NONLINEAR TRANSMISSION AND COLOUR-CENTRE DYNAMICS IN GERMANOSILICATE FIBERS AT 420-540 nm

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ABSTRACT

We report evidence in support of the view that induced loss and non-linear transmission in pure germanosilicate fibers at blue/green wavelengths are governed by the formation (via two-photon absorption), spontaneous and stimulated transformation and bleaching (via single-photon events) of Ge(1), Ge(2) and Ge(3) colour-centres. Using a tunable pulsed dye laser, the excitation spectrum of the induced absorption, its spectral attenuation and the effects of germania concentration and thermal annealing are investigated.

In the blue/green region of the spectrum, optical fibers frequently exhibit peculiar transmission characteristics at even modest average power levels. For example, Brown and his associates[1] report reversible non-linear transmission (NLT) at 488 nm and 514.5 nm in single-mode high-birefringence (Hi-Bi) fibers at CW powers of a few hundred mW. This phenomenon has however not yet been thoroughly studied or explained, even though single-mode fibers capable of transmitting large amounts of power in this spectral range are of interest in the development of measurement systems for dynamic light scattering, photon correlation, quasi-elastic light scattering and laser doppler velocimetry[2]. Such fibers are also of great interest in interferometer systems. As has already been pointed out[1], NLT in fibers cannot be explained by recourse to more usual non-linear phenomena such as stimulated Brillouin or Raman scattering. The short lengths of fiber involved, the low intensities and the fact that the effect is independent of laser line-width all confirm this conclusion.

The formation, transformation, and bleaching of colour-centres (CC's) are known to play an important role in the optical behaviour of many doped glasses. These effects were probably behind the photosensitivity observed by Kawasaki and coworkers[3] in germanosilicate fibers at blue/green wavelengths, and play a role in anomalously efficient generation of second-harmonic light by optical fibers[4]. Two-photon absorption (TPA) is known to induce CCs in bulk borosilicate glasses at 532 nm[5] and in a variety of glasses at UV wavelengths[6]. This encouraged us to examine the possibility that dynamic photon-driven changes in CC population

might lie at the root of NLT in fibers[7]. To our knowledge this loss mechanism has not yet been identified in germanosilicate glasses at blue/green wavelengths. In its simplest form, our model proposes that, at a given constant photon flux n_p, TPA leads to the creation of CC's (at a rate proportional to n_p^2), which at the same time can be bleached out by single-photon absorption (proportional to n_p). A balance is reached between these two rates at an equilibrium population of CC's (and hence attenuation) that is linearly proportional to n_p. This simple model agrees with our experimental observation that bleaching occurs only in the presence of light. Two-stage absorption can be discounted because the absence of an intrinsic single-photon absorption peak indicates that no intermediate level exists. Although the TPA coefficient is very small in silicate glasses (of the order of 10^{-6} cm/MW at 532nm in borosilicate[5]), TPA is thought be the cause of significant NLT for two reasons. There is a high probability that a single TPA event will lead to the creation of a loss centre, and the very long interaction lengths offered by optical fibers mean that small changes in loss can yield large changes in transmission.

Here we report the main results of a detailed investigation of this phenomenon in germanosilicate HiBi fibers exposed to pulsed light in the range 420 to 540 nm. Pulsed light of low average power enabled us to induce reproducible permanent changes in loss by permitting the effects of TPA (proportional to peak power) to dominate over those of single-photon bleaching (proportional to average power).

The experimental arrangements were as follows. A range of HiBi fibers with "bow-tie" geometry was fabricated using the MCVD process. Their germania concentrations were estimated from their refractive index profiles, and their cut-off wavelengths designed to lie below 440 nm so as to allow predominantly single-mode operation in the range 420-540 nm. Optical pulses of 6 ns width were delivered from a frequency-tripled Nd-YAG laser pumping a dye-laser to give the required wavelength tuning range. The pulse repetition rate was 30 Hz at peak pulse powers launched into the fibers of 10-100 W ±10 %. These pulses caused a gradual change in transmission in the fibers that eventually reached a steady-state level that depended on the peak power launched. The kinetics of this process were investigated as follows. We exposed the fiber to a few hundred pulses, then blocked the laser and measured the new transmission level with a separate low-power white-light system. This sequence was repeated until either a required pulse count was reached or the loss induced in the fiber had attained a steady-state level. Transmitted pulse-to-pulse stability was monitored using a calibrated silicon detector, and any errors due to drift in launch conditions were reduced to negligible levels.

