A planar refractive x-ray lens made of nanocrystalline diamond

L. Alianelli,[1,a)] K. J. S. Sawhney,[1] A. Malik,[2] O. J. L. Fox,[3] P. W. May,[3] R. Stevens,[2] I. M. Loader,[2] and M. C. Wilson[2]
[1]Diamond Light Source Ltd., Didcot OX11 0DE, United Kingdom
[2]STFC Micro-Nano Technology Centre, Didcot OX11 0QX, United Kingdom
[3]School of Chemistry, University of Bristol, Bristol BS8 1TS, United Kingdom

(Received 5 October 2010; accepted 15 October 2010; published online 23 December 2010)

Diamond has unique properties which make it the ideal material for use in synchrotron instrumentation. X-ray optics made of diamond are almost transparent, they possess strength, and are subject to very low thermal expansion; therefore they will be able to withstand the powerful beams generated by fourth-generation light sources without compromising brilliance. For this reason, several groups are attempting fabrication of refractive lenses and zone plates made of diamond. Lithography and, in general, microfabrication technology, are the ultimate tools for the innovation of synchrotron focusing optics. We propose to combine modern silicon microtechnology with advanced deposition methods to fabricate nanocrystalline-diamond lenses for third- and fourth-generation synchrotron sources. The fabrication method is described here and microfocusing synchrotron tests are illustrated. © *2010 American Institute of Physics*. [doi:10.1063/1.3517060]

I. INTRODUCTION

In the past two decades, lithography and semiconductor microfabrication techniques have represented the ideal tool for fabrication of high resolution synchrotron optics. Both refractive lenses and zone plates have been fabricated that are capable of focusing synchrotron light down to nanometer spot sizes.[1–7] Diamond is an attractive material for synchrotron optics due to its excellent thermal properties. In addition, it is a material with low x-ray absorption and can be exploited to make refractive lenses with better efficiency than currently used silicon lenses. Single crystal diamonds have been the subject of several studies using x-rays for a very long time due to their extremely interesting structural properties. In the last decade large diamond crystals of good quality have been characterized and exploited as synchrotron monochromators, polarizers, and beam splitters. Diamond is also used to make x-ray windows and monitors. It has been proposed that a plasma etch method would deliver a nonabsorbing diamond lens.[8,9] It seems, however, extremely difficult to etch diamond in a controlled way due to its physical hardness. Other methods like laser ablation and ion beam milling are not appropriate to fabricate lenses for synchrotron radiation, as these require a high aspect ratio. In this report, we discuss the properties of a diamond lens made with a novel fabrication method that could be exploited both in third- and fourth-generation x-ray sources. Extremely homogeneous layers of nanocrystalline diamond are grown on top of silicon molds using microwave chemical vapor deposition (CVD). The silicon is then easily removed by etching, leaving the freestanding diamond lens structures.

We present experimental data showing that the lens is focusing the x-ray beam to micrometer sizes without producing the beam splitting often observed when using single-crystal diamond. The focusing properties are not strongly affected by the use of a polycrystalline material for refraction of x-rays, although this is certainly responsible for unwanted scattering.

Efficiency and aperture of x-ray lenses are limited by absorption in the refractive material. Therefore, unlike lenses that work in the visible range, the x-ray lens effective aperture, defined as the aperture that transmits 75% of the full flux from the lens, can be much smaller than the total aperture. The effective aperture is an important parameter which permits to estimate lens efficiency and diffraction limited beam size of the focal spot. Our data show that the diamond lens efficiency increases proportionally to the illuminated aperture, therefore suggesting that the transmission is almost constant within this aperture. This is a consequence of low absorption and is observed in other microfocusing lenses like those made of beryllium. High quality nanofocusing lenses are made of silicon instead, which is more absorbing, and have much smaller effective apertures. A nanocrystalline diamond lens would be able to provide unprecedented efficiency for nanofocusing applications.

