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A revised version of the program VEC (visual computing in electron crystallography)*

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The program package VEC (Visual computing in Electron Crystallography) has been revised such that (i) a program converting one-line symbols to two-line symbols of (3+1)-dimensional superspace groups has been incorporated into VEC so that the latter can interpret both kinds of symbols; (ii) a bug in calculating structure factors of one-dimensionally incommensurate modulated crystals has been fixed. The correction has been verified by successfully matching the experimental electron microscopy image of an incommensurate crystal with a series of simulated images. The precompiled revised version of VEC and relevant materials are available on the Web at http://cryst.iphy.ac.cn.

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1. Introduction

VEC is a computer program package for "Visual computing in Electron Crystallography".[1−3] The package is written in C++ and FORTRAN and is for running under MS Windows 95 up to Windows XP. VEC can perform a series of unique crystallographic computations including direct-method image deconvolution and/or enhancement, *ab initio* solution of incommensurate structures, and tracing of one-dimensional modulation wave functions in (3+1)-dimensional Fourier maps. In dealing with incommensurate structures VEC takes advantage of superspace-group symbols to avoid manual input of symmetry operations. There are two kinds of superspace-group symbols,[4] i.e. one-line symbols and two-line symbols. For historical reasons, the original version of VEC can only recognize two-line symbols. However, one-line symbols are now commonly used. Hence there is a need to make VEC recognize one-line symbols also. VEC can simulate dynamical electron diffraction of incommensurate structures using the modified multislice algorithm proposed by Sha *et al* [5] (see also the VEC tutorials in the section "Simulation/multislice method: aperiodic crystals" at http://cryst.iphy.ac.cn). However, a bug has been found in the calculation of structure factors of incommensurate structures. This has been fixed in the present revised version.

2. Converting one-line symbols to two-line symbols

The definitions of and the relation between one-line and two-line symbols of (3+1)-dimensional superspace groups are described in detail by Yamamoto[4] (see Section 3.1 and Table 1 in his paper). The program SYMBOL1to2 has been written and incorporated into VEC for converting one-line symbols to two-line symbols. The program can distinguish two-line symbols from one-line symbols. When the input symbol is a two-line symbol, the program will invoke SPGR4D,[6] a program which derives (3+l)-dimensional symmetry operations from two-line symbols, and pass on directly the input symbol to it. SPGR4D will then derive and output the corresponding symmetry operations. On the other hand, if the input symbol is a one-line symbol, SYMBOL1to2 will derive the corresponding two-line symbol and then pass on the result to SPGR4D to get the symmetry operations. Figure 1 shows four outputs which correspond to four different but equivalent input (3+l)-dimensional superspace-group symbols.

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Input symbol: P[B 2/M]-1 S

-----------------------------------   1
SYMMETRY CLASS:     MONOCLINIC
SUPERSPACE GROUP:     P[B 2/M]-1 S
          +(0 0 0 0   1/2 0 1/2 0)

   1  0  0  0  .00   -1  0  0  0  .00    1  0  0  0  .00   -1  0  0  0  .00
   0  1  0  0  .00    0 -1  0  0  .00    0  1  0  0  .00    0 -1  0  0  .00
   0  0  1  0  .00    0  0  1  0  .00    0  0 -1  0  .00    0  0 -1  0  .00
   0  0  0  1  .00    0  0  0 -1  .50    0  0  0  1  .50    0  0  0 -1  .00

Input symbol: B 2/M(A B 0)0 S
Two-line symbol: P[B 2/M]-1 S

-----------------------------------   1
SYMMETRY CLASS:     MONOCLINIC
SUPERSPACE GROUP:     P[B 2/M]-1 S
          +(0 0 0 0   1/2 0 1/2 0)

   1  0  0  0  .00   -1  0  0  0  .00    1  0  0  0  .00   -1  0  0  0  .00
   0  1  0  0  .00    0 -1  0  0  .00    0  1  0  0  .00    0 -1  0  0  .00
   0  0  1  0  .00    0  0  1  0  .00    0  0 -1  0  .00    0  0 -1  0  .00
   0  0  0  1  .00    0  0  0 -1  .50    0  0  0  1  .50    0  0  0 -1  .00

