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High integrity interconnection of silver submicron/nanoparticles on silicon wafer by femtosecond laser irradiation

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Abstract

Welding of nanomaterials is a promising technique for constructing nanodevices with robust mechanical properties. To date, fabrication of these devices is limited because of difficulties in restricting damage to the nanomaterials during the welding process. In this work, by utilizing very low fluence (~900 μJ cm^{-2}) femtosecond (fs) laser irradiation, we have produced a metallic interconnection between two adjacent silver (Ag) submicron/nanoparticles which were fixed on a silicon (Si) wafer after fs laser deposition. No additional filler material was used, and the connected particles remain almost damage free. Observation of the morphology before and after joining and finite difference time domain simulations indicate that the interconnection can be attributed to plasmonic excitation in the Ag submicron/nanoparticles. Concentration of energy between the particles leads to local ablation followed by re-deposition of the ablated material to form a bridging link that joins the two particles. This welding technique shows potential applications in the fabrication of nanodevices.

Keywords: femtosecond laser, high-integrity interconnection, silver nanoparticles

(Some figures may appear in colour only in the online journal)

1. Introduction

To date, much effort has been devoted to developing nanodevices for applications in nanoelectronics [1], nanophotonics [2], and biosensors [3] based on plasmonic nanomaterials. In order to obtain tailored functional properties, nanodevices are usually fabricated via the assembly of nanoscale building blocks such as nanoparticles (NPs) and nanowires (NWs) [4, 5]. A variety of nanowelding methods including thermal heating [6], high energy electron beam irradiation [7], ultrasonic nanowelding [8], and nanosoldering [9] have been utilized for fabrication, but these techniques are limited by inadequate control over spatial heating, the high cost of high energy electron beam system, and the intrinsic complexity of the nanosoldering system.

Recently, welding of nanomaterials with localized surface plasmon (LSP) energy (plasmonic welding) has been shown to be a promising technology for the construction of nanodevices. The localization of LSP-induced effects such as enhancement of the optical gradient force [10] as well as site-specific heating [11] and ablation [12] can restrict damage in nanomaterials optimizing the welding process. The concept of plasmonic welding was first demonstrated by Mafuné *et al* [13] who soldered Pt NPs with Au NPs selectively melted by excitation of the LSP resonance in Au. The fs laser is an ideal tool for plasmonic welding, because the 'non-thermal' nature and high intensity of fs laser irradiation can produce highly localized structural transformations in nanomaterials as a result of plasmonic enhancement effects [11, 14]. Other experiments have shown that plasmonic welding of Ag NPs in aqueous solution can be optimized by controlling the location of the LSP-induced electric field enhancement (or hotspots) during irradiation with fs laser pulses [15, 16]. The random orientation and location of NPs in aqueous solution due to Brownian motion during irradiation precludes close control over welding conditions and

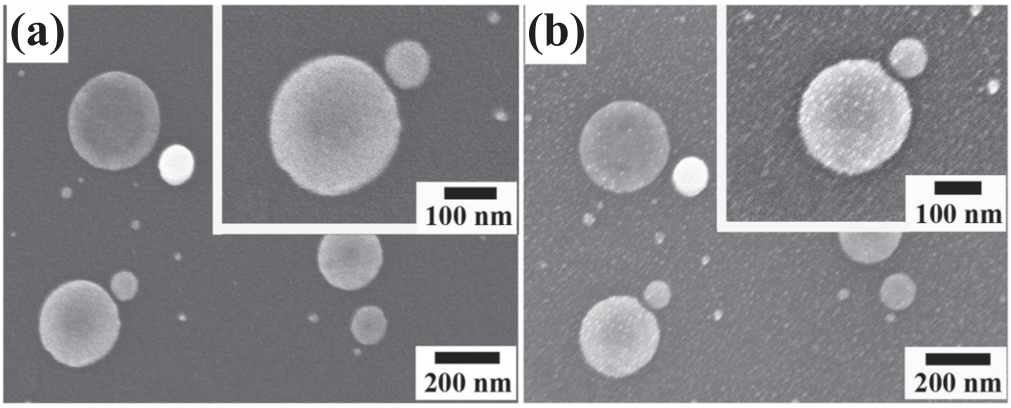

Figure 1. Scanning electron microscopy (SEM) images of the morphology of deposited Ag particles before and after fs laser irradiation at a fluence ~900 μJ cm^{-2} for 50 s. (a) Before irradiation; (b) same area after irradiation. The inset images are magnified view.

