ELECTRON REFLECTION AND ELECTRON TRANSMISSION APPLIED TO BILAYER FILMS

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Electron energy loss spectrum

Electron energy loss spectroscopy (EELS) has become one of the most powerful tools for investigating both the surface and the bulk properties of thin films /5/. When applied to multilayer systems it also provides important information about the structure of interfaces. Collective oscillations, as a description of long-wavelength correlations between electrons and ions in a medium /1,2/ are a suitable method for studying many interesting properties of materials. Here we shall adopt this approach to discuss the electron reflection and electron transmission applied to bilayer films. Let us note that those problems can also be solved in the framework of classical electrodynamics /7/. However, typical quantum-mechanical features, such as multiple-excitation processes or the Landau damping are naturally incorporated only in the quantum-mechanical approach.

The whole theory is rather tediuos and we shall only outline the main steps. In fact, it is a nontrivial generalization of similar approach applied to a thin film /4,5/. The full approach will be published elsewhere. Briefly, we study a system consisting of two adjancent parallel plates. The plates are denoted by n=1 and n=2, and their thicknasses by a and b, respectively, so the surfaces of the system are at z=-b,0,a. In the nonretarded, long-wavelength approximation, we take the electrostatic equations and assume the step-density profile for both electron and ion charge density in the bilayer system. The collective polarisation eigenmodes $\vec{P}_i$ are then decoupled into the surface and bulk modes /1,2/. Each surface mode has an appreciable electric field close to the corresponding surface. While the S surface modes, with a dispersion $\omega=\omega_s(k)$, s=1,2...S, reflect properties of the whole system, the dispersionless bulk modes (longitudinal $\omega=\omega_L$ and transverse $\omega=\omega_T$) are independent in each thin film. The wave vector $\vec{k}$ of surface modes is continuous and parallel to the surfaces, while the wave ve-

+Also at the "Rudjer Bošković" Institute, Zagreb, Yugoslavia ctor $\vec{q}=(\pi m/z_0)\hat{z}$ of bulk modes is discrete (m=1,2,3...) and normal to the surfaces of the system. Here z_0=a,b if n=1,2 respectively.

The dielectric function $\varepsilon_n(\omega)$ follows from the equation of motion for a dielectric medium. In the case of an ionic crystal $\varepsilon(\omega)$ for optical phonons is given by $\varepsilon(\omega)=(\omega^2-\omega_L^2)/(\omega^2-\omega_T^2)$ /1,2/. The same holds for plasmons ($\omega=\omega_p$) in metallic plate if we replace $\omega_T\to 0$, $\omega_L\to\omega_p$. There are four (three) surface modes in the case of two thin dielectric (metallic) plates. The inert dielectric is simply described by ε=const.,and if we add such a plate to the system the number of surface modes remains the same.

If the plates of the bilayer system are described by any of the named dielectric functions, we are able to diagonalize the hamiltonian of the free system. The interaction of a point charge e with collective oscillations of the bilayer system is then described by interaction matrix elements Γ_i which can be calculated for each mode.

In the trajectory approximation, the EEL spectrum $P(\omega)$ can be obtained in a closed form /5/. We shall devide it in two parts

$$P(\omega) = \Delta(\omega) + S(\omega) \tag{1}$$

The main contribution arises from $\Delta(\omega)$, which gives the standard Poisson distribution for EELS, with sharp peaks close to the asymptotic frequencies $\omega_{\infty i}$ of eigenmodes. The spectrum is normalized $\int P(\omega)d\omega=1$ so the probability of n-th order excitation of i-th mode is given by $P_i^{(n)}=P_0A_i^n/n!$. Here P_0 corresponds to the no-loss line while A_i is the interaction strength of i-th mode interacting with the external charge and it essentialy depends on the interaction matrix elements Γ_i. $S(\omega)$ represents the background for the sharp peaks in $\Delta(\omega)$, which is due to the strong dispersion of surface modes. If the dispersion is not very strong, $S(\omega)$ can be neglected.This is a case for thick films and small electron energies.

<u>Electron reflection</u>

The energy loss spectrum of reflected electrons /5/ is an important tool for investigating the properties of both the reflecting surface and the interface in a bilayer system. For simplicity we shall analyze only the electron beam specularly reflected from plate 1 (z=a) of a bilayer system. We neglect the penetration of electrons into the sample so only the surface modes are excited.

