FRESNEL PROJECTION MICROSCOPY, THEORY AND EXPERIMENT: ELECTRON MICROSCOPY WITH NANOMETER RESOLUTION AT 200 VOLTS

Vu Thien Binh*, V. Semet, S.T. Purcell and F. Feschet

Laboratoire d'Émission Électronique, DPM-UMR CNRS Université Claude Bernard Lyon 1, 69622, Villeurbanne, France

(Received for publication May 29, 1996 and in revised form October 2, 1996)

Abstract

Nanotips are used as electron point sources in the Fresnel projection microscope (FPM). The FPM allows imaging in direct space of free standing nanometric fibres at working voltages around 200 V with observation of details of less than one nanometer. These observations are made without irradiation damage, particularly useful for soft materials such as organic or biological molecules (polymers and RNA). Striking new observations of the images of non-opaque fibres in the FPM are presented for the first time. The experimental results are interpreted within the framework of electron optics by taking into account the properties of the nano-objects and the specific field emission properties of nanotips.

Key Words: Field-emission, nanotips, electron projection microscopy, Fresnel projection microscopy, coherent e-beams, Fresnel diffraction, polymers.

*Contact for correspondence: Vu Thien Binh, address as above.

> Telephone number: (33) 04 72 44 80 70 FAX number: (33) 04 78 89 74 10 E-mail: vuthien@dpm.univ-lyon1.fr

Introduction

In most conventional electron microscopes, the resolution limits are the result of aberrations introduced by the electron optic systems used to reduce the effective source to atomic dimensions. In the Fresnel projection microscope (FPM) (Binh et al., 1994), the atomic size of the source is given directly by the single-atom-apex of the nanotip used as the field emission source (Binh, 1988; Binh and Garcia, 1991; Binh et al., 1996a). The magnification process is due to the radial propagation of the e-beam from the point source, which is specific to the projection microscope (Morton and Ramberg, 1939). This lens-less set-up eliminates the aberrations related to the electron lenses. When combined with the piezo-electric control of source-sample distances, a technology introduced for the scanning tunnelling microscope (STM) {see, for example, the review on STM by Chen (1993)}, the ensemble constitutes a versatile microscope with a magnification in the 10⁶ range.

The imaging mechanisms and the resolution of the FPM are the result of the propagation of the coherent e beam from a nanotip in the near-field electron-optical and electrostatic environment specific to the nano-tip/ nano-object ensemble. In general, the experimental observations show a resolution of 0.5 nm in the 100 to 300 eV energy range. This article: (1) reviews the results previously presented for opaque objects, (2) shows experimental images that demonstrate directly the importance of the source quality, and (3) presents new images of non-opaque objects. The discussion highlights the limits of this microscopy. The effect of multiple diffraction is not considered because the wavelength λ of the e beam (~0.7 Å) is much smaller than the object size. Also, the influence of multiple scattering within the object can be ignored for the opaque objects.

The Fresnel Projection Microscope

The projection or shadow microscope (Morton and Ramberg, 1939) is essentially a lens-less microscope based on the radial propagation of an e beam from a point source as depicted schematically in Figure 1. The projection image has a magnification factor M given by:

$$M = (i/o) \approx (D/d) \tag{1}$$

where i and o are the image and object dimensions and D and d are the distances from the projection point to the screen and to the object, respectively. With the recent technological developments of the piezoelectric ceramics for controlled nanometric displacements (Fink $et\ al.$, 1990; Spence $et\ al.$, 1993; Binh $et\ al.$, 1994), the magnification can reach values in the range of 10^6 for d ~ 10 nm to 100 nm, and D ~ 10 cm.

The FPM is the result of the combination of a nano-tip and a projection microscope. The nanotips are pyramidal nano-protrusions grown on the tops of microscopic tips and ending in one atom. The field emitting area, which corresponds to the last atom of the protrusion, leads to specific properties for the field emitted e-beam (Binh *et al.*, 1994, 1996a). Among them, those of principal interest for projection microscopy are:

- (1) The size of the emitting area is limited to the last apex atom of the protrusion and the position of the virtual source, which is the projection point, is within nano-metric distance from the actual surface.
- (2) The total beam opening in the vicinity of the emitting atom is less than $\sim 20^{\circ}$ and it is self-focused to $\sim 5^{\circ}$ at microscopic distances away from the apex due to the focusing lens effect of the whole tip.
- (3) The field emission current is stable to within 0.25% for durations of up to ~10 hours inside ultra-high vacuum (UHV).
- (4) The energy spread is ~100 meV at room temperature due to field emission process from localised bands.

From a practical point of view, the FPM is essentially composed of a nanotip, mounted on a structure which permits heating, cooling and temperature measurement in front of an object holding system. Piezoelectric motors and tubes allow a three-dimensional (3D) movement between the nanotip and the object in the centimetre range with sub-nanometric steps. The displacement system makes possible the choice of the imaging zones over a large area by the x,y displacements and a continuous zooming on a chosen area by approaching the object to within nano-metric distances to the nanotip. Controlled bending of the piezoelectric tube gives the absolute calibration for the x,y scales at the object plane without any reference to the source object distance, which is a difficult parameter to determine. This means that the scales given in the FPM experimental images below are independent of the imaging process and are related only to the calibration accuracy of the piezo-electric tube. The images are projected on a fluorescent screen, equipped with a micro-channel plate, placed ~10 cm from the nanotip, and are recorded by a video camera with image processing on-line. The volume of the whole effective system including nanotip, object holder, and visualisation screen is around 1000 cm³ (1 liter) and it is inside an UHV chamber. The whole apparatus is isolated from

