Elementary Excitations at Magnetic Surfaces and Their Spin Dependence

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The elementary surface excitations are studied by spin-polarized electron energy loss spectroscopy on a prototype oxide surface [an oxygen passivated Fe(001)-$p(1 \times 1)$ surface], where the various excitations coexist. For the first time, the surface phonons and magnons are measured simultaneously and are distinguished based on their different spin nature. The dispersion relation of all excitations is probed over the entire Brillouin zone. The different phonon modes observed in our experiment are described by means of *ab initio* calculations.

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It was realized at the very beginning of the development of the quantum theory of condensed matter that, although solids are composed of atoms, they cannot be described using atomic properties only. The reason lies in the collective phenomena that appears in solids. They should undoubtedly be taken into account when excitations are studied. Collective excitations in solids can be described by their representative quasiparticles. For instance, the collective modes of the lattice vibrations are well described by phonons. Phonons are characterized by their dispersion relation, which links the energies of the excitations to their propagating wave vectors. In magnetic solids there is another kind of collective excitation that originates from the precession of the atomic spins around their equilibrium position. Such excitations have a totally different nature and are called spin waves or magnons. Spin waves are also characterized by their dispersion relation and more importantly they carry a spin of 1 $\hbar$. The above mentioned description of spin waves applies to the classical Heisenberg systems, where the spins are considered as localized moments. In itinerant ferromagnets, magnons can be also defined as electron-hole pair excitations, in which the spin orientation of the electron is opposite to the spin of the hole. From the energy point of view, phonons and magnons can have comparable energies and can coexist in a magnetic solid. The fundamental question in such a case is the following: How can one distinguish experimentally between these two different kinds of excitations, especially in low dimensional systems? The answer to this question is very important for deeper understanding of the lattice- and spin dynamics at surfaces and nano-objects.

In this Letter we report on the first simultaneous observation and unambiguous separations of phonons and magnons. Measurements are performed by means of a spin-polarized version of high resolution electron energy loss spectroscopy technique (SPEELS) [1–3]. We demonstrate that the spin degree of freedom of the incoming beam can be used for clarifying the nature of different types of excitations observed at the surface despite the fact that phonons show a significant spin dependence in the scattering of electrons. As an example we show the results of a high quality oxygen passivated Fe(100) surface [4,5] [see Fig. 1(a)].

The inelastic scattering of low-energy electrons is usually considered as a superposition of the contributions from dipole and impact scattering. Originating from the Coulomb interaction between the incoming electrons and the dipolar electric field at the surface, dipole scattering does not reveal any asymmetry with respect to the spin orientation of incident electrons. In the impact scattering, the incident electrons interact with electrons of the solid and such an interaction can involve exchange processes, which is usually referred to as exchange scattering. Within the exchange scatterings, the incident electron may transfer its energy to another electron in the sample, and the latter one is thus scattered out of the surface. In specular geometry ($\theta_i = \theta_f$), dipole scattering is usually a dominant process; however, at a magnetic surface the impact scattering can be rather pronounced and even comparable to the dipole scattering [6,7].

A schematic illustration of the scattering geometry in our SPEELS experiments is given in Fig. 1(b). In the experiment, an electron beam with energy E_i and spin σ_i is focused onto the sample surface. The angle- and energy-resolved intensity $I(\theta_f, E_f)$ of backscattered electrons is recorded and analyzed. The energy conservation law provides a direct access to the energy $\epsilon_{\hbar\omega}$ of excitations, which can be obtained from $\epsilon_{\hbar\omega} = E_f - E_i$, and zero energy is defined by the incident electron energy. A fraction of the backscattered electrons change their energy by $\epsilon_{\hbar\omega}$, thus surface excitations can be observed as peaks in intensity spectrum centered exactly at excitation energies $\epsilon_{\hbar\omega}$. An example of such spectra acquired for different spin directions of incident electrons at $E_i = 4.1$ eV, $\theta_i = 29.2°$,

