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

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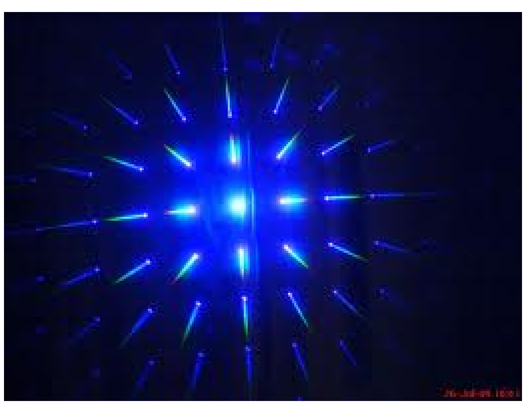
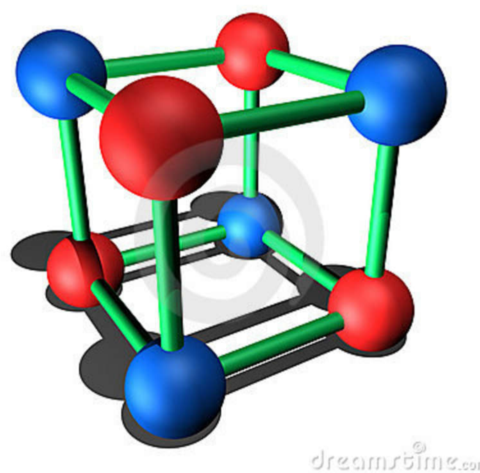
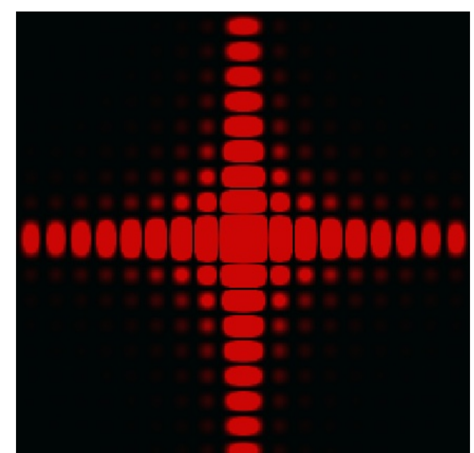






Diffraction imaging

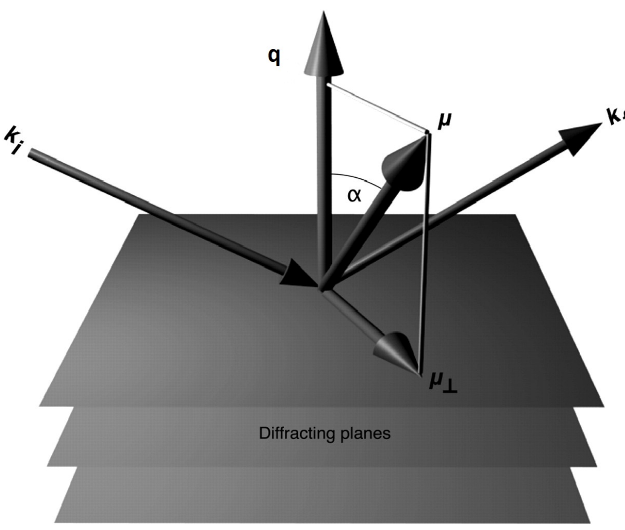
- The amplitude of the scattered wave can be calculated as:

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

where *f* is the atomic form factor defined as:

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

where $\rho_{at}(\vec{r}\,')$ is the atomic electron density



$$\boldsymbol{q} = \boldsymbol{k_f} - \boldsymbol{k_i}$$

- We then have an access to the intensity through:

