Diamond & Related Materials 65 (2016) 37–41

Contents lists available at ScienceDirect

Diamond & Related Materials

journal homepage: www.elsevier.com/locate/diamond

Large radius of curvature micro-lenses on single crystal diamond for application in monolithic diamond Raman lasers

Hangyu Liu, Sean Reilly, Johannes Herrnsdorf, Enyuan Xie, Vasili G. Savitski, Alan J. Kemp, Erdan Gu *, Martin D. Dawson

Institute of Photonics, University of Strathclyde, Department of Physics, Technology and Innovation Centre, 99 George Street, Glasgow G1 1RD, UK

ARTICLE INFO

Article history:
Received 1 December 2015
Received in revised form 19 January 2016
Accepted 20 January 2016
Available online 22 January 2016

Keywords:
Micro-lens
Radius of curvature
Inductively coupled plasma etching
Single crystal diamond
Monolithic diamond Raman lasers

ABSTRACT

The design and fabrication of large radii of curvature micro-lenses in single crystal chemical vapour deposition diamond is described. An optimised photoresist reflow process and low selectivity inductively coupled plasma etching are used to actualize a uniform array of micro-lenses with radii of curvature of 13 mm or more and a high quality surface of a root-mean-square roughness of 0.18 nm. The processes developed have the potential to achieve diamond micro-lenses with an even larger radius of curvature. These new diamond micro-lenses enable the pulse energy scalable monolithic diamond Raman laser where a large radius of curvature of the micro-lenses is critical.

1. Introduction

Diamond, as one of the most effective optical and optoelectronic materials, has long been studied [1]. The unrivalled properties of wide transparency range (from ~220 nm to 2.5 μm) [2], high thermal conductivity (2000 $Wm^{-1} K^{-1}$) [3] and large Raman gain coefficient (measured to be ~42 cm/GW at 532 nm [4], with previously reported values ranged between 20–75 cm/GW [1,5]), along with recent advances in the growth of single crystal synthetic diamond [6] have led the material being one of the best choices to be used in Raman lasers [7].

In biomedical applications, a diamond Raman laser that is compact and robust and converts widely available pulsed green laser to the yellow–orange spectral region is highly desired [8]. Conventionally, diamond Raman lasers are composed of a pump laser, diamond (the Raman gain medium), and external mirrors used to form a laser resonator [5]. However, by forming micro-lens structures onto the diamond surface and coating with dielectric mirrors, a simpler, more compact and robust monolithic diamond Raman laser can be achieved. This monolithic diamond Raman laser needs minimal alignments of mainly the laser beam through the micro-lens and has no further requirements of external mirrors [9].

To realise such monolithic diamond Raman laser, the optimisation of the fabrication process is of critical importance since the Raman cavity mode is primarily defined by the micro-lens structure [9]. A large cavity mode, and hence a large radius of curvature (ROC) (>10 mm) micro-lens, discussed in Section 2, is crucial to avoid laser induced damage to the diamond and the optical coatings deposited on the diamond surface.

Generally, diamond micro-lenses are fabricated by single-layer photoresist (PR) thermal reflow [10] to form PR micro-lenses and then Ar/O_2 inductively coupled plasma (ICP) etching to transfer PR micro-lenses [11]. This approach yields simplicity, high reproducibility and high lens surface quality. However, the ROCs of diamond micro-lenses fabricated using this method are limited to below 10 mm [11–15]. The difficulties of fabricating diamond micro-lenses with a larger ROC are threefold. Firstly, the PR layer may have a non-uniform thickness due to the edge bead effect during PR coating on a small sample surface [16]. Secondly, and discussed in more detail in Section 3, an edge bulge phenomenon occurs during thermal reflow when the applied PR is too thin. Finally, the conventionally used Ar/O_2 plasma etching does not have the optimal etch selectivity for the transfer of shallow micro-lens patterns.

