Appendix A

Differential magnetic-scattering cross section

Assuming a crystal with a one-atomic basis, which is described by the momentum and photon-energy dependent atomic form factor $f(\underline{q}, h\nu)$, the differential scattering cross section can be written as [67]

$$\frac{d\sigma}{d\Omega} = r_0^2 P^2 \left| f\left(\underline{q}, h\nu\right)\right|^2 \left| F\left(\underline{q}\right)\right|^2 = \frac{d\sigma_a}{d\Omega}\left| F\left(\underline{q}\right)\right|^2, \tag{A.1}$$

with the classical electron radius r_0 and the polarization dependence P. $d\sigma_a/d\Omega = r_0^2 P^2 |f(\underline{q}, h\nu)|^2$ is the atomic differential scattering cross section and $F(\underline{q})$ is the structure factor of, e.g. the lattice or as in the present case of the magnetic structure. Unlike for charge scattering, in magnetic scattering $d\sigma_a/d\Omega$ depends on the orientation of the polarization vectors of incident and scattered x-rays with respect to the magnetic-moment direction (equation 2.9), therefore the differential scattering cross section reflects the individual magnetic structure. The atomic quantity, which is independent of the special magnetic structure, is the atomic form factor $f(\underline{q}, h\nu)$.

For a crystalline solid, equation A.1 is only useful to some extent since an infinitesimal small detector size cannot be realized and since the intensity distribution of a Bragg reflection into a solid angle, and thus the intensity scattered into a small detector depends on the crystal size and quality and also on the collimation of the incident x-ray beam. More practical, one can calculate the differential scattering cross section from the total *integrated* intensity of a Bragg reflection $I^{integr.}$, where the integration covers the volume in momentum space, where the signal is not zero. Then the differential scattering cross section is written as

$$\frac{d\sigma}{d\Omega} = \frac{d\sigma_a}{d\Omega}\int_{Bragg}\left| F\left(\underline{q}\right)\right|^2 d\Omega = \frac{I^{integr.}}{\Phi_0},$$

where Φ_0 is the incident photon flux. The atomic form factor $f(\underline{q}, h\nu)$ is assumed to be constant, which is acceptable for diffraction peaks across which $\underline{q}$ does not vary much. The atomic differential scattering cross section can now be calculated from the experimental data by

$$\frac{d\sigma_a}{d\Omega} = \frac{I^{integr.}}{\Phi_0} (NL)^{-1} ,$$

where N is the number of scattering particles in the interaction volume V that $N = \rho V$ with the atomic density ρ. L is the Lorentz factor of the corresponding Bragg reflection arising from the integration over the structure factor

$$\int_{Bragg} \left| F\left(\underline{q}\right) \right|^2 d\Omega = NL. \tag{A.2}$$

When the photon mean-free path is longer than the sample thickness, the interaction volume is determined by the thickness of the sample d, the area A of the incident x-ray beam profile, and the scattering angle of the Bragg reflection $\widetilde{\Theta}_{Bragg}$ in specular geometry, measured with respect to the sample surface: $V = d \cdot A / \sin(\widetilde{\Theta}_{Bragg})$. The photon flux can be expressed by the intensity of the direct x-ray beam I_0 as $\Phi_0 = I_0/A$. Thus, the scattering cross section can be written as

$$\frac{d\sigma_a}{d\Omega} = \frac{I^{integr.}}{I_0} \frac{\sin\left(\widetilde{\Theta}_{Bragg}\right)}{\rho \cdot d \cdot L}. \tag{A.3}$$

While the film thickness d is precisely known from the scattering experiment and the atomic density ρ from literature, the remaining problems are a proper integration of the scattered intensity distribution and a proper determination of the Lorentz factor. Both problems are closely related to each other and will be discussed in the following.

Lorentz factor

Unlike the scattering from an electron or an atom, the intensity scattered from a crystalline solid is not a two-dimensional (ϑ, φ), but a three-dimensional quantity ($\widetilde{\Theta}$, φ, Θ). In the present case the magnetic structure is modulated along the surface normal leading to an intensity distribution essentially along the specular rod ($\widetilde{\Theta}$ direction), but with finite intensity scattered into the two remaining off-specular directions (φ, Θ). The integration of the experimental data over the smeared-out Bragg-peak contour causes the Lorentz factor L in equation A.3. This factor can be calculated according to equation A.2 for the present magnetic structure and the individual experimental geometry by integrating the corresponding structure factor in exactly the same way as the experimental data have been integrated.

