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A Proposal for Improved Helium Microscopy

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Abstract: Elimination of the electrostatic objective lens and alternative use of a C_c- and C_s-corrected quadrupole doublet may increase the useful working distance of the helium microscope, improve its resolution from 3 to 0.3 Å, and improve its optimum convergence angle from 0.4 to 4 mrad.

Key words: helium microscope, focused ion beam, C_s correction, C_c correction, objective lens

INTRODUCTION

The Need for Improvement

Although immense progress has been made in scanning ion microscopy by the commercialization of bright atom-emitting ion sources (Ward et al., 2006; Economu et al., 2012), there is still a need for improvement of the helium microscope. Existing instruments use an electrostatic objective lens, which suffers from the defect of chromatic aberration. Electrostatic lenses also limit the space for detectors around the instrumental focus, because a short focal length is required in order to keep chromatic aberration as small as feasible. The net result as shown at point C in Figure 1 is a minimum possible resolution around 3 Å for a lens with $C_c = 1.3$ cm (Hill et al., 2012: Fig. 11).

Use of the Quadrupole Doublet

A straightforward method for improvement is to adopt one of the mid-column chromatic (C_c) and third-order (C_s) correctors devised for electron microscopy. As time progresses, these designs will undoubtedly come into use, but their capability to deal with extended sources over an extended image plane is unncessary in a probe-forming lens where the source and focus are both single points. A simpler and less-expensive method, readily adaptable to existing design, is to use a quadrupole doublet as an objective lens. Figure 2 illustrates a focused ion beam system with a quadrupole objective composed of upstream (U) and downstream (D) quadrupoles. Such a doublet is essentially half of a Rose-type mid-column corrector (Rose, 1967, 1971; Bastian et al., 1971), and as such it has unequal focal lengths in its two principal sections and thus makes an elliptical image of a round source. As originally advocated for the scanning electron microscope (Crewe et al., 1967), the asymmetrical nature of the image does not matter so long as it is small. The doublet also has a long focal length, and, as originally pointed out by Zach, when C_c and C_s corrections are applied, there can be a long working distance and more room for instrumentation (Zach & Haider, 1995). A long working distance also enables greater depth of focus than is available from a round lens with short focal length.

For either ions or electrons, correction of chromatic aberration follows the Kelman–Yavor principle (Kelman & Yavor, 1962), as developed by Rose and others at Darmstadt. When an electric quadrupole and a magnetic quadrupole are interleaved, the derivative of the focal length with respect to velocity can be made zero. Like the Wien condition that occurs when electric and magnetic forces are equal, this happens at what may be called the Kelman–Yavor condition, when the magnetic force is twice the electric force at every point in the lens. For either ions or electrons, correction of third-order aberrations can be effected by octopoles, as originally advocated by Scherzer in Darmstadt (Scherzer, 1947). When three octopoles are used with a doublet, there are only five third-order coefficients, as may be seen from eikonal techniques (Rose, 1966) or by Newton's force law in the impulse approximation (Martin, 2014).

Quadrupole lenses have the disadvantage of complicated (factory-based) initial alignment, which has been solved in electron microscopy (Zach & Haider, 1995) and ion microbeams (Martin & Goloskie, 1995, 1998) by schemes for measurement in place. First-order focusing and second-order parasites consume much effort. "A reliable alignment procedure is an essential prerequisite. In fact it is mandatory to additionally compensate the second-order axial aberrations…" (Haider et al., 1998). Third-order adjustments can then take place. Iterated computer adjustment to produce minimum misalignment and minimum aberration (Uno, 2005) is becoming routine in electron microscopy. Corresponding schemes of measurement and adjustment are needed in ion microscopy.

