PICOSECOND DYNAMICS OF PHOTOEXCITATIONS IN AMORPHOUS MULTILAYER STRUCTURES

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ABSTRACT

We report on measurements of ultrafast relaxation processes in transmission and reflection in amorphous multilayer structures consisting of a-Si:H, a–SiN_x:H, a–SiO_x:H, and a-Ge:H. The decays recorded in transmission in the a–Si:H/a–SiN_x:H and a–Si:H/a–SiO_x:H multilayers depend strongly on the silicon sublayer thickness and are interpreted in terms of carrier transport to and trapping at interfacial defects. In the a-Si:H/a-Ge:H multilayers we observe oscillations in reflectivity due to standing acoustic waves with a frequency that depends on the repeat distance of the multilayer.

INTRODUCTION

Multilayer structures made from amorphous hydrogenated silicon (a-Si:H) and related amorphous semiconductors have provided a new tool to study the electronic and acoustical properties of these materials. A variety of effects like charge transfer doping [1], interfacial defects [1], built-in electric fields [2,3], an increase in disorder [4], folding of acoustic phonons [5], and quantum size effects [6,7] have been observed. We have studied ultrafast dynamics of photoexcitations in amorphous multilayer structures and obtained additional information about some of these effects and some fundamental properties of amorphous semiconductors. We have extended our measurements [8] of photoinduced changes in transmission (ΔT) in amorphous multilayer structures consisting of alternating layers of a-Si:H and a–SiN_x:H to multilayers containing a-Si:H and a–SiO_x:H. We also report on photoinduced changes in reflectivity (ΔR) in a-Si:H/a-Ge:H multilayers.

EXPERIMENT

Using the pump and probe technique we recorded ΔT and ΔR on a time scale between 0.1 psec and 1.8 nsec. The light source was a colliding-pulse modelocked ring dye laser [9] with a pulse length of 0.1 psec, an energy per pulse of 0.2 nJ, a photon energy of 2 eV, and a repetition rate of 108 MHz. The pump was modulated at 4 MHz and the illuminated spot had a diameter of 10 μm. The preparation method of the multilayers is given elsewhere [10,11]. The total thickness of the multilayers ranged between 0.5 and 1 μm. Because the absorption length of a-S:H at 2 eV is of the order of a few thousand Ångstroms (a–SiN_x:H and a–SiO_x:H are transparent at 2 eV) we measured ΔT in the a–Si:H/a–SiN_x:H and a–Si:H/a–$Si:O_x$:H multilayers. On the other hand due to the much shorter absorption length of a-Ge:H at 2 eV (about 300 Å) we could not measure ΔT in the a-Si:H/a-Ge:H multilayers, but recorded ΔR instead.

ELECTRONIC PROCESSES IN a–Si:H/a–SiN_x:H AND a–Si:H/a–SiO_x:H MULTILAYERS

In thin films of a-Si:H the change in transmission is proportional to the change in the absorption coefficient ($\Delta\alpha$) [12]. In a low defect sample of a-Si:H the response is an abrupt

Mat. Res. Soc. Symp. Proc. Vol. 77. 1987 Materials Research Society

increase in absorption ($\Delta T < 0$) immediately after zero time delay followed by a non-exponential decay with a time constant of about 1 nsec [8]. In Fig. 1 we show ΔT for a–Si:H/a–SiN_x:H multilayers with silicon sublayer thicknesses d_S of 204 Å, 78 Å and 25 Å at room temperature. The thickness of the silicon nitride layer was 1.2 d_S. We have also measured samples with d_S = 125 Å and 39 Å. The decay constant τ defined through $\Delta T(\tau) = \Delta T(0)/2$ is plotted as a function of d_S in Fig. 2. τ decreases from a value of 160 psec for d_S = 204 Å to 10 psec for d_S = 78 Å and then increases again to 160 psec for d_S = 25 Å. The minimum of τ occurs between 78 Å and 39 Å. The time constant is the same for d_S = 204 Å and 25 Å, but Fig. 1 shows that the decays in these two samples are very different. The decay in the multilayer with d_S = 25 Å is much faster for short times than the one for d_S = 204 Å, but at longer times (t > 400 psec) the decay for d_S = 25 Å becomes much slower. The same behavior is observed when one compares the decays of the multilayers with d_S = 39 Å and 78 Å. This indicates that the decay kinetics for small d_S is different from the one in samples with large d_S.

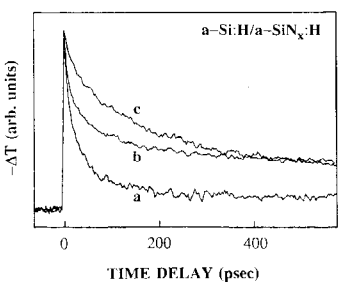

Fig. 1. Picosecond decays of ΔT in a–Si:H/a–SiN_x:H multilayers with different silicon sublayer thickness d_S (a: d_S = 78 Å, b: d_S = 25 Å, c: d_S = 204 Å) at 300 K.

