Pinhole Neutral Atom Microscopy

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Pinhole Neutral Atom Microscopy

by

Philip James Witham

A thesis submitted in partial fulfillment of the requirements for the degree of

Master of Science
in
Physics

Thesis Committee:
Erik Sánchez, Chair
John Freeouf
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Portland State University
2013
Abstract

This work presents a new form of microscopy, the instrument constructed to demonstrate it, the images produced and the image contrast mechanisms seen for the first time. Some of its future scientific potential is described and finally, recent work towards advancing the method is discussed.

Many forms of microscopy exist, each with unique advantages. Of several broad categories that they could be grouped into, those that use particle beams have proven very generally useful for micro and nano-scale imaging, including Scanning Electron, Transmission Electron, and Ion Beam microscopes. These have the disadvantage, however, of implanting electric charges into the sample, and usually at very high energy relative to the binding energy of molecules. For most materials this modifies the sample at a small scale and as we work increasingly towards the nano-scale, this is a serious problem.

The Neutral Atom Microscope (NAM) uses a beam of thermal energy (under 70 meV) non-charged atoms or molecules to probe an atomic surface. For several decades scientists have been interested in this possibility, using a focused beam. Scattering of neutral atoms provides a uniquely low-energy, surface-sensitive probe, as is known from molecular beam experiments.

We have developed a new approach, operating with the sample at a close working distance from an aperture, the need for optics to focus the beam is obviated. The demonstrated, practical performance of this “Pinhole” NAM exceeds all other attempts by great lengths by many measures. The unique images resulting and contrast mechanism discoveries are described. The future potential for nano-scale resolution is shown.
Acknowledgments

I'd like to thank to Professor Erik Sánchez for the use of his lab, vacuum equipment and advice. Without that I could not have done this wonderful project. And many thanks to Sean King and Intel corporation for support. Also Everett Lapp for donated RGAs, pumps and vacuum hardware, Professor Rolf Könenkamp for his reviews and insights, Greg Batty and the PSU microscopy center for sputter coater and FIB training, Rich Swinford for image processing expertise, Mike DeArmond for FIB help and help in general, and Dr. Derek Nowak for his LabVIEW™ expertise and paper review, and Portland State University for the use of their facilities. And not the least I'd like to thank George Schmermund for getting me into SEMs.
Preface

This project started out for me as a chance to finally do physics of my own devising, which I needed to demonstrate for the good of a career in physics. But the lucky success of the idea gave it a life of its own, based on it simply being a really interesting new thing, that really should be explored, for all the best reasons we do science, where we don't know where it will lead. I've been fortunate to have the chance to do this at PSU and to play the advocate for it. I set out to test the idea theoretically simply because I had never heard of it being done by any means, and thought I had a way to do it. After some basic math showed it should work I then did a literature search and was surprised that although many had tried other approaches for years, no one had tried this simplest method. So I was fortunate that people had assumed it required a difficult approach. This is the most important lesson I take from this project, that if the math says that a simple approach should work, you should try it. It'll be great to see where it goes.

Philip Witham

May 2013
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1 Introduction

A neutral atom beam created by room-temperature gas expansion into a vacuum has an energy of roughly 0.07 eV, and this energy is usually insufficient to penetrate a single atomic layer. Despite this low energy, the high mass results in a de Broglie Wavelength under 0.1 nm. This wavelength is, \( \lambda = \frac{h}{m_0 V} \), where \( h \) is Planck's constant, \( m_0 \) is the particle mass and \( V \) the velocity. Helium is one option, with a mass \( \approx 4 \) Atomic Mass Units. Expanding from high pressure at room temperature, the He molecules will reach an average velocity of approximately 1.7 km/s, thus \( \lambda \) equals 0.6 Angstrom, smaller than an atom width. As a result, the low energy of these atoms does not limit potential resolution. Magnetized samples can be imaged, and non-conductive samples without coating. Helium has no spin or chemical interaction with samples. With a surface binding energy of far less than the room-temperature thermal energy, helium atoms generally scatter from the sample without adsorption. The possibility exists therefore to image the very surface layer of a sample, including any water or contamination layer, and extremely fragile structures without damage. Operating in reflection mode means that samples need not be sectioned as for TEM.

Helium at this energy has a strong scattering interaction with surface hydrogens, which have a great importance to surface chemistry including catalysts and metallurgy.\(^2\,^3\) Since the binding energy of surface hydrogens can be as low as 0.1 eV, a NAM should be capable of performing interesting surface science on a hydrogen layer.
The first images from a neutral atom beam microscope was achieved in 2007\textsuperscript{1} by a group at the University of Bergen in Norway after years of work by a number of labs and investigators. The transmission mode silhouettes below were the only published images from the “Atomic de Broglie Microscope” prior to our publishing in 2011. This microscope used a micro-machined silicon zone plate to produce a focused beam spot. We have found no published reflection mode images prior to the present work.

![Figure 1.1 a,b, TEM grid images, 2 um resolution, M. Koch, S. Rehbein, G. Schmahl, T. Reisinger, G. Bracco, W. E. Ernst, and B. Holst, 2007\textsuperscript{1}](image1)

![Figure 1.2 a,b, TEM grid images demonstrating 0.8 um resolution, S Eder, T Reisinger, M Greve1, G Bracco and B Holst, Focusing of a neutral helium beam below one micron, 2012\textsuperscript{39}](image2)
2. **Review of Literature: Previous attempts and molecular beams**

2.1 **Focusing methods**

In comparison to charged particles, it is difficult to focus particles that have no net charge. Some of the methods that have been successfully used or proposed include those summarized in table 1 and the references. Simultaneously obtaining a high beam intensity and a small spot size is required in order to realize a useful microscope. Many focusing methods have been tried.

<table>
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<th>Table 1</th>
<th>Some focusing methods and notes.</th>
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<tr>
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<td>1.5</td>
<td>11, 12</td>
<td></td>
</tr>
</tbody>
</table>

*Information not given

The best published 2D image resolution obtained by focusing methods is 0.8 micrometers. Low resolution molecular scanners were reported previously, but using effusive samples rather than an atom beam²¹,²².
In both figures 2.1 and 2.2, the methods tested by researchers used a gradient of electric or magnetic fields to produce a weak force vector pointing away from the region.
of higher field intensity. This weak force is only effective for atoms of low velocity, i.e., ultra-cold atoms, such as can be obtained by laser cooling. As a result, so far, only a very low beam intensity has been achieved. Other structures using fields produced by laser illumination have been studied theoretically for the purpose of producing a lens.

![Diagram of curved atom mirror](image)

(a) An ellipsoidal Cartesian surface focuses each ray coming from the object point into an image point. (b) The coordinate system for the ellipsoidal mirror \((x, y, z)\) and for the image plane \((x', y', z')\). The origin of the mirror coordinate system is the point of reflection of the central ray on the mirror surface.

Figure 2.3, curved atom mirror formed by electrostatically bending a thin hydrogen-passivated Si(111)-(1 × 1) crystal sheet.\(^9,10\)

The method illustrated in Figure 2.3 was used to greater effect but still suffered a poor focus. Due to the extreme sensitivity of helium scattering to the atomic-scale
“surface” flatness, only ~1% of the incoming atoms reflect specularly (fig. 2.4 illustrates the meaning of specular and diffuse).

Figure 2.4, Gas scattering is mainly diffuse, not specular. [Cavendish Lab Surface Physics Group]³

Using a silicon surface bump hologram, the method of figure 2.5 was able to produce an arbitrary image pattern on a target, but with only a small fraction of the intercepted atoms reflecting into the desired image. Resolution was again poor.

Figure 2.5, Atomic reflection hologram ⁴
Fresnel zone plates for helium focusing were built through a collaboration of several labs and incorporated at Bergen into the first successful neutral helium microscope, mentioned previously. This produced a usable beam intensity and (to date) 0.8 µm resolution.

