Photoinduced second-harmonic generation in optical fibers

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A possible mechanism for photoinduced second-harmonic generation in glass optical fibers is analyzed. This mechanism is based on the onset of an orientational order in a system of defects having a local second-order susceptibility.

Despite the abundance of publications (e.g., Refs. 1–6) on photoinduced second-harmonic generation in quartz-glass optical fibers, the nature and mechanism of this effect remain unclear. In the present letter we propose a model for explaining the appearance of a second-order macroscopic dielectric susceptibility by intense light in a glass. This susceptibility does not exist under ordinary conditions because of the isotropy of the glass.

Let us assume that a glass contains defects which do not have a symmetry center and which therefore do have a nonzero second-order polarizability. Since the orientation of defects in the glass is random, the average macroscopic susceptibility vanishes in the ordinary state of the glass. Laser light causes an interaction between these defects, which in turn causes a transition of the system of defects into an ordered state characterized by a nonzero macroscopic second-order susceptibility.

Let us assume that an intense pump wave

$$\mathbf{E}^{\omega}(t, \mathbf{r}) = \mathbf{E}^{\omega} \exp[i(\mathbf{k}^{\omega}\mathbf{r} - \omega t)] .$$

is propagating through the glass. Because of the local second-order susceptibility, the field of the pump wave induces dipole moments at the defects. At the frequency of the second harmonic of this wave, these dipole moments have a component $d_i^{2\omega} = b_{ijk} E_j^{\omega} E_k^{\omega} \exp(2i\mathbf{k}^{\omega}\mathbf{r}')$ (here and below, a repeated index means a summation) and thus emit a wave at the frequency 2ω (in other words, the pump wave is scattered into

the second harmonic by the defects). The alternating induced dipole moment at a defect interacts with the fields of the same frequency, 2ω. When there is a pump wave in the glass, two waves of frequency 2ω exist: the second-harmonic wave scattered by the defects and the plane second-harmonic wave which arises from the pump wave by virtue of quadrupole and other effects. The effective Hamiltonian of the system of interacting induced dipoles in the presence of a pump wave and a second-harmonic wave can be put in the form $H = U + V$, where the terms

$$U = -(NbI^{\omega})^2 \left[J(\vec{\eta}\vec{\eta}) - J_k \frac{(\mathbf{k}^{\omega}\vec{\eta})(\mathbf{k}^{\omega}\vec{\eta})}{(k^{\omega})^2}\right]$$

and

$$V = -\gamma NbI^{\omega}(\vec{\eta}\mathbf{E}^{2\omega})\cos(\Delta\mathbf{k}\cdot\mathbf{r})$$

describe the interaction of the dipoles with each other and with the field of the second-harmonic wave, respectively. The second-order macroscopic susceptibility can be put in the form

$$\chi^{(2)}_{ijk} = Nb\vec{\eta}_i\delta_{jk},$$

where N is the concentration of defects, $\vec{\eta}$ is a unit vector (the order parameter), the dimensional coefficient b is approximately equal to the local susceptibility of a defect, I^{ω} is the intensity of the pump wave, k^{ω} is its wave number, γ is a dimensionless factor (unity in the case of linearly polarized light), J and J_k are dimensionless coefficients which depend on the relative distribution of defects, and $\Delta\mathbf{k} = 2\mathbf{k}^{\omega} - \mathbf{k}^{2\omega}$, where $\mathbf{k}^{2\omega}$ is the wave vector of the plane second-harmonic wave. Hamiltonian H is equivalent to the Hamiltonian of a ferromagnet; the vector $b\vec{\eta}$ serves as a magnetization, and the field of the second-harmonic wave serves as an external magnetic field. An order which is described by the vector $\vec{\eta}$ and which gives rise to a macroscopic susceptibility $\chi^{(2)}$ thus arises in the system of defects.

A redistribution of the electron density near a defect upon the appearance of an ordered state may be accomplished by an irreversible relaxation of the defect, with result that order is preserved even after the pump light is turned off. This mechanism should operate most effectively for defects which have a high polarizability, which have a low symmetry, and which readily relax. In connection with the results of Ref. 5, we might cite an oxygen vacancy ≡Ge—Si≡. An example of defects which are oriented easily would be bound hydroxyl groups ≡Si—OH, in which the relaxation upon the onset of an order reduces to rotations of the OH group (an effect of a hydrogen impurity on second-harmonic generation was observed in Ref. 8). Because of the spatial periodicity of the interaction V, we have $\chi^{(2)} \sim \cos(\Delta\mathbf{k}\cdot\mathbf{r})$. This result leads to a phase matching of the fundamental and second-harmonic waves upon excitation of the second-harmonic wave in a glass with a macroscopic susceptibility induced beforehand.

Let us look at some numerical estimates. We assume that the oscillator strengths of the dipole transitions between defect states are $J \sim 10^{-3}$. The matrix elements of the dipole transitions are then $d \sim 10^{-18}$ cgs unit, and for a pump wavelength $\lambda \approx 1$ μm the

susceptibility of an individual defect is $b \approx 10^{-30}$ cgs unit. Consequently, the value $\chi^{(2)} \approx 10^{-12}$ cgs unit observed in the experiments of Refs. 1–5 is reached at a defect concentration of only $N \approx 10^{18}$ cm^{-3}. From the obvious condition for the onset of an order, $H/T \sim 1$ at $T \approx 300$ K, and for the same concentration, we find the critical pump intensity to be $I^{\omega} \approx 10^{6}$ cgs units. This figure corresponds to a field $E \approx 10^{3}$ cgs units $\approx 10^{5}$ V/cm and to a power flux of 10^{9} W/cm^2. If the area of the illuminated region in the fiber is 10 μm^2, the power required here would be on the order of 100 W or about 10 W in the pulse for the pulse lengths and periods used in Refs. 1–6. The agreement with experiment is thus completely satisfactory.

In a sense, the interaction between defects which we have been discussing here is analogous to a van der Waals interaction between atoms or molecules.[7,8] The primary distinction lies in the nature of the electromagnetic field which induces the dipole moments: In our case, it is the field of a pump wave, while in the case of the van der Waals interaction it would be a fluctuation vacuum field.[9]

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