MECHANISMS OF ENHANCEMENT OF NONLINEAR OPTICAL RESPONSE IN NANOSTRUCTURED III-V COMPOUNDS

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It is well known that III-V compounds possess second order nonlinear optical coefficients several orders of magnitude higher than those of KDP, ADP and other materials used traditionally in frequency up-conversion. However, the utilization of large nonlinear susceptibilities of III-V compounds has not been possible due to high dispersion and lack of birefringence necessary for phase matching [1]. Electrochemistry proves to be a powerful tool for introducing the necessary optical anisotropy in semiconductor materials. Birefringence as high as $n_e - n_o = 0.25$ was measured recently [2] in porous GaP membrane with the degree of porosity close to 40 %. This birefringence was shown to meet the phase matching conditions for the second harmonic generation in III-V materials [3,4]. Another important effect of nanostructuring III-V compounds is the enhancement of nonlinear optical properties. Second harmonic generation (SHG) efficiency two orders of magnitude higher than that of bulk material was reported for porous GaP membranes [3-7]. A significant enhancement in both the radiated terahertz field and second harmonic radiation from the porous InP surface, relative to the bulk InP surface, was recently observed [8,9]. The enhancement factor is currently the subject of investigation, and has been attributed to such processes as local field enhancement within the porous network [3-5,8,9], and increased interaction of the radiation with the porous network due to scattering [6,7]. The analysis of the SHG experimental conditions for the InP porous samples excited by the 800 nm radiation wavelength suggested that the effect of increased interaction of the radiation with the porous network due to scattering is minor in comparison with local field enhancement [8]. For the case of THz emission, there is also virtually no scattering of the THz radiation as the wavelength in this case is much larger than the characteristic sizes of the porous skeleton entities.

As concerns the mechanism related to local field enhancement, using the special isotropic structure model of Bruggeman [10] for a non-linear metal-insulator composite, Bergman [11] was able to prove that fourth order fluctuations $\left\langle E^4 \right\rangle_{metal} / E_0^4$ even can diverge. Repeating this analysis for the dielectric case $\sigma_I = 0 \longrightarrow \varepsilon_1$, $\sigma_M = \longrightarrow \varepsilon_2$ with $\frac{\varepsilon_2}{\varepsilon_1} \geq 10$, the fourth order fluctuations diverge, too.

These divergences are connected with and due to a so-called percolation threshold, a relative concentration $0 < f_0 < 1$ at which the σ_M - resp. the ε_2-material forms connected paths through the sample. All simple structure models without such a percolation threshold do not result in large fourth order fluctuations. The Bruggeman model is based on spherical inclusions of both materials in a self-consistently calculated effective matrix.

For the experiments with SHG, an enhanced second order non-linearity requires strong third order field fluctuations. It was suggested, that the fields near sharp edges can result in large third order fluctuations, too, therefore explaining the SHG efficiency enhancement in the case of triangular-prism like pores [3,4]. However, this model does not explain the enhancement of SHG efficiency in the case of circular pores. Apart from that, the percolation model is developed for direct current experiments in highly disordered media. The experiments with SHG are related to optical frequencies, and some of the investigated porous structures are characterized by a high degree of order.

The goal of this paper is to propose a new mechanism of nonlinear optical properties enhancement in nanostructured semiconductors based on the concept of photonic crystals (PC). The photonic crystal is usually formed as a structure consisting of two or more optically transparent periodically arranged media. It is to be noted that not all photonic applications require a strictly ordered distribution. PC properties are also inherent for structures with a quasi-uniform distribution, called amorphous PC. Short range order in porous III-V is easily achieved with electrochemical dissolution. Long range order is also possible with appropriate electrochemical parameters. Single crystals of pores have been obtained by means of self organization processes in n-InP [12,13].

