Surface Science 630 (2014) 311–316

Contents lists available at ScienceDirect

Surface Science

journal homepage: www.elsevier.com/locate/susc

Magnetic excitations in ultrathin magnetic films: Temperature effects

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ARTICLE INFO

Available online 21 July 2014

Keywords:
Magnetic excitations
Magnons
Spin waves
Magnetic monolayers
Spin dynamics

ABSTRACT

The idea of investigating large wave-vector magnetic excitations in ultrathin films by spin-polarized electron spectroscopy is briefly reviewed. The historical background of the paper is based on the personal experience of the authors who collaborated and discussed with Douglas Mills regarding this subject. Douglas Mills' impact on the understanding of fundamental mechanisms involved in the excitation process and the development of the theory of magnetic excitations is outlined.

In addition, the temperature effects on the large wave-vector magnetic excitations in ultrathin Fe films are addressed. The experimental results of magnon excitations in the pseudomorphic Fe monolayer on W(110) are presented. The temperature dependence of the magnon dispersion relation is discussed.

1. Introduction

Magnetic excitations belong to one of the most important concepts in magnetism and have been intensively investigated over several decades, as they are the fundamental basis of understanding many phenomena in solids. Generally, in a given magnetic solid there is a large variety of magnetic excitations, depending on the wavelength of these excitations and the magnetic interactions involved. On the atomic length scales the dominating magnetic interaction describing the coupling of the neighboring moments is the magnetic exchange interaction. On these length scales the exchange dominated magnetic excitations with short wavelength (large wave-vector) are important. These types of excitations possess energies on the order of a few millielectron-volts up to a few hundred millielectron-volts and are sometimes referred to as high-energy magnetic excitations.

Experimentally, these excitations have already been investigated by means of inelastic neutron scattering in bulk magnetic materials [1–13]. The dispersion relation of elementary magnetic excitations (magnons) has been measured over a large fraction of the Brillouin zone (only a small part of the Brillouin zone, close to the zone center, has not been measured due to the limited energy resolution at that time). Nowadays one can perform inelastic neutron scattering experiments with an extremely high energy resolution, for example by using cold neutrons or the so-called neutron spin echo technique. The interaction between neutrons' spin and the magnetic moment of the unit cell leads to excitation of a magnon in the system. As this interaction is of dipolar nature, it is rather weak and hence neutron scattering cannot be employed to investigate the magnons in small size structures or ultrathin films.

Probing magnetic excitations in low-dimensional magnets has been of great fundamental importance for understanding their novel properties. For probing the uniform mode (zero wave-vector) and also short wave-vector magnetic excitations in low-dimensional magnets, techniques like ferromagnetic resonance and Brillouin light scattering have been developed [14–19]. However, still a novel experimental technique was required for probing the large wave-vector excitations. One idea was to use electrons as probe tools.

In this paper we briefly review the progress in the field of spin-polarized electron energy loss spectroscopy and its application to investigate the magnetic excitations in ultrathin magnetic films. The focus is put on Douglas Mills' impact on the development of the theory of electron scattering and its use for investigating magnetic excitations. In Section 2 the basic idea of using electrons for probing magnons is discussed. One of the main goals was to take advantage of strong electron–matter interaction to probe the magnetic excitations in an ultrathin film with the thickness of one monolayer (ML). In Section 3 the experimental results of magnon excitations in 1 ML Fe(110) pseudomorphically grown on W(110), measured at different temperatures, are presented. The influence of the temperature on the magnons' energy and lifetime is discussed.

2. Basic concepts: magnon excitations by SPEELS

Generally, when an electron is inelastically scattered from a surface, it can create elementary excitations in the system. The interaction involved in this process is of electrostatic Coulomb nature and hence it is rather strong. The process would allow probing elementary excitations

