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MJ4 Color-center model for second-order optical nonlinearities in germanosilicate optical fibers

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Second-order nonlinearities can be induced in germanosilicate (GS) glass fibers in a variety of ways: by coherent superposition of pump and second-harmonic light (a third-order dc polarization P_{dc}[3] is induced that breaks the inversion symmetry[1]); by excitation poling with blue light and an external dc field,[2] E_{pol}; and by strong E_{pol}'s in the absence of light.[3] There is as yet no satisfactory agreement over the nature of the underlying alignment process. From related detailed studies of color-center behavior in GS fiber,[4] we have formulated a self-consistent model as developed in the following points.

(1) If (as has been proposed[5]) $\chi^{(2)}$ is caused by a built-in space-charge field, then in excitation poling it should start large (being initially $\chi^{(2)} = 3E_{pol}\chi^{(3)}$), gradually falling off to zero as the core region charges up. Upon switching off E_{pol}, it should immediately reappear at its initial level, with its sign reversed. In practice $\chi^{(2)}$ simply increases continuously, tending asymptotically toward a steady level; this strengthens the case for color-center alignment.

(2) Breakage of oxygen deficient Ge–Si bonds by two-photon absorption in the blue (or by $E_{pol} \sim 500\ \mathrm{V}\,\mu\mathrm{m}^{-1}$ in the absence of light) creates GeE' centers and positively charged $\equiv Si^{+}$ hole traps, releasing electrons that drift off through the glass matrix and are captured at Ge(1) and Ge(2) precursor sites to form negatively charged color centers $Ge^{-}(1)$ and $Ge^{-}(2)$ (Fig. 1).

(3) Ge(1) precursors offer three [and Ge(2) precursors, two] equally likely trapping positions for electrons; E_{pol} or $P_{dc}{}^{(3)}$ distorts the local structure surrounding the Ge(1) and Ge(2) precursors prior to capture, favoring one trapping position and hence causing color-center alignment. After capture, the bond angles change significantly, so a strong latch-on effect occurs, whereby a small perturbation in the initial condition results in a large permanent change in final alignment.

(4) These aligned $Ge^{\cdot}(1)$ and $Ge^{\cdot}(2)$ centers behave like nonlinear molecules, with intrinsic $\chi^{(2)}$ values. Although a strong correlation between Ge–Si bond breakage and $\chi^{(2)}$ has been found both by electron spin resonance and nonlinear spectroscopy, the alignment of the resulting GeE' centers is random and fixed in the glass matrix, so they are unlikely to contribute to a macroscopic $\chi^{(2)}$. This is explained by the fact that the broken Ge–Si bonds are merely electron donors and the $\chi^{(2)}$ level is proportional to the population of electrons donated.

In conclusion, color-center alignment offers a viable explanation for induced $\chi^{(2)}$'s in GS glasses; the talk will provide more evidence in support of this view.

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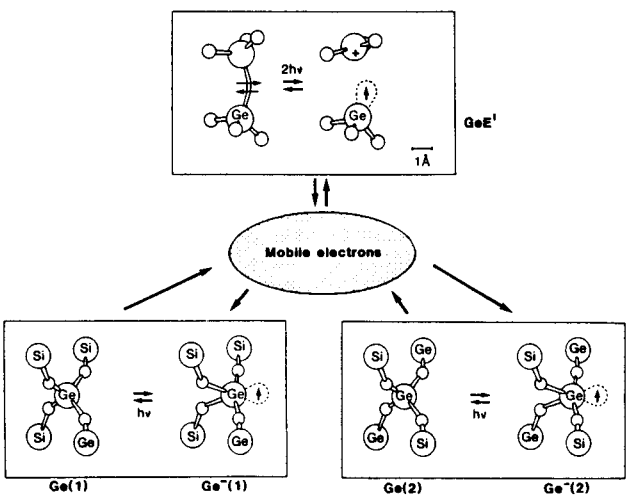

Fig. 1. Proposed color-center model. Electrons are passed among the Ge–Si bonds, GeE' centers, Ge(1) and Ge(2) precursors, and color centers. In the presence of a dc alignment bias [$P_{dc}{}^{(3)}$ or E_{pol}], preferential trapping occurs because of distortions in the electronic bond structure of the precursor molecules.