Figure 2.6, Fresnel zone plate lens. A small fraction of the incoming atoms are diffracted into the focal point.

Figure 2.7, Fresnel zone plate lens in silicon.
A “Quantum Stabilized” Pb surface mirror was developed\textsuperscript{12}, claimed to be one of the flattest mirrors ever made, with just a few surface atoms out of place. This achieved a helium (specular) reflectivity of $\sim 15\%$. It was found to be easily destroyed by contamination on exposure to air.

Recently (2011)\textsuperscript{32} it was shown that a graphene monolayer grown on Ru(0001) can be a better mirror for thermal heliums, achieving 20\% reflectivity. This mirror was also shown to be fairly unaffected by exposure to air.

By no means have all possible methods of focusing been tried, and one would expect that over time improvements will be made. However, calculations indicate that the only advantage of focusing is ultimately that the working distance between the sample and microscope components can be made much longer in the case of a focused system. (see section 3, Pinhole Neutral Atom Microscope concept.) That is potentially a significant advantage.

Figure 2.8 a,b, Helium Atom Scattering mechanism\textsuperscript{3}
(Cavendish Lab Surface Physics Group)

Thermal energy helium typically scatters elastically by long-range van der Wals forces.

“Trajectory 1 - elastic scattering, producing a diffraction pattern for surfaces of atomic periodicity;

trajectory 2 - inelastic scattering, involving energy exchange with a surface or adsorbate vibration;

trajectory 3 - elastic scattering into a resonant state;

trajectory 4 - accommodation or adsorption onto the surface, with the impinging atom becoming trapped near the bottom of the potential well. Dotted black lines illustrate the variation in potential corrugation with distance from the surface.”\textsuperscript{3}
2.2 Molecular beam sources

The most common source for creating a molecular beam is a “Free Jet” nozzle, admitting gas into a vacuum chamber. (fig. 2.9).

Free jets and beam formation are covered well in the literature.\textsuperscript{16-20} Gas is admitted to a vacuum through a small opening. The gas exits at Mach 1 velocity (for that gas at the local temperature) with roughly half of its original pressure, since it cannot obtain a higher velocity than this in a converging space. It then expands adiabatically into the vacuum, further accelerating, until the pressure has dropped to the point where scattering events between gas molecules are no longer likely. The imaginary more-or-less spherical surface within which the last usual gas-gas scattering events happen is called the "quitting surface". This is the apparent source illumination area from which all beam atoms can be traced (note, however, it is just a useful approximation.) From this point on, if the ambient pressure is kept low, the flow is free-molecular.

One then forms a beam by blocking off entrance to an adjacent vacuum chamber except through a small aperture (see fig. 3.3.) Often, molecular beam experimenters use additional "skimmer" apertures, chambers and vacuum pumps to improve beam quality.
A skimmer is ideally a narrow cone shape pointing upstream into the flow. The purpose of this shape is to obtain the highest beam quality by minimizing the back-scattering of gas into the beam path.

Formulae for adiabatic expansion found in Miller\textsuperscript{24} give us the Mach 1 limited conditions at the smallest point in the nozzle. First we calculate the throat pressure, $P_1$ (in Pa),

$$P_1 = P_0 \left( \frac{\gamma - 1}{2} + 1 \right)^{\frac{1}{1 - \gamma}},$$  \hspace{1cm} (1)

given the inlet pressure $P_0$ and $\gamma$, the gas ratio of specific heats (1.660 for monatomic gas). The throat temperature $T_1$ (in K) is calculated,

$$T_1 = T_0 \left( \frac{P_1}{P_0} \right)^{\frac{\gamma - 1}{\gamma}},$$  \hspace{1cm} (2)

derived from $T_0$, the inlet temperature. $v_1$, the throat average axial velocity (in m/s) is calculated,

$$v_1 = \sqrt{\frac{\gamma k T_1}{m_h}},$$  \hspace{1cm} (3)

using the Boltzmann constant $k$ (1.381×10^{-23} J/K) and helium mass $m_h$ (kg). Next, calculate the throat area $A_1$ (m$^2$),

$$A_1 = \frac{\pi D_1^2}{4},$$  \hspace{1cm} (4)

given its internal diameter $D_1$ (in m). From this the atom flow rate, $n$ (atoms/s) can be calculated,

$$n = \frac{P_1 A_1 v_1}{k T_1}.$$  \hspace{1cm} (5)
The actual flow rate of the source is reduced somewhat by viscous effects that depend on the internal size and shape of the nozzle, and these calculations do not predict that. Miller\textsuperscript{17} recommends correcting for this by using a measured flow rate and calculating back to an effective value for $D_1$. In that case, it would be necessary to use the corrected $D_1$ value consistently (in equations 4, 7, 9).

The ultimate axial gas velocity after full expansion into the vacuum, $v_{\text{inf}}$, is,

$$v_{\text{inf}} = \sqrt{\frac{2k}{m_h} \left( \frac{\gamma}{\gamma - 1} \right) T_0}.$$  \hspace{1cm} (6)

For example, with helium at 300 K temperature and 5.5 MPa (800 psia) pressure, and a 2 $\mu$m diameter nozzle, about $2.4 \times 10^{18}$ atoms per second exit into the source vacuum chamber at 882 m/s axial velocity, 226 K temperature, 2.7 MPa pressure, and $v_{\text{inf}}$ is 1770 m/s.

Next, the point where the pressure has dropped to where the mean free path prevents further collisions can be taken as the position of the quitting surface. Using the continuum approximation for the expansion we can obtain a function of Mach number versus distance from the nozzle exit, $M(x)$. Setting the ultimate Mach number $M_{\text{inf}} = M(x)$ we can determine $x$. Following Miller\textsuperscript{17} the formula below combines many details and experimental fits to estimate $S_{\text{inf}}$, the speed ratio for the gas after expansion (the mean velocity divided by the thermal spread of velocities). Helium is assumed from here on.

$$S_{\text{inf}} = 0.778 \left[ P_0 D_1 \left( 9.57 \times 10^4 \frac{s^2}{kg} \left( \frac{K}{T_0} \right)^{4/3} \right) \right]^{0.495},$$ \hspace{1cm} (7)

and from this we calculate the Mach number, $M_{\text{inf}}$. 

\[ M_{\text{inf}} = \sqrt{\frac{2}{\gamma}} S_{\text{inf}} \] \hfill (8)

Next calculate the position \( x \) from the nozzle exit where the quitting surface has been reached. To be conservative we can assume that \( x \) is equal in all directions (spherical approximation), maximizing the estimate of the quitting surface diameter, \( D_2 \) (\( D_2 = 2x \)),

\[ D_2 = 2D_1 \left( \frac{M_{\text{inf}}}{3.232} \right)^{\frac{1}{\gamma-1}} \] \hfill (9)

In our example the results are \( S_{\text{inf}} = 17 \), \( M_{\text{inf}} = 19 \), and \( D_2 = 58 \ \mu m \).

\( D_2 \) is used in section 3 below to calculate spot size.

Beam energy is straightforward, and equals 65 meV in our example.

\[ e_b = \frac{m_h v_{\text{inf}}^2}{2} \] \hfill (10)

This energy is largely driven by the source gas and nozzle temperature.

An experimental value for the centerline intensity downstream of a free jet is somewhat more than would be expected from a spherical expansion assumption, as detailed in Miller\textsuperscript{17}. This is due to a concentration along the nozzle axis. It is empirically calibrated with a constant "\( K_h \)" , equal to 2.0, and thus \( I_0 \) (in \( \text{atoms/s} \times \text{sr} \)) can be developed from \( n \) above,

\[ I_0 = K_h \frac{n}{\pi S r} \] \hfill (11)

\( I_0 \) is on the order of \( 1.5 \times 10^{18} \ \text{atoms/s} \times \text{sr} \) for the example conditions.

