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ELECTRICALLY STIMULATED LIGHT-INDUCED SECOND-HARMONIC GENERATION IN GLASS: EVIDENCE OF COHERENT PHOTOCONDUCTIVITY

P.G. Kazansky and V. Pruneri

Optoelectronics Research Centre,

University of Southampton,

Southampton SO17 1BJ,

United Kingdom

Tel +44 1703 593083

Fax +44 1703 593149

Abstract

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.

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 via photoinduced spatially-oscillating electrostatic fields in glass was probably the first observed phenomenon where coherent photocurrent was involved [4-6]. 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 [7], HCl molecular beam [8] and between five- and three-photon processes in mercury vapour [9], 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. Firstly, 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 [10]. Secondly, 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 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.

Normally, second harmonic (SH) generation is forbidden in glass due to the inversion symmetry of the glass matrix. However, when a sample of glass or glass fibre (e.g. Ge-doped 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 [11]. In experiments on optical glass fibres strong amplification of a weak seeding radiation at frequency 2ω, generated inside a fibre as a result of magnetic dipole or quadrupole effects, is observed after launching only intense light at frequency ω (without external seeding) [12]. The explanation for the phenomenon of photoinduced second harmonic generation is based on 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 [4]. Indeed, the probability P of simultaneous ionization of defect site by two photons at frequency ω and one photon at frequency 2ω is given by:

$$P \sim |a_2 E_\omega E_\omega + a_1 E_{2\omega}|^2 = |a_2|^2 I_\omega^2 + |a_1|^2 I_{2\omega} + 2\mathrm{Re}\{a_1^* a_2 E_\omega E_\omega E_{2\omega}^*\} \qquad (1)$$

where E_ω and $E_{2\omega}$ are the fields amplitudes at frequency ω and 2ω respectively, I_ω and $I_{2\omega}$ are

the corresponding intensities and a_1 and a_2 are two complex coefficients which determine the weight of the different processes in the probability P. The first two terms, proportional to even powers of the electric fields, describe the photoconductivity σ:

$\sigma \sim |a_2|^2 I_\omega^2 + |a_1|^2 I_{2\omega}$

and the last term, proportional to odd power of the fields, describes the coherent photocurrent j_{coh} or the modulation for the angular distribution of photoelectrons:

$j_{coh} \sim 2\mathrm{Re}\{a_1^* a_2 E_\omega E_\omega E_{2\omega}^*\}$

Therefore the coherent photocurrent spatially oscillates with a period Λ determined by the refractive index mismatch between light waves at frequencies ω and 2ω:

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

where λ is the wavelength in vacuum of the light at frequency ω, $n_{2\omega}$ and n_ω are the refractive indices at frequencies 2ω and ω respectively. This photocurrent gives rise to an electrostatic (dc) field E_g (photogalvanic field): $E_g \simeq j_{coh}/\sigma$. The magnitude of the photogalvanic field E_g in glass is typically $\sim 10^4$-10^5 V/cm and this field can induce a modulation in the second order nonlinear susceptibility ($\chi^{(2)}$) via the third-order nonlinearity:

$\chi^{(2)} = 3\chi^{(3)} E_g \sim \cos 2\pi z/\Lambda$. Assuming $\chi^{(3)} \simeq 10^{-22}$ $(\mathrm{m/V})^2$ for silica glass, the amplitude 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 (refractive index mismatch), thus making the second harmonic process efficient. In nonlinear optics the process that allows such compensation is known as quasi-phase-matching.

The coefficients a_1 and a_2 in the expression of the probability P can depend on the pump and second harmonic intensities: $a_1 = a_1(I_\omega^2, I_\omega^3, I_{2\omega})$, $a_2 = a_2(I_\omega^2, I_\omega^3, I_{2\omega})$. The physical

interpretation of such dependence consists in the following: the ground level of the defect site in the one- and two-photon ionizing transitions can be populated from other defect levels, lower in energy and more densely populated. The last transition could be produced by one-photon (2ω photon or 3ω photon, generated as a result of the third harmonic generation) or two-photon ($\omega + \omega$ or $\omega + 2\omega$) absorptions.

