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Induced Absorption in Silicon under Intense Laser Excitation: Evidence for a Self-Confined Plasma

A. Aydinli, H. W. Lo, M. C. Lee, and A. Compaan
Department of Physics, Kansas State University, Manhattan, Kansas 66506

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Time-resolved transmission and reflectivity of silicon on sapphire have been studied at $\lambda = 1152$ and 633 nm following excitation by a ~1 J/cm^2, 8 nsec pulse at 485 nm. In addition, the spectral dependence of the transient absorption was obtained for the range 1.3 to 3.3 eV with use of a pulsed–dye-laser probe. The results are inconsistent with a metallic molten state but suggest the existence of a self-confined dense carrier system similar to that proposed by Van Vechten and Wautelet.

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The behavior of semiconductors under intense laser excitation and therefore high carrier densities has been the subject of considerable interest since the advent of high power lasers. In particular, transient absorption due to laser-excited free carriers[1] and enhanced reflectivity[2] induced by intense laser pulses have been observed. Recently this subject has received renewed interest because of the excitement surrounding the technique of laser annealing.[3] In recent studies,[4] the onset of a high-reflectivity phase during laser annealing of ion-implanted silicon has been used to infer the presence of a transient molten phase at the surface. This inference is based on the similarity of the transient reflectivity values to those of steady-state molten silicon,[5] although some discrepancies have been noted.[6] However, this melting interpretation is now suspect since a direct measurement of lattice temperature by pulsed Raman scattering has shown only a very small (~300 °C) rise in lattice temperature for periods of tens of nsec after an ~8-nsec excitation pulse.[7] Other experimental evidence against a molten phase has been obtained from a transient–electrical-conductivity measurement[8] and from cw Raman line shape studies of laser-annealed silicon-on-sapphire (SOS) samples.[9] Furthermore, we have recently shown by a time-resolved transmission measurement at 1152 nm on bulk, single-crystal silicon[10] that this high-reflectivity phase is not accompanied by the absorption appropriate for molten silicon with its skin depth (absorption length) of ~100 Å. In this Letter we have used SOS samples to obtain the full wavelength dependence (from 1.1 to 3.3 eV) of the optical absorption induced during the enhanced-reflectivity phase. The results appear to confirm the importance of dense-plasma effects in pulsed-laser annealing.[11]

Time-resolved reflectivity and transmission measurements were performed with the 1152- and 633-nm lines of a He-Ne laser. These cw probe beams were focused to a spot size of ~30 μm on the SOS sample which was excited by the 8-nsec pulse from a N$_2$-laser-pumped dye laser ($\lambda = 485$ nm) focused to a spot of diameter ~200 μm. To obtain the spectral dependence of the transient absorption, we used a second dye laser pumped by the same N$_2$ laser. The energy density of this probe laser was attenuated to below 0.01 J/cm^2 and the pulse delayed optically by 25 nsec. Signals generally were detected by silicon or germanium *p-i-n* diodes or an RCA model-7102 photomultiplier and photographed on a Tektronix model-7904 oscilloscope. Special care was taken[10] to guard against light leakage around the excited region of the sample. Most experiments were performed on a 0.6-μm-thick sample although some data was also obtained on SOS samples with silicon thicknesses of 1.16, 1.8, 2.0 μm, and an SOS wedge ~0.05 to 0.3 μm thick.

The time-resolved reflectivity and transmission signatures of the 0.6-μm SOS sample are shown in Fig. 1 for an excitation pulse energy[12] of 0.8 J/cm^2. Signals could generally be reproduced for several consecutive laser shots on the same sample position although usually a fresh sample spot was used for each trace. The general features of both the reflectivity and transmission signals were independent of the various SOS thicknesses. We believe optical interference effects play no significant role in the results reported here.[13] Our major emphasis in this Letter is to examine the effects which occur during the initial high-reflectivity phase (0 to 70 nsec). A detailed discussion of the transmission and reflectivity signals at longer times will be presented in a subsequent publication.

During the enhanced-reflectivity phase (0 to 70 nsec) little absorption occurs at 1152 nm as shown in Fig. 1(a). A similar result was seen in bulk, crystalline silicon.[10] We find that any

