Laser-Induced Periodic Surface Structure on Solids: A Universal Phenomenon

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Laser-induced periodic structure on solid surfaces can be understood as a universal phenomenon which occurs for a broad range of wavelengths and different laser polarization states in both polariton-active and -inactive media. Circularly polarized light has been used, for the first time, to generate periodic structures; these show an interesting dependence on the sense of rotation.

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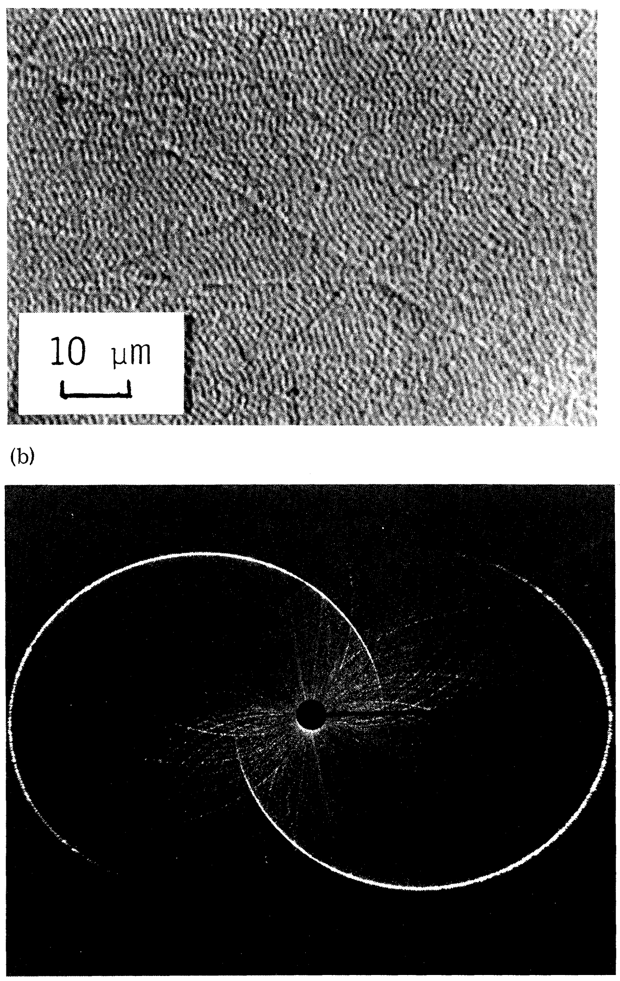
Over the past eighteen years it has been observed that single, intense linearly polarized laser pulses could induce permanent ripples on the surface of various metals, semiconductors, and insulators.[1-12] It is generally considered that the pattern results from inhomogeneous energy deposition associated with the interference of the incident beam with a surface scattered field, but the nature of this field has been the subject of some controversy.[3-7,9-14] In this Letter we report that laser-induced periodic surface structure (LIPSS) can be understood as a very general phenomenon; it can be expected on a wide variety of materials, polariton active and polariton inactive, with different surface microroughness, and over a broad range of wavelengths. With respect to beam polarization, we have observed for the first time that LIPSS can be produced on metals and semiconductors by circularly, as well as linearly, polarized light, contrary to earlier suggestions.[6,10]

We have observed LIPSS on Ge, Si, Al, and brass using 15-nsec pulses with wavelength $\lambda = 0.53$ or $1.06\ \mu$m. Although it is possible to form ripples with single pulses, the patterns so formed can be influenced by the presence of isolated defects or scratches. For multiple shots at constant fluence (typically 75 mJ/cm^2), a steady-state pattern emerges after about twenty shots which is independent of the number of additional shots and the initial surface conditions. Only this case is discussed here. The role of defects in connection with single-pulse LIPSS is discussed elsewhere.[14]

