronic structure and the contribution of different orbitals to the hybridization and exchange interaction. Our results suggest a way of tailoring magnetic excitations in low-dimensional magnetic structures.

One of the fascinating aspects in solid state physics is the magnetic response of a ferromagnet to the change of its atomic structure. Of particular interest is the response of the exchange interactions that determine both ground magnetic state and spin excitations. Since the excitations crucially influence the dynamic as well as thermodynamic properties of magnets, the interplay between atomic structure and exchange interactions is of great importance for the design of magnetic nanostructures with desired functionality.

Although the impact of the structural changes on the magnetic interactions has already been discussed theoretically for the case of bulk ferromagnets like Fe (see, for example, Refs. [1,2]), it has not been proven experimentally. The main reason for this is that in nature there exists only one stable Fe bulk phase, i.e., the body-centered cubic (bcc) phase.

The modern methods of the fabrication of low-dimensional structures open an inspiring possibility of creating materials of given chemical content with different atomic structures. Since the atomic structure influences essentially the properties of the system, this possibility strongly enhances the potential of designing materials with desired properties. A crucial step on this way is to understand the relation between the atomic structure and the electronic properties.

In this Letter, we report experimental and theoretical study of the magnon excitations in two Fe films of two monolayer (ML) thickness that differ in the atomic structure. On one hand, ultrathin Fe(110) films grow pseudomorphically on W(110) in bcc stacking [3,4]. On the other hand, by introducing an ultrathin Au buffer layer with the thickness of 2 ML, one can obtain an Fe(111) film with a close-packed structure [5,6].

Our measurements show a large difference in the magnon dispersion relation of the two films. Surprisingly, the intralayer exchange interaction increases when the distance between Fe atomic layers increases. The response of the exchange interaction to the change of the atomic structure and interlayer distance is investigated in detail. Our first-principles calculations confirm the experimental results and provide a deeper insight into the microscopic origin of this effect.

All the experiments are performed under ultrahigh vacuum. Prior to the film deposition, the surface of the W(110) substrate was prepared using our standard cleaning procedure [7]. The Au and Fe films were grown by molecular beam epitaxy at 500 and 300 K, respectively. Tungsten has a bcc structure and the (110) surface is composed of rectangular unit cells. Since the gold crystal structure is face-centered cubic (fcc), the most similar surface to W(110) is the Au(111) surface. The Au ultrathin films grow in fcc(111) structure on W(110) as is verified by our low-energy electron diffraction (LEED) experiments. The LEED patterns recorded for a 2 ML Au film on W(110) indicate that Au does not grow in the same structure as W(110). The most probable structure is Au(111) with the epitaxial relationship $\mathrm{Au_{fcc}}[1\bar{1}0] \parallel \mathrm{W_{bcc}}[001]$ known as the Nishiyama–Wassermann relationship. A similar observation is also reported by other groups [8,9].

The growth and the structure of the Fe films on the flat and vicinal Au(111) surfaces have been intensively investigated [10–12]. It has been shown by scanning tunneling microscopy that ultrathin Fe films grow pseudomorphically on Au(111) from the initial stage of growth up to a film thickness of about 2 ML. This is confirmed by our IV-LEED analysis. The evolution of the average interlayer distance with the number of Fe layers shows that for Fe thicknesses below 2 ML one sees a pseudomorphic growth of Fe on the Au(111) film. The films start to relax at thicknesses above 2 ML and finally, at an Fe thickness of

about 3.5 ML, the Fe film reaches the value of the layer spacing of the Fe bulk. A detailed analysis reveals that the LEED pattern is a slightly distorted hexagon. As the unit cell of Au possesses a threefold symmetry, this distortion results from the epitaxy of the Au(111) film on the W(110) surface. A similar observation is reported by Zdyb *et al.* [5,6].

Since for the Fe/Au/W(110) samples with the Fe film thickness above 2 ML, a structural transformation to the bcc structure occurs, we restrict our investigations to the samples composed of 2 ML Fe. The magnetic state of the samples was checked by means of the magneto-optical Kerr effect in longitudinal geometry with an external magnetic field applied along the W[1$\bar{1}$0] direction (Fe[$\bar{2}$11] direction). The rectangular hysteresis loop shows a typical easy axis behavior (see the inset of Fig. 1).