The results of a typical run are given in Figure 1. A 5m length of fiber with a GeO_2 concentration of 6.5 mol% was exposed to pulses of wavelength 470 nm in two 10 minute stages, first at 10 W/μm^2 and then at 1 W/μm^2 peak pulse intensity. The total induced loss is plotted as a function of time. For clarity, each stage is plotted above the same time axis. During the 10 W/μm^2 stage the loss rises to about 4.7 dB and almost reaches

saturation. However when the fiber is then exposed (after a short pause for re-adjustment, during which the laser is blocked and the loss rises slightly) to the lower power level, there is an initial rise in loss before the expected bleaching is seen.

The saturated absorption levels induced by exposure to various peak intensities are shown in Figure 2. The lower experimental points represent the loss immediately after blocking the laser light, and upper ones the loss measured one day later. Each pair of points was obtained from a fresh 5 m length of fiber (GeO_2 concentration 6.5 mol%).

To establish if these effects are Ge-related we fabricated fibers with a range of different Ge concentrations. Taking account of the different mode-spot sizes in each case, we launched peak intensities of 10 W/μm^2 at 470 nm into each fiber and allowed the transmission to stabilize. The induced attenuation at 460 nm scaled linearly with germania concentration at 170 dB/km per mol%.

ESR measurements on 900 strands of unexposed 40 μm core fiber showed a resonance typical of oxygen-deficient germanosilicates, confirming the existence of Ge(1), Ge(2) and Ge(3) colour-centres[8]. However, ESR was impractical as a means of quantifying the changes in CC population induced in our experiments because the signals were well below the detection limits of the ESR technique. This was especially true for single-mode fibers where, although NLT is more pronounced due to higher attainable in-core intensities, the actual volume of core material per fiber strand is extremely small.

For the present work the relevant precursors to the Ge(3) centres are the Ge-Si or Ge-Ge bonds that exist in oxygen-deficient germanosilicates[9]. They give rise to a characteristic absorption band centred at 240 nm[10]. It is known[11] that energetic photons of 240 nm can break these bonds, creating free electrons and Ge(3) centres. We postulate that this process is initiated by TPA. In the vicinity of the Ge centres the band-gap is locally narrower and the released electrons (see Figure 3) are free to move along strings of interconnected Ge defects which form "electron pathways" until they recombine with another Ge(3) defect or fall into Ge-related traps, creating Ge(1) and Ge(2) centres. Both thermal and stimulated single-photon bleaching of the Ge(1) and Ge(2) centres are also likely to occur. The full picture is thus of three different CC populations that trap and release charge via a mobile electron "cloud", at rates that are governed by two-photon and single-photon absorption, spontaneous recombination, and thermal bleaching.

This model is as yet tentative. We are at present fitting it analytically to the data, and receiving encouraging results; these will be presented elsewhere. However, as we shall now outline, it provides a plausible physical mechanism for the main features of the behaviour in Figure 1. During the 10 $W/\mu m^2$ stage, an increased population of free electrons appears, leading, at equilibrium, to an increased population of Ge(1) and Ge(2) centres and hence to increased loss. When the peak pulse intensity is reduced to 1 $W/\mu m^2$, the now excess free electron population will recombine to create more Ge(1) and Ge(2)

centres, leading to an initial surge in loss. On a longer time scale these new centres will slowly bleach out, the released charge eventually recombining with the Ge(3) centres until a new (lower) equilibrium loss is reached. The complexity of these processes means that the steady-state induced loss is unlikely to be a linear function of peak pulse power as suggested by the simple model. Axial variations in induced loss will also cause nonlinearities in this relationship, so that the nonlinear experimental plot in Figure 2 is perhaps not surprising. The slow increase in loss over 24 hours is probably the result of a very slow spontaneous transfer of charge between Ge(1), Ge(2) and Ge(3) centres.

To provide additional evidence that TPA plays a role in the phenomenon, we investigated the wavelength sensitivity of the absorption creation process. Pulses at wavelengths ranging from 420 to 540 nm were launched into a series of 5 m lengths of fiber (2.7 mol% GeO_2) for a period of 15 minutes each. Care was taken to launch the same core intensity of 10 W/μm^2 at each wavelength. The loss induced at 450 nm (arbitrarily chosen) is plotted against excitation wavelength in Figure 4. The greatest losses are induced at a wavelength 480 nm, indicating that TPA plays an important role in the phenomenon, since the Ge-Si absorption peak mentioned above appears at 240 nm in oxygen-deficient germanosilicates. The breakage of Ge-Si bonds may also be the mechanism that permits the direct holographic writing of gratings into fibers using light from a KrF excimer laser at 248 nm[12].