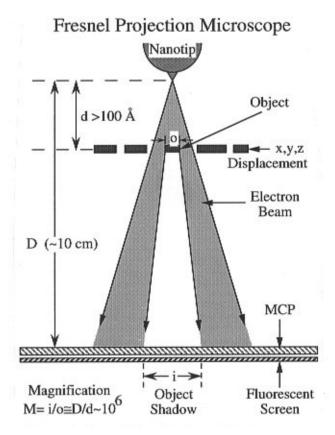


Figure 1. Schematic of the Fresnel projection microscope. The coherent projection source is a field emission W-nanotip emitting in the range of 200 V to 300 V. The image magnification is given in the first approximation by the ratio D/d, where d and D are the distances between the virtual projection point to the object and to the screen, respectively.

external vibrations with a pneumatic system. There is no need to incorporate magnetic shielding in the microscope chamber.

Specimen preparation consists of the deposition on a holey carbon grid of a liquid drop in which the material is diluted. The solvent is then evaporated leaving the material stretched across the carbon holes. We have found that for materials, such as, biological or synthetic polymers, that are susceptible to build nano-range diameter fibres, either alone or within a network, such a preparation procedure is always successful in providing free-standing nano-objects. Other techniques of preparation can be arranged for other materials.

The projection principle

The simple geometrical projection used to illustrate the projection microscope (Morton and Ramberg, 1939) ignores electrostatic deflections which result from the presence of strong local electric field distributions over the nanotip and

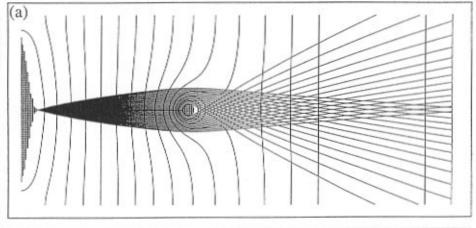
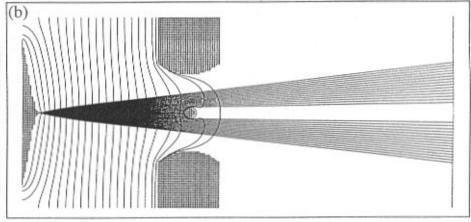


Figure 2. Calculation of the electrostatic potentials and electron trajectories in the projection geometry: (a) for an isolated object in front of the tip; (b) for the same object within a small hole in a plane at the same potential. Tip at -150 Volts, object at 0 Volts. The presence of the planar structure is necessary for the formation of shadow images.

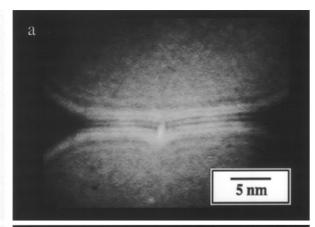


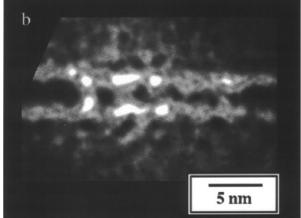
the nano-object. Such contributions are among the main limits for imaging isolated small objects due to the bending of the electron paths near the object (Piquet et al., 1971). This is demonstrated in Figure 2a which shows the results of calculations of the electrostatic potentials and electron trajectories from a point source with a small isolated sphere as the object. The electrons are strongly deviated from a simple rectilinear propagation from the point source by the strong fields near the object and do not form a shadow image of the object on a distant screen. These preliminary simulations of the electron trajectories for the FPM were carried out using the program SIMION by Dahl and Delmore (1988). The projection geometry can be maintained if the object is within an electrostatic environment which counters the deviation and the merging of the beam by the object. This is realised when the potential in the neighbourhood of the nano-object is physically defined at the same potential as the object potential as illustrated in Figure 2b. In practice, this is done by the object being stretched over the holes of a holey carbon grid whose thickness is in the range of 10 nm. The electrostatic induced deviation from straight propagation from the real point source is then taken into consideration by defining a virtual projection source localised at a different position than the

position of the physical source. The optical projection approximation can, therefore, be used to schematically represent the FPM principle.

Opaque nano-fibres

The incident wave front characteristics, which are defined by the dimension of the real source and the source object distance, are the main parameters in the diffraction process. Owing to its protruding geometry ending in one atom, nanotips as FE sources will favour the Fresnel diffraction from nanometric objects (Binh et al., 1994, 1995, 1996b), in contrast to Fraunhofer diffraction, giving then the possibility of observing, on the projection screen, the shape of the object and its localised defects in direct space. The experimental diffractograms of nanometric fibres of carbon, synthetic polymer and RNA reveal details well under 1 nm in the direct space (Fig. 3). The various factors limiting the resolution of the FPM have been previously considered (Binh et al., 1996b); they are: (1) the electron dose for image acquisition; (2) the source size; (3) the diffraction of the propagating electronwave by the nano-object; (4) the energy spread of the electron beam; (5) the blurring of the image due to time varying magnetic stray fields; and (6) the relative vibrations of the point-source object coordinates. The intrinsic resolution (points 1 to 4),





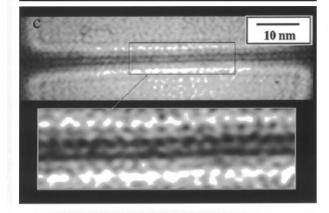


Figure 3. Examples of FPM images: (**a**) a carbon fibre with a diameter of ~1.5 nm, showing a localised defect in the middle; (**b**) a polymer fibre (PVP-PS) showing a periodic superstructure along the length of the fibre; (**c**) an RNA fibre showing periodic variation along the length of the fibre.

estimated to be \sim 0.5 nm, is mostly determined by the first three of these factors which we review below.

