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Annual Report for contract N00014-86-K-0711
(1 June 1988 - 31 May 1989)

Ultrafast processes and spectroscopy with free electron lasers

prepared by

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1. Objectives

The goal of this project is twofold:

1) to demonstrate that the SCA free electron laser (FEL) ~~at~~ Stanford U. is a useful tool for time-resolved spectroscopy; and
2) to use the FEL with other ultrashort pulse sources to uncover the electronic processes in disordered semiconductors.

Potentially, the SCA FEL ~~at Stanford~~ U. is a near-ideal source for ultrashort time-resolved spectroscopy. It delivers widely tunable, psec pulses with high peak power and at high repetition rate. However, pulse-to-pulse variations in duration, wavelength and phase within one micropulse may be a limitation for high quality spectroscopy. In addition, pulses below one picosecond and tunability around the laser wavelength are desirable. ~~We plan to~~ carry out essential experiments that will demonstrate how useful the SCA FEL is in ultrashort time-resolved spectroscopy, including pulse compression and generation of white light continuum. The second goal of this project is to uncover the electronic processes in disordered materials. Understanding these processes is the key to understanding the physics in materials such as amorphous semiconductors or liquid semiconductors. In turn, understanding the physics of these materials will lead to novel devices or improved device operation and will impact on our models for other materials. We are carrying out picosecond and femtosecond time-resolved measurements on a-Si:H and its alloys with Ge and C. We focus on the electronic properties of carriers in the vicinity of the mobility edge.

2. Facilities/Instrumentation

We have established one laboratory at the SCA FEL facility at Stanford U. The FEL is operated by the group led by Professor Schwettman, to which we

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bring our expertise in ultrashort pulses. The beam is delivered to our optical table for experiments. Although we are still in the process of expanding our laboratory, we are already equipped with a detection system that has allowed us to record our first results on the FEL. We are equipped to perform time-resolved transmission and reflectivity measurements in the near infrared region.

In addition, we have at Princeton U. a femtosecond facility which provides tunable $\sim$ 100 fsec pulses at 8 kHz. This laser source has been used to study a-Si:H and its alloys while we are developing our laboratory at Stanford U.

3. Results

For the first time, we are able to report results obtained with the SCA FEL. We have performed successfully a simple version of time-resolved spectroscopy, namely autocorrelation by second harmonic generation in a $LiNbO_3$ crystal. Figure 1 shows the result. The data were obtained by measuring the intensity of the frequency doubled light as a function of the relative time delay between the two pulses. In this particular experiment, the difference in frequency between the incident and measured light allows us to block the incident light with optical filters and make a background free measurement with improved signal to noise ratio. The measured autocorrelation shows that the pulse width is less than 3 psec. We then attempted to perform time-resolved reflectivity measurements (pump at 1.5 μm, probe at 0.75 μm) on an a-Si:H sample. This was not successfully accomplished within the time constraints, but as explained in section "Future Plans," the technical difficulties have been solved.

We have made strong advances in our understanding of carriers close to the mobility edge of amorphous semiconductors using our femtosecond laser system. We have found that carriers recombine nonradiatively in less than 10 psec if N $\geq 5 \times 10^{18} cm^{-3}$. Each pair loses 2 eV worth of energy in that time. Since the injecting photon energy is 2 eV, all the energy is lost to phonons. The temperature of the sample is monitored by the thermal variations of the index of refraction n and of the absorption coefficient α (above the bandgap only), which dominate the optical constants when recombination has taken place (see Figure 2). The microscopic nature of this recombination is presently under investigation. Before the carriers have time to recombine, we can also investigate the properties of carriers in the extended states. The free carrier absorption is fitted well with a Drude model in which the relaxation time $\tau \simeq 0.5$ fsec. This indicates that the mean free path between collisions is of the order of the interatomic distance and therefore the mobility is low ($< 10\ cm^2/V$ sec). This point is under intense investigation presently since our measurements represent the first determination of the extended state mobility that is independent of any knowledge of the density of states close to the mobility edge.