It is possible to interpret the coherent photocurrent in glass in terms of electronic wave-functions interference [3]. Indeed, in centrosymmetric media one-photon transition is allowed between states of different parity and two-photon transition between states of the same parity. If the ground state for one- and two-photon transitions is the same, then the parity of the corresponding excited states should be different. The interference of electronic wave-functions of different parity in the continuum of states (conduction band) results in asymmetric wavefunction, leading to photocurrent. The orthogonality of the wave-functions in the excited state of the one-and two-photon transitions in centrosymmetric media leads also to the absence of modulation of the total cross section of ionization (coherent photoconductivity) in this process.

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

$$P \sim |a_2 E_\omega E_\omega + b_2 E_o E_{2\omega}|^2 = |a_2|^2 I_\omega^2 + |b_2|^2 I_{2\omega} E_o^2 + 2\mathrm{Re}\{a_2 b_2^* E_o E_\omega E_\omega E_{2\omega}^*\} \quad (2)$$

where the symbols used have the same meaning as for equation (1). All terms in this

expression are even powers of the electric field regardless 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)). Unlike in the process without dc electric field considered above, the modulated part of the probability leads to modulation of the total ionization cross section and hence to a corresponding modulation of the photoconductivity:

$$\sigma = \sigma_o + \sigma_{coh},$$

where

$$\sigma_o \sim |a_2|^2 I_\omega^2 + |b_2|^2 I_{2\omega} E_o^2$$

is the uniform part of the photoconductivity and

$$\sigma_{coh} \sim 2\mathrm{Re}\{a_2 b_2^* E_o E_\omega E_\omega E_{2\omega}^*\} \sim \cos 2\pi z/\Lambda$$

is the part of the photoconductivity being dependent on the relative phase between E_ω and $E_{2\omega}$ (coherent photoconductivity).

The ohmic current ($j = \sigma E_o$) induced by the applied electric field separates photocarries which accumulate at the boundaries of the illuminated region and screen the applied electric field E_o. The resulting internal electric field inside the glass E_c evolves accordingly to:

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

where τ is the dielectric relaxation time and ϵ is dielectric constant of glass. An approximate solution for E_c in the limit $\sigma_{coh} << \sigma_o$, $\tau = \tau_o = \epsilon/\sigma_o$ is:

$$E_c \simeq \sigma_{coh}/\sigma_o \, E_o\{1 - \exp(-t/\tau_o)\}\exp(-t/\tau_o) + E_o\exp(-t/\tau_o) =$$

$$= E_c^{coh} + E_c^{o}, \qquad (3)$$

where E_c^{coh} and E_c^{o} are the modulated and uniform part of the photoconductivity. The modulated part of the electric field, proportional to σ_{coh}, and the corresponding

observations on photoinduced SHG in Ge-doped fibres [11] and may be explained by the appearance of a modulated second-order nonlinearity ($\chi^{(2)}$ grating) in the fibre 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 fibre and launched into the fibre (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 a 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 grows rapidly, within 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" behaviour 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 $\sim 2\%$ 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 ~ 4 times smaller SH signal and ~ 2 smaller growth rate in a fibre with electrode superposition $\sim$ 10 cm long, thus two times shorter than in the previous fibre used.

We excluded a simple explanation of this phenomenon by the EFISH seeding of $\chi^{(2)}$ grating

in a fibre on the basis of two experimental observations. Firstly, 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. Secondly, no SHG signal was observed in any of the used fibres after preparation for more than 1 hour 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 (Fig.4) and time, necessary to reach half of the maximum (Fig.5), when the voltage is switched-on and switched-off, 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 fibre length represent clear evidences of quasi-phase-matched SHG due to presence of a $\chi^{(2)}$ grating in the fibre. Moreover the enhancement of SHG in glass due to a strong applied electric field ($\sim 5 \times 10^6$ 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 experimental situation where no electric field is applied to the fibre. 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 $\sim 8\%$ in the applied electric field.