introduces additional damage to fabricated structures. In all cases, minimal damage to nanomaterials during plasmonic welding is obtained at low laser fluence.

The best condition for damage-free joining of NPs occurs when particles are 'pinned' to a substrate and then irradiated at low laser fluence. These conditions can be replicated by attaching Ag NPs to a Si substrate followed by irradiation at low fs laser fluence. In this report, we show that damage-free metallic interconnection of Ag submicron/nanoparticles on a Si wafer can be obtained via the excitation of LSP-induced hotspots at fs laser fluence as low as ~900 μJ cm^{-2}. We suggest that this new technique may have potential applications in the fabrication of nanodevices.

2. Experimental and simulation

Ag particles were deposited on a Si wafer (as received) by fs laser ablation of an Ag target. The laser (Coherent) is operated at a wavelength of 800 nm with a 35 fs pulse duration, and the repetition rate is 1 KHz. The fluence used to ablate the Ag target was ~1.4 J cm^{-2} and ablation proceeded for 10 s. After deposition, particles on the Si surface were then irradiated using the same fs laser at fluences ~900 μJ cm^{-2} or 1300 μJ cm^{-2} for 50 s. Both ablation/deposition and irradiation/joining operations were carried out in vacuum at a base pressure of ~10^{-6} Torr. The morphology of selected Ag particles before and after laser irradiation was compared using scanning electron microscopy (SEM, LEO 1550) and high magnification images of welded structures were obtained using transmission electron microscopy (TEM, JEOL, JEM-2010F). Commercial finite difference time domain (FDTD) software (Lumerical Solutions) was used to identify the electric field enhancement in the Ag particles irradiated at 800 nm.

3. Results and discussions

Figure 1 shows the morphology of deposited Ag submicron/nanoparticles before and after 50 s of laser irradiation at a fluence of ~900 μJ cm^{-2}. Comparison of these images shows that laser irradiation did not change the position of the deposited Ag particles, although the particles were slightly ablated (see the larger particle in the inset image of figure 1(b)). Under the same irradiation conditions, some closely-spaced Ag particles (~30 nm separation) become welded together (figures 2(a), (b)). There is no apparent damage to these particles apart from a slight change in morphology near the weld area. Since the particles did not move during the irradiation process, welding occurs as the result of material filling the small gap between the particles. This is shown in the inset images of figures 2(a), (b) where two separated particles are seen to be interconnected by filler material, forming dumbbell shaped structures. This filler material arises from ablation of the Ag particles (the inset image of figure 1(b)) due to LSP-induced hotspots in the vicinity of the gap between the particles as reported in a previous study [17]. This ablation is clearly observed in the welded Ag particles (see the left particle in figures 2(c), (d)) when the samples were tilted by 75° for clearer observation. Figures 2(c), (d) also show that the area below the weld did not contain filler material implying that the weld was formed by highly directional deposition of the ablated material.

