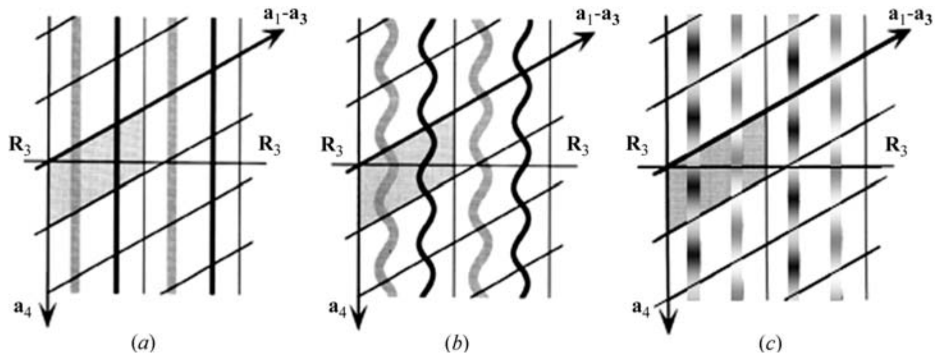


Fig. 2.5.3.17. The (3 + 1)-dimensional description of one-dimensionally modulated crystals. Atoms are shown as strings along the fourth direction $\mathbf{a}_4$. (*a*) No modulation, shown as straight strings. (*b*) Displacive modulation, shown as wavy strings. (*c*) Amplitude modulation with varying-density strings.

Tanaka (1993) clarified theoretically the interrelation between the symmetries of CBED patterns and the (3 + 1)-dimensional point-group symbols for incommensurately modulated crystals and verified experimentally the theoretical results for $Sr_2Nb_2O_7$ and Mo_8O_{23}. Terauchi *et al.* (1994) investigated dynamical extinction for the (3 + 1)-dimensional space groups. They clarified that approximate dynamical extinction lines appear in CBED discs of the reflections caused by incommensurate modulations when the amplitudes of the incommensurate modulation waves are small. They tabulated the dynamical extinction lines appearing in the CBED discs for all the (3 + 1)-dimensional space groups of the incommensurately modulated crystals. The tables were stored in the British Library Document Supply Centre as Supplementary Publication No. SUP 71810 (65 pp.). They showed an example of the dynamical extinction lines obtained from $Sr_2Nb_2O_7$. The point- and space-group determinations of the (3 + 1)-dimensional crystals are described compactly in the book by Tanaka *et al.* (1994, pp. 156–205).

Fig. 2.5.3.17 illustrates (3 + 1)-dimensional descriptions of a crystal structure without modulation (*a*), a one-dimensional displacive modulated structure (*b*) and a one-dimensional substitutionally modulated structure (*c*). The arrows labelled $\mathbf{a}_1$–$\mathbf{a}_3$ (**a**, **b** and **c**) and $\mathbf{a}_4$ indicate the (3 + 1)-dimensional crystal axes. The horizontal line labelled $\mathbf{R}_3$ represents the three-dimensional space (external space). In the (3 + 1)-dimensional description, an atom is not located at a point as in the three-dimensional space, but extends as a string along the fourth direction $\mathbf{a}_4$ perpendicular to the three-dimensional space $\mathbf{R}_3$. The shaded parallelogram is a unit cell in the (3 + 1)-dimensional space. The unit cell contains two atom strings in this case. In the case of no modulations, the atoms are shown as straight strings, as shown in Fig. 2.5.3.17(*a*). For a displacive modulation, atoms are expressed by wavy strings periodic along the fourth direction $\mathbf{a}_4$ as shown in Fig. 2.5.3.17(*b*). The width of the atom strings indicates the spread of the atoms in $\mathbf{R}_3$. The atom positions of the modulated structure in $\mathbf{R}_3$ are given as a three-dimensional ($\mathbf{R}_3$) section of the atom strings in the (3 + 1)-dimensional space. A substitutional modulation, which is described by a modulation of the atom form factor, is expressed by atom strings with a density modulation along the direction $\mathbf{a}_4$ as shown in Fig. 2.5.3.17(*c*).

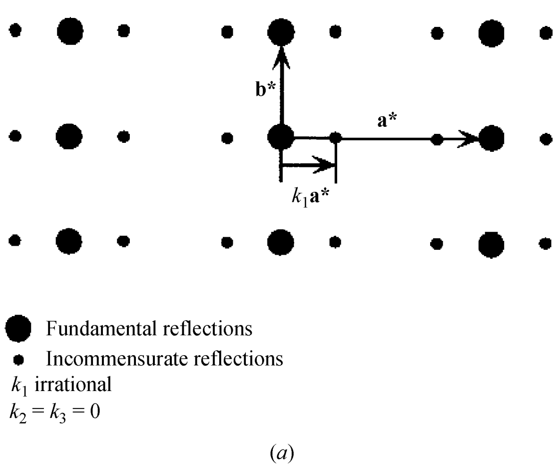
The diffraction vector **G** is written as

$$\mathbf{G} = h_1\mathbf{a}^* + h_2\mathbf{b}^* + h_3\mathbf{c}^* + h_4\mathbf{k},$$

where a set of $h_1h_2h_3h_4$ is a (3 + 1)-dimensional reflection index, and $\mathbf{a}^*$, $\mathbf{b}^*$ and $\mathbf{c}^*$ are the reciprocal-lattice vectors of the real-lattice vectors **a**, **b** and **c** of the average structure. The modulation vector **k** is written as

$$\mathbf{k} = k_1\mathbf{a}^* + k_2\mathbf{b}^* + k_3\mathbf{c}^*,$$

where one coefficient k_i (i = 1–3) is an irrational number and the others are rational. Fig. 2.5.3.18(*a*) shows a diffraction pattern of a crystal with an incommensurate modulation wavevector $k_1\mathbf{a}^*$ (k_2 and k_3 = 0). Large and small black spots show the fundamental reflections and incommensurate reflections, respectively, only the first-order incommensurate reflections being shown. It



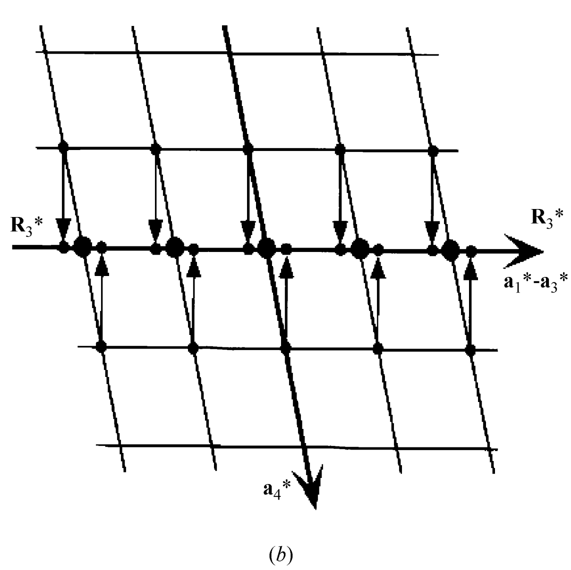


Fig. 2.5.3.18. (*a*) Schematic diffraction pattern from a modulated crystal. As an example, the wave number vector of modulation is assumed to be $k_1\mathbf{a}^*$, k_1 being an irrational number. Large and small spots denote fundamental and incommensurate reflections, respectively. (*b*) Incommensurate reflections are obtained by a projection of the Fourier transform of a (3 + 1)-dimensional periodic structure.