PROPOSED MATERIALS AND METHODS

Measurement Methods

It is proposed that detectors useful for scanning transmission ion microscopy (STIM) can be utilized to measure aberrations and enable zeroing of the five coefficients of a C_s-corrected quadrupole doublet. A position-sensitive detector can measure

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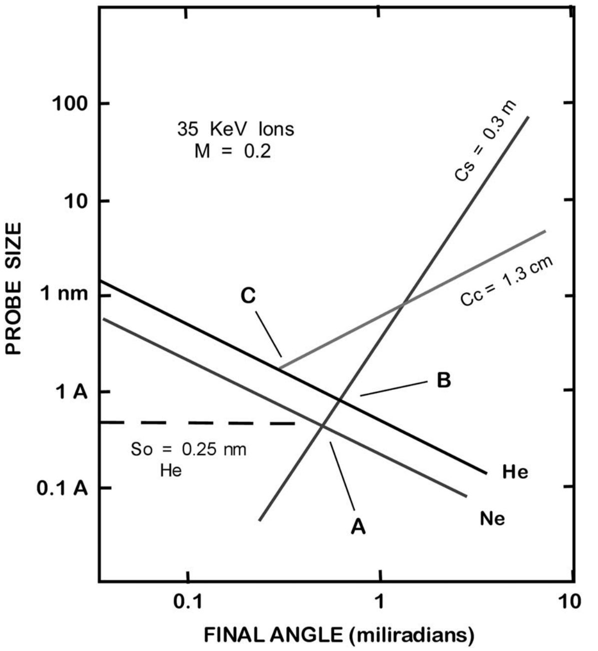

Figure 1. Probe size versus final convergence angle for He+ and Ne+ focused ion beams. At large angles, removal of chromatic aberration leaves significant spherical aberration. Correction of spherical aberration then moves the optimum resolution to smaller values and larger angles, with consequent greater beam current.

the rectangular (x, y) coordinates of the aberrations. For this purpose the beam can be divided into bundles by an electron microscope grid placed slightly downstream of the focal plane, and the deviations of the bundles from a rectangular array can be measured at the detector (Martin, 2013*a*). This method differs from schemes based on Fourier transform of the focused beam shape (Zach, 2003) or examination of Ronchigrams (Krivanek et al., 2003), as used in electron microscopy.

With the grid downstream of the focal plane, the aberrations are magnified according to the ratio of grid focal plane to grid-detector distances, which can be 1 mm relative to 50 cm, a ratio of 500. Thereby aberrations as small as 10 nm may be seen, if the detector has a resolution of 5 μm. If measurements at the edge of the lens aperture are made, and subsequently the lens is stopped down to 1/10 of its full aperture, third-order aberrations will fall by a factor of 10^3, resulting in correction to 10 pm, amounting to 0.1 Å. These measurements correspond to a third-order coefficient of magnitude 0.1 mm. The radial distance ratio of 10 is reasonable for a working distance of 4 cm and a half convergence angle as large as 12.5 mrad, resulting in a beam of radius 0.5 mm in a lens with radius 5 mm (Martin, 2013*c*).

Adjustment Methods

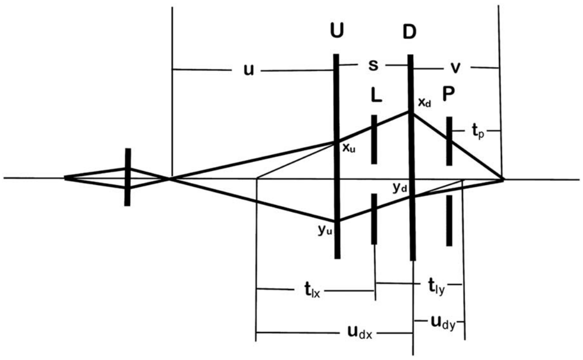

Figure 2. Ray diagram of a focused ion beam column containing an achromatic quadrupole doublet as used for preliminary experiments (Martin, 2012). The dark vertical lines separated by distance s represent the midplanes of the upstream (U) and downstream (D) quadrupole lenses. Rays emanating from a needle source at the left are focused by the condenser lens to a crossover, located at an object distance u upstream of the U lens. Between the quadrupole lenses, the rays aim at intermediate virtual line images with object distances (u_{dx}, u_{dy}) relative to the D lens. Downstream of the D lens, rays are focused to a point at the image distance v. The achromatic quadrupoles consist of four electrodes inside four insulated magnetic poles, a structure which can create a normal electric octopole as well as electric and magnetic quadrupoles. L is a 16-pole plate used to introduce one skew octopole in this preliminary experiment, and P is a postlens deflector oriented so as to introduce a second skew octopole, as required to null two of the five third-order coefficients of the assembly. The "throw distances" from these correctors to the various images are also indicated.