The cross-sectional morphology of welded Ag particles has been analyzed using transmission electron microscpoy (TEM). Samples were prepared from selected Ag-Ag dimers (figure 3(a)) by etching with a focused ion beam. It is evident that the bottom surface of the deposited Ag submicron particles dimer has been flattened, forming disc-like structures with a diameter/height ratio of ~3.7 (figures 3(a), (b)). Occasionally, some of these deposited Ag particles (not shown here) even appear to be donut-like. This is similar to the structure obtained in splat cooling [18–20], suggesting that Ag particles ablated from the sputtering target were in a softened or molten state [18–20] as they travelled at high speed prior to impacting on the Si wafer surface. As in splat cooling, quasi-liquid or liquid particles solidify immediately on reaching the surface of the Si wafer. Our studies have shown that when Ag particles were deposited on a carbon film under the same conditions, and then irradiated with fs laser pulses with same fluence (2.7 mJ cm^{-2}), adsorbed particles

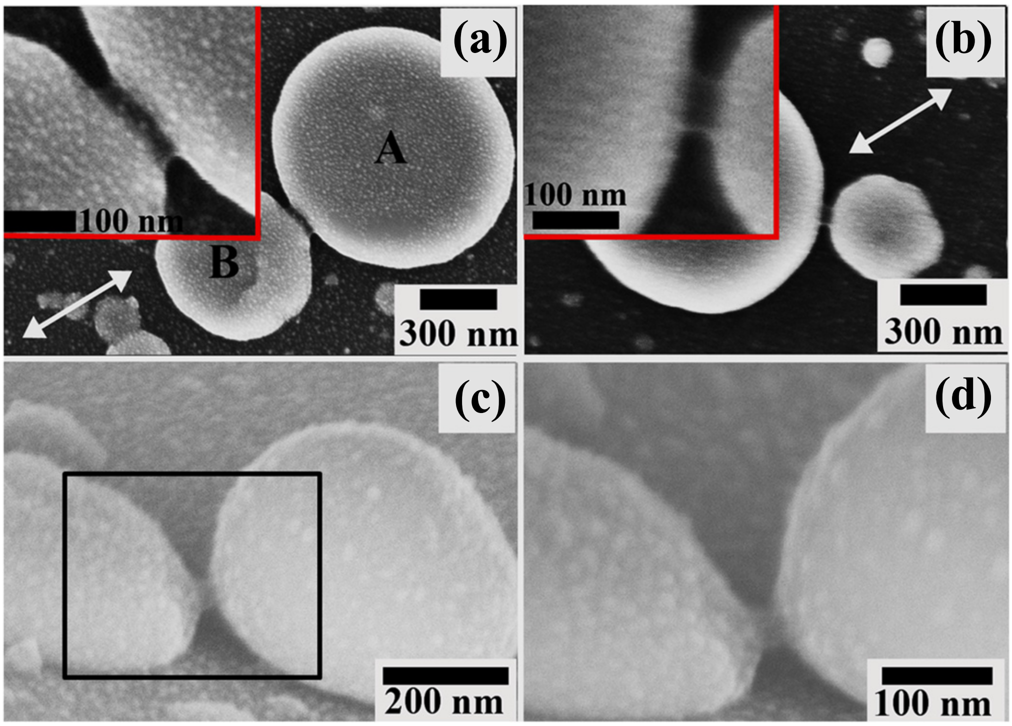

Figure 2. SEM images of the morphology of deposited Ag particles after fs laser irradiation at a fluence of ~900 μJ cm^{-2} for 50 s. (a), (b) Welded Ag particles after irradiation, the double ended arrows show the direction of laser polarization, and the inset images show a magnified view of the joint; (c) view of welded Ag particles after tilting by 75°; (d) magnified view of the square area in (c).

