Physics with neutrons 2

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Exercise sheet 4
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EXERCISE 4.1

A 2-dimensional hexagonal lattice with lattice constant a is given in the normal space. Draw the corresponding lattice in reciprocal space. How are the reciprocal lattice vectors determined? What does the first Brillouin zone look like?

Solution. The hexagonal lattice is determined by the two unit vectors

$$\vec{a}_1 = (a_{11}, a_{12}) = \frac{a}{2}(1, \sqrt{3}) \quad \text{and} \quad \vec{a}_2 = (a_{21}, a_{22}) = a(1, 0).$$

Between real-space unit vectors $\vec{a}_i$ and reciprocal unit vectors $vecb_j$, the following relation must hold, just as for 3-dimensional lattices:

$$\vec{a}_i \cdot \vec{b}_j = 2\pi\delta_{ij}.$$

From this, we can formulate a system of equations that needs to be satisfied:

$$\left(a_{ij}\right)^T \cdot \left(b_{ij}\right) = 2\pi I,$$

where (a_{ij}) and (b_{ij}) are the matrices formed by the components of the base vectors, and I is the identity matrix in two dimensions. Writing $A = (a_{ij})^T$, we can solve this system by multiplying with A^{-1}:

$$(b_{ij}) = 2\pi A^{-1},$$

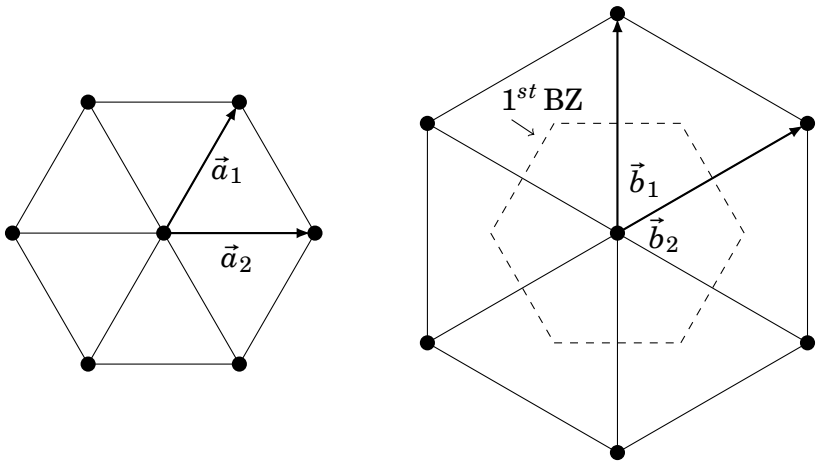
and A^{-1} is easily found as

$$A^{-1} = \frac{1}{\det A}\begin{pmatrix} a_{22} & -a_{12} \\ -a_{21} & a_{11} \end{pmatrix}.$$

This means that we have found our reciprocal unit vectors:

$$\vec{b}_1 = \frac{4\pi}{a\sqrt{3}}(0, 1) \quad \text{and} \quad \vec{b}_2 = \frac{2\pi}{a\sqrt{3}}(\sqrt{3}, -1),$$

which again form a hexagonal lattice. The first Brillouin zone therefore is a regular hexagon. The following sketch shows real and reciprocal unit cells:



□

EXERCISE 4.2

Diffraction of neutrons from argon monolayers absorbed on graphite basal planes indicated that an ordered, two dimensional triangular argon lattice is formed at low temperatures [Taub et. al. (1977)], see Fig. 1. There are two possible configurations for the argon monolayers, either commensurate with the graphite lattice or incommensurate corresponding to the closest packing.
(a) Explain the asymmetric sawtooth profile of the Bragg peaks in Fig. 1.
(b) Determine from the observed reflections displayed in Fig. 1 whether the Ar monolayers are commensurate or incommensurate with the graphite lattice. The C atoms have a nearest neighbor distance $a_C = 2.46$ Å in the hexagonal plane of graphite.

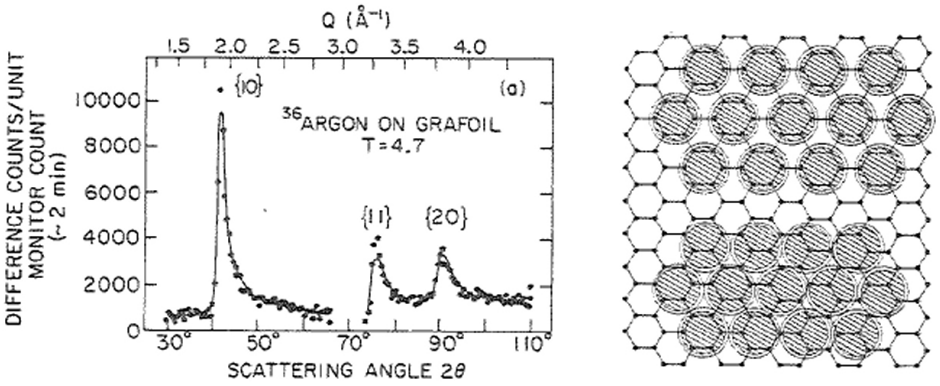


Figure 1: (left) Diffraction pattern from a two-dimensional Ar monolayer absorbed on graphite showing the Bragg reflections (1,0), (1,1) and (2,0). (right) Schematic representation of a commensurate (top) and incommensurate (bottom) Ar monolayer phase.

