Electrically Stimulated Light-Induced Second-Harmonic Generation in Glass: Evidence of Coherent Photoconductivity

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A strong electrostatic field applied to glass is spatially modulated by intense light at frequencies ω and 2ω. The phenomenon is explained in terms of photoconductivity being dependent on the relative phase of the light fields at different frequencies. [S0031-9007(97)02913-X]

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Recently, the interference between different quantum processes has been the subject of considerable attention in many areas of physics. One of the reasons for this growing interest is that such kinds of interference open a prospect of a new degree of freedom in the control of physical processes—not only by the intensity or the polarization of light, but also by the phase of light. It was observed in the experiments on rubidium atoms [1] and photoemission from Sb-Cs photocathodes [2] that the interference between the one- and two-photon transition moments changes the angular distribution of the photoelectrons and excites a phase dependent current (coherent photocurrent). Recently, coherent photocurrent via quantum interference of electrons produced by one- and two-photon bound-to-free intersubband transitions was observed in AlGaAs/GaAs quantum well superlattices [3]. However, second-harmonic generation (SHG) via photoinduced spatially oscillating electrostatic fields in glass was probably the first observed phenomenon where coherent photocurrent was involved [4–6]. Indeed, SHG is forbidden in glass due to the inversion symmetry of the glass matrix. However, when a sample of glass or glass fiber (e.g., Ge-doped silica glass) is illuminated for some time (preparation time) with intense light containing frequencies at ω (pump) and 2ω (second-harmonic seeding) and afterwards the seeding is removed, it is still possible to observe light at frequency 2ω (second harmonic) generated in the glass [7]. In experiments on optical glass fibers strong amplification of a weak seeding radiation at frequency 2ω, generated inside a fiber as a result of magnetic dipole or quadrupole effects, is observed after launching only intense light at frequency ω [8]. In this phenomenon the coherent photocurrent, excited in glass as a result of interference between one-photon ionization by light at frequency 2ω and two-photon ionization by light at frequency ω, gives rise to a spatially oscillating electrostatic field E_g (photogalvanic field): $E_g \simeq j_{\mathrm{coh}}/\sigma$, where j_{coh} is the coherent photocurrent and σ is the photoconductivity. The amplitude of the photogalvanic field E_g in glass is typically $\sim 10^4$–10^5 V/cm and this field can induce the second-order nonlinear susceptibility ($\chi^{(2)}$), responsible for SHG, via the third-order susceptibility: $\chi^{(2)} = 3\chi^{(3)}E_g$. Assuming $\chi^{(3)} \simeq 10^{-22}$ $(\mathrm{m/V})^2$ for silica glass, the magnitude of $\chi^{(2)}$ is $\sim 10^{16}$–10^{15} m/V. It turns out that the $\chi^{(2)}$ periodicity can compensate for the phase velocity mismatch, thus making the SHG process efficient. In nonlinear optics the process that allows such compensation is known as quasi-phase-matching.

Unlike mixing between one- and two-photon processes, which leads to a modulation in the angular distribution of photoelectrons, it was shown in experiments on mixing between one- and three-photon processes in xenon gas [9], HCl molecular beam [10], and between five- and three-photon process in mercury vapor [11] that the total cross section of the ionization transition can be directly modulated by changing the relative phase between two light fields oscillating at different frequencies. However, to our knowledge, two interesting aspects of the phenomenon have not been yet investigated. First, experiments on quantum interference have been carried out only in centrosymmetric media although it was already widely discussed that in media without inversion symmetry the interference between one- and two-photon transitions induced by light at frequencies 2ω and ω can lead to a modulation of the total cross section for the overall transition [12]. Second, the modulation of the total cross section of ionizing transitions due to quantum interference (coherent photoconductivity) has been observed only in atomic systems. In this Letter we report the observation of *efficient* second-harmonic generation in glass subjected to a strong external electrostatic field. The spatial periodic modulation of the applied electric field, responsible for the second-harmonic signal, arises from the interaction of the intense light at frequencies ω and 2ω with glass, which has its inversion symmetry broken by the applied field. The process could represent the first evidence of coherent photoconductivity in glass.

