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Characteristic energy loss of electrons in Mg with angle resolution

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Abstract. The article discusses electron energy loss of bulk and surface plasmons in solid polycrystalline Mg films. The experimental study of the electrons' characteristic energy loss is conducted and its results are compared in a wide range of electron incidence angles on the sample (0°–89°), dispersion angles (5°–165°) and electron energies (100–600 eV). Electron wave refraction at the vacuum–solid state interface is considered as one of the factors determining the intensity of surface plasmons.

Keywords: electron spectroscopy, bulk plasmons, surface plasmons, elastic electron scattering, electron wave refraction.

Introduction

Electron energy loss spectroscopy is an effective modern method for analysing near-surface solid state regions and obtaining data on the density of electronic states and plasmon excitations. Spectroscopy identifies special energy spectra features of non-elastically reflected electrons that arise from fixed energy loss of some low-loss electrons (elastic reflected electrons peak, EREP) and, therefore, are observed as separated from the EREP by the value of this loss. The nature of the loss may vary; it is connected with quasiparticle excitation in the solid state (phonons and plasmons), single-particle excitation of valence electrons (intraband and interband transitions), and ionisation of internal atomic levels. Electron energy loss spectroscopy can be carried out in a wide range of electron energies by various experimental methods, achieving the required resolution at very low and very high excitation energies. Most common methods use seed electrons with energies within hundreds of eV. In this energy range surface and bulk plasmons as well as intraband and interband transitions get excited (Iakoubovskii, Mitsuishi, Nakayama, Furuya 2008; Oura, Lifshits, Saranin et al. 2006). Energy resolution around tenths of eV, which does not require additional monochromatisation of the electron beam, is necessary to interpret visible features of the spectrum.

Experimental procedure

The experimental analysis was conducted using a universal electron spectrometer based on the USU-4 ultra-high vacuum set. The spectrometer is fitted with a quasi-spherical condenser which is a two-grid analyser that determines angularly integral parameters of elastically and non-elastically reflected electrons at normal incidence. It is also fitted with a mobile portable electron spectrometer used for studying angularly differential parameters of electrons within incident angles 0°–89° and scattering angles 5°–165°.

Angle-resolved electron energy loss spectroscopy is performed in order to vary the zone of examination by changing incident angles, emergent angles and electron energies, and to reveal changes in electron state density in passing from surface to volume.

In general, plasmons come into sharp focus in substances which meet two requirements simultaneously: (1) there is a fairly isolated (with respect to energy) group of valence electrons; and (2) plasmon energy corresponding to oscillations of electrons in this group is relatively far from the band-to-band transition energies typical for this substance (otherwise, plasmons quickly become damp due to strong plasmon-electron interactions). Mg films meet these criteria; therefore, they were chosen for this study. Features of energy loss spectrum for Mg films may only be determined by excitement of bulk and surface plasmons. Mg films were formed using the method of thermal evaporation on cooled mirrored glass base in vacuum ~10^{-7} Pa, so that the surface was rough enough.

Energy loss spectra for Mg were studied in the energy range E_p = 100 – 600 eV. They were analysed according to the following procedure. Firstly, an approximation of loss was made within the measured spectra independently of collective mode excitation similar to approximation (Iakoubovskii, Mitsuishi, Nakayama, Furuya 2008). The EREP and peaks related to the excitement of bulk and surface plasmons were approximated with the Gaussian distribution:

$$I_i = A \cdot e^{-\frac{(\Delta E_i - \Delta E)^2}{2\sigma_i^2}}$$

where ΔE_i is the plasmon excitation energy value; σi is the dispersion determined by EREP dispersion (σ_r) and plasmon dispersion (σ_p):

$$2\sigma_i^2 = 2\sigma_r^2 + 2\sigma_P^2$$

Then the loss rates and the EREP rates were defined as areas below the corresponding peaks.

Experimental results

Complex investigation of energy loss spectra including electron energies, their incident angles and angles of emergency from the surface can be carried out based on comparative analysis of results obtained from the following three types of studies: (1) measurement of energy spectra at a fixed angle of emergence depending on the incident angle ϕ; (2) measurement of energy spectra at a fixed incident angle depending on the angle of emergence α; (3) measurement of energy spectra at fixed incident and emergent angles depending on the energy of primary electrons.

