FUNDAMENTALS OF GLASS POLING: FROM SELF-ORGANIZATION TO ELECTRIC-FIELD POLING

Peter G. Kazansky and Valerio Pruneri

One decade has passed since the discovery of self-organized (photoinduced quasi-phase-matched) second-harmonic generation (SHG) in optical fibers [1-2]. This discovery has attracted considerable interest world-wide [3-16] due to the unusually strong $\chi^{(2)}$ (second-order susceptibility) gratings induced purely by optical fields in glass (10^{-15}-10^{-16} m/V, which were 4-5 orders of magnitude higher than one could explain by known physical processes). In 1991 the value of $\chi^{(2)}$ has been increased to a new level of 1 pm/V by thermal poling [26]. In this paper we discuss fundamentals of glass poling, trying to answer the question: What is the limit of a second-order nonlinearity in glass?

The mystery of photoinduced $\chi^{(2)}$ gratings was finally solved on the basis of a new phenomenon - the coherent photogalvanic effect [6]: quantum interference between the one- and two-photon ionization processes of defect states changes the angular distribution of photoelectrons and excites a phase dependent current (*coherent photocurrent*); this current gives rise to a spatially-oscillating electrostatic field ($E_g \simeq j_{coh}/\sigma$, where E_g is the photogalvanic field, j_{coh} is the *coherent photocurrent*, σ is the photoconductivity); the amplitude of the photogalvanic field E_g in glass is typically $\sim 10^4$-10^5V/cm and this field can induce experimentally observed values of $\chi^{(2)}$, via the third-order susceptibility: $\chi^{(2)} = 3\chi^{(3)} E_g$ (assuming $\chi^{(3)} \simeq 1.8\times10^{-22}$ $(m/V)^2$ for silica glass).

More recently, the interference between different quantum processes has been the subject of considerable attention in many areas of physics (e.g. electromagnetically induced transparency (EIT) and lasers without inversion are based on quantum interference [17]). 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 and chemical processes - not only by the intensity or the polarization of light, but also by the phase of light. *Coherent photocurrent* was observed in the experiments on rubidium atoms [18] and photoemission from Sb-Cs photocathodes [19] and in AlGaAs/GaAs quantum well superlattices [20]. However self-organized SHG in glass was the first observed phenomenon where *coherent photocurrent* was involved.

It is possible to interpret the *coherent photocurrent* in glass in terms of electronic wave-functions interference. 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 [20]. 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 (involving odd number of photons).

However, until recently, two interesting aspects of the quantum interference phenomenon have not been 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 [21]. Secondly, the modulation of the total cross-section of ionizing transitions due to quantum interference involving

even number of photons (*coherent photoconductivity*) has been observed only in atomic systems [22-24]. Recently, we reported the observation of efficient SHG in glass subjected to a strong external electrostatic field [25]. 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. We achieved $\sim 2\%$ conversion efficiencies for peak powers of $\sim$ 1 kW. These values give an amplitude of $\chi^{(2)} \simeq 10^{-14}$m/V, which corresponds to a modulation of $\sim 8\%$ of the applied electric field ($\sim 6 \times 10^6$ V / cm). The observed phenomenon represents the first evidence of *coherent photoconductivity* in solid state materials.

On the other hand during the past six years a number of glass poling techniques have emerged that produce permanent second-order nonlinearities (independent of the presence of optical fields of ω and 2ω) approaching 1 pm/V (very close to the value of nonlinearity in LBO, which is a crystal widely used for pulsed frequency conversion). These new poling techniques are: thermal poling at 250-300°C under an applied high voltage (the second-order nonlinearity appears in a thin layer just under the anode) [26], corona poling of glass waveguides [27] and charge implantation by exposure to an electron-[28] or proton-beam [29]. Most recently an electro-optic coefficient of ~ 6 pm/V has been reported in silica fibre poled under UV excitation [30]. However the latter result has not been confirmed by other groups. So far thermal poling is the most promising technique for practical applications: nonlinearity is strong enough and shows no degradation under illumination with intense visible and infrared light [31-50]. It was suggested that a high dc field ($\sim 10^7$ V/cm) in a thin region ($\sim$5-10 μm) depleted of cations under the anodic surface is responsible for this phenomenon [26]. This field can be frozen-in between the two layers of space charge: negatively charged depletion region and positively charged layer created as a result of ionization or diffusion in the high field between the depletion region and the anode [37, 44]. Charged distributions obtained in poled glasses by the laser induced pressure pulse (LIPP) [45] and the etching techniques [46] are in good agreement with this model. More complicated charge distributions (with the inverted charge regions) were also observed by the LIPP technique in poled samples [45]. It is still not clear whether $\chi^{(2)}$ is induced by high dc field via $\chi^{(3)}$ or orientation of dipoles. Indeed with the maximum dc field limited by the intrinsic breakdown of silica ($\sim 10^7$ V/cm) it is possible to justify the values of $\chi^{(2)} \leq .54$ pm/V via $\chi^{(3)}$ of silica. However about two times higher values have been measured experimentally. This discrepancy may have different explanations: by uncertainty in the thickness and distribution of the nonlinear layer, by a higher $\chi^{(3)}$ value in the poled layer, or by orientation of dipole defects (taking into account the number of defects available in silica glass for the latter to be true the hyperpolarizability of each defect must be unrealistically high).

New ideas and experiments are necessary to justify $\chi^{(2)}$ values $\sim$ 1pm/V and higher. A better understanding of the mechanisms of glass poling, which are still not fully understood, may help to improve the value of second-order nonlinearity in poled glasses, perhaps to values competitive with the best ferroelectric crystals.

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