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http://dx.doi.org/10.1016/j.susc.2014.07.011
0039-6028/

in a monolayer (or even sub-monolayer) of a ferromagnet. The substantial developments in the field of electron energy loss spectroscopy (EELS) [20] and its combination by spin-polarized electron sources [21] made the spin-polarized version of EELS a powerful tool for the investigation of large wave-vector excitations in ferromagnetic thin films and monolayers. The main idea to follow was to use the electrons to excite and probe the magnetic excitations at a ferromagnetic surface, similar to surface phonons by EELS. In this case the use of a spin-polarized beam would help to unambiguously reveal the nature of excitations. As the total angular momentum of a magnon is $1\hbar$, for exciting a magnon one should be able to transfer $1\hbar$ angular momentum to the sample. This would mean that if an incoming electron with the spin state of $-1/2$ has impinged onto the sample surface, it should be scattered into an electron with the spin state of $+1/2$ by creating a magnon. However, there have been a few open questions at the early stage of this idea: (i) Is such a process allowed? (ii) What is the main physical mechanism behind it? (iii) What are the fundamental interactions involved during the scattering? (iv) What is the typical timescale of such a process? (v) Is this timescale shorter than the lifetime of magnons?

To answer these questions, a series of experiments were performed in the 80s [23–27]. One of the experiments which shed light on these questions was performed in 1985 in which the intensity of the scattered beam was measured for different spin configurations of the incoming and scattered beam in a so-called "complete experiment". In that experiment, which was performed on an Fe(100) surface, a spin-polarized source was used and the spin polarization vector of the scattered beam was measured [25]. It turned out that the so-called "spin-flip" excitations, which lead to the change of the total angular momentum of the sample by $\pm 1\hbar$, can indeed take place. In addition, it was observed that the intensity of the processes in which the incoming spin state is of minority character ($-1/2$) and the scattered one of majority character ($+1/2$) is dominating the others. The theory of the inelastic scattering of spin-polarized electrons from a magnetic surface was developed by Douglas Mills and co-workers [28]. It was proposed that, in principle, electrons can efficiently contribute to the creation of magnetic excitations when they are scattered from a magnetic surface. The development of the theory of magnetic excitations in model ferromagnets was also done almost at the same time [29]. These two aspects were combined to understand the mechanisms involved in magnon excitations by spin-polarized electron energy loss spectroscopy (SPEELS) [30–33].

The first signature of large wave-vector magnon excitations in SPEELS was observed in the experiments performed in Halle [22]. The observation was explained based on the theory developed in the group of Douglas Mills. Fig. 1 shows the first experimental observation of magnetic excitations in spin-polarized energy loss spectra. A comparison to the calculated intensity spectra indicates that the observed excitation is of spin-flip nature. The experiment was performed using a primary beam with an energy of $E_i = 29$ eV and at a wave-vector transfer of $\Delta K = 0.68$ Å^{-1} ($|\Delta K| = |q|$, where, q is the wave-vector of the excited magnon). The spin-dependent excitation observed at a loss energy of about 100 meV was attributed to the magnetic excitations, in line with the theoretical calculations (see Fig. 1(b)). The total energy resolution was about 100 meV and hence it was rather difficult to resolve the magnon peak. The large drop in the asymmetry curve observed below 200 meV is caused by the tail of the quasielastic peak. The small negative value of the asymmetry in the region of quasielastic peak is due to the fact that electrons with different spin orientations experience a different scattering potential when they are quasielastically scattered from the sample surface. The nature of this asymmetry is different from the one in the loss region, where the magnon peak exists. The value and the sign of the asymmetry in the region of the quasielastic peak depend strongly on the energy of the primary beam and also the scattering angles. The sign of the spin asymmetry cased by magnon excitations in the energy loss region is always positive. This is due to the exchange process of minority electrons with majority ones which leads to the creation of magnons. It was hoped that with an improved energy resolution the effect caused by quasielastic scattering shall be suppressed, allowing a precise determination of the excitation energy. The degree of the spin polarization of the incoming beam for this experiment was about 30%. It was hoped that an improved spin resolution would substantially increase the value of the spin asymmetry.

