Why does the *hkl: h+k+l = 4n+2* reflections reveal intensity in Si [110]?

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For crystals with space group $Fd\bar{3}m$ like diamond, Si, Ge and Sn the *hkl: h+k+l = 4n+2* reflections are kinematically extinct. This can be shown by means of the squared structure factor, regarding the structure as two interpenetrating face centered cubic lattices displaced by a vector (¼ ¼ ¼):

$$\left|F_{hkl}\right|^2 = 4f^2\left[1+(-1)^{h+k}+(-1)^{h+l}+(-1)^{k+l}\right]\cos^2\left(\frac{\pi}{4}(h+k+l)\right)$$

It is clear that the cosine term is vanishing for *h+k+l = 4n+2*. One would expect that these reflections appear strongly by multiple diffraction. However, it is observed in dynamical simulations and in experiments that the reflections are still weak at thicknesses for which the diffraction already can be considered as highly dynamical. This can be explained by making use of the channelling theory, which describes in real space, in case of a zone-axis orientation, the dynamical scattering for an electron trapped in a column. Let us make this more clear by a concrete example; imagine a Si crystal in [110] orientation. Looking at the crystal structure (Fig. 1a) the projection of the potential on to the (001) axis has a periodicity, which is one fourth of the dimension of the unit cell, since in projection the projected (002) and (004) planes are identical. In reality the diffracted wave is not the Fourier transform of the crystal potential, except for thin crystals, but the Fourier transform of the exit wave of the crystal. Let us consider this exit wave in detail. If an electron, being trapped in a column is not affected by the neighboring columns, the exit wave at the column keeps the rotational symmetry. In that case the exit wave of the crystal is then the superposition of the constituting column exit waves. To a good approximation the exit wave can then be described as a linear combination of the 1S eigenfunctions of the columns [1,2]:

$$\psi(R,z) = 1 + \sum_i 2c^i_{1S}\sin\left\{\pi\frac{E^i_{1S}}{2E_0}kz\right\}\varphi^i_{1S}(R-R_i)\exp\left\{-i\frac{\pi}{2}\left(\frac{E^i_{1S}}{E_0}kz+1\right)\right\}$$

with, i the column index, E_0 the incident electron energy and k the inverse wavelength. The local rotational symmetry breaks down for larger thicknesses. For instance at 13 nm where the projection of the exit wave of Si [110] on the (001) axis has a periodicity of half the unit cell (Fig. 1b). The overlap of the potentials and eigenfunctions of the atom columns affect the local rotational symmetry. This effect is not sufficiently described by the simple channelling model, and increases with increasing thickness. This thickness dependence thus provides a good test for the validity of the simple channelling theory. Fig. 2 shows the normalized total intensity of all the forbidden reflections, calculated using a multislice program. The intensity is negligible below a thickness of 10 nm and increases to 16% of the total diffracted intensity only at a specimen thickness of 50 nm. It can be concluded that, for thicknesses in the range used for HREM, the local rotational symmetry, to a good approximation, is preserved and that the simple channelling theory provides a rather good, intuitive, simple description of dynamical electron diffraction for crystals in zone-axis orientation. Note that the symmetry breaking can be enhanced by a small tilt away from the zone-axis orientation [3].

References
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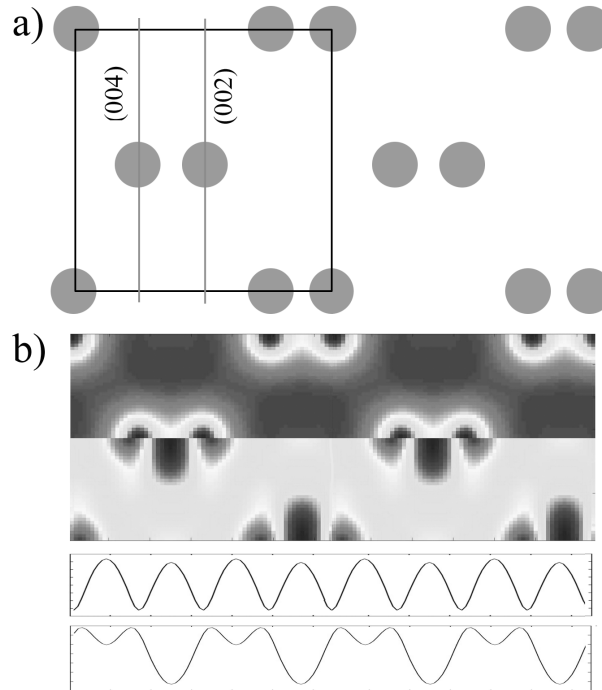

Figure 1 a) Si [110] (half of the cubic unit cell is shown by a rectangular), in the projection the periodicity along (001) is one fourth of the unit cell. b) The real (top) and imaginary (bottom) part of the complex exit wave of Si [110] at a specimen thickness of 13 nm. From the line plots of the projected real (top) and imaginary (bottom) part of the exit wave it is clear that the periodicity is doubled compared to a).
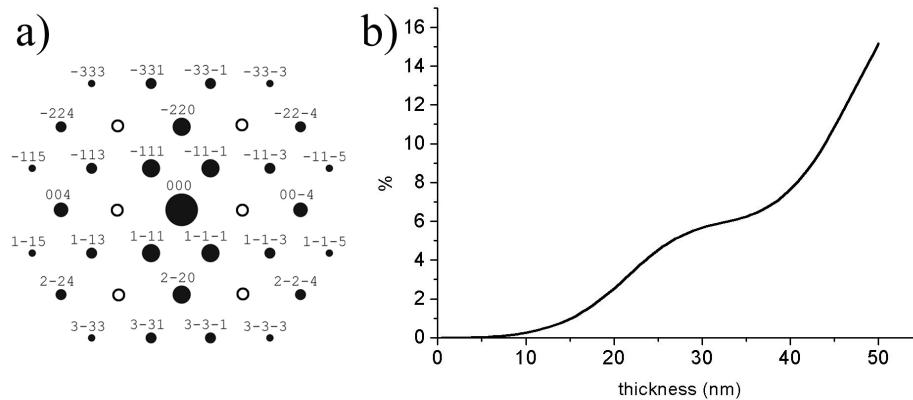

Figure 2 a) Diffraction pattern of Si [110] (kinematical theory). Circles mark the *hkl: h+k+l = 4n+2* reflections. b) The normalized total intensity of the *hkl: h+k+l = 4n+2* reflections for Si [110] (Debye Waller factor = 0.6 Å^2) in function of thickness.