Statistical image acquisition

In FPM, the instantaneous projected image of the electron beam is observed on the screen. The theoretical limit for the visual detectability of small objects in a statistical noisy image is given by the Rose equation (Rose, 1948):

$$\Delta a \ge 5 / \{C (fN)^{1/2}\}$$
 (2)

where Δa is the characteristic limit of the object size; C, the contrast factor relative to the immediate surroundings (in our case, we can assume it to be 1); f, the efficiency of "electron utilisation" (assumed to be 1); and N, the number of incident electrons per unit area.

The increase in the resolution with the electron dose in the image formation is demonstrated in Figures 4a to 4f. In this experiment, the total beam current is \sim 0.1 pA and the photos are taken for different exposure times from 40 ms to 4 seconds, which corresponds to a 100 time increase of the electron dose. These results show that for the FPM with a total beam current of 1 to 10 pA, sub-nanometer resolution is reached within 0.1 second exposure time range in agreement with the estimations given by Equation (2). In most of our experiments, we used a 1 to 5 pA total current e beam.

Source size limit

If one considers the projection source to have a diameter 2r and the boundary between two zones of different transparency, then the overlap casting area corresponding to the straight boundary will have a width w, whose value is given by the projection relation and the source diameter:

$$w = 2r \{(D-d)/d\}.$$
 (3)

At the object plane, this value corresponds to the limit of geometrical resolution

$$\Delta g = w (1/M) \approx 2r \tag{4}$$

Relation (4) means that Δg is of the order of the diameter 2r of the projection source.

One point should be emphasised since it supports the necessity of using a nanotip for the highest resolution. The position of the virtual source gives the magnification but the size of the real source of the nanotip controls the geometrical resolution limit. This is clearly seen in the calculation in Figure 2b where the trajectories passing closest to the object define the shadow edge, irrespective of the position of the virtual source.

Because of high coherence of the beam, which is due to the atomic size of the source, the relation between the quality of the source and the resolution obtained can be experimentally determined by the number of observable fringes present in the diffraction images. The last two adjacent fringes are just visible when the overlap of cast areas given by Equation (3) is

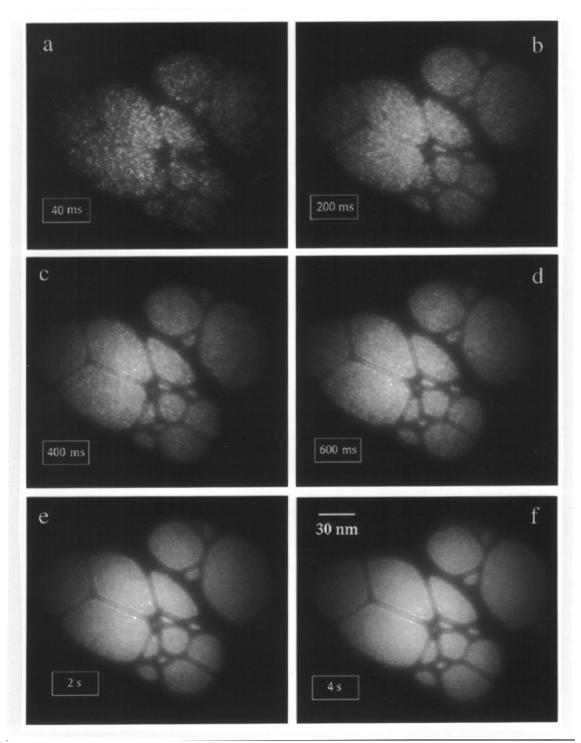


Figure 4. (a)-(f) Sequence of images of a nanometric carbon network irradiated with the same total beam current of 0.1 pA, for increasing exposure times.

less than half the inter-fringe, that is:

$$2r\{(D-d)/d\} \le (1/2)(X_{n+1} - X_{n-1}),$$

(5)

where X_n are the distances of the fringe maxima, or minima, to the position of the casting edge given by the geometrical projection. They are, at first approximation, given by (Bruhat,

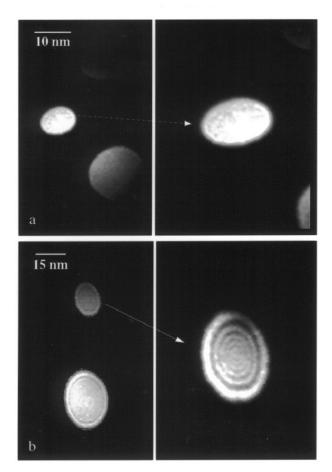
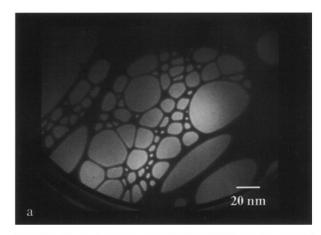


Figure 5. FPM images of: (a) nanometric-scale holes using a blunt nano-protrusion as a projection source; (b) nanometric-scale holes using a nanotip as a projection source. Note the increased clarity and number of Fresnel fringes. These are related to the atomic size of the nanotip apex.



