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Time-resolved evolution of laser-induced periodic surface structure on germanium

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We have time resolved the reflection and diffraction of a probe beam from germanium during creation of periodic surface structure by a 20-ns, 1.06-μm laser pulse. Large, transient diffracted signals typically precede the development of permanent structure while the maximum reflectivity is much less than the melt value. We conclude that inhomogeneous melting and resolidification of the surface occurs following the interference of the incident beam and a nonradiative electromagnetic field.

Single, high-intensity laser beams can be used to induce highly uniform, periodic surface structure in metals, semiconductors, and dielectrics.[1–5] This curious phenomenon has received considerable attention in connection with laser annealing of semiconductors,[6] reflectivity of high-energy laser pulses from metals,[7] and the development of periodic structures in laser photodeposited metal films.[8] Recently, we have employed optical diffraction techniques[4,5] to reveal the richness of the damage structures. We have also developed[9] a detailed theoretical model to describe the results in terms of interference between the usual refracted beam and the electromagnetic field scattered by microscopic surface roughness. In the case of metals and certain dielectrics, these fields are associated with surface plasmon or phonon polaritons.[3,8,10] However, even in the absence of surface electromagnetic modes (which, for example, do not exist on solid semiconductors in the visible) nonradiative field structures can still lead to the necessary interference condition and thereby explain the wide variety of structures produced.[5,11] We suggest that the interference effect leads to inhomogeneous melting of the surface, which on resolidification does not return to its original profile. Alternative models have also been proposed: Some state that at no time is the material molten[6,12,13] and some, developed to treat semiconductor surfaces in particular, require that the surface melt uniformly, allowing surface plasmons in the metallic, molten phase to interfere with the incident beam.[8,14] In this Communication we report the results of nanosecond time-resolved reflection and diffraction experiments which clarify the nature of the mechanism responsible for periodic surface damage.

Laser-induced periodic surface structures were produced on crystalline, intrinsic germanium using 20-ns, 1.06-μm laser pulses. In general, the surface structures, and therefore their diffraction patterns, are dependent on the angle of incidence and polarization of the damaging beam. By way of example, we will here consider the transient and permanent effects produced by an s-polarized beam incident at 30° from the normal. The actual transient reflectivity and first-order diffracted signals were obtained using 0.51-μm cw argon ion laser beam, at near-normal incidence. This beam, focused to a diameter of less than 200 μm, was centered on the region where uniform ripples were produced, which was typically greater than 1 mm in diameter. Smaller probe beam spot sizes were also used with no difference in results. Two photomultipliers were suitably located to record the reflected and diffracted beams in the far field. The outputs were recorded on storage oscilloscopes with system response times of better than 5 ns in each case. Figure 1 illustrates the Fraunhofer diffraction pattern produced by permanently induced structure at different fluences. The dominant feature

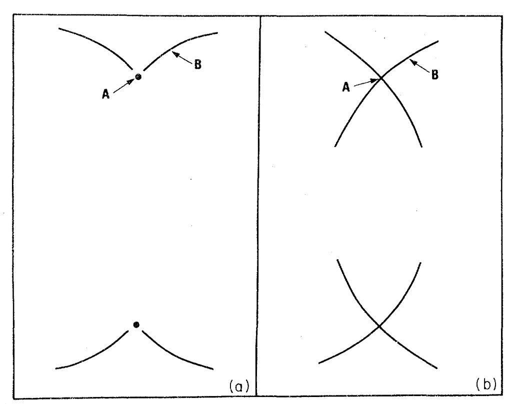

FIG. 1. Schematic illustration of the diffraction patterns from damaged Ge surfaces for which the specularly reflected signal was always less than (a), and greater than (b), the molten value of 0.74. Points A and B indicate two different positions at which the detector was located to monitor the transient diffracted signal.

in Fig. 1(a) (Ref. 5) is a pair of bright spots indicative of a ripple spacing of 1.06 μm/cos30°. These ripples are oriented perpendicular to the 1.06-μm beam polarization. Wings extend out from the bright spots which correspond to sets of ripples of slightly different orientation and spacing.

