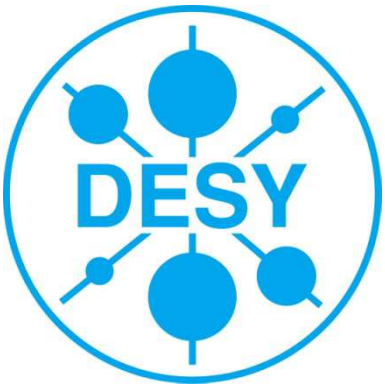

Coherent diffractive imaging of crystal with defects: can the structure of the undamaged crystal be recovered?

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Abstract

We have investigated the influence of the lattice defects on the diffraction image of crystals. The numerical tool was developed to calculate the intensity of the damaged crystals. Our analysis was performed for two types of uncorrelated defects – vacancies and displacements, and for two types of 2D lattices – square and hexagonal. Also, the analytical formulas for intensity were obtained for both types of defects. Finally, we obtained a general formula for intensity scattered from monoatomic crystals with uncorrelated defects, which allows us to recover the intensity from the undamaged crystal, if we know the underlying defect statistics.

Content

Introduction

When an incoming beam of x-rays interacts with a solid material, its scattering can be observed. There are two different types of scattering – elastic and inelastic. The former one is called Thomson scattering and it is the one used for structural investigations by x-ray diffraction. [1]

The incoming wave with the wave vector $\vec{k}_i$ scatters on an electron so the outcoming wave propagates with a wave vector $\vec{k}_f$, which has the same amplitude. The scattering vector $\vec{q}$ is then defined as

$$\vec{q} = \vec{k}_f - \vec{k}_i . \tag{1}$$

If we calculate the amplitude of the outcoming wave and sum it over all atoms in the crystal, we get

$$A(\vec{q}) = \sum_{j=1}^{N} f_j(\vec{q}) e^{i\vec{q}\vec{r}_j}, \tag{2}$$

where N is the number of atoms in the lattice, $\vec{r}_j$ is the position of the *j*-th atom and f_j is the atomic form factor defined as

$$f(\vec{q}) = \int \rho_{at}(\vec{r}') e^{i\vec{q}\vec{r}'} d^3\vec{r}', \tag{3}$$

where $\rho_{at}(\vec{r}')$ is the atomic electron density. Then we have an access to the intensity through

$$I(\vec{q}) \propto \left|A(\vec{q})\right|^2, \tag{4}$$

that is, unlike the complex amplitude, a measurable quantity.

It often occurs during diffraction imaging, that the measured crystal or biomolecule is slightly damaged, therefore the measured diffraction pattern does not correspond to the structure we wanted to determine – the undamaged crystal. The goal of this paper is to investigate, if it is ever possible to recover the intensity of the undamaged crystal from the intensity distribution of the damaged crystal.

I. Defects in crystal

Here we introduce the definitions of the crucial terms used in this paper.

Crystal – a solid with a spatial periodicity. [2]
Lattice – summarizes the geometry of the underlying periodic structure in crystal. [2] In 2D case, it is fully defined by the base vectors $\vec{a}_1, \vec{a}_2$. The linear combination of these vectors with the integer coefficients n_1, n_2 covers all the lattice points $\vec{r}$:

$$\vec{r} = n_1\vec{a}_1 + n_2\vec{a}_2 . \tag{5}$$

Defect – any region in the crystal, where the microscopic arrangement differs from that of a perfect crystal. [2]

$\boldsymbol{I_{ideal}}$ – the intensity obtained from an undamaged crystal, without defects.

$\langle \boldsymbol{I} \rangle_{\boldsymbol{R}}$ – the average intensity obtained from a crystal with some random defects.

The calculations were performed for two types of lattices – square and hexagonal (fig. 1) with the base vectors plotted in the figures.