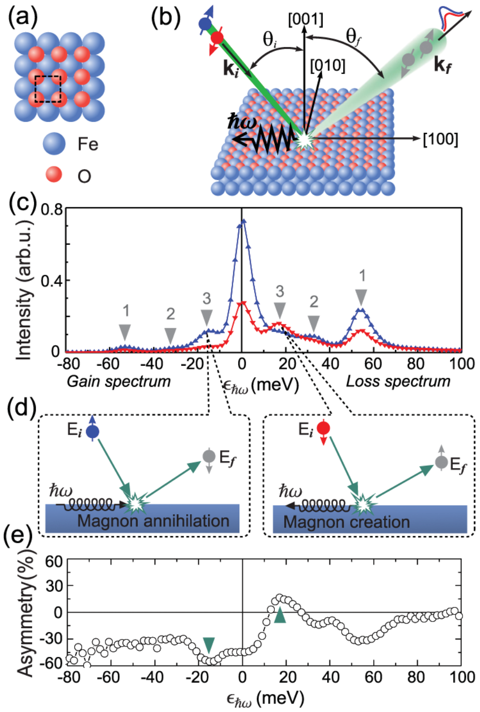

FIG. 1 (color). (a) Model of O/Fe(001)-$p(1 \times 1)$ surface. Surface $p(1 \times 1)$ unit cell is marked with dashed black square. (b) Scheme of SPEELS experiments. Electron beam with energy E_i, momentum $\mathbf{k}_i$, spin σ_i (real polarization of $70 \pm 10\%$) is aligned in the (010) plane with the angle θ_i to the surface normal. The intensity of backscattered electrons at angle θ_f is analyzed. The notched line represents an excited quasiparticle with energy $\hbar\omega$ created due to inelastic scattering events. (c) SPEEL spectra of the O/Fe(001)-$p(1 \times 1)$ surface measured at $\theta_i = 29.2°$, $\theta_f = 50.8°$. The blue and red curves are obtained with the spin of incident electrons parallel to that of the majority (spin-up) and minority (spin-down) electrons in the sample, respectively. The primary electron energy E_i is 4.1 eV. The twinned excitations marked by numbers are observed in loss and gain regions at room temperature. (d) Illustration of magnon creation and annihilation processes. Magnons can be created only by spin-down electrons, while they are annihilated only by spin-up electrons. (e) Asymmetry calculated using Eq. (1). Magnon excitations are marked by triangles.

$\theta_f = 50.8°$ is given in Fig. 1(c). Spectra with positive and negative energies $\epsilon_{\hbar\omega}$ are named as loss and gain spectra, respectively. This notation reflects the direction of the energy transfer during the scattering event. Excitation peaks in loss and gain spectra are "twinned" in a sense that electrons can both create certain quasiparticles losing energy $\epsilon_{\hbar\omega}$, and annihilate the thermally excited ones gaining energy $\epsilon_{\hbar\omega}$. The twinned loss and gain peaks are marked with the same numbers.

As a magnon excitation corresponds to a spin flip in the sample, if the incoming electron is of minority character with a spin of $-1/2\ \hbar$ then, according to total angular moment conservation rule, the outgoing electron has to be of majority character with a spin of $+1/2\ \hbar$, after the excitation of a magnon. It is worthwhile to emphasize that the apparent "spin flip" between the spins of incident and outgoing electrons occurs due to an exchange of the incident electron with one of the sample electrons instead of a real spin reversal [8,9]. An incident electron with minority character occupies a state above the Fermi level, and an electron with majority character from a state slightly below the Fermi level is scattered out. The interaction is of a pure Coulomb nature. The process occurs within a few attoseconds and without any energy dissipation. The time reversal process happens for the incidence of majority electrons, i.e., the annihilation of a magnon. The creation and annihilation processes of magnons are sketched in Fig. 1(d). The magnon creation peak in the loss spectrum appears only when incident electrons are of minority character, while the magnon annihilation peak in the gain spectrum is only produced by majority electrons. Since phonons are spin-independent quasiparticles, they can be created and annihilated by incident electrons of both spin characters. This particular dependence of magnon creation and annihilation on the spin of incident electrons, as opposed to phonons, is a fundamental feature, which allows us to distinguish between magnons and phonons in SPEELS experiments.

