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Excitation of plasmons in Ag/Fe/W structure by spin-polarized electrons

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Using Spin-polarized Electron-Energy Loss Spectroscopy (SPEELS), the plasmon excitations were probed in a few atomic layers thick Ag film deposited on an Fe layer or on a single crystal of W(110). The measurements were performed at two specular geometries with either a 25° or 72° angle of incidence. On a clean Fe layer (10 atomic layers thick), Stoner excitation asymmetry was observed, as expected. Deposition of a silver film on top of the Fe layer dramatically changed the asymmetry of the SPEELS spectra. The spin-effect depends on the kinematics of the scattering: angles of incidence and detection. The spin-dependence of the plasmon excitations in the silver film on the W(110) surface and on the ferromagnetic Fe film is suggested to arise from the spin-active Ag/W or Ag/Fe interfaces. © 2015 AIP Publishing LLC.

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Surface plasmons (SPs) are collective electronic excitations near the surfaces of metallic structures.^{1,2} SPs may be described by classical electrodynamics using appropriate material properties and boundary conditions.² Both non-propagating localized SPs (LSPs) and propagating or SP polaritons (SPPs) are possible. Localized surface plasmons usually exist in metal nanoparticles. Their resonance frequency is determined by the size, shape, and dielectric environment of these particles. The extinction spectra of metal nanoparticles exhibit LSP resonances, typically in the visible-UV range.³ In contrast, surface plasmon polaritons can be excited on metal surfaces or in thin metal films in a wide range of wavelengths using, for example, attenuated total reflection techniques² or electron impact.^{4,5} The evanescence, or confinement, of the field near the metal “plasmonic” surface allows for the possibility of near-field intensities that are significantly enhanced compared to the incident radiation.⁶

Plasmonic structures that support localized surface plasmon oscillations and propagating surface plasmon polariton excitations have become a topic of major interest in the last 10 years.^{7–10} These structures possess very unusual optical properties and can enhance the sensitivity of various molecular spectroscopies by several orders of magnitude and increase the resolution of optical microscopes beyond the wavelength limit. Recently, there have been observations of substantial enhancement of magneto optical effects in plasmonic structures combined with ferromagnetic components, thus a magneto-plasmonic coupling.^{11–14} This finding initiated the research of the coupling between plasmonic (charge oscillations) and magnonic (spin) degree of freedom in thin films and multi-layer structures.

Can magnetic fields modify the properties of electron plasmas? This question has been analyzed for surface plasmons at terahertz frequencies propagating at an interface between a semiconductor and a dielectric.¹⁵ It was demonstrated that a magnetic field could be used to control SPP

propagation, and so open the door for novel active plasmonic devices. Unfortunately, the magnetic field required to produce noticeable changes in noble-metal-based plasmonic structures in the visible spectral range is of the order of several Tesla and is therefore too large for realistic applications.

On the other hand, ferromagnetic layers exhibit large magneto-optical (MO) activity but support much less propagating SPP due to high ohmic losses. Combining magnetic and plasmonic counterparts in a magneto-plasmonic system would both enable high MO activity and support propagating surface plasmon modes. Orders of magnitude smaller external magnetic fields might be sufficient to manipulate the optical properties of their SPPs.¹⁶ This concept offers the potential of design and tailoring of the optical properties of complex composite magnetic materials where the coupling between plasmons and the magnetic field occurs through the magnetization of a ferromagnetic layer of a multi-layer structure.^{17,18} An example of such a structure is plasmonic microinterferometry^{19–21} that demonstrates active control of surface plasmons in a magneto-plasmonic gold-cobalt-gold multilayer film by a weak external magnetic field of a few milli-Tesla.²²

Similar but reversed “magneto plasmonic” coupling can be achieved when the plasmonic substrate enhances the magneto-optical effect.²³ In this experimental work and corresponding theoretical analysis, it was shown that in total reflection geometry, the intense and sharp surface plasmon resonance of noble metals can strongly increase the MO figure of merit.²⁴ This could provide a significant advantage to high-resolution near-field optical imaging of submicron magnetic domains for technological applications such as high-density data storage.²⁵