The magnons are probed by means of spin polarized electron energy loss spectroscopy (SPEELS), which has opened a possibility to measure the magnons in such ultra-thin structures [13–20]. The magnon dispersion relation is measured along the $\bar{\Gamma}$-$\bar{K}$ direction of the surface Brillouin zone. The SPEELS experiments are performed with an incident electron energy of 3.96 eV and a total energy resolution of about 14.9 meV. Figure 1 shows typical spin-down ($I_\downarrow$) and spin-up ($I_\uparrow$) SPEELS intensity spectra

recorded on 2ML Fe/2 ML Au/W(110) at room temperature and at a wave-vector transfer of $\Delta K_\parallel = 0.75$ Å^{-1}. $I_\downarrow$ ($I_\uparrow$) indicates the intensity of scattered electrons when incoming electrons have the spin polarization parallel (antiparallel) to the sample magnetization. $\Delta K_\parallel$ is the wave-vector transfer parallel to the surface of the film. It is determined by the momenta of the incident and scattered electrons and the scattering geometry [21]. The peak at 68 meV in the minority channel, $I_\downarrow$ (more clearly in the difference spectrum, $I_{\text{Diff.}} = I_\downarrow - I_\uparrow$) is due to the magnon excitation. The magnon peak shows a clear dispersion with the variation of the wave-vector transfer [see Fig. 1(b)]. In Fig. 1(b), the difference spectra for various in-plane wave-vector transfers are shown.

By plotting the excitation energy versus the wave vector, one obtains the magnon dispersion relation, as shown by solid circles in Fig. 2(a). The open circles are experimental data obtained earlier on a 2 ML Fe film directly grown on W(110) [16]. Comparison of the experimental magnon
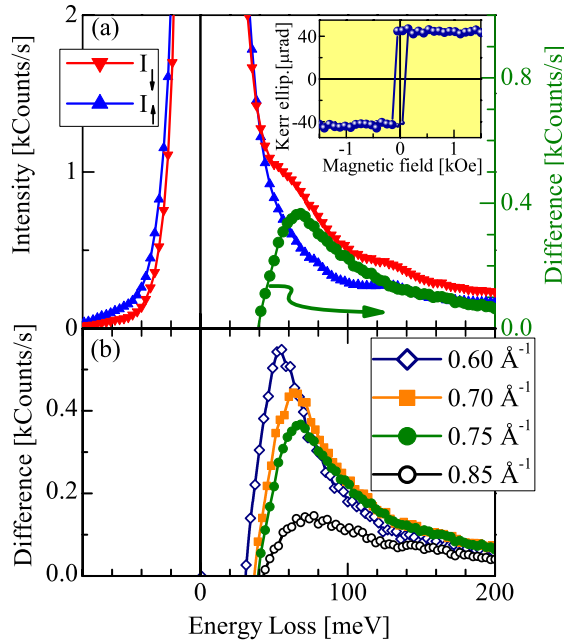

FIG. 1 (color online). (a) Typical spin polarized electron energy loss spectra recorded on 2 ML Fe/2 ML Au/W(110) at a wave-vector transfer of $\Delta K_\parallel = 0.75$ Å^{-1} and at room temperature. The difference spectrum ($I_{\text{Diff.}} = I_\downarrow - I_\uparrow$) is shown as solid circles. Inset shows a typical hysteresis loop recorded in longitudinal geometry with an external magnetic field applied along the W[1$\bar{1}$0]-direction (Fe[$\bar{2}$11]-direction). (b) Difference spectra measured at different wave-vector transfers from 0.6 to 0.85 Å^{-1}.
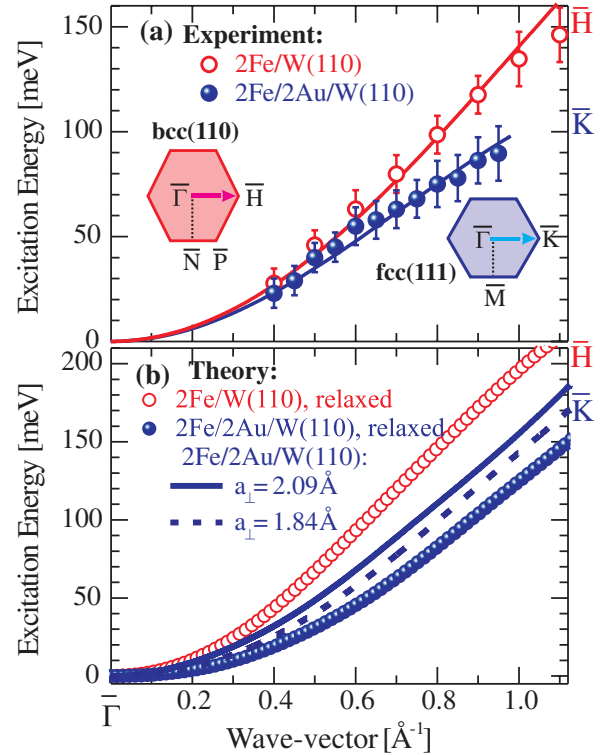