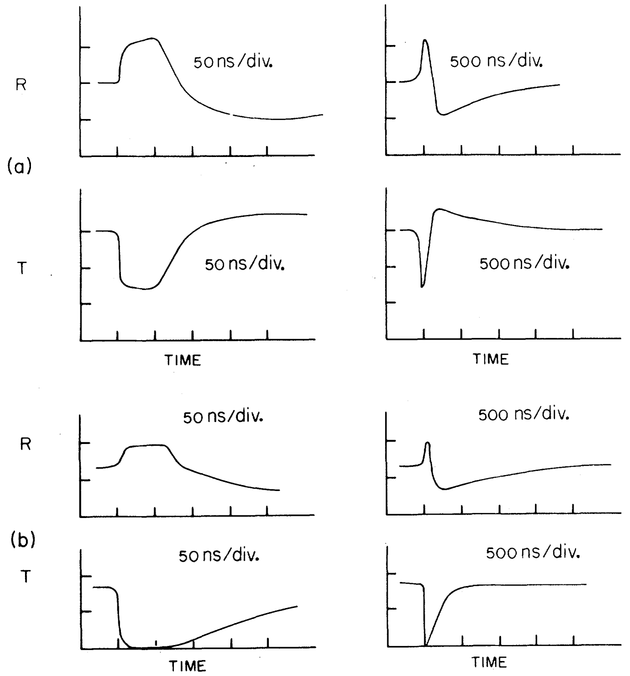

FIG. 1. Time resolved reflectivity (*R*) and transmission (*T*) of 0.6 μm SOS for probe wavelengths (a) 1152 nm and (b) 633 nm. Peak of the 8-nsec excitation pulse occurs on the first horizontal tick mark. Peak reflectivity at both wavelengths is approximately 0.6.

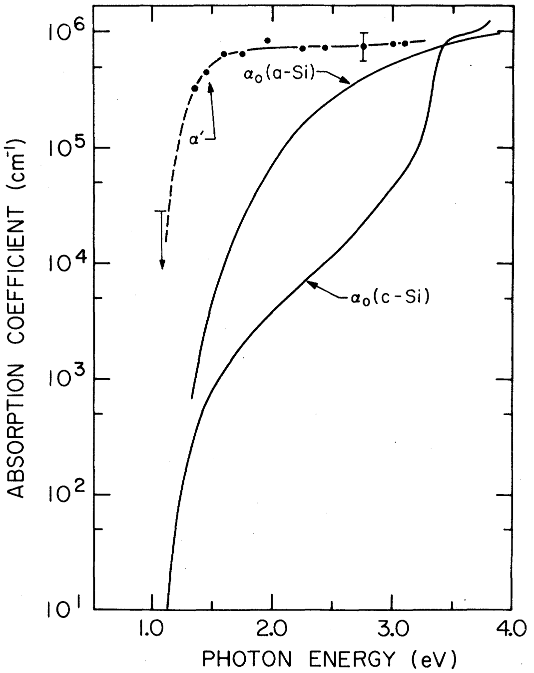

FIG. 2. Induced absorption coefficient (dots) measured 25 nsec after pulsed excitation. The curves for amorphous and crystalline silicon are taken, respectively, from Refs. 14 and 15. Typical error shown is largely due to uncertainty in the thickness of the absorbing layer. (See text.)

transmission loss not explained by the reflectivity rise must be less than 20%, i.e., $I_T/I_0 = 0.8$. At 633 nm, however, [Fig. 1(b)] there is indeed a large increase in absorption during this high-reflectivity phase as was seen at 1.06 μm by Auston *et al.*[4] As a first step toward understanding the physical origin of this phase we have obtained the spectral dependence of the induced absorption from 1.3 to 3.3 eV, using a second pulsed dye laser as a probe. The results of these pulsed transmission measurements are summarized in Fig. 2 where we show the absorption coefficient during the enhanced-reflectivity phase. The data have been corrected for the transient reflectivity change with use of the factor $(1-R')/(1-R_0) = 0.6$, which is assumed to be constant over the wavelength range studied. R' is the enhanced reflectivity and R_0 is the reflectivity before the excitation pulse arrives.

The data points in Fig. 2 have been calculated for an absorbing region 0.07 μm thick—considerably less than the full silicon thickness of 0.6 μm. This choice is explained as follows: For all sample thicknesses including the SOS wedge down to a thickness of 0.07 μm, the intensity ratio with (I_T) and without (I_T^0) the 485 nm excitation pulse was $I_T/I_T^0 = (8 \pm 4)\times 10^{-3}$ for $\lambda < 700$ nm. For a wedge thickness of 0.05 μm the ratio was 0.1. We believe these observations show the absorbing region to be *self-confined* to a thickness of 0.07 μm because carrier diffusion in normal silicon should easily be sufficient in 25 nsec to homogenize the carrier density throughout the 0.6 μm SOS thickness.[16]

We now demonstrate that the induced absorption shown in Fig. 2 is consistent with the values of enhanced reflectivity shown in Fig. 1 for the period 0-70 nsec: Knowledge of the change in the imaginary part of the index of refraction $\Delta n_I = (c/2\omega)\Delta\alpha$ over an extended wavelength range permits the use of a Kramers-Kronig analysis to calculate the real part of the change in refractive index or equivalently, the change in reflectivity. Taking $\Delta\alpha = 7\times 10^5$ cm^{-1} from 1.45 to 3.3 eV and falling smoothly to zero at 1.15 and at 3.5 eV, the Kramers-Kronig analysis yields $\Delta n_R = 3.6$ at 1152 nm and thus the enhanced reflectivity is $R' = [(1-n_R)^2 + n_I^2]/[(1+n_R)^2 + n_I^2] = 0.57$. Similar results are obtained at 633 nm.

