Regimes of laser-induced periodic surface structure on germanium: radiation remnants and surface plasmons

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We present experimental evidence showing that the period of the rippled surface structure induced on germanium by 1.06-μm laser pulses undergoes a discontinuous shift above a certain threshold intensity. The measured shift, as a function of the angle of incidence of the damaging beam, is quantitatively interpreted as a transition between a regime of inhomogeneous melting controlled by radiation-remnant field structures and a regime of ripple formation involving surface plasmons in an optically thick layer of liquid, metallic germanium formed at the surface.

During the past two years there has been a sharp increase of interest in the phenomenon of laser-induced periodic surface structure, which was first reported more than 15 years ago. In previous papers we thoroughly characterized the morphology of the laser-induced structure and commented on its universal nature; we also presented a detailed model that explains the effect as being due to inhomogeneous energy deposition, which results when the incident laser beam interferes with fields scattered from microscopic surface roughness. In the case of germanium irradiated by a low (<200-mJ/cm²) fluence of 1.06-μm, 20-nsec laser pulses, it was shown that the inhomogeneous energy deposition causes local melting of the surface, which does not recrystallize flat; so a permanent, localized structure is formed. Preliminary results indicated that at higher fluences the initial portion of the laser pulse caused uniform melting of the semiconductor surface, which then developed its own periodic structure. Molten germanium is a metal that can support surface-plasmon polaritons, and in this Letter we demonstrate the difference between the locally melted regime, which involves radiation remnants, and the uniformly melted case, which involves surface plasmons.

Because of the periodic nature of the induced surface structure, it is most convenient to work with its two-dimensional Fourier transform, which is easily obtained by observing the Fraunhofer diffraction pattern produced by scattering a weak cw probe beam from the surface. In germanium, the diffraction patterns that result from damage at low fluences consist of an underlying skeleton of two intersecting circles of radius \(\omega = \omega/\epsilon\), whose centers are translated with respect to the center of symmetry by the component of the incident wave vector parallel to the surface, \(\kappa_s\). With reference to Fig. 1, the Fourier components of the damage, \(\kappa_d = \kappa_i \pm \kappa_s\), are the same as those of an interference pattern that would be produced if the incident wave were to add coherently with waves scattered across the surface with wave vectors \(\kappa_s, |\kappa_s| \approx \omega\). To understand why the scattering is particularly strong for wave vectors of this magnitude, and to understand precisely which parts of this skeleton are actually filled in for a given angle of incidence, polarization, material, and wavelength, it is necessary to refer to the detailed electromagnetic calculation. For solid germanium \((\epsilon = 16 + 0.8i)\), the theory indeed predicts the detailed diffraction pattern observed. The theory also predicts that, for low-index insulators, wave vectors with \(|\kappa_s| = n\), where \(n\) is the refractive index, can also become important; such damage structures have been investigated by Soileau et al. In both cases, the phenomenological scattered waves are in fact found to be nonradiative field structures, which we have referred to as radiation remnants; they are closely related to the lateral waves referred to in other contexts. For polariton-active materials the theory predicts that there is a third possible value of \(\kappa_s\), namely, that associated with the dispersion relation for the surface polariton at the driving frequency. Because of the proximity of the light line to the surface-plasmon polariton line, it is particularly interesting to observe the scattering pattern for these wave vectors. The dashed region is shown in more detail in the inset of Fig. 3.

![Fig. 1. Schematic diagram of the diffraction patterns produced when an incident wave, with wave-vector component parallel to the surface, \(\kappa_s\), interferes with surface-scattered waves of wave vector \(\kappa_s\) or \(\kappa_s'\). \(\kappa_s\) represents the scattered wave vector on solid germanium, \(\kappa_s'\) is associated with the surface plasmons generated on molten germanium, and \(\kappa_d\) is a Fourier component of the damage induced on solid germanium. The dashed region is shown in more detail in the inset of Fig. 3.](image-url)
mon dispersion relation of most metals in the infrared and the visible, it is not clear from existing experimental data which field structure dominates the damage formed under such conditions, although the theory suggests that the polariton should. Ehrlich et al. have done work in the ultraviolet region of the light line; but the interpretation of their results is complicated by uncertainties in the dielectric constant at such frequencies. In order to demonstrate unequivocally a situation in which the surface plasmon is responsible for the modulation of the intensity, we consider the case of an optically thick layer of molten germanium; since the dielectric constant is known to be $\epsilon = -32 + 72i$ at 1.06 $\mu$m, the material is polariton active.

However, if the plasmon-dispersion relation is used, $\kappa_{sp} = \omega (\epsilon / c + 1)^{1/2} = \sqrt{2(1.003 + 0.006)}$; the plasmon resonance is centered at $\kappa_{sp} = 1.0036$, only 0.3% off the light line. But, because of the finite imaginary part of $\kappa_{sp}$, the maximum scattered field in phase with the incident field actually occurs at $\kappa_{sp} = R_{sp}$, with $R \approx 1.01$. It is possible to convert this 1% effect to a 3-6% effect by taking advantage of the interference that results in the periodic structure. To see this, refer to Fig. 1 and consider the consequence of leaving the incident wave vector unaltered while lengthening the surface-scattered wave vector by 1%. Some simple geometry shows that the separation of the intersection of the two circles in the two different cases is given by

$$|\Delta K| = \omega \left( R - \frac{1}{1 - \frac{\sin \theta}{K}} \right)^{1/2} \cos \theta,$$

where $\theta$ is the angle of incidence; thus, even though the surface plasmon is only 0.3% off the light line, a quite measurable shift of 3-6% in the position of the intersection of the two circles should occur if the germanium becomes uniformly molten.