Recently, using linearly polarized, 1.06-μm 15-nsec pulses incident on Ge, we studied the ripple patterns induced by s- and p-polarized light as a function of the angle of incidence θ.[9] It was noted that although the patterns in real space can be quite complex, their Fourier ($\vec{\kappa}$) spectra, as revealed by the Fraunhofer diffraction pattern of a cw Ar$^+$ laser, are quite simple; typically the damage spot size is 5 mm whereas the probe spot size is 1 mm. For $\theta = 0°$, well-defined ripples of spacing λ were formed with an orientation perpendicular to the polarization. In contrast, we report here that when circularly polarized light is used at $\theta = 0°$, well-defined fringes are not formed, but instead the surface appears to be speckled, as Fig. 1(a) shows. The corresponding observed diffraction pattern is a uniformly bright circle (as shown schematically in Fig. 2) whose radius in $\vec{\kappa}$ space is $2\pi/\lambda$. This indicates that the speckled surface actually consists of fringes of spacing λ formed isotropically along the surface. For $\theta = 30°$, the circularly polarized beam produces a much more complex diffraction pattern as seen in Fig. 2. In Fig. 2 we summarize schematically the diffraction patterns as seen on the various materials at the two different wavelengths. All the observed diffraction patterns are located on parts of two intersecting circles in $\vec{\kappa}$ space which satisfy $|\vec{\kappa} \pm \vec{\kappa}_i| = 2\pi/\lambda$, where $\vec{\kappa}_i$ is the component of the incident wave vector parallel to the surface. We wish to emphasize the asymmetrical patterns produced by circularly polarized light for $\theta \neq 0°$, and the fact that right- and left-circularly polarized light produce diffraction patterns which are mirror images of each other; the ability of optically inactive materials to produce these results for different senses of rotation is particularly interesting. We also note that, in general, the metals give more uniformly filled out circles and less asymmetry than the semiconductors.

We have been able to explain all the qualitative features of the diffraction patterns by means of a general scattered-field model.[9,13,14] Microroughness is assumed to exist in the "selvedge" region ($l_z > z > 0; l_z \ll \lambda$) which lies below vacuum ($z > l_z$) and above bulk material of dielectric constant ϵ ($z < 0$). If there were no material in the

(a)



(b)

FIG. 1. (a) Normarski microscope photograph of surface structure on Ge induced by 1.06-μm circularly polarized light at normal incidence. (b) Photograph of far-field diffraction pattern obtained in reflection from Ge surface with normally incident 0.46-μm Ar^+ laser light; Ge surface was damaged by right circularly polarized beam incident at 30°.

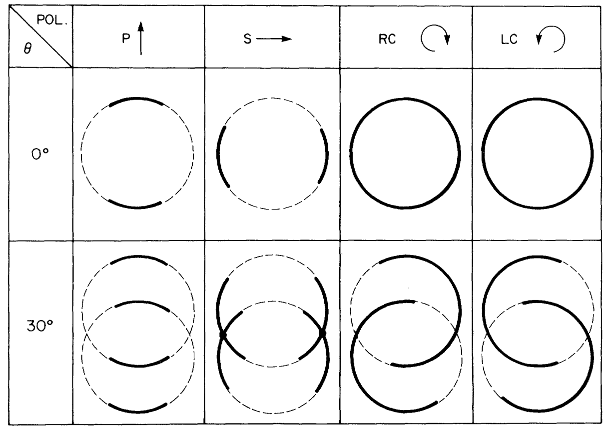


FIG. 2. Schematic summary of far-field diffraction patterns for Ge, Si, Al, and brass for various angles of incidence, θ, and polarization states of impacting beam. Solid curves indicate high-intensity regions.