One of the means for studying magneto-plasmonic coupling is Spin-Polarized Electron-Energy Loss Spectroscopy (SPEELS). In this technique, a spin-polarized incident beam is scattered from a sample surface and the energy distribution of scattered electrons is measured. Spin effects are usually

represented as an intensity asymmetry spectrum $A = (I^+ - I^-)/(I^+ + I^-)$, where I^+ and I^- are the SPEELS spectra measured with spin-up and spin-down polarization of the incident beam, respectively. The technique was successfully used for studying magnetic properties of surfaces, such as: Stoner excitations^{26–34} and spin waves.^{35–37} It was also exploited for investigating surface and bulk plasmon oscillations.^{4,5}

It turns out that the cross-section of plasmon excitation (in a ferromagnetic structure) by an electron impact may depend on the spin orientation of the incident beam that was predicted theoretically³⁸ and observed experimentally in strained Gd layer on Mo(112).³⁹ This effect shows up as intensity asymmetry in SPEELS at the energy loss corresponding to a Gd plasmon excitation. The qualitative theoretical explanation of this observation is the following.³⁹ The 4f electrons that are responsible for the Gd magnetic moment are localized (and cannot take part in low energy plasmon excitations) but can influence the spin dependent plasmons via the itinerant rare-earth 5d and 6s electrons. Essentially, the Gd 4f electron creates an exchange field that slightly polarizes the Gd 5d/6s conduction electrons and gives rise to different electron densities $n_\uparrow$ and $n_\downarrow$. This corresponds to different plasmon frequencies $\omega_\uparrow \propto n_\uparrow^{1/2}$ and $\omega_\downarrow \propto n_\downarrow^{1/2}$. Although the strained Gd layer is a local magnetic moment system, the spin-dependence of plasmon oscillations is related to the long-range magnetic order of the system (due to ferromagnetism): the spin-dependence disappears above the Curie temperature.

Another reason for a spin dependence in plasmon excitation can be the presence of a spin-active interface adjacent to a silver layer, where, in our case, the plasmons are excited. We call spin-active interface an interface between Ag layer and a ferromagnetic layer or between Ag layer and high-Z material. In such an interface, an exchange or spin-orbit interaction may lead to a spin-dependent electron scattering and, by consequence, to a spin-dependent plasmon excitation.

In the present work, we were looking for a spin-dependence of the plasmon excitation in thin Ag layer by spin-polarized electrons mediated by an adjacent ferromagnetic layer of Fe or spin-active Ag/W interface. We employed SPEELS and measured energy distributions of scattered electrons for spin-up and spin-down incident electrons. Two incident angles of 25° and 72° (in a specular geometry) were chosen to compare plasmon excitation at different kinematics. Geometrical arrangement of the experiment for 25° of incidence is shown in Fig. 1. For measuring energy distributions of scattered electrons, we used the time-of-flight technique that was described elsewhere.⁴⁰ In essence, this technique requires a pulsed incident beam. In our case, the pulse width was about 1 ns and the repetition rate of 4.5 MHz. Energy resolution of this technique depends on the energy to be measured and estimated to be about 0.5 eV in the energy range used. Spin-polarized incident beam was generated by a photoemission process from a strained GaAs photocathode activated by depositing Cs and by oxygen adsorption.⁴¹ A photoelectron beam excited by a circularly polarized laser light with a wavelength 830 nm was originally longitudinally polarized. A spherical electrostatic deflector then turned the photoelectron beam by 90° . Hence, the longitudinally polarized beam became

FIG. 1. Geometry of experiment.

transversally polarized. The polarization vector of the beam was perpendicular to the scattering plane defined by the incident and scattered electron momenta. The sign of polarization vector can be changed by changing helicity of the laser light, this was controlled by computer.

An electron optical system transported the beam into a scattering chamber. The base pressure in the chamber was in the 10^{-11} Torr range. The substrate (W(110) crystal) was cleaned prior to the deposition of films using well-established procedure.^{42,43} The iron films were deposited on a W(110) surface in UHV conditions at room temperature using OMICRON type evaporator (EFM-3). The Ag films were grown using ceramic crucible heated by a tungsten coil around the crucible. Deposition rate was calibrated using quartz microbalance and Auger electron spectroscopy.