Preliminary measurements (Martin, 2012) have been carried out using a commercial separated-isotope gallium liquid metal ion source (which is unsuitable for high resolution because its effective diameter is much larger than the emitting site on a cooled, gas-fed, single-crystal needle of the atom-emitting type). They have shown that measurement of the rectangular coordinates of third-order aberrations can be used to single out one of the five coefficients for adjustment. For example, the variable part of the third-order aberration $x_3(a,\ b)$ when the semi-angle b subtended by the lens aperture in the y direction is held constant is given by

$$x_3(a,\ b) - Db^3 = Aa^3 + 3Ba^2b + 3Ca\,b^2,$$

with a slope

$$dx_3/d\,a = 3\,A\,a^2 + 6\,B\,a\,b + 3\,C\,b^2,$$

and the slope at $a = 0$ measures the single coefficient C. When measurements are made off-center in the Y section, so that b is nonzero, the coefficient can be set to $C = 0$ by adjusting the slope to zero. The A term has no slope at $a = 0$ and forms "cubic wings" that rise for positive a and fall for negative a, so that eliminating these wings adjusts only A. The C term is even in a, causing a parabolic curvature centered at $a = 0$, and adjustment of C causes the remaining curvature to flatten out into a horizontal line. Similar sequences can be carried out for $x_3(a,\ b)$ with b kept constant, or $y_3(a,\ b)$ with either a or b kept constant.

Thus, there are 12 ways to measure five coefficients, and the experimenter can choose which way produces the largest effects. As such measurements are carried out to higher accuracy, it can be expected that higher order terms will become visible near the periphery of the lens.

During the above measurements, aberrations can be adjusted by trimming elements consisting of skew (22.5°) and normal (0°) octopole lenses added to the two quadrupoles. Adjustment of a single one of the five coefficients can be done without affecting the values of any of the four others. Two of the coefficients (*B* and *D*) are adjusted only by the skew octopoles, independently of the other coefficients. One skew lens can be used to set $B = 0$, and then two skew lenses used to make $D = 0$. The two lens strengths are varied simultaneously in the ratio required to keep $B = 0$. Similarly, the remaining three coefficients (*A*, *C*, and *E*) depend only on normal octopoles, and one coefficient at a time can be adjusted using normal lenses first singly, then two together, and finally three normal lenses at once. The ratios among lens strengths follow from the lens geometry (Martin, 2013*b*, 2014), and can be incorporated as constants in computer programs used to adjust the lenses.

ANTICIPATED RESULTS

Increased Optimum Angle and Improved Optimum Resolution

The estimated performance of an improved helium microscope is also shown in Figure 1, where the estimated probe size due to various aberrations is shown as a function of the convergence angle of the beam at the focus. The lines sloping downwards to the right show the effect of diffraction in the lens aperture. In order to produce the least diffraction, the helium ion energy is raised to the maximum energy of 35 keV available in a typical microscope. The line sloping steeply upward to the right shows the effect of aperture aberration, estimated assuming the largest of the five coefficients is a third-order coefficient of 0.3 m. The value of $C_s = 0.3$ m requires adjustment of electric lens voltages with an accuracy of one part in 10^5, a value within reasonable stability limitations (Martin, 2013*c*). Aperture aberration becomes equal to diffraction at the point of optimum resolution *B*, where spot size is 1 Å and convergence angle 0.8 mrad. In order to keep the geometrical image of the emitting site smaller than the aberrations, a magnification of 0.2 is utilized.

Point B in the graph moves downward and to the right as C_s is made smaller, and decreasing C_s below 0.1 m is unlikely to be necessary because the resolution would be limited by the geometrical image of the effective source size.