The spectrum $P(\omega)$ exibits the following features:

i) The mode ω_1 connected with the reflecting surface (z=a) exibits the strongest interaction with the scattered electron and we call it the dominant mode. We roughly estimate

$$A_1 \approx A_{\infty 1} \tag{2}$$

i.e. the electron interaction with the dominant mode (ω_1) is approximately the same as its interaction with the surface mode ($\omega_{\infty 1}$) of the semiinfinite plate 1.

ii) Two interface modes (ω_2, ω_3) connected with z=0 interface exhibit much weaker coupling than the dominant mode

$$A_{2,3}/A_1 \sim \exp(-2\omega_{\infty 2,3}a/v) \tag{3}$$

which means that we can detect the electron-interface mode interaction only for thin films and high electron energies: $\omega_{\infty 2,3}a/v \lesssim 1$.

iii) The surface mode ω_4 connected to the z=-b surface can be detected only if both films are very thin and the electron energy is very high

$$A_4/A_1 \sim \exp(-2\omega_{\infty 4}(a+b)/v). \tag{4}$$

Even in this case, the electron interaction with the interface and especially with the dominant mode mostly screens the interaction with the weak mode. In a good approximation, one can treat the electron reflection on two thin films as the electron reflection on film 1 placed on a semiinfinite substrate 2.

Figure 1 shows the spectrum $P_1(\omega)$ for the single surface plasmon excitation of a thin Al_2O_3 film on an Al (ω_p=15.0 eV) substrate. In the plasmon energy range, Al_2O_3 is described as an inert dielectric with ε=4. The increase of Al_2O_3 thickness broadens and gradually shifts the sharp maximum at ω_1=$\omega_p/\sqrt{2}$ (a=0) towards ω_2= $\omega_p/\sqrt{1+\varepsilon}$. At the same time, the peak becomes weaker since the surface plasmon is connected to the Al and not to (Al+Al_2O_3) reflecting surface. The same behaviour has been observed in experiment/8/.

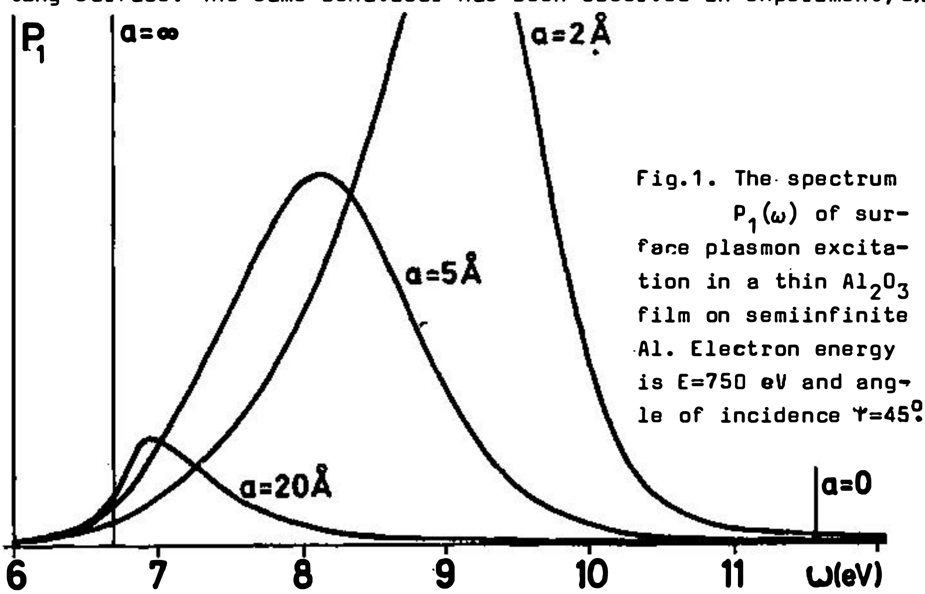

Fig.1. The spectrum $P_1(\omega)$ of surface plasmon excitation in a thin Al_2O_3 film on semiinfinite Al. Electron energy is E=750 eV and angle of incidence Ψ=45°.

