

Model for second-harmonic generation in glass optical fibers based on asymmetric photoelectron emission from defect sites

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Received December 19, 1990

We present a self-consistent calculation of anomalous second-harmonic generation in glass optical fibers. Quantum interference between multiphoton absorption processes leads to asymmetric photoelectric emission from defects, creating a spatially periodic space-charge electric field. The second harmonic is found to grow exponentially along the fiber, then saturate to a maximum value proportional to the square of the fundamental intensity. The predicted conversion efficiency is in reasonable agreement with experiments.

The observation of laser-induced growth of second-harmonic generation (SHG) in glass optical telecommunications fibers^{1,2} has inspired many efforts³⁻⁵ to elucidate the origin and growth of the responsible $\chi^{(2)}$ nonlinearity. In this Letter we present a calculation of SHG that has been motivated by several recent experimental and theoretical advances. In our model quantum interference between multiphoton absorption processes leads to asymmetric photoelectron emission from defects, creating a spatially periodic dc electric field in the fiber. This field, which we calculate using a simple charge-transport model, has both the correct periodicity and phase to produce self-consistent growth of the second-harmonic light in reasonable accord with observations.

Experimental evidence^{6,7} has supported the view that the photoinduced SHG is in fact the result of a strong transverse dc electric field that acts on the allowed $\chi^{(3)}$ nonlinearity of the glass, yielding an effective $\chi^{(2)}$. Also, it has been known for some time that an atom subject to harmonically related electric fields can exhibit a transition rate that, owing to quantum interference effects, is dependent on the relative phases between these fields.⁸ However, it has only recently been pointed out that in the case of an ionizing transition this interference can give rise to a phase-dependent preferred electron ejection direction.⁹⁻¹² Zel'dovich and collaborators have proposed that these effects may be responsible for the laser-induced SHG in glass fibers.^{9,12} In this context, Dianov and collaborators have advanced a phenomenological model^{13,14} of asymmetric electron ejection based on a coherent photovoltaic effect¹⁵ that results from an interference between one- and two-photon transitions.

While there is no experimental evidence that a second-harmonic photon (typically green light at 532 nm) has sufficient energy to ionize a defect, it is well known that there are germania-related defects that have a

broad absorption band centered at approximately 5 eV, which can be accessed with the equivalent energy of four 1.064- μm photons. Recent research¹⁶ points to the role of these defects as trap sites for charge carriers. We suppose, therefore, that the charge redistribution takes place among trap sites following photoionization by the two-, three-, and four-photon processes shown in Fig. 1.

We treat the fields as plane waves, copolarized along the x axis, propagating along the fiber z axis, and modulated by a slowly varying amplitude $E_{m\omega}$,

$$\mathcal{E}_{m\omega}(z, t) = E_{m\omega}(z, t) \exp[i(\phi_{m\omega} - m\omega t)] + \text{c.c.},$$

$$m = 1, 2, \quad (1)$$

where the phases have an unperturbed variation $\phi_{m\omega} = k_m z$.

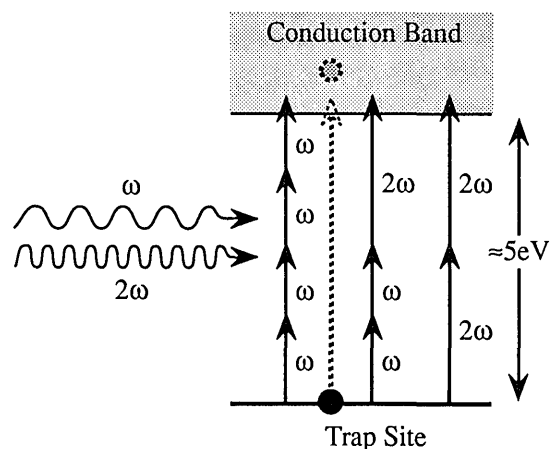


Fig. 1. Multiphoton paths to ionization, assuming a broad defect band at approximately 5 eV and a fundamental laser wavelength of 1.064 μm .

We use the standard formalism of perturbation theory,¹⁷ carried out to fourth order in the perturbation energy, to calculate the ionization rate for electrons ejected upward, $\dot{\rho}_+$, or downward, $\dot{\rho}_-$. After a straightforward but lengthy calculation, we find

$$\begin{aligned} \dot{\rho}_{\pm} = & R(|E_{2\omega}|^4 + \eta_3^2|E_{2\omega}|^2|E_{\omega}|^4 + \eta_4^2|E_{\omega}|^8) \\ & + \{\eta_4|E_{2\omega}|^2|E_{\omega}|^4 \exp[2i(\phi_{2\omega} - 2\phi_{\omega})] + \text{c.c.}\} \\ & \pm \{-i\eta_3|E_{2\omega}||E_{\omega}|^2(\eta_4|E_{\omega}|^4 - |E_{2\omega}|^2) \\ & \times \exp[i(\phi_{2\omega} - 2\phi_{\omega})] + \text{c.c.}\}. \quad (2) \end{aligned}$$

