Nanomaterial structure determination using XUV diffraction

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Abstract: Diffraction using coherent XUV radiation is used to study the structure of nanophotonic materials, in this case an ordered array of 196nm spheres. Crystal structure and defects are visible, and the nanomaterial dielectric constant determined.

Typical nanomaterials designed for visible and near infrared applications have structure on the 10-100nm length scale. By necessity, the detailed structure of such a material cannot be studied using these wavelengths. In this paper we describe the use of a femtosecond coherent XUV source to look at the detailed structure of a self-assembled array of polystyrene nanospheres. These arrays are often used as templates for nanophotonic or nanoplasmonic materials. XUV diffraction can give complementary information to electron microscopy techniques such as SEM or TEM. In this example we obtain information about the dielectric constant of the nanospheres in the XUV.

The samples for these experiments were self-assembled arrays of 196nm polystyrene spheres on a 50nm SiN substrate, which is transparent in the XUV. Coherent XUV radiation at 25-30nm was produced using high harmonic generation (HHG) from an argon gas cell source excited with 1mJ, 35fs laser pulses. Visible radiation was blocked with a thin (~200nm) aluminium filter. The XUV was focused to a spot of radius ~10µm using a Mo/Si multilayer mirror. Diffraction from the sample was recorded in transmission on an XUV CCD camera.

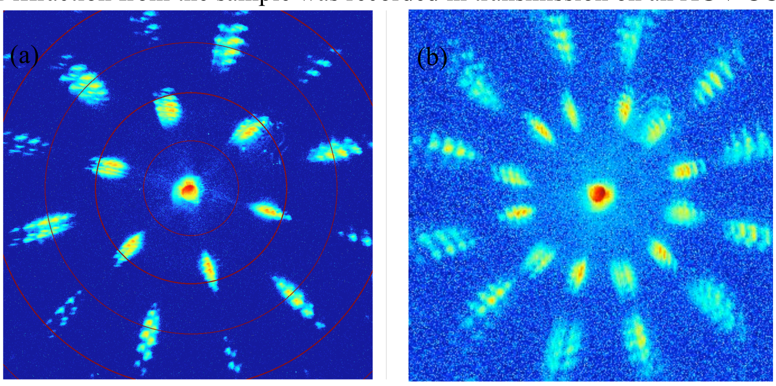

Figure 1: XUV diffraction patterns from (a) single and (b) multiple crystallites within the self-assembled nanosphere array.

Fig. 1(a) shows the XUV diffraction pattern from a well-ordered area of the self-assembled hexagonal sphere array. The radial structure in the pattern comes from the multiple wavelengths of the source. Other structure arises from defects in the hexagonal structure of the film. Fig. 1(b) shows the diffraction from an area of film with two crystallites, clearly showing the difference in angle of their hexagonal lattices. The structural information available from these patterns is more detailed than that available from SEM or TEM images, because large areas of film can be effectively sampled at ~λ/2 resolution, whereas SEM images are usually limited by camera resolution. Additionally, no preparation of the sample is necessary, and the technique is sensitive to the dielectric constant of the material.

Scattering intensities from the different wavelengths in fig. 1 can be used to determine the refractive index of the material, by using Mie scattering theory to calculate the form factor. Fig. 2 shows the variation of real and imaginary parts of the refractive index as a function of wavelength, compared with the values tabulated in the NIST and CXRO databases[1]. This direct determination of dielectric constant is considerably more accurate than the extrapolation from atomic scattering factors.

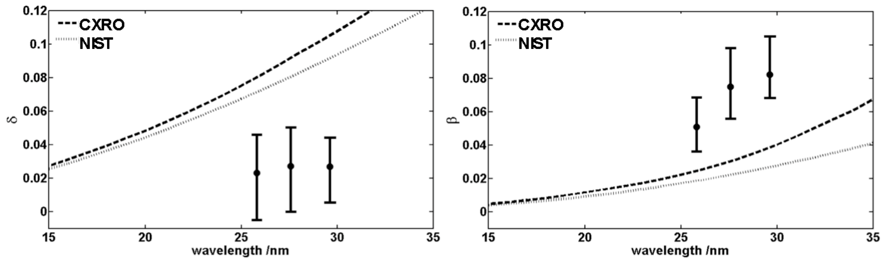

Figure 2: Variation of refractive index, *(1-δ) –iβ*, with wavelength.

In conclusion, we have used diffraction of coherent XUV radiation produced in the lab using HHG to study the structure of a self-assembled 2D nanocrystalline film of polystyrene spheres. The use of lab-based XUV sources will provide significant benefits in the characterization of nanomaterials because of the ability to look at optical diffraction at length scales close to the characteristic length scale of the nanomaterial itself.

[1] Chantler, C.T., J. Phys. Chem. Ref. Data **29(4)**, 597-1048 (2000); Chantler, C.T., J. Phys. Chem. Ref. Data **24**, p.71-643 (1995); B.L. Henke, E.M. Gullikson, and J.C. Davis , Atomic Data and Nuclear Data Tables **54(2)**, p181-342 (1993).