Point-source holographic imaging of nanostructures and interfaces with low energy electrons

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Abstract. A lensless holographic in-line point source microscope was envisioned more than half a century ago, but its realization with electron waves has come short due to not only difficulties inherent in Fresnel-type reconstruction methods, but also to the lack of an adequate (spatially and temporally coherent) point source. With the recent creation of ultrasharp nanotips, which can field emit electrons from a single atom at their apex, an extremely coherent electron source is available that provides a great boost to the holographic method. The spatial coherence of such nanotips is a few Å, while their temporal coherence is characterized by a value of energy dispersion (FWHM) as low as 0.1 eV. In this work we ascertain the use of such a microscope in the imaging of nanoscale structures and interfaces. The method is suitable for two- and three-dimensional imaging of solid nanoparticles, thin crystals, and surfaces, but also for biological entities. We show how improvements in the reconstruction method can be made by applying the rigorous Fresnel-Kirchhoff diffraction theory adapted to Electron Optics. Sub-nanometer resolution is achievable for beam energy between 100-200 eV.

1. Introduction
Recent advances in the fabrication of extremely coherent and bright electron sources featuring improved spatial and temporal coherence properties open up new horizons in low energy electron point-source (LEEPS) microscopy. The most coherent source of electrons seems to be found in ultrasharp nanotips [1-3]. Such nanotips end in a single atom at the apex of a nanoprolusion with a very small radius (a few nanometers) resting on the shank of the tip with a larger radius (hundreds of nanometers). Consequently, the beam of field emitted electrons generated from such tips present provocative characteristics such as high coherence and degeneracy, extreme brightness [4], strong focusing, very sharp energy spectra (sometimes with multiple peaks) [5, 6].

In this work we undertake a theoretical study for the simulation in atomistic detail of LEEPS holograms and their reconstruction. We investigate the possibility of obtaining both the amplitude and phase reconstruction for nanosized structures with ordered lattice, as well as disordered structures (e.g. defects in lattice). We propose a new reconstruction method for in-line holograms and analyze its performance with respect to previous reconstruction formulas.

2. Theory of Hologram Formation and Reconstruction
A holographic electron microscope has the potential to go beyond existing microscopic methods by avoiding irreducible lens aberration omnipresent in other electron microscopes. What is recorded at the hologram plane is the intensity of the total wave resulting from the interference of the reference and the scattered wave.
The first term of the RHS of (1) is just the background pattern obtained in the absence of any scattering object. The second term is the intensity of the scattered wave alone. Finally and most importantly, the last term is the holographic term, which results from the interference of the reference and scattered waves. The holographic regime is observed if the object scatters only a small fraction of the incident wave, so that we can neglect the second term in the RHS of (1). In addition, by subtracting the background, \( A_{\text{ref}}^2(s) \), from \( I_{\text{tot}}(s) \) we obtain the so-called contrast hologram, \( I_{\text{holo}}(s) \), which is usually used as input for digital reconstruction.

In order to simulate the formation for in-line holograms, we use the scattering theory in its integral form, i.e. Lippmann-Schwinger equation [9]. In this theoretical frame, the total wave function at point \( s \) on a detector placed at an asymptotically large distance from the sample is given by

\[
\psi_{\text{tot}}(s) = A_{\text{ref}}(s) \frac{e^{ik_g}}{g} + \sum_{l=2}^{N_{\text{term}}} A_{\text{ref}}(s) \frac{e^{ik_{lj}}}{r_j^l} \sum_{j=1}^{N_{\text{atoms}}} \left( 2l + 1 \right) \sin \theta_j \exp \left( il \theta_j \right) P_l \left( \cos \theta_j \right),
\]

where \( r_j \) are the positions of the atomic centers in the scattering object, \( l \) is the angular momentum number for each partial wave, \( P_l \) are the Legendre polynomials, \( \theta_j \) are the scattering angle for each atom, and \( \delta_l \) are the scattering phase shifts for each value of \( l \). We make the approximation that the values of \( \delta_l \) are given by the free atom phase shifts. These are provided by the NIST Standard Reference Database 64 [11]. For simplicity we assume a Gaussian beam, with a given width specified by its full width at half maximum (FWHM) angle, \( \gamma_{\text{beam}} \).