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

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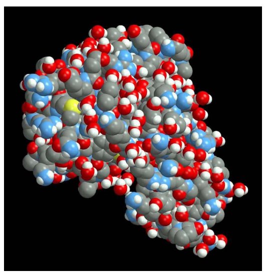
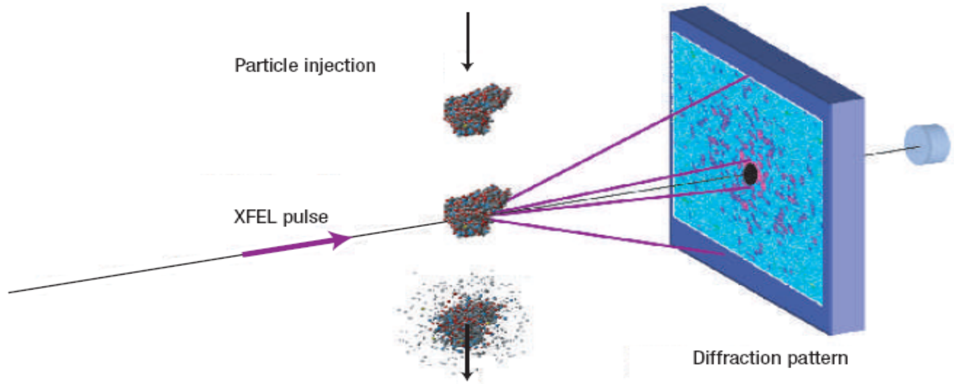
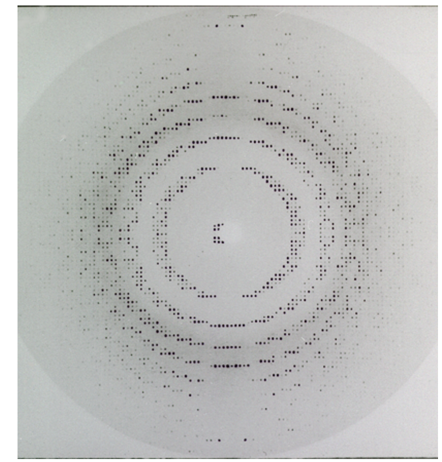
Structure determination through diffraction imaging?