We have calculated the PC properties of two porous structures with circular and square pores as illustrated in Fig. 1. The results of PC band structure calculation for these porous structures are presented in Fig. 2. One can see from Fig. 2 the existence of van Hoove singularities in the photonic band structure at specific frequencies. Notomi has shown [14]

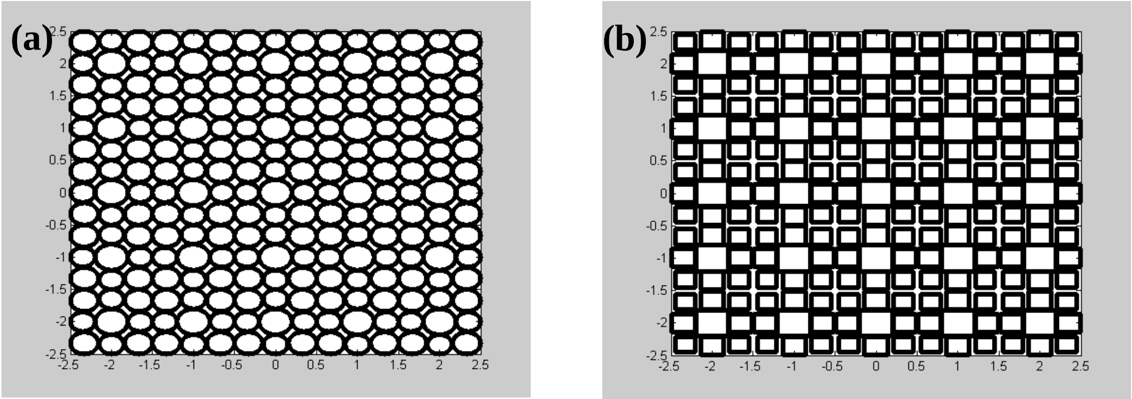

Fig. 1. Porous layers with circular (a) and square (b) pores.

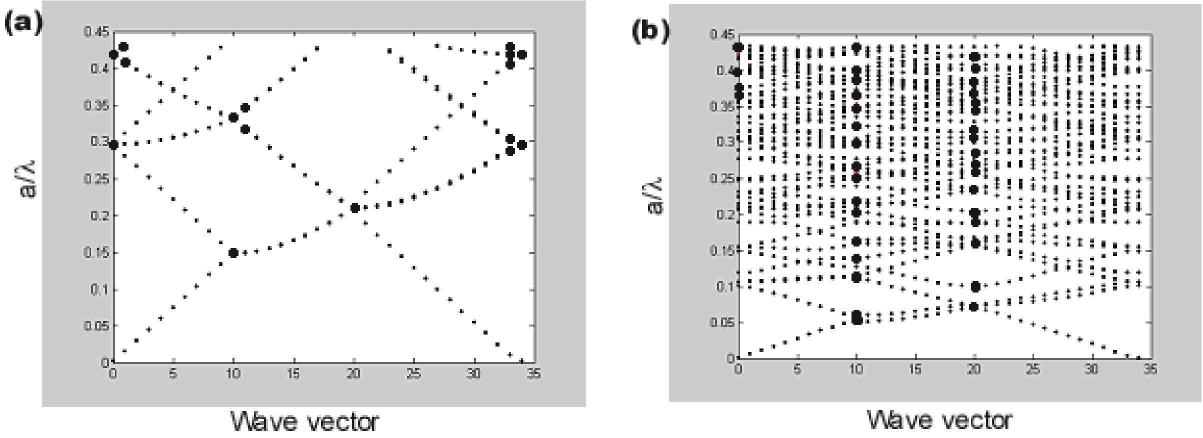

Fig. 2. PC band structure calculation for porous layers with circular (a) and square (b) pores.

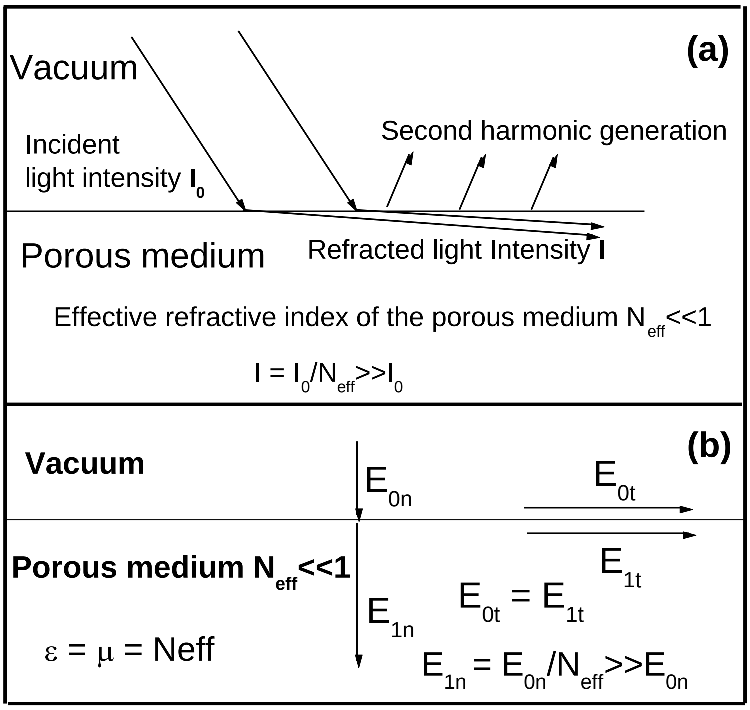

Fig. 3. Enhancement of the radiation intensity (a), and the normal component of the electrical field (b) inside the porous sample

