© 2009 OSA/CLEO/IQEC 2009

Laser-induced nano-corrugation of a dielectric surface

A. E. Kaplan and S. N. Volkov
Department of Electrical and Computer Engineering, The Johns Hopkins University, Baltimore, MD 21218
alexander.kaplan@jhu.edu

Abstract: We demonstrate that uneven distribution of evanescent field and sub-threshold ablation at a dielectric surface can explain experimentally observed creation of nanometer-sized surface grooves by femtosecond laser pulses. ©2009 Optical Society of America

OCIS codes: (050.6624) Subwavelength structures; (240.6690) Surface waves

Recent experiments demonstrated that femtosecond laser pulses can create nearly-periodic nanometer-scale grooves on the surface of solid-state films [1, 2]. The physical mechanism behind the phenomenon is still unclear. This paper suggests a viable mechanism based on properties of the field at the corrugated surface. Localized ablation at the surface is assumed to play an important role, whereby the surface field is stronger in the grooves formed by the ablation, than at the ridges. The grooves then deepen with each laser shot, because of the positive feedback for the ablation process, provided *via* this distribution of the evanescent field. The importance of the subject is in part due to potential applications of nano-structures with controlled growth, such as nano-scale computer components, diffraction gratings and reflectors for extreme UV and soft X-rays.

The well known regular surface ripples [3] formed by much longer (nanosecond) laser pulses, each exceeding the ablation threshold, under normal incidence have spacing of the order of the laser wavelength λ. They are due to the interaction of the incident light with the standing "surface-scattered wave" [4] of *the same wavelength*. In the case of sub-λ nano-gratings laser pulses are much shorter and each of them has a sub-ablation energy fluence [1, 2]. This greatly emphasizes the role of the intensity distribution at the surface with a corrugation period $\Lambda \ll \lambda$. Such surfaces have been investigated in many works (see e. g. [5]), with the focus on far-field reflectivity and transmittivity properties and little attention to the field distribution at the surface itself. Most of the far-field results were obtained by assuming that, instead of lateral corrugation, the boundary is formed by a smooth change of the material density in the direction normal to the surface. This approach cannot adequately describe the near-field area.

We consider a laser beam incident in the negative y direction on a periodically corrugated surface with a grating wavevector $\mathbf{K} = 2\pi\hat{\mathbf{e}}_x/\Lambda$. The surface is an interface between two semi-infinite dielectrics with the dielectric constants ϵ_1 (the "upper" dielectric) and ϵ_2 (the "substrate"). Since $\Lambda \ll \lambda$, the incident beam can be treated as a plane wave, resulting in a 2D (cylindrical) problem geometry, and also resulting in a quasi-static problem, whereby the magnetic field can be neglected, while the electric field will follow from the solutions of 2D Laplace equations for the *dc* potential U in each of the materials, $\partial^2 U/\partial x^2 + \partial^2 U/\partial y^2 = 0$. We verified this quasi-static approach by computer simulations using a full EM-wave model. If the electric field $\mathbf{E}_\mathrm{L}$ of the incident EM wave is normal to the grating (i. e. $\mathbf{E}_\mathrm{L} \parallel \mathbf{K}$), then it is coupled to the evanescent surface wave, whereas this is not so for $\mathbf{E}_\mathrm{L} \perp \mathbf{K}$. Thus, we will only consider the case of $\mathbf{E}_\mathrm{L} \parallel \mathbf{K} \parallel \hat{\mathbf{e}}_x$, so that the field components near the corrugated surface are $E_x = -\partial U/\partial x$, $E_y = -\partial U/\partial y$, $E_z = 0$.

We assume sinusoidal corrugation of the boundary, $Ky_\mathrm{bnd} = f(\xi) \equiv g\cos(\xi)$, with $\xi = Kx$. The fields in both "upper" and "substrate" dielectrics are determined from the relation $U + E_0 x = -\sum u_n(y)\sin(n\xi)$. The Fourier coefficients $u_n(y)$ for the spatial harmonics are $u_n(y) = C_n \exp(\pm nKy)$, where C_n are integration constants. Then the fields in both subspaces are as

$$\frac{1}{E_0}\begin{bmatrix} E_x^{(1)} \\ E_x^{(2)} \end{bmatrix} = 1 + \sum_{n=1}^{\infty} \begin{bmatrix} A_n \mathrm{e}^{-nKy} \\ B_n \mathrm{e}^{nKy} \end{bmatrix} \cos(n\xi), \qquad \frac{1}{E_0}\begin{bmatrix} E_y^{(1)} \\ E_y^{(2)} \end{bmatrix} = \sum_{n=1}^{\infty} \begin{bmatrix} -A_n \mathrm{e}^{-nKy} \\ B_n \mathrm{e}^{nKy} \end{bmatrix} \sin(n\xi). \tag{1}$$