The form of this expression is fairly general, but in our simplified treatment, where the defect is taken to be a one-dimensional potential with a single bound state and free particle excited states, we find that

$$\eta_3 = \frac{8e\kappa_3}{\hbar\omega k}, \quad \eta_4 = \frac{8e^2\kappa_4}{\hbar\omega^3 m}. \quad (3)$$

Here $e = |e|$ is the elementary charge, m is the mass of the electron, k is its wave number, c is the speed of light, and \hbar is the reduced Planck's constant. κ_3 and κ_4 are factors that are less than but of the order of unity, and they are weak functions of the ionized electron's momentum.

The first set of terms within the parentheses in Eq. (2) is recognized as the individual contributions from two-, three-, and four-photon ionization rates. The second set of terms, within the first set of braces, represents the interference between the two- and four-photon processes. We drop these since they are modulated at twice the phase mismatch spatial frequency. The last set of terms, within the second set of braces, which stems from the interference of the three-photon process with both the two- and four-photon interactions, is the set of interest.

We model the process of charge redistribution with a simple band charge-transport model wherein a photoejected carrier propagates for a time τ_m , after which it no longer "remembers" its original direction of propagation. It remains in the conduction band for a total time τ_R , before it becomes retrapped at an empty site. In this picture the anisotropic excitation gives rise to an anisotropic photogenerated current,

$$j_a = -en_d v_0 \tau_m (\dot{\rho}_+ - \dot{\rho}_-), \quad (4)$$

where n_d is the number density of filled sites and v_0 is the initial velocity of the ejected electron. At the same time the glass develops a local conductivity proportional to the total number of charges in the conduction band,

$$\sigma = en_d \mu_e \tau_R (\dot{\rho}_+ + \dot{\rho}_-), \quad (5)$$

where μ_e is the electron mobility. The redistribution of charge develops a space-charge field that increases until the photocurrent is balanced by the conduction current. In the steady state, then,

$$E_{dc} = -\frac{j_a}{\sigma} = \frac{v_0 \tau_m}{\mu_e \tau_R} \frac{\dot{\rho}_+ - \dot{\rho}_-}{\dot{\rho}_+ + \dot{\rho}_-} \equiv E_0 \frac{\dot{\rho}_+ - \dot{\rho}_-}{\dot{\rho}_+ + \dot{\rho}_-}. \quad (6)$$

We see that the field is determined by the ratio of

the anisotropic to the isotropic photoelectron ionization rates and that the ionization rate constant R [Eq. (2)] does not appear. Now, from Eq. (2), and given the indices of refraction $n_{m\omega}$ and the permittivity ϵ_0 and permeability μ_0 of free space, the dc electric field is written in a convenient form using the intensities $I_{m\omega} = 2n_{m\omega} \sqrt{\epsilon_0/\nu_0} |E_{m\omega}|^2$,

$$\begin{aligned} E_{dc} = & -iE_0 \frac{\eta_3}{\eta_4} \frac{1 - I_{2\omega}/I_{sat}}{1 + \beta I_{2\omega}/I_{sat} + (I_{2\omega}/I_{sat})^2} \\ & \times \frac{|E_{2\omega}|}{|E_{\omega}|^2} \exp[i(\phi_{2\omega} - 2\phi_{\omega})] + \text{c.c.}, \quad (7) \end{aligned}$$

where $\beta = \eta_3^2/\eta_4$ and the saturation intensity $I_{sat} = \eta_4 \sqrt{\mu_0/\epsilon_0} (n_{2\omega}/2n_{\omega}^2) I_{\omega}^2$.

The space-charge field gives rise to growth of the second harmonic through the allowed $\chi^{(3)}$ nonlinearity of the glass. In the usual slowly varying envelope approximation,¹⁸

$$\frac{\partial E_{2\omega}}{\partial z} = \frac{i\omega}{n_{2\omega} c} 3\chi^{(3)}(0, \omega, \omega, \omega) E_{dc} |E_{\omega}|^2 \exp(2i\phi_{\omega}). \quad (8)$$

We use Eq. (7) for E_{dc} , after dropping the asynchronous complex conjugate term. Writing the evolution of the second harmonic in terms of its intensity, we get the expression for second-harmonic growth and the key result of the model,

$$\frac{dI_{2\omega}}{dz} = G_0 \frac{1 - I_{2\omega}/I_{sat}}{1 + \beta I_{2\omega}/I_{sat} + (I_{2\omega}/I_{sat})^2} I_{2\omega}. \quad (9)$$