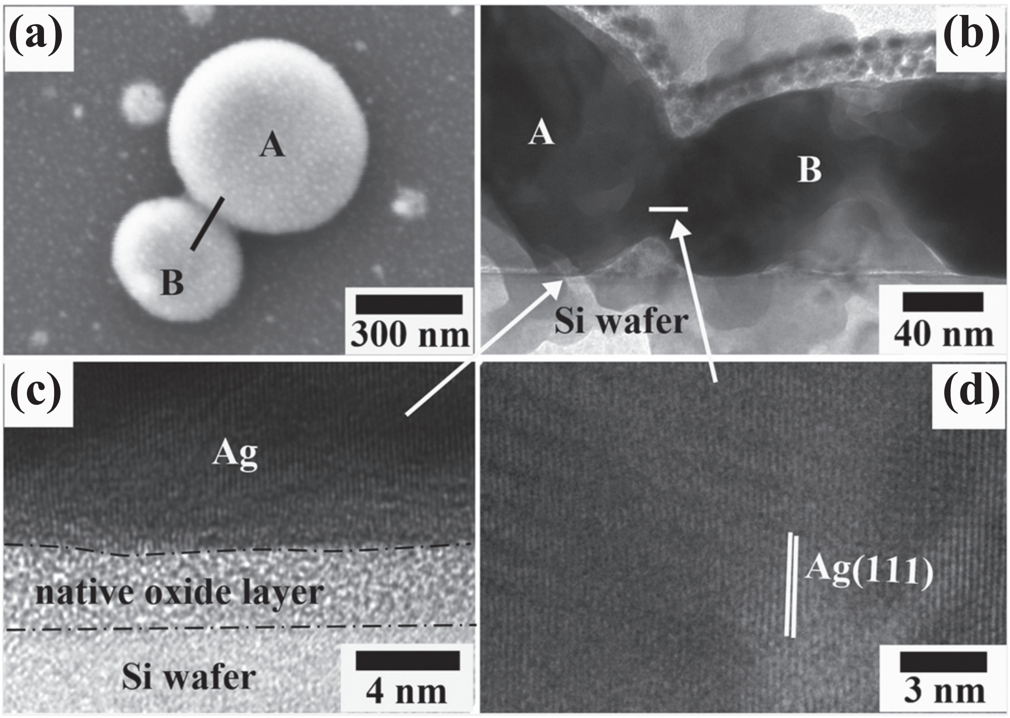

Figure 3. Morphology of a pair of welded Ag particles. (a) SEM Images; (b) TEM image of the cross-sectional view of the Ag pair in (a) along the the black line; the small particles on the top of particles A and B are re-deposited material as a result of the TEM sample preparation process; (c) HRTEM image of the Ag-Si wafer interface; (d) HRTEM image of the area across the junction between two Ag particles as shown by the white bar in figure (b).

were ejected from the surface due to the dewetting between Ag and carbon [21]. This is not seen when Ag particles were deposited on a Si wafer, implying that the Ag particles bond to the Si wafer during deposition. High resolution TEM (HRTEM) scans at the interface between the Ag particle and the Si wafer (figure 3(c)) show that bonding occurs in the contact area between the Ag particle and the ~3 nm thick amorphous native oxide (SiO_x) layer [22] on the surface of the Si wafer. HRTEM analysis also shows (figure 3(d)) that the Ag(111) lattice planes in both particles are aligned across the junction.

Figures 2(a)–(d) show that interconnection between the two particles is generated by the formation of a bridge comprising filler material from ablated submicron particles. This ablation can be attributed to LSP-induced hotspots which are accompanied by localized electron and ionic emission [12]. To investigate the possible contribution of this localized emission to the formation of the interconnection, FDTD calculations (Lumerical) have been used to simulate the electric field distribution (E^2) and identify the location of hotspots near and in between a pair of Ag submicron particles. The two-particle structure shown in figure 2(a) is selected as representative, and both particles A and B are assumed to be discs having a diameter/height ratio of 3.7. This geometry approximates relplicates that seen in the TEM scans. The laser beam (with a wavelength of 800 nm) is incident normal to the Si wafer supporting particles A and B. The angle between the laser polarization and the common axis of the two particles is the same as used in the experiments. The simulation results show that a hotspot with an electric field enhancement factor ~180 appears between the two discs (figures 4(a), (b)) close to the top edge of the lower disc (figure 4(a)). This simulation is in good agreement with the experimental observations (figures 2(a)–(d)), implying that LSP-induced hotspots facilicate the interconnection of pairs of Ag particles.

