Towards Realization of an Atomic de Broglie Microscope: Helium Atom Focusing Using Fresnel Zone Plates

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A neutral, ground-state, ^{4}He atom beam has been focused using Fresnel zone plates. At only moderate demagnification (0.40×), a focused spot diameter of $\leq 2.0\ \mu$m is achieved at a signal intensity of 500 counts/s. This is an improvement over previous work by a factor of 10 in resolution, 10^3 in signal intensity, and 10^8 in focused beam density, and allows the full intrinsic intensity profile of a focused atomic Fresnel spot to be measured for the first time. Raster scans of 15 and 25 μm wide slits across the focused spot demonstrate transmission scanning helium de Broglie microscopy of micron resolution.

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Essentially all focusing microscopy employs either photons or electrons as the means whereby an object is imaged. This is largely due to the ease with which these two species can be focused, either to demagnify an incident beam (raster microscopy) or to magnify a transmitted or scattered beam (imaging microscopy). Apart from this, a helium atom might well be the preferred particle by virtue of attributes which have made ^{4}He scattering an indispensable surface science tool [1]. Specifically, a ground-state thermal energy helium atom (5–100 meV) is completely nonpenetrating, chemically inert, and produces essentially no mechanical damage. Intense, highly monochromatic beams are easily generated via supersonic free-jet expansion. Helium atoms are well matched in momentum and energy to the reciprocal lattice and vibrations of a single-crystal surface, offering both elastic scattering (specular, diffraction) and inelastic scattering (single phonon excitation/deexcitation) as possible microscopy contrast mechanisms. The de Broglie wavelength of a thermal energy helium atom is small, at most a few angstrom, providing much better diffraction-limited resolution than that of optical microscopy or of electron microscopy at comparable incident energies. Therefore microscopy based on helium atoms—helium atom de Broglie microscopy—potentially offers substantial improvements in surface sensitivity, spatial resolution, contrast mechanisms, and reduced sample damage.

Focusing of a neutral atomic beam is difficult, however, and while molecular de Broglie microscopy has been demonstrated [2], this was done through aperturing rather than focusing. The very low resultant intensities make aperturing a doubtful approach for general de Broglie microscopy. Focusing is particularly challenging with ^{4}He atoms, for which low polarizability and lack of spin preclude standard techniques [3] of electromagnetic focusing. One approach has been to focus ^{4}He beams via reflection from a perfect, monolithic, bent single-crystal mirror [4], and focal spots as small as 210 μm have been attained in this fashion [5]. Bent-crystal mirrors offer both achromatic focusing and large numerical aperture, but bending a crystal surface accurately on atomic length scales remains problematic, and maintaining the mirror surface atomically clean for long periods is difficult.

These complications are avoided with free-standing Fresnel zone plates, which deflect and focus via diffraction through a concentric grating of interspaced open and opaque annular rings [6]. Zone plates provide a universal focusing technique, applicable with even reactive, fragile [7], or electronically excited species [8]. This is understood, in a classical picture, by virtue of the beam particles never physically contacting the zone plate structure as they pass through the open annuli. Focusing nonetheless takes place, a purely quantum mechanical effect and a direct manifestation of the wave nature of the atom. Seminal zone plate focusing of atoms was carried out by Carnal *et al.* [8] using electronically excited metastable ^{4}He* and attaining a spot size of 15–20 μm at about 0.5 counts/s. The current work employed custom-made zone plates to focus a neutral, ground-state ^{4}He beam for the first time. Advanced beam techniques, notably the use of microskimmers [9], improved the spatial resolution by a factor of 10, the signal intensity by 10^3, and the focused beam density by 10^8 relative to previous work. This made it possible to measure the intrinsic profile of a focused atomic Fresnel spot for the first time. Raster scans of the focused beam across a slit demonstrated the first true scanning de Broglie microscopy.

The limiting resolution of a zone plate is set by its outermost zone width, 100 nm for the state-of-the-art, free-standing plates used in this work [10]. Scanning electron micrographs of one such zone plate are presented in Fig. 1. The beam apparatus, depicted schematically in Fig. 1, is similar to equipment described in the literature [11].

