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Numerical simulation of valence losses in MgO cubes

J Aizpurua*, B Rafferty†, F J García de Abajo§ and A Howie†

* Facultad de Químicas, UPV/EHU, Aptdo. 1072, 20080 San Sebastián, Spain
† Cavendish Laboratory, Madingley Road, Cambridge CB3 OHE, UK
§ Facultad de Informática, UPV/EHU, Aptdo. 649, 20080 San Sebastián, Spain

ABSTRACT: Valence losses can be conveniently computed using classical dielectric excitation theory. In the non-relativistic case, the losses can be simulated using a distribution of surface and interface charges. These charges interact self-consistently with each other as well as with the incident electron. This method is used to analyse experimental STEM energy loss spectra for various incident beam positions near MgO cubes. The characteristic contributions of the cube edges are identified.

1. INTRODUCTION

Classical dielectric theory by Fermi (1940) deals very adequately with electron energy loss spectra (EELS) in scanning transmission electron microscopy (STEM). This approach has mainly been based on analytical solutions obtained for planar interfaces (Ritchie 1957), spheres (Ferrell and Echenique 1985) and other simple structures (Rivacoba *et al* 1996). In practice however, much more complex geometries are often encountered such as an interface passing normally through a thin film and allowing contributions from (possibly coupled) interface, surface and edge modes (Dobrzynski 1972, Davis 1976). Such structures can now be tackled using the boundary charge method together with numerical computation. This method is based on a distribution of interface charges which interact self-consistently with the field created by the incoming electron at the interfaces of the structure. This induced charge distribution produces a field at the fast electron position which generates the energy loss. It is convenient to place a greater number of interface charges near the beam as well as in sharp regions where the electric field can vary rapidly. The interface charge approach has already been employed by Fuchs (1975) to determine the normal modes of a cube and by Ouyang and Isaacson (1989a, 1989b) to compute energy losses for a spherical particle supported on a film. Standard packages allow simulation of many real-sample geometries observed in STEM such as cubes, truncated slabs or junctions. In particular we use the boundary charge method to deal with valence losses when the electron beam passes near an MgO cube. Marks (1982) studied the energy loss functions in this material for several electron trajectories and found new excitations associated with the edge effect. Here we simulate this effect in the cube energy loss spectrum and calculate the necessary corrections in terms of appropriate loss functions.

2. SIMULATIONS AND EXPERIMENTAL OBSERVATIONS

2.1 Parallel trajectory

As a first approximation to the edge effect in an MgO cube and to compare with observed spectra, we have simulated the energy loss probability per unit length for the case of the electron beam travelling parallel to an infinitely long, square cylinder of MgO. The dielectric response function used was derived from the bulk loss function in the same material extracted from energy loss data after deconvolution. In Fig. 1 we plot the simulated spectra for two different trajectories just outside a 100nm cube as shown in the inset and compare them with the experimental observations under the same conditions. In trajectory a the beam is travelling near the centre line of the cube face and the energy loss spectrum resembles that of a single planar interface. Three main peaks are excited at 11 eV, 14 eV and about 20 eV. The simulations agree well with the experimental observations although the latter could not be normalised

because of saturation of the zero loss signal. In trajectory b, where the beam travels near and parallel to the edge of the cube, the losses are shifted down in weight. The peaks are less clear than in the planar case and the peak at 20 eV almost disappears in the simulation though remaining more visible in the observations. This could be due to an error in the beam position since the 20 eV peak is restored if the trajectory is moved 2nm along the face and away from the edge. From the simulations, we can conclude that the main effect of the edge is to increase the relative weight of the low energy excitations and even completely remove the 20 eV peak for the trajectory exactly along the edge.