Since I_{sat} is a constant (in the nondepleted pump approximation), the second harmonic is seen to grow as a saturated exponential with a small-signal gain,

$$G_0 = \left(\frac{6\hbar\omega^3}{en_{2\omega} C} \right) \left(\frac{\kappa_3}{\kappa_4} \frac{1}{\mu_e} \chi^{(3)} \right) \left(\frac{\tau_m}{\tau_R} \right). \quad (10)$$

We have grouped this result into three distinct factors. The first factor contains quantities that are completely known to us. We will take a fundamental wavelength of 1.064 μm and an index of refraction of 1.5.

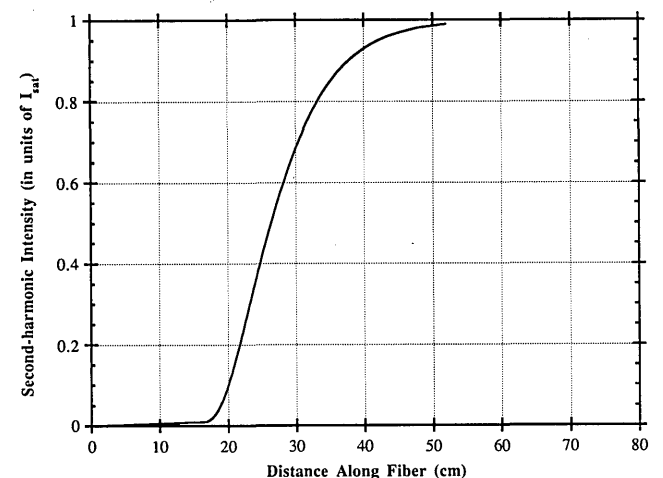


Fig. 2. Steady-state growth (in space) of the SHG calculated from Eq. (9), using the experimental parameters of Ref. 2: $I_{2\omega}(z=0)/I_{sat} = 1.2 \times 10^{-6}$. The experimental data are qualitatively fit with $\beta = 2$, and $G_0 = 70 \text{ m}^{-1}$.

The second factor contains quantities for which we have reasonable estimates. We use the measured¹⁹ electron mobility in silica, $\mu_e = 2.0 \times 10^{-3} \text{ m}^2/\text{V}\cdot\text{s}$. Ignoring dispersion, we use $\chi^{(3)} = 1.8 \times 10^{-22} \text{ m}^2/\text{V}^2$, which has been measured by nearly degenerate four-wave mixing.²⁰ Since κ_3 and κ_4 are both less than but of the order of unity, we choose unity for the ratio. The last factor is unknown, except that $\tau_m/\tau_R \leq 1$. Therefore we can reasonably estimate a maximum small-signal gain G_0 of 4500 per meter, or 17 dB/cm for a self-seeding fiber.

Equation (9) can be integrated, and $I_{2\omega}(z)$ is plotted in Fig. 2, where we have used the experimental parameters of Ref. 2, which reports the highest self-seeded conversion efficiency to date. For a qualitative fit we use $\beta = 2$ and find that G_0 need only be $\approx 70 \text{ m}^{-1}$ to obtain reasonable qualitative agreement with experiment, which is much less than our upper-bound estimate.

In our model, saturation of the second harmonic follows directly from the competition between the interference of the three- and four-photon process, which has the correct phase for growth, and the interference between the three- and two-photon process, which does not [see Eq. (2)]. We estimate that $I_{\text{sat}} = (4.8 \times 10^{-17} \text{ cm}^2/\text{W})I_\omega^2$. For, e.g., $I_\omega = 3 \times 10^{10} \text{ W}/\text{cm}^2$, the maximum conversion is 1.5%, which is in surprisingly good accord with the 5% of the experiment,² considering the elementary way in which we have modeled the defects. We note that I_{sat} is insensitive to our choice of transport model, if there are no other saturation effects taking place there such as defect depletion.

In this model we have found reasonable values of gain and predict an intensity-dependent maximum in conversion efficiency. Future research needs to consider a more realistic (e.g., three-dimensional) defect potential and ionized electron states along with a perhaps more refined transport model.

Dana Z. Anderson thanks B. Ya. Zel'dovich for stimulating and valuable discussions. Victor Mizrahi thanks T. Leskova for extensive help with the Soviet literature. The research of Dana Z. Anderson is supported by the U.S. Air Force Office of Scientific Research (Grant AFOSR-90-0198) and by the National Science Foundation (Grant PHY 9012244). The re-

search of John E. Sipe is supported by the Natural Sciences and Engineering Research Council of Canada and by the Ontario Laser and Lightwave Research Center.

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