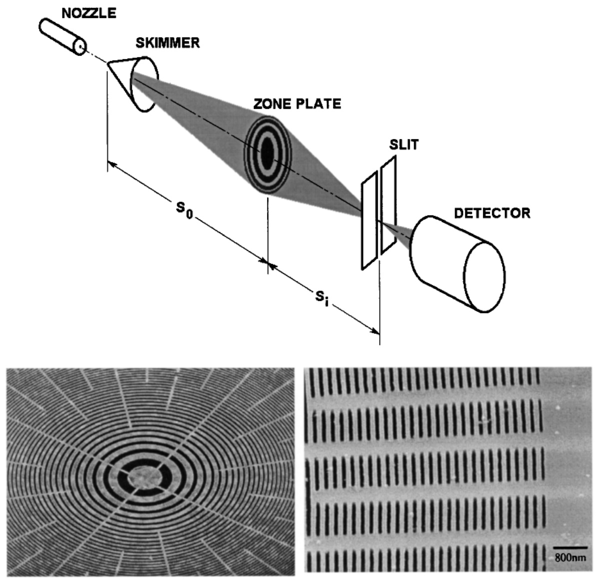

FIG. 1. Top: Schematic depiction of apparatus. Distance from skimmer apex to zone plate was $S_0 = 1.069$ m and from zone plate to detector slit, $S_i = 0.430$ m. Detector slit was 15 or 25 μm wide by 5 mm high. Bottom: Scanning electron microscope micrographs of the zone plates, showing the free-standing annular rings and radial array of support struts. The 0.27 mm diameter zone plate pattern had a design focal length of 150 mm for atoms of 1.80 Å, yielding 307 mm at 0.88 Å. Bottom left: Enlarged view of zone plate center. Opaque central Fresnel zone is 10.4 μm in diameter. Bottom right: Enlarged view of zone plate periphery. Outermost Fresnel zone (smallest, outermost annular slit) is 100 nm wide.

The beam was generated via supersonic free-jet expansion through a 5 μm diameter nozzle, the nozzle temperature T_0 being adjustable from 4 to 300 K and the pressure P_0 from 0 to 150 bars. Measurements were made at various beam velocity distributions, generally $\Delta v/v \sim 1\%$ FWHM but ranging up to $\sim 10\%$. An electron bombardment ionization mass spectrometer (magnetic sector field) detected the helium atoms. Crucial to the focusing measurements were the high absolute positioning accuracy of the detector (well under 1 μm) and a correspondingly low level of apparatus vibrations.

Within a free-jet expansion, the true source of molecular beam "rays" lies far downstream of the nozzle, in a region in which the expanding jet undergoes transition from continuum to free-molecular flow. This transition region typically has an intrinsic diameter of several hundred microns, which is far too large to critically test zone plate focusing. Accordingly glass microskimmers [12], ranging in diameter from 1 to 14 μm, were used to provide a more nearly pointlike beam source. These skimmers are fabricated by carefully drawing class capillary tubing to rupture at a fine neck [9].

Atom diffraction from a linear grating of period a can be described semiclassically as the transfer to the atom of one or more quanta $2\pi/a$ of transverse momentum. Approximating the zone plate locally as a linear grating, this momentum transfer is radially inward or outward and three primary channels are anticipated: (i) momentum transfer inward, deflecting atoms towards the beam axis and thus onto converging trajectories, (ii) no momentum transfer and therefore no deflection of the atom, and (iii) momentum transfer outward, deflecting atoms onto diverging trajectories. First order diffraction dominates, and the zone plate thus separates the transmitted beam into three main channels, the focused $(+1)$ channel, the undeflected (0) channel, and the "defocused" (-1) channel.

A measured transverse intensity distribution of a focused helium beam spot is shown in Fig. 2. Notice that the ordinate is logarithmic. The main features are (i) a narrow central maximum of 1000 counts/s and 25 μm width, (ii) a broad underlying plateau of 250 counts/s and 380 μm width, and (iii) a still broader and lower plateau of 50 counts/s and 760 μm width. These are signatures of the $(+1)$, (0), (-1) channels, respectively. The widths and intensities of these features are in good agreement with both exact quantum mechanical calculations and with simple ray tracing based on the 0.27 mm outer diameter of the zone plate. To characterize focusing, it is the narrow, central $(+1)$ peak which is of interest. This contains the demagnified image of the microskimmer aperture convoluted with the detector slit function plus any apparatus smearing (e.g., vibration).