Our experimental results can be qualitively interpreted in the 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 SH generation. 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. 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 equation (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 an index change due to Kerr effect, thus a phase mismatch between pump and SH waves together with a remaining weak $\chi^{(2)}$ grating. Initially, the SH signal quickly drops to zero as a result of this phase-mismatch. However due to the fact that the internal electric field increases almost to its initial value (with opposite polarity), the process of electrically stimulated growth of $\chi^{(2)}$ grating via coherent phoconductivity repeats itself for a second time.

In conclusion we observed electrically stimulated light-induced second harmonic generation in glass due to 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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Figure Captions

1. Cross-section of a Ge-doped silica fibre with internal electrodes. The fibre had 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 fibre over length varying from 5 mm to 500 mm.

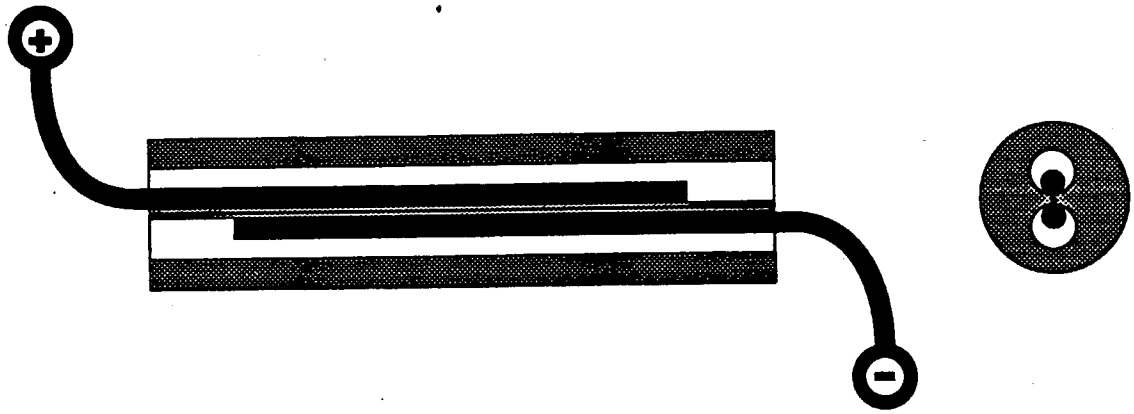
2. Time dependence of SH signal in a fibre with external SH seeding of $\sim$40 μW average power (opened triangles). The seeding was launched at t =0. The SH growth was monitored by blocking the SH seeding. Time dependence of SH signal in a fibre with applied voltage of 5 kV (filled triangles). The voltage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