Here, a novel fabrication process is presented that enables the production of diamond micro-lenses with larger ROCs. This process uses an optimised thermal reflow of multiple-layer PR to achieve a large ROC micro-lens PR mask combining with a low selectivity Ar/Cl_2 ICP etching to transfer the micro-lens PR mask to diamond. Profilometry and optical measurements have shown that the ROCs of the 400 μm diameter diamond micro-lenses are around 13 mm. The fabricated micro-lenses on a single crystal CVD diamond have a root-mean-square roughness of 0.18 nm on an area of 3 × 3 μm^2 of the micro-lens top surface. Such high quality diamond micro-lenses have enabled

* Corresponding author.
E-mail address: erdan.gu@strath.ac.uk (E. Gu).

http://dx.doi.org/10.1016/j.diamond.2016.01.016
0925-9635/

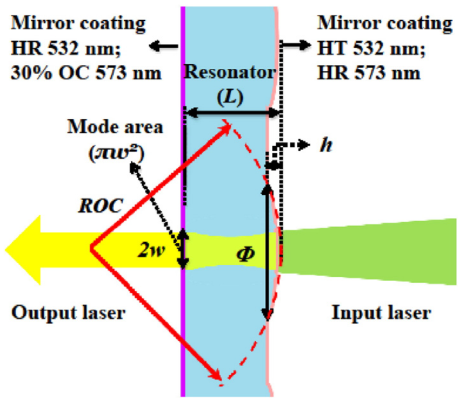

Fig. 1. Schematic of the resonator based on a diamond micro-lens illustrating the key parameters of: beam radius on the plane surface w, mode area πw^2, resonator length L, lens diameter Φ and lens height h. Specifications of mirror coatings: HT 532 nm, high transmission at 532 nm; HR 532/573 nm, high reflectivity at 532/573 nm; 30% OC 573 nm, 30% output coupling at 573 nm.

us to achieve monolithic diamond Raman lasers with a high conversion efficiency.

2. Design of the diamond micro-lenses

In a monolithic diamond Raman laser, two sides of a diamond are coated with dielectric mirror coatings to form a resonator. These coatings can be damaged if the optical field intensity exceeds a certain threshold. Such intense fields are within the realms of monolithic diamond Raman laser operation and therefore this aspect needs careful consideration. The peak intensity (P_{max}) of the pump laser is determined by the mode area:

$$P_{max} = \frac{E}{\Delta\tau \times (\pi w^2)} \quad (1)$$

where w represents the beam radius of the mode area (πw^2) on the plane surface, E denotes the pump energy and $\Delta\tau$ is the pump pulse duration. These key parameters are illustrated in Fig. 1. Hence, a large mode area is critical to enable higher pulse energies without damage. The beam radius w on the plane surface of the micro-lens resonator depends on the ROC of the micro-lens, the wavelength of the laser emission λ, the length of the diamond micro-lens resonator L and the refractive index of diamond n (~2.42) [15]:

$$w = \sqrt{\frac{L \times \lambda}{n \times \pi}} \times \sqrt{\frac{ROC \times n}{L} - 1} \quad (2)$$

From Eq. (2) it can be seen that a large mode area can be achieved by fabricating micro-lenses with a large ROC. In particular, a micro-lens ROC larger than 10 mm is critical if a micro-joule-class diamond Raman laser is required [9].

The ROC in turn depends on the diameter Φ and height h of the micro-lens:

$$ROC = \frac{\Phi^2 + 4h^2}{8h} \quad (3)$$

Thus, to achieve the largest possible ROC, a large aspect ratio of diameter to height is needed. We target a diamond micro-lens of 400 μm in diameter which was chosen as a trade-off between the fabrication challenges described here and the usage of sample area. This choice allows us to fit an 8 × 8 array of micro-lenses on the 4 × 4 mm² area of the diamond sample. Therefore, the ROC of the micro-lens is limited by the achievable minimum height h of the micro-lens. Diamond micro-lenses as shallow as possible with a diameter of 400 μm are desired. Section 3 describes the fabrication process for such large ROC micro-lenses.