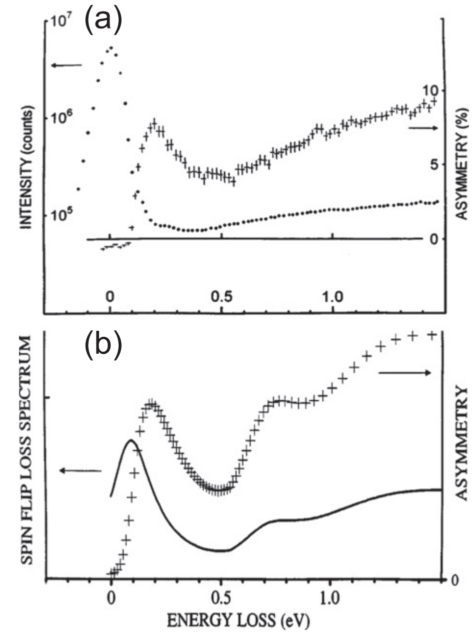

Fig. 1. The first experimental observation of magnetic excitations in spin-polarized energy loss spectra. (a) The experimental spin averaged intensity ($I_{Av.} = I_\downarrow + I_\uparrow/2$) and asymmetry ($Asy. = I_\downarrow - I_\uparrow/I_\downarrow + I_\uparrow$) spectra measured by SPEELS on an ultrathin Fe(110) film grown on W(110). (b) The calculated spin-flip loss and asymmetry spectra.
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This successful experiment was a strong motivation to build up a SPEELS set-up with a better energy and spin resolution [34]. The new set-up allowed the measurement of the magnon dispersion relation over the entire surface Brillouin zone for ultrathin Co(001) [35–38] and Co(0001) [39] films. The technique was successfully employed to investigate the magnetic excitations in ultrathin Fe(110) [40–45], Fe(111) [46], Fe(001) [47] and very recently in FeCo(001) films (with an out-of-plane easy axis) [48]. In addition to the magnons' energy, the lifetime of excitations could also be studied [49–51]. The technique is also capable of probing magnons and phonons, simultaneously [52]. Recently, an EELS set-up (without spin resolution) is used to investigate the magnetic excitations in Co(001) films [53–55]. The observed loss features in the spectra are attributed to the magnetic excitations. This interpretation is based on the knowledge of spin-resolved measurements obtained earlier by SPEELS [35].

3. Magnons in 1 ML Fe(110)/W(110)

One of the main goals of SPEELS experiments was to investigate the magnons in a ferromagnetic film with the thickness of 1 ML. Probing the magnon dispersion relation in a ferromagnetic monolayer would help for a better understanding of magnetism in ultrathin ferromagnetic films. The successful experiment was performed in 2009 on a pseudomorphic Fe(110) film on W(110) [41]. It is well-known that an Fe(110) monolayer grown on W(110) shows rather good structural and morphological properties and a high thermodynamic and chemical stability. Hence it can be regarded as a prototype ferromagnetic monolayer. The system is ferromagnetic at temperatures bellow 223 K [56].

The magnon dispersion relation was measured at 120 K along the main symmetry $\overline{\Gamma}-\overline{\mathrm{H}}$ direction of the surface Brillouin zone. It was found that the excitation energies are rather low and the peaks associated with magnon excitations were rather broad. For instance, at $q = 1.1$ Å^{-1} an energy of about 60 meV was measured. The results of the experiments were in sharp contrast to the results obtained by the theory based on the empirical tight-binding approach [57–59]. The calculated magnon energies within this theory were very high (up to 600 meV, at the Brillouin zone boundary) compared to the results of the experiment [59]. It was suggested that one reason for this discrepancy may originate in the initial tight-bounding parameters used as the starting point of the calculations, i.e. the Fe bulk parameters. A more appropriate way of describing the magnon dispersion relation in ferromagnetic films adsorbed on a substrate would be to start with the parameters calculated for the same system in which all the magnetic, structural and chemical relaxations are taken into account.

A way of improving the theory was later suggested in Ref. [60] in which the tight-binding parameters were obtained by other methods e. g. from real-space linear muffin-tin orbital atomic sphere approximation calculations or by fitting the Korringa–Kohn–Rostoker-based electronic-structure calculations. The results obtained in this way were in a much better agreement with the experiment [60]. This approach could also allow the investigation of the relativistic spin–orbit effects on the magnons' energy and lifetime.