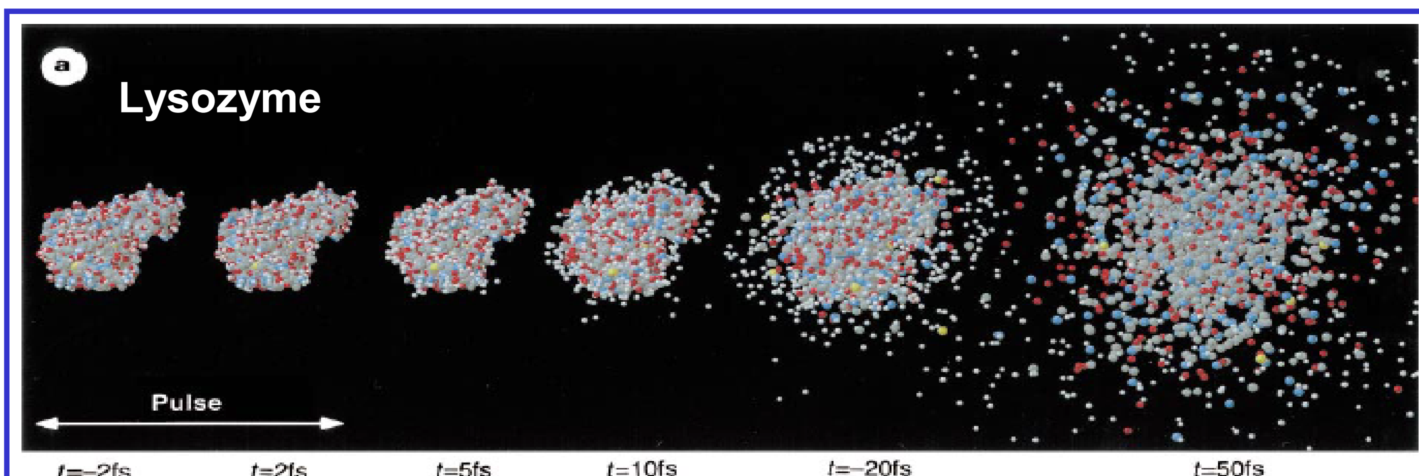
Molecules at atomic resolution





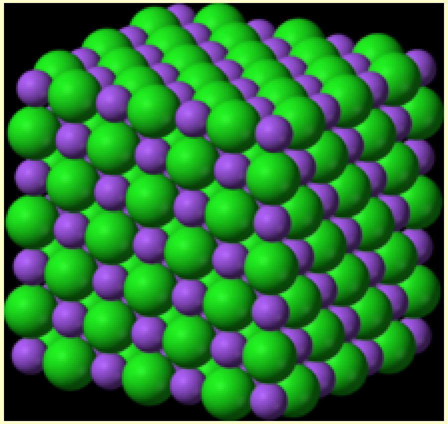
Crystal





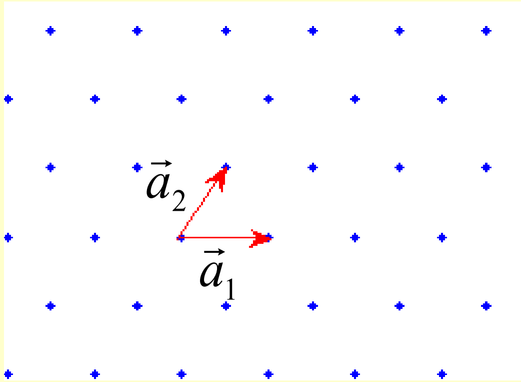
[R. Neutze, R. Wouts, D. van der Spoel, E. Weckert, J. Hajdu: Nature 406, 752 (2000)
Radiation damage and Coulomb explosion]

Defects in crystals

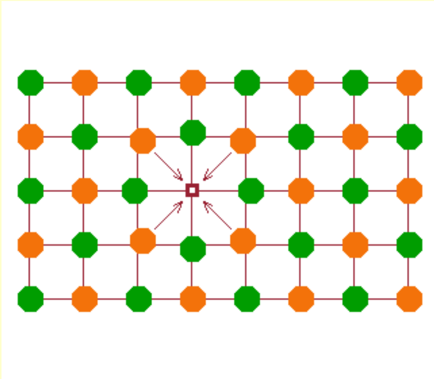


- Crystal – a solid with a spatial periodicity

- Lattice – summarizes the geometry of the underlying periodic structure in crystal $\vec{R} = n_1\vec{a}_1 + n_2\vec{a}_2$ in 2D.



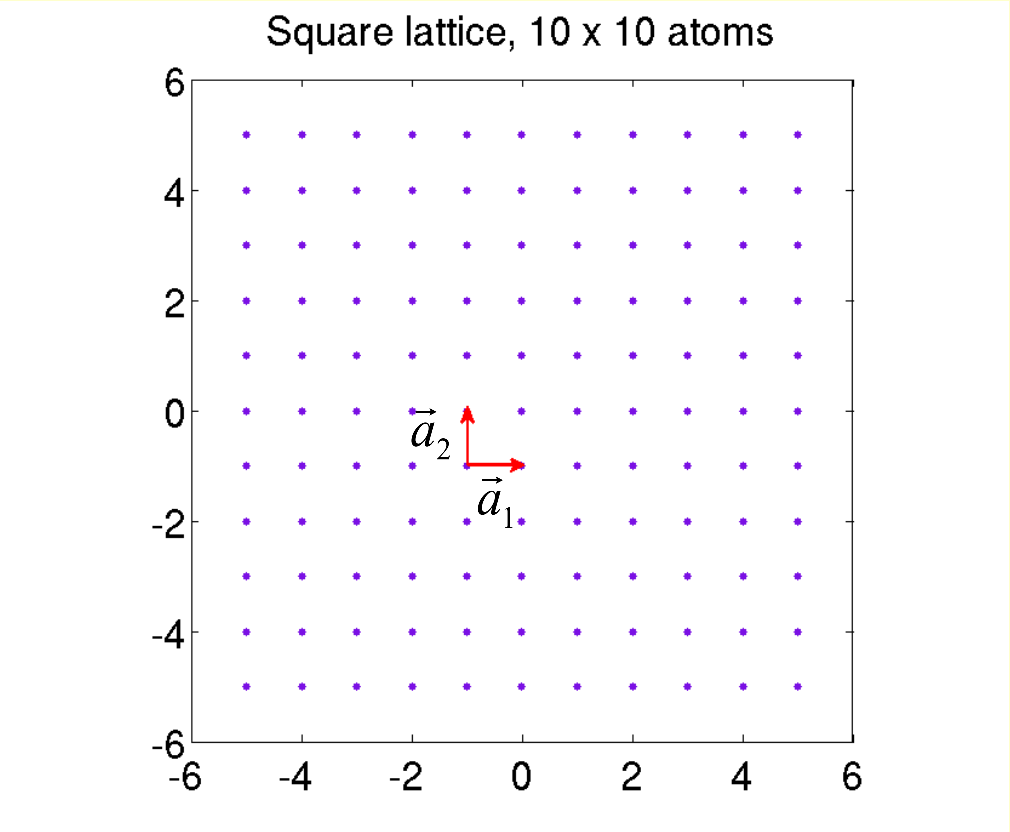
- Defect – any region in crystal, where the microscopic arrangement differs from that of a perfect crystal

