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Resonant soft X-ray and extreme ultraviolet magnetic scattering in nanostructured magnetic materials: Fundamentals and directions☆

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ABSTRACT

Theoretical and practical aspects of resonant magnetic and charge scattering in the soft X-ray and extreme ultraviolet spectral ranges are reviewed. Intensity-only measurements are considered because they are more efficient than polarization-resolving measurements. Two very different approaches are discussed and compared; transmission small-angle scattering described by a simple kinematical scattering model and specular reflection described by more complex yet standard magneto-optical formalisms. In both cases the scattered intensity is seen to contain distinct terms resulting from pure-charge scattering, pure-magnetic scattering, and charge-magnetic cross-terms, and emphasis is placed on distinguishing these contributions via their energy spectra and its dependence on incident polarization. Combined with measurements vs. scattering vector q, both approaches provide significant capability to resolve magnetic and chemical structure down to nanometer length scales. The role of and need for modeling to obtain reliable information from data is discussed, as are current directions and opportunities.

1. Introduction

The discovery of strong resonant magnetic terms in the atomic scattering factor expansion [1,2] has prompted a great deal of research using X-ray scattering to study magnetic materials that continues to this day. Dipole-allowed transitions to empty, spin-polarized 3*d* and 4*f* resonant intermediate states of magnetic transition metal (TM) and rare-earth (RE) elements originate from spin-orbit split 2*p* and 3*d* core levels, respectively, whose energies $h\nu$ of roughly 400–2000 eV (corresponding to wavelength $\lambda = 3$–0.6 nm) fall within the soft X-ray spectral range typically served by grating monochromators. Transitions to the same 3*d* and 4*f* resonant intermediate states originate from the 3*p* and 4*d* core levels, respectively, with $40 < h\nu < 200$ eV ($30\text{ nm} < \lambda < 6\text{ nm}$) in the extreme ultraviolet (EUV) spectral range. Large element-specific magneto-optical (MO) effects at these resonances facilitate their application to study the spatial distribution of magnetization via angle-resolved scattering. Using radiation from synchrotron and emerging free electron laser & lab-based higher harmonic generation sources [3,4], these element-specific MO effects bring significant new experimental opportunities. Following introductory comments, this paper briefly reviews some practical theoretical and experimental considerations of intensity-only resonant soft X-ray & EUV magnetic scattering in the context of recent applications, and comments on current and future directions for extended application of these approaches.

While resonant magnetic scattering mechanisms are essentially the same across all spectral regions, several practical aspects distinguish their application in the soft X-ray range [5]. One is that long soft X-ray wavelengths limit spatial resolution and usually, but not always, position crystalline Bragg peaks beyond the accessible range of scattering vector $\bar{q}$. Low-q techniques such as small-angle scattering and specular reflectivity are thus well-suited for soft X-ray studies. Another is that strong soft X-ray absorption significantly limits penetration into samples and requires in-vacuum measurement. This strong absorption also means that the

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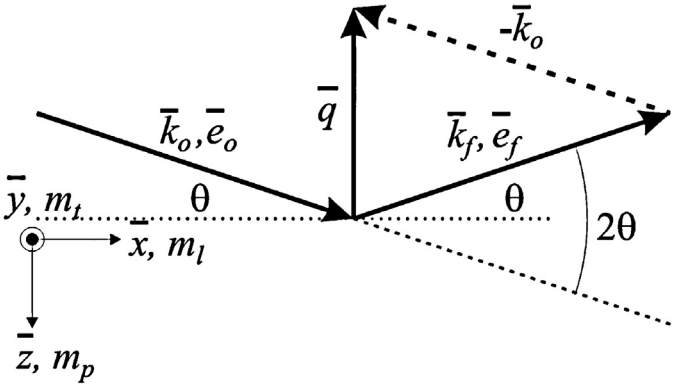

Fig. 1. A generalized scattering diagram in which the difference of incident and scattered wavevectors defines the scattering vector $\bar{q} = \bar{k}_f - \bar{k}_o$ and scattering plane. The incident and scattered polarization vectors $\hat{e}_{o,f}$ are typically described in terms of orthogonal linear components in (p) and perpendicular to (s) the scattering plane. The atomic magnetization vector $\hat{m}$ can in general have longitudinal (m_l), transverse (m_t) and polar (m_p) components. The $\hat{m}$ components are defined such that m_l and m_p are in the scattering plane perpendicular and parallel to $\bar{q}$, respectively, and m_t is out of the scattering plane. This 2-dimensional scattering plane is often a good approximation when scanning an aperture detector about an axis. When using a 2-dimensional detector one must consider the appropriate range of scattering planes, vectors, and moment projections.

imaginary part of relevant scattering factors or optical constants must be included in calculations of scattering effects, while in the hard X-ray range absorptive terms are often small and ignored. These attributes make soft X-ray resonant scattering well-suited to study magnetic behavior in nanostructured materials, thin films and layered heterostructures, and near-surface regions of bulk magnetic and other correlated electron systems.