Meanwhile a computational scheme was developed based on the time-dependent density functional theory [61]. The results obtained in this framework were in a good agreement with the experiment [62]. A combination of density functional theory and atomistic spin dynamics simulations showed that in fact if the magnetic, structural and chemical hybridizations are properly taken into account the theory and experiment would agree very well [63]. Another parameter which would be of importance in determining magnons' energy is the effect of temperature [63,64]. This effect will be discussed in detail in the following section. The conclusion of the theoretical results was that the electronic hybridizations of the film with the substrate, chemical and magnetic relaxations are of great importance in determining the magnons' energy. The main contribution to the electronic hybridizations originates from the hybridizations of the W(110) surface states with the Fe *d*-states. The electronic hybridizations influence not only the magnons' energy but also their lifetime [65]. It was also observed in the experiment that the excitation peaks are rather broad, meaning that the lifetime of the magnons excited in the Fe film is rather short. In addition to that the spin–orbit coupling of the surface state in W(110) is rather large. This has also an effect on the magnons excited in the Fe film. Since in the case of Fe monolayer the inversion symmetry is broken, the large spin–orbit coupling can, in principle, lead to a large antisymmetric interaction, known as the Dzyaloshinskii–Moriya (DM) interaction [66,67]. If the DM interaction is strong enough, it can change the properties of the high-energy magnetic excitations in the system [68]. The consequence of the DM interaction on the magnon dispersion relation in ferromagnetic Fe films grown on W(110) is to lift the energy degeneracy of magnons [68,60,63,69]. In addition, the spin–orbit coupling influences the magnons' lifetime [60]. Both effects were observed in the experiment [43,44,50].

4. Temperature dependence of the magnons' energy

In Fig. 2 the SPEELS spectra recorded on the Fe monolayer on W(110) at $T = 10$ K and at $q = 0.55$ Å^{-1} are presented. The magnon excitation appears as a triangle-like feature near the quasielastic peak in the minority ($I_\downarrow$) spectrum. It shows up as a peak in the difference ($I_\downarrow - I_\uparrow$) and asymmetry ($I_\downarrow - I_\uparrow/I_\downarrow + I_\uparrow$) spectra. The small peaks around 70 meV in both minority ($I_\downarrow$) and majority ($I_\uparrow$) spectra originate from the vibrational states of adsorbed oxygen physisorbed on the surface. At low temperatures a very small amount of the residual gases in the chamber adsorbs on the surface and since low energy electrons are extremely surface sensitive, one observes the peaks caused by vibrational excitations of the adsorbed gases. These peaks do not exist in the first measurement scan. Their intensity grows with time. Such peaks do not influence the magnon excitation peak as they do not show any spin dependence. The magnon excitation peak can be clearly observed in the difference and asymmetry spectra, shown in Fig. 2(b). When electrons of different spin characters are quasielastically scattered from a magnetic surface, they experience a different scattering potential. Hence the intensity of the quasielastic peak for each spin orientation is different. This fact leads to a non-zero difference ($I_\downarrow - I_\uparrow$) at the energy loss of 0 meV (and consequently a non-zero asymmetry, in this case of about -3%). The value and the sign of this difference depend on the energy of the incoming beam and also on the scattering geometry. The magnon excitation peak, centered at the loss energy of about 30 meV is rather broad, meaning that the lifetime of the excitation is relatively short. In order to investigate the effect of the temperature on the magnon excitation peak the same spectra are recorded at 120 K. Fig. 3 provides a direct comparison between the difference spectra measured at different temperatures. The spectra presented in Fig. 3 are recorded at $q = 0.6$ Å^{-1} and at $T = 10$ K and 120 K. It is apparent that the excitation peak moves towards lower energies when temperature increases from 10 to 120 K. The peak broadening is almost unchanged. This implies that magnons' energy in the ferromagnetic monolayer is temperature dependent in line with the prediction by spin dynamics simulations [63]. The lifetime does not drastically change when changing the temperature. Note that the intensity of the difference spectra cannot be compared, since in these experiments the intensity of the primary beam is different.

In Fig. 4 the magnon dispersion relation measured at different temperatures is presented. The magnons' energy decreases as temperature increases. Since the magnons are strongly damped in the Fe monolayer on W(110), the excitation does not show up as a well-defined peak in $I_\downarrow$. The only way to extract the magnon energy from the measured experimental spectra is to follow the peak in the difference spectra. For