Solution. 1. The reciprocal lattice of a two-dimensional crystal consists of an ordered array of rods aligned normal to the scattering plane. In diffraction experiments there is for

each Miller index pair (h,k) a minimum value of the scattering angle $2\theta = 2\theta_B$ where θ_B corresponds to the Bragg angle

$$\lambda = 2d\sin\theta_B. \tag{1}$$

Depending on the size of the array, there will be a continuous distribution of diffracted intensity for scattering angles greater than $2\theta_B$. This produces a characteristic *sawtooth* line shape with a sharply rising leading edge on the low-angle side, followed by a trailing edge extending to larger scattering angles. The maximum of the diffracted intensity I_{hk} occurs at the scattering angle $2\theta_B$. For a detailed calculation of the line shape we refer to the basic article by Warren [Warren (1941)]:

$$I_{hk} = \frac{m_{hk}|S_{hk}|^2 L(\theta)}{\sin\theta\sqrt{\sin^2\theta - \sin^2\theta_B}},$$

where m_{hk} denotes the multiplicity of the Bragg reflection (h,k,l), $L(\theta)$ is the Lorentz factor.

2. Following EXERCISE 5.2, the reciprocal lattice vectors of a two-dimensional heagonal lattice are given

$$\tau_1 = \frac{2\pi a}{f_0}(\sqrt{3}/2, 1/2), \tau_2 = \frac{2\pi a}{f_0}(0,1), f_0 = \frac{\sqrt{3}}{2}a^2,$$

where a is the nearest-neighbor distance. By identifying the peaks observed at $Q = 1.9, 3.3$ and 3.8 Å^{-1} as the $(1,0),(1,1)$ and $(2,0)$ Bragg reflections, we find from Eq. (1) $a_{\mathrm{Ar}} = 3.82$ Å. This is significantly smaller than the distance $\sqrt{3}a_C = 4.26$ Åfor the commensurate phase sketched in Fig 1, thus the Ar monolayer is clearly incommensurate with that of the underlying graphite basal plane. $a_{\mathrm{Ar}} = 3.82$ Åcorresponds to the minimum of the Lennard-Jones potential $\Phi(r) = -Ar^{-6} + Br^{-12}$ known for solid Ar. We conclude that the structure of the Ar monolayer is primarily determined by the couplings between the Ar atoms, and interactions with the substrate appear to play a minor role.

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EXERCISE 4.3

Prove the lattice sum equation:

$$\sum_{\mathbf{v}_{mnp}} \exp(i\mathbf{Q}\cdot\mathbf{v}_{mnp}) = \frac{(2\pi)^3}{V_{UC}} \sum_{\mathbf{G}_{hkl}} \delta(\mathbf{Q} - \mathbf{G}_{hkl})$$

Solution. Let $\mathbf{v} = m\mathbf{a}_1 + n\mathbf{a}_2 + p\mathbf{a}_3$ be a lattice vector in real space and $\mathbf{G} = h\mathbf{a}_1^* + k\mathbf{a}_2^* + l\mathbf{a}_3^*$ a reciprocal lattice vector, with m,n,p,h,k,l integer. Also let $\mathbf{Q} = q\mathbf{a}_1^* + r\mathbf{a}_2^* + s\mathbf{a}_3^*$ be an arbitrary scattering vector.
Now we can write the left hand side of the equation as

$$\sum_{m,n,p} \exp(i\mathbf{Q}\cdot\mathbf{v}) = \sum_{m,n,p} \exp(2\pi i(qm + rn + sp))$$

(using $\mathbf{a}_i^* \cdot \mathbf{a}_j = 2\pi\delta_{ij}$, the defining relation of the reciprocal lattice vectors).

This sum can further be separated in three parts:

$$\left(\sum_m e^{2\pi iqm}\right)\left(\sum_n e^{2\pi irn}\right)\left(\sum_p e^{2\pi isp}\right),$$

of which we now only look at the first. Assume that the crystal has N (without loss of generality we set N odd) unit cells in the direction of $\mathbf{a}_1$. Then the sum becomes

$$\sum_{m=-(N-1)/2}^{(N-1)/2} e^{2\pi iqm} = \sum_{m=0}^{(N-1)/2} e^{2\pi iqm} + \sum_{m=0}^{(N-1)/2} e^{-2\pi iqm} - 1 = (\ldots) = \frac{\sin N\pi q}{\sin \pi q}.$$

For large N, this expression effectively becomes a sum of delta functions at points where q is integer. Applying this to all three dimensions, we get that $\mathbf{Q} = \mathbf{G}_{hkl}$, i.e.

$$\sum_{\mathbf{v}_{mnp}} = C \cdot \sum_{\mathbf{G}_{hkl}} \delta(\mathbf{Q} - \mathbf{G}_{hkl})$$

with some constant C, which can be determined by integrating both sides over the unit cell:

$$\int_{cell} d^3q \sum_{\mathbf{v}_{mnp}} = \int_{cell} d^3q\, C \cdot \sum_{\mathbf{G}_{hkl}} \delta(\mathbf{Q} - \mathbf{G}_{hkl}) = C.$$

To evaluate the left-hand side, we use

$$\int_{cell} d^3q\, e^{i\mathbf{q}(\mathbf{v}-\mathbf{v}')} = \frac{(2\pi)^3}{V_{UC}} \delta_{\mathbf{v}\mathbf{v}'}$$

and get

$$C = \frac{(2\pi)^3}{V_{UC}} \sum_{\mathbf{v}} \delta_{\mathbf{v}\mathbf{v}'} = \frac{(2\pi)^3}{V_{UC}}.$$

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