Femtosecond, Electronically-Induced Disordering of GaAs

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Picosecond laser pulse experiments on semiconductors show that a thermal melting model can explain the behavior of these materials when irradiated with such pulses.[1] However, femtosecond experiments suggest that the thermal model breaks down if the energy of the laser pulse is deposited into the electronic system on a time scale which is short compared to the electron-lattice energy relaxation time (2-3 ps).[2,4-7] In this case, the bond breaking which occurs through the excitation of an electron-hole plasma can lead directly to ionic disordering while the lattice is still cold.[3] We have performed a series of measurements on GaAs using 160-fs, 620-nm pulses which show that above a threshold pump fluence of 0.1 J/cm^2, the material takes on a centrosymmetric structure within the pulse duration in a process which is directly driven by the electronic excitation.

We chose GaAs for this experiment because its lack of inversion symmetry results in a non-zero bulk, dipole, second-order susceptibility. Because disordering leads to a loss of this second-order susceptibility, measuring the reflection second-harmonic (SH) signal generated by a probe beam as a function of pump-probe delay time monitors the symmetry of the sample. The sample was aligned such that SH generation from the pump beam was suppressed, and the polarization of the probe beam was orthogonal to that of the probe beam. The sample was translated between shots to assure a fresh spot for each measurement.

Figure 1 summarizes the immediate response of the material for an incident fluence of 0.36 J/cm^2, which is about 3.6 times the damage threshold, on the (110) GaAs sample. Both the reflectivity and the SH signal are normalized to unity for no pump excitation. The SH efficiency drops to zero with a decay time of 90 fs while the linear reflectivity increases to a plateau 44% greater than that of the unpumped material with a rise time of 170 fs. The sudden rise in the reflectivity is evidence for a transformation to a metallic state. The vanishing of the SH suggests that the electronic system takes on a centrosymmetric character consistent with disordering of the lattice. This transition to a disordered phase occurs an order of magnitude faster than the electron-phonon energy relaxation time of 2 ps.[2,5] From the reflectivity results,[5-7] the dielectric constant of the transient disordered phase is determined to be $-0.31+i0.34$, which is inconsistent with a Drude model. This suggests a phase which is not an equilibrium liquid.

Information on the carrier density upon disordering can be derived from the results on the (100) GaAs sample. Figure 2 shows the results on the (100) GaAs sample at a pump fluence of 0.58 times the damage threshold. The SH efficiency rises to 2.4 while the reflectivity decreases to 0.85 by about 130 fs after the pump pulse peak. This rise in the SH is not due to changes in

Springer Series in Chemical Physics, Vol. 55 **Ultrafast Phenomena VIII**
Editors: J.-L. Martin · A. Migus · G.A. Mourou · A.H. Zewail
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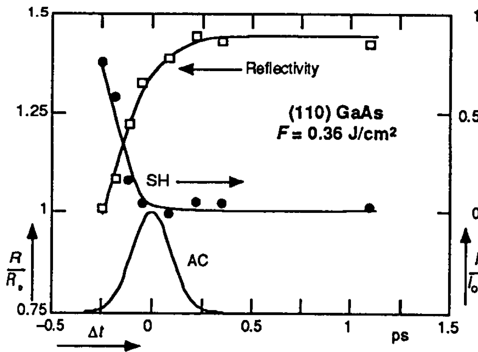

Fig. 1 Time response of second harmonic and linear reflectivity in (110) GaAs to 160-fs laser pulse. Second harmonic vanishes with 90-fs decay time while reflectivity rises to a plateau with 170-fs rise time.

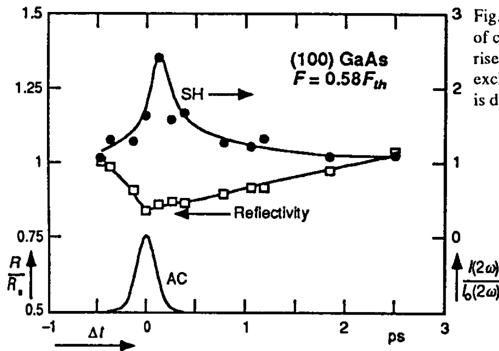

Fig. 2 Excitation at 0.58 times damage threshold shows effects of changes in linear dielectric constant. Second harmonic rises to peak of 2.4 while reflectivity drops to 0.85 due to excitation of free carriers. Quick recovery of second harmonic is due to fast Auger recombination.

the second-order susceptibility, but to changes in the linear dielectric constant caused by the excited free carriers, providing a very sensitive probe of the free carrier density. In this regime, we can use a Drude model for the dielectric constant, and fits to both the SH and linear reflectivity results yield a free carrier density of about 1×10^{22} cm^{-3}. The fast decay of the rise in the SH signal indicates an Auger recombination time of 400 fs. This rapid recombination puts a cap on strong nonlinear absorption, dominated by impact ionization driven by free-carrier absorption. Thus, the carrier density saturates at about 1×10^{22} cm^{-3} before the second-order susceptibility begins to drop due to the onset of disordering as the incident fluence approaches the damage threshold. This experimentally determined critical free carrier density agrees with that predicted by Van Vechten for electronically-induced disordering.[3]

This work was supported by the Joint Services Electronics Program and the Material Research Laboratory Program under contracts with Harvard University. YS acknowledges a Ph.D. fellowship from the National Science Foundation.

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