3D MICROSCOPY PROVIDES THE FIRST DEEP VIEW

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Abstract: Coherent X-ray diffraction microscopy has recently been developed to image nanostructures and biological systems in two and three dimensions. A highest resolution of 7 nm has been achieved. We anticipate this 3D imaging technique will find broad applications in the burgeoning field of nanoscience and technology.

Keywords: oversampling, coherent X-ray diffraction, synchrotron radiation

Visualizing the arrangement of atoms has played a crucial role in understanding the microscopic world. There are already a few ways of imaging atomic structures, but each has its limitations. Scanning probe microscopes are limited to imaging atomic structures at surface. Transmission electron microscopes can resolve individual atoms but only for samples thinner than ~ 30 nm. Crystallography can reveal the globally averaged 3D atomic structures based on the diffraction phenomenon, but requires crystals. These limitations can in principle be overcome by coherent X-ray diffraction microscopy, shown in Fig. 1.

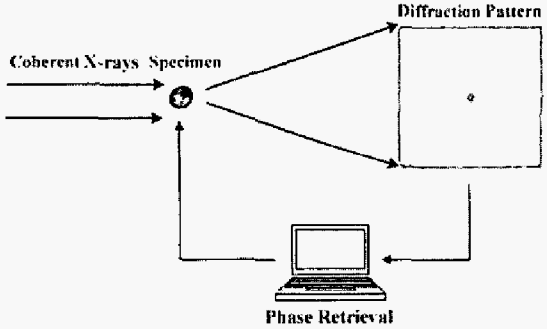

Fig. 1 The principle of coherent X-ray diffraction microscopy.

When a coherent wave of x-rays illuminates a noncrystalline specimen, the far-field diffraction intensities are continuous and weak. This continuous diffraction pattern can be sampled at a spacing finer than the Nyquist frequency (*i.e.* the inverse of the specimen size), which corresponds to surrounding the electron density of the specimen with a no-density region [1]. The higher the sampling frequency, the larger the no-density region. When the no-density region is larger than the electron density region, it has been shown that the phase information is uniquely encoded inside the diffraction pattern [1] and can be recovered directly by an iterative process that takes advantage of information such as the fact that electron density outside the object is zero and within the object is positive [2].

The first experimental demonstration of x-ray diffraction microscopy was carried out in 1999 [3]. Since then, it has been successfully applied to imaging a variety of samples ranging from nanocrystals, biomaterials to double wall carbon nanotube in both two and three dimensions by a number of groups [4,5]. A biennial international workshop series has been organized to discuss the current progress and the future potential of coherent diffraction imaging.

While coherent diffraction microscopy can in principle achieve the atomic resolution which is limited only by the X-ray wavelengths, the highest resolution currently achievable is ~ 7 nm [6], shown in Fig. 2. The specimen, made by electron beam lithography, consists of a single layer of Au pattern with a size of 2.5 x 2 x 0.1 μm, and was supported by a silicon nitride membrane window, which is transparent to X-rays. Fig. 2(a) shows a SEM image of the specimen. A high-resolution X-ray diffraction pattern was recorded from the specimen, which has a resolution of 7 nm at the edge, shown in Fig. 2(b). An iterative algorithm was used to retrieve the phases directly from the oversampled diffraction pattern of Fig. 2(b). Each iteration of the algorithm consists of the following four steps. (i). A complex reciprocal space array was constructed from the current phase set and the square root of the measured diffraction intensity on a suitable grid. For the initial cycle, a random phase set

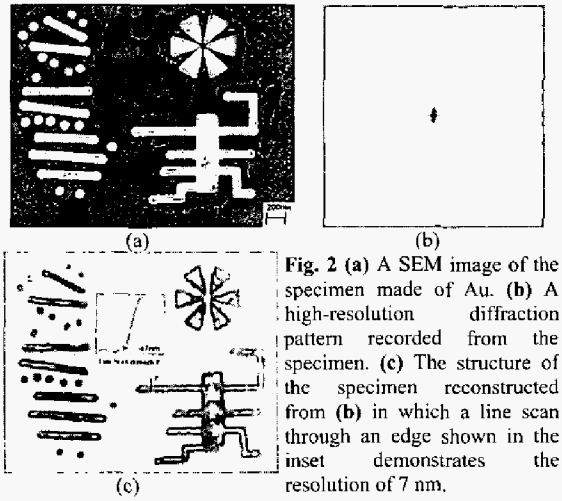

Fig. 2 (a) A SEM image of the specimen made of Au. **(b)** A high-resolution diffraction pattern recorded from the specimen. **(c)** The structure of the specimen reconstructed from **(b)** in which a line scan through an edge shown in the inset demonstrates the resolution of 7 nm.