Input symbol: C 2/M(A 0 G)0 S
Two-line symbol: P[C 2/M]-1 S :B

-----------------------------------   1
SYMMETRY CLASS:     MONOCLINIC
SUPERSPACE GROUP:     P[C 2/M]-1 S :B
          +(0 0 0 0   1/2 1/2 0 0)

   1  0  0  0  .00   -1  0  0  0  .00    1  0  0  0  .00   -1  0  0  0  .00
   0  1  0  0  .00    0  1  0  0  .00    0 -1  0  0  .00    0 -1  0  0  .00
   0  0  1  0  .00    0  0 -1  0  .00    0  0  1  0  .00    0  0 -1  0  .00
   0  0  0  1  .00    0  0  0 -1  .50    0  0  0  1  .50    0  0  0 -1  .00

Input symbol: C 2/M(0 B G)0 S
Two-line symbol: P[C 2/M]-1 S :A

-----------------------------------   1
SYMMETRY CLASS:     MONOCLINIC
SUPERSPACE GROUP:     P[C 2/M]-1 S :A
          +(0 0 0 0   1/2 1/2 0 0)

   1  0  0  0  .00    1  0  0  0  .00   -1  0  0  0  .00   -1  0  0  0  .00
   0  1  0  0  .00    0 -1  0  0  .00    0  1  0  0  .00    0 -1  0  0  .00
   0  0  1  0  .00    0  0 -1  0  .00    0  0  1  0  .00    0  0 -1  0  .00
   0  0  0  1  .00    0  0  0 -1  .50    0  0  0  1  .50    0  0  0 -1  .00
```

Fig.1. Outputs of Symbol1to2-SPGR4D from different but equivalent input (3+1)-dimensional superspace group symbols.

For a thorough examination of the combination of SYMBOL1to2 and SPGR4D, the 775 non-equivalent one-line symbols of (3+1)-dimensional superspace groups listed in the International Tables for Crystallography[7] were input to SYMBOL1to2 and the resulting symmetry operations from SPGR4D were compared with those listed by Ivan P. Orlov on the Web at http://superspace.epfl.ch/groups/. There are 229 superspace groups among the total of 775, for which symmetry operations derived by SPGR4D are different from those of Orlov. This is due to the shift of unit-cell origin and/or the direction choice of the 4$^{\mathrm{th}}$ axis. Symmetry operations of the 229 superspace groups from SPGR4D can be made coincident with those of Orlov via the following operation:

$$S_{\mathrm{Orlov}} = T^{-1} S_{\mathrm{SPGR4D}} T, \tag{1}$$

where

$$S_{\mathrm{Orlov}} = \begin{pmatrix} R_{\mathrm{Orlov}} & \boldsymbol{t}_{\mathrm{Orlov}} \\ \boldsymbol{o} & 1 \end{pmatrix},$$

$$S_{\mathrm{SPGR4D}} = \begin{pmatrix} R_{\mathrm{SPGR4D}} & \boldsymbol{t}_{\mathrm{SPGR4D}} \\ \boldsymbol{o} & 1 \end{pmatrix},$$

$$T = \begin{pmatrix} R & \boldsymbol{t} \\ \boldsymbol{o} & 1 \end{pmatrix}$$

are 5×5 matrixes, $\{R_{\mathrm{Orlov}}|\boldsymbol{t}_{\mathrm{Orlov}}\}$ and $\{R_{\mathrm{SPGR4D}}|\boldsymbol{t}_{\mathrm{SPGR4D}}\}$ are 4-dimensional symmetry operations listed by Orlov and derived from SPGR4D respectively, $\{R|\boldsymbol{t}\}$ is a 4-dimensional operation defining the shift of unit-cell origin and/or the direction choice of the 4$^{\mathrm{th}}$ axis, and $\boldsymbol{o} = (0,\ 0,\ 0,\ 0)$. The rotation matrix R can only be one of the following forms:

$$R_0 = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix},$$

$$R_1 = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & -1 \end{pmatrix}$$

and

$$R_2 = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 1 & 1 & 1 & -1 \end{pmatrix}.$$

For a detailed description of transformations of symmetry operations, the reader is referred to Ref.[8]. Tables are available at http://cryst.iphy.ac.cn, in which one-line symbols are listed alongside of the corresponding two-line symbols for the 775 non-equivalent (3+1)-dimensional superspace groups together with values of $\{R|\boldsymbol{t}\}$ for the 229 superspace groups. The stand-alone program combination of SYMBOL1to2 + SPGR4D is also available on the website.

3. Fixing the bug in calculating structure factors of incommensurate modulated structures

In the 4-dimensional representation of (3+1)-dimensional incommensurate structures (see the VEC tutorials in the section "Phasing diffraction data/Theory: 4D-representation" at http://cryst.iphy.ac.cn), the position vector of atoms is expressed as

$$\boldsymbol{x} = x_1\boldsymbol{a}_1 + x_2\boldsymbol{a}_2 + x_3\boldsymbol{a}_3 + x_4\boldsymbol{a}_4, \tag{2}$$

where $\boldsymbol{a}_1$, $\boldsymbol{a}_2$, $\boldsymbol{a}_3$ and $\boldsymbol{a}_4$ are vectors defining the 4-dimensional unit cell in direct space; x_1, x_2, x_3 and x_4 are respectively the fractional coordinates in directions of $\boldsymbol{a}_1$, $\boldsymbol{a}_2$, $\boldsymbol{a}_3$ and $\boldsymbol{a}_4$. Coordinates x_1, x_2, x_3 and x_4 are in turn expressed as

$$x_1(\bar{x}_4) = \bar{x}_1 + u_1(\bar{x}_4), \tag{3}$$

$$x_2(\bar{x}_4) = \bar{x}_2 + u_2(\bar{x}_4), \tag{4}$$

$$x_3(\bar{x}_4) = \bar{x}_3 + u_3(\bar{x}_4), \tag{5}$$

$$x_4(\bar{x}_4) = q_1x_1(\bar{x}_4) + q_2x_2(\bar{x}_4) + q_3x_3(\bar{x}_4). \tag{6}$$

In Eqs.(3)–(6), $\bar{x}_1$, $\bar{x}_2$, $\bar{x}_3$ and $\bar{x}_4$ are average values of x_1, x_2, x_3 and x_4 respectively, while u_1, u_2 and u_3 are components of the modulation wave function in directions of $\boldsymbol{a}$, $\boldsymbol{b}$, and $\boldsymbol{c}$ respectively which define the unit cell of the basic structure. Finally q_1, q_2 and q_3 are components of the modulation wave vector

$$\boldsymbol{q} = q_1\boldsymbol{a}^* + q_2\boldsymbol{b}^* + q_3\boldsymbol{c}^*, \tag{7}$$

where $\boldsymbol{a}^*$, $\boldsymbol{b}^*$ and $\boldsymbol{c}^*$ are vectors defining the reciprocal unit cell of the basic structure. In the original version of VEC, equation (6) is wrongly implemented as