Of note here is the very effective “cooling” of the gas flow indicated by the speed ratio. By operating at a high source pressure, a simple leak into a vacuum (“free jet
nozzle”) produces gas flow with very little energy in the random motion of the atoms, the energy is mainly axial. This also means that the flow is fairly monochromatic. The later part of this expansion/cooling process, beyond the quitting surface, is termed “geometric cooling”.

The most highly monochromatic molecular beam sources use higher source pressures than the 5.5 MPa (800 psia) pressure of our example. This is done by using a boost bump to raise the pressure above the gas bottle pressure.

Deuterium has been shown to be an interesting alternative gas to helium. Source intensities of 2-3x greater than helium sources have been shown. Other gasses can be used, with little or no change in the source design. Gasses with deeply cryogenic boiling temperatures are most interesting as these do not easily form dimers or clusters during expansion from room temperature. Helium does not forms dimers until below ~20K. This is one reason the noble gas series are all of interest.

Figure 2.10 a,b, SEM images of free jet source nozzles constructed by laser heating and pulling of a silica micro-capillary tube.
If one desires a beam with lower energy, all that is necessary is to cool the source gas and nozzle, for instance by use of a liquid nitrogen cold finger in contact with an insulated source nozzle assembly. Beam energy is linearly proportional to the absolute temperature of the gas entering the nozzle. This makes obtaining helium beam energies of ~17 meV straightforward, and lower energies possible.

The effect of the shape of the nozzle downstream of the throat is not clear from the literature. At higher pressures and larger scales the use of a de Laval (converging-diverging) nozzle shape would clearly be ideal as in rocket exhaust nozzles. This has been mentioned elsewhere.\textsuperscript{23,29} The goal here being better collimation of the flow, leading to higher center-line intensity. The expanding section continues expansion through supersonic velocities with the angle of the surface being within the Mach cone for that local velocity. The author expects that this remains true at the micro-scale and lower exit pressures until some limiting pressure where scattering interactions with the wall have a more negative effect than the beneficial effect of (increasingly rare) gas-gas scattering. Finding a method to construct an ideal nozzle exit shape has the potential to increase the source center-line intensity by a large factor. A simple cone shape has been shown\textsuperscript{40} to increase the centerline intensity by 20x.
3. **Pinhole Neutral Atom Microscope concept**

The approach presented in this work is analogous to pinhole optics rather than refractive or reflective (focusing) optics. A beam of gas particles is formed by a source nozzle and an aperture, operating in high vacuum and free molecular flow (fig. 3.1). The beam is directed against a sample located close to the aperture, where the beam has not yet diverged far.

The sample (or aperture) is mechanically scanned in two dimensions. A mass filtered detector produces the image (pixel) intensity signal from a gas partial pressure. In reflection mode, contrast is produced *at least* by differential scattering due to topography. For instance with the detector optimized for sensing those gas atoms scattered toward one side of the sample, a sample area sloped towards the detector would generally be "brighter". Transmission mode could also be used, but was not the goal of this project.

![Conceptual diagram of a pinhole NAM (not to scale).](image)
Fundamental questions for this concept include: How small is the beam spot on the sample? (which determines resolution), and what is the intensity of the beam on that spot? (which affects how fast image pixels can be collected.)

Given the estimate for the source quitting surface diameter (eq. 9) and the other parameters for the beam geometry (Fig. 3.3), the approximate spot size can be calculated by,

\[
D_{\text{Spot}} = D_2 \frac{WD}{L_{\text{Sep}}} + D_3 \frac{WD + L_{\text{Sep}}}{L_{\text{Sep}}}.
\]  

(12)
Where $D_2$ is determined using equations (1-9). The calculated spot size for the current microscope, at the ~25 µm working distance of the nearest sample areas, is ~0.25 µm. This matches closely the estimated resolution of the images.

From the aperture diameter $D_3$ and source to aperture distance $L_{sep}$, and $I_o$ from (eq.11) the beam flow rate can be predicted,

$$n_{beam} = I_o \frac{\pi}{4} \left( \frac{D_3}{L_{sep}} \right)^2 \text{sr}.$$  \hspace{1cm} (13)

Alternate system configurations can then be compared for spot size and intensity ($n_{beam}$). Examples of optimized configurations determined this way follow in section 6.

For the configuration used to capture recent images figures such as figure 5.1, calculated $n_{beam}$ was approximately $10^{10}$ atoms/s. He partial pressure measurements roughly agree with this. In SEM or FIB terms, multiplying this by the electron charge $q$ gives an equivalence to a probe current of 1.6 nA.

This intensity appears to be quite high, though a direct comparison with focusing efforts is difficult. The lack of any loss of the beam by a mirror or lens is only one factor in this. Mirror experiments by D. Barredo et al \textsuperscript{11,12} indicated a count rate (presumably detector counts) of somewhat under $10^7$ per second, but it is difficult to relate this to the atom intensity at a given spot size. One would presume that in time focusing methods can achieve higher intensities than pinhole optics, but this does not currently appear to be the case.
Aperture diffraction can be calculated as $a = \text{asin}(1.22\lambda/D_i)$ where
\[
\lambda = \frac{h}{m \times v_{inf}} ,
\]
$\lambda$ is the Airy disc half angle and $v_{inf}$ is the particle velocity from (6). This adds little to the spot size at the current working distance with the current aperture and is not yet significant. An interesting question remaining to be answered is the effect of van der Waals interactions with the aperture walls. Such effects would be angular, hence like the Airy angle, have less effect on the spot size at close working distances to the sample.

The goals for optimization are to minimize the spot size while maintaining a constant spot intensity ($n_{beam}$ in atoms/s) as necessary for a constant signal to noise (S/N) ratio. This requires obtaining the highest possible beam intensity at the aperture, while minimizing the included angle from the aperture to the quitting surface, minimizing beam divergence.

Some general rules are as follows. Decreasing the source to aperture distance increases the beam intensity by the inverse square, but increases the beam divergence angle. Increasing the gas flow pushes up the pressure in the source chamber, and at some point causes excessive scattering. But increasing gas flow does not necessarily increase beam intensity since it forces moving the source farther from the aperture for a constant spot size.
4. Prototype microscope design, construction and software

4.1 Overall design and vacuum system.

A compact NAM was built, occupying under \(1 \text{ m}^2\) of bench space, plus space for electronics and two small mechanical fore pumps (Fig. 2). Two adjacent chambers are evacuated, the "source" and "sample" chambers. A capillary tube injects helium from a regulated pressure helium source, through a small free-jet nozzle.

![Figure 4.1 (a) Early photo of the NAM. (b) Later configuration.](image)

Pressure in the source chamber is maintained between \(10^{-2}\) and \(10^{-1}\) Pa by a 300 l/s turbo pump (Pfeiffer TPH-330), operating against the source gas load. A replaceable aperture separates the source chamber from the sample chamber. The distance between the source and aperture is adjustable from 0 to 250 mm. Pressure in the source chamber is measured by a cold cathode gauge. Fore-line pressure is measured by thermocouple vacuum and capacitive manometer gauges and is from 1 to 7 Pa due to the gas load.
The sample chamber is separately pumped by a smaller turbo pump pair, in series (Leybold TMP 50) and a second mechanical fore pump. This combination was necessary to achieve a sufficiently high compression ratio for the low background Helium partial pressure desired. During source operation, He partial pressure in the sample chamber is from $10^{-10}$ to $10^{-11}$ Torr and total pressure is between $10^{-7}$ and $10^{-6}$ Torr, as measured by an Inficon Transpector 2 Residual Gas Analyzer (RGA). A netbook style PC performs all of the computing tasks. The beam is projected against the surface of a sample located 10 µm to 100 µm from the aperture outlet. The sample is scanned in two dimensions perpendicular to the beam axis by an electromechanical scanner. The RGA, off to one side, produces the partial pressure signal used to form images. An op-amp circuit shown in appendix B drives the scanner. A small data acquisition box (Measurement Computing USB-1208FS) connects this to a personal computer (Samsung NC-10). A LabVIEW™ program (appendix A) was written to produce the scan waveforms and collect the image data over RS232 from the RGA.