FIG. 2 (color online). (a) Experimental magnon dispersion relation measured on a 2 ML Fe on 2 ML Au on W(110) at room temperature. The results of a 2 ML Fe directly grown on W(110) are also shown [16]. The symbols represent the experimental results. The solid lines are the guide to the eyes. (b) Theoretical magnon dispersion relation of 2 ML Fe(111)/2 ML Au(111)/W(110) and 2 ML Fe(110)/W(110). The symbols represent the results of the calculation for the relaxed structure. The lines without symbols are the results for the Fe/Au/W(110) system calculated for different values of Fe interlayer spacing, $a_\perp$.

dispersions of the two systems reveals a strong softening of the magnon dispersion relation in the sample with a Au buffer. For instance, the softening at $\Delta K_\parallel = 0.9$ Å^{-1} reaches a value of 35 meV. This result demonstrates a way of tailoring magnetic excitations in low-dimensional structures by materials engineering.

Our next aim is to understand the microscopic nature of the observed effect on the basis of first-principles calculations. The calculations are performed within the generalized gradient approximation of the density functional theory [22]. The crystalline structure of Fe/W(110) was taken from the surface x-ray diffraction experiment [4], while the atomic positions in Fe/Au/W(110) were obtained using the VASP code, well known for providing accurate total energy and forces [23,24]. The structural information serves as an input for calculations of electronic and magnetic properties using a self-consistent Green function method, which is specially designed for layered semi-infinite systems [25]. The Heisenberg exchange parameters were determined employing the magnetic force theorem, likewise implemented within the Green function method [26].

The calculated magnon dispersion relation for both systems is presented in Fig. 2(b). The calculations are in good agreement with the experimental results, apart from the fact that they differ slightly in the absolute values of the energy. This means that the magnon softening is a consequence of adding the Au buffer. The analysis of the exchange parameters shows a strong anisotropy of exchange interaction for the systems we consider (see Fig. 3). The strongest interaction takes place between atoms of different layers. On the other hand, in the analysis of the magnon energies one should take into account that the number of the nearest neighbors within the layers is much larger than between the layers. In the following, the exchange interaction between atoms within the same atomic layer is referred to as *intralayer* interaction and the interaction between atoms from different layers is referred to as *interlayer* interaction.

The comparison of the exchange interactions for two systems, Fe/W(110) and Fe/Au/W(110), shows that the interlayer and intralayer interactions feature opposite trends. As an example, the largest interlayer interaction, $J_\perp$, increases from 27 meV in Fe/W(110) to 69 meV in Fe/Au/W(110) whereas the largest intralayer exchange interaction, $J_\parallel$, decreases from 20 to 14 meV for the interface layer and from 13 to 9 meV for the surface layer. The analysis shows that the softening of the magnons in Fe/Au/W(110) is the consequence of the decreased intralayer interactions that overcomes the opposite trend of increasing interactions between the layers.