In our first experiment, laser fluences between 100 and 150 mJ/cm^2 were used, and a phototube was located at point A in Fig. 1. The diffracted signals observed for the first five shots consist of a large 30-ns transient pulse. Little permanent structural change was observed following these pulses. During the following two to four pulses the transient signal increases dramatically and is followed by significant permanent damage, indicated by an increase in the dc level of the diffracted signal. A typical diffracted signal obtained during this stage of development is shown in Fig. 2. Although the ratio of the transient to dc signals may be as high as 10 during this stage, for subsequent pulses this ratio is reduced so that after approximately 10 shots no large transient peaks are observed, but only small positive dc shifts. After about 20 pulses an approximately "steady state" is achieved, which is characterized by very small transient diffracted signals and gradual changes of the induced structure upon further irradiation. During the entire development the reflectivity rises from an initial value of 0.52 to a maximum value significantly less than 0.74, the value characteristic of homogeneously melted germanium.[15] The reflectivity rises, in the example of Fig. 2, to a value of 0.65.

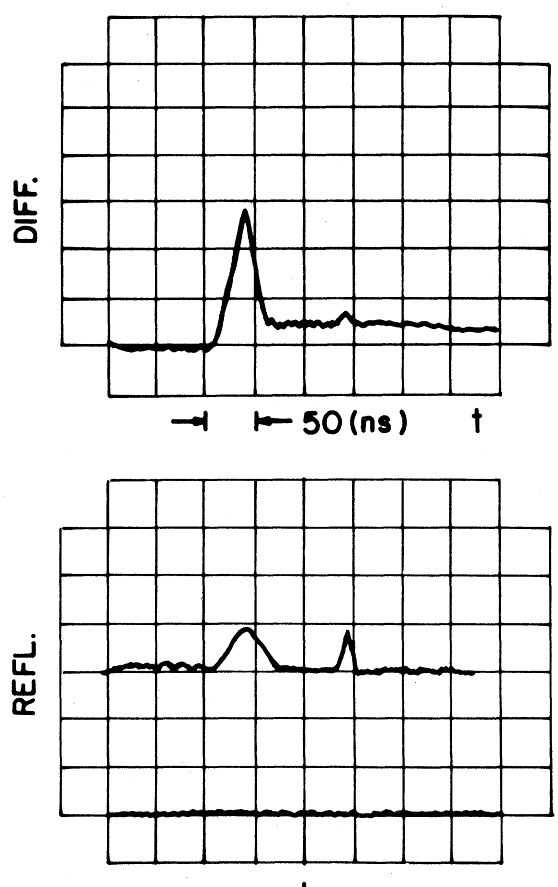

FIG. 2. Oscilloscope traces of the transient diffracted (top picture) and specularly reflected (lower picture) signals obtained at a fluence of ~110 mJ/cm^2. The diffraction detector was placed at point A (Fig. 1). The base line on the lower picture corresponds to zero reflectivity. Note the timing signal ~150 ns after the pulses which allows synchronization of the two traces.

For laser fluences in excess of 150 mJ/cm^2 the reflectivity does reach the value characteristic of homogeneously melted germanium, and both the transient and permanent diffracted signals are dramatically different from the case mentioned above. For example, the diffraction pattern of the permanent damage changes from that of Fig. 1(a) to that of Fig. 1(b). In addition, for the detector at point A, large peaks in the transient signals are absent and only a monotonic shift in the dc level is observed. If the phototube is located in the wings (point B) of the diffraction pattern, transient signals as illustrated in Fig. 3 are observed. The signal consists of two large peaks during the rising and falling edges of the reflectivity with a minimum, observed while the reflectivity is flat topped at a value of 0.74. Small dc level shifts also remain following the second peak. For both detector locations, when the laser fluence is sufficient to quickly melt, and keep the entire surface molten for tens of nanoseconds, all large peaks disappear and only monotonic shifts in the dc level of the diffracted signals are observed.

In summary, we have demonstrated that permanent periodic surface structure can be produced on germanium with single 1.06-μm laser pulses under conditions such that the specular reflectivity of a 0.51-μm probe beam remains significantly below the 0.74 value characteristic of a uniformly melted surface. When the fluence of the 1.06-μm damaging beam is increased to ~150 mJ/cm^2, the specular reflectivity acquires a flat-top structure at a value of 0.74 and fringes are still produced. The diffracted signal changes in three dramatic ways when the reflectivity reaches 0.74 at some point during the pulse. First, the permanent diffraction pattern no longer consists of a discontinuously bright spot at the intersection of two smooth arcs (see Fig. 1). Second, the large peaks in the transient diffracted signal observed at point A of Fig. 1 are absent. Third, the diffracted signal at point B of Fig. 1 consists of two large peaks during the rising and falling edges of the reflectivity, with comparatively little signal in between while the reflectivity has a value of 0.74.