selvedge region, the electric field there would be $\vec{E}_s = \vec{E}_i + \overleftrightarrow{r} \cdot \vec{E}_i$, where $\vec{E}_i$ is the incident field and $\overleftrightarrow{r}$ the matrix of Fresnel reflection coefficients. To lowest order in l_z/λ, the polarization at points in the selvedge where material is present is given by $\vec{P} = \overleftrightarrow{\gamma} \cdot \vec{E}_s$, where the response tensor $\overleftrightarrow{\gamma} = \gamma_z \hat{z}\hat{z} + \gamma_t (\hat{x}\hat{x} + \hat{y}\hat{y})$ is given by $4\pi\gamma_z = (\epsilon - 1)/\epsilon$, $4\pi\gamma_t = \epsilon - 1$, in the limit $l_t/l_z \gg 1$, where l_t is the correlation length of the roughness in the xy plane; this is a standard result.[15] We have extended this treatment to take into account local-field corrections that are important when $l_t/l_z \lesssim 1$; we find[13]

$$4\pi\gamma_z = (\epsilon - 1)\{\epsilon - (1-F)(\epsilon - 1)[a(s) + Ra_I(s)]\}^{-1},$$
$$4\pi\gamma_t = (\epsilon - 1)\{1 + \tfrac{1}{2}(1-F)(\epsilon - 1)[a(s) - Ra_I(s)]\}^{-1}, \tag{1}$$

where $s \equiv l_t/l_z$, $R = (\epsilon - 1)/(\epsilon + 1)$, F is the fraction of the selvedge filled with material, and

$$a(s) = (s^2+1)^{1/2} - s,$$
$$a_I(s) = \tfrac{1}{2}[(s^2+4)^{1/2} + s] - (s^2+1)^{1/2}. \tag{2}$$

Once the polarization in the selvedge is known, the field that it generates in the bulk is easily determined.[13,16] This field interferes with the usual (zeroth order) refracted field to produce an inhomogeneous absorption given by $I(\vec{\kappa}) \sim \eta(\vec{\kappa}; \vec{\kappa}_i) \times |b(\vec{\kappa})|$, where $b(\vec{\kappa})$ is the Fourier component of surface roughness at $\vec{\kappa}$, and $\eta(\vec{\kappa}; \vec{\kappa}_i) = |\mu(\vec{\kappa}_+) + \mu^*(\vec{\kappa}_-)|$ with $\vec{\kappa}_\pm = \vec{\kappa}_i \pm \vec{\kappa}$ and $\mu(\vec{\kappa}) = \vec{E}_t^* \cdot \overleftrightarrow{h}(\vec{\kappa}) \cdot \overleftrightarrow{\gamma} \cdot \vec{E}_s$; here $\vec{E}_t$ is the amplitude of the usual refracted field, and the nonvanishing components of $\overleftrightarrow{h}$ are found to be

$$h_{ss} = 2i\tilde{\omega}(w_0 + w)^{-1}, \quad h_{\kappa\kappa} = 2iww_0\tilde{\omega}^{-1}(w_0\epsilon + w)^{-1},$$
$$h_{zz} = 2i\kappa^2\tilde{\omega}^{-1}(w_0\epsilon + w)^{-1},$$
$$h_{z\kappa} = 2i\kappa w_0\tilde{\omega}^{-1}(w_0\epsilon + w)^{-1}, \tag{3}$$
$$h_{\kappa z} = 2iw\kappa\tilde{\omega}^{-1}(w_0\epsilon + w)^{-1},$$

where $\hat{s} = \hat{\kappa} \times \hat{z}$, $w_0 = (\tilde{\omega}^2 - \kappa^2)^{1/2}$, $w = (\tilde{\omega}^2\epsilon - \kappa^2)^{1/2}$, and $\tilde{\omega}$ is the incident-beam wave number. If the bulk material supports surface polaritons the "efficacy factor" $\eta(\vec{\kappa}; \vec{\kappa}_i)$ exhibits peaks due to these excitations, whose dispersion relations are given by $w_0\epsilon + w = 0$; but even in polariton-inactive materials there are peaks due to nonradiative field structures that we have called "radiation remnants,"[9] which for the materials studied here occur at $\kappa \simeq \tilde{\omega}$. Since these peaks are so sharp (cf. Fig. 3), the detailed spectra of the surface roughness [$b(\vec{\kappa})$ as a function of $\vec{\kappa}$] is unimportant

