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Multiscale Surface Roughness Measure for Dressed-Photon–Phonon Etching

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Abstract– Dressed-photon–phonon (DPP) etching is a disruptive technology in planarizing material surfaces because it completely eliminates mechanical contact processes. However, adequate metrics for evaluating the surface roughness and the underlying physical mechanisms are still not well understood. Here we propose a two-dimensional multiscale surface roughness measure that represents the effectiveness of DPP etching while conserving the original two-dimensional surface topology. Also, we created a simple physical model of DPP etching that agrees well with the experimental observations, which clearly shows the involvement of the intrinsic multiscale properties of dressed photons, or optical near-fields, in the surface processing.

1. Introduction

Nanophotonics, which exploits light–matter local interactions on the nanometer scale, has been intensively investigated from a variety of aspects, ranging from fundamental science, such as atom/molecule and optical near-field interactions [1], and spectroscopy [2], to a number of practical applications, including information security [3], computing [4], the environment and energy [5], and healthcare [6], to name a few. What we particularly address in this paper is nanofabrication involving optical near-field processes [7,8].

The idea is to induce a nanofabrication process, either in the form of material desorption or deposition, selectively in a region where optical near-fields are present. Nowadays, the optical near-field is understood as a virtual photon coupled with an excited electron, called a dressed photon (DP). DPs can interact with phonons in the crystal lattice structure of nanomaterials in a coherent manner [7]. The combined coupled state of a DP and a coherent phonon, which is referred to as dressed-photon–phonon (DPP), has a higher energy than those of the DP and the incident photon [7]. Therefore, a DPP can induce photochemical reactions even under irradiation with a low photon energy at which photochemical reactions are conventionally inactive [8,9]. Such a process has been called a phonon-assisted process and has been applied to numerous demonstrations, including photochemical vapor deposition [10], photolithography [11], among others [12,13].

In particular, the DPP-based surface etching, or flattening, proposed by Yatsui *et al.*, which planarizes the surfaces of devices without any mechanical contact processes, is an interesting and industrially important technique [8,9,14,15]. It selectively induces photochemical reactions in regions on a surface where DPPs are excited, namely, in the vicinity of regions possessing fine-scale rough structures, leading to reduced surface roughness. Planarization of surfaces is important for various devices such optical elements [8], solid-state materials, such as silicon wafers, diamonds [9], and so on.

There are, however, several unsolved, important issues associated with DPP etching. The first concerns a suitable metric for evaluating the surface roughness. Conventionally, the roughness average, R_a, defined as the average of the absolute values of the deviation from the average height, has been widely used. However, by definition, R_a depends on the size of the region-of-interest (ROI). Furthermore, with such a measure, the effects provided by DPPs are concealed, and so it has been difficult to obtain physical insights into the underlying mechanisms which would serve to reveal the ultimate limitations of the method and to improve/optimize fabrication processes.

In fact, with a view to resolving these issues, we have previously proposed a parametric statistical spectrum analysis method for evaluating surfaces flattened by DPP etching [15]. Furthermore, in Ref. [9], we developed another measure, namely, the standard deviation of the height differences of two adjacent areas averaged over every l pixels, inspired by the Allan variance [16] that is widely applied in evaluating the stability of time-domain signals. Specifically, letting $\overline{z_k^{(l)}}$ be the average height over every l pixels, the measure was defined as $R(l)=\sqrt{\left\langle \left(\overline{z_{k+1}^{(l)}}-\overline{z_k^{(l)}}\right)^2/2\right\rangle}$. These methods allowed us to see the reduction in surface roughness correlated with spatially finer/coarser structures [9,15].

Nevertheless, important concerns still remain. One is that both of the above-mentioned methods must convert the original two-dimensional (2D) surface profile data to one-dimensional (1D) data, on which the analysis is made. Namely, the topology inherent in the experimental data is destroyed, which is a large impediment to gaining an accurate physical understanding of the mechanisms involved. By overcoming these weaknesses, we will be able to understand the fundamental mechanisms of DPP etching.

In this paper, we propose a metric for evaluating surface roughness while preserving the original topology, what we call the *two-dimensional, multiscale surface roughness measure*, or MRM for short. Furthermore, taking into consideration the intrinsic multiscale properties of dressed photons, we propose a physical model representing the principal attributes of DPP etching. The resultant data agrees well with experiments.