The term scattering encompasses many different processes and types of measurements and can mean rather different things to different researchers. Here we take scattering to mean resonant elastic scattering measured as a function of scattering angle 2θ, X-ray energy $h\nu$, incident and possibly scattered X-ray polarization, applied magnetic field H, temperature, and possibly other parameters. Angle-resolved scattering is practiced in several different scattering geometries, all of which are relevant to resonant magnetic scattering. In all cases structural correlations are probed along the scattering vector $\bar{q} = \bar{k}_f - \bar{k}_o$ where $\bar{k}_o$ and $\bar{k}_f$ are the wave vectors of the incident and scattered wave fields as in Fig. 1. Below we consider the cases of scattering in transmission from thin films and specular reflectivity as in Fig. 2(a) and (b).

Finally, thinking in terms of resonant *magnetic* scattering can be misleading because resonant (and non-resonant) *charge* scattering always accompanies the magnetic part. Thus we must consider the interplay between charge and magnetic scattering amplitudes when planning and interpreting experiments. This interplay is both ubiquitous and strongly dependent on specific samples and different parameters as detailed below. The combined sensitivity to chemical and magnetic properties is significant as the two are usually highly correlated in space and it is frequently the imperfections in these correlations that are of interest in specific material systems. The ultimate utility of resonant soft X-ray magnetic scattering will thus depend on how well we can measure and model often-subtle details in terms of mixed chemical and magnetic heterogeneity.

2. Theoretical and practical considerations

The resonant atomic scattering factor $f(h\nu)$ expansion [1] provides the fundamental description of the distinct charge and magnetic X-ray scattering amplitudes as

$$f = (\hat{e}_f^* \cdot \hat{e}_o) f_c - i(\hat{e}_f^* \times \hat{e}_o) \cdot \hat{m} f_{m1} + (\hat{e}_f^* \cdot \hat{m})(\hat{e}_o \cdot \hat{m}) f_{m2} + \ldots . \quad (1)$$

Here $\hat{e}_o$ and $\hat{e}_f^*$ are unit polarization vectors of the incident and scattered wavevectors, respectively, and $\hat{m}$ is a unit vector along the axis of local magnetization of the scattering atom. The f_c, f_{m1}, and f_{m2} terms describe distinct linear combinations of matrix elements between initial and resonant intermediate states resolved according to spherical harmonics Y_{LM} and hence angular momentum increment of the transitions involved. Only electric dipole (EL with $L = 1$, $\Delta M = -1, 0, 1$) terms are retained in (1). We group the non-resonant charge term (approximately the atomic number

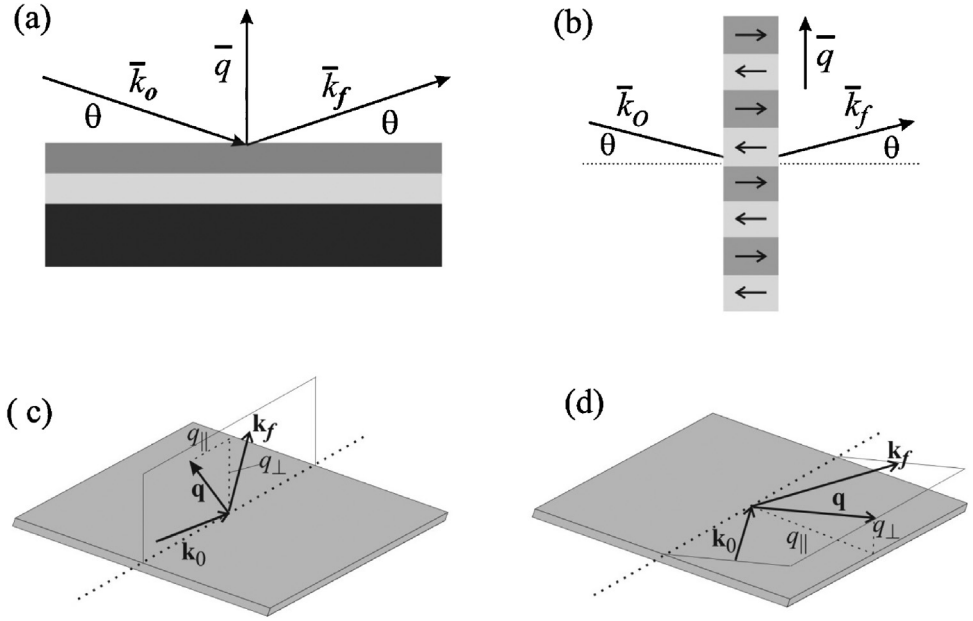

Fig. 2. Common scattering geometries and the associated wave- and scattering-vectors. Specular reflection (a) and symmetric transmission (b) geometries position $\bar{q}$ along the surface normal and in the film plane and optimize coupling to in-depth, and in-plane structure, respectively. Off-specular reflectance (c) and grazing incidence (d) geometries provide both in-plane and out-of-plane $\bar{q}$ components. In general scattered intensities can be measured using a point detector corresponding to a small volume of $\bar{q}$-space, or via 2D detectors sampling a larger range of $\bar{q}$-space for a fixed $\bar{k}_o$.