Electron transmission

When an electron is transmitted through a bilayer film it excites both surface and bulk collective oscillations /3/. This modes are well defined for $K<K_c$ where $\vec{K}_c(\vec{k}_c,\vec{q}_c)$ is an (averaged) three-dimensional cutoff wave vector and $\vec{k}_c$, $\vec{q}_c$ are (averaged) surface and bulk cutoff wave vectors /10/. By defining $q_c=\pi N/z_0$ we perform the summation over bulk modes to $m\leq N=[K_c z_0/\pi]$. The condition $N\geq m\geq 1$ implies that $z'=\pi K_c^{-1}\approx 3$-5 Å is the limiting film thickness below which bulk collective oscillations cannot exist, and it obviously corresponds to the distance between atomic layers in a crystal. Let us note that the summation over bulk modes can also be performed with the help of the closure relation /5/. This procedure assumes an infinite summation over m, i.e. $q_c\rightarrow\infty$, which is not always a good approximation /6/.

The interaction strength of bulk modes is proportional to $(\omega_{Ln}^2-q^2v^2)^{-2}$. It increases with increasing film thickness or decreasing electron energy. In both cases it reaches the resonant points when the electron velocity v is equal to the phase velocity ω_{Ln}/q of some bulk oscillation. This effect (which is not obtained in the closure-relation approach) explicitely shows the discrete nature of bulk collective modes. However it has not been detected yet. The reason might be that in transmission experiments neither the energy nor the film thickness are continuously changed in regions which are wide enough. One can easily find out that the resonance conditions

$$m = \frac{z_0\omega_{Ln}}{\pi v} = 0.08\frac{\omega_{Ln}(eV)z_0(Å)}{\sqrt{E_e(eV)}} = 1,2,...,N \qquad (5)$$

are quite realistic. or instance, for an Al film 100 Å thick, we obtain resonances at E_e=15 keV (m=1), E_e=3.7 keV (m=2), E_e=1.6 keV (m=3), etc.

As an example, figure 2 shows the EEL spectrum (Poisson distribution) for electrons transmitted through a thin Al+Mg sample. The first-order bulk losses in an Al+Mg system are a superposition of such losses in a thin Al and a thin Mg film. This is not true for surface losses, since the surface plasmon dispersion is drastically changed when Al and Mg films are put together. The new surface plasmon mode connected with the Al-Mg interface is detected. In the second-order bulk losses there is also a line describing the excitation of bulk plasmons in both Al and Mg films. All the results are in good agreement with experiment /9/.

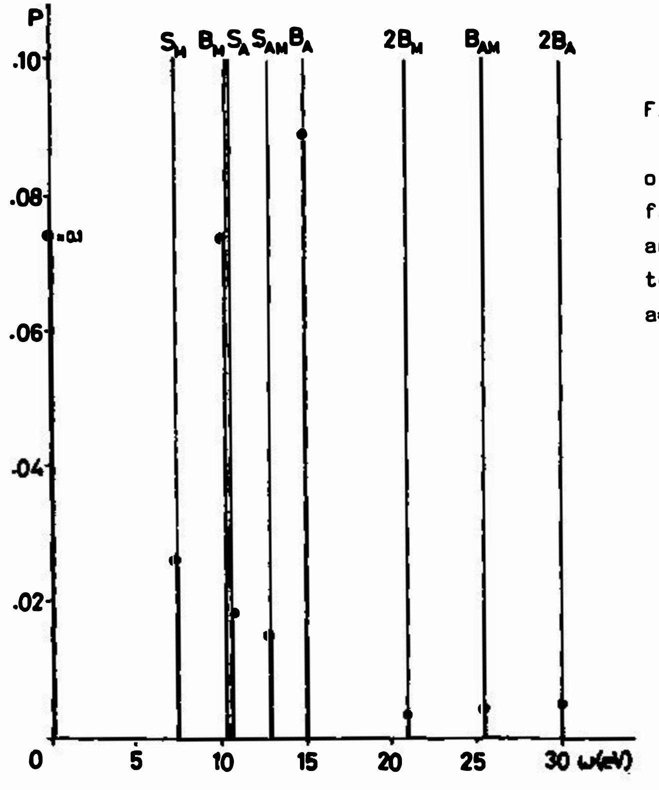

Fig.2. Excitation probabilities of bulk (B) and surface (S) plasmons in an Al(A)+Mg(M) system. We have chosen a=b=250 Å, E_e=34 keV.

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