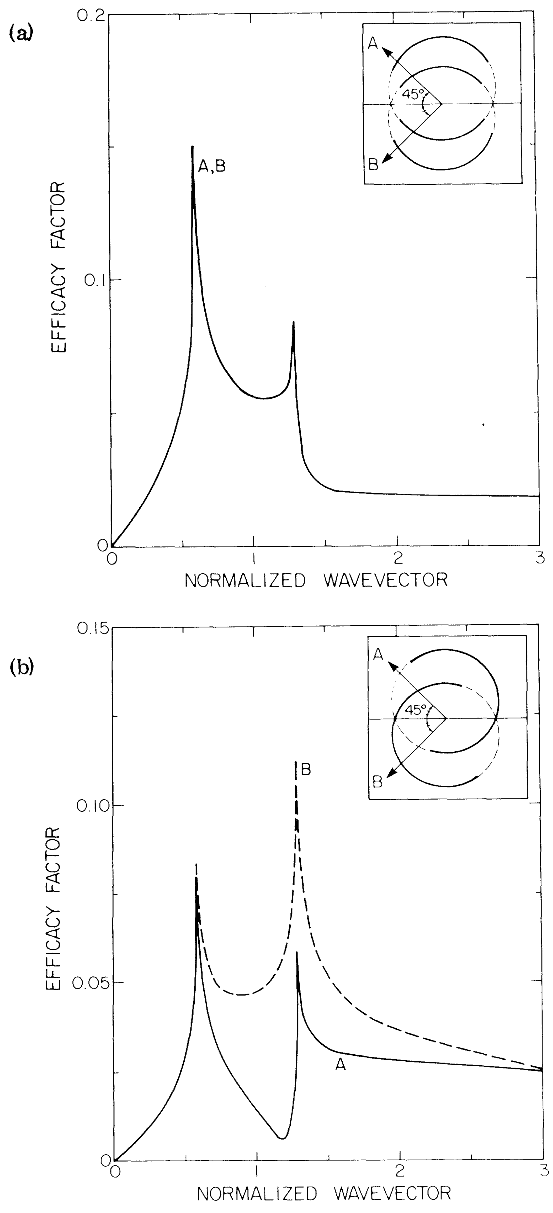


FIG. 3. Theoretical efficacy factors for Ge, $\lambda = 1.06$ μm, $\theta = 30°$, as a function of wave vector, normalized by $2\pi\lambda^{-1}$: (a) p-polarized light, (b) right-circularly polarized light. The inset depicts schematically the actual diffraction patterns with A, B indicating the two directions over which the theoretical plots were obtained.

in determining where in $\vec{\kappa}$ space the energy deposition is the strongest. Neglecting any feedback processes, we have found that not only the underlying circles shown in Fig. 2, but also the variations in intensity around these circles, are qualitatively given by the peak positions and peak heights in $\eta(\vec{\kappa};\vec{\kappa}_i)$. For example, we calculate that for circularly polarized light at $\theta = 0°$, the diffraction pattern should be a uniformly illuminated circle, as indeed it is. As another example, in Fig. 3 we show the relative inhomogeneous energy deposition in $\vec{\kappa}$ space for p-polarized and right-circularly polarized light in the case of 1.06-μm light incident on germanium (refractive index $= 4.1 + 0.1$) at 30°. The theoretical curves are constructed under the assumption that the surface can be modeled by isolated islands of roughness ($F \ll 1$) of comparable height and width ($s \simeq 0.4$) with both much less than λ. The plots show the magnitude of the energy deposition along two directions in $\vec{\kappa}$ space as indicated in the inset to the figure. In the case of p-polarized light the diffraction pattern is symmetric about the line of intersection of the circles (perpendicular to the polarization) and the theoretical plots in the two indicated directions are also identical. In the case of right-circularly polarized light the diffraction pattern is asymmetric and this is also borne out by the two plots shown for the two directions. The same symmetry characteristics are obtained for both types of polarized light in other directions in $\vec{\kappa}$ space and the relative intensities around the circles are correctly given in both cases. The overall agreement between theory and experiment is excellent, considering the simplicity of the theoretical model for surface roughness and the general nonlinear nature of laser damage. Similar agreement is obtained for other angles of incidence and polarization, and other target materials; the results are dependent on the values chosen for s and F, but not in a critically sensitive way.[14]

