Reconstruction of X-ray Waveguide Fluorescence Holography for Evolving Nanostructures in Thin Films

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Fluorescence is the emission of light or radiation by certain substances as a result of absorbing incident radiation of a shorter wavelength. Applications of fluorescence such as in spectroscopy and microscopy do not utilize its steradian sensitivity because the directly emitted fluorescence is an isotropic outgoing spherical wave. However, an anisotropic power distribution of the fluorescence can be induced when the fluorescence is modulated by local environmental inhomogeneities near its emitting source. This concept has been explored as the X-ray fluorescence holography (XFH) on oriented crystalline samples, where local atomic structures can be reconstructed from fluorescence holograms with sub-atomic spatial resolution. Besides crystals, an electromagnetic waveguide can also modulate the internally emitted fluorescence, creating a concentric cone-shaped hologram when the fluorescence leaves the waveguide. In this work, we illustrate the principle of XFH for a waveguide and demonstrate that when applied to a thin film consisting of fluorescence substances, it becomes an *in-situ* and time-resolved imaging technique – X-ray waveguide fluorescence holography (XWFH) – for embedded nanostructures and their kinetics in the film.

Conventional XFH is done in either the normal or inverse mode. The holographic reconstruction in either mode is achieved via a back-propagation of the far field hologram in the framework of kinematic approximation. During the holographical reconstruction, these Fourier-transform based algorithms often find difficulties for actual XFH experimental conditions, causing ghost patterns or false atomic images due to the presence of non-kinematic effects such as mode mixing, self-interference, multiple scattering, extinction, and etc. In contrast, these effects are inherently significant in a waveguide for the XWFH, requiring the reconstruction to be performed in the framework of the dynamical theory. By applying an iterative holographical reconstruction algorithm to a mixed-mode XWFH carried out at both grazing-incidence and exit angles, we can take many advantages of the dynamical scattering effects and treat them as additional constraints for better convergence and reconstruction qualities.

Samples of gold nanoparticle monolayers embedded in supported polymer films were selected to illustrate the working principle of the mixed-mode XWFH. Buried nanostructures in thin films have often been measured with forward scattering-based techniques such as grazing-incidence small-angle X-ray scattering (GISAXS) and reflectivity. With XWFH, we were able to monitor the diffusion kinetics of the nanoparticles because the broadened nanoparticle distribution upon thermal annealing alters the waveguide conditions which were detected as the variation of the angular dependence of the fluorescence hologram. The advantages of performing XWFH on nanostructured thin films emerged when the dynamically reconstructed gold atomic number density distribution was compared to the result from the reflectivity and the structures from model fitting of the simultaneously collected GISAXS.