$$x_4(\bar{x}_4) = \bar{x}_4 + [q_1x_1(\bar{x}_4) + q_2x_2(\bar{x}_4) + q_3x_3(\bar{x}_4)]. \tag{8}$$

This error has now been corrected. The revised version of VEC has been tested in multislice calculation of electron microscopy images. The superconductor $Bi_{2.31}Sr_{1.69}CuO_{6+\delta}$ was used as an example, crystals of which belong to the (3+1)-dimensional superspace group B2/b($\alpha\beta$0) (one-line symbol) or equivalently P[B2/b]–1 1 (two-line symbol) with basic-structure cell parameters $a = 5.37$ Å, $b = 5.43$ Å, $c = 24.53$ Å (1 Å=0.1 nm); $\alpha = \beta = \gamma = 90°$ and the modulation wave vector $\boldsymbol{q} = 0.223\boldsymbol{b}^* + 0.618\boldsymbol{c}^*$. The incommensurate structure model[9] was obtained using the program DIMS on the VEC platform.[2] Figure 2 shows a series of nine simulated electron microscopy images with the experimental image on top for comparison. As is seen, the experimental image is very well matched with the simulated one which has the defocus value at –380 Å and sample thickness of 32 Å.

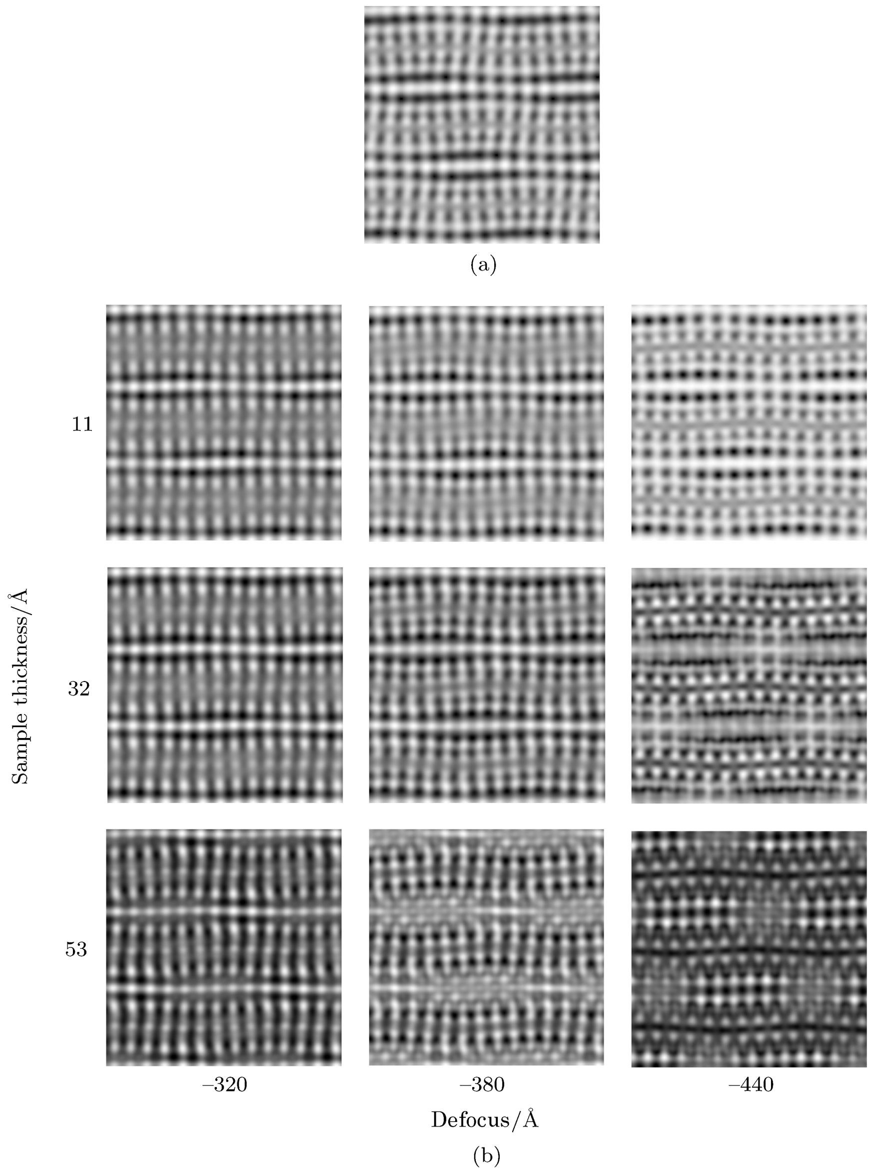

Fig.2. [100] electron microscopy images of the superconductor $Bi_{2.31}Sr_{1.69}CuO_{6+\delta}$, (a) experimental image taken near Scherzer focus with a JEOL 2010 high-resolution electron microscope under the condition: accelerating voltage $U = 200$ kV, spherical aberration $C_s = 0.5$ mm, defocus spread $D = 70$ Å, defocus value $\Delta f = -380$ Å and divergence half-angle $\alpha = 0.01$ mrad; (b) a series of nine multislice simulated images with different defocus and/or sample thickness calculated with $U = 200$ kV, $C_s = 0.5$ mm, $D = 100$ Å and $\alpha = 0$ mrad.

4. Concluding remarks

The revised version of VEC provides considerably better performance in dealing with incommensurate crystal data. The ability to accept both one-line and two-line (3+1)-dimensional superspace-group symbols avoids the manual input of complicated symmetry operations. This is beneficial to calculations involving incommensurate crystals. The modified multislice algorithm for incommensurate structures makes the simulation of electron microscopy images simpler and more accurate for incommensurate crystals. Otherwise a large approximate 3-dimensional unit cell should be chosen, which is an inaccurate representation and is inconvenient to deal with.

For publications involving the use of the program VEC and relevant materials, please cite the present paper and Ref.[10].

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