Optical nanolithography using a scanning near-field probe with an integrated light source

James W. Kingsley,[1] Sumon K. Ray,[1] Ali M. Adawi,[1] Graham J. Leggett,[2] and David G. Lidzey[1,a)]
[1]*Department of Physics and Astronomy, The University of Sheffield, Sheffield S3 7RH, United Kingdom*
[2]*Department of Chemistry, The University of Sheffield, Sheffield S3 7HF, United Kingdom*

(Received 27 August 2008; accepted 3 November 2008; published online 24 November 2008)

An ultracompact near-field optical probe is described that is based on a single, integrated assembly consisting of a gallium nitride (GaN) light-emitting diode (LED), a microlens, and a cantilever assembly containing a hollow pyramidal probe with a subwavelength aperture at its apex. The LED emits ultraviolet light and may be used as a light source for near-field photolithographic exposure. Using this simple device compatible with many commercial atomic force microscope systems, it is possible to form nanostructures in photoresist with a resolution of 35 nm, corresponding to $\lambda/10$.
 [DOI: 10.1063/1.3032912]

Scanning near-field optical microscopy (SNOM) can be used to image[1–5] and pattern[6–11] surfaces with a resolution significantly better than conventional far-field optical techniques and is often used to study systems in which the characteristic length scale of interest is below the optical resolution limit of standard or confocal microscopes. Examples of molecular systems imaged by SNOM include conjugated polymers,[12–14] chromosomes,[15] and DNA (Ref. 16) with a resolution typically less than 100 nm. SNOM based lithography has also been performed on conjugated polymers[13,17,18] and self-assembled monolayers[19–22] with a resolution as fine as 9 nm on certain substrates.[23]

While there are several different ways to implement SNOM, cantilever based designs offer many advantages as their construction is similar to regular atomic force microscopes. In such a configuration, an aperture is positioned at the apex of a probe located at the end of a flexible cantilever. The sample of interest is then placed in contact or very close to the aperture, which is scanned by a piezostage. A laser and segmented photodiode monitors the deflection of the cantilever as it moves across the surface providing feedback to the piezostage such that a constant force can be applied (for a review of SNOM systems, see Hecht *et al.*[1]). The major difference between a cantilever based SNOM system and a standard atomic force microscope (AFM) is that an optical path is provided for a second laser beam to illuminate the aperture, and, in the case of imaging, to collect the light emitted or scattered from the tip/sample. However, the alignment of the illumination laser and collection optics adds significantly to the complexity of both the system and its operation. As such, cantilever based SNOM techniques have not been as widely adopted as other scanning probe microscopy (SPM) techniques despite their other attractive features.

Various approaches have been explored to create scanning probes with integrated light sources. These include organic light-emitting diodes (LEDs) fabricated on the end of a probe,[24] sharpened silicon LEDs,[25] semiconductor nanoparticles incorporated into an electrode gap,[26] and gallium arsenide based heterostructures engineered into a cantilever.[27] However, none of these approaches combines the attributes of high spatial resolution in imaging or lithography together with the use of ultraviolet (UV) light emission.

In the present work we report a SNOM probe with monolithically integrated UV LED and demonstrate its use to expose a standard photoresist, generating feature sizes down to 35 nm full width at half maximum (FWHM), better than $\lambda/10$. The integration of a light source with the probe addresses the practical problems encountered previously in cantilever SNOM systems and greatly facilitates the near-field excitation of a surface. Because the device we describe is capable of being used in many commercial AFM systems, it may enable powerful near-field optical techniques to be employed more widely than is currently possible. The light source we used here also operates at UV wavelengths where near-field photolithography of many photoresists is possible and where there is much interesting photochemistry that can be observed through spectroscopic imaging.

Figure 1(a) shows schematically the operation of the system. It is fabricated from three stacked substrates consisting of (i) a LED fabricated on a GaN wafer, (ii) a microlens to focus the light from the LED into an image with a spatial filter incorporated to block stray light, and (iii) a cantilever style SNOM probe with its aperture positioned at the focus of the image produced by the microlens.