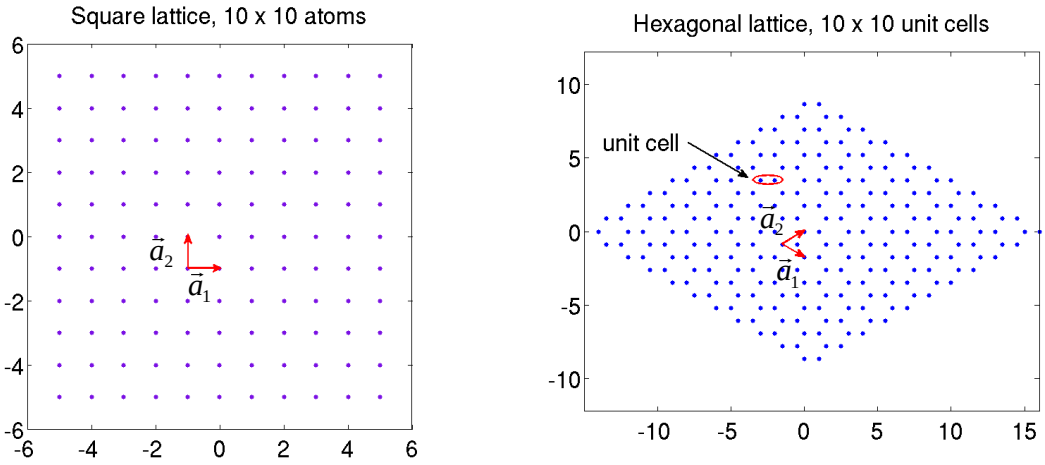

Fig. 1. The types of lattices used in calculations

All calculations were made using the interatomic distances as units. Atomic form factor was that of neutral carbon.

II. Imaging of monoatomic crystals with vacancies

The first type of the investigated defects were vacancies. Vacancies occur, when an atom is missing from a lattice site. To implement this type of defect, one can make the atomic form factor to be a random variable with a discrete probability distribution,

$$f_j(\vec{q}) = \begin{cases} 0, & p_v \\ f(\vec{q}), & 1-p_v \end{cases}, \tag{6}$$

where p_v stays for the probability of the vacancy. Once the form factor of a given atom equals zero, its contribution to the sum in the formula (2) becomes zero, i.e., the atom is missing. For this special case the analytical calculation of the intensity was performed with the result

$$\langle I(\vec{q})\rangle_R = (1-p_v)^2 I_{ideal}(\vec{q}) + N\left|f(\vec{q})\right|^2 p_v(1-p_v). \tag{7}$$

Fig. 2 shows the intensity as a function of the scattering vector $\vec{q}$ for two values of parameter p_v. First one is the intensity of the undamaged crystal ($p_v = 0$) and the second one is rather extreme case ($p_v = 0.8$) calculated numerically. In agreement with the analytical formula, it can be seen, that the intensity peaks are much brighter in the first case because in the second case the scaling factor $(1-p_v)^2$ and also the addition of a background term blur the diffraction pattern. For a detailed look, fig. 3 shows the intensity along the red line depicted in the right part of fig. 2. The periodically distributed Bragg peaks are still dominating over the background, but their magnitude decreases due to the vacancies. We also compared the results obtained by analytical and numerical approach to assure, that they are the same. For the numerical calculation we averaged over 100 realizations of random defects, and found them in agreement with the analytical formula.

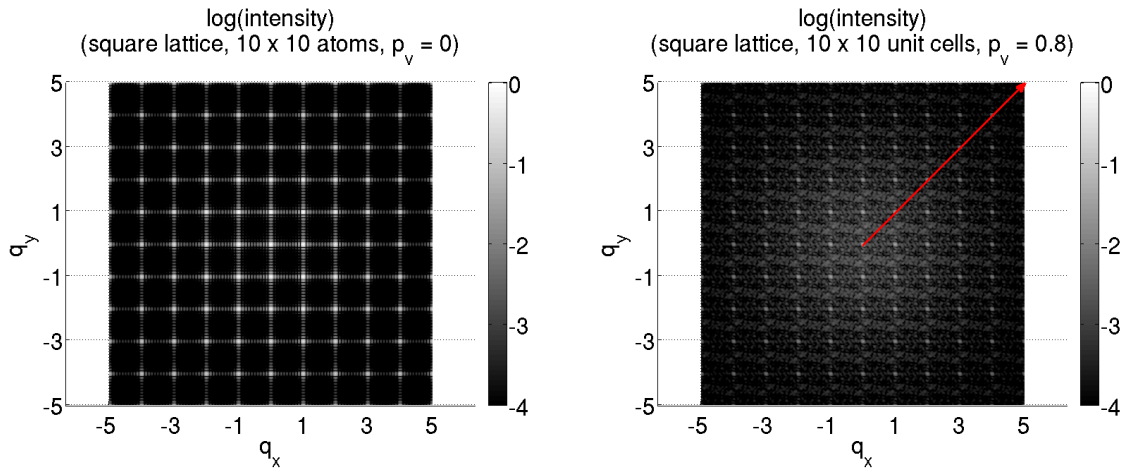

Fig. 2. Logarithm of intensity in 2D q-space for different probabilities of vacancy: $p_v = 0$ (left), $p_v = 0.8$ (right).