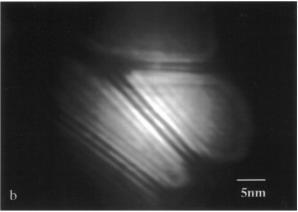


Figure 6. FPM image of a carbon fibre network using a: (a) microtip as an electron source, this is the largest magnification possible with such a tip; (b) nanotip as an electron source, the number of fringes and magnification are both increased.

1954):

$$X_n^2 = {\lambda D (D - d) / d} {n - (1/4)},$$
 (6)

with maxima for n = 1, 3, 5, ..., and minima for n = 2, 4, 6, ...Taking into account Equation (6), and for values of n not too close to 1, Equation (5) can be rewritten as

$$2r \le 0.5 \{\lambda D (D - d) / d\}^{1/2} (1/n^{1/2})$$
 (7)

FPM images of holes obtained either with a blunt nanoprotrusion or with a nanotip are shown in Figure 5. Diffraction fringes are clearly observable only with the nanotip (Fig. 5b), in contrast with the image of the hole obtained with a blunt nano-protrusion (Fig. 5a). The dimension of the field emitting area from the nano-tip, and hence limit of the geometrical resolution in the FPM, is then estimated within the FPM experimental conditions (d \approx 50 nm, D \approx 10 cm, $\lambda \approx$ 0.07 nm) and from the number of observable fringes in the FPM images. One gets:

$$\Delta g \approx 2r \le 0.5 \text{ nm}$$
 (8)

which is in agreement with the known geometry of the nanotips.

Further experimental confirmation of the prominent role of the geometry and size of the tip in the resolution of projection microscopy can also be seen in Figure 6 in which the projection images of carbon networks in a holey carbon grid are obtained either with a microtip or with a nanotip. When the field emission area decreases towards a single atom area characteristic of a nanotip, well-separated diffraction fringes are observed. Moreover, Figure 6a shows the maximum magnification

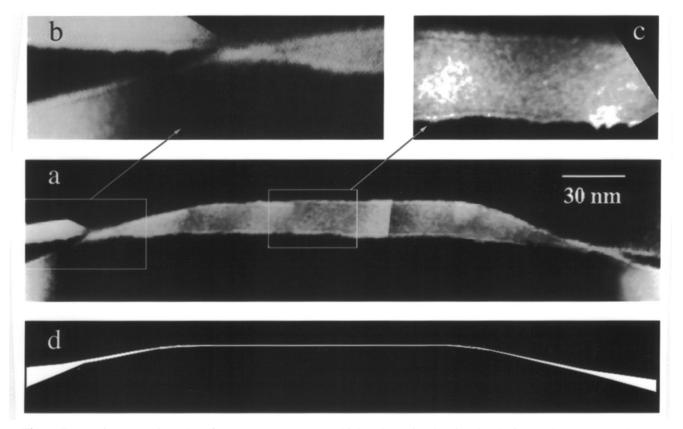


Figure 7. FPM images (**a**, **b**, and **c**) of non-opaque nanometer-thick carbon wire showing that the images have a reversed contrast with respect to opaque wires. (**a**) A composite of high magnification images taken along the carbon showing the passage from opaque to non-opaque zones where the carbon wire gets thinner. (**b**) Detail of the opaque to non-opaque transition zone (magnified 4 times from Fig. 7a). (**c**) Detail of the non-opaque zone showing some fringes inside the bright area (magnified 4 times from Fig. 7a). (**d**) A sketch representing the shadow mask of the carbon nanowire which gives the FPM image at Figure 7a.

obtained with the blunt microtip before the crashing of the tip on the sample, because the virtual source for a micro-tip is far back from the real surface of the tip (Binh *et al.*, 1995). Conversely, higher resolution is possible with nanotips as illustrated in the images in Figure 6b and discussed below.

Resolution limit by diffraction

In presence of diffraction fringes, the geometrical projected edge can be localised within a distance equal to half the distance of the first bright fringe (see Fig. 5b):

$$X_1 \approx D(\lambda/d)^{1/2},$$
 (9)

a value which depends on d, i.e., on the magnification factor. This distance, taken back at the object plane, defines the resolution limit due to the diffraction, which is:

$$\Delta d = (1/2) (X_1/M) \approx (1/2) (\lambda d)^{1/2}.$$
 (10)

Within the experimental conditions of FPM, Δd varies in the range between 0.4 nm to 1 nm for source-object distance varying from 10 nm to 50 nm.

Non-opaque objects

The discussions above have been developed for opaque or nearly opaque objects immersed in e-beams with energy in the range of 200 eV, which implies sample thicknesses greater than 1 to 2 nm {a compilation of electron free mean paths for different materials is given by Seah and Dench (1979)}. Under this assumption, the formation of the Fresnel diffraction images is well described by the Fresnel-Kirchhoff integral (Binh *et al.*, 1996b) for an opaque mask using a virtual source to reflect the lens effects due to deflections by the nanotip and the nano-object. Good agreement was found between experimental observations and calculated Fresnel diffractograms (Binh *et al.*, 1995, 1996b).