The two dimensional image scan time has been 10 minutes to 18 hours, and image resolution in pixels from $30^2$ to $800^2$. The beam intensity appears to be similar to the best achieved to date by neutral atom focusing methods. Long scan times are typical when the system is optimized for best resolution. Note that no focus or astigmatism adjustments are needed, thus the first scan of a sample produces a sharp image and scan time is less of a problem than one might expect from experience with other microscopes.
4.2 Source

The source gas pressure is 0.1 to 6.9 MPa, depending mainly on the nozzle being used. A 3 nm filter is used to stop particles that might clog the source nozzle or aperture. Fused silica free jets such as Fig's 11 and 17 were pulled from 50 µm ID micro-capillary tubing using a pipette puller. This is a standard technique mentioned elsewhere\textsuperscript{23, 24, 25}. They were selected for tip ID by SEM imaging. A load lock is provided to exchange the source nozzle, which is mounted on the end of a tube extending into the vacuum through an o-ring seal. Transverse alignment of the source nozzle is done by the use of a rubber gasket at the mounting flange, the three mounting bolts are not quite tight, adjusting them aligns the source. This is far from ideal as the source nozzle is located within $\sim\frac{1}{2}$ mm of contacting on all three axis in the current operating location and the nozzle is on the end of a 12” long tube.
4.3 Aperture holder and aperture

A difference from a typical molecular beam apparatus is the design of the “skimmer”. Here the beam quality is not as critical as clearance on the sample side, and the aperture is on the end of a wide angle cone shape, (Fig. 4.4) pointing down-stream (reversed from normal practice).

The aperture was formed in a small graphite sheet, ~0.5 mm diameter by ~1 um thickness, obtained by pealing a Highly Oriented Pyrolytic Graphite monochromator (as used for STM or TEM samples) with adhesive tape. This is glued over a 0.35 mm hole drilled in the tip of a thin aluminum or copper cone, stamped from sheet, about 12 mm in diameter (fig. 4.4a). The graphite is then drilled using a Focused Ion Beam system (FEI Co.) to the desired aperture diameter. The aperture cone is then glued with epoxy to a machined conical aluminum holder. Aperture inside diameters have decreased over time from 2 µm to ~0.3 µm as detector sensitivity has improved.
The aperture design faces several requirements. It must place the aperture hole itself close to the sample. The hole must not scatter the beam by creating a high pressure area, as would occur if it was a long cylinder in shape (hole diameter smaller than the material thickness.) Next, gas atoms scattering from the sample should ideally travel to the detector directly, with as few additional scatterings as possible. Each additional scattering event in this pathway will dilute the probability of the atom reaching the detector, and thus reduce the image signal to noise ratio. So optimally, the aperture is at the narrow end of a cone or wedge shape to maximize open area leading from the sample (fig. 4.5, 6.1). However, the source side of the aperture must not concentrate the gas flow excessively, or it would scatter the beam.
Background pressure at the source side produces a random gas particle flow through the aperture. As a result, the beam is superimposed on this "spray" flow. It was found that if the source nozzle is retracted too far, the spray flow exceeds the beam flow through the aperture. This would produce a diffuse image. In addition there is Beer-Lambert law attenuation of the beam, greater scattering with greater beam length. Both of these affects become significant at about the same background pressure.

Thus the source nozzle must be close enough for the beam to dominate. This is easily seen by watching changes in the sample chamber helium partial pressure with changes in the nozzle to aperture distance ($L_{sep}$). Above some distance, the pressure no longer follows an inverse square relationship to the distance, indicating beam flow no longer dominates over non-beam flow. At high flow rates in the experimental microscope, this occurred at only a few cm distance.

At the reduced flow rates and very close $L_{sep}$ now used, this is not a problem. This is a Knudsen number question ($\lambda/L$), with the mean free path being increased during development from 30 mm to over 100 mm by reducing the flow rate and $L_{sep}$ (the distance the gas must traverse to reach the aperture) reduced from 6 mm to <1 mm. So the Knudsen number increased from 5 to >100. Knudsen numbers of at least 10 are considered free molecular flow. Alignment of the source nozzle to the aperture has become critical at this distance, however. Misalignment causes a large degradation in the contrast and quality of images.

The effect of the reflection of the source from the back side of the aperture deserves discussion. Logically, if the flow rate is low enough, the source flow and its
reflection superimpose on each other without interaction - gas to gas scattering is not common. At some higher flow rate, the scattered reflection would increase the gas density enough to cause excessive gas-gas scattering, and the flow would no longer remain free-molecular. Momentum from the source would begin to push gas towards the aperture and holder, and a standing shock wave may form. Counter-intuitively, if the source is moved closer - but flow reduced to maintain constant intensity at the aperture - the problem is reduced - the Knudsen number is raised. This question has the same answer as the “spray” flow problem, at a low enough flow rate the Knudsen number is high, and this reflection is not a problem. This is why a conventional “skimmer” pointed upstream is unnecessary.

4.4 Scanner.

Another advantage a neutral beam is the freedom to incorporate strong magnetic fields in the sample area. Electromechanical scanners can be used. These give the advantages of a wide maximum scan range, simplicity, repeatability and low hysteresis. For this experiment the lens focus and tracking actuator of a used CD-ROM drive serves as the scanner. This is a flexure mounted coil assembly with about 1 mm of maximum range in each of two axis (Fig. 4.6 a,b) A rare-earth permanent magnet provides the stator field. Approximately 20 mA at 0.1 V is required to reach full deflection. The scanner's resonant frequency is on the order of 10-60 Hz (depending on the mass mounted to it). Either viscous or electronic damping of the scanner is needed to reduce vibration sensitivity at resonance. Electronic damping was provided for the prototype, using a negative
resistance driver circuit to (mostly) eliminate the scan coil resistance (appendix B). This was needed to damp the scanner motion electromagnetically. Use in vacuum forced removing the small silicone damping pots originally located at the base of the scanner's flexure wires. This left the system well-damped but temperature sensitive! This circuit also produces XY vibration measurement outputs integrated from the coil velocities. A vibration isolation platform is built underneath the microscope, consisting of a sheet of plywood, tennis balls and a 12” diameter tire inner tube.

A very precise two-axis manual positioner was used to adjust the sample Z approach to the aperture. This is monitored using a stereo optical microscope looking through a vacuum window perpendicular to the beam.

Figure 4.6 (a) Scanner, (b) scanner assembly and (c) complete sample chamber arrangement on the right. The detector inlet is towards the top. Note the detector nozzle located above the tip of the aperture cone and sample surface.

4.5 Detector.

The mass-spectrometer (RGA) samples gas through a nozzle located to one side of the sample area (fig. 4.5, 4.6c) and produces a helium partial pressure measurement which is collected in an array. When the full frame has been collected, the software scales the minimum to maximum range of readings to black and white image limits. The range (contrast) from black to white was originally about 6 % of the average pressure after
removing noise. In recent scans this has been improved to 25-55 %, depending on the sample topography.

The RGA’s ability to reject the nearby mass 2 and 3 lines from the mass 4 signal was tested to determine if residual gasses were adding noise to the He signal, particularly hydrogen from water. With the source shut off, the H$_2$ pressure measured 6×10$^{-6}$ Pa and He pressure measured 1.3×10$^{-11}$ Pa. With the source turned on, He pressure was 2.8×10$^{-8}$ Pa, ~2000× higher than the background including any RGA selectivity “leakage”. Thus ultra high vacuum is not needed for basic imaging, due to the mass selectivity of the RGA.