The exchange parameters between Fe moments of the Fe/Au/W(110) system do not change significantly if we repeat the calculations for a free standing Fe film (keeping the atomic arrangement of the Fe/Au/W(110) system).
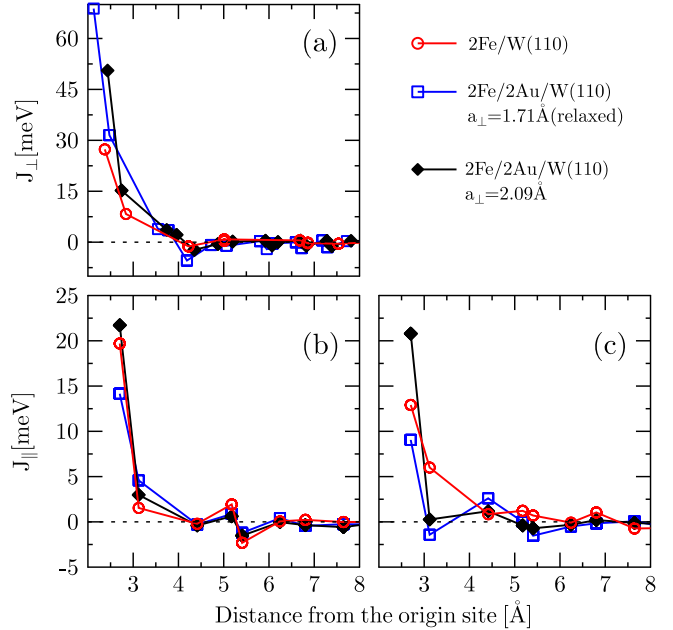

FIG. 3 (color online). Calculated site-resolved interlayer, $J_\perp$, (a) and intralayer, $J_\parallel$, (lower panels) exchange constants for the atoms located in the interface layer (b) and in the surface layer (c). Open and filled symbols represent the calculations for the relaxed and expanded structures, respectively.

Therefore, the role of the Au buffer is mostly reduced to the modification of the Fe atomic lattice. The change in the exchange parameters is the consequence of the change in the electronic structure caused by the modification of the Fe lattice.

In the epitaxial growth, the interatomic distance within the layers is determined by the atomic structure of the underlying lattice whereas the interlayer distance is governed by complex interplay of different interactions and can vary strongly from film to film. It is instructive to study the dependence of the exchange interactions and the magnon dispersion relation on the distance between Fe layers of the Fe/Au/W system. The prediction of the dependence of the effective interatomic exchange interactions on the interatomic distance is a difficult task because of a complex competition of various effects. On one hand, increasing distance between atoms leads to the narrowing of electron bands and increased atomic moments. On the other hand, it leads to a weaker overlap of the states of the atoms and produces the trend to decreasing interatomic exchange interaction [27].

Our calculations show that increasing the interlayer distance leads to an expected decrease of the interlayer exchange parameters. This results from the fact that a relatively small increase of the atomic moments is over-compensated by decreasing interlayer hybridization. Unexpectedly, at the same time we observe an increase in the energies of the acoustic magnons that become closer to the corresponding energies of the Fe/W(110) magnons.

Further analysis of the calculated exchange parameters shows that the origin of the increased magnon energies is in the strong increase of the intralayer exchange parameters. We emphasize that this increase takes place for unchanged interatomic distances within the layers. It is a consequence of complex reconstruction of the electronic structure due to the increase of the interlayer distance. Such a behavior cannot be understood without detailed first-principles calculation of the electronic structure. While changing the interlayer distance of Fe layers in Fe/Au/W(110) structure from 1.71 to 2.09 Å, the nearest neighbor intralayer coupling increases by a factor of 1.5 and 2.3 for the atoms in the interface and surface layer, respectively. This increase in the values of the intralayer exchange parameters is much larger than the increase of the atomic moments.

To understand the microscopic mechanism of the formation of effective interatomic exchange parameters, it is necessary to consider the consequence of the deviation of the atomic moments from the ground state directions. The larger the increase in the energy of the system following the deviation of the moments, the larger are the effective interatomic exchange parameters. The change in the total energy is a cumulative effect of the changes in the energies of individual electronic states. In a multiple-band real system the change in the electronic structure is complex and the reduction of the change in the total energy to a small number of "hot spots" in the electronic structure is usually not possible. Instead, we perform the analysis of the features of the density of states (DOS) that can contribute to the discussed effect. An important part of the response of the electronic systems to the deviation of the atomic moments is the hybridization of the spin-up and spin-down electron states of the collinear ground state of the system. The strength of the hybridization depends on the energy distance between hybridized states and on the overlap of their orbital wave functions. Since the hybridization leads to the formation of binding and antibinding states, the changes in the electronic energies strongly compensate each other if both binding and antibinding levels are occupied. However, if the hybridization involves the states lying close to the Fermi level, the compensation can be disturbed when certain unoccupied states become involved, since unoccupied states do not contribute to the total energy.