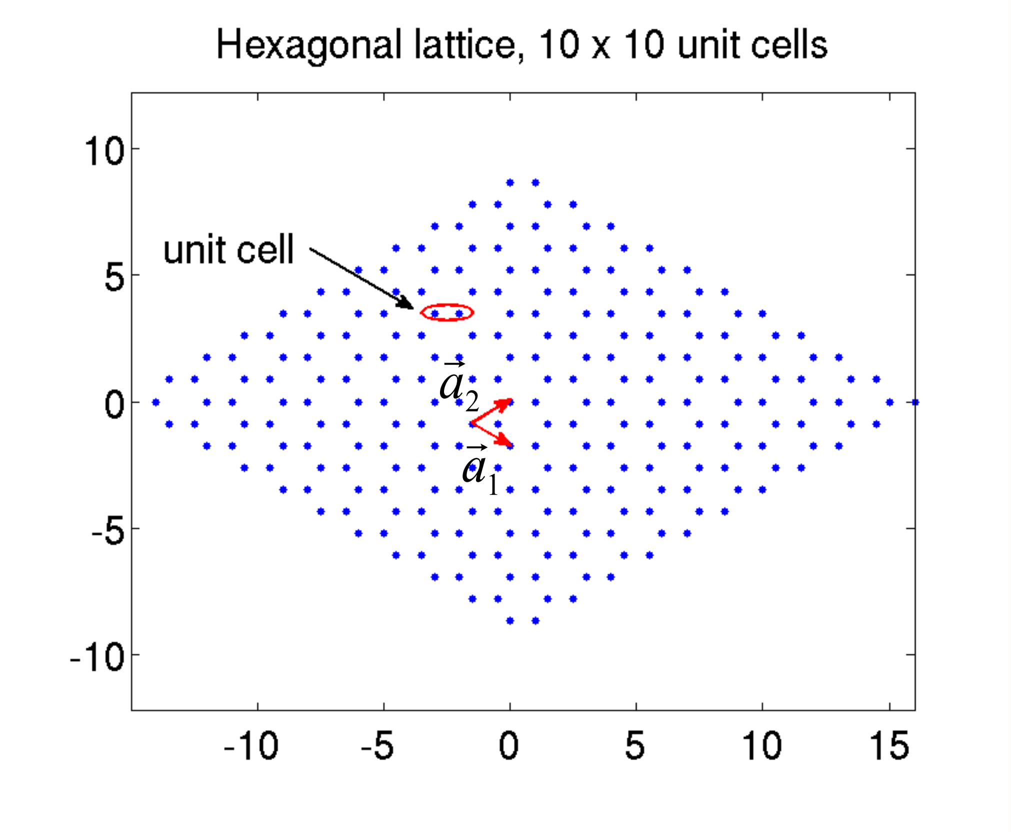


- I_{ideal} - the intensity obtained from an undamaged crystal

- $\langle I \rangle_R$ – the average intensity obtained from a crystal with some random defects