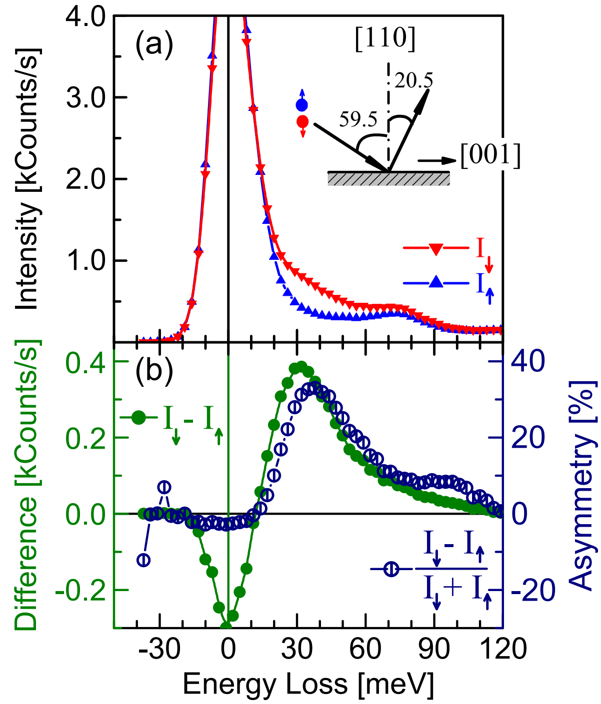

Fig. 2. (a) The experimental SPEELS spectra recorded for the incoming spin state of minority ($I_\downarrow$) and majority ($I_\uparrow$) states on the Fe(110) monolayer grown on W(110) at $q = 0.55$ Å^{-1}. Inset shows the measurement geometry. (b) The difference ($I_\downarrow - I_\uparrow$) and asymmetry ($I_\downarrow - I_\uparrow/I_\downarrow + I_\uparrow$) spectra. The data are recorded at $T = 10$ K. The energy of the primary beam was set to 4.4 eV.

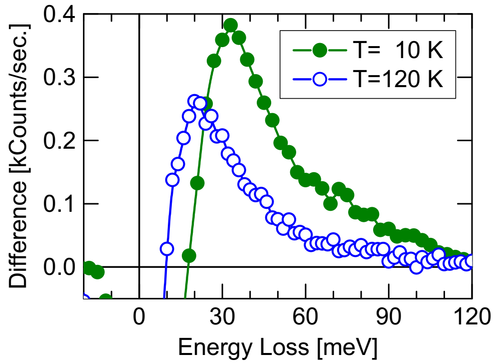

Fig. 3. A comparison between the difference spectra ($I_{\downarrow} - I_{\uparrow}$) recorded at $T = 10$ and 120 K. The data are measured at $q = 0.6$ Å^{-1}. The peak position moves towards lower energies when temperature increases from 10 to 120 K. The peak broadening is almost unchanged while changing the temperature. The data are recorded using a different primary beam intensity, therefore the intensity of the magnon peak cannot be compared.

temperatures higher than 190 K the intensity of the difference spectra becomes very low such that it is very difficult to determine the magnon energy from the measured spectra.

We now attempt to estimate the effective interatomic exchange parameter using a solution of the effective Heisenberg spin Hamiltonian. As one expects for a spin system with magnetic anisotropy, the experimental data suggest a gap in the magnon dispersion at $q = 0$ [59,68,70,71]. Hence we add a term in the spin Hamiltonian representing the effective magnetic anisotropy K_{eff} of the system with an easy axis along the unit vector $\hat{n}$. For the Fe monolayer the easy magnetization axis is along the $\left[1\bar{1}0\right]$ direction. The extended Heisenberg spin Hamiltonian can be written as: $H = -(1/2)J\sum_{<i,j>} \mathbf{S_i} \cdot \mathbf{S_j} - K_{eff}\sum_i (\mathbf{S_i} \cdot \hat{\mathbf{n}})^2$, in which J denotes the effective isotropic interatomic exchange coupling constant between spins S_i and S_j. For the pseudomorphic monolayer and considering only the nearest neighbor interactions the dispersion relation along the $\bar{\Gamma}-\bar{\mathrm{H}}$ direction of the surface Brillouin zone can be written as: $\hbar\omega = 4JS[1 - \cos(qa_0/2)] + 2K_{eff}S$, where S denotes the magnitude of the atomic spin (and can be set to 1) and $a_0 = 3.165$ Å is the lattice constant of the pseudomorphic Fe monolayer grown on W(110) (it is the same as the W substrate). In order to reduce the fit parameters and for simplicity we do not take into account the DM term in our spin Hamiltonian. The best way of estimating the DM term is to measure the so-called energy asymmetry [$\Delta E = E(q) - E(-q)$]. Such data are measured for an Fe double layer on W(110) in Ref. [43]. The best fits to the experimental data are obtained with $JS = 11 \pm 1$ meV and $2K_{eff}S = 4.6 \pm 2.5$ meV for the data recorded at $T = 120$ K and $JS = 13 \pm 1$ meV and $2K_{eff}S = 8 \pm 3$ meV for the data recorded at 10 K. The fits are presented in Fig. 4 as solid lines. The obtained values of effective interatomic exchange constant are in very good agreement with the JS^2 value estimated from the analysis of the magnetic domain walls in 1 ML Fe grown on the vicinal surface, where a $JS^2(S \equiv 1)$ of 14 meV (at $T = 14$ K) is measured [72]. The obtained anisotropy constant $K_{eff}S$ is similar to the effective anisotropy ($K_{eff} = 4.2$ meV/atom, at $T = 14$ K) reported by Pratzer et al. [72]. This analysis suggests that both the effective magnetic anisotropy and the effective exchange coupling are temperature dependent and decrease as temperature increases.