Let us consider the interaction of light beams of frequencies ω and 2ω in glass when a strong dc electric field ($E_0 \gg E_g$) is applied. The probability of simultaneous ionization of the defect site by two photons at frequency ω and one photon at frequency 2ω in the presence of E_0 is given by

$$\begin{aligned} P &\sim |a_2 E_\omega E_\omega + b_2 E_0 E_{2\omega}|^2 \\ &= |a_2|^2 I_\omega^2 + |b_2|^2 I_{2\omega}^2 E_0^2 \\ &\quad + 2\,\mathrm{Re}(a_2 b_2^* E_0 E_\omega E_\omega E_{2\omega}^*), \end{aligned} \tag{1}$$

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where E_ω and $E_{2\omega}$ are the field amplitudes at frequency ω and 2ω, respectively, I_ω and $I_{2\omega}$ are the corresponding intensities, and a_2 and b_2 are two complex coefficients which determine the weight of the different processes in the probability P. All terms in this expression are even powers of the electric field regardless of the fact that they are uniform (first two terms) or modulated [being dependent on the relative phase of the fields at frequencies ω and 2ω (the last term)]. The modulated part of the probability leads to modulation of the total ionization cross section and hence to a corresponding modulation of the photoconductivity:

$$\sigma = \sigma_0 + \sigma_{\mathrm{coh}}, \tag{2}$$

where

$$\sigma_0 \sim |a_2|^2 I_\omega^2 + |b_2|^2 I_{2\omega} E_0^2$$

is the uniform part of the photoconductivity and $\sigma_{\mathrm{coh}} \sim 2\,\mathrm{Re}(a_2 b_2^* E_0 E_\omega E_\omega E_{2\omega}^*)$ is the part of the photoconductivity being dependent on the relative phase between E_ω and $E_{2\omega}$ (coherent photoconductivity). Therefore the coherent photoconductivity spatially oscillates with a period Λ determined by the refractive index mismatch between light waves at frequencies ω and 2ω:

$$\sigma_{\mathrm{coh}} \sim \cos 2\pi z/\Lambda, \qquad \Lambda = \lambda/2(n_{2\omega} - n_\omega),$$

where λ is the wavelength in vacuum of the light at frequency ω and $n_{2\omega}$, and n_ω are the refractive indices at frequencies 2ω and ω, respectively. It should be pointed out that the process of quantum interference considered above is qualitatively different from the already observed process of quantum interference in glass without strong ($E_0 \gg E_g$) dc electric field applied [4–8]. In the latter case a modulation of the angular distribution of photoelectrons (*coherent photocurrent*) rather than a modulation of the total cross section of ionization (*coherent photoconductivity*) takes place.

In fact the coefficients a_2 and b_2 in the expression of the probability P can depend on the intensities of light at frequencies 2ω and ω: $a_2 = a_1(I_\omega^2, I_\omega^3, I_{2\omega}), b_2 = a_2(I_\omega^2, I_\omega^3, I_{2\omega})$ and this can result in even higher order nonlinear processes [5].

The Ohmic current ($j = \sigma E_0$) induced by the applied electric field separates photocarries which accumulate at the boundaries of the illuminated region and screen the applied electric field E_0. The resulting internal electric field inside the glass E_c, assuming intensities of light waves at frequencies 2ω and ω to be constant, evolves accordingly to

$$dE_c/dt = -E_c/\tau, \qquad \tau = \epsilon/\sigma,$$

where τ is the dielectric relaxation time and ϵ is the dielectric constant of glass. An approximate solution for E_c in the limit $\sigma_{\mathrm{coh}} \ll \sigma_0, \tau = \tau_0 = \epsilon/\sigma_0$ is

$$\begin{aligned} E_c &\simeq \sigma_{\mathrm{coh}}/\sigma_0 E_0[1 - \exp(-t/\tau_0)] \\ &\quad \times \exp(-t/\tau_0) + E_0 \exp(-t/\tau_0) \\ &= E_c^{\mathrm{coh}} + E_c^0, \end{aligned} \tag{3}$$

where E_c^{coh} and E_c^0 are the modulated and uniform parts of the internal dc electric field. The modulated part of the electric field, proportional to σ_{coh}, and the corresponding $\chi^{(2)}$ grating ($\chi^{(2)} = 3\chi^{(3)} E_c^{\mathrm{coh}}$) are zero at the beginning ($t = 0$), reach maximum values at $t = \tau_0 \ln 2$, and finally decrease to zero in the steady state condition ($t = \infty$). The $\chi^{(2)}$ periodicity compensates exactly for the phase velocity mismatch (refractive index mismatch), thus making the second-harmonic generation process efficient [$I_{2\omega} \propto (\chi^{(2)})^2 l^2 (I_\omega)^2$, where l is the interaction length]. In fact, the evolution of the dc electric field can be more complicated since the second-harmonic electromagnetic field and the $\chi^{(2)}$ grating, i.e., modulated dc electric field itself, can vary in time and along the sample. However, even in a more general case, Eq. (3) still provides a qualitative physical description (in particular at the beginning of the process and in the steady state condition). A more detailed mathematical analysis for the general case will be presented elsewhere.