In the case of objects which can no longer be considered as opaque to the impinging electrons, i.e., objects

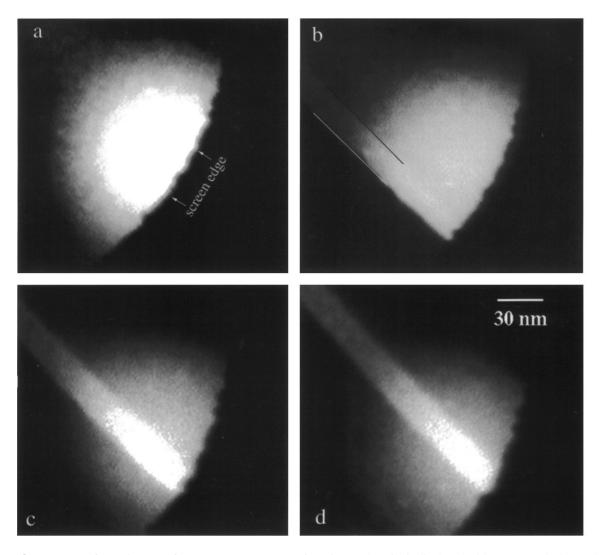


Figure 8. Sequence of FPM images of non-opaque nanometer-size wires as the wire is displaced with respect to the beam showing the modifications of the beam size and intensity resulting from the sucking-in effect of the object. (a) Beam size without the object; (b) to (d) progressive displacement of the nanowire across the beam, coming from the bottom of the image. In (b), the position of the bright image due to the interaction between the beam and the nanowire is indicated by broken lines.

whose dimensions are in the order of 1 nm or less, and with a large electron mean free path due to their composition, the role of the electric field distribution around the objects becomes more important. As a consequence, the electrostatics comportment at the tip and, in particular, at the object, can no longer be taken into account by considering a virtual projection source whose position differs from the real field-emitting surface, as for the opaque objects. This results in the striking observation that thin molecular-scale wires project a bright positive image rather than the usual dark shadow surrounded by fringes of the projection image of an opaque wire. Several examples are shown for carbon fibres and

polymethyl methacrylate (PMMA) networks (Figs. 7, 8, 9 and 10).

Some elementary analysis is necessary to distinguish the various mechanisms at play in this new type of image and, in particular, to be convinced that the slit-like bright images reflect a transmission through the molecular-scale wire. In detail:

(1) The electric field distribution around the nanoobjects will lead to larger deformations of the electron trajectories towards the object. This will have two consequences: (i) the virtual source position must be redefined within the nanotip-nano-object field distribution, and (ii) the

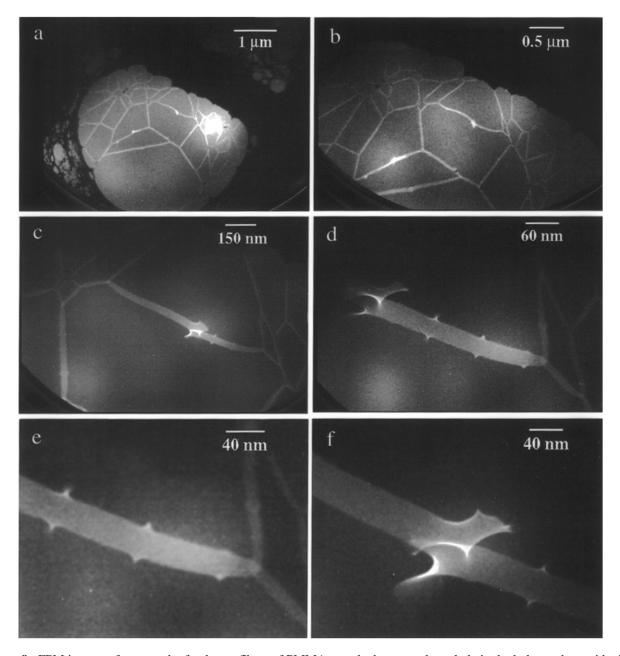


Figure 9. FPM images of a network of polymer fibres of PMMA stretched across a large hole in the holey carbon grid with increasing magnification from (a) to (f). The whole micrometer-scaled network is composed of nano-meter diameter fibres, non-opaque to ~200 V electrons. Localised features in the images along the nano-fibres which represent periodic defects (e) or a fibre connection (f) are clearly visible as bright extrusions.

density of electrons hitting the object is more important than the density of the remaining surrounding e beam due to a sucking-in effect of the electron flux by the local electric field of the object.

(2) As the object dimensions become smaller and

smaller, the transmission coefficient of the object will become more and more important. The objects, for example, the nanofibres, which are opaque masks for dimensions greater than ~2 nm (i.e., electron density transmitted through the fibres is much less than the outside beam) will be "slits" when their

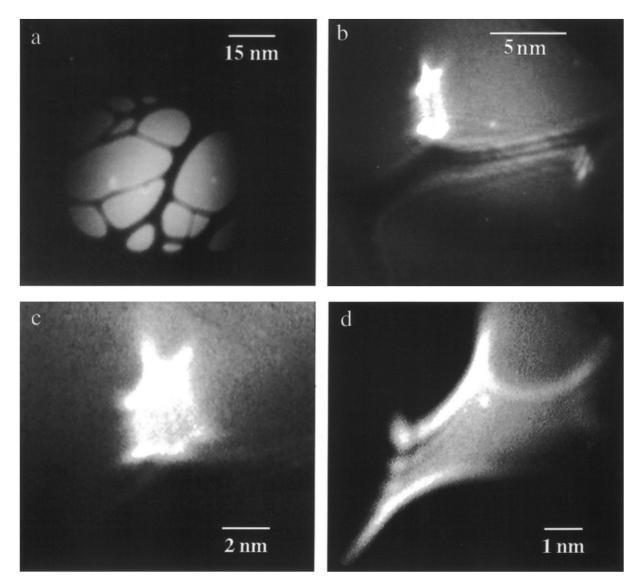


Figure 10. Bright images for a small carbon prickle protruding from a thicker carbon fibre with increasing magnification from (a) to (d). Periodic variation of fringe intensity within the bright projection images (c) and (d) is related to local field variation along the small carbon prickle.

diameters become smaller than a nanometer, i.e., the electron density transmitted through the fibres is much greater than the electron density in the outside beam due to the sucking-in effect. The Fresnel diffraction patterns will then evolve from small wire diffraction pattern towards a pattern showing bright lines with internal diffraction fringes which is characteristic of Fresnel diffraction from narrow slits, i.e., slit-like diffraction patterns.