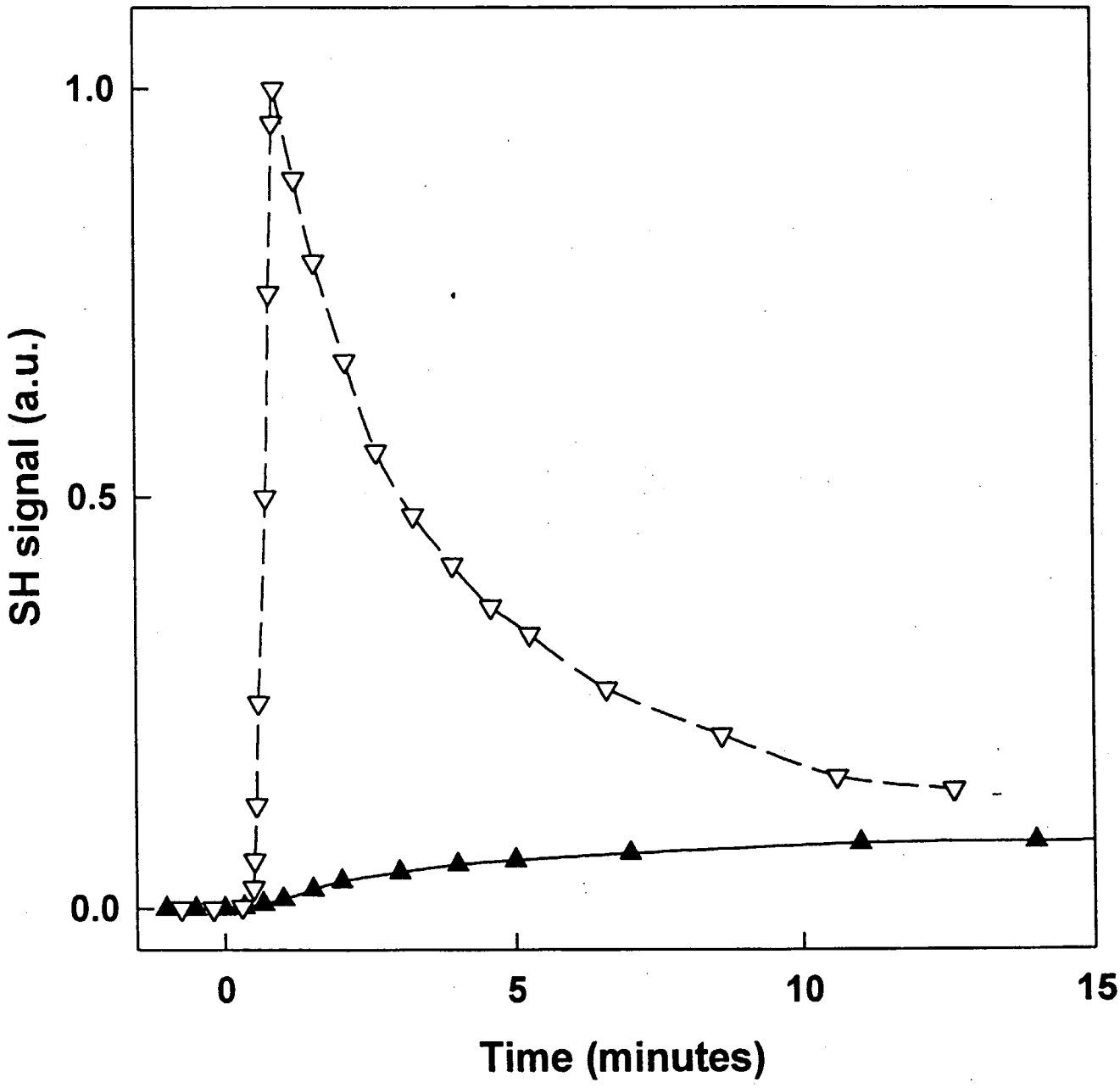
3. Time dependence of SH signal in a fibre when an applied voltage of 5 kV was repeatedly switched on and off. The instants when the voltage is on and off are shown by arrows.