3. Fabrication of diamond micro-lenses with a large ROC

The sample used in this work was a 2 mm thick (resonator length L), 4 × 4 mm² high purity single crystal CVD diamond from Element Six Ltd.

To fabricate a shallow micro-lens in diamond using thermal resist reflow and ICP etching, two aspects are crucial: 1. a shallow and uniform micro-lens PR mask and 2. a low selectivity ICP etching to transfer the micro-lens PR mask to diamond. A schematic of the diamond micro-lens fabrication process is illustrated in Fig. 2. To fabricate a shallow and uniform micro-lens PR mask, there are two main issues. The first is the "edge bead effect" [16] which occurs when spin coating the PR on the surface of small samples. This "edge bead effect" affects the uniformity of the spin coated PR and the control of the PR thickness. The second issue is the deformation of the PR pattern, namely the edge bulges, after reflow. This deformation happens due to insufficient surface tension.

To minimise the "edge bead effect", we developed a UV curable optical adhesive holder which is made of Norland 81. The diamond sample was sandwiched in between two glass slides and surrounded with the optical adhesive. The optical adhesive was then UV-cured to form the holder. By removing the glass slides, a free standing holder with the diamond embedded is obtained. This effectively extends the boundaries of the diamond and shifts the edge bead from the diamond to the holder. The edge bead on the holder was then cleaned using a cleanroom swab to ensure that the PR patterns developed on the diamond sample surface had a uniform thickness after using a standard lithography process.

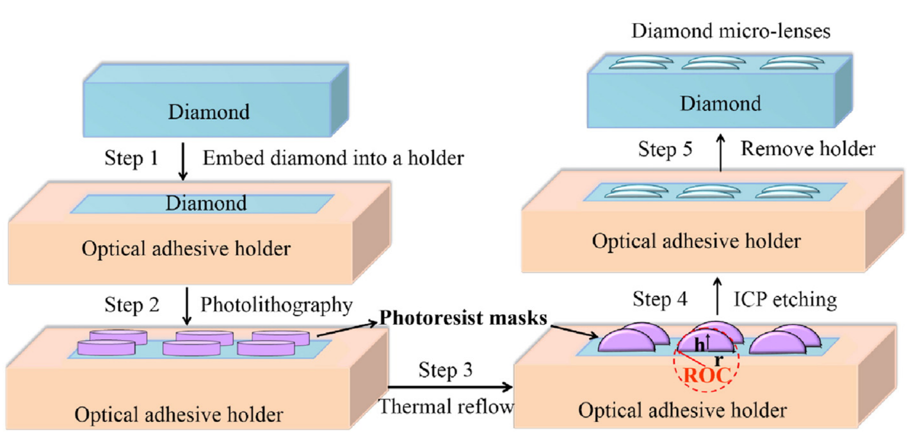

Fig. 2. Schematic of the fabrication process of diamond micro-lenses.