4.6 Improvements during development

Improvements were made on the miniaturization front. First, the aperture diameter was reduced. Then the working distance was reduced (from 25-100 µm to 10-30 µm) by using a more precise sample positioner. Working distance is currently limited by the stereo (optical) microscope used to view the sample and avoid contact with the aperture. A higher resolution optical microscope would allow a smaller working distance.

The source nozzle inside diameter and flow rate were reduced. In addition, the distance from the source to the aperture was reduced, to 0.3-0.6 mm currently. The cone shaped “aperture holder” component was formed to a sharper point. This modification has improved image contrast by providing more clearance from the sample for scattered beam particles to reach the detector nozzle inlet. Alignment of the source to the aperture was improved.
The sample and scan plane were tilted somewhat towards the detector, effectively raising the illumination angle in the images.

It was also found important to electrically ground the sample platform since a buildup of electrical charge can cause movement of the sample due to electrostatic attraction to the aperture.

With smaller source nozzle ID's, the inlet pressure was raised, to 500-1000 PSI. This improved all image characteristics, and theoretically produces a more collimated and monochromatic beam as shown above.

Temperature sensitivity was a problem due to the scanner damping method effectively turning the scan coils into highly sensitive RTD's (resistance temperature detectors), in fact I was able to clearly see coil temperature changes of ~10mK in an experiment. This was improved by changing the driver circuit to damp near the resonant frequency and operate as a voltage to current amplifier at near DC frequencies. This still left it with a limited ambient temperature range it could accommodate.

The largest improvements were in detection, as the original RGA was replaced with an Inficon Transpector I and then a Transpector II, and as the detector nozzle was improved as the nature of and requirements for detection were learned. (More on this in section 6.)
5 Image results, contrast mechanisms discovered

5.1 Images and contrast mechanisms

Images gradually improved from barely recognizable to spectacular as the microscope improved. Here they are presented in no particular order. Each of these is interesting for some unique reason.

Figure 5.1, Gold foil on mica.

~50 nm thick sputter-coated foil was pealed off and flipped over to expose the smoother side as coated. Image strongly suggests predicted metal versus insulator contrast mechanism: scattering from metal is more specular than from an insulator.

Figure 5.2, Gold foil on mica.

Higher resolution mosaic of area above, including anomalously bright area in fold of metal.
Figure 5.3, Uncoated Crocosmia pollen grains.

Figure 5.4 a,b, Resolution test with Crocosmia pollen grain.

0.26 µm edge transition measurement by 12% to 88% criteria for a Gaussian beam profile. Taken by average of the four edges marked by white lines at the top of (a) and plotted in (b). Adjusting for the S/N ratio and scale calibration precision, resolution is 0.35 µm.
The image of Fig. 5.5 demonstrates another predicted contrast mechanism, surface roughness contrast. Much of the image shows areas that are darker than their surroundings, not because of topographic contrast (which is also present), but because the surface has a rough texture at a scale smaller than the microscope's resolution. Argon plasma etching produced this effect. This is confirmed by imaging the same sample by SEM. The raised rows have a cauliflower-like texture to the much better resolution limit of the SEM. The bright areas are smooth with small debris particles. The large-scale speckle noise throughout the image is actual sample texture, not microscope noise (which is also present, but of a finer scale). See also fig. 5.9.
Figures 5.6 and 5.7 demonstrate the ability of the NAM to image highly magnetized samples, such images could not be done by SEM for instance.

Figure 5.6. He scattering images of a crushed high-field NdFeB magnet. (a) An area with fractured planes at three depths and small particles. (b,c) Magnified areas of the same location. (d) Below, a test of a black-body color map on this image.

Figure 5.7. Tall magnet particle extending >100 µm from surface, with additional particles attached magnetically. This is a good illustration of the reduction in sharpness with distance from the aperture.
All images show topological contrast related to the angle of the surface relative to the detector and beam as one would expect from a diffuse reflecting surface, with the highest brightness corresponding to the specular geometry. The detector is towards the top of fig's 5.1-5.6, and the top left of fig. 5.7. The detector nozzle opening is just above the "horizon" of the sample surfaces. More specifically, the brightness at each point appears to roughly match the expected cosine distribution for diffuse scattering, corresponding to the visible included angular area of the beam spot as seen from the direction of the detector inlet (fig. 5.8). Shadowing of the visibility of the detector inlet area from the beam landing point also produces contrast as expected if you consider the detector inlet as the apparent source of “illumination”.

Figure 5.8 a,b, (a) normal cosine distribution of beam scattering from sample surface. (b) illustration of this as the apparent area of the beam landing spot as seen from the direction of the detector, for several surface angles.

Figure 5.9 is the same sample of figure 5.5, prior to etching the surface. This illustrates the lack of penetration of the beam into the sample, the test pattern cannot

Figure 5.9, Silicon integrated circuit test sample, corner. Pattern of low-k dielectric and copper is invisible to the NAM.
be seen (i.e., topographic contrast only.)

One important difference from the behavior of light is that He atoms do not undergo significant absorption. The detected partial pressure, converted to image intensity, is literally a mixed reflection to the detector (the source of “white”) and to the vacuum pumping system (the source of “black”).

![Figure 5.10](image)

Generally, sample areas from which beam particles undergo multiple scattering events on the path to the detector have a mid-level grey intensity. Grey intensity indicates more balanced probabilities of scattered particles reaching the vacuum system or the detector inlet first (fig. 5.10). In a similar situation for a light image, the multiple reflections would typically result in high absorption, these areas of the sample would be darker. This effect can especially be seen in narrow indented areas, such as point 1 within a grain in fig. 5.10a, and other obscured areas such as points 2, 3 and 4 of fig. 5.10b. On the upper right side of fig. 5.3, a simplified case of this effect can be seen where one pollen grain overlaps another. The reflection off of the closer grain produces a diffuse illumination of the grain below it. At the same time, contrast is reduced in that area. This varying contrast effect can be understood as a quality of the illumination of the area. A direct view of the
detector nozzle inlet or the vacuum system produces direct, high contrast illumination, while areas where escape requires multiple scattering are diffusely illuminated. Note that significant diffuse illumination is reflected from the detector side of the conical aperture holder, just above the sample.

Figure 5.11. Diagram illustrating a few of many possible scattering paths between the beam landing spot and the detector given a deep (aspect ratio) sample surface topography. Areas without a direct view from the detector inlet have reduced contrast due to multiple scattering.

Fig's 5.3, 5.6 and 5.7 show small areas and patches which have greater brightness than surrounding areas at the same angle and depth. This is likely to be the surface roughness contrast mechanism that was proven later in the image of figure 5.5.

The images of fig. 5.1 and 5.2 are exciting in comparison to the diffuse appearance of the mica background, or mica alone (fig 5.12). Theory and experiments predict that the reflection from a smooth metal surface should be more specular (shiny) than from an insulator due to the less tightly bound, shared conduction band electrons. This causes the reflection to occur farther from the surface atom cores. This might be a weak effect, producing, rather than a cosine scattering angle distribution, cosine \(^N\) distribution, where \(N\) is some fractional power, perhaps 1.5 to 2.5 for example. This “Metal versus Insulator” contrast would be very useful for imaging integrated circuit samples. Work needs to be done to verify and quantify this effect, and this may be simply surface roughness contrast.
In comparison to fig. 5.1, the mica surface above does not have a similar specular appearance despite a variety of angles being presented to the beam and detector. Also of note, edges of mica layer steps are clearly visible. Most likely these are not single crystal layer steps but thicker layers.

Note the thin vertical strand extending from the edge debris (probably copper).
On fig. 5.13, the line profile taken across the thin strand shows the beam profile was essentially Gaussian. This was done at an earlier stage with lower resolution then present.