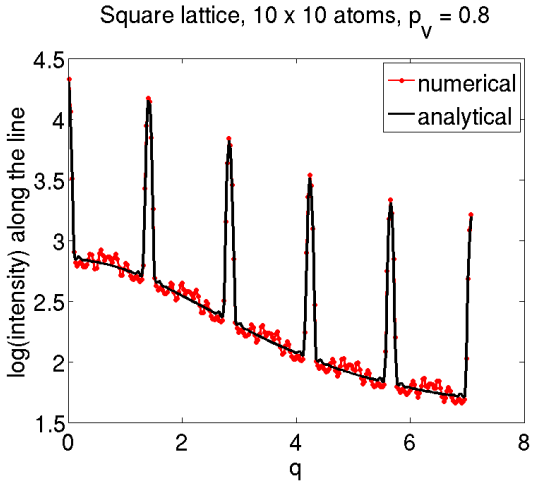

Fig. 3. Logarithm of the intensity along the line plotted in fig. 2 right as a function of $q = \sqrt{q_x^2 + q_y^2}$ *. Figure gives the comparison of the analytical and numerical results.*

III. Imaging of monoatomic crystals with displacements

The second type of defects we investigated were displacements. In our simplified model, the atom position can be displaced by a constant vector $\Delta\vec{r}$ with a given probability, thus it is a random variable with the probability distribution

$$\vec{r}_i = \begin{cases} \vec{r}_{i0} + \Delta\vec{r}, & \frac{p_d}{2} \\ \vec{r}_{i0}, & 1 - p_d \\ \vec{r}_{i0} - \Delta\vec{r}, & \frac{p_d}{2} \end{cases}, \tag{8}$$

where $\vec{r}_{i0}$ is the position of the i-th atom in the equilibrium position and p_d is the probability of the displacement. After the analytical calculation of the intensity one can obtain

$$\left\langle I(\vec{q})\right\rangle_R = \left[1-p_d+p_d\ \cos(\vec{q}\Delta\vec{r})\right]^2 I_{ideal}(\vec{q}) + N\left|f(\vec{q})\right|^2\left\{1-\left[1-p_d+p_d\cos(\vec{q}\Delta\vec{r})\right]^2\right\} \quad (9)$$

Fig. 3 shows the scattered intensity as a function of the scattering vector $\vec{q}$ for two values of parameter p_d .The first one corresponds to a crystal without defects (using the hexagonal lattice) and the second one is an extreme case with all the atoms displaced from their equilibrium position. According to formula (9), in the case of $\vec{q}$ vector perpendicular to the constant vector $\Delta\vec{r}$ (which has been chosen as (0.1; 0.1) in the figure), the multiplier of the I_{ideal} equals one and therefore for such $\vec{q}$ values there is no difference of damaged pattern when compared to undamaged crystal. On the other hand, for $\vec{q}$ values parallel to $\Delta\vec{r}$, the periodic behavior is present due to the cosine term and also the background term becomes stronger. Fig. 5 shows, similarly as in the case of vacancies, the intensity dependence along the red line depicted in the right part of fig. 4. Again, the numerical and analytical results converge for sufficient number of realizations.