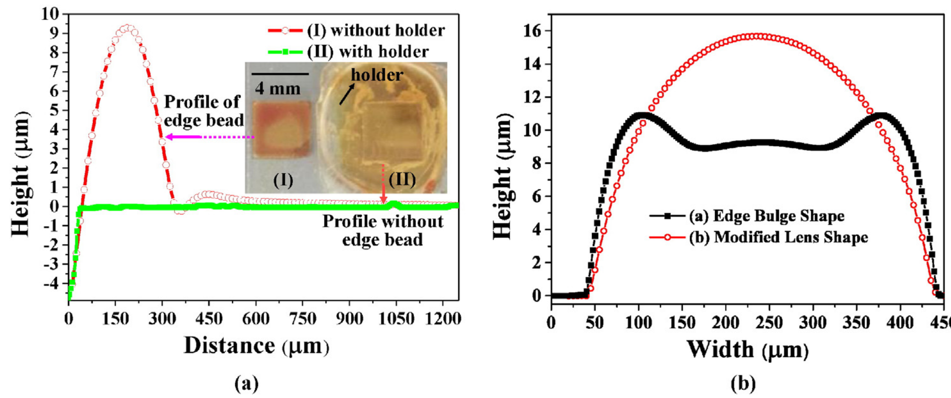

Fig. 3. Measured profiles of: (a) the spin coated PR on surface of two diamond samples with and without a holder, inset are images of the spin coated PR on diamond samples; (b) the measured profiles of edge bulges from single-layer PR reflow and a modified PR micro-lens shape after the optimisation of the PR initial thickness and reflow temperature, initial thicknesses used were 7.5 µm and 11.5 µm, respectively.

Shipley 220 7.0 positive PR was spin coated with an optimised spin speed of 3400 rpm. A Dektak surface profilometer was used to characterise the profiles of the PR. A comparison between the spin coated PR profiles on the diamond surface with and without the holder is shown in Fig. 3(a). Corresponding optical images of the spin coated PR on surface of two diamond samples of the same size with and without holder are shown as inset. It can be seen that the usage of the holder greatly improves the uniformity of the PR layer and thus the usable area.

During our experiments, edge bulges occurred when PR patterns developed using single-layer PR were reflowed. This effect is illustrated in Fig. 3(b). These edge bulges are formed due to the lack of surface tension during PR reflow at inadequate PR thicknesses (i.e. 7.5 µm illustrated in Fig. 3(b)). It is found that at a certain reflow temperature, there is a minimum PR thickness above which the reflowed PR will form the desired micro-lens shape. A detailed examination of this phenomenon will be reported elsewhere. It is desirable to fabricate micro-lenses using a PR layer close to the minimum thickness since this minimum thickness allows the minimum h of the reflowed PR micro-lens, and hence the largest possible ROC at a certain diameter as expressed in Eq. (3). An optimised PR thickness of 11.5 µm, which is close to the minimum thickness at a reflow temperature of 135 °C, is used to provide a high uniformity and pattern fidelity of the lithographically defined PR patterns. This PR thickness is not possible by deposition of a single PR layer and therefore a double-layer deposition was employed. After 3 min reflow of the PR patterns on a hotplate, a uniform array of PR micro-lenses was formed on the surface of the diamond sample. The resulting PR micro-lens had an ROC as large as 1.3 mm. The PR micro-lenses were then transferred to diamond using an ICP etching via a multiplex ICP etch tool (SPTS Technologies Ltd.).

ICP etching of diamond is conventionally performed using an Ar/O_2 plasma. However, the selectivity of this etch recipe, which is defined as the ratio of the etch rate in diamond to the etch rate in PR, is higher than 0.22 [11]. This selectivity limits the ROC of transferred diamond micro-lenses to less than 9 mm. Thus, an etch recipe with a lower selectivity is needed. The chlorine-based diamond ICP etch developed by our group has a selectivity of 0.1, half that of the conventional Ar/O_2 etching under the same conditions [17]. Transferring the micro-lens PR mask to diamond is therefore performed using ICP etching with an Ar/Cl_2 plasma. This enables shallow etched structures and therefore a large ROC of the resulting diamond micro-lens. The etch recipe used here was as follows: a pressure of 5 mTorr, an Ar flow rate of 25 sccm, a Cl_2 flow rate of 40 sccm, a platen power of 300 W and a coil power of 400 W, which generated an etch rate of 75 nm/min.

4. Results and discussion

The fabricated diamond micro-lenses were characterised and applied in a monolithic diamond Raman laser. Optical images of the fabricated large ROC diamond micro-lenses are shown in Fig. 4.

