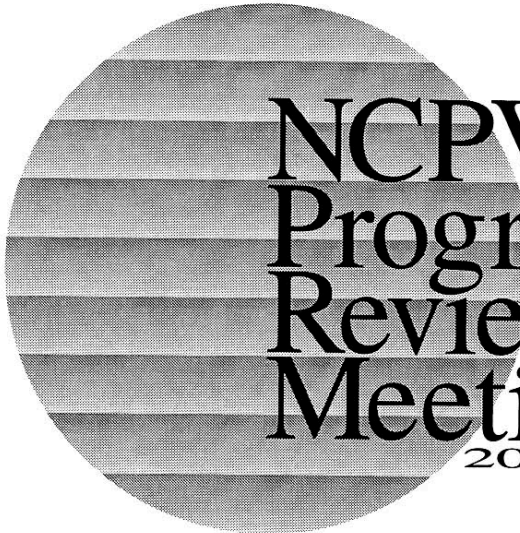


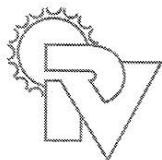
ADDENDUM TO THE PROCEEDINGS



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Ultrafast Dynamics of Photoexcited Carriers in HWCVD a-Si:H and a-SiGe:H

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ABSTRACT

We present femtosecond studies of photoexcited carrier dynamics in hydrogenated amorphous silicon and silicon-germanium alloys grown by the hot-wire assisted chemical vapor deposition (HWCVD) technique, which is promising for producing high-quality device-grade materials. We have used wavelength-resolved femtosecond pump-probe techniques to study the dynamics of photoexcitations in these materials. Femtosecond dynamics measurements have been carried out on thin film samples under experimental conditions with varying sensitivity to carriers in extended states or in band tail states. The relaxation dynamics of carriers associated with extended states show a strong dependence on excitation density and follow a bimolecular recombination law, consistent with a number of earlier studies on related amorphous materials. In contrast, measurements involving band tail states reveal significantly altered dynamics, characterized by a marked deviation from simple bimolecular recombination.

1. Introduction

Thin-film amorphous silicon and silicon-germanium alloys are promising materials for low-cost, high-efficiency solar cells, yet a range of issues relevant to their development and application as photovoltaic materials are not yet fully understood. Important unresolved issues include the detailed nature of the electronic states in these disordered semiconductors, and in particular, the relation of the band tail and extended states and their influence on the carrier dynamics. In this work, we investigate the dynamics of photoexcited carriers in a series of a-SiGe:H alloys. By varying the relative energy of the photoexcitation and the band gap, we selectively excite carriers into extended states or into progressively deeper band tail states, and we monitor the time evolution of the carrier distribution using femtosecond pump-probe techniques.

2. Experimental

Thin films of a-SiGe:H were grown on glass substrates by the hot-wire assisted chemical vapor deposition (HWCVD) technique [1]. For the experimental results presented in this paper, the Ge concentration of the films varied from 10 to 50 atomic percent. The optical band gap of the materials can be characterized in terms of the

transition energy at which the absorbance reaches a value of 104 cm^{-1} , and this value corresponds to energies of 1.76 eV, 1.60 eV, and 1.42 eV for samples of 10, 30, and 50 atomic percent Ge, respectively. The samples were grown with a nonuniform thickness so that an optimal thickness for the thin film etalon response could be chosen for the timeresolved optical measurements [2]. Approximate sample thicknesses varied from $\sim 350 \text{ nm}$ for the 50% Ge sample to $\sim 780 \text{ nm}$ for the 10% Ge sample, giving optical densities at the pump wavelength of < 0.25 .

Time-resolved measurements of the carrier dynamics were carried out using an optical pump-probe technique, in which a short pump pulse excites carriers in the sample and a time-delayed probe pulse measures the resulting change in the optical properties as a function of the pump-probe delay time. Measured changes in the transmission and reflectivity can be related to changes in the optical absorption coefficient and the index of refraction of thin film samples for comparison with theoretical models. In these initial measurements, the thin film samples were excited with pulses 35 fs in duration centered at a wavelength of 800 nm , corresponding to a transition energy of 1.55 eV. The pulses were generated by an amplified Ti:sapphire laser system operating at a repetition rate of 1 kHz. Probe pulses of variable wavelength were produced using a femtosecond white-light continuum generated by self-phase modulation in a sapphire plate, followed by prism dispersion compensation to give a temporal resolution typically $< 50 \text{ fs}$.

3. Results

Measurements of the time-resolved change in transmission following excitation at 1.55 eV for a series of a-SiGe:H alloys are displayed in Figures 1-3 [3]. The data are presented as the negative of the differential transmittance, corresponding to an induced absorbance that results from the photoexcited carrier population. In all cases, the carrier response was probed at a detection wavelength of 900 nm.