Fig. 2 shows scattered electron energy distributions as a function of the Ag film thickness. Spectra were recorded in a specular geometry with 25° angle of incidence. It is seen that for film thickness above 1 mono layer (ML), the plasmon energy loss at about 4 eV is already observable. It becomes stronger as the film thickness increases. The plasmon energy corresponds fairly well to the previous EELS measurements on Ag.^{44–46}

Fig. 3 represents SPEELS spectra measured on 4 ML Ag film on W(110) at 22 eV primary energy for spin-up and spin-down polarizations of the incident beam. Spin-up and spin-down spectra show unequal intensities in the region of the plasmon peak, and the asymmetry spectrum has a maximum around the plasmon energy.

The spin-dependence of plasmon excitation in this case may be due to the involvement of the Ag/W interface in the process of the plasmon excitation. Indeed, plasmon excitation may occur before or after scattering back.⁴⁴ The scattering back occurs at two interfaces: at Ag/vacuum and at Ag/W. The scattering back at the Ag/W interface may occur after the plasmon excitation (Fig. 2(a)) or before the excitation (Fig. 2(b)), i.e., at energy E_0 or at energy $E_1 = E_0 - E_{pl}$, where E_0 is the primary electron energy and E_{pl} is the

FIG. 2. Energy loss spectrum of a Ag film on W(110) as a function of the film thickness. Angle of incidence is 25° , angle of detection is 25° .

plasmon energy. In our case $E_0 = 22$ eV and E_1 is about 18 eV. We can assume that elastic scattering at the Ag/W interface is spin-dependent, and therefore, the plasmon energy loss may be enhanced for one polarization of the incident beam compared to the opposite polarization. The intensity asymmetry of elastic scattering at zero energy loss (22 eV kinetic energy of electrons) is about twice as small as at the plasmon energy loss (kinetic energy of electrons is about 18 eV). These unequal asymmetries at 22 eV and at 18 eV kinetic energy may indicate that plasmon excitation in this case occurs predominantly *before* scattering back. This conclusion stems from the previously studied energy dependence of the asymmetry of elastic scattering of the (00) beam

FIG. 3. Energy loss spectra of 4 ML Ag film on W(110) recorded with spin-up and spin-down polarization of the incident beam at primary energy 22 eV. Asymmetry spectrum is also shown in the figure.

from W(110)⁴⁷ where the asymmetry at 18 eV was about twice larger than at 22 eV.

One should understand, of course, that the Ag/W interface has different scattering properties compared to the vacuum/W interface in terms of the shape of the potential barrier and multiple scattering conditions. Therefore, the above speculations are suitable only for qualitative explanation of the observations.

A much more complicated modification of the asymmetry of SPEELS occurs in the case of plasmon excitation in Ag film deposited on a Fe layer. First, we check the magnetic state of the Fe layer by measuring the Stoner excitation asymmetry. Fig. 4(a) shows the Stoner excitations asymmetry measured on the 10 ML iron film at the film magnetization M1 (magnetic moment of the layer is parallel or anti-parallel to the electron beam polarization). Measurements for opposite magnetization of the Fe layer (M2) give opposite sign of asymmetry. It indicates the magnetic nature of the asymmetry and shows the magnetic state (in-plane magnetization) of the Fe film. When a 6 ML Ag film is deposited on top of the Fe layer (magnetization M1), the EELS spectrum exhibits the plasmon excitation energy loss at about 3.8 eV (Fig. 3(b)), and asymmetry spectrum changes dramatically: negative asymmetry in the plasmon region (see Fig. 3(a)) becomes positive with a small peak at plasmon energy (~ 4 eV) (Fig. 4(b)). Figures 4(c)–4(f) represent two sets of measurements on the 6MLAg/10MLFe/W(110) structure: (i) in specular geometry with 72° of incidence, and (ii) in specular geometry with 25° of incidence. The primary energy in both cases was 17 eV. Figures 4(c) and 4(e) show a comparison of EELS spectra recorded on Fe film and on Ag/Fe structure. The appearance of a plasmon peak is obvious in both cases. However, asymmetry spectra for 72° of incidence (Fig. 4(d)) recorded for two opposite magnetizations of the Fe film (in the experiment, to change the magnetic moment orientation the sample with the double-layer structure was azimuthally rotated around the normal to the surface by 180°) are very similar. A strong (and wide) asymmetry maximum at the plasmon energy (3.8 eV) reaches a positive value of about 7% for both magnetizations of the substrate (Fe film). The similarity of the asymmetry spectra for two opposite magnetizations of the Fe film in Fig. 4(d) is usually interpreted as being predominantly due to the spin-orbit interaction. The exchange effect in this case is negligible. In contrast, in the case of 25° angle of incidence, the asymmetry spectra for magnetization M1 and magnetization M2 of the Fe film are very different as shown in Fig. 4(f). This means that both spin-orbit and exchange contributions to the measured asymmetry spectra are present.⁴⁸ If the asymmetries measured for magnetizations M1 and M2 are denoted as A_{M1} and A_{M2} then following⁴⁸ the spin-orbit (A_{SO}) and exchange (A_{ex}) components of the asymmetry spectrum can be extracted: $A_{SO} = 1/2(A_{M1} + A_{M2})$ and $A_{ex} = 1/2(A_{M1} - A_{M2})$.