In the pursuit of contrast that would reveal the test pattern on Low-k/ copper test IC samples, in-situ low-energy laser desorption and heating was tested. Because the vacuum condition in the sample chamber is not nearly UHV, and scans are long, all sample surfaces must be considered contaminated and covered with a water layer at the least. As a result, a method for continuously cleaning sample surfaces during scans, without damaging energy or charges is needed. A 532 nm, 75 mw laser was aimed at a silicon IC sample through one optic of a stereo microscope. The second side of the microscope allowed monitoring the laser aim (at low power settings!) This caused some heating and perhaps provided some energy for photo-desorption as well, without resorting to UV energy. Tests in air showed under 100 C sample temperature, but is it not clear what temperature was reached in the sample chamber vacuum.

Figure 5.14, In situ low-energy cleaning experiment.
This produced only a mild improvement in the visibility of the test pattern, which did slightly appear as a series of horizontal lines in contrast enhanced images (fig. 5.15).

![Figure 5.15](image)

Figure 5.15, silicon IC test sample, during laser heating / cleaning test. Raised lines appear, but are actually only a weak contrast effect.

### 5.2 Resolution

Resolution has reached 0.35 µm as illustrated in figure 5.4. An edge sharpness measurement is taken across the four edges marked by white lines at the top of (5.3a) and plotted in (5.3b). These sites are chosen because the sample topography presents bright linear edges facing the detector, folded under and contrasting against a dark shadowed background. These nearly sharp edges provide a reasonable resolution measurement.

Beam Full Width Half Maximum (FWHM) measurement is often used for resolution. The FWHM can be measured using a 12%-88% line step transition measurement in cases where the beam has a Gaussian profile. The edge 12%-88% transition distance is 0.260 µm, averaged over the four line profiles, with the 10%-90% figure being 0.274 µm. The image scale is calibrated using a 127 µm spacing Quantifoil TEM grid. Our resolution estimate, 0.35±0.05 µm, is more conservative than the FWHM to account for image noise and the scale calibration precision.

Vibration measurements done using the electromagnetic sample positioner/scanner indicate that vibration (mainly due to vacuum pumps) is a significant
component of the present resolution limitation, perhaps 100 nm. A primary factor in the vibration magnitude is the low fundamental resonant frequency of the XY scanner, which could be easily replaced.

Figure 6.1, Detector nozzle optimization (a) larger inlet, (b) smaller inlet, (c) enclosed detector
6 Discussion of future directions and ultimate performance limits

Section 3 discussed how to calculate the spot size and beam intensity. Combined with detector sensitivity, these are what determines the microscope's resolution, speed, and image S/N ratio performance. Currently the scan times are long and the resolution only matches far-field optical microscopes. How far can this be improved and how?

![Figure 6.2, resolution improvement versus time.](image)

Roughly 28x improvement in one year, validating our method for predicting resolution.

Noise limits all practical performance limits in NAM. All other performance parameters such as aperture size, beam divergence and speed can be almost arbitrarily improved, except the S/N ratio, which limits all. This is a statistical particle counting problem, limited by the beam intensity and detector sensitivity. It is clear that one could simply FIB drill a nearly arbitrarily small aperture in graphene or other thin material, until reaching the single digit or low tens of nm level, when contamination and mobility of the aperture atoms could make smaller apertures difficult. Holes as small as a single carbon atom have been drilled in graphene at PNNL using a helium ion FIB.
Consideration of the equations presented above, and basic Poisson noise statistics and the function of the detector nozzle (described below), leads to the following relation:

\[
\frac{\text{Source Brightness} \cdot \text{Detector Sensitivity} \cdot \text{Scan Time}^2 \cdot \text{Spot Size}^4}{\text{Number of Pixels}^2 \cdot \text{Pixel S/N ratio}^2 \cdot \text{Working Distance}^2} = \text{constant}
\]

The various performance factors, including scan time, resolution (spot size), number of image pixels, and S/N are traded off between each other, and optimized for particular goals. To make an overall improvement, the remaining factors: source brightness, detector sensitivity, and working distance must be improved.

Better optics for monitoring the aperture to sample distance would allow the working distance to be reduced. That then allows a reduction in the source to aperture distance without excessive beam divergence. This allows reducing the aperture diameter while maintaining a constant beam intensity. Active monitoring and control of the aperture-sample distance could be introduced to approach even closer, as in probe microscopes, while still remaining out of contact with the sample.

Improving the source intensity offers some possible improvement, as mentioned in section 2.2. Replacing our present vibration sensitive scanner would improve resolution by approximately 100 nm.

Detector sensitivity is the particularly important question, in part because commercial devices such as our excellent Inficon RGA perform so poorly. The efficiency of its ionizer is its major limitation, at approximately \(1/10^6\) of the theoretical limit. This subject will be expanded below and then we will conclude with estimates of the future performance.
6.1 Detectors for reflection-mode NAM

Producing image contrast by the reflection of beam atoms from a surface required a long process of experimentation and theorizing. Although the literature covers gas detection very well, nothing was found covering the logical path between the scattering of the beam and the detector. Experiment showed that it was not effective to simply place the RGA to one side of the sample. It turned out to be very important to be clear about what you are detecting and how that relates to a partial pressure within the detector.

First, the sample chamber is pumped to maintain a low background pressure, and all flow is free-molecular (reference 28 is a good primer on this). In reflection mode, image contrast (signal) is created by some surface characteristic that changes the angular distribution of the beam scattering. Other effects such as changes in velocity or spin might be detected, but those methods would be much more difficult.

Simply placing the detector to one side would produce a higher probability that an atom scattered in that direction would enter the detector, and a corresponding lowering of that probability for atoms scattered in the opposite direction. But it is not a large change in probability as it turns out, because on average the gas particles will undergo many (wall) scatterings before reaching the small detector ionizer inlet, and they have a greater probability of reaching the large vacuum pump inlet and being removed first. Atoms that do reach the detector have a probability of leaving as well. This leads to the realization that the detector is measuring through a network of vacuum conductances, which must be optimized for the highest pressure change in the detector with some sample contrast mechanism.
A good approach is to select some general scattering direction from the sample, an included angular area, that will be detected. We can call these atoms "contrast positive". Isolating this subset of the scattered gas with a detector nozzle was tried and found to be effective for maximizing the pressure signal. One of the reasons for this is the presence in the sample chamber of gas scattered in the opposite of the chosen "contrast positive" direction ("contrast negative"). These would reduce the contrast if detected. There are also "contrast neutral" atoms which would only increase the background pressure and noise, for example scattered in directions roughly perpendicular to a side-side contrast axis. There are many ways to think about this, I'll start with the most thorough I know first.

A good analogy can be drawn to a resistor network (Figures 6.3, 6.4). Conductance in units of l/s is simply the inverse of resistance, for which the units would be s/l. Voltage is analogous to pressure, and current to flow (throughput, in Pa×l/s). W. Schwarz provides a treatment of this technique\textsuperscript{28}.

We'll assume we are trying to create image contrast in reflection mode by differential scattering along one axis perpendicular to the beam. Call those atoms scattered generally toward the detector at one side "contrast-positive" atoms, and those scattered away "contrast-negative". Those scattered in any direction roughly perpendicular to the detector we'll call "contrast-neutral". Since the beam flow is constant, the total of these three flows is constant. In the steady state, this is the same as the flow reaching the vacuum system and thus the pressure at the entrance to the vacuum system is constant.
In the schematics the three flow categories are drawn as constant current sources. This is not an exact analogy since there are "beaming" effects in the free molecular gas flow. We can divide the system into three volumes connected by some set of vacuum conductances. The detector is in one of these, the sample chamber a second, and the vacuum pump a third.