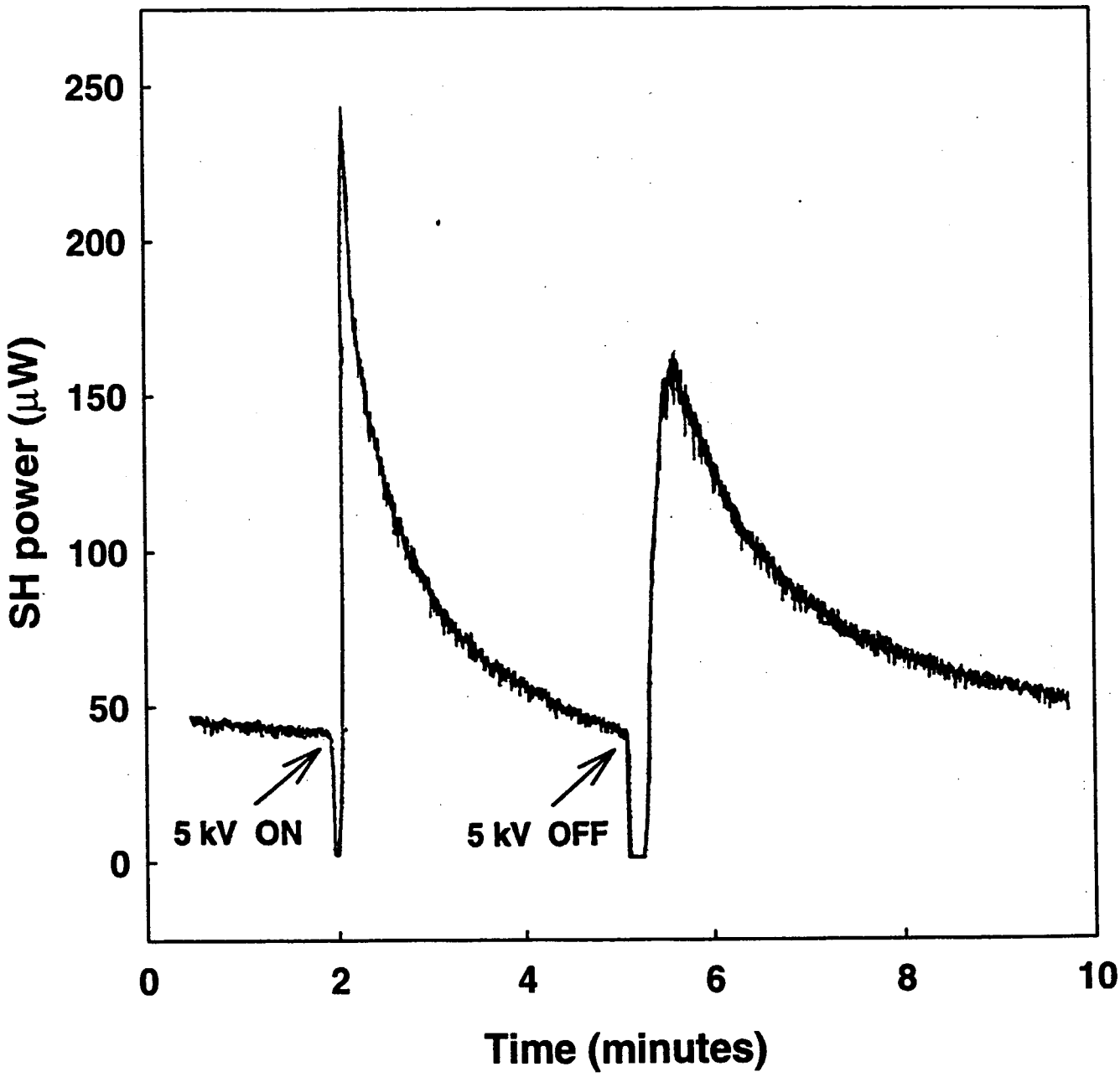
4. Dependence of the maximum SH signal on applied voltage when the voltage is switched ON and OFF respectively.