Extensive studies have shown that the generation of hotspots arising from LSP will produce localized ablation and electron/ion emission from the particles, and that the material ablated from the particles will re-deposit near the ablated surface within the hotspots [23]. This re-deposition of ablated material can gradually reduce the gap distance between the particles resulting in the formation of an interconnecting link. In our case, the electic field enhancement factor is ~13.4 giving an root square enhancement factor of ~180, corresponding to the presence of a local electric field $E \sim 5 \times 10^9\ \mathrm{V\ m^{-1}}$ near the surface of the Ag particle. This field is close to the measured threshold for field ion emission [24], suggesting that ions can be emitted from the Ag particles under the effect of LSP-induced hotspots. These ions contribute to the formation of an interconnection between the particles and facilitate the welding process.

To explore the connection between ablation and ionic emission at hotspots in irradiated particles, the laser fluence was increased to $\sim 1300\ \mu\mathrm{J\ cm^{-2}}$ in order to mimic the enhancement that occurs in LSP-induced hotspots. Figure 5(a) shows SEM images of the morphology of freshly deposited Ag submicron/nanoparticles. These images reveal that the surface of these particles was almost smooth prior to irradiation. Particles labeled 1 and 2 are seen to be closely separated. After these particles were irradiated for 50 s by fs laser pulses at a fluence of $1300\ \mu\mathrm{J\ cm^{-2}}$ (figure 5(b)), the particle morphology is seen to change and the surface become rough, indicating that ionic emission (or ablation) of the particles occurs under these conditions. Emitted material is also seen to form a bridge between the two particles 1 and 2. This suggest that the electric field enhancement in the hotspots facilitates the generation of a weld between adjacent Ag particles. It should also be noted that, under fs laser irradiation, the isolated particles tend to be ablated at the two poles in the direction parallel to the laser polarization. Ablated material from these locations is re-deposited at the poles of the particles (see particle 3 in figure 5(b)). The location of the ablated area in these particles is consistent with that of LSP-induced hospots [12], indicating that laser induced hotspots selectively enhanced ablation of the particle. This is apparent in the SEM scan of the sample after rotation by 90° followed by further irradiation by fs laser pulses for another 50 s at the same fluence. The SEM image (particle 3 in figure 5(c)) clearly shows that, when the sample was rotated by 90°, the ablated area appeared at the other poles whose direction was parallel to the laser polarization. This direction is perpendicular to that of the previous poles. For particles 1 and 2, the dependence of ablation on laser polarization is less significant because the gap distance between the two particles is so small that it becomes the main factor in the induction of hotspots due to LSPs. These data confirm that LSP-induced hotspots generated by fs laser irradiation contribute to localized ablation of the particle and then to the welding produced between two adjacent particles.

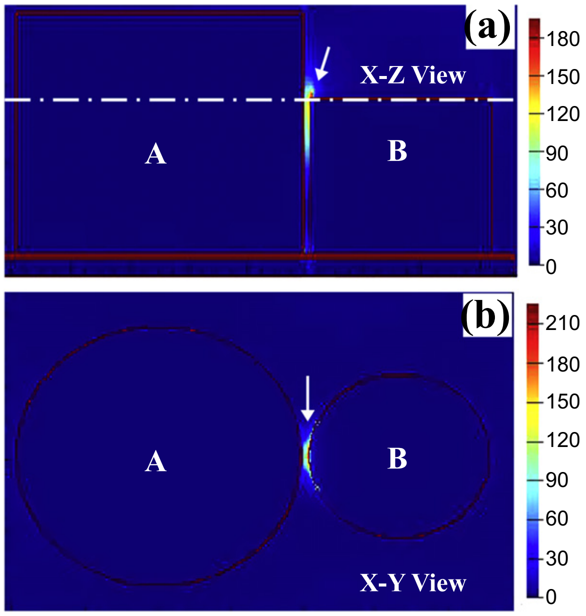

Figure 4. Simulated electric field (E^2) distribution in two adjacent Ag particles as observed in figure 2(a). (a) View of the *x-z* plane crossing the axes of the two particles; (b) view of the *x-y* plane at the same height in particle B, as indicated by the dashed line in (a). The arrows show the locations of the hotspots.