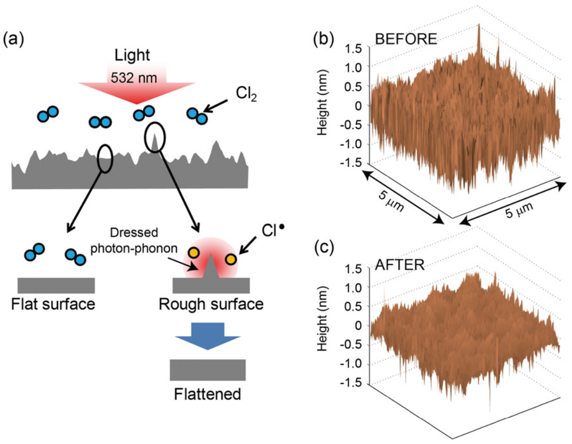

Fig. 1 (a) Schematic illustration of the dressed-photon–phonon etching. (b,c) Surface height profiles of a (001) GaN substrate (b) before and (c) after 30-minute irradiation with continuous-wave (CW) light at a wavelength of 532 nm in a Cl_2 atmosphere at 200 Pa. The roughness average, Ra, decreased from 0.23 nm to 0.14 nm.

2. Multiscale Roughness Measure

We first review the principles of DPP etching proposed by Yatsui *et al* [8]. The surface of a device to be etched is irradiated with a light beam. If the surface is rough, DPPs will be generated in the vicinity of the corresponding rough structures, and photochemical reactions can be induced selectively in the regions where the DPPs are generated [8,9]. More specifically, in the case of the experiments described below, chlorine gas (Cl_2) is filled in the space around the device to be processed, and chlorine radicals ($Cl^\bullet$) are selectively produced from the Cl_2 gas in the regions where DPPs are generated. The $Cl^\bullet$ then reacts with and etches the material, decreasing the local surface roughness, or bumps. Once the rough structures are eliminated, the DPPs will disappear, thus automatically terminating the photochemical reaction process. This physical principle is schematically shown in Fig. 1(a). In the experiment, the device under study was a (001) GaN substrate, which was subjected to 30 minutes of irradiation with continuous-wave (CW) light having a wavelength of 532 nm in a chlorine gas (Cl_2) atmosphere at 200 Pa. Figures 1(b) and (c) respectively show surface profile images taken by an atomic force microscope (AFM) before and after the etching process. The surface heights were measured at 256 × 256 equally spaced sampling points in a 5 μm × 5 μm area. The value of R_a decreased from 0.23 nm to 0.14 nm.

Now, assume that the height of a surface profile is given by $h(i,j)$, where the indexes (i,j) specify the position among $N \times N$ sampling points. Both i and j are positive integers ranging from 1 to N. The idea of the proposed measure, MRM, is (1) to evaluate the average height over $l \times l$ pixels (denoted by $h_P^{(l)}$); (2) to see how $h_P^{(l)}$ differs from the average height of its four neighbors, namely, the north ($h_N^{(l)}$), south ($h_S^{(l)}$), east ($h_E^{(l)}$), and west ($h_W^{(l)}$) areas; and then (3) to calculate the variance of such differences in the entire region of the sample. A schematic illustration of $h_P^{(l)}$ is shown in Fig. 2(a). As a function of the scale parameter, or the size of the local area (l), the MRM is defined by

$$\sigma_{2D}^2(l) = \left< \left(h_P^{(l)} - \left(\frac{h_N^{(l)} + h_S^{(l)} + h_E^{(l)} + h_W^{(l)}}{4} \right) \right)^2 \right> \qquad (1)$$

where < > means taking the average over all areas of a given sample. For simplicity, we assume that N and l are both powers of 2. When the size of a single local area is given by $l=2^k$, there are in total $(N/l)^2$ areas at the corresponding scale, and the average height in an area specified by (s,t), corresponding to $h_P^{(l)}$ in eq. (1), is given by

$$h^{(l)}(s,t) = \sum_{m=1\cdots,l,} \sum_{n=1,\cdots,l} h(l \times (s-1) + m, l \times (t-1) + n) \Big/ l^2 . \qquad (2)$$

Here, $h_N^{(l)}$, $h_S^{(l)}$, $h_E^{(l)}$, and $h_W^{(l)}$ respectively correspond to $h^{(l)}(s,t+1)$, $h^{(l)}(s,t-1)$, $h^{(l)}(s+1,t)$, and $h^{(l)}(s-1,t)$. The indexes s and t are natural numbers ranging from 1 to $N/2^k$.

In an experimental demonstration using GaN as the sample, N was 256, and we used scale parameters l of 1, 2, 4, 8, 32, and 64 pixels, corresponding to physical lengths of about 19.5, 39.0, 78.1, 156.2, 312.5, 625, and 1250 nm, respectively. Figures 2(b) and (c) schematically represent the scale- and position-dependent metric $E_P^{(l)} = h_P^{(l)} - (h_N^{(l)} + h_S^{(l)} + h_E^{(l)} + h_W^{(l)})/4$, which we call the *multiscale etching score* (MES), before and after the optical processing, respectively. As demonstrated in Fig. 3(a), we can clearly see that the dashed curve, which is the MRM of the surface after DPP etching, is reduced compared with the solid curve, which is the MRM of the surface before the etching.