Although a separated-isotope feed gas would be required for use with achromatic lenses, the use of more massive Ne would decrease diffraction and enable better resolution. For $M = 0.2$, the Ne ions reach the geometric limit at point A, where the optimum resolution is about 0.3 Å.

Possible Benefits

The effects of increased resolution of focused ion beams cannot be adequately forseen. Combined with cold ion sources (Hill & McClelland, 2003; Knuffman et al., 2013), compensated quadrupole doublets might be used to introduce arrays of single phosphorus atoms in silicon for quantum computing purposes (Pla et al., 2012). A likely use is in application of STIM to thin specimens. STIM requires megavolt energy ions to produce images of the interior of biological cells (Watt et al., 2013), and kilovolt-energy ion beams give high-resolution images only of the surface of biological cells (Chen et al., 2011). However, increased resolution at kilovolt ion energy may enable extension of STIM techniques to specimens of monolayer thickness mounted on thin carbon films. The strong interaction of lower energy ions may actually be an advantage in this instance, enabling registration of a single atom by the first projectile which strikes it as measured in STIM geometry. For example, the ionization cross-section of oxygen gas impacted by 30 keV helium ions is at its peak value of 1.4×10^{-15} cm^2 (Rudd et al., 1985). STIM geometry is also readily adaptable to the high-angle annular-detector technique for detecting target atoms of high atomic number as pioneered by Crewe, which has been successful in locating labelled bases in a single DNA molecule as long as 7,000 base pairs in electron microscopy (Bell et al., 2012).

CONCLUSION

Correction of chromatic and spherical aberration may be applied to the helium microscope. The simplest system has a quadrupole doublet objective lens with correcting octopoles. In addition, the doublet has the advantage of long working distance, enabling better working space around the focal point. All aberration correctors have the need for computer-based initial adjustment, which in the doublet involves only five third-order coefficients. With the advent of inexpensive computers for initial feedback alignment, as proven in electron microscopy, and solution of the problem of brightness by atom-emitting ion sources, as are now available commercially, the opportunity at last exists for useful progress in ion microscopy.

REFERENCES

BASTIAN, B., SPENGLER, K. & TYPKE, D. (1971). Ein elektrisch-magnetisches Octopol-Element zur sphärischen und chromatischen Korrector von Elektronenlinsen. *Optik* **33**, 591–596.

BELL, D.C., THOMAS, W.K., MURTAUGH, K.M., DIONNE, C.A., GRAHAM, A.C., ANDERSON, J.E. & GLOVER, W.R. (2012). DNA base identification by electron microscopy. *Microsc Microanal* **18**(5), 1049–1053.

CHEN, X., UDALAGAMA, C.N.B., CHEN, C.-B., BETTIOL, A.A., PICKARD, D.S., VENKATESAN, T. & WATT, F. (2011). Whole-cell imaging at nanometer resolutions using fast and slow focused helium ions. *Biophys J* **101**, 1788–1793.

Crewe, A.V., Eggenberger, D.N., Welter, L.M. & Wall, J. (1967). Experiments with quadrupole lenses in a scanning microscope. *J Appl Phys* **38**, 4257–4266.

Economu, N.P., Notte, J.A. & Thompson, W.B. (2012). The history and development of the helium ion microscope. *Scanning* **34**, 83–89.

Haider, M., Rose, H., Uhlemann, S., Schwan, E., Kabius, B. & Urban, K. (1998). A spherical-aberration-corrected 200 kV transmission electron microscope. *Ultramicroscopy* **75**, 53–60.

Hill, R., Notte, J.A. & Scipioni, L. (2012). Scanning helium ion microscopy. In *Advances in Imaging and Electron Physics*, Hawkes P.W. (Ed.), pp. 77ff. Amsterdam: Elsevier.

Hill, S.B. & McClelland, J.J. (2003). Atoms on demand: A fast, deterministic source of single Cr atoms. *Appl Phys Lett* **B2**(18), 3128–3130.