- Two types of 2D lattices were used





- The interatomic distance was chosen as unit
- Atomic form factor of neutral carbon was used

Imaging of monoatomic crystals with vacancies

- The crystal without defects

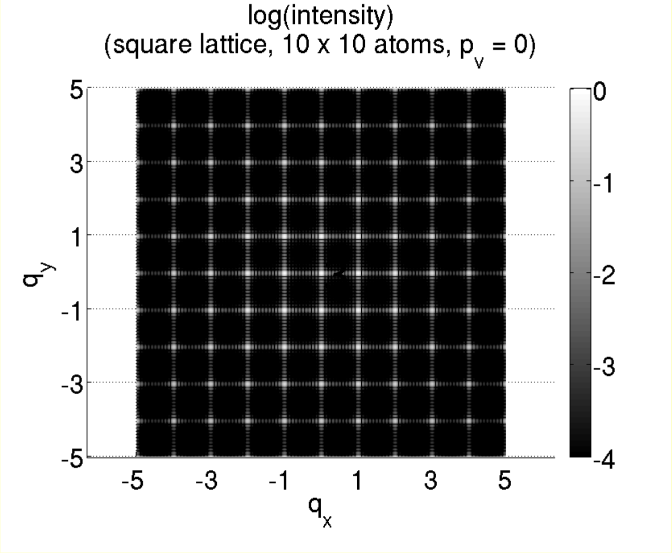
$$A(\vec{q}) = \sum_{j=1}^{N} f_j(\vec{q})\, e^{i\vec{q}\vec{r}_j} \qquad I_{ideal}(\vec{q}) \propto \left|A(\vec{q})\right|^2$$

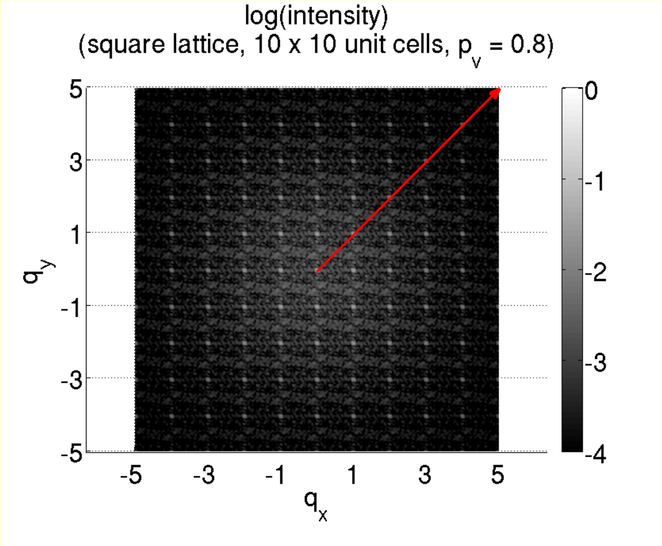
- The form factor is a random variable

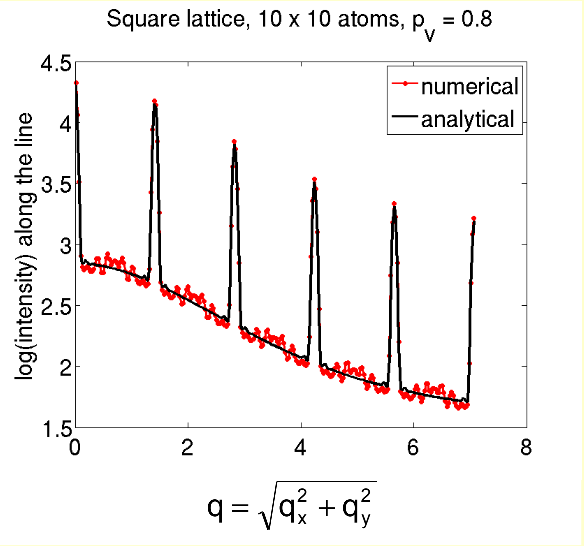
$$f_j(\vec{q}) = \begin{cases} 0, & p_v \\ f(\vec{q}), & 1-p_v \end{cases}$$

