Indirect Two-Photon Transitions in Si at 1.06 μm

John F. Reintjes* and James C. McGroddy
IBM Thomas J. Watson Research Center, Yorktown Heights, New York 10598
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We have studied the nonlinear optical absorption in silicon at 1.06 μm using picosecond pulses. By measuring the time dependence of the induced absorption we are able to separate the effects of indirect simultaneous two-photon absorption and stepwise absorption processes. Values of the two-photon absorption coefficient at 1.06 μm were measured to be 1.9 cm/GW (20 K) and 1.5 cm/GW (100 K).

Nonlinear absorption in semiconductors has received attention in the past, partially because of its potential applications in optical limiting. In silicon, nonlinear absorption has been reported at temperatures[1-3] between 80 and 300°K using Q-switched pulses from a Nd-doped yttrium-aluminum-garnet laser. The observed results have been attributed variously to two-photon absorption[1] and to two- and three-photon stepwise absorption processes,[2,3] in which an electron makes a transition from the valence band to the lowest Δ_1 conduction band via a linear (one-photon) indirect absorption process and is subsequently raised to a higher conduction-band state by the absorption of one or more additional photons. In this Letter we report studies of nonlinear absorption in Si with picosecond pulses from a Nd:glass laser. By using such short pulses we are able to observe, in addition to the stepwise processes above, the true nonlinear absorption due to the simultaneous absorption of two photons accompanied by the creation or annihilation of a phonon. By making time-resolved measurements of the nonlinear absorption we have been able to measure the relative importance of the instantaneous and stepwise absorption processes.

The experimental arrangement is shown in Fig. 1. The pulse train from the mode-locked laser passes through a pulse selector which transmits five successive pulses, the separation between successive pulses being 10 nsec. These pulses are amplified in a Nd:glass amplifier and then divided into two beams with a 3% glass beam splitter. After passing through appropriate optical delays, the two beams are recombined in the sample at a small angle (about 1°). The main (strong) beam is used to generate the nonlinear absorption in the sample and the probing (weak) beam is used to sample the resulting nonlinearity. Time-dependent measurements are obtained by varying the relative time of arrival of the two beams with the variable path delay. Only the first pulse of each set of five is used for measurements, since the stepwise absorption due to free carriers accumulates during the pulse train.

The sample is a 1-cm-long single crystal of undoped Si oriented so that the beam is polarized

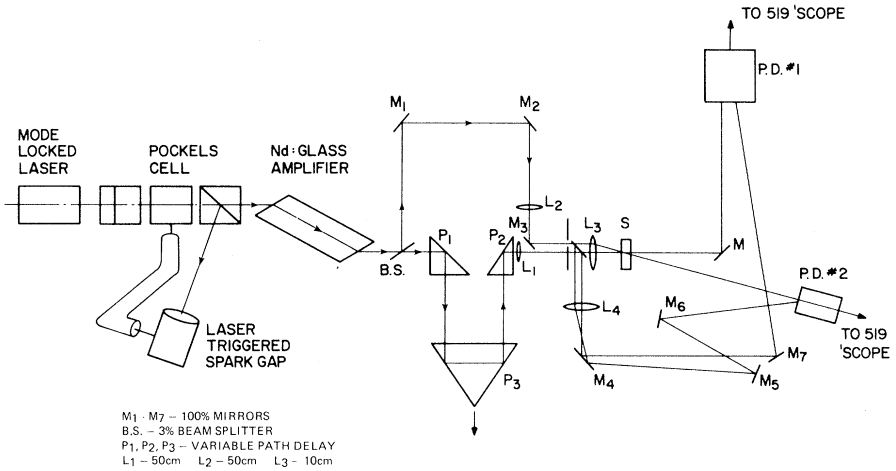

FIG. 1. Experimental arrangement for measuring nonlinear absorption.

along a ⟨100⟩ or ⟨110⟩ direction. No orientation dependence of the effects was observable. The sample was tilted slightly from normal incidence so that multiple reflections were not detected.

The nonlinear absorption was observed by measuring the intensity-dependent attentuation of the light transmitted through the Si. Signals were detected with a planar photodiode and a Tektronix 519 oscilloscope. The main and probing beams were observed on separate detectors and suitably delayed reference pulses were displayed for each beam on each laser shot. The beam profile was measured using an array of 256 Si photodiodes, and the intensity distribution over the beam cross section was found to be considerably more uniform than a Gaussian distribution.

Figure 2 shows the results of measurements of the transmitted fraction of the main beam as a function of incident intensity at 20°K. The vertical scale plots "absorbance" (inverse of transmittance) normalized to 1 at low intensities, and the horizontal scale plots incident laser intensity. A pulse duration of 20 psec, determined by separate two-photon fluorescence measurements, was used to convert the measured pulse energy to intensity. The dots are the measured points and the straight line is a plot of the curve $I_0/I = 1 + \beta L I_0$ which describes pure two-photon absorption for a beam with a uniform distribution of intensity. A reasonably good fit is obtained with a value of $\beta = 1.9$ cm/GW, which is more than 5 times smaller than the smallest previously reported value,[1-3] indicating that previous results were due primarily to stepwise absorption. Band-structure calculations[4] as well as optical absorption data[5] indicate that the threshold for direct transitions in Si is above the energy of two 1.06-μm photons. Thus the two-photon absorption processes must be phonon assisted. Such indirect two-photon transitions are allowed energetically over a substantial volume of the Brillouin zone.