Here the superscripts (1,2) denote fields in the upper dielectric and the substrate, respectively, and the coefficients A_n and B_n are to be found from the boundary conditions at $y = y_\mathrm{bnd}(x)$: $E_\parallel^{(1)} = E_\parallel^{(2)}$ and $\epsilon_1 E_\perp^{(1)} = \epsilon_2 E_\perp^{(2)}$, where the subscripts "$\parallel$" and "$\perp$" designate field components tangential and normal to the boundary, respectively. They are $E_\parallel = E_x\cos\psi + E_y\sin\psi$, $E_\perp = -E_x\sin\psi + E_y\cos\psi$ at each side of the boundary, with $\tan\psi = df/d\xi \equiv f'$.

In the limit of a shallow grating, $g \ll 1$, we find $\mathcal{E}_\parallel \equiv (E_\parallel^{(1)}/E_0) = (E_\parallel^{(2)}/E_0) \approx 1 - \eta f$ and $\bar{\mathcal{E}}_\perp \equiv (E_\perp^{(1)} + E_\perp^{(2)})/(2E_0) \approx -f'$, where $\eta = (\epsilon_2 - \epsilon_1)/(\epsilon_2 + \epsilon_1)$ is a dielectric contrast factor. For relatively large modulation amplitudes g, we obtained approximate solutions for $\mathcal{E}_{\parallel,\perp}$:

$$\mathcal{E}_\parallel \approx (1 - F) \Big/ \sqrt{1 + (f')^2} \quad \text{and} \quad \bar{\mathcal{E}}_\perp \approx -[f' \exp(-F^3/2)] \Big/ \sqrt{1 + (f')^2}. \tag{2}$$

© 2009 OSA/CLEO/IQEC 2009

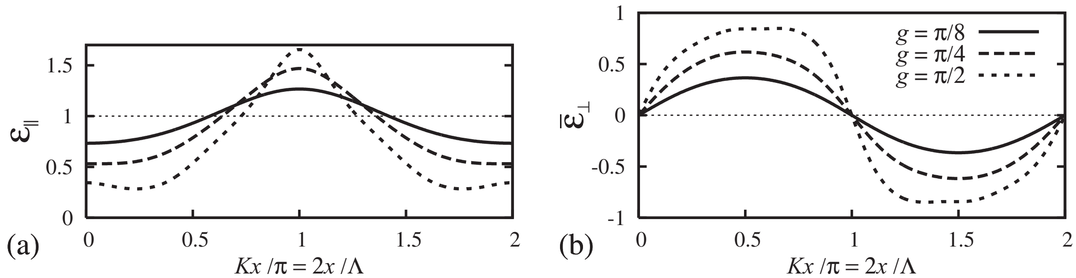

FIG. 1: Normalized electric fields at the corrugated boundary for $\epsilon_2/\epsilon_1 = 6$.

where $F = \eta \tanh(f)$. The spatial profiles $\mathcal{E}_{\|}(\xi)$ and $\mathcal{E}_{\perp}(\xi)$ are plotted according to these approximate formulae in Fig. 1 for different corrugation depths g and for the ratio ϵ_2/ϵ_1 close to that of an air–diamond interface. We found that for $1 < \epsilon_2/\epsilon_1 \lesssim 10$ the approximate solutions (2) are consistent with the results of our numerical simulations up to $g = \pi/2$, which corresponds to the total corrugation depth of $\Lambda/2$, providing thus the range of depths exceeding those observed in [1, 2]. We also found that the approximation (2) still gives a good overall prediction of the numerically generated field profiles, especially the one for $\mathcal{E}_{\perp}$, up to $g \approx 2\pi$ (the depth of 2Λ).