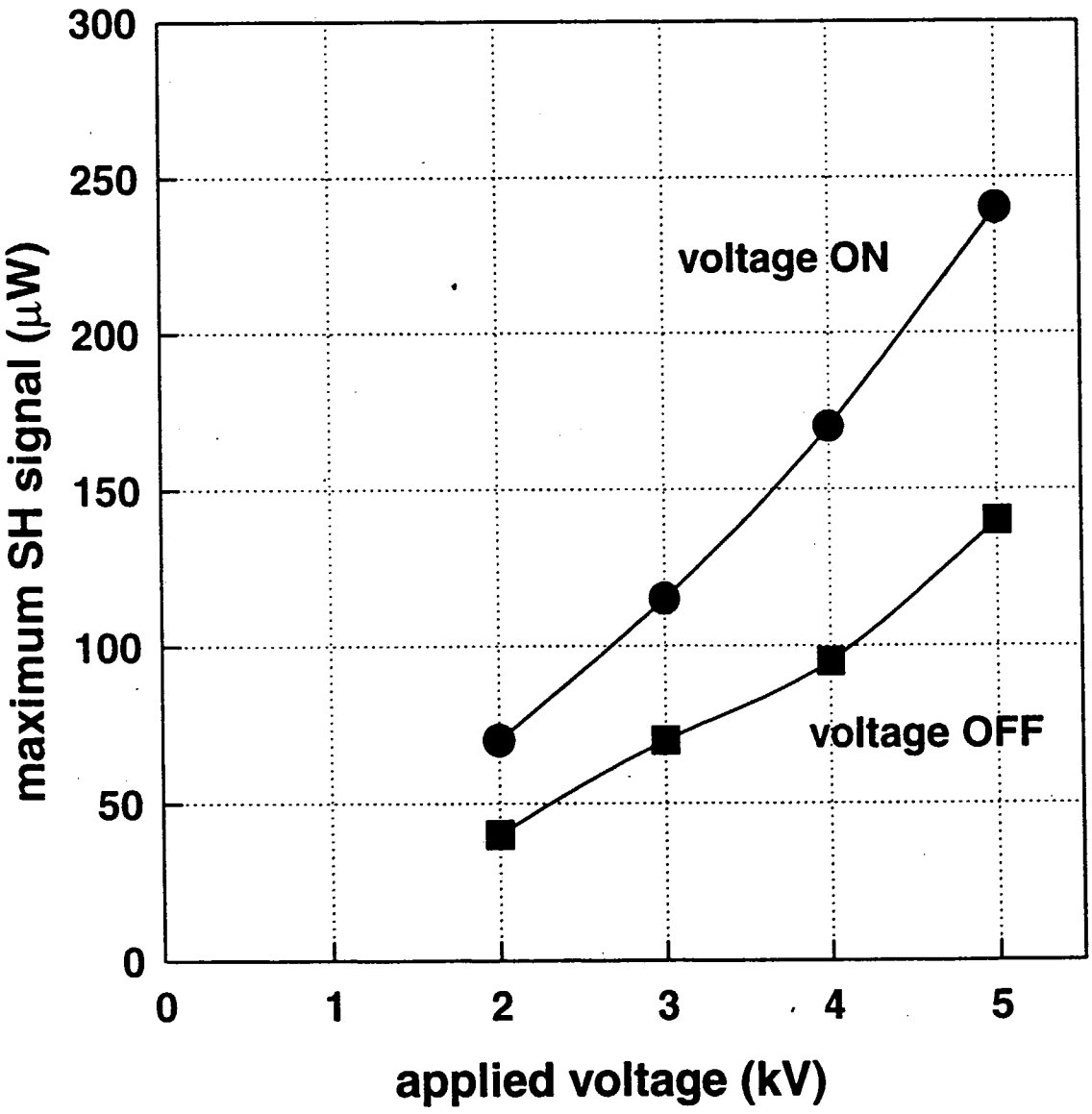
5. Dependence of the time necessary for reaching half of maximum SH signal on applied voltage when the voltage is switched ON and OFF respectively.