As mentioned above, the surface etching is autonomously induced at locations where the DPPs are generated by the roughness of the sample under study. We may envisage a physical picture in which etching is preferentially induced in regions where a tiny bumps exist.

Namely, the etching may be dominated by the MES of the tiniest scale, $E_P^{(1)}$, concerning the surface roughness at the tiniest scale. Based on such a picture, we can construct a physical model of the DPP etching such that the position that gives the maximum $E_P^{(1)}$ decrease the height of that position by an amount Δ_h. Note that the MES *at the tiniest scale* is considered.

We applied the above surface etching strategy to the original, unpolished surface profile of the GaN device. Through such modeling, the resultant surface roughness indeed decreased as compared with the original one, but the resulting MRM, $\sigma_{2D}^2(l)$, was as shown in Fig. 3(b-1), which did *not* agree with the experimental reality shown in Fig. 3(a).

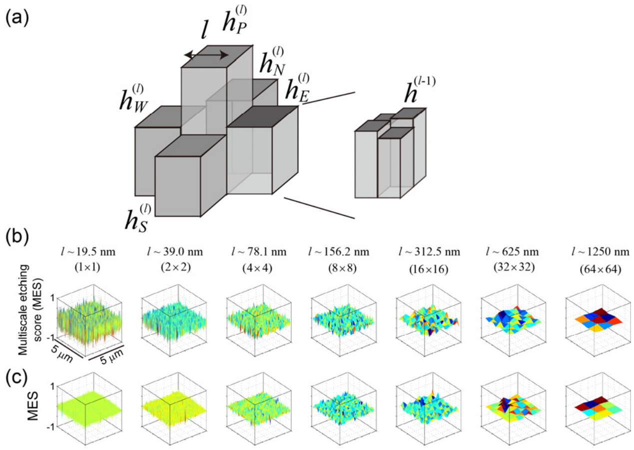

Fig.2 (a) Schematic diagram of the proposed two-dimensional multiscale surface roughness measure (MRM). (b,c) The two-dimensional distribution of the multiscale etching score (MES) (b) before and (c) after the DPP etching.

3. Multiscale Property of Optical Near-Field

We should consider that this is clear evidence that the DPP etching does *not* depend only on the finer structures. In fact, dressed photons, or optical near-fields, appear in a hierarchical manner depending on the spatial fine/coarse structures [17,18]. Suppose that there are two spheres whose radii are respectively given by a_P and a_S (Fig. 4(a)). The near-field optical potential is given by [18]

$$V(r) = \sum_{i=S}^{P} \frac{\exp(-\pi r / a_i)}{r}. \tag{3}$$

The scattered signal obtained from the interaction between these two spheres is given by

$$I(r_{SP}) \equiv \left| \iint \nabla_{r_P} V(|\boldsymbol{r}_P - \boldsymbol{r}_S|) d^3 r_S d^3 r_P \right|^2 \tag{4}$$

$$\propto \left[a_P^3 \left\{ \frac{a_S}{a_P} \cosh\left(\frac{\pi a_S}{a_P}\right) - \frac{1}{\pi} \sinh\left(\frac{\pi a_S}{a_P}\right) \right\} \left(\frac{1}{r_{SP}} + \frac{a_P}{\pi r_{SP}^2} \right) \exp\left(-\frac{\pi r_{SP}}{a_P} \right) \right]^2$$

where the center positions of the two spheres are respectively given by $\boldsymbol{r}_S$ and $\boldsymbol{r}_P$, and the distance between $\boldsymbol{r}_S$ and $\boldsymbol{r}_P$ is given by r_{SP} [18]. We can consider that a_S represents the surface roughness, whereas a_P indicates the size of the environment, containing chlorine radicals, that

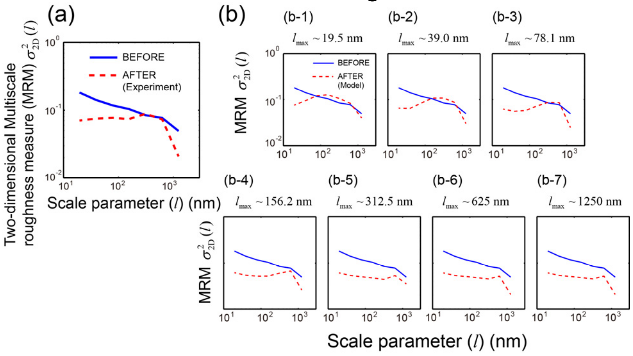

Fig. 3 The MRM, $\sigma_{2D}^{(l)}$, based on experimental results for GaN surfaces. (b) Evaluation of MRM, $\sigma_{2D}^{(l)}$, with respect to the calculated surface profiles derived through the proposed DPP etching model for different values of the maximum physical scale, $l_{\max}$.