The heights of these micro-lenses were measured to be 1.7 µm or less using a Dektak surface profilometer. Fig. 5(a) shows the measured profiles of three adjacent micro-lenses indicated by the black dotted arrow in Fig. 4. These measured profiles have a uniform height and the ROCs of these micro-lenses are calculated to be 12 mm using Eq. (3). Fitting with an equation for a circle as shown in Fig. 5(a) yields slightly larger ROCs of 13 mm or more. For this fit, only data points close to the centre of the micro-lenses were taken into account. This restriction of the fit region was made because the lens profiles deviate from the spherical shape at the bottom of the profiles. Since the laser mode is confined to a region close to the centre of the micro-lens, the ROC value obtained from this fit is more relevant than the one obtained by Eq. (3). The deviation from the spherical shape results from the original high aspect ratio PR micro-lens and the subsequent low selectivity etching process and is not thought to influence the laser operation.

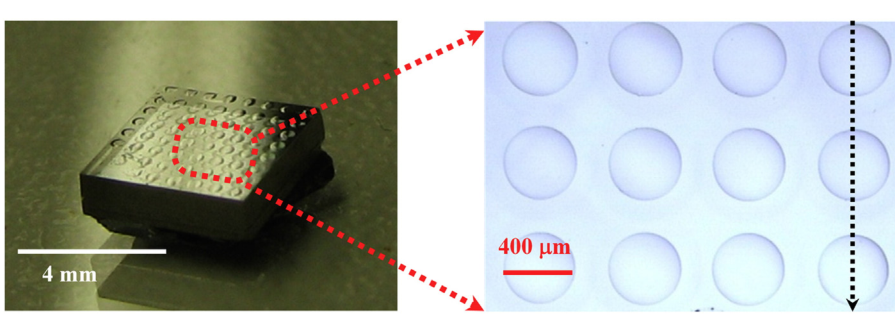

Fig. 4. Optical images of fabricated diamond micro-lenses with large ROCs.

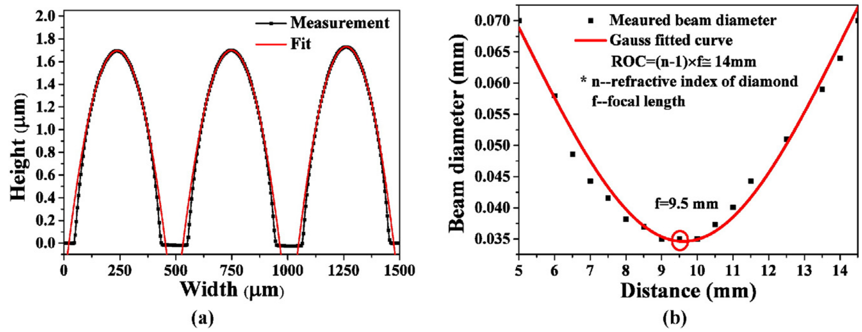

Fig. 5. (a) Measured and fitted profiles of a column of fabricated diamond micro-lenses as shown in Fig. 4 along the black dotted arrow and (b) focal length measurement of a representative diamond micro-lens fabricated with the height of 1.7 μm or less, the distance was measured in reference to the place where the diamond sample was located.

For further characterisation, the focal length of a representative diamond micro-lens was measured optically. The collimated beam from a He–Ne laser was passed through the micro-lens and the beam diameters were measured as a function of position in reference to the place where the diamond sample was located along the principal axis using the knife-edge technique [18]. The measurement yielded a focal length of 9.5 ± 0.5 mm as shown in Fig. 5(b). The ROC was then calculated using the thin lens approximation:

$$\mathrm{ROC} \cong (n-1) \times f \tag{4}$$

where n is the refractive index of the diamond (2.42) and f is the focal length. The ROC calculated from Eq. (4) is 13.5 mm, which is consistent with the values obtained from the profile measurement.

Atomic force microscopy measurement shows that the diamond micro-lenses have a root-mean-square roughness of only 0.18 nm on an area of 3 × 3 μm^2 measured on the top surface of micro-lenses. The unetched diamond surface was measured to have a roughness of around 1 nm. The obtained micro-lens roughness value was comparable to the value reported by Lee et al. [17] and the work reported by M. Karlsson et al. [13]. Such a smooth surface is important for mirror coating so as to form a high quality laser resonator.