To construct the system, all three components are fabricated individually using standard processes and are then bonded together producing a stack with total thickness of around 1 mm. Our design permits cantilever deflection to be measured using a laser as it occurs in a standard AFM. A scanning electron microscope image of a cantilever aligned over a microlens and spatial filter is shown in Fig. 1(b).

The LEDs were fabricated using a commercial UV gallium nitride (GaN) LED wafer grown on a sapphire substrate (Nitride Semiconductors NS385W) using standard processes that resulted in the structure shown in Fig. 2(a). The light emitting region is based on a series of epitaxial quantum wells positioned between *p*- and *n*-doped GaN. Individual LEDs are made by creating mesas through which a current is passed. The mesas were created with an inductively coupled plasma using a photoresist as an etch mask while electrical connections to the wafer are provided by Ohmic contacts that consist of Ni/Au (*N*-type) and Ti/Au (*P*-type) deposited by thermal evaporation. The edges of the mesa were coated in silicon nitride to insulate them from the *p*-contact electrical

a)Electronic mail: d.g.lidzey@sheffield.ac.uk.

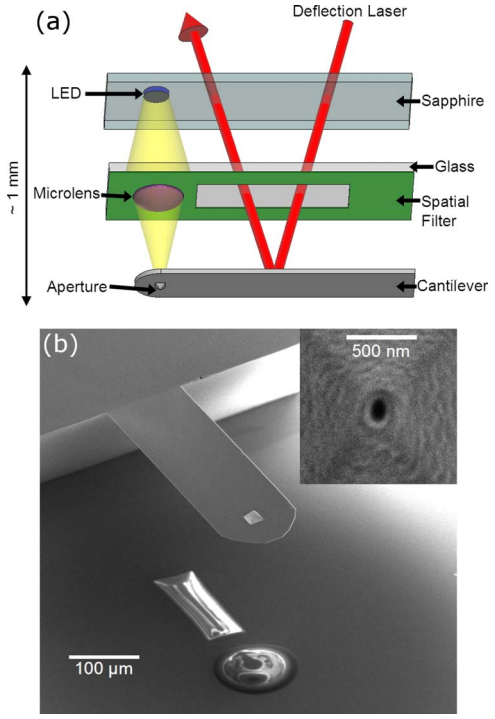

FIG. 1. (Color online) Layout of the integrated SNOM probe. (a) Schematic showing the three substrates: LED (top), microlens with spatial filter (middle), and SNOM cantilever (bottom). (b) A SEM image of the cantilever above the microlens and deflection laser window with the inset showing the SNOM aperture.

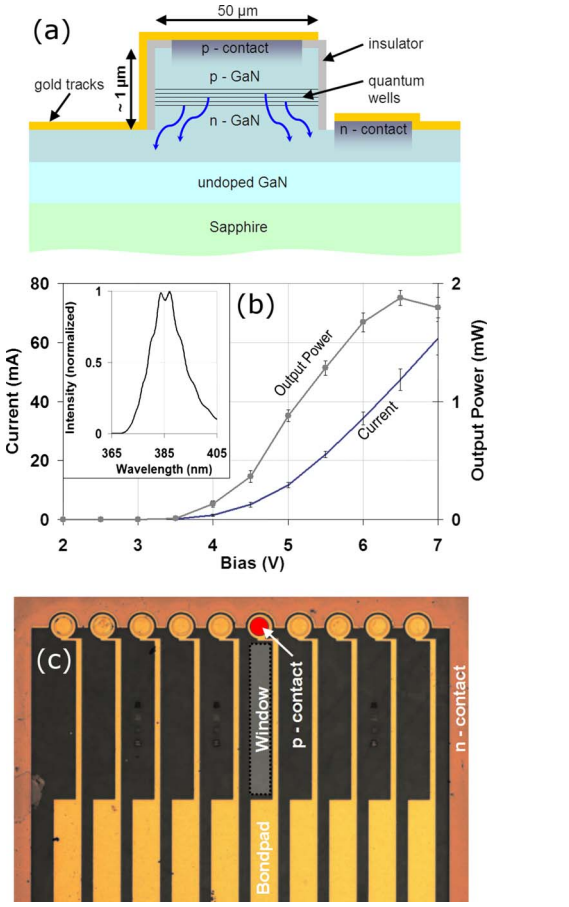

FIG. 2. (Color online) LED structure and performance. (a) Schematic of the LED layout. (b) Input current and output power as a function of applied bias with (inset) the electroluminescence spectra. (c) Optical micrograph of a LED array showing the optical window for the deflection laser.

track that links the LED to the bonding pads. Finally, the underside of the wafer was polished to an optical finish.