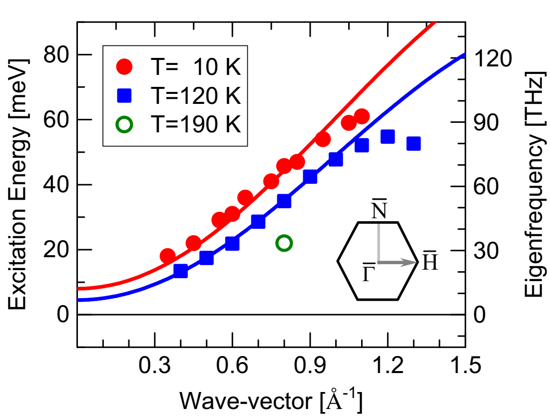

Fig. 4. The magnon dispersion relation measured for a monolayer of Fe(110) film on W(110) at different temperatures. The symbols represent the experimental data. The solid lines are the fits based on the nearest neighbor Heisenberg model. The data at 120 K are taken from [41]. The error bars in energy are: about ± 6 meV for low wave-vectors ($q < 0.5$ Å^{-1}), ± 4 meV for intermediate wave-vectors ($0.5 < q < 1$ Å^{-1}) and ± 10 meV for the large wave-vectors ($q > 1$ Å^{-1}). Inset shows the surface Brillouin zone.

At this point we would like to mention that this simple analysis does not provide the actual value of the magnetic exchange parameters of the system. The reason is twofold: first, in this model only the nearest neighbor interaction is taken into account. In reality one would expect a relatively complex pattern of exchange parameters and the interactions between neighbors with longer distances are also important (see for example Ref. [47]). Second, this simple analysis does not provide a separation of the temperature dependence of local moment from the temperature dependence of the interatomic exchange parameter, as only the quantity JS can be estimated from the experimental data. In reality both J and S are temperature dependent and their temperature dependence may be different. However, the effective exchange coupling extracted from the experimental data using this approach describes the "stiffness" of magnons. The effect of the temperature is to soften the magnons. The ratio of the effective exchange coupling, measured at different temperatures, quantifies the magnon softening caused by temperature.

5. Conclusions

Similar to many other breakthroughs that happened in physics, the collaboration between the experiment and theory eventually made the measurement of high wave-vector magnons in ultrathin ferromagnets possible. This could greatly deepen our understanding of magnetism in low-dimensional magnets. High wave-vector magnetic excitations can be efficiently excited by inelastic scattering of spin-polarized electrons from the sample surface. The fundamental mechanism involved in the excitation process is the exchange scattering mechanism. As the exchange process is mediated by the electrostatic Coulomb interaction, it is very efficient. This fact allows one to investigate magnons in very thin magnetic structures such as ferromagnetic monolayers.

The experimental results of magnon excitations in Fe monolayer revealed that the magnons' energy decreases as temperature increases whereas the magnon's lifetime does not show a strong temperature dependence. A simple analysis of the measured magnon dispersion relation in Fe monolayer on W(110) showed that the effective interatomic exchange parameter JS and the effective magnetic anisotropy $K_{eff}S$ increase by about 2% and 70%, respectively, when temperature is decreased from 120 to 10 K.

Acknowledgment

Fruitful discussions with A. Bergman and A. Taroni are gratefully acknowledged.

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