To judge the type of the excitations on their spin-flip or non-spin-flip natures, the straightforward way is to know the spin orientations of the electrons before and after the scattering event. This might be realized by using a spin-polarized electron source and a spin detector after scattering [9]. However, such an experiment demands a rather high feeding current for the analysis of outgoing spins. One has to sacrifice the energy resolution to achieve enough beam intensity, which then loses the information concerning the low-energy excitations. In the present work we will show that, for low-energy excitations where the energy is comparable to the thermal energy, it is possible to discriminate phonons and magnons without further spin analysis of the backscattered electrons. The differentiation procedure based on the nature of the magnons and phonons is discussed below.

Quantitatively, the spin dependence of backscattered intensity can be characterized by the spin asymmetry defined as

$$A(\epsilon_{\hbar\omega}) = \frac{I_{\downarrow}(\epsilon_{\hbar\omega}) - I_{\uparrow}(\epsilon_{\hbar\omega})}{I_{\downarrow}(\epsilon_{\hbar\omega}) + I_{\uparrow}(\epsilon_{\hbar\omega})}, \qquad (1)$$

where $I_{\downarrow}(\epsilon_{\hbar\omega})$ and $I_{\uparrow}(\epsilon_{\hbar\omega})$ are SPEEL spectra measured with spin-down and spin-up polarizations of incident electron beams, respectively. To reveal the nature of an excited quasiparticle, one has to compare the signs of spin asymmetries shown in Fig. 1(e) at energies of peaks in loss and gain spectra. Asymmetries of loss and gain spectra of excitation "3" at energies of -16 and 16 meV are of opposite signs as it is marked with arrows in Fig. 1(e); therefore, peaks are associated with magnon excitations. Asymmetry of excitation "1" and "2" at ±53 and ±33 meV have the same sign and almost identical magnitudes; hence, these excitations are phonons [10].

Interestingly, in our case the asymmetries of phonon peaks are always negative. It is most likely related to the strong exchange scattering between the incoming electrons and the electrons at the sample surface. To clarify the origin of the spin asymmetries observed on phonons, we measured the spin-resolved intensities for elastically and inelastically ($E_{\mathrm{loss}} = 53$ meV) scattered electrons with respect to the incident electron energy (see the upper and lower panels of Fig. 2(a), respectively). It is clear that presented intensities are very similar, except for the fact that the intensity of the inelastically scattered electrons is 2 orders of magnitude smaller. It is known that the spin asymmetry of elastically scattered electrons has to be caused by the exchange scattering, as dipolar scattering does not involve the electron's spin. In this case the exchange scatterings only involve the electrons with the same spin orientation. Otherwise, it would lead to a change of energy (as the incident electron must come to an unoccupied state above the Fermi level, the one with the opposite spin leaves the system from a state below the Fermi level and this leads to an energy change). As it is demonstrated in Fig. 2(b), the asymmetry curves of the inelastic excitation at 53 meV are almost identical to those of the elastic peak in all scattering geometries. Therefore, it is highly possible that the physical origin of the observed large spin asymmetries for both the elastic and inelastic intensities are the same, mainly the exchange scattering of electrons with the same spin. Therefore, the similarity of the spin asymmetry for the elastic and inelastic scatterings strongly suggests that the inelastic peak at 53 meV is also of the non-spin-flip nature and the observed high asymmetry is due to the exchange of the electrons with the same spin character. The same arguments apply to the other phonon excitations.

