Diffraction of X-rays by Structured and Evolving Targets (10 points)

Note
1. Vectors are denoted by bold symbols (e.g., $\mathbf{r}$, $\mathbf{q}$).
2. Assume that absorption is neglected and that the electric field is polarized perpendicular to the plane of incidence.

X-ray diffraction patterns arise because many tiny "wave sources" within a crystal interfere, and we can predict this by adding their complex amplitudes with the correct phases. Consider a monochromatic wave characterized by a (real) amplitude $A \geq 0$ and a phase ϕ. We define the **complex amplitude** $\tilde{A}$ as

$$\tilde{A} = Ae^{i\phi},$$

so that the (real) amplitude of the wave is the magnitude (absolute value) of $\tilde{A}$, and ϕ is its phase. Thus $\tilde{A}$ conveniently encodes both magnitude and phase in a single complex number. In this problem set, we define a dimensionless intensity by the squared magnitude of the total complex amplitude:

$$I = |\tilde{A}|^2 = A^2.$$

Common experimental and geometric proportionality factors, such as detector response, incident-beam normalization, and common propagation factors, are absorbed into this definition. By contrast, the single-electron scattering amplitude f_0 is retained explicitly as the amplitude scale for scattering from one point electron.

When a crystalline material is exposed to an incident wave, the wave is diffracted by the crystal lattice and the diffracted parts interfere with one another. The intensity of the resulting diffracted wave can be calculated by adding the complex amplitudes of the individual diffracted waves, taking into account the phase differences between them, and then computing the squared magnitude of the resulting total complex amplitude. Diffraction primarily arises from interactions with electrons, and contributions from heavier particles such as nuclei are typically negligible. The amplitude of the wave diffracted by a single point electron depends only on $R = |\mathbf{R}|$, the distance from the electron to the detector. Since R is much larger than the dimensions of the sample, its variation across the sample can be neglected. Therefore, the total diffracted wave can be accurately determined by properly accounting for the phase differences between individual diffracted waves, while their amplitudes are assumed to be constant.

Let $\mathbf{k}_i$ and $\mathbf{k}_f$ denote the wavevectors of the incident and diffracted waves, respectively. An incident plane wave with wavevector $\mathbf{k}_i$ is diffracted into a wave with wavevector $\mathbf{k}_f$. The **momentum transfer** is defined as

$$\mathbf{q} = \mathbf{k}_f - \mathbf{k}_i$$

and their magnitudes are assumed to be equal, because the wavelength is unchanged:

$$k \equiv |\mathbf{k}_i| = |\mathbf{k}_f| = \frac{2\pi}{\lambda}.$$

Part A: Diffraction from two electrons treated as point particles (2.0 pts)

Consider two point electrons located at positions $\mathbf{r}_1$ and $\mathbf{r}_2$, and define $\mathbf{r} \equiv \mathbf{r}_2 - \mathbf{r}_1$. A detector is located at $\mathbf{P}$, and we define $\mathbf{R} \equiv \mathbf{P} - \mathbf{r}_1$. A plane wave $E_i(\mathbf{r}) \propto e^{i\mathbf{k}_i \cdot \mathbf{r}}$ is incident on the two electrons, and the diffracted wave is observed in the far field along the direction $\mathbf{k}_f$. In part **A**, the common time-dependent factor $e^{-i\omega t}$ is suppressed, since only relative spatial phases are relevant.

A.1 Under the far-field approximation, $R \equiv |\mathbf{R}| \gg |\mathbf{r}|$, keep only the leading-order term in $|\mathbf{r}|/R$ and write the outgoing geometric path difference, $\Delta L_{\text{out}} \equiv |\mathbf{P} - \mathbf{r}_1| - |\mathbf{P} - \mathbf{r}_2|$, in terms of $\mathbf{r}$ and $\mathbf{k}_f$, or equivalently $\mathbf{k}_f/k_f$. 0.6 pt

A.2 Using your result from A.1 and accounting for the position-dependent phase of the incident wave at $\mathbf{r}_1$ and $\mathbf{r}_2$, find the phase difference between the two diffracted

contributions at the detector, expressed in terms of $\mathbf{q}$ and $\mathbf{r}$. The phase difference is defined as $\Delta\phi \equiv \phi_1 - \phi_2$, where ϕ_1 and ϕ_2 are the phases of the contributions from the electrons at r_1 and r_2. 0.4 pt