The absorption spectra of the intrinsic and induced loss for 5 m of fiber (6.5 mol% GeO_2) before and after 15 minutes exposure to pulses of 3 W/μm^2 peak intensity at 463 nm are given in Figure 5. The induced absorption is strong in the UV, with a tail that extends at least to the red end of the visible spectrum. This resembles the optical loss associated with Ge(1) defects[9].

This fiber was then thermally annealed for equal periods of time (15 minutes) at selected discrete temperatures (starting with a low temperature). At the end of each period, the spectrum of the induced loss was measured after the fiber had been removed from the furnace and cooled to room temperature. These measurements show that the induced absorption increases steadily between room temperature and about 250°C, where it peaks. Then beyond 250°C it gradually returns approximately to its level prior to thermal annealing. If the initial and final loss are caused by the same colour-centres, then the TPA-induced absorption is highly temperature stable.

In summary, there is a substantial body of evidence to support the view that TPA lies at the root of NLT in fibers; the steady-state induced loss scales with the photon flux n_p, bleaching out when n_p is reduced; the greatest losses are induced at a wavelength of 480 nm, corresponding via TPA to the 240 nm absorption band characteristic of oxygen-deficient germanosilicates; ESR results confirm the presence of Ge(1), Ge(2) and Ge(3) centres; the induced absorption spectrum is close to that associated with Ge(1) defects. Conclusive proof will be possible if a technique is found that is sensitive enough

to detect extremely small changes in defect populations over long path lengths in tiny total volumes of material.

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FIGURE CAPTIONS

1. Dynamics of the induced loss. Curve A shows the increase in loss with time for 5m of fiber exposed to 10 W/μm^2 at 470 nm. Curve B shows the subsequent evolution of the induced loss after the intensity is reduced to 1 W/μm^2.

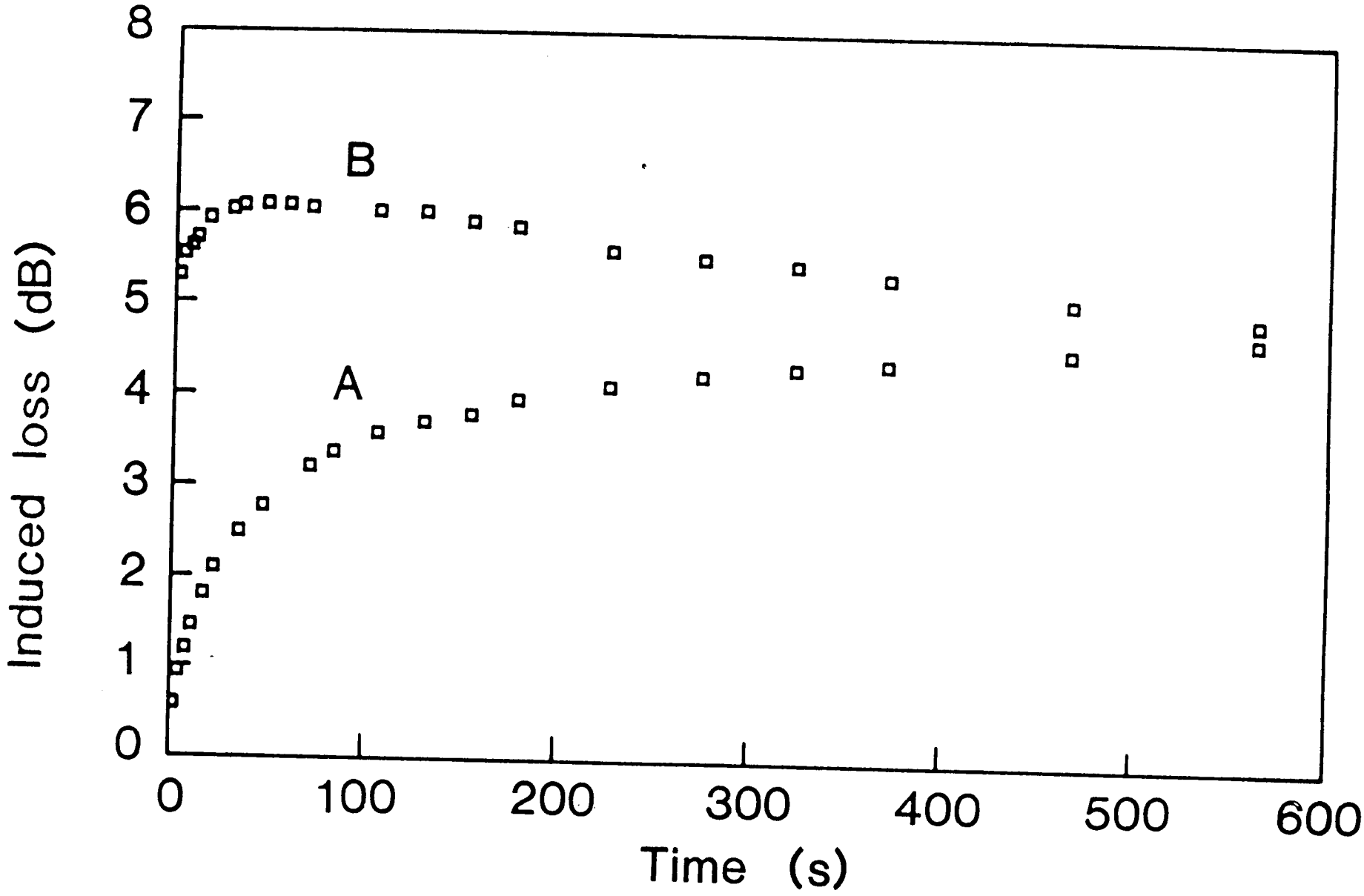
2. Saturated absorption levels for various launched intensities in 5m lengths of fiber. The lower points represent the loss immediately after exposure, the upper ones the loss measured one day later.