The passage from a wire like Fresnel diffraction (opaque objects) towards a slit-like Fresnel diffraction (non-opaque objects) can be observed along a carbon wire whose

diameter decreases regularly. Such an observation is shown in Figure 7. The high magnification observations of the bright zones (Figs. 7b and 7c) reveal the presence of fringes. The sucking-in effect and increased transparency is clearly observed at high magnification and by moving the fibre across the beam. In Figure 8, experimental FPM images are shown that demonstrate the modifications of the geometry and intensity of the e beam when a transparent object is placed at various positions in the beam. Supporting calculations of electron trajectories in the case of opaque and non-opaque objects are shown in Figure 11. The existence of an increased

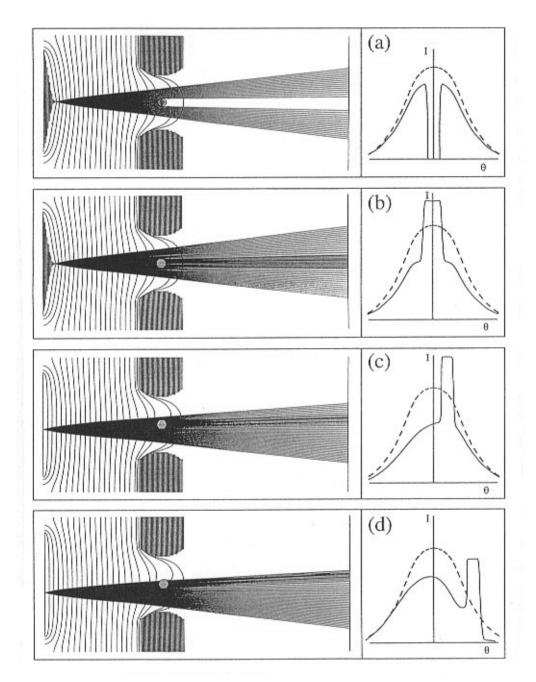


Figure 11. Simulation of the electron trajectories, electrostatic potentials and projected intensities along a line of the image (sketches to the right) in the projection microscope for (a) opaque and (b)-(d) non-opaque objects in the approximation of geometrical optics. Clearly, there is a bright intensity behind the nonopaque object. Several positions of the object with respect to the beam are simulated which are in agreement with observed images in Figure 8.

intensity behind the transparent object is evident for all relative beam-sample positions. Note that the goal of these preliminary simulations is limited to showing that the modification in the object transparency con-curs with the mutation from a wire like image to a slit-like image for the same tip-object geometry. Clearly, the wave property of the beam has not been considered.

The presence of fringes specific to diffraction from a narrow slit is undeniable in the high magnification im-ages (see, Figs. 7b and 7c). However, highly-resolved fringes are difficult to obtain because of the vibration of the nano-fibres

due either to external mechanical excita-tions or to electrostatic force interactions between the fibres and the nanotip. Another possible explanation for the lack of well-resolved fringes could be the non-uni-form phase shifts of the beam as it passes through an ob-ject of non-uniform thickness. A simple estimation shows that the electrons at such low energies can have appreciable phase shifts even when passing through a nano-meter-thick object.

Because the role of local electric field distribution becomes progressively more important as the dimensions of the fibres decrease, the bright diffraction patterns of narrow

fibres must then also reflect the local field distribution along the fibres and, in particular, the existing localised defects which can strongly modify this local field distribution. This is illustrated in Figure 9 which shows images of a network formed from PMMA. The fibres in the network are imaged as bright lines with localised structures along the fibres which we attribute to the presence of localised defects. The presence of defects and their influence in the imaging process is very well illustrated in Figures 9c and 9d which show localised periodic defects along a nano-fibre and a connection of two polymer fibres, respectively. Note, also, that the variations of the lateral magnification are different from those of the longitudinal magnification during the zooming process, due to the variation of the local field distribution and orientation along the fibres for different tip-sample distances and sample diameters.

The capability to probe local distributions of the electric field is also demonstrated by imaging a narrow carbon fibre with a finite length. Recent calculations have predicted periodic field distribution along nano-metric size carbon fibres (Devel *et al.*, 1996). Such a free standing carbon structure can be encountered as carbon prickle along some larger carbon fibres. Figure 10 shows the presence of such a nanostructure which appears as a slit-like image sticking to a more conventional wire like carbon fibre diffraction image. At high magnification, such a carbon prickle image shows longitudinal periodic structure with a periodicity of about 1.5 nm (Fig. 10d). Such a structure, in the high magnification image, may be a confirmation to the presence of the predicted periodic field distribution.

The interaction of a coherent radial beam and a narrow fibre is the main physical principle for the Möllenstedt biprism (Möllenstedt and Düker, 1954). Its configuration appears, at first glance, to be very similar to the projection geometry microscope imaging a fibre stretched across a small hole. However, our calculations show that the beam splitting and interference approach is not in agreement with the experimental results we obtained either with large or with small diameter fibres shown in the preceding figures.