- This will result in

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







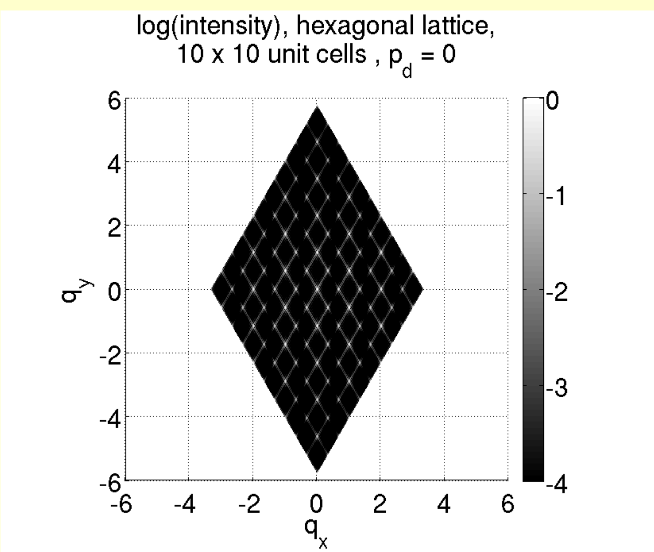
Imaging of monoatomic crystals with displacements

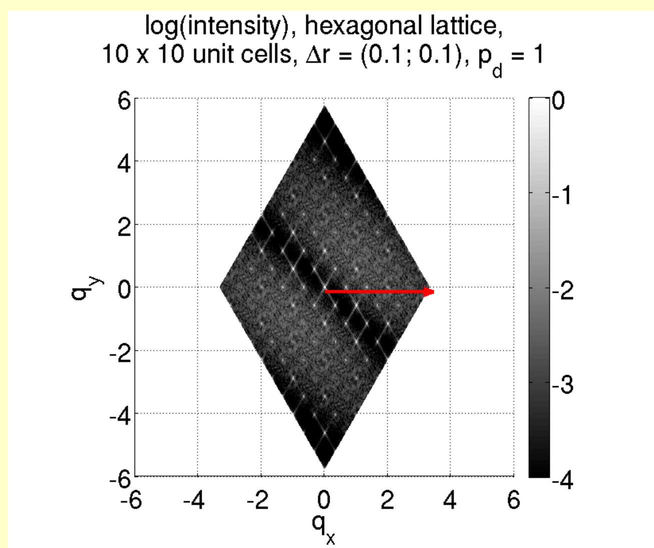
- The atom position is a random variable (can be displaced by $\Delta\vec{r}$)

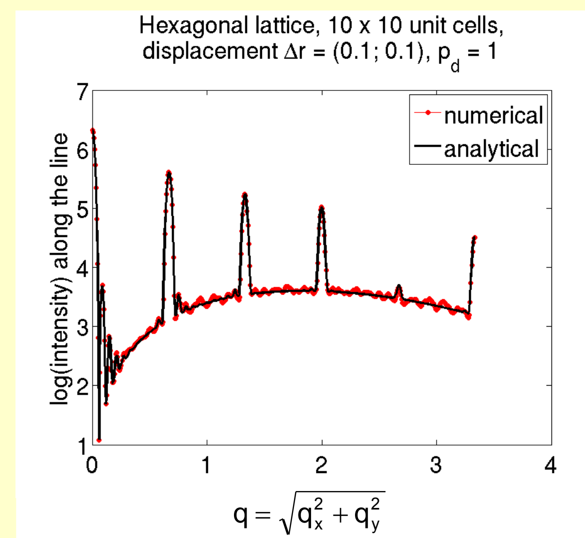
$$\vec{r}_i = \begin{cases} \vec{r}_{i0} + \Delta\vec{r}, & \dfrac{p_d}{2} \\ \vec{r}_{i0}, & 1 - p_d \\ \vec{r}_{i0} - \Delta\vec{r}, & \dfrac{p_d}{2} \end{cases}$$