could interact with the surface roughness. The effect of the interactions involving these two spheres is defined by the quantity given by eq. (4) divided by the square of the total volume of the two spheres, so that the evaluation is made in the dimension of per unit area. The solid, dotted, and dashed curves in Fig. 4(a) indicate the normalized quantity given by eq. (4) divided by $(a_P^3 + a_S^3)^2$, which represents the strength of the interaction between the two spheres, as a function of a_P when a_S is given by 10, 20, and 40 nm, respectively. Notice that the peak of the signal is obtained when the sizes of the two spheres are comparable. It turns out that, by regarding a_S as the spatial fine/coarse surface roughness of the structure, the interaction may be stronger at a coarser scale, rather than at a finer scale, when the roughness of the sample dominates on a large scale. Therefore, we should modify the DPP etching model above by taking into account the hierarchical attributes of dressed photons, so that the surface flatness is evaluated on multiple scales, and a reduction in surface roughness may be induced in a region that gives the maximum MES.

Specifically:

(a) Calculate the MES at each of the regions of a given device *and* at multiple scales: l=1,2,...,$l_{\max}$. The maximum scale considered is given by $l_{\max}$.

(b) Find the area that gives the maximum $E_P^{(l)}$ among all of the calculated positions and scales. Decrease the height of the corresponding area by an amount Δ_h.

(c) Repeat the process until all values calculated at step (a) are smaller than a certain threshold value.

In this model, we modify the maximum scale to be considered, $l_{\max}$, in step (a) and investigate the physical scales that affect the surface etching. The threshold at step (c) was assumed to be sufficiently small, and was set at 0.1. Figure 3(b) shows the MRM, $\sigma_{2D}^2(l)$, for different values of l_{max}, namely, 1, 2, 4, 8, 16, 32, and 64. We can see that the resultant $\sigma_{2D}^2(l)$ is consistent with the experimental demonstrations shown in Fig. 3(b-3) when l_{max} is 4, corresponding to a physical length of 78.1 nm. Also, Fig. 4(b) shows the simulated time evolution of R_a depending on different l_{max} values. The resultant R_a in the experiment was 0.14 nm, whereas the converged R_a obtained through the modeling was 0.13 nm when l_{max} was 78.1 nm. This is another indication of the consistency between the proposed model and the experiment. These results suggest that the hierarchical properties of dressed photons inevitably affect the surface etching. With this model, we can predict the converged surface characteristics and the achievable flatness of given initial surfaces.

Finally, we remark on the converged surface characteristics. Minimizing the multiscale etching score (MES) to zero, in the best case, indicates that the height at a particular area is equal to the average of its surrounding areas. This is the property of the so-called harmonic functions, which are solutions of Laplace's equations [19]. What is particularly different from the conventional harmonic functions is that the solutions should minimize the MES at multiple scales (not just at a single scale). Nevertheless, such considerations inspired by harmonic functions support the fact that completely flat surfaces may *not* be the only converged pattern of DPP etching.

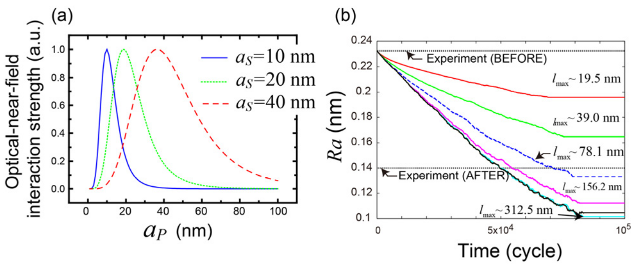

Fig. 4 (a) The hierarchical, or multiscale, nature of near-field interactions based on two-sphere model where the radii are respectively given by a_S and a_P. (b) Simulated time evolution of the roughness average (R_a) with respect to the maximum physical scale in the DPP etching model.

4. Conclusion

In conclusion, we proposed a two-dimensional multiscale surface roughness measure (MRM) for evaluating the surface roughness of a surface planarized by DPP etching. Taking into account the intrinsic hierarchical properties of dressed photons, we built a simplified physical model of the DPP etching, which agreed well with the experimental demonstration. It clearly demonstrates that the DPP etching involves multi-scale structures of the rough surface being processed. This study has unveiled one fundamental mechanism of nanofabrication, and we consider that the method described here will help to predict the achievable performance of nanofabrication and to optimize fabrication processes [20]. The application of the proposed measure to other fields, such as image processing [21], will also be an interesting future issue.

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