Utility for Calculating Diffuse Scattering for RMCProfile Configurations

The routine **diffuse.exe** calculates x-ray/electron diffuse scattering for an atomic configuration described by the RMCProfile *.cfg file.

The procedure implemented in this routine follows that described by Butler and Welberry (*J. Appl. Cryst., 25, 391-399, 1992*). The total scattering amplitude $F_T(\mathbf{q})$ is calculated by a direct summation over atomic coordinates according to

$$F_{\mathrm{T}}(\mathbf{q}) = \sum_{n=1}^{N}\sum_{j=1}^{M} f_j(q)\exp\left(i\mathbf{q}\left(\mathbf{R}_n + \mathbf{r}_{nj}\right)\right) \tag{1}$$

where $\mathbf{R}_n$ are the coordinates of the n^{th} unit cell, $\mathbf{r}_{nj}$ are the coordinates of j-th atom in the n^{th} unit cell, $N=N_xN_yN_z$ is the total number of unit cells in the box, and M is the total number of atoms in the unit cell.

The average structure amplitude is calculated as

$$F_{\mathrm{A}}(\mathbf{q}) = \frac{1}{N}\sum_{n=1}^{N}\sum_{j=1}^{M} f_j(q)\exp\left(i\mathbf{q}\mathbf{r}_{nj}\right) \tag{2}$$

Then, the diffuse scattering amplitude is

$$F_{\mathrm{D}}(\mathbf{q}) = F_T(\mathbf{q}) - F_A(\mathbf{q})\psi(\mathbf{q}), \tag{3}$$

where the interference function is

$$\psi(\mathbf{q}) = \sum_{n=1}^{N}\exp\left(i\mathbf{q}\mathbf{R}_n\right) = \frac{\left[\exp\left(i2\pi hN_1\right)-1\right]\left[\exp\left(i2\pi kN_2\right)-1\right]\left[\exp\left(i2\pi lN_3\right)-1\right]}{\left[\exp\left(i2\pi h\right)-1\right]\left[\exp\left(i2\pi k\right)-1\right]\left[\exp\left(i2\pi l\right)-1\right]} \tag{4}$$

and $\mathbf{q}=(h,k,l)$.

The resulting diffuse intensity is

$$I_D(\mathbf{q}) = \left|F_D(\mathbf{q})\right|^2. \tag{5}$$

Values of $\mathbf{q}_{ijn}$ are selected on a rectangular mesh in the reciprocal lattice plane defined by the three points (000), $(h_1k_1l_1)$, and $(h_2k_2l_2)$:

$$\begin{aligned}&\mathbf{q}_{ijn} = (a_ih_1 + b_ih_2, a_jk_1 + b_jk_2, a_nl_1 + b_nl_2).\\ &a_i=a_{\min}+(i\text{-}1)(a_{\max}\text{-}a_{\min})/(N_{\mathrm{a}}\text{-}1),\ i=1\ldots N_a\\ &b_j=b_{\min}+(j\text{-}1)(b_{\max}\text{-}b_{\min})/(N_{\mathrm{b}}\text{-}1)\ j=1\ldots N_b\end{aligned} \tag{6}$$

The Parameters h_1, k_1 , l_1, h_2, k_2, l_2, a, b, N_a, and N_b are defined in the input file **diffuse.inp** (see below).

Atomic scattering amplitudes are calculated according the formula (*L-M. Peng, Acta Cryst., A54, 481-485, 1998*)

$$f(q) = 0.023934\Delta Z / q^2 + \sum_{j=1}^{5} \alpha_j \exp\left(-\beta_j q^2\right), \qquad (7)$$

where ΔZ is an ionicity of the atom in the compound and the coefficients α_j and β_j are defined in the input file **diffuse.inp**.

Periodic boundary conditions assumed for an RMC atomic configuration imply that the scattering intensity can be averaged by consecutive transposing of the unit cell layers from one box boundary to the opposite side of the box (equivalent to sliding a boundary of the sampling box used in the scattering calculations from one side of the RMC box to another). The numbers of times this transformation is applied along each box axis (P_x, P_y, and P_z) are defined in the input file **diffuse.inp**.

An example of the **diffuse.inp** file for the atomic configuration containing two types of atoms is given below.

```
initial.cfg                  ! File containing atomic coordinates before refinements
final.cfg                        ! File containing atomic coordinates after refinements
4 8 1                            !Px Py Pz
25 25 25                         ! Nx Ny Nz
-0.1 1.1 300 -0.1 1.1 100        ! amin amax Na bmin bmax Nb
0  1  4                          ! h1k1l1
1  0  0                          ! h2k2l2
5                                !Z1
0.0828 0.271 0.654 1.24 0.829    α1 α2 α3 α4 α5
0.0369 0.261 0.957 3.94 9.44      β1 β2 β3 β4 β5
-2                                !Z2
0.0421 0.210 0.852 1.82 1.17
0.0609 0.559 2.96 11.5 37.7
```

The file **initial.cfg** corresponds to the initial atomic configuration constructed from the average structure.

Three output files are generated: **diff_scatt** (diffuse scattering only), **tot_scat** (total scattering), and ref_scatt (Bragg only). The calculated intensities are presented as tables N_a(lines)×N_b(columns).

Four files are required in the directory used for the calculations: **diffuse.inp**, **diffuse.exe**, **initial.cfg**, and **final.cfg**. The program window displays (1) the number of layers that are transposed from one side of the box to another for averaging (i.e. P_x, P_y, and P_z), (2) the

lengths of reciprocal lattice vectors and (3) the relative lengths for the basis vectors $\delta_1[h_1k_1l_1]^*$ and $\delta_2[h_2k_2l_2]^*$, where $\delta_1=a_{max}-a_{min}$ and $\delta_2=b_{max}-b_{min}$; the latter information is used to obtain a correct scale while generating plots of diffracted intensity (vector $[h_1k_1l_1]$ is always vertical). The results of calculations can be visualized using any suitable software (e.g. Origin). Rename the files to avoid overwriting.