Let us consider the Mg spectra behaviour with changing electron incidence angle (Fig. 1). Bulk plasmon energy is 10.5 eV, which corresponds with the previous research data (Raether 1980). As the incident angle increases and, therefore, the penetration depth of the initial beam into the sample decreases, quick weakening of the bulk loss peak is observed. Fig. 2 shows the dependence of the second type on the incident angle α. According to the spectral analysis, it is possible to conclude that at relatively small incident and emergent angles of electrons the ratio A_{S1}/A_r (where A_{S1} is the intensity of the first surface loss, A_r is the intensity of EREP), to the accuracy of experimental precision that does not exceed approximately 5%, is defined by the following formula (with the assumption of two-part process of the surface loss development):

$$\frac{A_S}{A_r}(\varphi, \alpha) = \frac{B}{\cos\alpha} + \frac{C}{\cos\varphi}$$

which corresponds to the perceptions of dielectric formalism (B is a constant defined by the intensity of primary electron beam; C is defined by the intensity of EREP) (Stern, Ferrell, 1960). However, at reasonably large incident angles ϕ> 80° the surface loss intensity does not change significantly.

The effect discovered may be interpreted by electron wave refraction at the solid state–vacuum interface:

$$\frac{\sin\varphi}{\sin\varphi'} = \frac{\sin\alpha}{\sin\alpha'} = \sqrt{\frac{E + eU_0}{E}}$$

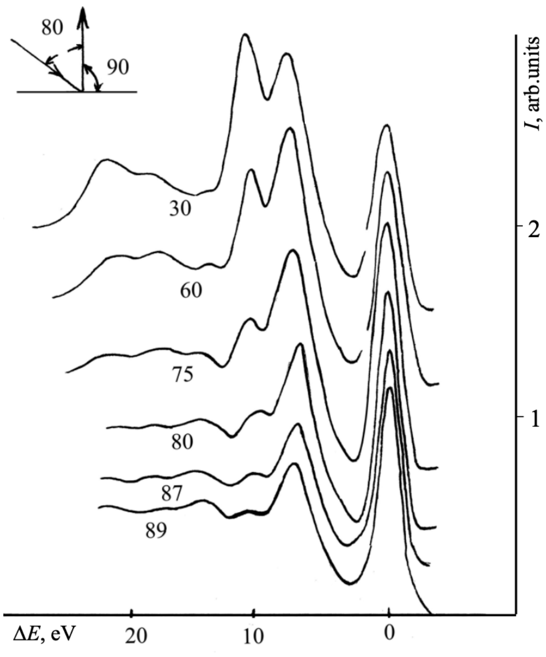

Fig. 1. Electron energy loss spectra for Mg. E_p = 200 eV. α = 0°. Incident angles ϕ in degrees are specified near the curves