A monolithic diamond Raman laser based on these micro-lenses was reported in [9]. This compact device achieved near quantum limited conversion of pulsed green laser to the yellow–orange spectral region. A picture of the resulting compact and robust monolithic diamond Raman laser in operation is shown in Fig. 6. This monolithic diamond Raman laser was pumped by a 532 nm pulsed laser with 1.5 ns pulses. The threshold pump pulse energy was 1.5 μJ. A conversion efficiency from pump to the combined Raman output (1st, 2nd and 3rd order Raman shifts) of over 80% was achieved, with a maximum Raman energy of 13.4 μJ at a pump energy of 16 μJ.

By choosing the appropriate mirror coatings on the facets of the micro-lensed resonator, it should be possible to operate at any wavelength range throughout diamond's large window of transparency if an appropriate pump laser is available. The large ROC diamond micro-lenses fabricated here may also be used in application in higher power monolithic semiconductor disk lasers [15]. The technique developed in this work can also be used to fabricate diamond micro-lenses with even larger ROCs with a bigger lens diameter than the ones reported here. Furthermore, this technique might also find use in developing other diamond components such as solid immersion lenses for photon collection [19,20] and aspheric lenses.

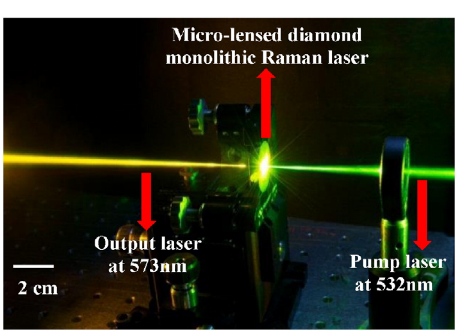

Fig. 6. Demonstration of the monolithic diamond Raman laser based on the micro-lens resonator using a pulsed laser of 1.5 ns duration at a pump pulse energy of 1.5 μJ.

5. Conclusion

Diamond micro-lenses with large ROCs of 13 mm or more were achieved using PR thermal reflow and ICP etching. The ROCs of these diamond micro-lenses were measured and confirmed by profilometry and optical measurements. These diamond micro-lenses have a very smooth surface of 0.18 nm root-mean-square roughness. By mirror coating these diamond micro-lenses, a resonator is formed which enables compact and robust monolithic diamond Raman lasers with high conversion efficiency.

Prime novelty statement

In this work, we developed a novel process of fabricating diamond micro-lenses with large radius of curvatures. These diamond micro-lenses were applied to form a stable resonator and enabled a compact and robust monolithic diamond Raman laser.

Acknowledgement

Hangyu Liu and Sean Reilly acknowledge a China Scholarship Council studentship (File No. 201206950018) and an EPSRC DTA studentship, respectively. Alan Kemp acknowledges a Fraunhofer UK/RAEng Research Chair. Thanks to the funding from the European Research Council (DiaL: 278389) and the UK Engineering and Physical Sciences Research Council (EPSRC; EP/I029141/1). Thanks to Ricardo Ferreira and Dr. Ross Leyman for useful discussions.

References

[1] R. Mildren, J. Rabeau, Optical Engineering of Diamond, Wiley, 2013 Chapter 1.
[2] A. Tallaire, A.T. Collins, D. Charles, J. Achard, R. Sussmann, A. Gicquel, M.E. Newton, A.M. Edmonds, R.J. Cruddace, Characterisation of high-quality thick single-crystal diamond grown by CVD with a low nitrogen addition, Diam. Relat. Mater. 15 (2006) 1700–1707.
[3] S.E. Coe, R.S. Sussmann, Optical, thermal and mechanical properties of CVD diamond, Diam. Relat. Mater. 9 (2000) 1726–1729.
[4] V.G. Savitski, S. Reilly, A.J. Kemp, Steady-state Raman gain in diamond as a function of pump wavelength, IEEE J. Quantum Electron. 49 (2013) 218–223.
[5] R.P. Mildren, J.E. Butler, J.R. Rabeau, CVD-diamond external cavity Raman laser at 573 nm, Opt. Express 16 (2008) 18950–18955.
[6] I. Friel, S.L. Geoghegan, D.J. Twitchen, G.A. Scarsbrook, Development of high quality single crystal diamond for novel laser applications, Proc. SPIE 7838 (2010) 783818–783819.