In our experiments we used Ge-doped silica fibers with built-in capillaries on both sides of the core, suitable for introducing wire electrodes (Fig. 1). A typical fiber used in our experiments had a 0.32 numerical aperture, 3 μm core diameter, 165 μm outer diameter, 50 μm hole diameter, and 9 μm interhole spacing. Metal wires of 25 μm diameter were inserted into the fiber over length varying from 5 to 500 mm. A mode-locked (76 MHz repetition rate, 300 ps pulse duration) and Q-switched (1 kHz repetition rate, 300 ns envelope duration) Nd-YAG laser operating at 1064 nm was used as the pump source.

First, we launched only infrared light (pump) of $\sim$12 mW average power (1 kW peak power) for $\sim$1 h into the fiber, $\sim$25 cm long. No SHG was observed in the fiber. Then we launched simultaneously the pump of $\sim$12 mW average power and the SH seeding of $\sim$40 μW average power, generated in a ($KTiOPO_4$) crystal. The SH seeding was removed after preparation of $\sim$10 min and a SH signal (being generated in the fundamental LP_{01} fiber mode) of $\sim$20 μW average power was observed, corresponding to a conversion efficiency of $\sim$0.16%. This was confirmed by monitoring the growth of the SH signal in the fiber when the SH seeding was blocked for a short time during the preparation (Fig. 2). Our result is in good agreement with previous observations on photoinduced SHG in Ge-doped

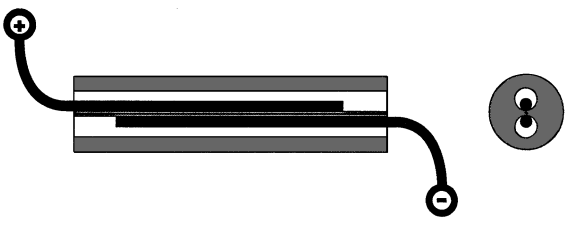

FIG. 1. Cross section of a Ge-doped silica fiber with internal electrodes. The fiber had a 0.32 numerical aperture, 3 μm core diameter, 165 μm outer diameter, 50 μm hole diameter, and 9 μm interhole spacing. Metal wires of 25 μm diameter were inserted into the fiber over length varying from 5 to 500 mm.

fibers [7] and may be explained by the appearance of a modulated second-order nonlinearity ($\chi^{(2)}$ grating) in the fiber as a result of the charge separation due to coherent photocurrent.

We then applied voltages up to 10 kV (corresponding to electrical fields $\sim 10^7$ V/cm, which are probably among the highest values ever applied to glass materials) across the electrodes inside the fiber and launched into the fiber (25 cm long) IR pump light of ~1 kW peak power. The length of the region over which the electric field had been applied (electrode superposition) was ~20 cm long. For applied voltages greater than ~2 kV a strong increase of weak electric-field-induced second-harmonic (EFISH) signal of ~10 nW (i.e., generated immediately after the electric field was applied) was observed (Fig. 2). In fact the SH signal (being generated in the fundamental LP_{01} mode) grows rapidly, within a 1 min time scale, reaching the maximum value of ~250 μW average power and then slowly decreases (Fig. 2). After this first growth and decrease an interesting "echo" behavior was observed when the voltage was repeatedly switched off and on (Fig. 3). After disconnecting the voltage, the SH signal rapidly drops, remaining zero for a short time period, then increases, reaching almost 60% of the maximum value when the voltage was applied and finally slowly decreases again (Fig. 3). During these experiments we achieved conversion efficiencies as high as ~2% for a peak pump power of ~1 kW, which is ~10 times higher than in our experiments carried out without applying an external electric field. We detected a ~4 times smaller SH signal and ~2 times smaller growth rate in a fiber with electrode superposition ~10 cm long, thus two times shorter than in the previous fiber used.