Kelman, J.M. & Yavor, S.Y (1962). Achromatic quadrupole electron lenses. *Soviet Phys Tech Phys* **6**, 1052–1054.

Knuffman, B., Steele, A.V. & McClelland, J.J. (2013). Cold atomic beam ion source for focused ion beam applications. *J Appl Phys* **114**, 044303-1-7.

Krivanek, O.L., Dellby, N. & Lupini, A.R. (2003). Auto adjusting charged-particle probe-forming apparatus. US Patent 6,552, 340 B1.

Martin, F.W. (2012). A large-aperture ion-beam lens corrected for both chromatic and spherical aberration. *Abstracts Archive for the International Conference on Electron, Ion, and Photon Beam Technology and Nanofabrication (EIPBN12)*, poster P07-08. Available at http:// eipbn.omnibooksonline.com (retrieved 2013-06-03).

Martin, F.W. (2013*a*). Particle-beam column corrected for both chromatic and spherical aberration. US Patent application 13,849496, laid open October 10, 2013.

Martin, F.W. (2013*b*). Second and third order aperture aberrations of the compensated quadrupole doublet. *Microsc Microanal* **19**(Suppl 2), 388–389.

Martin, F.W. (2013*c*). On achromatic probe-forming lenses for FIB columns and helium microscopes. *Microsc Microanal* **19**(Suppl 2), 1194–1195.

Martin, F.W. (2014). Cc, Cs and parasitic correction in quadrupole probe-forming lenses. *Optik* **125**, 1311–1315.

Martin, F.W. & Goloskie, R. (1995). Experimental compensation of parasitic octopole aberrations in small-bore quadrupole lenses. *Nucl Instr Meth Phys Res* **B104**, 59–63.

Martin, F.W. & Goloskie, R. (1998). Simultaneous compensation of 2nd-order parasitic aberrations in both principal sections of an achromatic quadrupole lens doublet. *Nucl Instr Meth Phys Res* **B30**, 242–247.

Pla, J.J., Tan, K.Y., Dehollan, J.P., Lim, W.H., Morton, J.J., Jamieson, D.N., Dzurak, A.S. & Morello, A. (2012). A single-atom electron qubit in silicon. *Nature* **489**, 541–545.

Rose, H. (1966). Allgemeine Abblildungseigenschaften unrunder Elektronenlinsen mit gerader Achse. *Optik* **24**, 36–59.

Rose, H. (1967). Uber den spharischen und den chromatischen Fehler unrunder Elektronenlinsen. *Optik* **25**, 587–597.

Rose, H. (1971). Abbildungseigenschaften sphärisch korrigierter electronenoptischer Achromate. *Optik* **33**, 1–24.

Rudd, M.E., Goffe, T.V., Itoh, A. & DuBois, R.D. (1985). Cross sections for ionization of gases by 10-2000 KeV He + ions and for electron capture and loss by 5-350-keV He + ions. *Phys Rev* **A32**, 829–835.

Scherzer, O. (1947). Spharische und chromatische Korrectur von Electronen-Linsen. *Optik* **2**, 114–132.

Uno, S., Honda, K., Nakamura, N. & Matsuya, M. (2005). Aberration correction and its automatic control in scanning electron microscopes. *Optik* **116**, 438–448.

Ward, B.W., Notte, J.A. & Economu, N.P. (2006). Helium ion microscope: A new tool for nanoscale microscopy and metrology. [unpublished abstract, EIPBN06 conference, no online archive].

Watt, F., Chen, C.N.B., van Kan, J.A. & Bettiol, A.A. (2013). Whole cell structural imaging at 20 nanometre resolutions using MeV ions. *Nucl Instr Meth Phys Res* **B306**, 6–11.

Zach, J. (2003). Method for detecting geometrical-optical aberrations. US Patent 20030001102.

Zach, J. & Haider, M. (1995). Aberration correction in a low voltage SEM by a multipole corrector. *Nucl Instr Meth Phys Res* **A363**, 316–325.