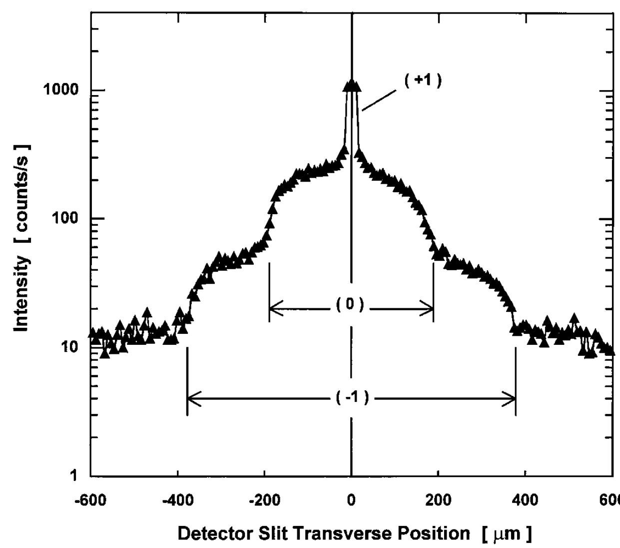

FIG. 2. Transverse scan of detector slit in steps of 6.9 μm through the focused He atom beam. The nozzle temperature was $T_0 = 124$ K to yield a de Broglie wavelength $\lambda = 0.88$ Å, giving a zone plate focal length of 307 mm. Under these conditions the 4 μm diameter beam skimmer should be optimally focused onto the 25 μm detector slit. The central peak is due to the focused $(+1)$ diffraction channel. Underlying plateaus corresponding to the undiffracted (0) channel and defocused (-1) channel are identified on the basis of the expected widths of these features, as marked.

Beam focusing was explicitly verified by varying the beam wavelength to alter the focal length of the zone plate. This changes the image distance S_i, shifting the apex of the converging cone of $(+1)$ diffraction rays to a point upstream or downstream of the detector slit plane and thereby increasing the width of the $(+1)$ peak measured in that plane. Such $(+1)$ peak widths are plotted (triangular points) in Fig. 3. The anticipated broadening with wavelength is readily apparent, outside of a region near optimum focusing at $\lambda = 0.88$ Å. Here the focused spot diameter becomes appreciably smaller than the 15 μm detector slit and the $(+1)$ peak width "bottoms out" at the slit width.

When the beam spot size is much smaller than the detector slit width, a high resolution transverse scan through the beam spot yields a trapezoidal profile for the $(+1)$ peak as illustrated in Fig. 4. Information on the spot size is then contained in the intensity rise at the edge of the $(+1)$ peak and can be extracted by fitting the peak profile. This was done by treating the focused spot as a circular disk of uniform intensity, convoluting this with a slit detector function and taking as free parameters the peak intensity, the background intensity, the spot diameter, and the slit width. An optimum fit for this scan, plotted in Fig. 4 as a solid line curve, yields a spot diameter of 4.6 ± 1.0 μm. Since this 4.6 μm beam spot is much smaller than the 25.2 μm slit width, Fig. 4 constitutes a true raster helium atom microscope scan of the slit.

Similar high resolution transverse scans were carried out under various nozzle conditions for skimmers of 1, 4, and 14 μm diameter. Values of the beam spot diameter at optimum focus, extracted from fits to the $(+1)$ profile as discussed above, are tabulated in Table I along with the "expected" diameter (0.40× of the skimmer diameter). Also listed are the focused beam intensity—equal to the $(+1)$ peak intensity less the (0), (-1), and background intensities—and the corresponding nozzle pressure P_0. Focused spot diameters extracted in this manner for a 1 μm skimmer are plotted as square points in Fig. 3.

The measured spot diameters of Table I contain the actual beam diameter plus instrumental broadening due to zone plate imperfections, uncertainties in skimmer diameter, apparatus vibrational smearing, and detector slit imperfection. The diffraction-limited resolution of the zone plate is nominally 100 nm and should not be a factor. Chromatic dispersion of the beam velocity distribution should also play only a minor role, given that a slit detector is used [13]. A known imperfection is waviness of the slit edge, which scanning electron micrographs show to be about 0.5 μm in amplitude over micron length scales.