The experiments were carried out at angles of incidence between 45 and 65 deg. The 1.06-$\mu$m beam was $s$ polarized, and the transient diffraction was monitored at the positions in $\kappa$ space where the two circles intersect. At fluences between 250 and 350 mJ/cm$^2$, the permanent diffraction patterns produced were of the form previously reported for such irradiation conditions, while the specular reflectivity never reached the uniform melt level. For fluences between 350 and 700 mJ/cm$^2$, the diffraction from the permanent structure produced in the central portion of the laser spot fills in the region around the intersection of the two circles. The 350-mJ/cm$^2$ level corresponds to the threshold at which the specular reflectivity during the first few shots reaches the characteristic flat-topped uniform melt value. Little structure is formed in the central region during the first few shots, but substantial periodic structure is formed in an annular region about the central spot where the local intensity is below the 350-mJ/cm$^2$ threshold. After 8 to 10 shots the specular reflectivity no longer reaches the uniform melt value, and significant growth (the formation) of periodic structure begins in the central region. This phase we have discussed previously; it is caused by strong modulation of the central intensity by the periodic structure built up in the annular region. The typical transient first-order diffraction from the central region is shown in Fig. 2(a). The slight dc-level shift represents a permanent increase in the height of the periodic structure caused by localized melting during the laser pulse. It should be emphasized that, below fluences of 700 mJ/cm$^2$, the diffraction patterns correspond to those produced by radiation-remnant types of field structures with $|\kappa_{p}| \approx \omega$. Viewed with a scanning electron microscope, the fringes formed in this regime result from localized restructuring of an otherwise flat surface.

Above 700 mJ/cm$^2$ the transient specular reflectivity always reaches the uniform melt value, and it remains at that level for longer than approximately 120 nsec. Again, little transient or permanent diffraction occurs from the central region for the first few shots. Once structure develops, the transient diffracted signal is of the form shown in Fig. 2(b), and it is detected at a position in $\kappa$ space corresponding to the intersection of two circles of radii $|\kappa_{p}|$ near the surface-plasmon wave

![Fig. 2. Typical transient diffraction signals observed at the intersection of the circles of radii (a) $|\kappa_{p}|$ and (b) $|\kappa_{p}'|$ shown in Fig. 1. The top traces in both (a) and (b) represent zero diffraction; the signals increase in the downward direction.](image)

![Fig. 3. A plot of the normalized wave vector of induced damage as a function of the 1.06-$\mu$m beam's angle of incidence. The vertical extremities of the data points along the top (bottom) curve correspond to the measured positions of points A and B (C and D) shown in the inset. The inset is a magnified view of the dashed portion of the diffraction pattern shown in Fig. 1. The solid lines represent points at which detailed electrodynamic calculations predict maximum inhomogeneous energy deposition in the case of (top) a liquid and (bottom) a solid germanium surface.](image)
number. The oscillations in the transient diffraction are due to the oscillation of capillary waves on the molten surface; this has been conclusively verified at smaller angles of incidence where the period of the capillary waves is much smaller. Under such conditions the period is so short and the damping so great that the capillary-wave activity dies out before the surface resolidifies, so that little permanent record is impressed upon the surface. However, at the larger angles of incidence where the current work was done, the resolidification takes place while the capillary wave is still oscillating, and hence a weak periodic structure with a wavelength of the capillary wave is frozen into the solid surface. This allows us to get accurate measurements of the shift in the intersection of the circles by positioning the probe beam so that the diffraction from the central and annular regions is superimposed. An example of such a pattern is shown in the inset of Fig. 3, which shows a plot of the experimentally measured position of the intersections of the two sets of different radii circles as a function of the angle of incidence. Viewed with a scanning electron microscope, the surface profile produced in this regime is sinusoidal and smoothly varying.

We note that the width of the curves that make up the patterns are approximately 30% broader in the surface-plasmon regime than in the lower-intensity case. The precise reason for this is not completely understood, but the measured width of the plasmon lines is within 30% of the calculated range in spacings obtained from Eq. (1) by taking into account the nonzero width of the plasmon resonance. The vertical range of the experimental points plotted in Fig. 3 represents this nonzero width rather than the experimental reading error, which was approximately 15% of the linewidths. The horizontal bars do represent experimental uncertainty. The solid line drawn through the lower set of data points is from an extension of our earlier electrodynamic calculation to treat periodic roughness and indicates at which wave numbers the maximum inhomogeneous energy deposition occurs for completely solid germanium; it follows the light line to within 1%. The other line is the corresponding predicted maximum for a liquid surface with a small-amplitude ripple and is associated with the surface-plasmon dispersion relation. Agreement in both cases is excellent. In the latter instance, the inhomogeneous energy deposition occurs at the troughs of the ripple, suggesting that an etching effect is responsible for building up the structure.

In conclusion, we have confirmed the role of radiation remnants in the formation of laser-induced periodic surface structure on germanium and identified the role of surface plasmons: these electromagnetic field structures are important, respectively, at low intensities when the material melts only locally and at high intensities when it melts uniformly to an optically thick (>1000-Å) depth. Finally, we note that the model for laser-induced periodic surface structure of Ehrlich et al., which invokes a uniform molten layer of germanium, does not seem to apply to the phenomena described here. That model, which relies on a ripple at the liquid–solid interface to lead to a ripple at the liquid–air interface, predicts good fringe formation, with surface-plasmon-controlled spacing, at low fluences when the molten layer is optically thin; at higher fluences, as the molten layer becomes optically thick, the coupling between the interfaces becomes weak, and poorer fringes should result. This is not the scenario that we observe.

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References
16. The fluences quoted in the text correspond to s-polarized 1.06-μm radiation incident at 60 deg.