- This will result in

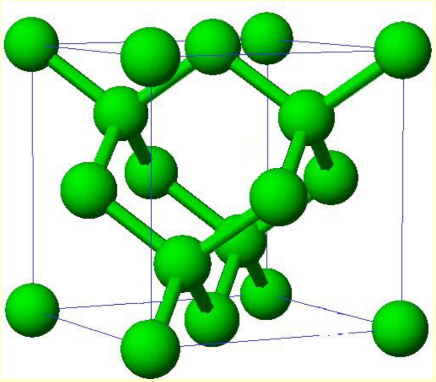
$$\left\langle I(\vec{q}) \right\rangle_R = \left[1 - p_d + p_d \cos(\vec{q}\Delta\vec{r})\right]^2 I_{ideal}(\vec{q}) + N\left|f(\vec{q})\right|^2 \left\{1 - \left[1 - p_d + p_d \cos(\vec{q}\Delta\vec{r})\right]^2\right\}$$







Monoatomic crystals with uncorrelated defects



If these conditions are satisfied:

- We deal with monoatomic crystals
- The defects are uncorrelated

then

- It can be proven that it always leads to

$$\left\langle I(\vec{q}) \right\rangle_R = \underbrace{S(\vec{q}, X)}_{\text{Scaling factor}} \cdot I_{ideal}(\vec{q}) + \underbrace{B(\vec{q}, X)}_{\text{Background}}$$

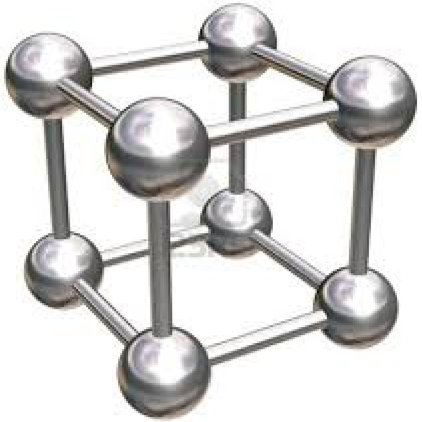
where parameters *X* depend on the defect statistics

Discussion

- If one knows the statistical distribution of defects, one can extract I_{ideal} from the average recorded intensity $\langle I \rangle_R$ for any kind of uncorrelated defects in monoatomic crystals.

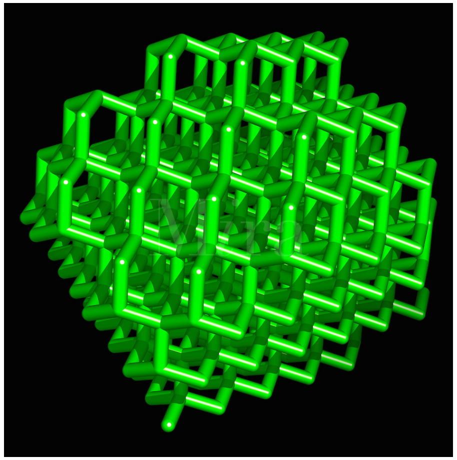
- This becomes non-trivial, if crystal is not monoatomic. Even worse, if defects are correlated.

- Our numerical tool can perform analysis of defects for any 2D crystal geometry. Extension to 3D is straightforward.



Summary

- We have studied the influence of the lattice defects on the diffraction image
- Two types of uncorrelated defects were considered – vacancies and displacements
- The results obtained for both analytical and numerical approach were in agreement
- The scattering intensity from a damaged crystal allows us to recover the intensity from the undamaged crystal, if we know the underlying defect statistics



Thank you!

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- Especially to my supervisors

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- Thank you for your attention!