Conclusions

In this article, we have reviewed previous results on opaque objects for which the main imaging mechanism is shown to be Fresnel diffraction, pointed out the important role of the source quality in image formation and shown new results for the projection imaging of non-opaque objects. Previously, we had shown that there is a good agreement between simulated FPM images and the experimental observations for simple opaque objects such as carbon or RNA fibres (Binh *et al.*, 1995, 1996). Here, we have shown a new type of image for very small diameter fibres that can be explained by taking into consideration the non-opaqueness

of the objects in concomitance with local electric field modifications of the electron trajectories in the object region. These results show that the FPM is a tool to probe field distributions with spatial resolution in the nanometer size. It is clear from the above discussions that a full quantitative explanation and exploitation of the images in the FPM will require extensive simulations of electron trajectories and diffraction patterns. New unsettled questions for the transmission of low energy electrons through nanometric fibres have been opened for analysis.

In considering object reconstruction from FPM im-ages, different options with phase recovery have recently been critically assessed by the Cavendish group (Bleloch *et al.*, 1996). They are based on either the super resolu-tion algorithm developed for STEM (Rodenburg and Bates, 1992), or the Kendogram inversion (Kreuzer *et al.*, 1992), or the k-vector averaging (Terminello *et al.*, 1993), or the Gabor in-line reconstruction (Gabor, 1948), or the modified Gerchberg-Saxton iteration (Gerchberg, 1972). The best prospects have been identified and a object reconstruction algorithm by iterative Fresnel propagation has been proposed (Bleloch *et al.*, 1996). Their calculations are currently in progress and they estimate that a resolution value of 0.1 nm can be attained by a super resolution reconstruction approach.

As a final note, the images show the existence of micrometer-length fibres with nanometer diameter. Their existence, and the fact that they can now be easily observed with the FPM, and their mechanical properties analysed, for example, via the amplitude of vibration and the frequencies, open new opportunities in nano-mechanics using the FPM.

Acknowledgements

This work has been supported by European Community Contracts (HCM and BRITE) and by French Government Agencies (DRET). It is a joint effort with Prof. N. Garcia. We also acknowledge discussions with Prof. Howie's group at Cambridge University (U.K.) about the object reconstruction mechanisms and their recent calculations. We acknowledge the technical assistance from the Service Central d'Analyse du CNRS-Département Instrumentation.

References

Vu Thien Binh (1988) *In-situ* fabrication and regeneration of microtips for scanning tunneling microscopy. J Microsc **151**: 355-361.

Vu Thien Binh, Garcia N (1991) Atomic metallic ion emission, field surface melting and scanning tunneling microscopy tips. J Phys (France) I 1: 605-612.

Vu Thien Binh, Semet V, Garcia N (1994) Low-energy-electron diffraction by nano-objects in projection microscopy without magnetic shielding. Appl Phys Lett **65**: 2493-2495.

Vu Thien Binh, Semet V, Garcia N (1995) Nano-metric observations at low-energy by fresnel projection microscopy: Carbon and polymer fibres. Ultramicroscopy **58**: 307-317.

Vu Thien Binh, Garcia N, Purcell ST (1996a) Electron field emission from atom-sources: Fabrication, properties and applications of nanotips. In: Advances in Imaging and Electron Physics, Vol. **95**. Academic Press, London. Chap. 2, pp. 63-153.

Vu Thien Binh, Semet V, Garcia N, Bitar L (1996b) Nearfield diffraction microscopy with a coherent low-energy ebeam: Fresnel projection microscope. In: Optics at the Nanometer Scale. Nieto-Vesperinas M Garcia N (eds.). NATO-ASI Series E, Vol. 319. Kluwer Acad Pub, Dordrecht, Netherlands. pp. 277-296.

Bleloch AL, Howie A, James EM (1997) Amplitude recovery in Fresnel projection microscopy. Appl. Surf. Sci. **111**: 180-184.

Bruhat G (1954) Optique (Optics). Masson et Cie, Paris, France. pp. 163-204.

Chen CJ (1993) Introduction to Scanning Tunneling Microscopy. Oxford Univ Press, New York. pp. 213-235.

Dahl DA, Delmore JE (1988) SIMION 4.0. EE&G Idaho Co, Idaho Falls, Idaho, USA.

Devel M, Girard C, Joachim C (1996) Computation of electrostatic fields in low-symmetry systems: Application to STM configuration. Phys Rev B **53**: 13159-13168.

Fink HW, Stocker W, Schmid H (1990) Holography with low-energy electrons. Phys Rev Lett **65**: 1204-1206.

Gabor D (1948) A new microscopic principle. Nature **161**: 777-778.

Gerchberg RW (1972) Holography without fringes in the electron microscope. Nature **240**: 404-406.

Kreuzer HJ, Nakamura K, Wierzbicki A, Fink HW, Schmid H (1992) Theory of the point source electron microscope. Ultramicroscopy **45**: 381-403.

Möllenstedt G, Düker H (1954) Fresnelsher Interferenzversuch mit einem Biprisma für Elektronenwellen (Fresnel interference of electron waves with a biprism). Naturwiss **42**: 41.

Morton GA, Ramberg EG (1939) Point projector electron microscope. Phys Rev **56**: 705.

Piquet A, Roux H, Vu Thien Binh, Uzan R, Drechsler M (1971) Réalisation et étude d'un microscope à ombre par émission de champ (Realisation and study of a field emission shadow microscope) Rev Phys Appl **6**: 105-109.

Rodenburg JM, Bates RHT (1992) The theory of super-resolution electron microscopy via Wigner-distribution deconvolution. Phil Trans Roy Soc (Lon) A **339**, 521-553.