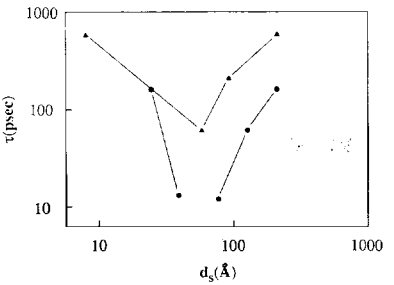

Fig. 2. The decay constant τ at 300 K as a function of silicon sublayer thickness d_S for a–Si:H/a–SiN_x:H (●) and a–Si:H/a–SiO_x:H (▲) multilayers.

In Fig. 3 we plot ΔT as a function of time delay on logarithmic scales for d_S = 25 Å at different temperatures. The decays exhibit a power law t^{-m} (m < 1) over about two decades in time and the exponent m increases linearly with temperature. The multilayers with $d_S \geq 78$ Å do not exhibit a simple power law dependence over two decades in time in our time range and the exponents obtained for these samples from the asymptotic behavior at long times differ from the ones in thin multilayers ($d_S \leq 39$ Å). This supports the observation mentioned above that the decay kinetics is different in the thinner multilayers.

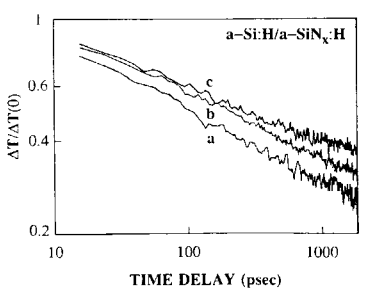

Fig. 3. Picosecond decays of ΔT in a–Si:H/a–SiN_x:H multilayer with d_S = 25 Å at different temperatures (a: T = 300 K, b: T = 240 K, c: T = 180 K).

The picosecond decay of $\Delta\alpha$ in a-Si:H has been attributed to carriers trapped in states with a lower absorption cross section [12]. In the multilayers these deeper traps are probably at the interfaces. For $d_S \geq 78$ Å it will take the photoexcited carriers some time before they arrive at the interfacial defects. The transport of these carriers can be by diffusion or drift due to built-in electric fields [2,3]. Since τ increases monotonically with d_S both processes (diffusion and drift) can explain the dependence of τ on d_S. The decay in the thinner multilayers has a different origin. Since for the thin multilayers d_S is of the order of the interface width, diffusion or drift do not play a significant role anymore. The different exponent of the power law decay in the thin multilayers is probably due to a change in the structure of the a-Si:H layer. Raman scattering experiments [4] have shown an increase in disorder. This broadens the band tail and slows down the deep trapping process. The broadening of the band tail has been also observed in optical absorption [6], photoluminescence [6], and photomodulation spectroscopy [8].

We have also measured ΔT in a–Si:H/a–SiO_x:H multilayers with d_S between 8 Å and 220 Å with different oxide layer thicknesses d_0. The dependence of τ on d_S is shown in Fig. 2 and is qualitatively the same as in the a–Si:H/a–SiN_x:H multilayers. The values of τ in the oxide multilayers are considerably larger (between 60 and 600 psec) than in the nitride multilayers. A possible explanation is a smaller density of interfacial defects for the oxide multilayers. One has also to take into account that the built-in electric fields result from different charge distributions in these two sets of multilayers [3].

ACOUSTIC EFFECTS IN a-Si:H/a-Ge:H MULTILAYERS

In the oxide and nitride multilayers we studied the silicon sublayer and the interfaces. In the a-Si:H/a-Ge:H multilayers carriers are excited in both layers, but due to the high absorption coefficient of germanium the density of excited carriers is much higher in the a-Ge:H layer. In thin films of a-Si:H we have observed a monotonic decay of ΔR [13], whereas ΔR in a-Ge:H shows an oscillatory response [14] superimposed on a decay. This additional response has been attributed to a travelling acoustic wave in the Ge film. The generation and detection process is described in detail in [14].

In Fig. 4 we show a typical result of a measurement of ΔR on a multilayer structure consisting of a-Si:H and a-Ge:H, when the light is incident on the free surface. The repeat distance $d = d_S + d_G$ was about 1100 Å and the ratio of the relative widths of the individual layers was $d_S/d_G = 1.13$. A smoothly decaying background to the response has been sub-

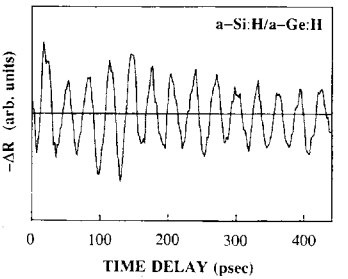

Fig. 4. Oscillatory component of ΔR in a-SiH/a-Ge:H multilayer with a repeat distance d = 1100 Å at 300 K.