The carrier response shows a nonexponential time dependence that is strongly dependent on excitation density. When carriers are initially excited into the extended states well above the band gap, as is the case for 1.55 eV excitation of the 50% Ge sample shown in Fig. 1, the relaxation dynamics can be well described by a

bimolecular recombination law, consistent with previous time-resolved measurements on similar systems at comparable excitation densities. Fits to the solution of a bimolecular rate equation $dn/dt = -kn^2$ are shown superimposed on the data traces, and give values for the bimolecular recombination constant $k \sim 5 \times 10^{-9} \text{ cm}^3/\text{s}$, consistent with previous work on a-Si:H in which carriers were excited into extended states at comparable energies relative to the band gap [4].

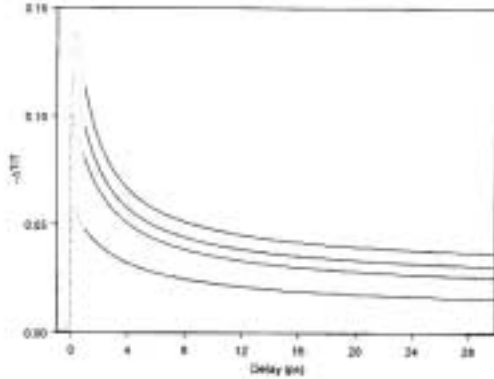


Fig. 1. Time-resolved negative differential transmittance of an a-Si_xGe_{1-x}:H thin film with $x = 0.50$. The data traces correspond to initial carrier densities of approximately $3 \times 10^{20} \text{ cm}^{-3}$, $2.5 \times 10^{20} \text{ cm}^{-3}$, $1.8 \times 10^{20} \text{ cm}^{-3}$, and $9.2 \times 10^{19} \text{ cm}^{-3}$.

In contrast, the time-resolved response for carrier excitation at lower energies relative to the optical gap exhibits a marked deviation from simple bimolecular recombination at short times. Representative responses for carriers excited progressively farther into the band tail states are presented in Figs. 2 and 3. For pump-probe delay times longer than ~ 1 ps, the response is well characterized bimolecular recombination, as shown by the fits to the data traces. At early times, a significant delay is observed in the peak of the induced absorbance that becomes more pronounced for initial excitation farther into the band tail. (The sharp feature at $t = 0$ includes contributions from nonlinear effects due to the temporal overlap of the pump and probe pulses and is neglected in the interpretation of the carrier dynamics.) The short-time response provides evidence for a rapid thermalization process on a subpicosecond time scale.

Acknowledgments

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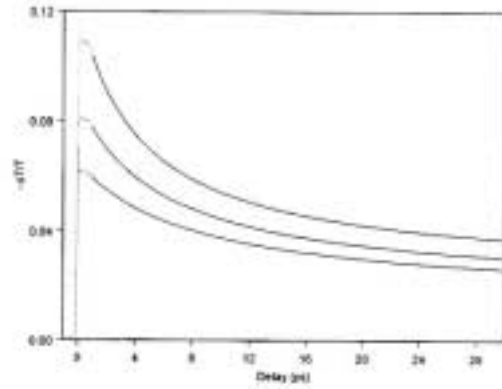


Fig. 2. Time-resolved negative differential transmittance of an a-Si_xGe_{1-x}:H thin film with $x = 0.70$ and initial carrier densities of approximately $5.5 \times 10^{19} \text{ cm}^{-3}$, $4.4 \times 10^{19} \text{ cm}^{-3}$, and $3.3 \times 10^{19} \text{ cm}^{-3}$.

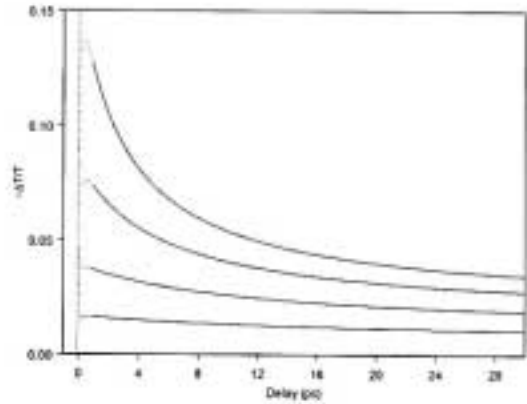


Fig. 3. Time-resolved negative differential transmittance of an a-Si_xGe_{1-x}:H thin film with $x = 0.90$ and initial carrier densities of approximately $6.6 \times 10^{19} \text{ cm}^{-3}$, $4.4 \times 10^{19} \text{ cm}^{-3}$, $2.6 \times 10^{19} \text{ cm}^{-3}$, and $1.3 \times 10^{19} \text{ cm}^{-3}$.

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