The trends in spot diameter versus nominal skimmer diameter are suggestive and currently interpreted as follows: About 1 μm of the measured spot diameter arises through apparatus smearing, specifically some 500 nm each due to apparatus vibration and slit edge waviness. The remaining diameter is a true, demagnified source image. This is larger than the demagnified skimmer diameter because the

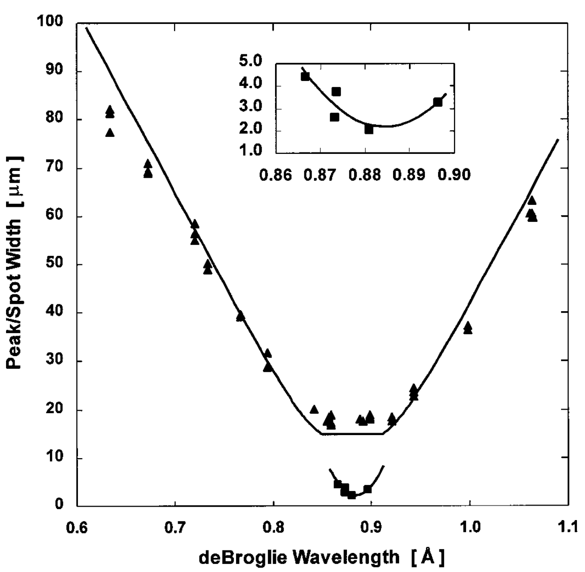

FIG. 3. Zone plate focusing of a helium beam as demonstrated by measuring the $(+1)$ peak width (FWHM) as a function of the helium beam wavelength. Triangular points are data recorded with a 14 μm diameter skimmer and a 15 μm detector slit. The solid line curve through these points is a fit based on simple modeling which treats the beam spot as a disk of uniform intensity shuttered with the detector slit. Once the beam is focused to a spot diameter smaller than the detector slit, the peak width no longer decreases (flat-bottomed portion of curves near optimum focusing at 0.88 Å). In this regime the spot diameter can be extracted from the edge rise of the transverse peak profile. The beam spot diameter measured in this fashion for a 1 μm skimmer is marked with square points in the main figure and also, enlarged, in the inset. The solid line curve through these square points is a quadratic least squares fit.

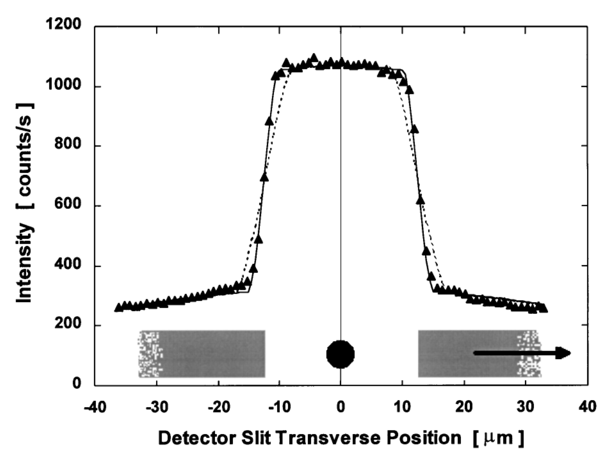

FIG. 4. High resolution transverse scan through the $(+1)$ diffraction peak for a 0.87 Å wavelength beam, using a 4 μm skimmer and scanning the 25 μm detector slit in steps of 0.91 μm. The flat, broad, trapezoidal shape of the peak shows conclusively that the focused beam is much smaller than the 25 μm slit, as indicated schematically at the bottom of the figure. The solid line curve is a calculated intensity distribution for a 4.6 μm diameter disk of uniform intensity shuttered by a 25 μm slit. Dashed line, to gauge the sensitivity of the modeling, is the corresponding curve for twice this diameter.

TABLE I. Measured beam spot size and intensity at optimum focus.

Skimmer Dia. (μm)	Focused Spot Dia. (μm) Expected	Measured	I_{peak} (counts/s)	P_0 (bars)
1	0.4	2.0	530	140
4	1.6	4.6	420	50
14	5.6	6.2	1260	40

free-jet expansion continues through the skimmer opening. The transition to free-molecular flow thus occurs downstream of the skimmer opening, and the size of this transition zone, rather than the skimmer opening, determines the true source size. At sufficiently small skimmer diameter the measured spot diameter must then approach the demagnified skimmer size, as appears to transpire below about 2 μm. Given this interpretation, a moderate increase in demagnification should yield a submicron spot diameter.

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[13] Even an incident beam of broad wavelength distribution yields a radial spot intensity profile $I(r)$ in the detector plane, the peak intensity corresponding to that wavelength λ_0 which is in optimum focus and the tails being comprised of wavelengths which are progressively further removed from λ_0. Provided the detector is approximately 0D (small aperture) or 1D (narrow slit), a profile will thus be measured as the detector is rastered through the beam. Such a broadband source would be of no use for microscopy on a 2D array of transmitting/scattering centers, however. Relatedly, a small aperture passes only a narrow range of velocities centered around the focused velocity, monochromatizing the transmitted beam.