We used two different methods to numerically model our problem. In the first one is based on a plane EM-wave expansion method for solving the Maxwell equations. Our second, quasi-static, numerical approach was to solve Eq. (1) truncated to a finite number of coefficients A_n and B_n; it works well for $g \lesssim 1$, but it becomes divergent at $g \gtrsim \pi/2$ if $\epsilon_2/\epsilon_1 \gtrsim 2$. The agreement of the first method with the second one as well as with Eq. (2) reaches just a few percents of the maximum field value at $g = \pi/2$, and is much better at smaller g.

At the bottoms of the grooves and at the ridges, where $f' = 0$, we have $E_{\perp} = 0$, and the tangential fields at those points are $(\mathcal{E}_{\|})_{\text{bott, ridge}} = 1 \pm \eta \tanh(g)$, where the "+" sign corresponds to the bottoms (maxima) (e. g. $\xi = \pi$), and "–" sign – to the ridges (e. g. at $\xi = 0$ or $\xi = 2\pi$), respectively. If $\epsilon_2/\epsilon_1 \gg 1$, as e. g. for air–diamond or air–semiconductor boundaries, the ratio of the field amplitudes at the bottom and at the ridge can be significant. The main physical factor, however, is that $E_{\|}$ in the grooves can significantly exceed the incident field (up to a factor of 2), which is of great consequence to the ablation processes. Even if the incident field intensity, E_0^2, is lower than the characteristic ablation intensity, $E_{\text{abl}}^2 \propto F_{\text{abl}}/\tau_{\text{p}}$ (where F_{abl} is the critical ablation fluence, and τ_{p} is the laser pulse duration), the field at the bottom can *exceed* E_{abl} and – by selectively heating and chiseling away atoms from there – make the grating grow deeper. According to Eq. (2), as the depth g of the grating increases, the sizes of the areas with high $E_{\|}$ become smaller, which results in stronger localization of ablation near the lowest points of the grooves.

The significant normal component of the electric field, $E_{\perp}$, on the groove slopes may provide another potential mechanism of grating growth. If $E_{\perp}$ is strong enough, the electrons accelerated normally to the corrugated surface may be ejected from of the dielectric and then re-enter it in the next half-period of the light wave, hitting and ejecting atoms from the surface.

In conclusion, we have shown that the coupling of the incident laser light to an evanescent quasi-static electric field *via* a surface nano-grating can be a potential mechanism of formation of nano-gratings on a dielectric surface by *fs* laser pulses. The strongest surface field, exceeding the incident field by up to a factor of 2, occurs at the bottoms of the grooves, while the field at the ridges is weaker than the incident field. The resulting contrast in the field can promote sub-ablation growth of the grating due to preferential localized ablation at the bottoms.

This work is supported by AFOSR.

[1] J. Bonse, *et al.*, Appl. Phys. A **71**, 657 (2000); Q. Wu *et al.*, Appl. Phys. Lett. **82**, 1703 (2003); A. Borowiec and H. K. Haugen, *ibid*, **82**, 4462 (2003); F. Costache, M. Henyk, and J. Reif, Appl. Surf. Sci. **208**, 486 (2003); T. Q. Jia *et al.*, Phys. Rev. B **72**, 125429 (2005).

[2] K. Miyazaki *et al.*, Appl. Phys. A **80**, 17 (2005); G. Miyaji and K. Miyazaki, Appl. Phys. Lett. **89**, 191902 (2006).

[3] M. Birnbaum, J. Appl. Phys. **36**, 3688 (1965); D. C. Emmony, R. P. Howson, and L. J. Willis, Appl. Phys. Lett. **23**, 598 (1973); J. F. Young, J. S. Preston, H. M. van Driel, and J. E. Sipe, Phys. Rev. B **27**, 1155 (1983) and references therein and in [4].

[4] Z. Guosheng, P. M. Fauchet, and A. E. Siegman, Phys. Rev. A **26**, 5366 (1982); J. E. Sipe, J. F. Young, J. S. Preston, and H. M. van Driel, Phys. Rev. B **27**, 1141 (1983).

[5] R. A. Hurd, Can. J. Phys. **32**, 727 (1954); R. S. Elliott, IRE Trans. AP-2, **71**, (1954); S. M. Rytov, Zh. Exp. Teor. Fiz., **29**, 605 (1955); *Electromagnetic theory of gratings*, ed. R. Petit, (Berlin, Springer, 1980).