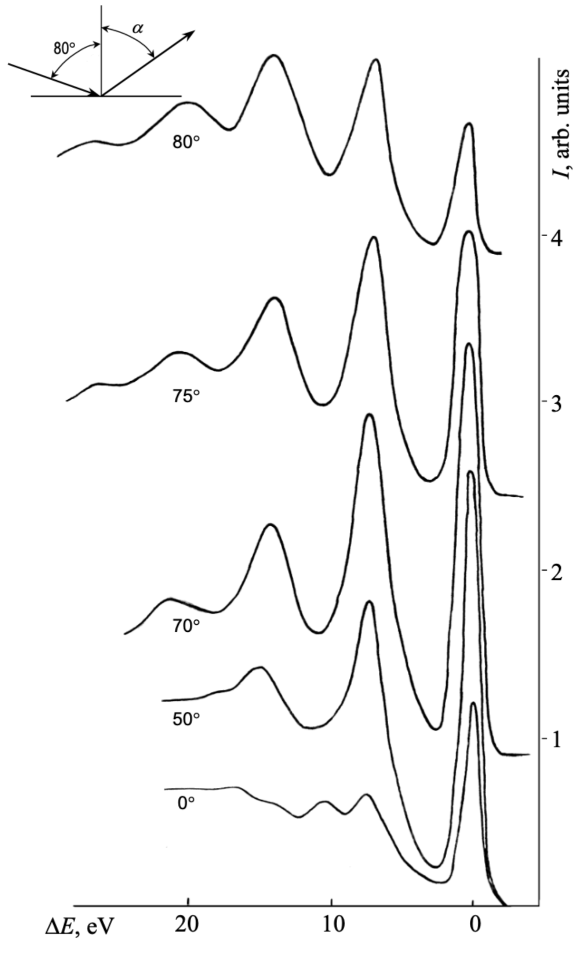

Fig. 2. Electron energy loss spectra for Mg. E_p = 100 eV. ϕ = 80°. Emergent angles α are specified near the curves

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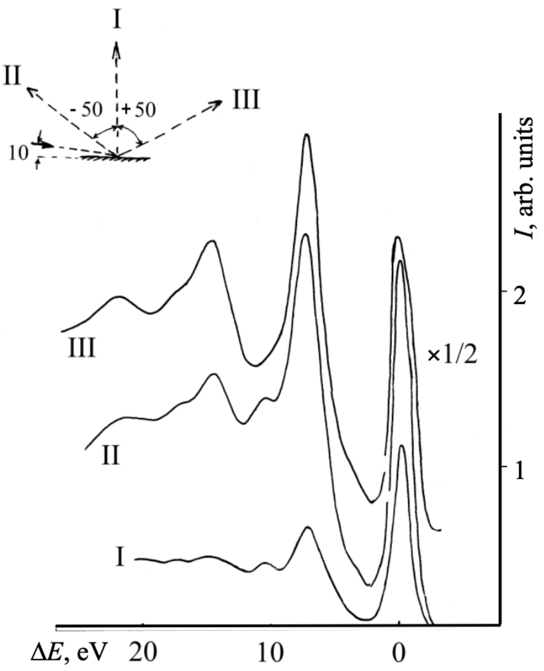

Fig. 3. Electron energy loss spectra for Mg. E_p = 200 eV. ϕ = 80°. Emergent angles: I correspond to 0°; II, III corresponds to 50°

where α' and ϕ' are the inner angles of electron incidence and emergence, U_0 is the effective inner potential of solid state. With increasing incident angle ϕ from 80° to 89° (Fig. 1), the inner incident angle changes from 74° to 79°, which leads to insignificant difference in the electron energy loss spectrum.

Fig. 3 illustrates the comparison of electron energy loss spectra at the electron incidence angle ϕ = 80° at two symmetrical angles of emergence corresponding to very different scattering angles. In this case, the intensity of surface loss peaks at small scattering angles and significantly exceeds those for big scattering angles where bulk plasmon peaks are observable. The result seems to derive from the fact that electrons with small scattering angles elastically reflected once and the electrons that experienced multiple scattering contribute differently to the electron energy loss intensity peaks. The electrons reflected once at small angles from the surface of solids (their contribution is 70–80 % of the total amount of reflected electrons) play the main role in forming the electron energy loss spectrum in the region of small scattering angles. For large angles this contribution is substantially smaller, and the role of electrons emerging from relatively large depths becomes significant as they cause volume loss (bulk plasmon).

Fig. 4 illustrates changes in electron energy loss spectra depending on the changes in incident electron energy. As energy increases and the depth of the primary beam penetration distance grows, the intensity of the bulk loss peak also grows.

Energy dependences $\frac{A_{S1}}{A_r}(E_p)$ corresponding to the experimental data in the entire given interval of energies with relatively small angles ϕ and α are close to the theoretically predicted probability of surface loss that is in accordance with the perceptions of dielectric formalism (van Attekum, Trooster 1979):

$$W_S \sim \frac{1}{E^{1/2}}$$

However, for big incident and emergent angles, the dependence $\frac{A_S}{A_r}(E_p)$ (fig. 5) is different.

This result can be explained by the above-mentioned phenomenon of electromagnetic wave refraction at the solid state–vacuum interface. From this perspective, experimental dependences $\frac{A_S}{A_r}(E)$ may be described as:

$$\frac{A_S}{A_r}(E) = \frac{C}{E^{1/2}} \cdot F(\alpha', \varphi') \tag{1}$$

where α' and ϕ' are inner emergent and incident angles.