Figure 2(b) shows the current-voltage and optical power curves as measured by a calibrated photodiode and averaged over eight LEDs. The devices emit a maximum optical power output of nearly 2 mW at an applied bias of 6.5 V, above which thermal rollover[28] occurs and output decreases. We note, however, that even at maximum output power the LEDs have been operated for several tens of hours with no degradation in performance. As can be seen in the inset of Fig. 2(b) the device emission peaks at 385 nm. An optical micrograph of a series of LEDs is shown in Fig. 2(c). In this work, only a single LED from the linear array of devices was used; however, such a LED array could be integrated with an array of microlenses and cantilevers, permitting parallel lithography to be performed.

The design of the LED is such that the majority of light is emitted downward [relative to Fig. 2(a)] through the wafer. This allowed easy attachment of the LEDs to a glass slide upon which a microlens and spatial filter were fabricated. The spatial filter is used to reduce stray light and consists of a continuous layer of thermally evaporated aluminum into which two apertures were defined by photoresist lift-off. The first circular aperture was placed in the location where the microlens was to be fabricated while the second rectangular aperture was placed directly below the cantilever to allow transmission of the deflection laser.

The microlenses were fabricated above the spatial filter and made using a standard photolithographic technique[29–32] by creating cylinders of photoresist that were reflowed such that the surface tension pulled them into a spherical shape. Cantilevers were bought commercially from Witec Ltd (Alpha 300 S) and used as received except for their apertures being enlarged slightly to 150 nm using a focused ion beam (FIB) in order to increase their transmission.

In order to maximize the amount of light incident on the aperture, it is necessary to accurately focus the LED image onto the back of the cantilever. For the cantilevers used here, the image was approximately 400 μm below the microlens substrate. Focusing was achieved by optimizing the focal length of the microlens and adjusting the thickness of a spacer placed between it and the LED. For the results reported here a lens diameter of 130 μm was used with a focal length of approximately 300 μm.

Construction of the system was performed manually under a microscope using a vacuum pickup tool and micromanipulator to align the components before they were bonded in place. The integrated probe was then placed on the piezostage of a Witec Alpha SNOM and the photoresist sample approached until the cantilever deflected. Once in contact, the piezo was used to move the probe across the surface.

To test the capability of the system, lithography was performed on glass substrates spin coated in BPRS 100 photoresist at 4000 rpm for 30 s and then baked on a hotplate at 60 °C for 60 s. After exposure, the samples were developed for 60 s using Fujifilm PLSI mixed 1:3 with water and then imaged using a Veeco Multimode AFM.

Using low exposure doses, an average feature size of below 100 nm was achieved. Figure 3(a) shows an array of dots produced with a periodicity of 200 nm with a cross section taken through the image shown in Fig. 3(b). The average linewidth is 33 nm as measured using the FWHM

