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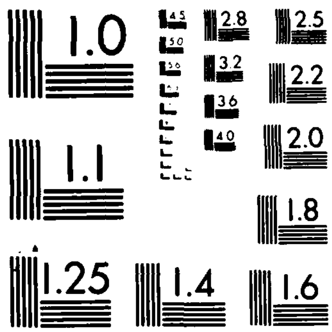


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| Semiconductors | Phonon | |
| Silicon (Si) | Optical absorption | |

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

This project explored the uses of pulsed laser Raman scattering as a technique for studying vibrational or phonon temperatures in Si with nanosecond time resolution. Raman scattering is unique in its ability to probe in a highly selective way populations of specific phonon modes. Knowledge of how phonon populations evolve in time is of fundamental importance in understanding how semiconductors respond to intense laser pulses such as in pulsed laser annealing of semiconductor surfaces. This information is crucial for

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understanding the dynamics of the rapid laser-induced melting.

Raman scattering was performed with a heat/probe sequence derived from a frequency doubled Nd:YAG laser and from YAG-laser-pumped or $N_2$-laser-pumped dye lasers. Initial results showed low phonon populations, consistent with only a 300-400 K temperature rise after a 10 nsec heat pulse with energy sufficient to melt the Si surface. Final results, however, indicate a population rise equivalent to a 1500±300 K phonon temperature. This is consistent with the normal 1685 K melting temperature of Si. These results were confirmed in second order Raman scattering with 150 nsec pulses from a 10 kHz frequency-doubled Nd:YAG laser. Thus we measure melting-temperature phonon populations in several phonon branches.

Additional experiments were done to measure the optical absorption in transiently heated silicon-on-sapphire. We found absorption consistent with that of molten Si from 0.8 eV to 3.1 eV.

A strong effort was placed on understanding the temperature-dependence of resonance Raman effects in Si. Our cw Raman study of oven-heated Si uncovered a strong decrease in Raman efficiency at high temperatures. Therefore the final pulsed Raman experiments were slightly redesigned to accommodate this drop in scattering efficiency. A complete analysis of the Raman line position and shape indicates significant modification of the Raman line due to phonon anharmonicity and to the influence of photoexcited free carriers with a density of $\sim 2 \times 10^{19}$ cm$^{-3}$.

FINAL REPORT

ONR Contract # N00014-80-C-0419

"Raman Studies of Surface Temperature in

Laser-Heated Semiconductors"

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This project systematically analyzed the applicability of pulsed Raman scattering as a measure of phonon populations (lattice vibrational temperatures) in pulsed-laser-heated silicon. The results and conclusions of this work are detailed in the publications listed as references 1-21. Additional important supporting work was accomplished while the principal investigator, Alvin Compaan, was on sabbatical leave at the Max Planck Institute für Festköperforschung in Stuttgart. Published results of this work appear as references 22-25. The principal findings and conclusions of this work are summarized below. Detailed analyses may be found in the cited references.

## A. DEVELOPMENT OF EXPERIMENTAL TECHNIQUES

The measurement of Raman scattering under the high power excitation conditions of pulsed laser annealing required the development of new experimental techniques. Throughout we have used a high intensity heat pulse/weak probe pulse configuration. The original system used two dye lasers operating at different wavelengths and pumped by a single pulsed $N_2$ laser. Significant improvements were made by adding a frequency-doubled Nd:YAG laser and then a YAG-laser-pumped dye laser. Further improvements were obtained by implementing an optical multichannel detector (OMA) vidicon to obtain parallel data acquisition. Raman signals are now regularly obtained with 100 $\mu$ Watts of average power or less (see references 1,2,16&19). We believe this heat/probe technique for time-resolved Raman scattering is quite generally useful and powerful. Its temporal resolution is limited only by the laser pulse durations (10 nsec in the work reported here).

## B. RAMAN STOKES/ANTI-STOKES RATIOS

Initial pulsed Raman results were obtained with two $N_2$ laser-pumped dye lasers and were restricted to spot sizes of $\sim$100$\mu$m because of the limited $N_2$ laser power. Nevertheless results were obtained on both single crystal Si[1,4] and ion-implantation-amorphized Si.[2] Improved signals were obtained as the more powerful Nd:YAG laser[7,14,16] and finally the YAG-laser-pumped dye laser were installed.[19-21] This permitted larger spot sizes on the sample and a smaller ratio of probe diameter to heat spot diameter. All the early Raman results were obtained with very low probe energy densities (typically 0.06 J/cm$^2$) to keep the probe as non-intrusive as possible. The results consistently showed Brillouin Zone center optic phonon populations characteristic of 300-400$^{o}$C.