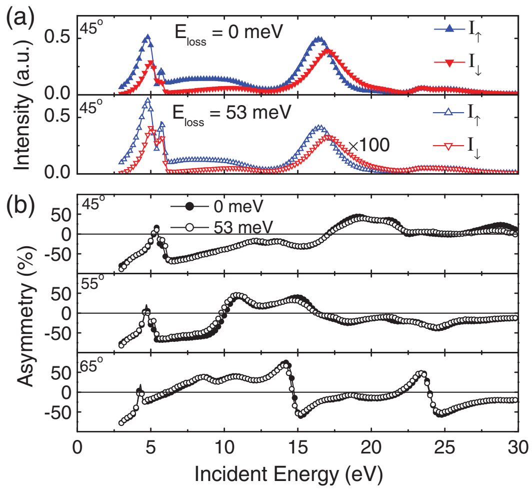

FIG. 2 (color). (a) Intensity of backscattered electrons as a function of the incident energy in the specular geometry recorded for elastically ($E_{\mathrm{loss}} = 0$ meV, upper panel) and inelastically ($E_{\mathrm{loss}} = 53$ meV, lower panel) scattered electrons. The incident angle is 45° and marked in the graphs. (b) The asymmetry curves for the elastically (●) and inelastically (○) scattered electrons measured at specular geometry and the angles of $\theta_i = \theta_f = 45°, 55°$, and $65°$.

The momentum conservation law provides a direct access to the quasiparticles momenta $\mathbf{q}_{||} = \mathbf{k}_i \sin\theta_i - \mathbf{k}_f \sin\theta_f$. It is important to emphasize that this relation is valid only for momenta lying in the incident plane, and SPEELS does not probe excitations with momenta perpendicular to this plane. In our experiments, the incident plane of the electron beam is (010) plane and only surface excitations with nonzero x- and z- momenta lying at the $\bar{\Gamma} - \bar{X}$ section of the reciprocal space are probed. To plot the entire excitations band structure we examined the backscattered intensities of both loss and gain spectra. The intensity spectra recorded for different wave vectors are presented in Figs. 3(a) and 3(b). A careful fitting of the spectra with a superposition of Gaussian peaks revealed five excitation branches in both energy loss and energy gain spectra. Centers of the fitted peaks are marked in Figs. 3(c) and 3(d) with symbols. Phonon branches labeled 1, 2, 4, and 5 are represented by open symbols, while a magnon branch labeled 3 is plotted by filled symbols. The nature of those quasiparticles is clarified from the asymmetries of the gain and loss peaks as it is sketched in Figs. 1(c) and 1(d). Magnon branch clearly disperses from 16 to about 40 meV as the wave vector increases from 0.2 to 0.7 Å^{-1}. Magnon excitation peaks become much more broad at high wave vectors. This broadening can be explained by a strong decay of magnons in the itinerant electron system. In this case, the simple Heisenberg model cannot be applied to describe the spin wave dispersion in metal thin films, especially in the wave vector range that SPEELS measures [3,11].

To shed light on the origin of all the observed phonon modes we performed calculations of the harmonic phonons of the O/Fe(001)-$p(1 \times 1)$ surface by means of the direct calculations of the force matrix [12,13]. Surface was simulated with a slab built from 11 layers of bcc Fe stacked in the [001] direction. One side of the slab was covered by a layer of oxygen placed in hollow sites of Fe(001) and the whole system was relaxed. Hellmann-Feynman forces used to construct the force matrix were calculated from the first principles by means of the Vienna *ab initio* simulation package (VASP) [14]. The phonon band structure calculated with the help of the force matrix was projected on surface atoms of the system and on particular polarizations (or Cartesian directions) of oscillations. Figures 3(c) and 3(d) show the spectral densities of phonons projected on the oxygen and the topmost Fe atoms. Only phonons with x- and z polarization (displacement direction) lying in the incident (010) plane are taken into account in Figs. 3(c) and 3(d) because the SPEELS is sensitive exactly to these excitations due to the selection rules [15]. Based on the theoretical calculations, we conclude all the phonon excitations as follows: the phonon branch 5 with the lowest energy [right-oriented triangles in Figs. 3(c) and 3(d)] originates from the acoustical z-polarized transversal