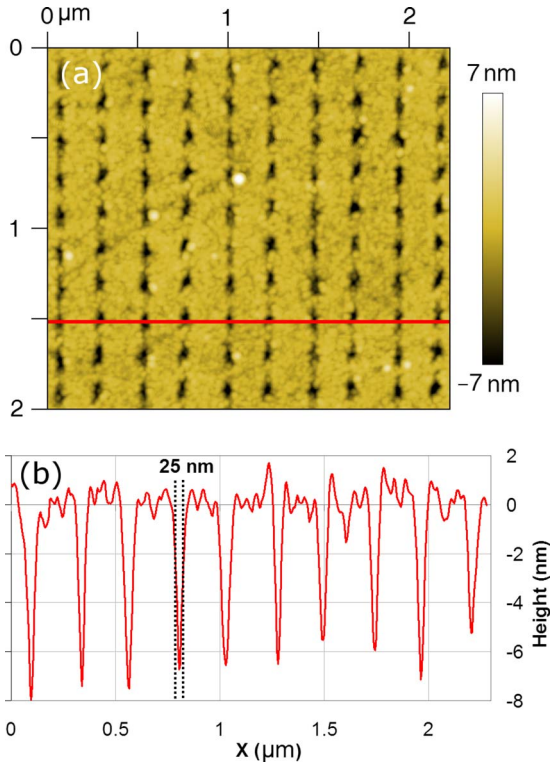

FIG. 3. (Color online) Photoresist exposed by the integrated SNOM probe. (a) An AFM image of an array of dots with 200 nm period. (b) A cross section of the cut shown in (a).

with a standard deviation of 5 nm (the minimum recorded feature size was 25 nm). In the vertical direction [relative to Fig. 3(a)] the average FWHM is slightly larger at an average of 85 nm with a standard deviation of 18 nm. We believe that this effect results from a combination of two factors. First, the LED was not switched off in between writing individual dots—instead the probe was moved as quickly as the piezostage would allow between sites, leading to a small amount of unwanted exposure in the scan direction. Second, due to issues with the FIB milling software, the enlarged probe apertures were slightly elliptical with their major and minor axes being 200 and 150 nm, respectively (as imaged using a SEM). As the major axis coincided with the scan direction, it is likely that this ellipticity partially contributed to the coarser patterning resolution achieved parallel to the scan direction. For completeness we also scanned the photoresist using identical tips that had not been enlarged and therefore transmitted very little light. This did not produce any discernable features, eliminating scratching as the surface patterning mechanism and also suggesting that the tip temperature during operation is below the melting point of the photoresist at ~100 °C.

As reported elsewhere,[2] the features produced using this probe are smaller (FWHM) than the aperture size resulting from inherent nonlinearity of the exposure process. It is also noticeable that the lithography is approaching the limit of the photoresist since the grain size of the films is approximately 20 nm as is partially visible in Fig. 3(a).

In conclusion, we have developed a scanning probe with an integrated light source suitable for use in SNOM. The probe requires no external optics and so can potentially be used in a standard atomic force microscope. Light is provided to the cantilever by a high brightness GaN LED along with a microlens to focus the light. Using this probe, a standard photoresist was patterned with a resolution comfortably below 100 nm. Finally, we emphasize that the single cantilever devices presented here could be readily adapted for use in a multiple cantilever based system, potentially allowing surfaces to be patterned in parallel at high resolution and at high speed.

We would like to thank Research Councils U.K. for funding this work through a Basic Technology award for the SNOMIPEDE project. We also thank the ERA foundation, Sheffield University, and Yorkshire Forward for financial support. We also thank Rob Airey and Peter Parbrook for technical assistance and useful discussions.