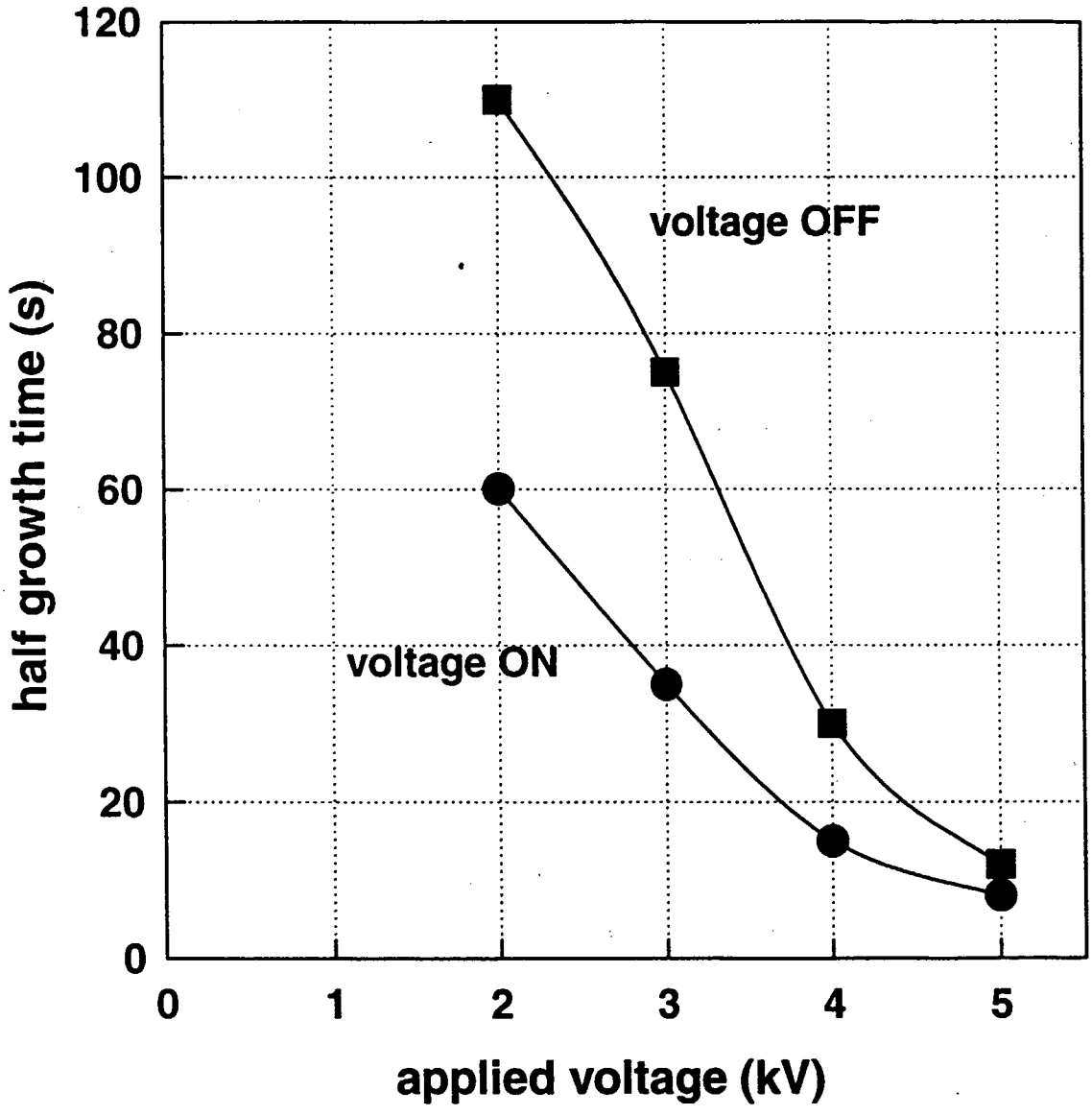
P.G. Kazansky and V. Primeri "Electrically stimulated light-induced second-harmonic generation in glass ...", FIG.1




1.0
0.5
0.0
SH signal (a.u.)
0
5
10
15
Time (minutes)


250
200
150
100
50
0
SH power (μW)
5 kV ON
5 kV OFF
0
2
4
6
8
10
Time (minutes)


300
250
200
150
100
50
0
maximum SH signal (μW)
voltage ON
voltage OFF
0
1
2
3
4
5
applied voltage (kV)


120
100
80
60
40
20
0
half growth time (s)
voltage OFF
voltage ON
0
1
2
3
4
5
applied voltage (kV)