II. DESIGN AND FABRICATION OF DIAMOND LENSES

We have designed and fabricated diamond lens chips which can provide a large choice of focal lengths in the hard x-ray energy spectrum. A photograph of a chip carrying 16 arrays of nanocrystalline diamond structures is shown in Fig. 1. The array is a planar compound refractive lens (CRL). Each CRL is designed to provide a focal length of exactly $f=0.55$ m for energy values $E=20$ (top), 19, 18, ….., 5 keV (bottom). The curvature of the refractive parabolic surfaces is adjusted and fixed for each array. The CRL can be used to obtain different focal lengths from the design value by adjusting the x-ray energy according to the thin lens formula

a)Electronic mail: lucia.alianelli@diamond.ac.uk.

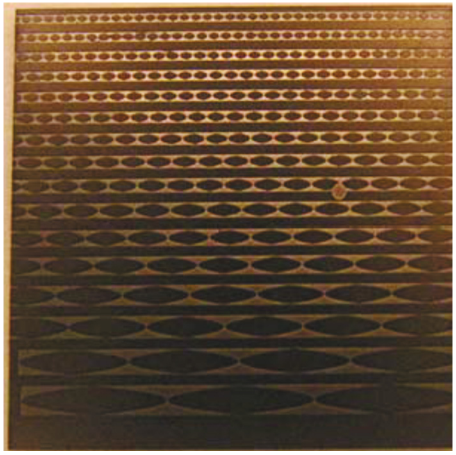

FIG. 1. (Color online) Optical microscope image of a silicon template chip with diamond deposited on top. The chip dimensions are 20 mm ×20 mm.

$$f = R/N\delta, \tag{1}$$

where N is the number of refractive surfaces and δ is the refractive-index real-part decrement,

$$\delta = \mathrm{Re}(1 - n) \approx 10^{-6} - 10^{-5}. \tag{2}$$

For the chip illustrated here the radius at the parabola apex is in the range $R = 50-60$ μm. A silicon template is made using standard lithographic and silicon etch tools; the lens arrays are patterned onto a silicon wafer using SU8 resist and UV lithography. The deep silicon etch of the molds is carried out in an inductively coupled etcher from Surface Technology Systems. The etch recipe is optimized to minimize scalloping and verticality of the walls. Diamond deposition is carried out using a microwave plasma CVD reactor at the School of Chemistry of the University of Bristol.[10,11] Prior to deposition, the samples were seeded using electrospray deposition of 5 nm diamond particles in methanol.[12] The reactor parameters, including reactor power density and substrate temperature, were adjusted in order to provide uniform diamond deposition without large crystalline domains or polycrystalline morphology as well as increasing the growth rate. While the use of typical nanocrystalline diamond deposition chemistry, where the majority of the CVD gas mixture is argon with small amounts of hydrogen and methane, is too slow to achieve the required film thickness of >20 μm on a feasible time scale, faster growth rates can be obtained by addition of 0.2% N_2 to the standard H_2/CH_4 gas mixture. The presence of N_2 both increases the growth rate of diamond, so that thick coatings can be deposited onto the lens substrates, and induces the required nanocrystalline morphology.[13]

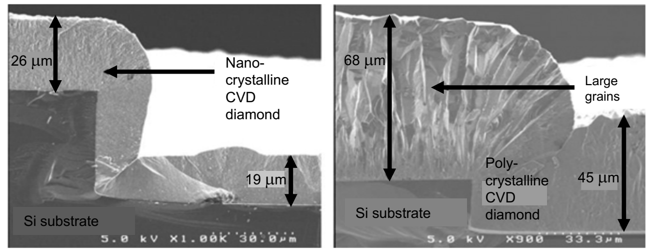

FIG. 2. SEM images of sliced diamond layers deposited on silicon molds. These images illustrate the different quality of diamond material achieved with different deposition chemistry. Left: nanocrystalline diamond, believed to provide better lenses due to reduced unwanted scattering, and used for experiment described in this paper. Right: microcrystalline diamond unsuitable for synchrotron optics.