The structure factor of the magnetic helical structure of Ho metal has the form

$$F\left(\underline{q}\right) = \sum_{\underline{R}_n} \exp\left(i\left(\underline{q} - \underline{\tau}\right) \cdot \underline{R}_n\right) ,$$

with the magnetic modulation vector $\underline{\tau} = (0,\, 0,\, \tau)$, and the momentum transfer expressed in terms of scattering angles

$$\underline{q} = \left(\frac{2\pi}{\lambda}\left(\cos\left(2\widetilde{\Theta}_{Bragg} - \Theta\right) - \cos\Theta\right),\ \frac{2\pi}{\lambda}\tan\varphi,\ \frac{4\pi}{\lambda}\sin\widetilde{\Theta}\right) ,$$

where the first two components are the in-plane values of the momentum transfer caused by a variation of the sample rotation around the Bragg-peak position (Θ), and perpendicular to the scattering plane (φ). The third component corresponds to the momentum transfer normal to the surface achieved in specular geometry ($\widetilde{\Theta}$). $\underline{R}_n$ denotes the lattice vector, specifying the location of the atom (or unit cell) within the hexagonal lattice, $\underline{R}_n = (n_1 a + n_2 a/2,\ n_2 a\sqrt{2}/2,\ n_3 c/2)$, with the lattice constants of Ho: a =3.578 Å and c =5.618 Å. Thus, we obtain the structure factor by summation over $N_1 N_2 N_3$ lattice sites:

$$F\left(\Theta,\varphi\right) = \sum_{n_1=0}^{N_1-1}\sum_{n_2=0}^{N_2-1} \ldots,$$

which describes the structure factor within the basal plane of the hexagonal lattice, and

$$F\left(\widetilde{\Theta}\right) = \sum_{n_3=0}^{N_3-1} \exp\left(i \cdot n_3 \frac{c}{2}\left(\frac{4\pi}{\lambda}\sin\widetilde{\Theta} - \tau\right)\right),$$

which corresponds to the form factor of the magnetic helical structure. Thus, equation A.2 can be rewritten for the present case as

$$\int\int \left|F\left(\Theta,\varphi\right)\right|^2 d\Theta d\varphi \cdot \int \left|F\left(\widetilde{\Theta}\right)\right|^2 d\widetilde{\Theta} = NL,$$

which was evaluated numerically to yield the Lorentz factor L.

Integrated intensity

The integration along the off specular direction perpendicular to the scattering plane (φ) is ensured by the large acceptance angle of the detector. A rocking or Θ scan through the Bragg condition leads basically to a variation of the scattering vector parallel to the sample surface to detect the off-specularly scattered x-rays within the scattering plane. Both the Θ and the specular $\widetilde{\Theta}$ scan are in principle independent of each other since they describe linearly independent paths in momentum space. But one has to take into account that a finite detector width already accepts a fraction of the off-specular scattered x-rays in Θ direction along a path in momentum space corresponding to a detector scan (ϑ scan), as shown in figure A.1. The different scans in momentum space and the accepted volume of the Bragg-peak contour are displayed in figure A.1. The integration in Θ direction can be reproduced by calculating the fraction of off-specular scattered intensity, $\Delta_{||}$, which is accepted by the detector size in the present specular geometry. In the present case the Θ scans reveal a Lorentzian-type intensity distribution with a constant line width $\Delta\Theta$ for all $\widetilde{\Theta}$ values of the Bragg-peak contour. This transforms to a width in momentum space of $W_{q,||} = 4\pi/\lambda \cdot \sin(\widetilde{\Theta}_{Bragg}) \cdot \Delta\Theta$, corresponding to the inverse of the magnetic coherence length. The detected off-specular intensity fraction can now be calculated by the integration of the Lorentzian over the acceptance of the detector slit in $q_{||}$ direction normalized to the entire area of the Lorentzian

$$\Delta_{||} = \int_{-\Delta q_{||}/2}^{\Delta q_{||}/2} \frac{W_{q,||}}{4q_{||}^2 + W_{q,||}^2} dq_{||} \Bigg/ \int_{-\infty}^{\infty} \frac{W_{q,||}}{4q_{||}^2 + W_{q,||}^2} dq_{||}\ ,$$