Figure 6.3 Full detector conductance network

Figure 6.4, Optimized network
Call the pressure beyond the vacuum pump “ground” (zero), since atoms that reach it do not return. The pump conductance (in liters/second) combined with the conductance of all plumbing from the chamber to the pump become a single value, the effective conductance to "ground". Between the detector and the sample chamber is some tube and/or aperture area with a total conductance value. If the detector is directly in the chamber, this value could be very high, but still finite. To maximize contrast we must isolate the detector from the contrast negative flow and also from the contrast neutral flow. If that is done, the network ideally approaches the simpler one of fig. 6.4. This could be called "differential conductance contrast." Since this configuration ideally can't detect contrast-neutral and contrast-negative pressures, assume their conductance to the chamber can be ignored. The contrast-positive flow, however, has a conductance to the chamber, and one to the detector. We want to maximize the conductance of this flow to the detector, and minimize the conductance from the detector back to the chamber. One would also want to minimize the conductance for the same contrast-positive flow directly to the chamber.

6.2 Nozzle

Aiding us in this is that atoms scattering off the sample radiate from a specific source location, in this way the gas does not follow the electrical analogy. A way to use this is to build a detector nozzle which exclusively connects the detector to a limited area close to one side of the sample, which intersects an optimum cross-section of contrast-positive flow. An approximate cone shaped nozzle was used in the experimental microscope (the aluminum foil shape visible in the top of fig. 4.6c). This expands in cross-section towards
the detector to maximize the conductance to the detector while minimizing the conductance back to the chamber (fig. 6.1). Another approach would be to place the detector inlet as close as possible to the sample, and again enclosing the detector such that the inlet is also the only outlet for the detector.

Consider now the entrance area of the detector nozzle, at one side of the sample. If it is expanded, the conductance from the detector to the chamber is increased, reducing the detected pressure signal. But at the same time, the conductance of scattered atoms towards the detector is improved. If these atoms are contrast positive, a signal increase occurs as well. Finding an optimum detector nozzle opening shape is something that will take experimentation and/or Monte Carlo simulation. Note that the conical aperture holder, sample and flat aperture disc area are also involved here, up close to the beam landing point (fig. 4.5). Scattering of some contrast positive flow occurs off of these, into the detector nozzle. You can see that looking radially around the sample, there is some optimum included angle that the detector nozzle should be open to, perhaps 90 degrees or less, for collecting contrast-positive atoms.

![Figure 6.5, Radial view of detector nozzle. Sample and beam in center.](image-url)
The nozzle opening in the prototype now is a rectangle roughly 2 mm in Z, by 3 mm wide. Adding a second detector and nozzle on the opposite side, collecting contrast-negative flow, could be an improvement, providing a differential signal pair. A detector at 90 degrees to the first could provide quadrature (a sin & cosine pair) for detecting scattering angle, and produce useful two-channel color images or alternate illumination.

Ultimately, the apparent illumination of the sample in the resulting images is "from" the direction of the detector nozzle opening, similar to the effect of the location of the secondary electron detector in SEM. Choosing the detector nozzle opening area will also be a choice of "illumination" direction and shape. Sample surface slopes on the axis perpendicular to the detector direction produce little sample contrast. The overall illumination apparently has some diffuse component, possibly from scattering off of the aperture and other nearby objects.

During scanning, pressure at the detector is not in steady state but responds with some time delay. This is related to the enclosed volumes and conductances. In the electrical analogy, this is equivalent to capacitances to ground representing the detector and chamber volumes. The detected pressure has roughly a first order low-pass filter response.

_A more simple way of looking at all of this now is as follows._ The nozzle is a sheet metal cone shape with a narrow inlet area located close to the sample. Its large end connects to a volume enclosing the detector's ionizer and mass spectrometer head. The small inlet area facing the sample is the only area open to the vacuum system, and is thus also the outlet for any gas which enters the nozzle. In effect this has a large partial
pressure “multiplying” effect since the scattered beam particles radiate from the point of impact on the sample in free molecular flow. The detector nozzle inlet area must always cover an optimum included angle for the highest contrast. (fig. 6.1a, b) Using a small inlet area allows the inlet to be placed closer to the beam landing point where the pressure from scattered beam particles (impingement rate per unit area) is higher. Therefore a smaller inlet can sample a higher pressure. The tradeoff is in pressure response time, essentially a first order low pass filter with a time constant $\tau = v/C$, where $v$ is the empty detector volume enclosed behind the nozzle, in L, and $C$ is the vacuum conductance through the nozzle, in L/s. For an estimate we can treat the nozzle opening as an aperture.

$$C = k \cdot a_i \cdot \sqrt{\frac{g}{M_b \cdot mol}} \quad \text{(15)}$$

Where $M_b$ is the beam particle mass in g/mol, $a_i$ is the nozzle open area and $k = 630$ m/s at 300 K, which equals $(R \cdot T \cdot \text{mol} / (2\pi \cdot g))^{1/2}$.

Assuming a constant optimum included angle at the nozzle opening, the inflow of particles is consistent regardless of the open area. Also assuming the conductance of the vacuum chamber is much higher than the nozzle, the pressure contrast seen by the detector is inversely proportional to the area of the inlet. For our current microscope, the effective detector inlet area without the nozzle is ~380 mm$^2$. The current inlet area is 6 mm$^2$. The increase in pressure is then roughly 63:1. A “beaming” effect is ignored here which should increase the effectiveness of the nozzle, gas radiating into the nozzle from the beam landing point, on average, reaches deep within the nozzle before scattering
since the nozzle area expands behind the inlet. Gas returning randomly to the chamber faces the conductance limitation (higher scattering probability) of a decreasing area cone.

The result is a tradeoff which has been optimized for a response time similar to the present pixel sample time of 0.15 s. An unexpected result of this tradeoff between sensitivity and speed is that the optimized performance of this detector system is inversely proportional to its unfilled internal volume.

Unfortunately, if one desired to use a time-of-flight (TOF) detection method which required rapid sampling at the detector, the optimized detector nozzle method of increasing the pressure signal could not be used. An example of that would be the use of a source beam chopper and TOF-specific sampling to select a narrow range of particle energies. The slow pressure response would eliminate the TOF information. Selecting a particular beam energy by some method is important to many molecular beam experiments.

6.3 Ionizer

Detecting the pressure (density) of neutral gas atoms in a vacuum involves ionizing the atoms and then counting ions or measuring the current. The atoms can be mass-filtered to reject the background of other gasses. Helium is particularly useful as the background pressure is low at mass 4. Ionization by electron spray impact ionization is common, and field ionization has also been tried for possible use in a neutral atom microscope\textsuperscript{13}. Mass filtering can then be done using a magnetic sector as in helium leak detectors, or by a quadrupole mass filter, etc.
The ionizer has a limited input area and also does not ionize all of the Helium atoms entering it. As a result only a small fraction of the target atoms are detected. Detected ions are neutralized and return to the same gas volume from which they came, and may be detected again. Inherently, this is a partial pressure sensor with some noise level, best considered as an equivalent noise pressure. Some commercial quadrupole mass spectrometers have a noise level near $10^{-14}$ Torr (measured as the standard deviation in one second He partial pressure measurements).

Detector Noise Performance, Equivalent Area.

Fundamentally, a best case detector could count the rate of impingement of (correct mass) gas atoms on the detector nozzle inlet area ($n/sec.$) The result is the same if we count the number of atoms in a detector volume, by setting that volume to give a pressure response time constant equal to the sampling period ($\tau=\text{volume/conductance}$). The Poisson noise that results would have a standard deviation equal to square root($n$/sample period). An actual detector will have a higher noise level than this.

It is useful to compare a detector's performance using the standard deviation of its measurements (noise), to that of an ideal detector of some inlet area. To do this, first we start with the rate of particle impingement on an area (based on multiplying the particle velocity by the density of particles)$^{21}$. If we set the impingement rate equal to one per sample period (standard deviation of 1), we obtain a detector equivalent area,

$$a_{d} = \frac{\sqrt{2\pi m_{h} k 300 K}}{p_{n} \tau},$$

(14)
where $p_n$ is the noise (standard deviation) equivalent pressure (in Pa) and $t$ is the sample period.