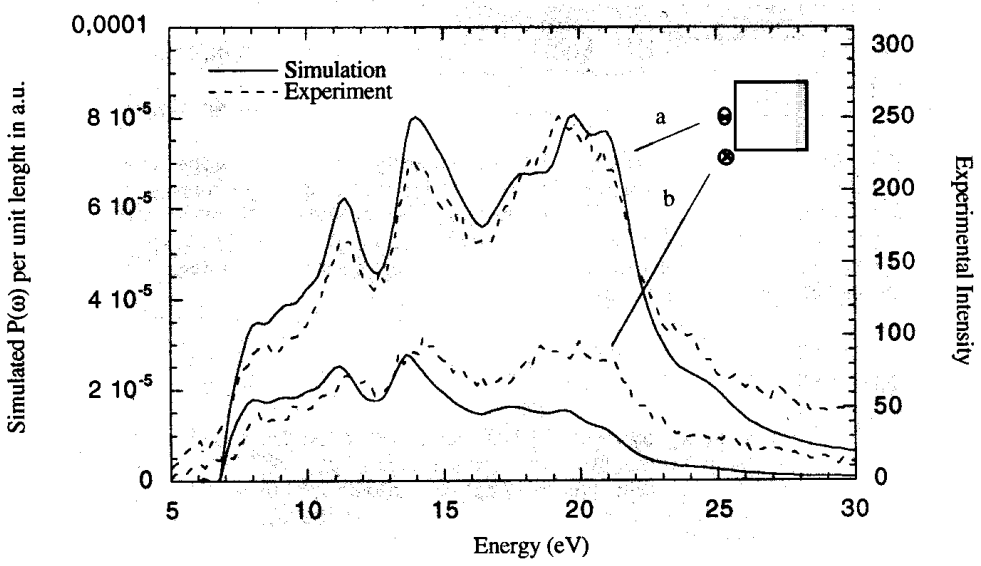

Fig 1. Energy loss probability per unit length for a 100 keV electron beam passing near an MgO cube as shown in the inset. The impact parameter is 2 nm. Solid lines correspond to the square cylinder simulations and dashed lines to the experimental observations.

2.2 Perpendicular trajectory

We can also study by experiment and simulation the edge effect in the loss spectra of MgO cubes when the beam travels perpendicularly to the edge. Since the exact position of the edge is not clear in all experimental situations, there can be a bulk contribution at 22 eV due to some beam penetration of the cube. In fig. 2 we present both the STEM observed energy loss spectrum and, for the identical trajectory, the total simulated energy loss probability $dP(\omega)/d\omega$ which agrees in absolute terms to a factor of about 0.7. The simulation does not reproduce the excitations observed in the band gap which must arise either from defects or from relativistic effects. In this case, the beam is travelling 2 nm just inside the edge and the contribution of the 22 eV bulk loss appears together with the other excitations from the top and bottom surfaces as well as from the edge. In the simulations we plot separately the uncorrected bulk contribution and the correction. It is clear that the main effect of the correction to the bulk is to produce an enhancement in the contribution of the low energy peaks at 11 eV and 14 eV. The planar contribution (20 eV) is not so strong in this case although it is also present in the spectrum. We cannot associate separately every peak to edge or planar excitations since they all appear mixed up, but it is possible to state that the low energy peaks are very sensitive to the edge while the higher one at 20 eV is not. This behaviour is related to the complex dielectric function which characterises MgO. A cleaner separation would be possible in the case of a free electron metal. As mentioned above, the total energy loss probability shows an important contribution in the high energy range (22 eV) due to the penetrating trajectory. The correction due to the nearby cube surfaces and edge introduces not only new characteristic peaks but also diminishes the bulk contribution via the Begenzungs effect. This bulk peak does not mask the surface and edge correction which is of the same order of magnitude for the impact parameter used.

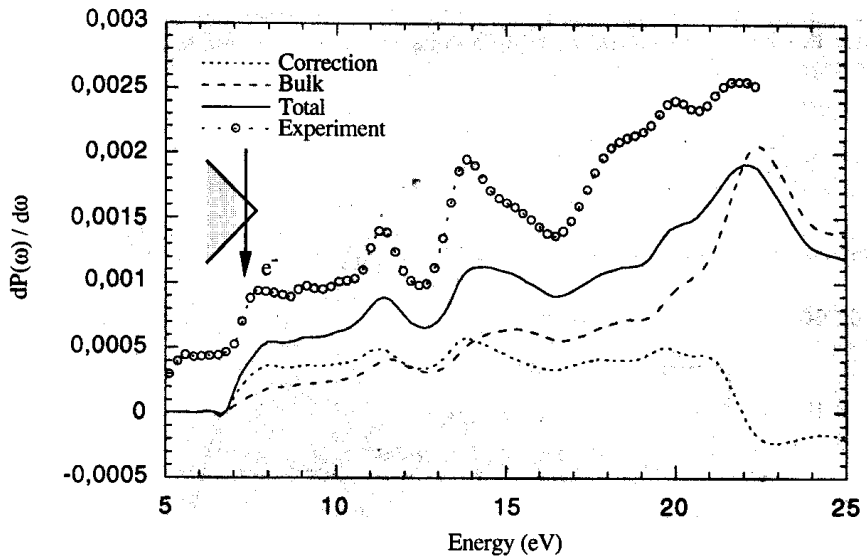

Fig 2. Energy Loss probability for a 100 keV electron impinging on an MgO edge as shown in the inset. The impact parameter is 2 nm from the edge and the different contributions to the losses are plotted together with the experimental observation.

3. CHARACTERISTIC ENERGY LOSS FUNCTIONS

For situations in which the beam in the STEM passes through a complex structure constructed from two dielectric media A and B with bulk regions, planar interfaces, edges or even corners, the loss probability can (in the non-relativistic case) be described as a sum over characteristic energy loss functions.

$$\frac{d^2P}{d\omega\, dZ} = \frac{2e^2}{\pi\hbar v^2}\sum_{j=0}^{n} A_j \,\mathrm{Im}\left[\frac{-1}{\alpha_j\varepsilon_A + (1-\alpha_j)\varepsilon_B}\right] \qquad (1)$$

In this eqn. each j value represents the contribution of a different mode which, in simple situations, is recognised as a bulk mode (α_j = 1 or 0) or a planar interface mode (α_j = 0.5). In our analysis of MgO cubes, we can take material A to be MgO and B to be vacuum so that ε_B=1. For larger cubes of dimension L>>v/ω, the cube edges do not interact significantly (except near the corners) and the value α_j = 0.3 seems to describe the characteristic edge function. Losses in smaller cubes can still be described by eqn(1) but interactions between edges could shift the mode eigenvalues and thus the values of α_j.