A.3 Express the total complex amplitude of the diffracted wave from these two electrons in terms of $\mathbf{q}$ and $\mathbf{r}$. You may ignore any overall common phase factor, since it does not affect the intensity. Assume that the real amplitude of a diffracted wave from a single point electron is a constant f_0, independent of position. 0.6 pt

A.4 Express the intensity of diffracted waves from those two electrons in terms of $\mathbf{q}$ and $\mathbf{r}$. 0.4 pt

Part B: Finite longitudinal coherence (phase-jump model) (2.3 pts)

Consider two point-like electrons located at positions $\mathbf{r}_1$ and $\mathbf{r}_2$, with $\mathbf{r} \equiv \mathbf{r}_2 - \mathbf{r}_1$. A beam of wavelength λ_0 (so $k = 2\pi/\lambda_0$ and $\omega = 2\pi c/\lambda_0$) illuminates the electrons, and the diffracted wave is observed in the far field along $\mathbf{k}_f$. We model the incident field as a plane wave with a time-dependent random phase,

$$E_i(\mathbf{r}, t) = A \ \exp[i\,(\mathbf{k}_i \cdot \mathbf{r} - \omega t + \phi(t))]\,,$$

where $\phi(t)$ is *piecewise constant* and undergoes *random phase jumps* at regular time intervals of duration

$$t_0 \equiv \frac{L_0}{c},$$

with L_0 the (given) longitudinal coherence length. At the beginning of each interval of length t_0, $\phi(t)$ is reset to a new independent value uniformly distributed on $[0, 2\pi)$. Let I_0 denote the (time-averaged) intensity at the detector that would be obtained from a *single* electron in the same geometry. In this problem, the detector is assumed to measure a time-averaged intensity. That is, it does not resolve the individual random phase jumps. Instead, it records the average of the instantaneous intensity over a time much longer than the phase-jump interval t_0: $\langle I\rangle_t \equiv \left\langle |E(t)|^2 \right\rangle_t$. Here $\langle\cdots\rangle_t$ denotes an average over many phase-jump intervals.

B.1 Using the phase-jump model above, derive the *time-averaged* total intensity $\langle I\rangle_t$ at the detector from the two electrons. Your final result should be written in terms of I_0, $\mathbf{q} \equiv \mathbf{k}_f - \mathbf{k}_i$, the separation vector $\mathbf{r}$, the wavelength λ_0, and the coherence length L_0. Assume the detector averages over times much longer than t_0. 2.3 pt

Part C: Non-point particle effect (1.0 pts)

An electron is often treated as a classical point particle, but in a more realistic model its charge may be regarded as being distributed over a finite spatial region. Consider two idealized charge distributions: (i) an ideal point charge located at $\mathbf{r} = 0$, whose scattering amplitude is $A_1(\mathbf{q}) = Q_0$, (ii) an extended Gaussian charge distribution $\rho_2(\mathbf{r}) = \rho_0 \exp\left(-\frac{r^2}{R_0^2}\right), \qquad r = |\mathbf{r}|.$

The constants Q_0 and ρ_0 are chosen so that the total charge is the same in both cases:

$$Q_0 = \int \rho_2(\mathbf{r})\, d^3r.$$

Here d^3r denotes the volume element in three-dimensional space. In Cartesian coordinates, $d^3r = dxdydz$ and $\int_{\mathbb{R}^3}$ means integration over all space. Useful identities (may be used without proof):

$$\int_0^\infty e^{-r^2/R_0^2}\, 4\pi r^2\, dr = \pi^{3/2} R_0^3, \int_{\mathbb{R}^3} e^{-\alpha r^2} e^{i\mathbf{k}\cdot\mathbf{r}}\, d^3\mathbf{r} = \left(\frac{\pi}{\alpha}\right)^{3/2} \exp\left(-\frac{k^2}{4\alpha}\right), \qquad \alpha > 0.$$

C.1 Obtain the relation among Q_0, ρ_0, and R_0. 0.4 pt

C.2 Evaluate the amplitude 0.4 pt

$$A_2(\mathbf{q}) \equiv \int_{\mathbb{R}^3} \rho_2(\mathbf{r}) e^{i\mathbf{q}\cdot\mathbf{r}}\, d^3\mathbf{r}$$

and compare it with the point-charge amplitude $A_1(\mathbf{q}) = Q_0$.