Rose A (1948) Television camera tubes and the problem of vision. Adv Electronics Electron Phys 1: 131-166.

Seah MP, Dench WA (1979) Quantitative electron spectroscopy of surfaces: A standard data base for electron inelastic mean free paths in solids. Surf Interf Anal 1: 2-11.

Spence JCH, Qian W, Melmed AJ (1993) Experimental low-voltage point-projection microscopy and its possibilities. Ultramicroscopy **52**: 473-477.

Terminello LJ, Barton JJ, Lapiano-Smith DA (1993) Holographic imaging of atoms using multiple-wave-number electron angular distribution. Phys Rev Lett **70**: 599-602.

Discussions with Reviewers

M. Ichihawa: When you get FPM images, is there any problem about electric charging effect of polymer samples which are not so conductive? What is the effect of secondary electron emission from non-opaque objects on the local electric field in objects? The spatial variation of the secondary electron emission in objects may cause the change of the local electric field.

Authors: During the observations of polymer fibres, the images were stable in time. No fluctuation in the images due to charging effect was noticed in general during the irradiation.

We distinguish here two cases: (1) the charges due to the applied potential between the tip and the object, and (2) the additional charges induced by the electron beam impinging on the sample. In the second case, the electron beam induced charges may be due either to deposited charge directly from the beam or generated from secondary electron emission. Any effect that causes a change of the local electric field will lead to a local modification of the FPM images. This is illustrated, for example, in Figures 7, 9 and 10. For these examples, the local modifications are very stable under observation; it is then difficult to identify, among different mechanisms, the origin of these local electric fields. The relaxation of a localised charge created by the interaction between the e beam and the object must be considered in relation to the size of the fibres and the different possibilities of electron conduction through molecular configurations over the very small length scales of the illuminated areas. For example, quantum conductivity and the Coulomb blockade behaviours through nano-wires, must be considered. These notions are still under investigation and, therefore, it is very difficult to predict quantitatively the different behaviour for the polymer fibres. Within this field, the FPM is revealed to be a good candidate as a tool for nanowire conduction study.

U. Valdrè: As I understand, the magnification is measured by moving the specimen in the x,y plane by a known amount and by comparing it with the corresponding displacement on the fluorescent screen. What is the accuracy of the method and how do you take into account the fact that the image may protrude out of the x,y plane. Do you also use standard specimens, if so which one?

Authors: The displacement is done by the bending of a ceramic-piezo tube with a quadrant electrode system developed in STM. The accuracy of the calibration is directly

related to the accuracy of the piezoelectric ceramics (PZT) displacements which is a technology very well known in STM. It can be better than one angstrom.

This is not a 3D calibration. This calibration gives only information on the scale of the object dimensions projected on the x,y plane.

No, we have not calibrated the microscope with standard specimens until now.

U. Valdrè: It is unclear to me how the fringes observed with transmission specimens are produced, since the range of 200 eV electrons in carbon is of the same order of the fibre thickness. Could they not be due to the interference of the external beams deflected towards the fibre by local charging? Authors: As stated in the text, the mechanism of interference of the external beams deflected towards the fibre by the local field, which follows the physical principle of the Möllenstedt biprism, has been discarded because the interference results obtained by a simulation of biprism are not in agreement with the observed experimental images. The sucking-in-effect approach we proposed in this article shows no disagreement between predicted and experimental results. However, the first approach taken here needs to be extended by taking into account the interaction between the beam and the object, as well as the effect of phase changes for the electrons passing through the wire on the diffraction process.

U. Valdrè: Would you explain what you mean by "localised defects". Do you include insulating, spurious nanoparticles which charge up?

Authors: We have observed many types of nanometric scale defects and objects with the FPM, some of which give large local distortions of the image that appear to be charged regions. Examples of such localised defects are given in Figure 9 where there are apparent breaks in an otherwise straight wire. Yes, we included such nano-particles.

U. Valdrè: Presumably, "total" beam current means the current extracted from the tip. It would be of interest to know the value of the current flowing through the extraction electrode and/or through the specimen.

Authors: The electron source used is a single nanotip, the current extracted from the tip is then only the field emission current from the apex of the nanotip. This is the current that flows through the extraction electrode and corresponds to the current of the whole spot on the image. The specimen current is too low to provide significant data by a direct measurement. At first approximation, it can be estimated from the image by considering the area ratio. However, there will be a significant error in this estimation due to the sucking-in effect at high magnification.

U. Valdrè: What is the cause of the elliptical illuminated field

shown in Figure 3?

Authors: This happens when the nanotip axis is not perpendicular to the object plane.

U. Valdrè: Please give details of the pneumatic anti-vibration system you use.

Authors: It is a commercial system (Stabilizer Vibration Isolators PL-2000; Micro-Controle Newport Co., Irvine, CA, USA) used in most of the optical tables, with a cutting frequency in the range of few hertz. The whole microscope ensemble is then vibration isolated.

U. Valdrè: How do you prepare the PMMA films?

Authors: We use a two-step procedure: (1) dissolution of the PMMA in chloroform at a concentration of a 10 mg/l; and (2) deposition of a drop (\sim 2 μ l) of this solution on a holey carbon grid. After evaporation of the solvent, the probability of having the polymer stretching across a hole is large.

However, the dissolution concentration given here is only an indication because during the evaporation of the solvent the concentration in the drop will increase by a factor that is not controlled by this preparation technique. Other techniques for specimen preparation can be proposed and adapted to the particular systems to be studied.