It is obvious that the spin effect in SPEELS of Ag/Fe structure is due to the presence of the Fe layer (or Ag/Fe interface). Indeed, first, SPEELS applied to a single crystal of Ag(100) does not show any difference between spin-up and spin-down energy loss spectra, although the plasmon peak is clearly observed.⁴⁹ In contrast, the presence of magnetic substrate under the Ag film leads to a spin asymmetry in

FIG. 4. (a) SPEELS and asymmetry of Stoner excitations on Fe layer on W(110); (b) SPEELS and asymmetry spectrum of Ag/Fe/W structure; (c) Comparison of EELS spectra on Fe film and on Ag/Fe/W structure at specular geometry, 72° of incidence; (d) Asymmetry spectra of Ag/Fe/W structure for two magnetizations of the Fe layer, specular geometry, 72° of incidence; (e) and (f) the same as (c) and (d) but angle of incidence is 25°.

SPEELS in a wide range of energy losses around the energy of a plasmon excitation. Second, the change of the Fe film magnetic moment orientation from M1 to M2 (opposite) leads to a change of the asymmetry spectrum (Fig. 4(f)). It suggests that these spin effects are due to a spin-dependent scattering (or multiple scattering) of incident electrons from the Ag/Fe interface after, before or during the plasmon excitation in the Ag film. Spin-dependent electron scattering at the interface between magnetic and nonmagnetic metals is well established and is responsible for the Giant Magneto Resistance effect in ferromagnetic/nonmagnetic multilayered structures.⁵⁰ An even more complicated scenario may take place when an incident spin-polarized electron dynamically polarizes the local environment of valence electrons in the Ag film and Ag/Fe interface (through an exchange-correlation hole) and propagation through the film excites a thin film plasmon mode. The mechanism of spin-dependent plasmon excitation in the Ag/Fe structure might be similar to the one discussed in Ref. 39 with an involvement of a strong exchange field of the ferromagnetic layer (Fe layer), which may penetrate into the adjacent Ag film (proximity effect) and influence plasmon excitation by spin-polarized electrons.

Spectacular difference between two geometries, 25° and 72° incident angles might be due to the different contributions

of the “dipole” and “binary” mechanisms of the electron scattering at these two geometries. Indeed, at 72° of incidence primary electrons undergo almost “forward” scattering with predominantly “dipole” mechanism of electron excitation that reduces the probability of electron exchange. In contrast, at 25° of incidence primary electrons are “back-scattered,” when the exchange effect is much more probable. Therefore, in this geometry, both the exchange and the spin-orbit contributions are observed in the measured asymmetry spectra.

Preliminary investigation shows that the spin effects in SPEELS of the Ag/Fe/W system strongly depend on parameters of the electron scattering (energies and angles). Systematic measurements with various primary energies, film thicknesses, and geometries, as well as a theoretical analysis, are required to identify the mechanism of observed spin-effects in electron scattering from a magneto-plasmonic system of Ag/Fe/W.

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