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EELS of Bloch waves in wedge-shaped crystals

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ABSTRACT: Wedge-shaped crystals give rise to thickness fringes in the image and a splitting of spots in the diffraction pattern. The splitting arises because different Bloch states experience different refractive indices. These Bloch states might be expected to show energy loss spectra that correspond to the atomic species on which they are located. Calculations show favourable candidates for the effect include InP and MgO in 2-beam condition. Preliminary experimental results show that the splitting may not be maintained in the loss electrons.

1. WEDGE-SHAPED TEM SPECIMENS

Flat and uniformly thin is how microscopists generally prefer their specimens. But a wedge-shaped specimen has its uses, especially when the thickness increases uniformly and rapidly. MgO smoke crystals form perfect cubes and as early as 1946, Cowley and Rees observed splitting in the spots in the diffraction pattern caused by refraction. The refraction fine structure of the diffraction spots has been recorded as a function of crystal orientation allowing branches of the dispersion surface to be plotted directly (Lehmpfuhl and Reissland, 1968).

In the image, the effect is manifested as thickness fringes and the intensity distribution can be used for measuring Fourier coefficients of the crystal potential: see Cowley (1969) for a review of the early work. Thickness fringes in AlGaAs were examined by Kakibayashi and Nagata (1985) and others. A perfect 90° specimen can be made by cleaving on non-parallel 110 planes and the fringe profile, which varies with the Al content, can be compared with calculations for a composition measurement. HREM studies have taken advantage of wedge-shaped specimens to obtain through-thickness series in, for example, MgO (O'Keefe *et al.* 1985). With a crystal in an orientation such that there is no strong diffraction, the refraction effect caused by the mean inner potential may be used to generate topographical contrast in specimens (Cullis and Maher, 1975).

The effect we investigate in this work involves tilting the crystal to a strongly diffracting condition, then using the wedge shape of the specimen to refract electrons by different angles according to the potential (*i.e.* refractive index) they experience.

2. BLOCH WAVE LOCALISATION

The standard diagram depicting 2-beam Bloch waves shows one wave on the atom sites and the other sitting between atom sites (see *e.g.* Williams and Carter 1996). In order to increase our chances of seeing differences in the loss spectra, we examined MgO and InP, in which the Bloch waves may sit on different atom species. Projected structures having separated atom strings are found at <110> in MgO

and <100> in InP. We performed Bloch wave calculations for these cases (see fig. 1 for MgO) in zone axis orientation and in a 2-beam orientation (s=0, full systematic row calculation) using g=111 for MgO and g=002 for InP. These planes have alternating sheets of the different atom types. The 2-beam cases show, as expected, two strongly excited Bloch waves located on different atoms; the zone axis cases may show additional excited but delocalised Bloch states depending on incident kV.

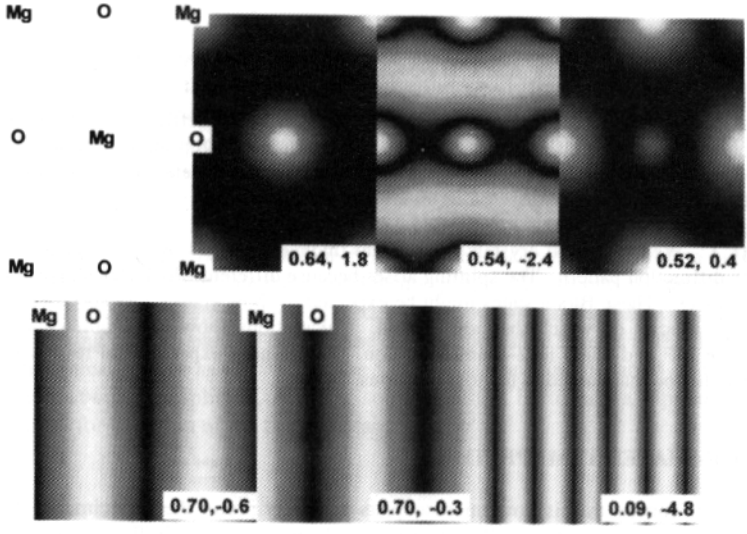

Fig 1. Location of first three Bloch states for MgO at zone axis and 2-beam with g=111. White indicates high density of electrons, numbers are (C_0, Δk). C_0 is excitation, Δk is increase in length of wavevector relative to that given by the mean inner potential alone, unit = 10^{-3}Å^{-1}.

3. EXPERIMENT AND RESULTS

Lying behind the choice of MgO and InP as suitable materials was the practicability of preparing nice wedges with appropriate zone axes along the beam direction when the wedges are aligned with the leading edge horizontal. The MgO was prepared in the traditional way by burning magnesium ribbon in air and collecting smoke particles on holey carbon film. InP was prepared by cleaving a previously thinned wafer on non-parallel planes and mounting upright on a slot grid with silver-loaded glue. The MgO was examined at 200kV in a JEOL 2010F, crystals were found that were both at <110> and rotated correctly so that the split spots were perpendicular to the dispersion direction in a Gatan imaging filter. The InP specimen was examined at 300kV in a Philips CM300FEG. The splitting found at the diffraction spots was examined by energy-filtered imaging and the "spectrum-diffraction pattern" method.