We excluded a simple explanation of this phenomenon by the EFISH seeding of the $\chi^{(2)}$ grating in a fiber on the basis of two experimental observations. First, we observed that the second-harmonic growth took place only in the region where the electric field was applied (where the electrodes superposed) and the SH output was quadratic dependent on the length of this region. Second, no SHG signal was observed in any of the used fibers after preparation for more than 1 h with a weak external SH seeding of the same and even an order of magnitude higher level of power compared to the EFISH maximum signal. The maximum SH signal and the time, necessary to reach half of the maximum, when the voltage is switched on and switched off (Fig. 4), clearly depend on the applied voltage: The maximum SH signal increases with the applied voltage whereas the half-growth time decreases with the applied voltage.

The high conversion efficiency measured in our experiments and the observed quadratic dependence of the SHG efficiency on the fiber length represent clear evidences of quasi-phase-matched SHG due to the presence of a $\chi^{(2)}$ grating in the fiber. Moreover, the enhancement of SHG in glass due to a strong applied electric field ($\sim 5.5 \times 10^6$ V/cm) was confirmed by experimental observation that the amplitude of the $\chi^{(2)}$ grating increases at least ~3 times (corresponding to an increase of conversion efficiency ~10 times) compared to the experimental situation where no electric field is applied to the fiber. From our measurements we can also estimate the amplitude of the second-order susceptibility $\chi^{(2)} \simeq 10^{-14}$ m/V, which corresponds to a modulation of ~8% of the applied electric

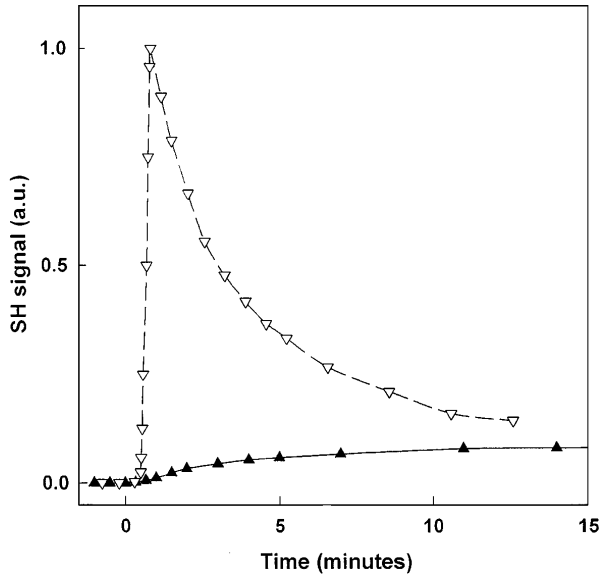

FIG. 2. Time dependence of a SH signal in a fiber with external SH seeding of ~40 μW average power (opened triangles). The seeding was launched at $t = 0$. The SH growth was monitored by blocking the SH seeding. Time dependence of a SH signal in a fiber with applied voltage of 5 kV (filled triangles). The voltage was switched on at $t = 0$. The length of the fiber is ~25 cm and the superposition of the electrodes is ~20 cm. Average pump power is ~12 mW.

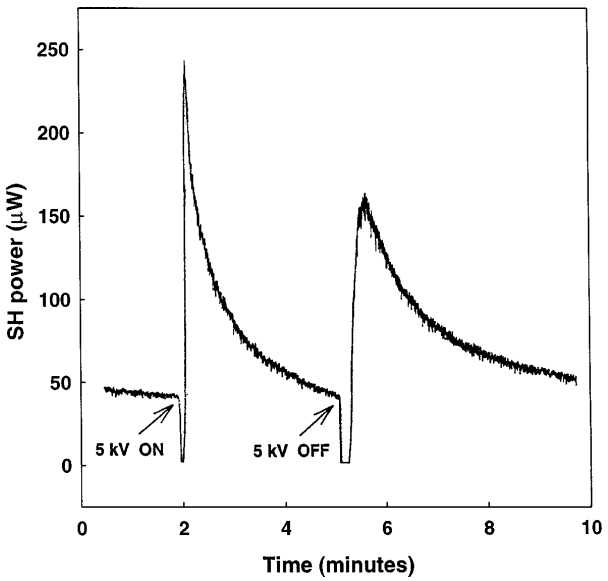

FIG. 3. Time dependence of a SH signal in a fiber when an applied voltage of 5 kV was repeatedly switched on and off. The instants when the voltage is on and off are shown by arrows.