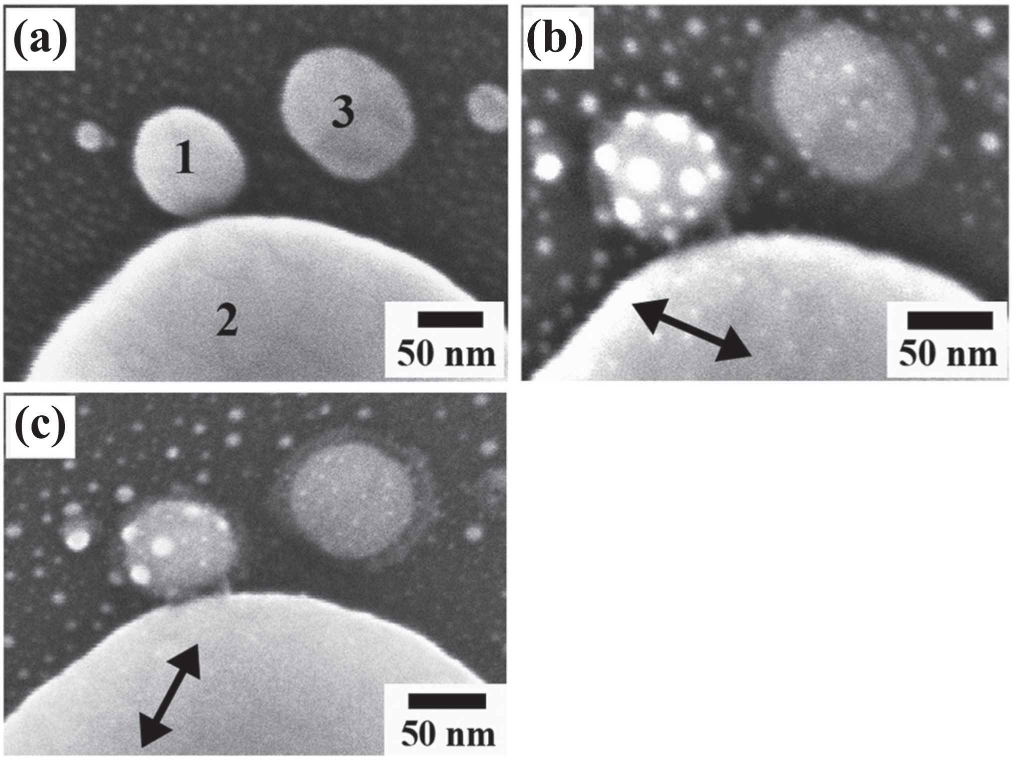

Figure 5. Development of joint between two Ag particles under fs laser irradaition at a fluence ~1300 μJ cm^{-2}. (a) Ag particles before irradiation; (b) same Ag particles after 50 s laser irradiation with polarization (double-ends arrow) shown; (c) same Ag particles after another 50 s laser irradiation with the polarization direction perpendicular to that in (b).

It is significant that, due to the use of low fluence combined with the 'non-thermal' nature of the fs laser–matter interaction [15], the irradiated particles are virtually unaffected and have no apparent damage after welding (figures 2(c), (d) and 5). This suggests that fs laser irradiation is a promising technique for precision fabtrication in the 'non-destructive' interconnection of submicron/nanodevice parts for applications in nanoelectrics.