MgO oriented at or near to a 2-beam condition on g=111 has the 111 spot split by 0.23mrad (fig. 2), equivalent to a thickness fringe spacing of 11nm. For comparison, the diffraction angle between the unscattered beam and g=111 is 10.3mrad. These spots were admitted by the imaging filter entrance aperture and dispersed to form the double spectrum (fig 2). In principle, we were looking for differences between the two spectra, especially around the Mg and O K edges at 1305 and 532eV loss respectively. Although at zero loss the spots are well separated (as seen in the upper part of fig 2), at higher losses the two spectra are less distinct. A Mg $L_{2,3}$ signal (at ~50-80eV loss) may be visible in fig 2, but further observations are required to determine if that signal is split between the two spots.

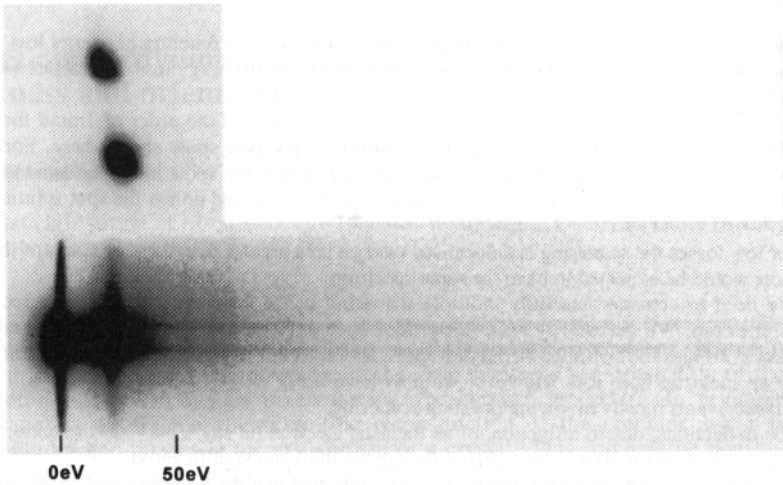

Fig 2. Split g=111 spot in MgO: image and "spectrum-diffraction pattern"(inverted contrast).

Observations of split spots in InP showed a similar spreading of the spots as we move to electrons at higher energy loss, see fig. 3. Instead of exact 2-beam conditions, diffraction conditions of g(4g) were chosen to increase the g=200 spot splitting (in the image, the thickness fringe spacing decreases). The spot splitting is 0.15mrad, thickness fringe spacing is 13nm. This weak beam condition may also help boost the effect by further localising the Bloch states onto atom cores.

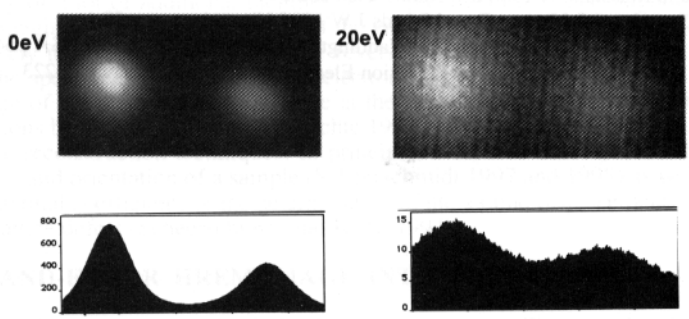

Fig 3. Portion of InP filtered diffraction pattern showing g=200 spot at zero loss and at 20eV loss

4. DISCUSSION

There are several reasons for the diffraction spots broadening as a function of energy loss.

1. The spectrometer focus and microscope focus both vary with energy loss. In practice we believe this is a small effect in this case.

2. For core loss scattering the associated angular scattering is of the order of 1mrad for OK and 3mrad for MgK, both greater than the ~0.2mrad splitting of the two spots shown here. For 200 kV electrons the energy loss above which the angular scattering makes the spots indistinguishable is about 60eV meaning that only very low energy core loss edges can be examined unless the spot splitting can be increased.

3. For low losses the scattering is delocalised over an area greater than the separation of the atoms, thus both spots would be expected to have the same spectrum.

4. We need to consider carefully multiple scattering in the specimen (which the effect we are looking for relies on). We necessarily need fairly thick specimens in order to have a wedge containing enough thickness fringes to form split diffraction spots, and need one elastic scattering event (diffraction) followed by an inelastic core loss. However we also have a big range of thickness and will also see multiple inelastic events mostly involving plasmon scattering.

5. The deflections, due to refraction, of an inelastic electron (at say 199kV) at the bottom surface and of the elastic electron (at 200kV) may differ, though we think not significantly.

It is interesting to note that Midgley *et al.* (1995) reported that the branches in the first order Laue zone of the convergent beam pattern of TiO_2 remained separated at energy losses up to 600eV. While this is, to some extent, due to the greater separation of the two branches (~8mrad), it may also indicate additional elastic scattering after the inelastic losses.

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