It is interesting to note that synchrotron radiation has not yet been used to characterize nanocrystalline diamond. We will show results of such characterization, in terms of phase content, and particle size measurement, in a future paper. In this paper scanning electron microscope (SEM) images clearly show the improvement in material quality achieved during this work. The images in Fig. 2 have been recorded after slicing two different chips made with different deposition chemistry. The actual lenses presented in this work have a nanocrystalline morphology, and the microcrystalline sample is shown for comparison only.

Figure 3 displays images of the freestanding lenses obtained after silicon removal. The geometrical aperture is $A = 300$ μm while the maximum deposition thickness used so far is $D = 40$ μm. The lens dimensions are not intrinsically limited, and thicker diamond structures would be obtained using longer deposition runs with better control of overhanging diamond growth at the corners of the mold.

III. EXPERIMENTAL

The B16 Test beamline at Diamond Light Source is a versatile instrument used for a range of experiments in material science, technique development, and instrumentation development.[14] The beamline exploits a bending-magnet x-ray source and is equipped with a range of optics, detectors, and ancillary equipment, such as high-resolution stages which are ideal for lens tests. In this experiment, the beamline optical layout includes a double-crystal silicon monochromator and slits to vary the size of the incident beam. The diagnostics system is a high-spatial-resolution detector based on a 5 μm Europium doped lanthanum aluminium garnet $La_3Al_5O_{12}$ (LAG) scintillator, a microscope, and a PCO4000 charge coupled device camera. The detector allows measurement of beam details with spatial resolution of 1.3 μm. The x-ray image shown in Fig. 4 was obtained by illuminating

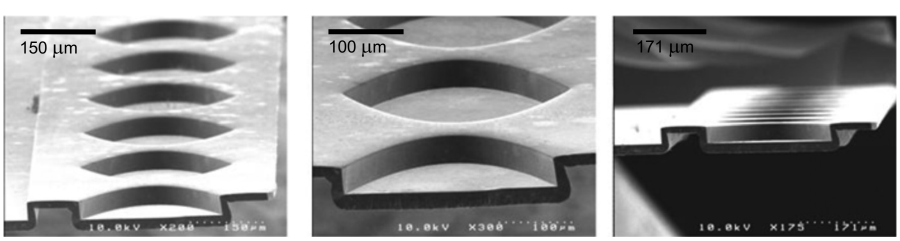

FIG. 3. SEM images of the freestanding nanocrystalline diamond lenses after silicon etch and removal.

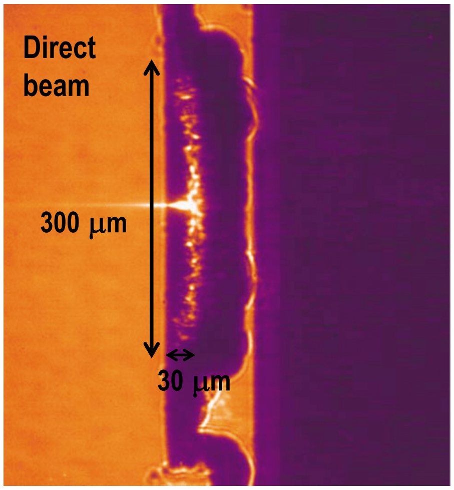

FIG. 4. (Color online) X-ray image of the diamond lens structure. The bright spot in the middle is the microfocused beam. The orange area is the unfocused monochromatic incident x-ray beam. The purple areas are the diamond lens structure, easily recognized by looking at the SEMs in Fig. 2, and the diamond carrier chip to which the nanocrystalline diamond was glued before the silicon etch.