[1] B. Hecht, B. Sick, U. P. Wild, V. Deckert, R. Zenobi, O. J. F. Martin, and D. W. Pohl, J. Chem. Phys. **112**, 7761 (2000).
[2] D. W. Pohl, Philos. Trans. R. Soc. London, Ser. A **362**, 701 (2004).
[3] D. Courjon, *Near-Field Microscopy and Near-Field Optics* (Imperial College Press, London, 2003).
[4] J. W. P. Hsu, Mater. Sci. Eng., R. **33**, 1 (2001).
[5] A. Rasmussen and V. Deckert, Anal. Bioanal. Chem. **381**, 165 (2005).
[6] A. A. Tseng, Opt. Laser Technol. **39**, 514 (2007).
[7] S. Wegscheider, A. Kirsch, J. Mlynek, and G. Krausch, Thin Solid Films **264**, 264 (1995).
[8] Y. Lin, M. H. Hong, W. J. Wang, Y. Z. Law, and T. C. Chong, Appl. Phys. A: Mater. Sci. Process. **80**, 461 (2005).
[9] M. M. Alkaisi, R. J. Blaikie, S. J. McNab, R. Cheung, and D. R. S. Cumming, Appl. Phys. Lett. **75**, 3560 (1999).
[10] Y. Lin, M. H. Honga, W. J. Wang, Z. B. Wang, G. X. Chenc, Q. Xie, L. S. Tan, and T. C. Chonga, Sens. Actuators, A **133**, 311 (2007).
[11] S. J. Kwon, Y. M. Jeong, and S. H. Jeong, Appl. Phys. A: Mater. Sci. Process. **86**, 11 (2006).
[12] A. J. Cadby, R. Dean, C. Elliott, R. A. L. Jones, A. M. Fox, and D. G. Lidzey, Adv. Mater. (Weinheim, Ger.) **19**, 107 (2007).
[13] D. Richards and F. Cacialli, Philos. Trans. R. Soc. London, Ser. A **362**, 771 (2004).
[14] A. Cadby, R. Dean, A. M. Fox, R. A. L. Jones, and D. G. Lidzey, Nano Lett. **5**, 2232 (2005).
[15] R. M. Baylis, S. H. Doak, and M. D. Holton, Ultramicroscopy **107**, 308 (2007).
[16] T. Yoshino, S. Sugiyama, S. Hagiwara, D. Fukushi, M. Shichiri, H. Nakao, J. Kim, T. Hirose, H. Muramatsu, and T. Ohtani, Ultramicroscopy **97**, 81 (2003).
[17] R. Riehn, A. Charas, J. Morgado, and F. Cacialli, Appl. Phys. Lett. **82**, 526 (2003).
[18] F. Cacialli, R. Riehn, A. Downes, G. Latini, A. Charas, and J. Morgado, Ultramicroscopy **100**, 449 (2004).
[19] S. Kramer, R. R. Fuierer, and C. B. Gorman, Chem. Rev. (Washington, D.C.) **103**, 4367 (2003).
[20] S. Sun, K. S. L. Chong, and G. J. J. Leggett, J. Am. Chem. Soc. **124**, 2414 (2002).
[21] S. Sun, M. Montague, K. Critchley, M.-S. Chen, W. J. Dressick, S. D. Evans, and G. Leggett, Nano Lett. **6**, 29 (2006).
[22] P. Burgos, M. Geoghegan, and G. J. Leggett, Nano Lett. **7**, 3747 (2007).
[23] M. Montague, R. E. Ducker, K. S. L. Chong, R. J. Manning, F. J. M. Rutten, M. C. Davies, and G. J. Leggett, Langmuir **23**, 7328 (2007).
[24] Y. Zhao, K. H. An, S. Chen, B. O'Connor, K. P. Pipe, and M. Shtein, Nano Lett. **7**, 3645 (2007).
[25] K. Hoshino, L. J. Rozanski, D. A. Vanden Bout, and X. Zhang, Appl. Phys. Lett. **92**, 131106 (2008).
[26] K. Hoshino, L. J. Rozanski, D. A. Vanden Bout, and X. J. Zhang, J. Microelectromech. Syst. **17**, 4 (2008).
[27] S. Heisig, O. Rudow, and E. Oesterschulzea, Appl. Phys. Lett. **77**, 1071 (2000).
[28] M. P. Liao, IEEE Photonics Technol. Lett. **19**, 2000 (2007).
[29] H. Ottevaere, R. Cox, H. P. Herzig, T. Miyashita, K. Naessens, M. Taghizadeh, R. Volkel, H. J. Woo, and H. J. Thienpont, J. Opt. A, Pure Appl. Opt. **8**, S407 (2006).
[30] P. Nussbaum, R. Volkely, H. P. Herzigy, M. Eisnerz, and S. Haselbeckz, Pure Appl. Opt. **6**, 617 (1997).
[31] H. Hocheng, C.-T. Pan, and C.-C. Cheng, Jpn. J. Appl. Phys., Part 2 **42**, 4044 (2003).
[32] C. W. Jeon, E. Gu, C. Liu, J. M. Girkin, and M. D. Dawson, IEEE Photonics Technol. Lett. **17**, 1887 (2005).