4. Future Plans

Regarding the SCA FEL laboratory, we plan the specific experiments and improvements;

a) improvement of our detection system:

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We will use an acoustooptic stabilizer to control fluctuations in light intensity during the macropulse. Experiments will be performed using high frequency modulation to avoid 1/f noise from the laser and the environment. In addition, we will use a reference signal to employ differential detection. This gives a pseudo background free signal from the experiment. Finally, data acquisition is synchronized with the repetition rate of the macropulse to eliminate "jitter" in the data.

b) study of pulse distortion and spreading in infrared fibers, pulse compression, and generation of a white light continuum. Pulses will be chirped in fibers and recompressed using prisms. They will then be focussed in a medium where we will broaden their spectrum and generate a white light continuum. The goal is to achieve sub-psec pulses.

c) pump and probe ultrafast spectroscopic measurements will be performed on c-Si and a-Si:H. The first experiment will reproduce the results obtained at Princeton; then we will tune the pump energy to inject carriers well above the mobility edge (to study hot carrier relaxation) and just below the mobility edge (to study the mobility of carriers in weakly localized states).

Regarding the Princeton facility, we plan to continue our on-going experiments. The extended states mobility will be measured in a-Si:H and its alloys with Ge and C; the nature of the psec non-radiative recombination will be established; and experiments on superlattices and nanocrystal will be initiated. Some relatively minor improvements on our present facility will be required.

5. Personnel

Graduate students and postdoctorals supported under this research grant for the year ending May 31, 1989;

Ian H. Campbell
Ting Gong
Yanming Liu
William L. Nighan, Jr.
Jason Yee

(All the above are graduate students)

6. Publications

1. "Hot carrier dynamics in amorphous semiconductors," P.M. Fauchet, K. Gzara, and I.H. Campbell in SPIE vol. 942, 92-98 (1988).
2. "Determination of carrier-carrier and carrier-phonon relaxation times from ultrafast photoinduced absorption in amorphous semiconductors," P.M. Fauchet and K. Gzara, Phys. Stat. Sol. (b) **148**, K71-K75 (1988).

3. "Free-carrier and temperature effects in amorphous silicon thin films," C. Tanguy, D. Hulin, A. Mourchid, P.M. Fauchet and S. Wagner, Appl. Phys. Lett. **53**, 880-882 (1988).

4. "Self-diffraction: A new method for characterization of ultrashort laser pulses," W.L. Nighan, Jr., T. Gong, L. Liou and P.M. Fauchet, Optics Commun. **69**, 339-344 (1989).

5. "Ultrafast carrier relaxation in hydrogenated amorphous silicon," P.M. Fauchet and D. Hulin, Opt. Soc. of Am. **B**, 1024-1029 (1989).