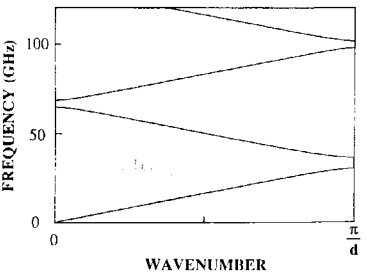

Fig. 5. Calculated dispersion of longitudinal acoustic phonons in a-Si:H/a-Ge:H multilayer. The parameters are given in the text.

tracted out, and the remaining signal shows oscillations with a frequency of 33 GHz. It can be seen from Fig. 4 that the amplitude varies from cycle to cycle, and in addition there is some fine structure which is reproducible from run to run. As the repeat distance becomes smaller the frequency of these oscillations increases. For d = 470 Å the frequency is 92 GHz and for d = 280 Å it is 127 GHz. The observed oscillations are interpreted as standing acoustic waves. We have never observed acoustic waves in experiments with a-Si:H, and so we conclude that the oscillations of the superlattice are detected via the changes they cause in the optical properties of the a-Ge:H layers.

Rytov [15] has calculated the dispersion relation for a multilayer and the result is shown in Fig. 5. We have used d_G/v_G as an adjustable parameter and set $v_S/v_G = 1.64$ (crystalline value), $\rho_S/\rho_G = 0.44$ (crystalline value) and $d_S/d_G = 1.13$. Computer simulations of the strain distribution in this system and its contribution to the reflectivity also show persistent oscillations and demonstrate that the major contribution to these oscillations comes from zone-boundary phonons of the lowest two branches of the dispersion relation (Fig. 5). The frequency of the oscillations is roughly equal to the average of the frequencies of the two branches at the zone-boundary. We are extending these simulations to include in detail the way a spatially-varying strain field in the superlattice changes the reflectivity. We expect that this will enable us to understand the fine structure of the oscillatory part of ΔR.

The frequency of the observed phonons can be tuned by changing the repeat distance d. The attenuation of acoustic phonons in glasses above 100 GHz as a function of frequency is an interesting problem in ultrasonics. We hope to use our experimental method to study the attenuation of these high frequency acoustic phonons in real time.

ACKNOWLEDGEMENT

The authors would like to thank T. R. Kirst for technical assistance. The work was supported in part by the National Science Foundation grant DMR 82-09148 and the NSF Materials Research Laboratory Program at Brown Univeristy.

REFERENCES

1. T. Tiedje and B. Abeles, Appl. Phys. Lett. 45, 179 (1984).
2. C. B. Roxlo, B. Abeles, and T. Tiedje, Phys. Rev. Lett. 52, 1994 (1984).
3. C. B. Roxlo and B. Abeles, Phys. Rev. B34, 2522 (1986).
4. N. Maley and J. S. Lanin, Phys. Rev. B31, 5577 (1985).
5. P. Santos, M. Hundhausen, and L. Ley, Phys. Rev. B33, 1516 (1986).
6. B. Abeles and T. Tiedje, Phys. Rev. Lett. 51, 2003 (1983).
7. N. Ibaraki and H. Fritzsche, Phys. Rev. B30, 5791 (1984).
8. H. T. Grahn, H. A. Stoddart, T. Zhou, Z. Vardeny, and J. Tauc, Proc. 18th Int. Conf. on the Physics of Semiconductors, Stockholm, 1986, in press.
9. R. L. Fork, B. I. Greene, and C. V. Shank, Appl. Phys. Lett. 38, 671 (1981).
10. B. Abeles, L. Yang, P. D. Persans, H. S. Stasiewski, and W. Lanford, Appl. Phys. Lett. 48, 168 (1986).
11. P. Persans, B. Abeles, J. Scanlon, and H. Stasiewski, Proc. 17th Int. Conf. on the Physics of Semiconductors, edited by J. D. Chadi and W. A. Harrison (Springer Verlag, New York, 1985), p. 499.
12. J. Tauc, in Tetrahedrally-Bonded Amorphous Semiconductors, edited by D. Adler and H. Fritzsche (Plenum Press, New York, 1985), p. 345.

13. H. T. Grahn, C. Thomsen, and J. Tauc, Opt. Com. 58, 226 (1986).
14. C. Thomsen, H. T. Grahn, H. J. Maris, and J. Tauc, Phys. Rev. B34, 4129 (1986).
15. S. M. Rytov, Akust. Zh. 2, 71 (1956) [Sov. Phys. Acous. 2, 67 (1956)].