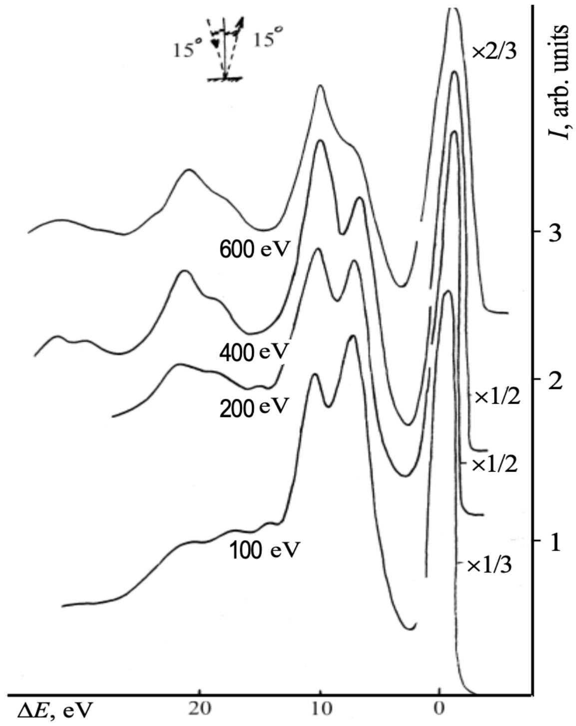

Fig. 4. Electron energy loss spectra for Mg. ϕ = α =15°. Primary electron energies are specified near the curves. The size of elastic reflected electron peaks is reduced in proportion to the figures indicated

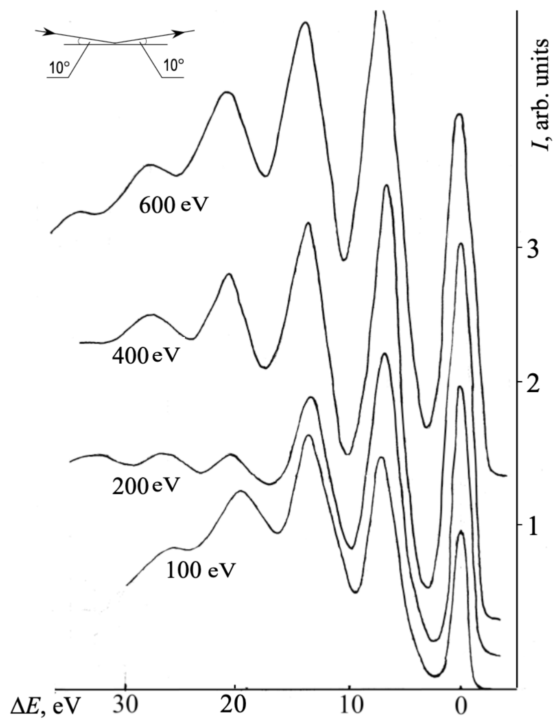

Fig. 5. Electron energy loss spectra for Mg. ϕ = α = 80°. Primary electron energies are specified near the curves

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It appeared that the dependences calculated using the formula (1) correspond with the experimental results (Fig. 6).

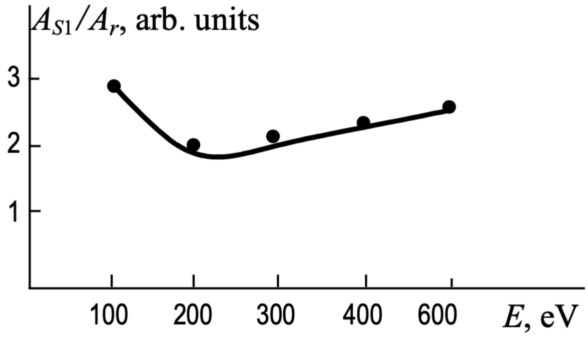

Fig. 6. Dependence of ratio of first surface plasmon intensity to EREP intensity from primary electron energy for Mg. ϕ = α =80°. The solid curve corresponds to the experiment, points correspond to the calculations.

Conclusion

Detailed investigation of angle-resolved electron energy loss spectra showed that for correct reading of experimental results it is essential to take into account the character of elastic electron scattering and the influence of electron wave refraction at the solid state–vacuum interface.

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