Second-Harmonic Efficiency and Reflectivity of GaAs During Femtosecond Melting

J.-K. Wang, P. Saeta, Y. Siegal, E. Mazur, and N. Bloembergen

Division of Applied Sciences and Department of Physics,
Harvard University, Cambridge, MA 02138, USA

1. Abstract

Second-harmonic and reflectivity probes of photoexcited GaAs demonstrate melting on a 200-fs timescale and partial recrystallization of the molten material.

2. Introduction

Studies of the ultrafast melting of semiconductor surfaces under intense laser irradiation at frequencies above the bandgap reveal changes in the dielectric properties on the sub-picosecond timescale. Reflectivity measurements of silicon with femtosecond pulses demonstrate significant changes within 300 fs [1]. Second-harmonic measurements on silicon further show that the top 70–130 Å of the silicon surface loses its cubic symmetry within 150 fs [1]. In this paper we report on simultaneous reflectivity and second-harmonic generation (SHG) measurements during the melting of GaAs with 160-fs laser pulses. The results show bulk melting to a depth of at least 90 atomic layers within the pulsewidth.

3. Experimental

Amplified 200-μJ pulses at 620 nm are split into a pump and a probe beam with an energy ratio of 20:1. The *p*-polarized probe pulse is incident upon a (100) GaAs surface at 45°; the pump is orthogonally polarized and enters along the normal. The probe interrogates the central one-tenth of the pumped area. We determine the origin of time by attenuating the pump beam and maximizing the reflection sum frequency signal of the pump and probe beams. For each laser shot, the reflected intensity and the second-harmonic intensity are measured. In addition, we check for the presence of broadband ultraviolet radiation from a surface plasma. The pump beam by itself generates no spurious reflection or second-harmonic signal.

4. Results and Discussion

Figure 1 summarizes the reflectivity measurements normalized to the appropriate value for negative times. The dip in reflectivity at low fluence in the temporal range

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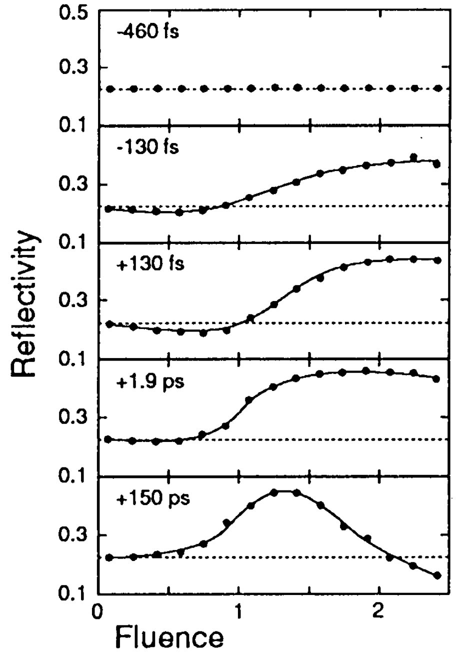

FIG 1. GaAs reflectivity vs fluence for various time delays. A fluence of 1 indicates where the SH efficiency first drops to zero.

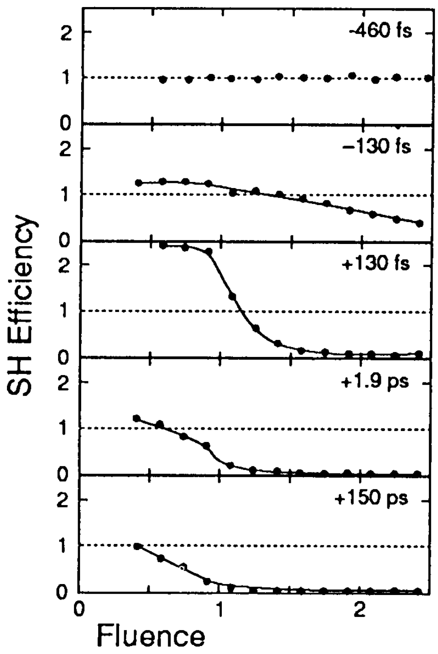

FIG 2. GaAs second-harmonic efficiency vs fluence for various time delays. A fluence of 1 indicates where the SH efficiency first drops to zero.

of −130 fs to +1.9 ps can be attributed to a dense free-carrier plasma created by photoexcitation [2]. This plasma decreases the real part of the dielectric constant, leading to a drop in reflectivity. By +130 fs the reflectivity at high fluence rises to roughly twice the crystalline value, consistent with the value predicted by a Drude model for metallic molten GaAs. After 50 ps the reflectivity at high fluence drops dramatically. This drop is probably due either to heating of the molten material far above the melting temperature [3], or to the delayed formation of a plasma cloud in front of the surface [4,5].

The second-harmonic signal is divided by the square of the probe intensity to obtain the second-harmonic (SH) efficiency, normalized to unity for unpumped material. Figure 2 shows a rapid, dramatic decrease to zero in SH efficiency as early as +130 fs, coincident with the observed plateau in reflectivity. The absence of any second-harmonic signal in this region indicates melting to a depth of at least 90 atomic layers, the absorption depth of the second harmonic.

Concurrent with the observed dip in reflectivity at short times and low fluence one finds a marked increase in SH efficiency. This increase is caused by a change in index of refraction due to the presence of the free-carrier plasma. First, lowered reflectivity results in an increased probe intensity beneath the surface. Second, with a decrease in the real part of the index, the angle between the probe *E*-field inside the material and the $<0\bar{1}0>$ crystal axis increases from 10° to about 15°, resulting in approximately a factor 2 increase in SH efficiency.

We also measured the reflectivity and SH efficiency after complete equilibration of the material. The reflectivity rises again and settles at a value of 0.24, slightly higher than the value of crystalline GaAs (0.22), but lower than that of amorphous GaAs (0.28); the SH efficiency recovers to roughly half its original value. The net increase in reflectivity rules out any ablation of surface material, while the second harmonic from the resolidified material suggests a partial recrystallization.

5. Conclusion

In summary, we have performed femtosecond pump-probe measurements of the second-harmonic efficiency and reflectivity of GaAs during melting. Within 200 fs the second-harmonic signal drops to zero at high fluence, indicating melting to a depth of at least 90 atomic layers.

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