For example, consider the previously mentioned noise level of $10^{-14}$ Torr, using a one second sample period. This is equivalent to an ideal detector sampling an area of only $10 \mu m^2$. The available nozzle inlet area is $2 \text{ mm} \times 3 \text{ mm}$, $6 \times 10^5$ greater. This is an estimate of just how much room for improvement there is in the detector system.

Conventional mass spectrometers have small ionizer inlet areas, and only ionize a few thousandths of a percent of helium atoms. Many things have been tried and at least one achieved a 70% detection efficiency, but only for a 2 mm diameter collimated incoming molecular beam. Nonetheless, that is a substantial improvement over the very good RGA used in this experiment, perhaps $2000 \times$ improvement, based on measured sensitivity of $497 \text{ mA/Torr}$ versus the $0.2 \text{ mA/Torr}$ specification for the RGA used on the prototype.

As explained in section 6.2, the performance of the combined detector and nozzle, with an inlet area optimized for the pixel sampling rate, is inversely proportional to the unfilled internal volume of the detector as well. This allows the possibility of improving both volume and ionization efficiency, and the combination should be assessed together. A detailed look at a particular design (section 7) shows that expecting a combined improvement of $1000 \times$ would be somewhat conservative.
6.4 Practical limits: examples

Improving the detector is a main variable for improving resolution. Some improvement in the source intensity is possible. A useful figure of merit for the system performance as a whole is obtained by multiplying the detector equivalent area by the source intensity \( I_0 \).

The source intensity can be increased, by perhaps 20x through the use of a de Laval or conical nozzle\(^{40}\) and possibly by using an additional source vacuum chamber, pump and “skimmer” aperture, and operating at a higher source pressure.

The combined physical limit appears to be at least \( 10^6 \times \) better than the current microscope based on the previous analysis, with a practical long-term goal based on known technologies being \( 10^3 \times \) to \( 10^4 \times \) based on better detectors and previously attained\(^{23-27}\) source intensities.

Removing the vibration component of the present resolution limit (upgrading the scanner) would be a simple and significant improvement. Within these known improvement possibilities, the practical limits currently are illustrated in table 3. The resolution and scan time values are based on using eq.(14), given the current microscope's performance, and improving the detector and source by a combined factor of \( 10^4 \). Aperture diffraction has also been included here using the Airy formula (p.18).

These are a few of the possible configurations, and all assume a 500 x 500 pixel\(^2\) image size and constant S/N ratio.

<table>
<thead>
<tr>
<th>Working Distance ((\mu m))</th>
<th>Scan Time (min.)</th>
<th>Resolution (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>25</td>
<td>6</td>
<td>250</td>
</tr>
<tr>
<td>25</td>
<td>60</td>
<td>89</td>
</tr>
<tr>
<td>2.5</td>
<td>6</td>
<td>80</td>
</tr>
<tr>
<td>2.5</td>
<td>60</td>
<td>29</td>
</tr>
<tr>
<td>1</td>
<td>60</td>
<td>19</td>
</tr>
</tbody>
</table>

Table 2, Estimated practical performance limits using known improvement methods.
Other improvements over the prototype are also desirable.

- Build a new system with UHV compatible components, able to stand baking to 150 C. This is important for surface science, quite possibly, sample surfaces are masked by surface contamination and absorbates. If so, the scattering seen is off of the contamination layer and not the underlying sample.
- Incorporate a gentle method for continuous cleaning of the water surface layer.
- Better control of vibration by adding an optical table, maglev turbopump, and by replacing our home-brew sample scanner with an UHV compatible piezoelectric scanner.
- Better sample positioning and control: more degrees of freedom with a piezoelectric sample positioner. In particular, the angle of the sample to the beam and detector needs to be controllable to quantify and maximize the brightness of specular reflections from the sample surface. This would increase the contrast produced by differences in the specularity of sample areas, while perhaps reducing the effect of topography. This should be of use in observing surface characteristics.
- Load locks for sample exchange and maintenance tasks, making it much easier to use.
- Better optics for monitoring the sample-aperture distance, so the working distance can be reduced, or a non-optical method for active control of the distance.
- Two or more detectors at angles around the sample, allowing simultaneous collection of images from a variety of illumination directions.
6.4 Future work

Aside from the drive for higher resolution and speed, there are many other fruitful avenues to pursue. Demonstrating atomic diffraction effects in images from crystal surfaces is important and in the case of Lithium Fluoride, should only require imaging that sample with the current microscope. Other crystals may require chilling the source nozzle by LN$_2$ cold finger to see diffraction. Quantitative measurement of brightness versus surface angle has yet to be done. This would allow proving (or disproving) metal-vs-insulator contrast. Imaging of clean hydrophilic and hydrophobic surfaces on the same sample, before and after the introduction of water vapor to the vacuum would also be interesting, as would images taken at various stages of monolayer growth in general. Testing the effect of different source gasses, such as Krypton and Deuterium would be very interesting and simply requires changing the gas bottle.

In conclusion, this technique shows enormous promise for future development and interesting, potentially very useful science.
Improved detector: concept, simulation, construction and testing

A search of the literature for better ionizers for neutral helium microscopy finds many results.\textsuperscript{13,14,15,17} The best of these found\textsuperscript{17,14} use a solinoidal magnetic field to trap electrons in a large cylindrical ionization volume. Simulation was done of a smaller version of this concept using Simion\textsuperscript{®} 8.1, replacing the liquid cooled electromagnet of the published prototype\textsuperscript{17} with a NdFeB permanent magnet. The results were promising and a breadboard prototype was constructed for operation in a bell-jar (a custom turbo-pumped evaporative coater system previously built by F. deArmond and E. Sanchez).

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure71.png}
\caption{Ionizer simulation.} \label{figure7.1}
\end{figure}

Electrons (black) are emitted by a ring-shaped filament hidden on the left, and cycle through the center following magnetic field lines.

Magnet (brown) is at a high positive potential relative to the filament and surroundings.

Generated ions (green) exit to either end of the cylinder.

Red lines are electric equi-potentials.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure72.png}
\caption{Ionizer prototype in operation.} \label{figure7.2}
\end{figure}
Testing of the ionization efficiency showed performance of 25 to 58 mA/Torr for Helium at $10^{-6}$ to $10^{-5}$ Torr. This is 125 to 290x the efficiency of our reference RGA's ionizer efficiency specification. Simulation of a 90° magnetic sector ion mass filter was then done, in combination with the ionizer. Preliminary testing showed that this worked but had a broad focus and poor separation of the adjacent masses. Following the re-invention of the “ExB” crossed field mass filter configuration, a 90° electrostatic sector was added, co-located within the magnetic sector. This essentially nulls the sensitivity to ion energy around the average value, resulting in a sharper focus (near zero chromatic aberration).

Figure 7.3, Ionizer and mass filter simulation. Ions are red: charge to mass ratio 3 (such as HD), green:4, blue: 5 (which does not exist except as multiply ionized heavier species).

This was simulated, built and tested and shown to function fairly well. The crude test power supplies were then replaced with a set of custom regulated power supplies to produce the numerous voltages needed. A “Raspberry-Pi” ARM (Advanced RISC
Machine) based linux computer board monitors the system, replacing the previous stack of digital volt meters. Refinement of electrode positions, filament position, and identification of various problems, has proceeded well and currently the detector prototype functions reliably and rejects the background gasses (primarily water and “cracked” hydrogen from water) by about a $10^5$ ratio. This is measured as a background current at the settings optimized for detection of the helium line, relative to the total pressure (current equivalent). End to end helium ionization efficiency is also quite good. More work needs to be done before packaging it for use on a microscope.

Figure 7.4, Detector electronics.

Figure 7.5, Detector (upper right) undergoing testing in a bell jar.

Black device on the lower left is an RGA serving as a reference.
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