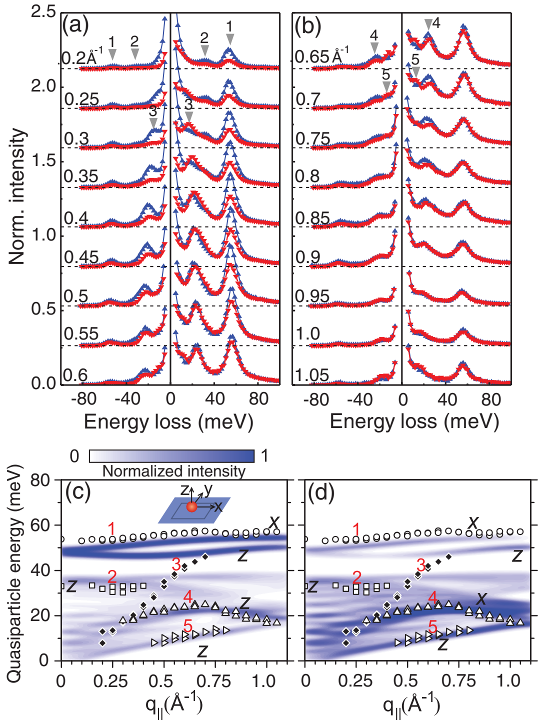

FIG. 3 (color). (a) and (b) SPEELS spectra measured on O/Fe(001)-$p(1 \times 1)$ with the spin of incident electrons parallel to that of the majority (blue curve) and minority (red curve) electrons. The corresponding in-plane wave vectors are marked in the graphs. The spectra have been shifted to have a clear view. Theoretically calculated phonon spectral density maps projected on (c) the oxygen layer and (d) the topmost Fe layer. Symbols represent the phonon and magnon peaks in the experimental data. Open symbols denote the phonon branches numbered by 1, 2, 4 and 5; filled symbol denotes the magnon branch and is numbered by 3. Letters "x", "y", and "z" near each of the pronounced phonon bands denote displacement directions of the corresponding phonons. The corresponding directions are illustrated in the inset of (c).

oscillations of the topmost Fe and O layers. It is the so-called Rayleigh wave of the O/Fe(001)-$p(1 \times 1)$ surface, which has been also observed in the He-atom scattering experiments [16]. The next phonon branch 4 shown in Figs. 3(c) and 3(d) with upward-oriented triangles is also localized in the two topmost layers. In the Fe layer branch 4 is a longitudinal acoustic phonon with x polarization, while in the O layer it is transversal with z polarization. Phonon branch 2 spreading off the $\bar{\Gamma}$ point at the energy of $\simeq 33$ meV plotted with squares in Figs. 3(c) and 3(d) is a z-polarized surface resonance of the phonons of the Fe slab. Two high-energy branches at $\simeq 50$ meV are optical z-transversal and x-longitudinal phonons localized mostly at the oxygen atoms. The agreement with experimental results for these excitations is not so sharp as in the above cases, but these phonons can still be associated with the excitation branch 1 at $\simeq 55$ meV plotted in Figs. 3(c) and 3(d) with circles. The differences between the experiment and the theory can be explained by the anharmonic contribution to oxygen vibrations, which couples the x- and z-polarized phonon modes. Our theory has not revealed any traces of excitation branch 3, which indicates that it is not a phonon as was also confirmed by our experiments.

In summary, by presenting the example of the O/Fe(001)-$p(1 \times 1)$ surface, we have demonstrated the fundamental possibility to identify the spin-flip and non-spin-flip nature of different excited quasiparticles among the vast variety of loss and gain features observed in SPEELS. This ability is achieved by controlling the spin of incident electrons. The magnon and phonon are unambiguously distinguished based on their spin nature and their dispersion relation is measured over the whole Brillouin zone. Our *ab initio* calculations successfully described all the observed phonon branches. We hope that our results could open a way towards a better understanding of the quasiparticles involved in lattice- and spin dynamics and their possible coupling, in particular, in the multifunctional complex hybrid and oxide materials as well as strongly correlated electron systems.

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