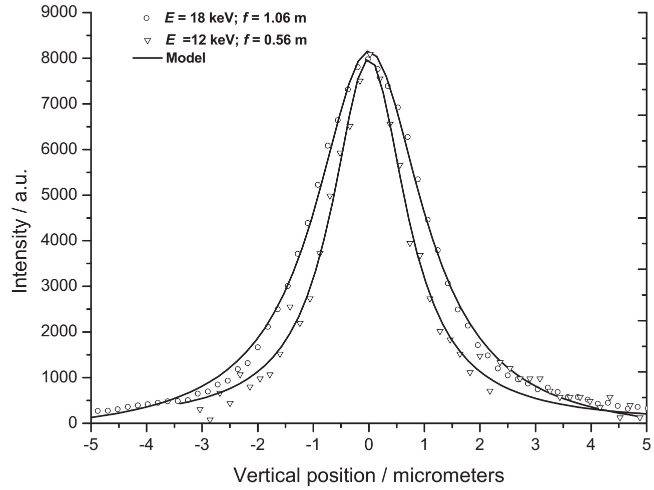

FIG. 5. Vertically focused beam profiles extracted from a detector image. The data are the results from two different experiments, at different values of the energy and of the focal length as summarized in Table I. Two different lenses on the same chips were used. The model (solid line) is a Lorentzian curve that fits the data and provides the following values for the focused beam FWHM: 2.2 μm (open circles) and 1.6 μm (triangle) as reported in Table I.

the diamond lens with a very wide beam, and positioning the scintillator in the lens focal plane. The image shows the presence of a linear focus as expected. Additional details are visible, for instance diffraction effects from lens borders, a topographic image of the lens structure and of the carrier chip to which the lens was glued after growth of nanodiamond.

The experiment is located at distance $P=47$ m from the bending-magnet source. The lens chip is illuminated by a monochromatic beam of x-rays, with energy values of $E=18$ keV (Lens A) and 12 keV (Lens B). The chip is mounted in vertical focusing geometry as seen in Fig. 4, and the detector placed at the focal distance $Q=1.06$ m (Lens A) or $Q=0.56$ m (Lens B). A minimum full-width-half-maximum (FWHM) of the focused beam profile is reached of 2.2 μm and 1.6 μm for the two different focal lengths, respectively, as summarized in Table I. The theoretical source size on B16, in the vertical plane, is $S_{\mathrm{FWHM}}=56$ μm, while the measured effective source is larger, of order 80–100 μm. The focused beam-profile plots, extracted from the imaging detector data, are shown in Fig. 5. Despite the Gaussian profile of the beam incident on the lenses, in this case the focused beam data have to be fitted by purely Lorentzian profiles; this is a problem often encountered with focusing optics, and we believe that scalloping produced by the Bosch process is partly responsible for it. Scalloping in these lenses has submicrometer amplitude which causes unwanted x-ray scattering and the appearance of tails in the focused beam profile. We were able in the past to obtain focused beams with Gaussian profiles by using a fourth-generation silicon etcher for different types of lenses.[15] A possible improvement of the diamond lens would consist in using advanced machines with faster etch cycles; in this way silicon molds would be fabricated with scalloping amplitude of 20 nm or less. High resolution SEM shows that the CVD diamond replicas have high form fidelity; therefore reduced silicon scalloping would result in smoother lenses. Another inherent cause of scattering is the nonperfect crystal material; the influence of the polycrystalline morphology on the lens efficiency will be assessed in a future work using x-ray small-angle scattering data.

The linear focus from a planar lens is normally a uniformly bright line: in Fig. 4 we observe instead some additional features across the lens aperture. These features are the results of lens imperfections, and are visible due to the low absorption in diamond. We suggest they may be caused by bending of the lens along its optical axis. Bending is caused by the lens itself being very thin and free-standing. This structural problem will be solved in future by increasing the robustness of the diamond layer; by adding a frame to the chip, or by growing a thicker diamond layer. It is possible that lens distortion (i.e., departure from the shape etched into silicon) happens during growth, and this may also produce the observed aberrations.

We recorded the focused beam intensity versus lens ap-

TABLE I. Summary of microfocusing lens test results.