The contribution of each excitation mode is weighted with the A_j factor which depends on the position of the incident beam. Thus, in the simple case of the beam travelling in vacuum at distance x parallel to and just outside an infinite planar surface, eqn(2) would consist of just one term with α_j = 0.5 and $A_j = K_0(2\omega x/v)$. For the beam passing normally through a slab of thickness L >>v/ω eqn(1), after integrating over the path length z, becomes

$$\frac{dP}{d\omega} = \frac{2e^2}{\pi\hbar v^2} L\left[A_1 \,\mathrm{Im}\left(\frac{-1}{\varepsilon_A}\right) + A_2 \,\mathrm{Im}\left(\frac{-1}{0.5\varepsilon_A + 0.5}\right)\right] \qquad (2)$$

where $A_1 = \ln(k_c v/\omega)$ - 0.5 A_2, A_2= const/L (Ritchie 1957). This illustrates the Begrenzung effect whereby the strength of the new surface excitation comes from a compensating reduction in the bulk excitation. More generally when the beam crosses the interface between two media, the Begrenzung reduction is shared between the bulk losses on either side.

We now return to the trajectory a of Fig. 1 where the beam is just 2nm outside the face centre for MgO cubes with L >>v/ω. The most important losses are then the planar loss and the

losses from the top and bottom edge excitations. The edges on either side are too remote to be relevant and, for this beam position near an MgO cube of size L, the results are to quite a good approximation expressed by the eqn.

$$\frac{dP}{d\omega} = \frac{2e^2}{\pi\hbar v^2} L \left[A_1 \mathrm{Im}\left(\frac{-1}{0.5\varepsilon_A + 0.5}\right) + A_2 \mathrm{Im}\left(\frac{-1}{0.3\varepsilon_A + 0.7}\right)\right] \quad (3)$$

where $A_1 = K_0 (2\omega x/v) - 3/L$; $A_2 = 6.75/L$ (L is expressed in nm). These impact parameter dependent coefficients are obtained for the case of MgO with an accuracy of 10 per cent.

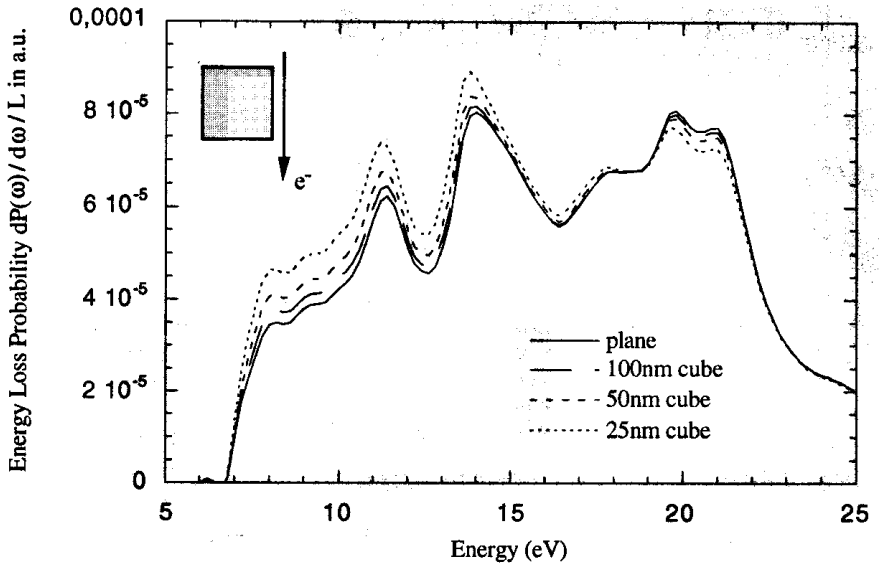

Fig 3. Energy loss probability simulated for a 100 keV electron passing near an MgO cube as shown in the inset. The probability is normalised to the length of the cube L for different sizes. The impact parameter is 2nm in all the cases.

The simulated spectra for different sizes of cube and the fixed 2nm impact parameter are shown in fig.3. Due to the presence of the edges, the planar contribution is reduced to compensate the enhancement of the low energy peaks. This effect is clearer as the cube size is smaller. In the case of a free electron material, unlike MgO, the bulk, surface and edge modes all give sharp and well separated losses. It is then possible to identify contributions from some of the other, weaker edge modes with an accuracy of 1% in the A_j coefficients.

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