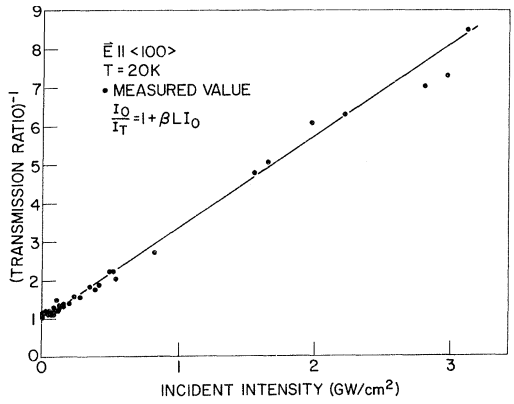

FIG. 2. Dependence of the inverse of the normalized optical transmission on incident intensity, at about 15 K. The solid line is a fit to the two-photon curve with $\beta = 1.9$ cm/GW.

These data were also analyzed with a least-squares analysis of the solution of the equation $dI/dX = -\beta I^2 - \gamma I^3$ to account for a possible three-photon process. This analysis yielded a value of β which was essentially identical to that obtained with the simpler two-photon equation and a value of γ which was zero within experimental accuracy. Linear absorption was not included in the above analyses since it is negligible in Si at 1.06 μm below about 25°K.

Similar data were obtained at about 100°K. The fit to a combined one- and two-photon absorption curve was quite good, with a value of $\beta = 1.5$ cm/GW. These results indicate that the major source of nonlinear absorption of these pulses arises from the two-photon process.

In order to distinguish between simultaneous and stepwise process, we can compare the strength of the effects at 100 and 20°K. The stepwise processes must follow the temperature dependence of the linear optical absorption as has been observed in other studies.[3] Here, however, the strength of the effect is comparable at the two temperatures although the linear absorption coefficient α varies by at least an order of magnitude, indicating that a simultaneous two-photon absorption process is involved.

This conclusion can be confirmed by making time-resolved measurements of the nonlinear absorption as shown in Fig. 3. Here the relative transmission of the weak probing beam is plotted as a function of its time of arrival relative to the main beam. The curve has three distinct regions.

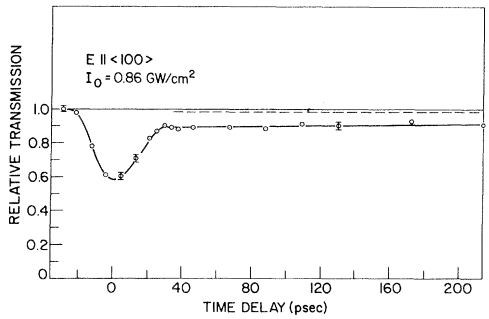

FIG. 3. Time dependence of the transmission of a weak probing pulse at 100 K. The dashed line indicates the value measured after approximately 10 nsec.

For arrival times well before the main pulse, the probe beam experiences linear transmission. The major absorption occurs near $t = 0$ and has an almost symmetric distribution about zero time delay. Since the lifetime of the carriers at 100°K is expected to be considerably longer than the laser pulse duration, this indicates that the major absorption process is an instantaneous one. For times greater than about 30 psec, the absorption does not return to its equilibrium value immediately, but recovers gradually during the time between laser pulses. The value of the absorption coefficient for times about 50 psec after the main pulse at 100°K is consistent with free-carrier absorption due to both electrons and holes if we assume that one pair is produced for each two photons absorbed, and that the cross section[6] per pair is about 5×10^{-18} cm^2.

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Screening-Enhanced Optical Absorption and the Search for M_3 Critical Points

John D. Dow*

Department of Physics and Materials Research Laboratory, University of Illinois at Urbana-Champaign, Urbana, Illinois 61801

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The optical absorption spectrum near an M_3 critical point of a model doped semiconductor or metal is calculated as a function of screening wave vector, using a realistic effective-mass model and a statically screened final-state interaction, but neglecting exchange. Wavelength-modulation line shapes are derived by direct (analytic) differentiation, and it is shown that screening *enhances* the absorption spectrum and its derivatives near an M_3 threshold. Experiments to detect M_3 critical points should concentrate on materials with heavy band masses near the conduction-band minimum and light (negative) masses at the band maximum.

Since one-electron, energy-band theory forms the foundation for our understanding of the electronic states of solids and their optical properties, it is astounding that no optical data unambiguously support one of the theory's principal predictions: Each interband transition energy (as a function of wave vector $\vec{k}$) has a maximum (M_3 critical point) which should give rise to a characteristic square-root dependence of the interband density of states on energy[1] [see Fig. 1(a)]. Although strong evidence for M_0 critical points (interband minima) has been widespread,[2] and some experiments have suggested that M_1 saddle-points exist,[3] no M_3 critical points have been unambiguously identified[4] in all the careful studies of the optical properties of semiconductors. Even modulation-spectroscopic studies of well-understood materials such as Si, Ge, and GaAs have failed to turn up a single M_3 critical point.

In this Letter, we show that M_3 critical points, if they exist, can best be observed in heavily doped materials, and that the doping *enhances* the optical absorption near an M_3 interband threshold. The effects of doping are treated analytically and exactly, using a statically screened electron-hole interaction and an effective-mass exciton model.

The model assumes the applicability of Wannier-Mott-Elliott exciton theory[5] and takes the optical absorption[6] $\epsilon_2(\omega)$ to be proportional to the transition density of states times the probability