Lens	X-ray energy (keV)	Focal length (m)	Source demagnification	Theoretical focus size FWHM (μm)	Measured beam size FWHM (μm)
A	18	1.06	44	1.8–2.25	2.2
B	12	0.56	84	0.95–1.2	1.6

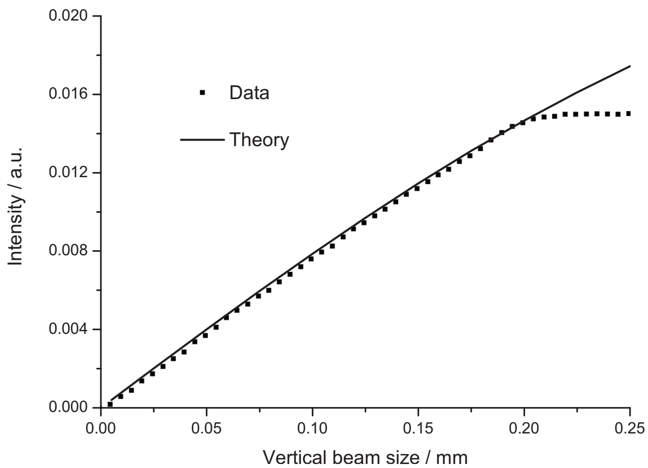

FIG. 6. Intensity of the focal spot as a function of size of the incident beam, with $E=18$ keV, and $f=1.06$ m. The theoretical values are the product of three factors: theoretical transmission; beam size; arbitrary scaling factor. The experimental data are the integrated flux in the focal spot.

erture by varying the vertical size of the beam incident upon the lens. The results are shown in Fig. 6, where the theoretical data are the product of theoretical transmission by incident beam size, scaled by an arbitrary factor. The expected transmission by the lens with $f=1.06$ m, at $E=18$ keV, is $T=75\%$, for a 200 μm aperture. The data in Fig. 6 demonstrate that the focused beam intensity is approximately proportional to the beam size up to an aperture of 200 μm, i.e., the transmission by the lens is approximately constant within this aperture. A supplementary test of local lens efficiency is repeated by scanning a pencil beam across the lens aperture: the result is plotted in Fig. 7 and shows that the maximum variation in efficiency is less than 15%. As already discussed, the flux is in good approximation proportional to the incident vertical beam size, specified in the labels.

IV. SUMMARY AND PROSPECTS

Diamond microfocusing lenses have been developed with a novel deposition technique allowing satisfactory control of the lens growth. This technique potentially improves the quality of diamond lenses compared to previous work made using single-crystal diamond. We suggest that growth of nanodiamond is more reliable than etching of single-crystal diamond due to the extreme hardness and the presence of imperfections in single crystals, such as stacking faults. The lenses obtained with deposition have good aspect ratios and uniform efficiency over a wide aperture. Unlike etching, this technique will permit growth of thicker lenses which can actually be used on beamlines. We plan to produce lenses with reduced focal length to demonstrate submicrometer focusing with nanodiamond, and improve robustness of the free-standing lens layer. Once the fabrication study is completed, nanodiamond lenses will allow better focusing efficiency and easier heat load management compared to other microfabricated optics.

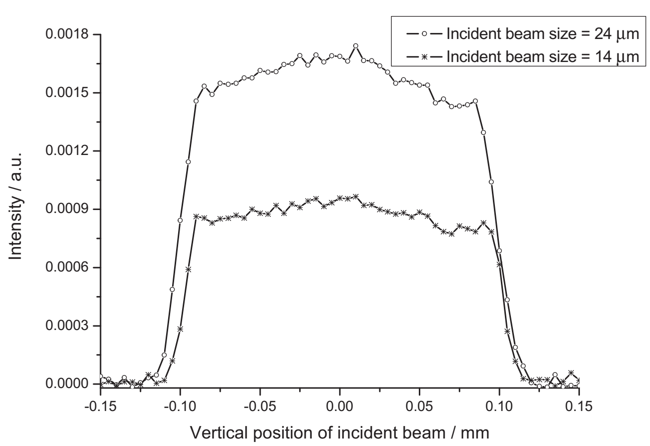

FIG. 7. Pencil beam scan of the lens aperture. The data are the integrated flux in the focal spot vs pencil-beam position for incident vertical beam sizes as indicated in the legend.