[7] V.G. Savitski, I. Friel, J.E. Hastie, M.D. Dawson, D. Burns, A.J. Kemp, Characterization of single-crystal synthetic diamond for multi-watt continuous-wave Raman lasers, IEEE J. Quantum Electron. 48 (2012) 328–337.
[8] A.D. Greentree, S. Prawer, Yellow lasers: a little diamond goes a long way, Nat. Photonics 4 (2010) 202–203.
[9] S. Reilly, V.G. Savitski, H. Liu, E. Gu, M.D. Dawson, A.J. Kemp, Monolithic diamond Raman laser, Opt. Lett. 40 (2015) 930–933.
[10] Z.D. Popovic, R.A. Sprague, G.A. Connell, Technique for monolithic fabrication of microlens arrays, Appl. Opt. 27 (1988) 1281–1284.
[11] H.W. Choi, E. Gu, C. Liu, C. Griffin, J.M. Girkin, I.M. Watson, M.D. Dawson, Fabrication of natural diamond microlenses by plasma etching, J. Vac. Sci. Technol. B 23 (2005) 130–132.
[12] E. Woerner, C. Wild, W. Mueller-Sebert, P. Koidl, CVD-diamond optical lenses, Diam. Relat. Mater. 10 (2001) 557–560.
[13] M. Karlsson, F. Nikolajeff, Diamond micro-optics: microlenses and antireflection structured surfaces for the infrared spectral region, Opt. Express 11 (2003) 502–507.
[14] E. Gu, H.W. Choi, C. Liu, C. Griffin, J.M. Girkin, I.M. Watson, M.D. Dawson, G. McConnell, A.M. Gurney, Reflection/transmission confocal microscopy characterization of single-crystal diamond microlens arrays, Appl. Phys. Lett. 84 (2004) 2754–2756.
[15] N. Laurand, C.L. Lee, E. Gu, S. Calvez, M.D. Dawson, Power-scaling of diamond microlensed microchip semiconductor disk lasers, IEEE Photon. Technol. Lett. 21 (2009) 152–154.
[16] T. Dobroth, L. Erwin, Causes of edge beads in cast films, Polym. Eng. Sci. 26 (1986) 462–467.
[17] C.L. Lee, E. Gu, M.D. Dawson, I. Friel, G.A. Scarsbrook, Etching and micro-optics fabrication in diamond using chlorine-based inductively-coupled plasma, Diam. Relat. Mater. 17 (2008) 1292–1296.
[18] A.E. Siegman, M.W. Sasnett, T.F. Johnston, Choice of clip levels for beam width measurements using knife-edge techniques, IEEE J. Quantum Electron. 27 (1991) 1098–1104.
[19] J.P. Hadden, J.P. Harrison, A.C. Stanley-Clarke, L. Marseglia, Y.-L.D. Ho, B.R. Patton, J.L. O'Brien, J.G. Rarity, Strongly enhanced photon collection from diamond defect centers under microfabricated integrated solid immersion lenses, Appl. Phys. Lett. 97 (2010) 241901.
[20] W. Pfaff, B.J. Hensen, H. Bernien, S.B. van Dam, M.S. Blok, T.H. Taminiau, M.J. Tiggelman, R.N. Schouten, M. Markham, D.J. Twitchen, R. Hanson, Unconditional quantum teleportation between distant solid-state quantum bits, Science 345 (2014) 532–535.