The analysis reveals important trends in the density of states. The $3d$ states responsible for the interlayer hybridization (d_{xz}, d_{yz}, and d_{z^2}) appear substantially higher in energy than the states responsible for the intralayer hybridization ($d_{x^2-y^2}$ and d_{xy}). Figure 4 shows the spin-resolved DOS of $3d$ electrons calculated for different interlayer distances. In order to see the contribution of different $3d$-orbitals to the exchange interactions, their projected DOS are plotted in Figs. 4(c) and 4(d). With increasing interlayer distance all states move to lower energies as a
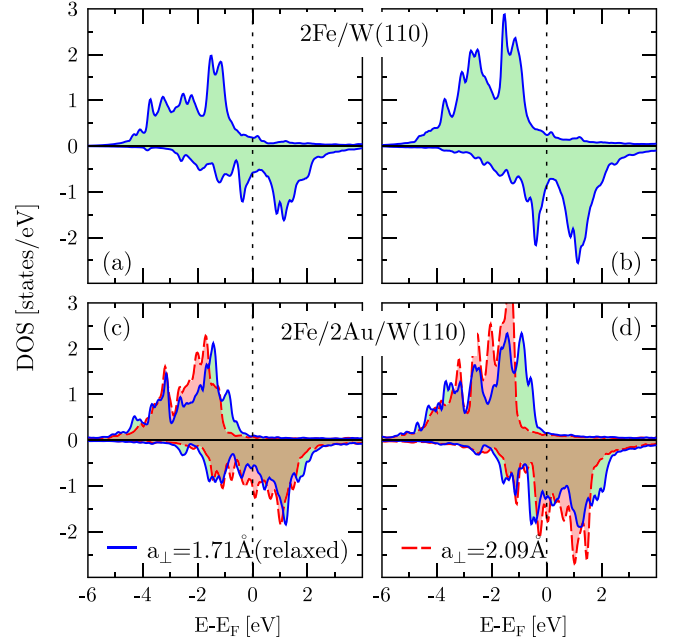

FIG. 4 (color online). Spin-resolved DOS of $3d$ states in 2Fe/W(110) (upper panels) and 2Fe/2Au/W(110) (lower panels) systems, separated in a part with d_{xy} and $d_{x^2-y^2}$ states (a),(c) and a part corresponding to d_{xz}, d_{yz} and d_{z^2} orbitals (b), (d). The solid and dashed lines represent the results for relaxed and expanded structures, respectively.

consequence of decreasing $3d$ band width. However, this shift is more important for d_{xz}, d_{yz}, and d_{z^2} states, since $d_{x^2-y^2}$ and d_{xy} are located well below the Fermi level for the minimal value of the interlayer distance. On the other hand, increasing the interlayer distance increases the spin-down density of $3d$ states on the Fermi level. The appearance of a large number of states near the Fermi energy makes the energy of the system sensitive to the deviation of the atomic moments and constitutes an important factor in the enhancement of intralayer exchange interactions. The discussion above indicates that the magnetic properties of complex systems cannot be understood without careful microscopic study of the exchange interactions. While the interlayer exchange parameters decreased as expected, the intralayer exchange parameters turned out to increase, upon increasing the interlayer distance, due to the reconstruction of the electronic structure. Clearly, the evolution of the electronic structure cannot be separated into features related to the interlayer and intralayer distances since the influences of both distances are strongly interconnected.

In summary, we have shown both experimentally and theoretically that the engineering of the atomic structure of low-dimensional magnets leads to a strong modification of the exchange interaction that provides a route to the design of the materials with desired magnetic properties. The analysis of the variation of the Heisenberg exchange parameters with the variation of the interlayer distances shows an unexpected effect of the strong increase of the

intralayer exchange interaction overcompensating the decrease of the interlayer exchange parameters. This behavior is not restricted to the systems studied here. It is expected also for ultrathin Fe films grown on other fcc surfaces. This demonstrates the necessity of the combined experimental-theoretical approach to the complex physical properties of the real materials and reveals strong potential of such studies for the design of new materials with desired properties.

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