The latest Raman results, however, were obtained with much higher probe energy densities.[19-21] This change was dictated by the discovery that the Raman strength drops rapidly at high temperatures in Si. (See ~~the next section~~ Section D. and reference 22.) Thus the higher energy probe pulse holds the sample temperature high for most of the duration of the Raman data acquisition (equal to the 10 nsec probe pulse duration). This change in experimental design is the principal factor accounting for the fact that the final results indicate higher phonon populations (characteristic of lattice temperatures of 1100±300 C). This is within experimental error of the melting point of Si (1412$^{o}$C).

Early measurements were performed at a probe wavelength of 405 nm, the later results utilized both 405 nm and 532 nm probes with consistent results.

## C. TRANSIENT TRANSMISSION AND REFLECTIVITY

Reliable interpretation of Raman Stokes/anti-Stokes ratios requires knowledge of the optical constants of Si not just at room temperature but also under conditions of intense laser heating. Therefore we pursued complementary measurements of transient transmission and reflectivity.

The transient reflectivity rise is a widely used indicator of the annealing phase and we used either a HeNe or Ar laser throughout all of the Raman and other measurements as a check on experimental conditions (e.g. power density, beam overlap, etc.).

Additionally we used a 1.15μm HeNe laser,[3,4] a 1.58μm Raman shifted pulse,[15] and the tunability of the dye laser[5,6,7,11] to probe the absorptivity of the transient, high reflectivity, "molten" phase over the photon energy range 0.78 eV to 3.1 eV. The experiments used silicon-on-sapphire samples both with crystalline Si and ion-implantation amorphized Si.[13] In the latter case the recrystallization and cooling was followed over a broad time scale from 10 ns to 10 μsec. The transmission during the high reflectivity phase was shown to be less than $10^{-5}$ of the value of crystalline Si at $\lambda$=514.5 nm.

For these measurements during the high reflectivity phase, the spectral dependence of the absorption coefficient and its magnitude were shown to agree well with the values measured for molten Si.

## D. COMPLEMENTARY MEASUREMENTS MADE DURING SABBATICAL LEAVE

From July 1982 to June 1983, I was on sabbatical leave at the Max Planck Institute für Festkörperforschung-Stuttgart with the group of Prof. Manuel Cardona. During this period I completed a thorough study, with H. J. Trodahl, of the high temperature behavior of resonance Raman scattering in Si.[22] This work significantly strengthened our understanding of and confidence in the temperature dependence of all the factors which control the Raman intensity.

One consequence of the work was the discovery that the Raman strength decreases at high temperatures for all probe wavelengths. Consequently the latest pulsed Raman experiments were redesigned to avoid as much as possible this problem and the analysis fully accounted for the drop in Raman strength (see section B above).

While on sabbatical, I was also strongly involved in Raman studies of ultra-heavily doped Si prepared with ion implantation and pulsed excimer laser annealing.[23-25] This work layed a foundation for understanding carrier-phonon interactions in highly photoexcited Si. These results were also included in our final analyses of the pulsed Raman results.[21] The carriers give extra broadening and asymmetry to the Raman line.

Finally, a pulsed Raman experiment was begun on sabbatical and completed in January 1984 on a return trip to Stuttgart. This was a study of second-order Raman scattering with 150 nsec pulses from a 10 kHz, repetitively Q-switched, frequency-doubled Nd:YAG laser.[19-21] These data gave transient phonon populations for several other phonon branches, acoustical and optical. Phonon populations were found to be characteristic of T=1200±300 C for all phonon modes studied.

## E. ANALYSIS OF RESULTS, SOURCES OF ERROR, POTENTIAL OF THE RAMAN TECHNIQUE

The Raman results have stimulated much discussion of potential sources of difficulty in interpretation. This has prompted thorough analysis of laser pulse-to-pulse fluctuations,[14,17] beam homogeneity,[9] timing jitter,[14,17,18,22] electron densities,[8] possible phonon hot spots[10] and laser-induced strain effects.[21]

We have found that these factors either have minor influence on the

Raman results or have been controlled adequately. However the strong reduction of Raman strength at high temperatures[22] was unknown at the beginning of these studies. We now know that it accounts for much of the difficulty in the early experiments.

We find that when this drop in Raman strength is properly accounted for (applying the results of ref. 22) then the Raman data give results consistently showing phonon populations appropriate for T=1100-1200 C with an uncertainty of $\pm 300^{0}$C. This consistency applies to Stokes/anti-Stokes ratios, Raman line shift and line width of the first order Raman scattering, and also to second-order ratios.[19-21]

We believe the understanding of the Raman effect and its temperature- and carrier-density-dependence in Si is now easily well enough understood to permit its use as a reliable probe of phonon populations on a picosecond as well as nanosecond time scale. However the drop in Raman strength at high temperatures seriously reduces its potential as a convenient and easily applied technique. Nevertheless Raman scattering remains a technique capable of ultrafast time resolution (limited only by laser pulse durations). Also Raman scattering is unique in its ability to provide not just an average lattice temperature but a direct measure of phonon populations at well defined points in the individual phonon branches. Consequently it is an important tool for studies of phonon thermalization in solids and in general the response of solids to high power laser pulses.

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