In our metal calculations the efficacy peaks are due to surface plasmons, which at the wavelengths studied here occur at $\kappa \simeq \tilde{\omega}$; in Si and Ge the peaks are due only to radiation remnants, since we have assumed no change in the material dielectric constants from their room-temperature values ($\epsilon_{\mathrm{Si}} = 11.8$, $\epsilon_{\mathrm{Ge}} = 16.1$). Of course, it is clear that any material will undergo significant structural change (e.g., melting) as the fringes are formed; however, our results indicate that the damage patterns can be understood largely in terms of the interference pattern set up before the optical properties of the material change significantly. Additional experiments and calculations are underway to clarify the role of the temperature dependence of the material and optical properties.

In summary, we have shown that laser-induced periodic structure is a universal phenomenon with respect to materials, wavelength, beam polarization, and details of surface roughness. This universality results because the class of surface resonances normally thought of must be extended to include radiation remnants, in addition to the usual surface polaritons. Since even a polariton-inactive material exhibits resonantlike response due to these field structures, fringe formation can be understood, and to some extent even expected, on every material.

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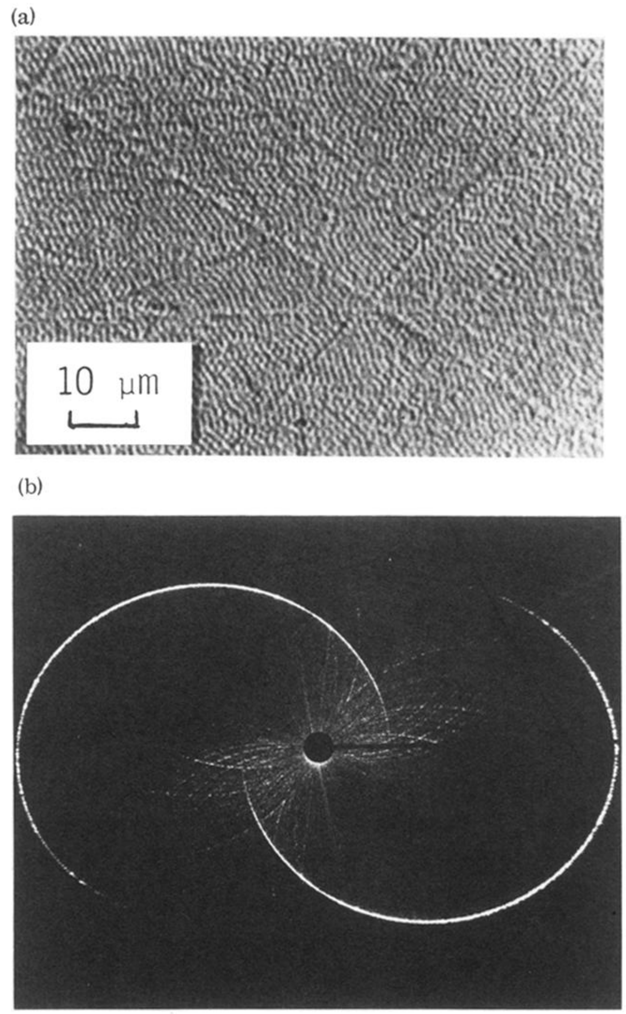

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