C.3 Estimate the ratio of the diffracted intensities, $\dfrac{I_2}{I_1}$, for these two idealized cases when $q = \dfrac{2}{R_0}$. 0.2 pt

Part D: Diffraction from a film with non-flat surface morphology (2.4 pts)

Imagine that the surface of a film (i.e., the top atomic layers) is not perfectly flat, but exhibits surface roughness. A common model is to assume that the local film thickness (measured in monolayers) follows a Gaussian distribution. Let N denote the local number of completed monolayers within a lateral coherence area of the beam. We assume that N varies across the surface and is normally distributed with mean $\bar{N}$ and standard deviation σ (both in units of monolayers):

$$P(N) = \frac{1}{\sqrt{2\pi}\sigma} \exp\left[-\frac{(N-\bar{N})^2}{2\sigma^2}\right].$$

(For the purpose of evaluating averages, you may treat N as a continuous variable.) Let d be the spacing between adjacent atomic layers (monolayers), and let q_z denote the component of the scattering vector $\mathbf{q} = \mathbf{k}_f - \mathbf{k}_i$ normal to the flat surface of the film, i.e., $q_z = \mathbf{q} \cdot \hat{\mathbf{z}}$. For an integer number of monolayers N, the scattering amplitude is

$$A_N(q_z) = \sum_{n=0}^{N-1} e^{iq_z nd} = \frac{1 - e^{iq_z Nd}}{1 - e^{iq_z d}}.$$

When averaging over the Gaussian thickness distribution, we treat N as a continuous variable and use the closed-form expression

$$A_N(q_z) \equiv \frac{1 - e^{iq_z Nd}}{1 - e^{iq_z d}}$$

as the corresponding continuous extension. Assume the measured diffraction intensity is obtained from the *coherent* average amplitude,

$$I(q_z) \equiv \left|\langle A(q_z)\rangle\right|^2, \qquad \langle A(q_z)\rangle = \int_{-\infty}^{\infty} P(N)\, A_N(q_z)\, dN.$$

D.1 Calculate the intensity ratios 2.4 pt

$$\frac{I(q_z, \sigma = 0.4, \bar{N} = 5)}{I(q_z, \sigma = 0, \bar{N} = 5)}$$

at

$$q_z = \frac{\pi}{2d} \qquad \text{and} \qquad q_z = \frac{2\pi}{d},$$

respectively. If needed, evaluate the second case by taking the appropriate limit.

Part E: Diffraction from a film with evolving surface morphology (2.3 pts)

Imagine a thin film with a simple cubic structure grown on a substrate in a layer-by-layer mode, *i.e.*, each monolayer is completed before the next monolayer starts to grow. Let d be the spacing between adjacent atomic layers (monolayers), and let q_z denote the component of $\mathbf{q} = \mathbf{k}_f - \mathbf{k}_i$ normal to the flat surface of the film, *i.e.,* $q_z = \mathbf{q} \cdot \hat{\mathbf{z}}$. As the film is being grown, the thickness of the film changes, and so does the diffraction intensity at $\mathbf{q} = \frac{\pi}{d}\hat{\mathbf{z}}$. The calculation is performed at the specified out-of-plane momentum transfer, and all monolayer contributions are coherently summed.

E.1 If the film starts to grow at $t = 0$ and the time required to complete one monolayer is t_0, obtain the diffraction intensity ratio 2.3 pt

$$\frac{I(t = 0.8t_0)}{I(t = 3.6t_0)}$$

measured at $\mathbf{q} = \frac{\pi}{d}\hat{\mathbf{z}}$. Assume that during each monolayer-growth interval, the fractional coverage of the top layer increases linearly from 0 to 1, so that at time $t = (N + \theta)t_0$, there are N completed monolayers and a fractional coverage θ of the next monolayer.