4. Conclusions

We report a technique for the interconnection of two adjacent Ag submicron/nanoparticles on a Si wafer without the use of additional filler material. The Ag particles self-bonded to the Si wafer following deposition by laser ablation. Adjacent Ag particles separated from each other by a small gap were found to be welded together after fs laser irradiation for 50 s at fluences between ~900 and 1300 μJ cm^{-2}. SEM and TEM scans show that the metallic interconnection linking the particles was generated by the re-deposition of ablated material in the gap between the particles. The integrity of both particles is conserved in this process and the overall damage to the non-bonded component of the Ag nanoparticles was found to be negligible. FDTD simulation indicates that the ablation of the Ag submicron/nanoparticles and the formation of the interconnection can be attributed to LSP-induced hotspots. This study points the way to the development of a high integrity technique in the fabrication of submicron and nanodevices.

Acknowledgments

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References

[1] Lu W and Lieber C M 2007 *Nat. Mater.* **6** 841–50
[2] Guo X, Qiu M, Bao J, Wiley B J, Yang Q, Zhang X, Ma Y, Yu H and Tong L 2009 *Nano Lett.* **9** 4515–9
[3] Hao Q, Wang B, Bossard J A, Kiraly B, Zeng Y, Chiang I-K, Jensen L, Werner D H and Huang T J 2012 *J. Phys. Chem.* C **116** 7249–54
[4] Patolsky F, Timko B P, Zheng G and Lieber C M 2007 *MRS Bull.* **32** 142–9
[5] Gandra N, Abbas A, Tian L and Singamaneni S 2012 *Nano Lett.* **12** 2645–51
[6] Hu A, Guo J Y, Alarifi H, Patane G, Zhou Y, Compagnini G and Xu C X 2010 *Appl. Phys. Lett.* **97** 153117
[7] Wu Y and Yang P 2001 *Adv. Mater.* **13** 520–3
[8] Chen C, Yan L, Kong E and Zhang Y 2006 *Nanotechnology* **17** 2192–7
[9] Peng Y, Cullis T and Inkson B 2009 *Nano Lett.* **9** 91–6
[10] Xu H and Kall M 2002 *Phys. Rev. Lett.* **89** 246802
[11] Valev V K *et al* 2012 *Adv. Mater.* **24** OP29–35

[12] Plech A, Kotaidis V, Lorenc M and Boneberg J 2006 *Nat. Phys.* **2** 44–7
[13] Mafune F, Kohno J, Takeda Y and Kondow T 2003 *J. Am. Chem. Soc.* **125** 1686–7
[14] Liu L, Peng P, Hu A, Zou G, Duley W W and Zhou Y 2013 *Appl. Phys. Lett.* **102** 073107
[15] Huang H, Liu L, Peng P, Hu A, Duley W W and Zhou Y 2012 *J. Appl. Phys.* **112** 123519
[16] Huang H, Hu A, Sivayoganathan M, Duley W W, Huang Z H and Zhou Y 2014 *Mater. Res. Express* **1** 025022
[17] Wang Z B, Luk'yanchuk B S, Guo W, Edwardson S P, Whitehead D J, Li L, Liu Z and Watkins K G 2008 *J. Chem. Phys.* **128** 094705
[18] Tran A T T, Hyland M M, Shinoda K and Sampath S 2011 *Thin Solid Films* **519** 2445–56
[19] Brossard S, Munroe P R, Tran A T T and Hyland M M 2010 *Surf. Coat. Technol.* **204** 2647–56
[20] Cui C and Li Q 2011 *J. Shanghai Jiaotong Univ. (Sci.)* **16** 347–51
[21] Habenicht A, Olapinski M, Burmeister F, Leiderer P and Boneberg J 2005 *Science* **309** 2043–5
[22] Al-Bayati A H, Orrman-Rossiter K G, van den Berg J A and Armour D G 1991 *Surf. Sci.* **241** 91–102
[23] Kaempfe M, Seifert G, Berg K J, Hofmeister H and Graener H 2001 *Eur. Phys. J.* D **16** 237–40
[24] Chang C S, Su W B and Tsong T T 1994 *Phys. Rev. Lett.* **72** 574–7