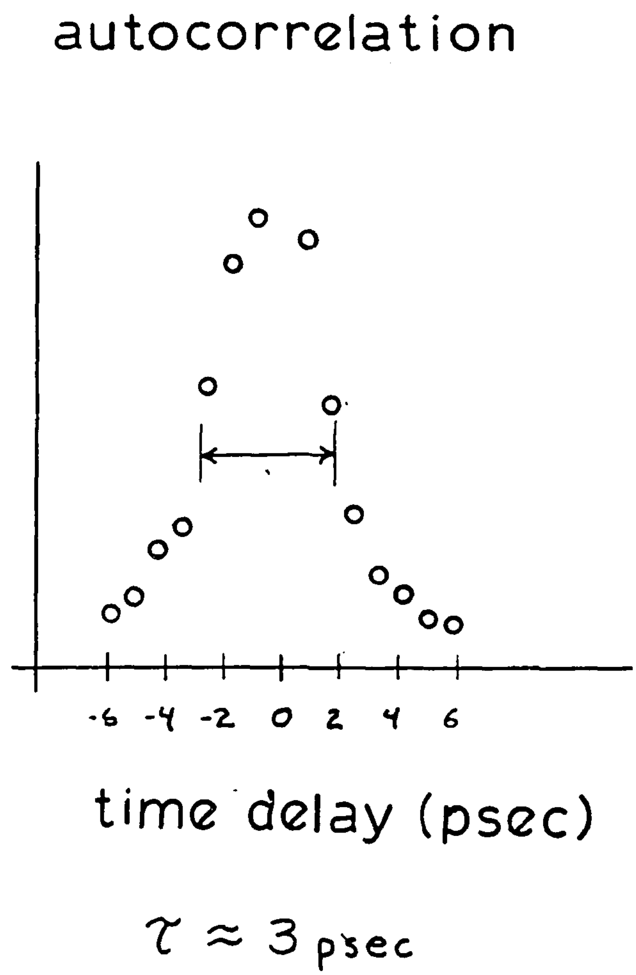

Figure 1

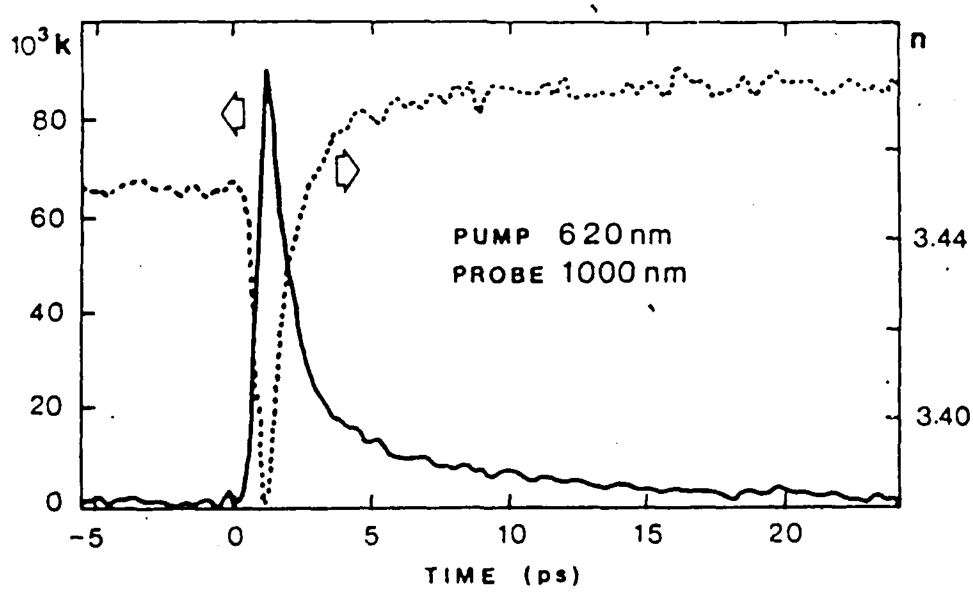

Figure 2

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18. SUBJECT TERMS (Continue on reverse if necessary and identify by block number)
Free electron laser; time-resolved (picosecond and femtosecond) spectroscopy; amorphous semiconductors; laser-induced phase transitions.

19. ABSTRACT (Continue on reverse if necessary and identify by block number)

Femtosecond laser spectroscopy has been used to study carrier relaxation times in amorphous silicon. We find a relaxation time of 1 picosecond above the mobility edge and a relaxation time of 10 picoseconds in the bandtail states, after which temperature effects dominate the optical properties. Theoretical modeling of femtosecond spectroscopic measurements has also helped define what is measurable and what is not.

Picosecond time-resolved reflectivity measurements have been performed during laser-induced phase transitions. The dielectric function of molten Si has been measured and superheating in the liquid phase has been observed at least to 10 picoseconds.

Work continues in both areas. We expect to expand the experimental program to other wavelengths thanks to the free electron laser.

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