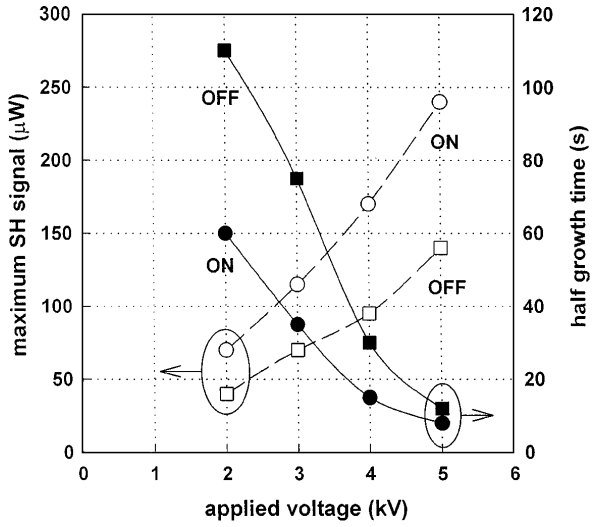

FIG. 4, Dependencies of the maximum SH signal and the time necessary to reach half of this signal on applied voltage when the voltage is switched on and off, respectively.

field. The possibility of a $\chi^{(2)}$ grating due to the interference between different fiber modes at the same frequency of light [13] was excluded since both the SH and the pump signals propagated in the fundamental LP_{01} mode.

Our experimental results can be qualitatively interpreted in light of the mechanism presented above, i.e., on the basis of photoconductivity being dependent on the relative phase between interacting waves at frequencies ω and 2ω, which provides quasi-phase-matching for the SHG. More important this model is consistent with the increase in conversion efficiency with the applied voltage and is also in good agreement with the experimental time dependence of the SH signal growth to a maximum value followed by a gradual decay. It is possible to estimate from (2), assuming $a_2 \simeq b_2$, and using the experimental values for the electric fields ($E_0 = 5.5 \times 10^6$ V/cm, $|E_\omega| = 3.2 \times 10^6$ V/cm, $|E_{2\omega}| = 4.5 \times 10^5$ V/cm), the theoretical ratio $\sigma_{\text{coh}}/\sigma_0 \simeq 2E_0|E_\omega|^2|E_{2\omega}|/(|E_\omega|^4 + |E_{2\omega}|^2E_0^2) \simeq 0.4$. Experimental estimation of the ratio $\sigma_{\text{coh}}/\sigma_0$ can be obtained from (3): $\sigma_{\text{coh}}/\sigma_0 \simeq 4E_m^{\text{coh}}/E_0$, where E_m^{coh} is the maximum amplitude of the modulated dc electric field. Using experimentally obtained $E_m^{\text{coh}}/E_0 \simeq 0.08$, the experimental ratio $(\sigma_{\text{coh}}/\sigma_0)_{\text{exp}} \simeq 0.3$ is in good agreement with the theoretical prediction. Moreover, an interesting feature in the time dependence of the SH signal—the origin of an unexpected growth (echo) of the SH signal after switching off the voltage (Fig. 3)—can be explained. Indeed, as follows from Eq. (3) in steady state conditions the internal electric field E_c is zero, which means that the applied dc electric field $\mathbf{E}_0$ is locally compensated by the electric field $\mathbf{E}_s$ due to the screening charges, accumulated at the boundaries of the illuminated region: $\mathbf{E}_c = \mathbf{E}_0 + \mathbf{E}_s \simeq 0, \mathbf{E}_0 \simeq -\mathbf{E}_s$. After switching off the voltage ($E_0 = 0$) the internal electric field rapidly increases almost to its initial value $\mathbf{E}_c \simeq \mathbf{E}_s \simeq -\mathbf{E}_0$. This rapid increase induces in the glass, due to the dc Kerr effect, a change in the phase mismatch between pump and SH waves which cannot be compensated any longer by the remaining weak $\chi^{(2)}$ grating of a period given by the previous phase mismatch (before switching off the voltage). The lack of quasi-phase-matching makes the SH signal drop quickly to zero. However, due to the fact that the internal electric field increases almost to its initial value (with opposite polarity) a new quasi-phase-matching grating (with a new period given by the new phase mismatch) will start growing and the process of electrically stimulated growth of $\chi^{(2)}$ grating via coherent photoconductivity repeats itself for a second time.

In conclusion we observed electrically stimulated light-induced second-harmonic generation in glass due to the modulation of a strong applied electrostatic field induced by light at frequencies ω and 2ω. This phenomenon in glass is interpreted as the first evidence of photoconductivity being dependent on the relative phase between light waves of different frequencies.

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