ACKNOWLEDGMENTS

The fabrication of focusing lenses is funded by STFC through Grant No. ST/F001665/1. O.J.L.F. was funded by EPSRC Grant No. EP/D074924/1. We acknowledge Diamond Light Source Ltd. for providing beamtime on B16.

[1]C. G. Schroer, M. Kuhlmann, J. Patommel, U. T. Hunger, T. F. Gunzler, O. Kurapova, S. Feste, M. Drakopoulos, A. Somogyi, A. S. Simionovici, A. Snigirev, I. Snigireva, C. Schug, and W. H. Schroder, Appl. Phys. Lett. **82**, 1485 (2003).
[2]C. G. Schroer, O. Kurapova, J. Patommel, P. Boye, J. Feldkamp, B. Lengeler, M. Burghammer, C. Riekel, L. Vincze, A. Van der Hart, and M. Kuchler, Appl. Phys. Lett. **87**, 124103 (2005).
[3]K. Evans-Lutterodt, J. Ablett, A. Stein, C. Kao, D. Tennant, F. Klemens, A. Taylor, C. Jacobsen, P. Gammel, H. Huggins, G. Bogart, S. Ustin, and L. Ocola, Opt. Express **11**, 919 (2003).
[4]K. Evans-Lutterodt, A. Stein, J. M. Ablett, N. Bozovic, A. Taylor, and D. M. Tennant, Phys. Rev. Lett. **99**, 134801 (2007).
[5]E. Di Fabrizio, F. Romanato, M. Gentili, S. Cabrini, B. Kaulich, J. Susini, and R. Barrett, Nature (London) **401**, 895 (1999).
[6]K. Jefimovs, J. Vila-Comamala, T. Pilvi, J. Raabe, M. Ritala, and C. David, Phys. Rev. Lett. **99**, 264801 (2007).
[7]L. Alianelli, K. J. S. Sawhney, R. Barrett, A. Malik, I. Pape, M. C. Wilson, and I. M. Loader, "High efficiency nano-focusing optics for synchrotron radiation" (unpublished).
[8]B. Nöhammer, J. Hoszowska, A. K. Freund, and C. David, J. Synchrotron Radiat. **10**, 168 (2003).
[9]F. Isakovic, A. Stein, J. B. Warren, S. Narayanan, M. Sprung, A. R. Sandy, and K. Evans-Lutterodt, J. Synchrotron Radiat. **16**, 8 (2009).
[10]P. W. May and Yu. A. Mankelevich, J. Phys. Chem. C **112**, 12432 (2008).
[11]P. W. May, "Chemical vapour deposition – A route to microcrystalline, nanocrystalline, ultrananocrystalline and single crystal diamond films," Chapter 6, pp. 145–176, in Carbon Based Nanomaterials, Materials Science Foundations (monograph series), Vol. 65–66, edited by N. Ali, A. Öchsner, and W. Ahmed (Trans Tech Publications Inc., Switzerland, 2010).
[12]O. J. L. Fox, J. O. P. Holloway, G. M. Fuge, P. W. May, and M. N. R. Ashfold, "Electrospray deposition of diamond nanoparticle nucleation layers for subsequent CVD diamond growth," in Diamond Electronics and Bioelectronics – Fundamentals to Applications III, edited by P. Bergonzo, J. E. Butler, R. B. Jackman, K. P. Loh, and M. Nesladek (Mater. Res. Soc. Symp. Proc., Warrendale, PA, 2010), Vol. 1203, paper J17-27.
[13]J. E. Butler and I. Oleynik, Philos. Trans. R. Soc. London, Ser. A **366**, 295 (2008).
[14]K. J. S. Sawhney, I. P. Dolbnya, M. K. Tiwari, L. Alianelli, S. M. Scott, G. M. Preece, U. K. Pedersen, and R. D. Walton, AIP Conf. Proc. **1234**, 387 (2010).
[15]L. Alianelli, K. J. S. Sawhney, I. Snigirev, and A. Snigirev, AIP Conf. Proc. **1234**, 633 (2010).