that at this specific frequency the PC can be ascribed an effective refractive index less than 1 or even negative values.

Therefore, if one assume that in some frequency ranges the effective refractive index of investigated porous structures can be ascribed an effective refractive much less than 1, then according to Fig. 3 one can explain the enhancement of the radiation intensity, and the normal component of the electrical field inside the porous sample, which results in turn in the enhancement of nonlinear optical properties, including the SHG efficiency. The bold dots in Fig. 2 show the frequencies at which, according to calculations, there is an enhancement of the SHG efficiency comparable with the experimentally observed one.

On the basis of the PC approach one can explain also the higher sensitivity of the SHG intensity to the polarization in porous semiconductor structures as compared to the bulk material observed in the experimental investigations illustrated in Fig. 4.

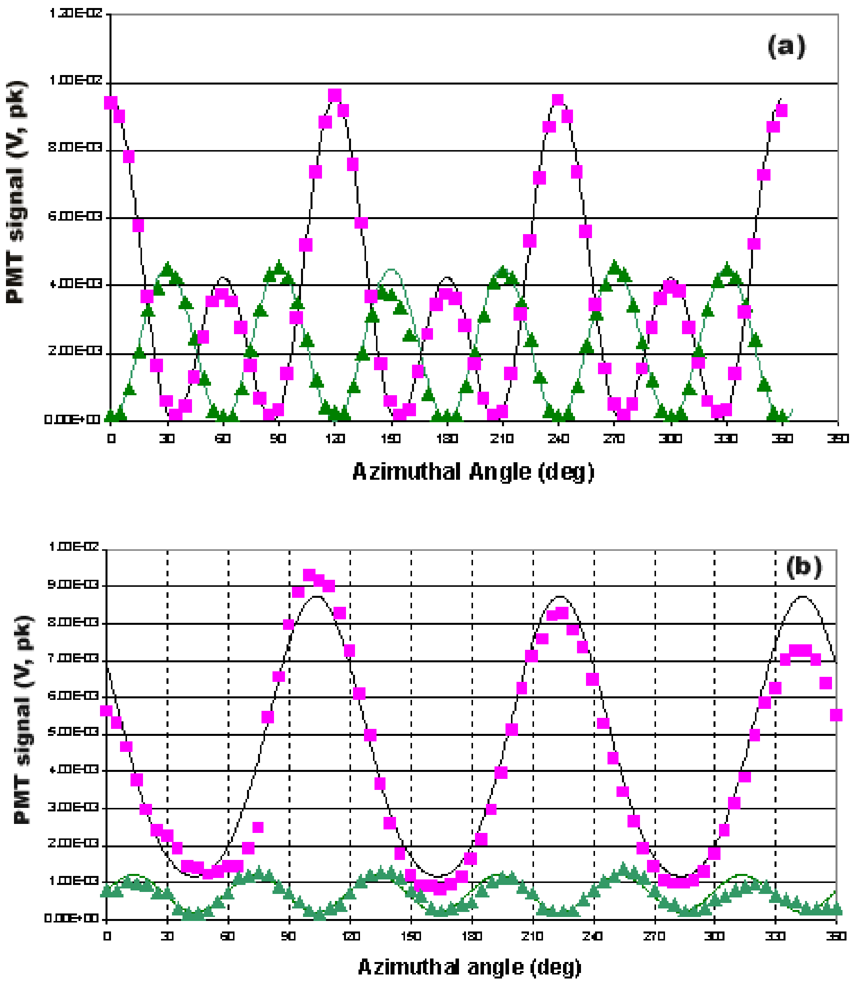

Fig. 4. SHG intensity in bulk (a) and porous (b) InP samples measured in p-p (squares) and p-s (triangles) polarizations.

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