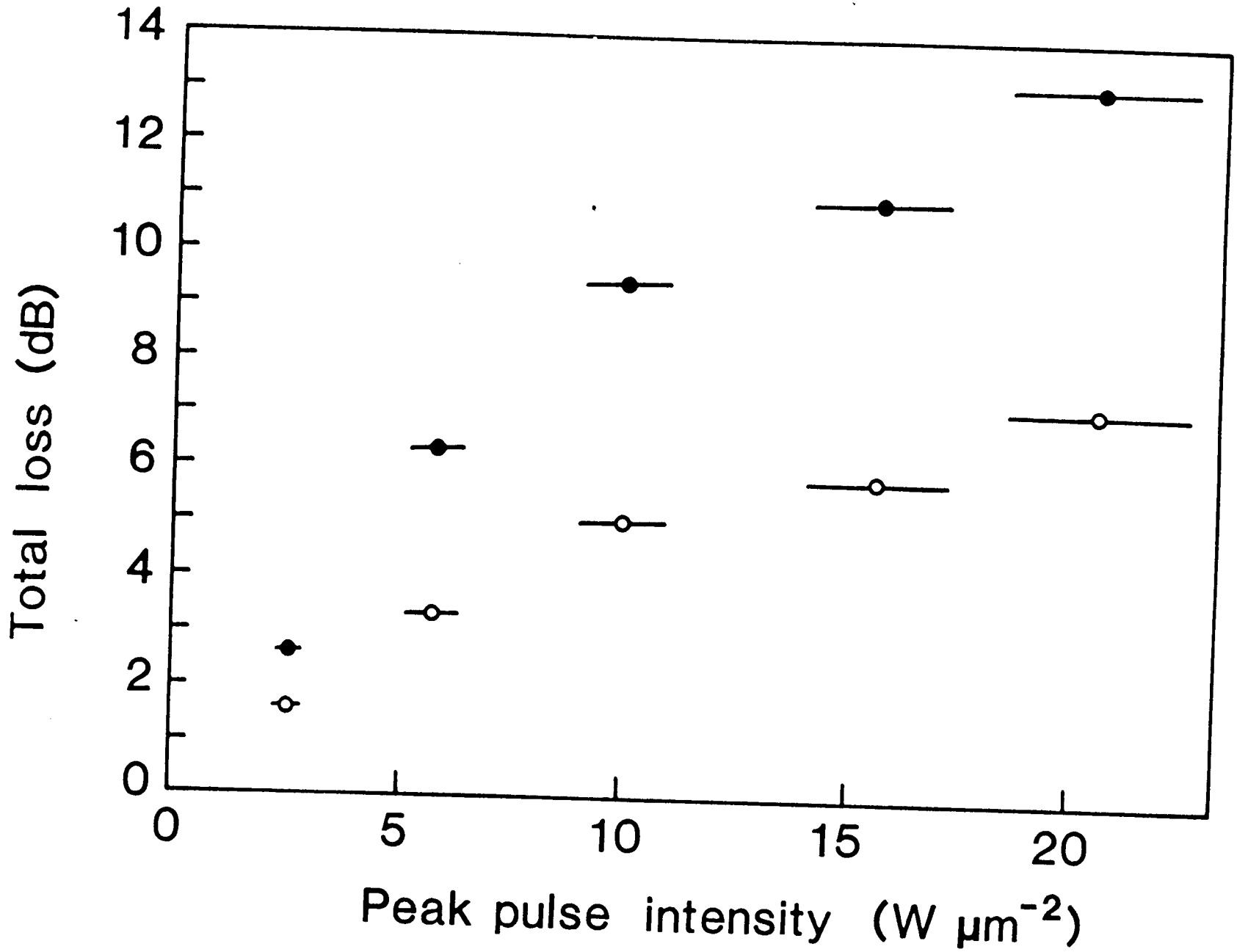
3. Schematic diagram of the different mechanisms of charge release and trapping. Thermal bleaching is represented by kT. TPA breaks the Ge-Si bonds to generate Ge(3) centres and free electrons:

$$\equiv Ge\text{-}Si\equiv \quad \underset{\longleftarrow}{\overset{2h\nu}{\longrightarrow}} \quad \equiv Ge^{\bullet} + Si\equiv + e^{-}$$

4. Attenuation at 450 nm in 5m lengths of fiber after exposure to 10 W/μm^2 at various pump wavelengths

5. Absorption spectra and effects of thermal annealing for 5m of fiber exposed to 3 W/μm^2 peak power at 463nm.


Induced loss (dB)
8
7
6
5
4
3
2
1
0
B
A
0
100
200
300
400
500
600
Time (s)


14
12
10
8
6
4
2
0
Total loss (dB)
0
5
10
15
20
Peak pulse intensity (W μm⁻²)

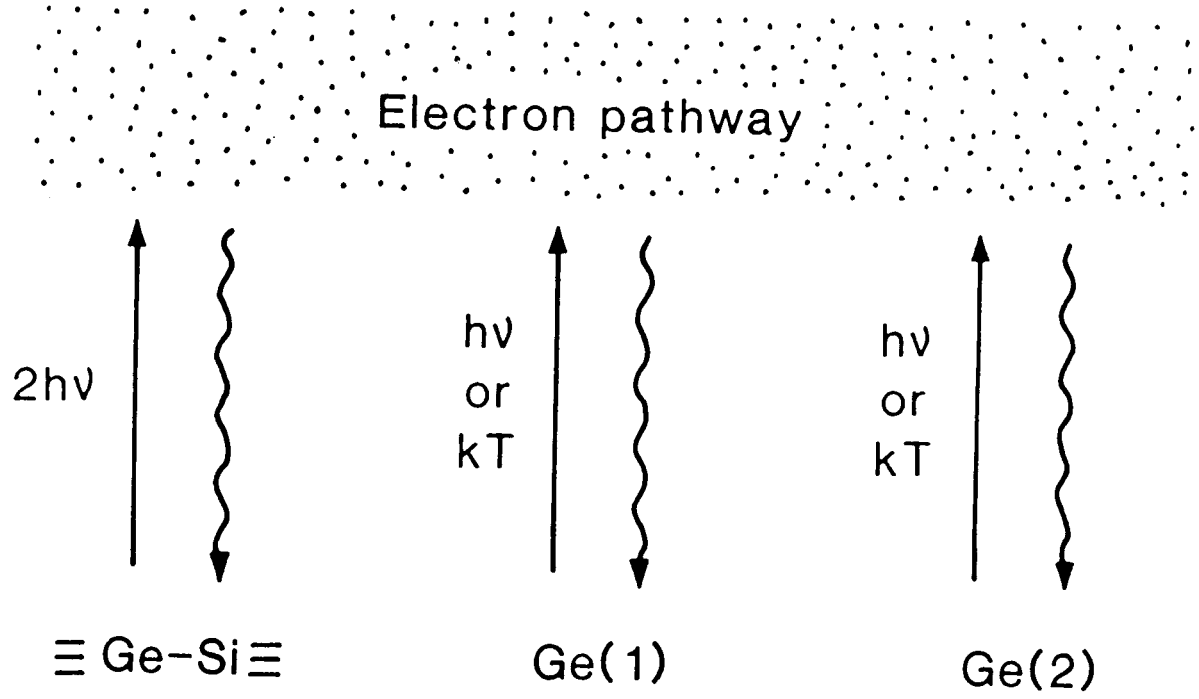
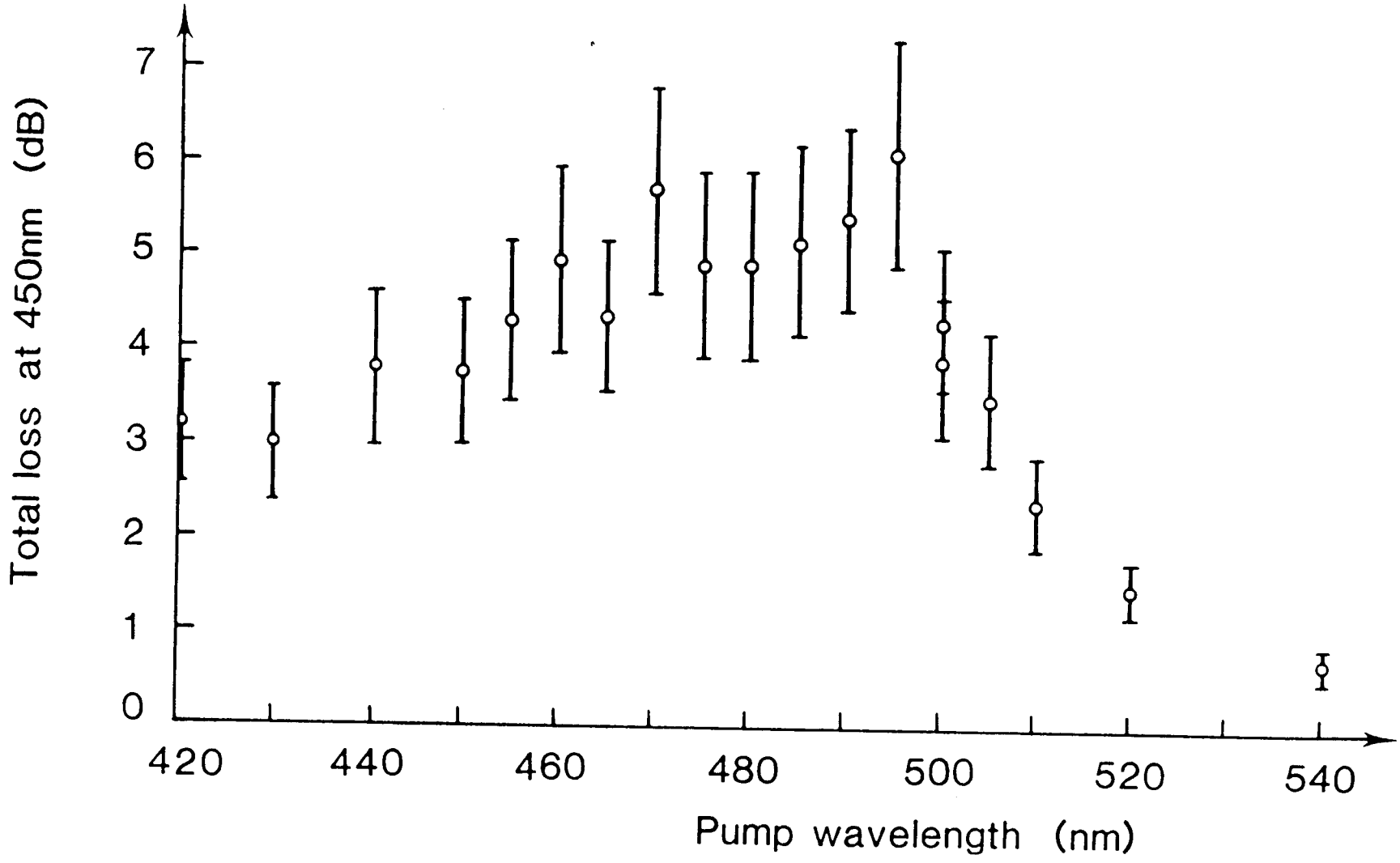