In this study we adopt two reconstruction formalisms of in-line holograms:

i. Helmholtz-Kirchhoff formula, previously used in LEEPS holography [9] as well as in LEED and photoelectron holography [10]

\[
\psi_{\text{rec}}(r) = \int_{\text{screen}} ds_x ds_y \psi_{\text{holo}}(s) \exp \left( ik_x s_x + ik_y s_y \right), \quad (3)
\]

ii. Fresnel-Kirchhoff theory of diffraction [7], in which the reconstruction of the hologram can be performed by the formula

\[
\psi_{\text{rec}}(r) = \int_{\text{screen}} ds_x ds_y \psi_{\text{holo}}(s) \frac{e^{ik_{d} e^{-ik_{d} |s|}}}{|s|}, \quad (4)
\]

3. Results - Reconstruction of Nanostructures from Simulated Holograms

In the following we present the results obtained by reconstructing holograms simulated using the above methodology. An ideal sample for our microscope is graphene (single atomic layer of graphite). Not only does it offer a well known (2-dimensional) crystalline structure, but it also insures that the holographic regime mentioned above is verified by allowing a reference wave (through and around the sample) much stronger than the scattered wave. In Fig. 1 we present the reconstruction of graphene from a simulated hologram, using the reconstruction formula (3) and (4), respectively. The hologram was simulated assuming an electron energy of 200 eV, a beam angle, \( \gamma_{\text{beam}} = 50 \text{ deg.} \), source-object distance \( d = 40 \text{ nm} \), source-screen distance \( L = 1 \text{ cm} \), and a micro-channel plate detector with 12 micron pore-pore distance.

The hexagonal structure of graphene is recovered well in both intensity and phase reconstructions. Atoms appear dark in the amplitude images and bright in the phase image. The phase image crop of the central region of the sample (top middle inset in Fig. 1) also seems to have a slightly better resolution than the amplitude images. Notice that eq. (4) performs better in terms of the consistency of contrast throughout the sample. This improvement is achieved at the expense of lower speed, as only formula (3) is amenable to evaluation by fast Fourier Transform techniques.
As a second sample we consider a short section of a 16 × 16 carbon nanotube capped at one end, which contains 2000 atoms. Its amplitude reconstruction is shown in Fig. 2 (top). The lattice structure is not easily apparent in this reconstruction due to the fact that the walls are curved while the reconstruction is done in a plane.

Figure 1: Reconstruction of a rectangular cut of a graphene sheet (shown in the bottom middle inset): amplitude reconstructions with the Helmholtz-Kirchhoff formula, eq. (3) (leftmost image) and Fresnel-Kirchhoff formula, eq. (4) (right-most image). The top middle inset is a central crop of the phase reconstruction with eq. (3) highlighting the hexagonal structure. The scale bar applies to the amplitude images only.

Our holographic method is not restricted to periodic structures. Disordered structures are amenable to imaging as well. Defects (e.g. missing atoms) in the lattice can also be imaged by this method. In the case of graphene, we randomly removed atoms from the structure and simulated the hologram and its reconstruction. The results are shown in Fig. 2 for one atom missing (b), and eight atoms missing (c).

In the above results, the source was considered to be ideal, i.e. the incoherent wave (thought of as noise) emitted from the terminal atom of the nanotip is negligible when compared to the coherent wave (signal). In practice, we expect incoherent emission of electrons from a finite source to contribute to the image formation. The incoherent source size will thus play an important role in determining the accuracy and resolution of the reconstruction. A quantitative analysis of the effect of the source size will be undertaken shortly.

4. Discussion and Conclusion
Recent progress in fabrication of tungsten ultrasharp nanotips has prompted us to exploit their field emission beam for electron holography. Such nanotips are very stable emission sources, extremely coherent and bright. Their use in point-source holography greatly improves the resolution and magnification of the method. It also allows for the simultaneous imaging of a large areas because of the lateral coherence of the electron wave is also increased as compared to other nanotips used in commercially available microscopes. The increased brightness of nanotips allows for shorter hologram exposure times, therefore minimizing the distortive effects of stray fields on the recorded image. A full holographic procedure including phase retrieval is possible if the distortion effect of magnetic fields via the Aharonov-Bohm effect on hologram formation is rigorously controlled [7]. Thus, the main contributions to the resolution limit are the virtual (incoherent) source size and the degree of magnetic field shielding of the microscope.
Figure 2: (a) Amplitude reconstruction of a capped carbon nanotube segment with the Fresnel-Kirchhoff formula, eq.(4); (b,c) amplitude reconstruction with eq.(3) of graphene with vacancies: one vacancy in (b), and eight vacancies for the sample in (c).

Although here we present results only for the transmission mode, the reflective mode is also possible as recently outlined [12]. Numerical simulations of electron holograms of various samples presented here show the possibility of attaining atomic resolution by lensless low energy electron holography. The possibility of obtaining phase images opens up the way of obtaining elemental contrast across a sample. This is due to the fact that different elements/species yield different phase shifts ($\delta$ in eq. 2) of a scattered electron wave. Hologram simulation and reconstruction of samples with elemental contrast will be undertaken shortly.

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