Defect Creation and Two-Photon Absorption in Amorphous SiO_2

In a recent Letter,[1] Tsai, Griscom, and Friebele have reported results on E_1' (oxygen vacancy) defect creation in bulk, amorphous SiO_2 using 6.4-eV pulsed laser radiation. Their results give clear evidence for the role of two-photon absorption and suggest that excitonic mechanisms are involved in new defect creation rather than precursor transformation.

We have previously reported[2] dose-dependent E_1' defect creation studies in low OH Suprasil Wl using 4.8-eV photons from a pulsed excimer laser. Assuming two-photon processes relevant and a two-photon absorption coefficient,[3] β at 4.8 eV of 4.5×10^{-5} cm/MW, we show in Fig. 1 the E_1' defect creation as a function of the density of two-photon excitations. The value for β was measured[3] at 4.5 eV, but is expected to vary slowly with photon energy in this range.[5] For the 6.4-eV data,[1] we assume that the E_1' creation efficiency is independent of wavelength and, from the 4.8-eV data, that 5×10^{14} E_1'/cm^3 are created by 1.8×10^{18} two-photon excitations per cm^3. The 6.4-eV data from Ref. 1 is plotted in Fig. 1. To compare the laser results with defects created by γ rays, we first estimate the number of oxygen displacements produced by Compton electrons ($E_{th}\sim150$ keV). For 10 Mrad of ^{60}Co γ rays, if each γ produced a Compton electron of 1 MeV, then the number of displaced oxygens per cm^3 would be[6] $3\times10^{15}<n_0<6\times10^{15}$. Comparison of the number of displaced oxygens per implanted ion with the E_1' creation per implanted ion in ion-implantation experiments[7] indicates that for every 37 displacements only one "permanent" E_1' is created. For 10 Mrad of γ rays, the maximum number of E_1' defects is then $8\times10^{13}<n_{E'}<1.6\times10^{14}$ per cm^3, which is much less than we have observed. In Fig. 1 we thus plot the E_1' density we have observed as a function of density of electron-hole pairs, assuming a creation rate of (8×10^{12} pairs)/cm^3/rad.[8] We deduce a power-law fit to both the laser and γ-ray data of the form $n_{E'}=1.4\times10^4 n_p^{0.57}$ where n_p is either the density of electron-hole pairs or of two-photon excitations. For high OH silica, our γ-ray data and the 6.4-eV laser data[1] coincide but with a different power law for n_p (we have no 4.8-eV laser data).

We can approximately estimate the two-photon absorption coefficient at 6.4 eV noting that for the same number of incident pulses at 4.8 and 6.4 eV, incident intensities of 15 MW/cm^2 (Ref. 3) and 6 MW/cm^2 (Ref. 1) are required, respectively, to produce the same density of defects. We assume that the fraction of energy lost in two-photon processes is $E_2=(1-e^{-2\alpha z})I_0\beta/\alpha$, where α is the one-photon coefficient in cm^{-1}, z is the sample thickness in cm, and I_0 is the incident intensity; then with[4] $\alpha(4.8)=0.002$ and $\alpha(6.4)=0.017$, we deduce $\beta(6.4)/\beta(4.8)=2.5$ in excellent agreement with theoretical predictions[5] of ~3.

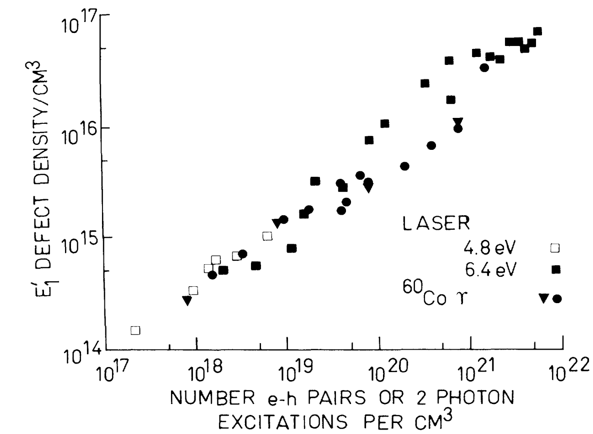

FIG. 1. E_1' defect creation in low OH silica by 4.8- (Ref. 3) and by 6.4-eV (Ref. 1) laser radiation and ^{60}Co γ rays as a function of density of electron-hole pairs or two-photon excitations. γ data from Ref. 4 (▼) and from the present authors (●). All irradiations were carried out at room temperature.

The results presented indicate a unique power law for E_1' creation whether by ionizing radiation or two-photon absorption, suggesting the same mechanism of defect creation independent of the radiation responsible for cross-band-gap excitation. Such defect studies may enable a crude estimate of two-photon absorption coefficients for short-wavelength laser radiation.

R. A. B. Devine
Centre National d'Etudes des Telecommunications
BP 98, 38243 Meylan, France

Received 18 August 1988
PACS numbers: 61.70.At, 61.80.Ba, 76.30.Mi

[1] T. E. Tsai, D. L. Griscom, and E. J. Friebele, Phys. Rev. Lett. **61**, 444 (1988).

[2] R. A. B. Devine, C. Fiori, and J. Robertson, Mater. Res. Soc. Symp. Proc. **61**, 177 (1986).

[3] P. Liu, W. L. Smith, H. Lotem, J. H. Bechtel, N. Bloembergen, and R. S. Adhav, Phys. Rev. B **17**, 4620 (1978).

[4] E. Dooryhee, Ph.D. thesis, University of Paris, 1987 (unpublished).

[5] H. S. Brandi and C. B. de Arauja, J. Phys. C **16**, 5929 (1983).

[6] O. S. Oen and D. K. Holmes, J. Appl. Phys. **30**, 1289 (1959).

[7] R. A. B. Devine, in *The Physics and Chemistry of SiO_2 and the Si-SiO_2 Interface,* edited by B. E. Deal (Plenum, New York, to be published).

[8] J. R. Srour, S. Othmer, O. L. Curtis, Jr., and K. Y. Chiu, IEEE Trans. Nucl. Sci. **23**, 1513 (1976).