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was used. (ii). By applying the fast Fourier transform, an electron density distribution on a second grid was calculated from the reciprocal space array. (iii). A "support constraint" was applied to separate the electron density from the no-density region. The electron density outside the support and the negative electron density inside the support were pushed close to zero. (iv). A new reciprocal space array was calculated by applying the inverse fast Fourier transform to the new electron density. The phases of the new reciprocal space array were then adopted in the next iteration after setting the phase of the central pixel to zero. By using a 2.53 x 2.06 μm square as the "support", the phases were directly recovered from the diffraction pattern. Fig. 2(c) shows the reconstructed image, in which both the shapes of individual nano-structures and the variation of the electron density inside the nano-structures are visible.

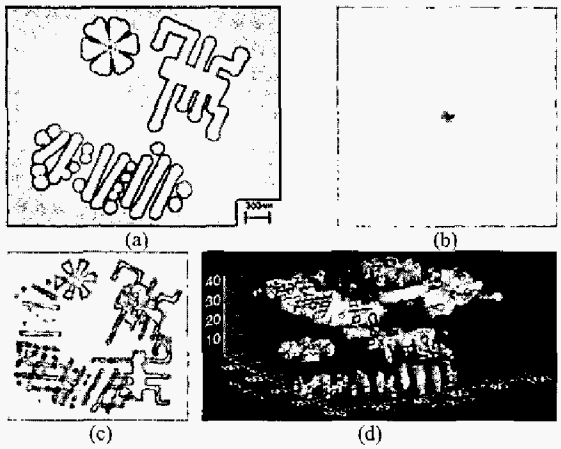

Fig. 3 (a) A SEM image of a buried nanostructure, showing the pattern in the top layer, but not the pattern in the bottom layer. (b) A coherent X-ray diffraction pattern with a rotation angle equal to 0°. (c) An image reconstructed from (b) with a resolution of 8 nm where both the top and bottom layered patterns are clearly seen as overlapped in this 2D image. (d) The reconstructed 3D structure displayed in iso-surface rendering. The finest division corresponds to 27.5 nm.

3D imaging of a buried nanoscale material has also been carried out [7]. The sample consists of two single-layered Ni patterns (each with a size of 2.5 x 2 x 0.1 μm) rotated relatively 65° to each other in-plane and separated by a distance of 1 μm, which was supported by a thin silicon nitride membrane window. Fig. 3(a) shows a SEM image of the sample. Due to the 1 μm separation of the two layers, the SEM image shows the pattern in the top layer, and the pattern in the bottom layer is visible only as a soft blur. By using coherent X-rays with a wavelength of 2 Å, a series of thirty-one 2D diffraction patterns were recorded from the sample with the rotation angles ranging from −75° to 75° in 5° increments. Fig. 3(b) shows a coherent diffraction pattern with a rotational angle of 0°. The oversampled diffraction pattern was directly converted to a high-resolution image by using the iterative algorithm, shown in Fig. 3(c). The top and bottom layered patterns are clearly seen as overlapped in this 2D image, and the variation of the electron density on the nanometer scale is also visible. Five more reconstructions with different random initial phase sets were done and the reconstructed images consistently and faithfully reproduced the original sample pattern. Due to the longer penetration length of X-rays than of electrons, it can be seen that coherent diffraction imaging can image much thicker specimens at high resolution. This is probably beyond the capability of both scanning probe microscopy and transmission electron microscopy.

To obtain a 3D image of the nanoscale material, we first mapped the thirty-one 2D diffraction patterns onto a 3D diffraction pattern by using interpolation [7]. A 3D image was reconstructed from the assembled diffraction pattern using the iterative algorithm described above. Fig. 3(d) shows a 3D iso-surface rendering of the reconstructed image. The finest division in z-axis corresponds to 25 nm and the distance between two patterns is about 1 μm, which is consistent with the known characteristics of the sample.

The potential of 3D X-ray diffraction microscopy is enormous. It can be applied to investigate a wide range of systems such as synthetic nanostructure, composite materials and biological systems. Furthermore, with the appearance of X-ray free electron laser facilities, it may be able to image single particles at the near atomic resolution with the sub-picosecond time resolution.

Acknowledgments

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