Thus, the absorption is consistent with the observed reflectivity and together they provide the first direct experimental evidence of a self-confined carrier system in highly excited silicon. The possibility of such a confinement occurring was first raised by Van Vechten and Wautelet, who have given detailed thermodynamical arguments for the existence of a confined plasma when electron and lattice temperatures are far from equilibrium.[17] Our earlier Raman measurements of temperature show that this is indeed the case. The lattice remains quite cool (~300 °C) and therefore, the electron system must be very hot since it retains most of the absorbed laser energy. The absorption measurements now provide clues to the electronic structure of the enhanced reflectivity phase. The optical band gap shows little shift although it now closely suggests a direct-band-gap semiconductor. Alternatively the dense plasma may give rise to a strong carrier-assisted band-to-band absorption[18] replacing the usual phonon-assisted process in normal silicon.

Beyond 100 nsec, the reflectivity at both 1152 and 633 nm drops far *below* that of unexcited room-temperature silicon and consequently the transmission in the transparent region at 1152 nm increases *above* the value before excitation.[19] (See Fig. 1.) The amplitude and duration of this depressed reflectivity diminish as the silicon thickness is increased to 2 μm and this feature is not present at all in bulk silicon.[10] We believe that the behavior in this regime might be explained by the optical response of a dense plasma frequency, ω_p, slightly less than the probe frequencies.[20] Confinement of the plasma apparently ceases with the end of the enhanced reflectivity, and at this point rapid carrier diffusion returns and carrier density decreases rapidly. In bulk silicon the carrier density may drop by orders of magnitude in times of the order of nanoseconds but in SOS the sapphire provides an effective barrier thus prolonging the reduced-reflectivity period. Using a background dielectric constant of $\epsilon = 11$, we estimate a plasma density of the order of 5×10^{21} cm^{-3} would be required to produce the depressed reflectivity observed near 200 nsec in Fig. 1. During the high-reflectivity phase the volume is smaller by 0.07 μm/0.6 μm so that the density would be of the order of 4×10^{22} cm^{-3}. However, the Kramers-Kronig analysis shows that N_R increases from 3.6 to 7.2 at 1152 nm so that $\omega_p{}^2$ should not rise in proportion to the plasma density. Furthermore, the data of Fig. 2 indicate in the dense phase the optical absorption is more complex than that of a simple free-carrier plasma plus normal interband transitions. Van Vechten and Wautelet[17] have discussed some of these effects.

In conclusion, we believe that these experiments have provided the first good evidence for the nature of the high-reflectivity phase which occurs during laser annealing of silicon. We think that this state is substantially the same whether the material begins amorphous as in most laser annealing experiments or crystalline as is the case in these experiments. Preliminary experiments performed on ion-implantation-amorphized SOS samples appear to confirm the similarity. This enhanced reflectivity phase is characterized by high carrier density, high carrier temperature, low vibrational temperature, and self-confinement of the plasma to a region of the order of 0.07 μm. The experiments at this point provide no evidence for how the normally fast (~10^{12} sec^{-1}) electron-phonon scattering is suppressed which is a necessary requirement for decoupling the electron system from the vibrational system. Additional theoretical and experimental work is clearly required.

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Study of Random Magnetic Alloys near Their Critical Concentrations under High Pressure

C. W. Chu, M. K. Wu, B. J. Jin,[(a)] and W. Y. Lai[(b)]
Department of Physics and Energy Laboratory, University of Houston, Houston, Texas 77004

and

H. S. Chen
Bell Laboratory, Murray Hill, New Jersey 07974
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The low-field ac magnetic susceptibility of the amorphous (FeMn)PBAl and (FeNi)-PBAl, and the crystalline (PdFe)Mn random magnetic alloys near their critical concentrations was measured under pressure up to 20 kbar between 1 and 300 K. The results support the proposed existence of reentrant ferromagnetism, and show the important role of magnetic clustering in the magnetic behavior of these alloys.

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In a random magnetic metallic system, the delicate balance between various interactions dictates the magnetic structure at low temperature. The system[1] can be made of a Kondo system, a spin-glass, or a ferromagnet by continuously changing the magnetic-impurity concentration.