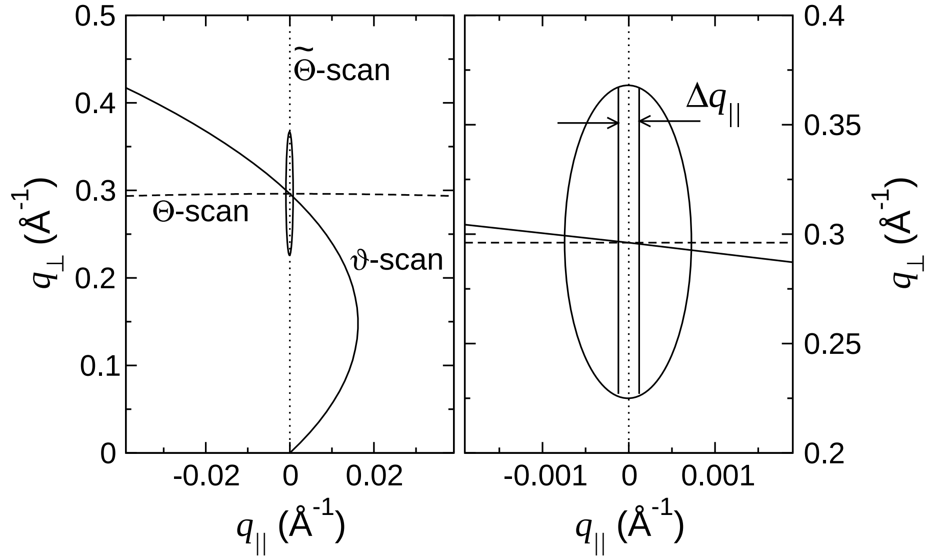

Figure A.1: Scans in momentum space. The left panel shows a Θ scan (dashed), a $\widetilde{\Theta}$ scan in specular geometry (dotted), and a detector scan (ϑ) with the sample rotation being in Bragg-peak position (solid line). While a Θ and a specular $\widetilde{\Theta}$ scan are perpendicular to each other, a detector scan across the Bragg peak describes a linear combination of both. The right panel shows a zoom-in of the left figure. The elliptical contour represents the magnetic $(000{-}\tau)$ superstructure satellite. The stripe in the middle of the contour represents the fraction in momentum space, $\Delta q_{||}$, which is accepted by a $\widetilde{\Theta}$ scan.

with the acceptance in momentum space parallel to the surface corresponding to the angular acceptance of the detector $\Delta\vartheta$: $\Delta q_{||} = 2\pi/\lambda \cdot \sin(\widetilde{\Theta}_{Bragg}) \cdot \Delta\vartheta$. The integrated intensity of the Bragg reflection can then be expressed as $I^{Integr.}_{exp.} = I_{\widetilde{\Theta}} \cdot \Delta_{||}^{-1}$, where $I_{\widetilde{\Theta}}$ is the integrated intensity experimentally obtained from a $\widetilde{\Theta}$ scan.

With the strong photon absorption in the maximum of the Ho M_V absorption threshold, the different atomic layers contribute with different weight to the scattering signal. Absorption is corrected for by simulating the integrated intensity $I = I_0 \cdot \exp(-\mu l)$, where l describes the optical way through the absorbing medium and $\mu = 4\pi\beta/\lambda$:

$$A\left(\beta\right) = \int \left| \sum_{n_3=0}^{N_3-1} \exp\left(i\frac{c}{2}n_3 \cdot \left(\frac{4\pi}{\lambda}\sin\widetilde{\Theta} - \tau\right)\right) \exp\left(-\frac{4\pi}{\lambda}\frac{c}{2}n_3\frac{\beta}{\sin\widetilde{\Theta}}\right)\right|^2 d\widetilde{\Theta}.$$

Using the experimentally obtained data for N_3, τ and $\beta\left(h\nu\right)$ (see figure 4.4), the experimentally obtained integrated intensity $I_{\widetilde{\Theta}}$ was corrected to yield the required total-integrated intensity of the Bragg reflection according to

$$I^{integr.} = I_{\widetilde{\Theta}} \cdot \Delta_{||}^{-1} \cdot \frac{A\left(0\right)}{A\left(\beta\right)}. \tag{A.4}$$