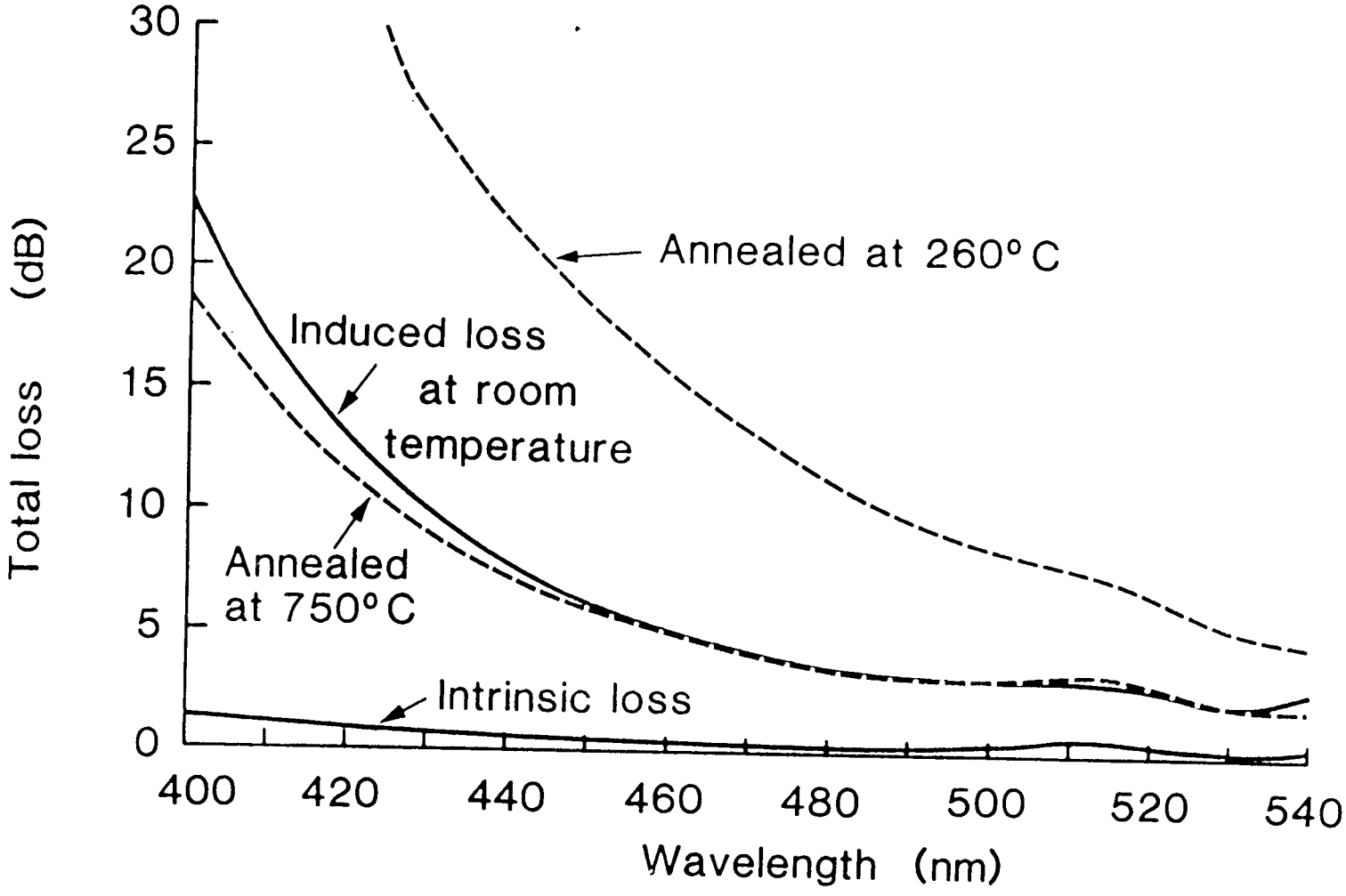


Figure 2


Total loss at 450nm (dB)
0
1
2
3
4
5
6
7
420
440
460
480
500
520
540
Pump wavelength (nm)


30
25
20
15
10
5
0
Total loss (dB)
Annealed at 260° C
Induced loss
at room
temperature
Annealed
at 750° C
Intrinsic loss
400
420
440
460
480
500
520
540
Wavelength (nm)