The simplest explanation for the low maximum reflectivity mentioned above would be that the ger-

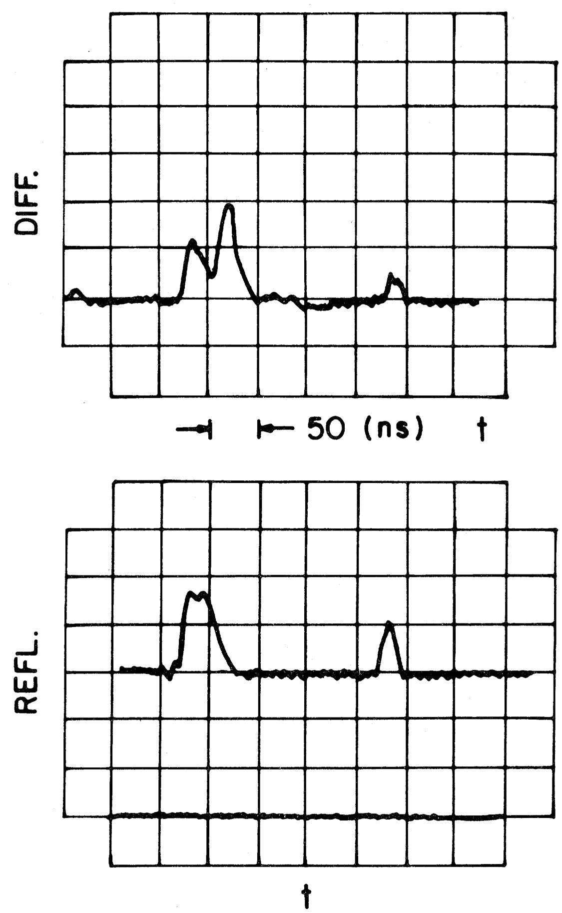

FIG. 3. As in Fig. 2, but for the diffraction detector at point B (Fig. 1) and the fluence at ~ 160 mJ/cm^2.

manium indeed melts uniformly, but only in a layer of thickness less than the skin depth of the probe beam. We consider this unlikely because of (a) the discontinuous behavior, described above, at the point when the reflectivity does reach 0.74, (b) the similar rise and fall times in the reflectivity (Fig. 2) and the independence of these times on the intensity as long as the peak reflectivity is less than 0.74, and (c) the difficulty in understanding particularly the double peak diffraction signal (Fig. 3) in the context of a uniformly melted layer. In any case, we note that even if such a thin uniformly melted layer did exist, the surface would not be polariton active: The calculated Fresnel coefficients, at 1.06 μm, of a solid substrate covered with a molten layer of the right thickness (~ 60 Å) to raise the reflectivity at 0.51 to 0.65 μm are very similar to those of a solid substrate, and do not exhibit a metallic-type surface plasmon resonance. Thus, at least in our low fluence experiments, it seems clear that surface plasmons are not involved in the formation of transient or permanent fringes.

Although additional work is required in order to completely understand the nature of the observed behavior, we feel that our results are consistent with a model in which the surface first melts nonuniformly in a periodic array due to inhomogeneous energy deposition in the solid. Such a scenario does not require surface plasmons, and the nature of the electromagnetic fields involved has been discussed earlier.[4,5,9,11] The large spike in the diffracted signal would then be expected, since alternating regions of high and low reflectivity should provide an efficient grating compared to small periodic structural defects on a uniform solid or liquid surface; the second of the two peaks in the double peak diffraction signal could be attributed to the alternating regions that reappear when the less deeply melted regions of the surface solidify first. At high fluences, the melt extends over the whole surface very quickly, and so quenches the strong diffraction signal.

Finally, we mention that in some instances inhomogeneous energy deposition could lead to permanent periodic damage without even the nonuniform melting we feel is important here, as in the regrowth of amorphous films,[12] or through nonequilibrium phase transitions induced by picosecond pulses.[13]

ACKNOWLEDGMENT

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