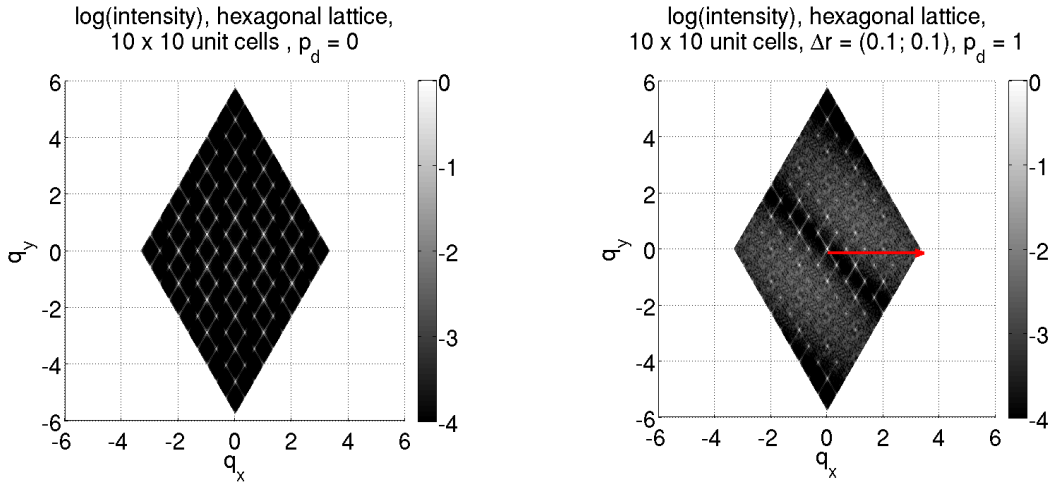

Fig. 4. Logarithm of intensity in 2D q-space for different probabilities of displacement: p_d = 0 (left), p_d = 1 (right)

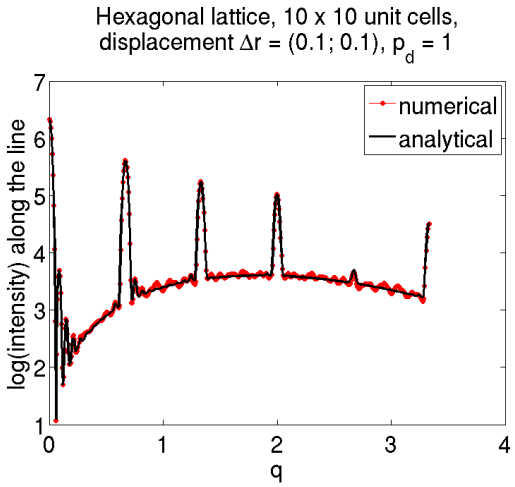

Fig. 5. Logarithm of the intensity along the line in fig. 4 right. The figure compares the analytical and numerical results.

IV. Monoatomic crystals with uncorrelated defects

After coping with two special cases of uncorrelated defects, one can ask whether there is any general formula concerning the relationship between the intensity of the damaged and undamaged crystal. Indeed, it can be proven, that for any kind of the uncorrelated defects occurring in a monoatomic crystal:

$$\left\langle I(\vec{q})\right\rangle_R = S(\vec{q}, X)\cdot I_{ideal}(\vec{q}) + B(\vec{q}, X), \tag{10}$$

where $S(\vec{q}, X)$ is a scaling factor and $B(\vec{q}, X)$ is a background term, both dependent on $\vec{q}$ vector and X, which depend on the defect statistics.

Now we can proceed to answering the question proposed in the title of this paper. Once we know the defect statistics of the uncorrelated defects in a monoatomic crystal, with the formula (10) it is always possible to recover the intensity of the undamaged crystal from the intensity of the damaged crystal. This becomes non-trivial, if the crystal is not monoatomic and even worse, if the defects are correlated.

The numerical tool we have written can perform analysis of defects for any 2D crystal geometry and can be easily extended to other kinds of defects as well as to 3D lattices.

V. Conclusions

The aim of this paper was to study the possibilities of recovering the diffraction image of an undamaged crystal from the diffraction image of the damaged crystal. We have performed the calculations of the scattered intensity for two special cases of defects on two types of 2D lattices – square and hexagonal. The first type of defect were vacancies and the second type were displacements of atoms from their equilibrium positions. Both cases were calculated analytically and numerically. Moreover, the general formula for intensity for any kind of the uncorrelated defects was derived as a scaling factor multiplying the intensity of the undamaged crystal with the addition of a background term. Using this formula one can recover the intensity of the undamaged crystal, if we know the underlying defect statistics. This formula can be a useful tool to handle the damage, which happens to a crystal during a diffraction process. On the other hand, its applicability is limited, since it is valid just for the case of monoatomic crystals with the uncorrelated defects. Therefore, the challenge for the future is how to deal also with the correlated defects.

Literature

[1] BIRKHOLZ, M. 2006. Thin Film Analysis by X-Ray Scattering. WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim, ISBN: 3-527-31052-5.
[2] ASHCROFT, N.W., MERMIN, N.D. 1976. Solid state Physics. Saunders College, Michigan university, ISBN: 9780030493461.