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Enhancement of the Performance of Bulk and Nano-structured Crystalline Materials for Control of Laser Radiation

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In this paper we focus on improvements of the efficiency of optoelectronic devices for nonlinear optical (NO) and electro-optical (EO) control of laser radiation. This can be done while improving performance of active solid-state crystalline elements of the NO and EO cells, which are made of specialized dielectrics and III-V semiconductors that comprise both bulk- and nano-structures. Enhancing of the NO and EO interactions in the bulk and nano-structured materials is achieved basing on novel techniques for 3D spatial analysis of the crucial parameters of those optical effects. Moreover, further enhancement of the NO parameters of III-V semiconductors can be achieved due to improvement in the spatial homogeneity of quantum dots and quantitative control of their characteristics. The latter should enable comprehending the effects of size, shape and density of nanoscale crystals.

Keywords: Electro-optics, Nonlinear optics, Anisotropy, bulk and Nano-structured dielectrics and semiconductors, Control of laser radiation, Optoelectronic devices.

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1. INTRODUCTION

Many crucial components of optoelectronic devices and systems are based on nonlinear optical (NO) and electro-optical (EO) effects (see, e.g., [1–7]). The former include devices related to light-emitting sources, optical frequency conversion (optical parametric oscillators, parametric amplifiers [8, 9] and optical harmonics generators [10]), optical Kerr effect, optical switching, self-phase modulation, laser mode-locking, memory cells, photovoltaic cells, etc.

The latter devices facilitate control of the intensity and phase of light beams, e.g. in multi-channel laser systems, where electrical energy modulates the incident laser beam [11, 12]. In view of an industrial importance, the research work aimed at finding out new efficient optical materials for such purposes or increasing the performance of the existing materials seems to be urgent (see, e.g., [13, 14]).

Until now, only 'extensive' methods for improving the efficiency of NO or EO cells, consisting in going over different optical materials, has been used the most often. Moreover, there have been a few technologies which could considerably improve this efficiency at the stage of design of the optical cells, one of which being switching over to quantum-confined structures. However, this most common way for achieving better performance, i.e. searching for newer materials with the best characteristics, does not represent the cheapest and the fastest method.

In the present report we would like to describe two novel and interrelated ideas which should solve the problem.

2. WORKING IDEAS AND TECHNIQUES

2.1 Employing of spatial anisotropy of NO and EO parameters

The first idea described in this paper relies upon anisotropic (tensorial) character of NO and EO properties. It can ensure utilization of anisotropic crystalline materials in the most efficient manner and, moreover, provide better stability of their performance characteristics (see [15]). The method exploits comprehensive 3D analysis of spatial anisotropy of practical NO and EO characteristics in a given material and often enables achieving far better performance with the same crystal. In other words, most of (either already approved or new) crystalline materials still have some hidden resources, which have yet not been used and even studied quantitatively.

Crystalline materials mainly exhibit essential spatial anisotropy of their optical properties induced by external fields. We have already extensively studied the spatial anisotropy of some of these effects (e.g., piezo-optic [16–21], EO [22–24], elastic, photo-elastic [17, 21, 25–27] and acousto-optic [25, 28–30]) for a large variety of crystalline materials. It has been shown that, in general, the global maxima of these field-induced effects do not correspond to the principal (crystallographic) axes [16, 17, 23, 24, 29, 30]. One of the relevant examples is illustrated in Fig. 1. The so-called indicative surfaces shown in Fig. 1 illustrate well spatial sensitivity of the EO effect in widely used lithium niobate, $LiNbO_3$. It is evident that these crystals [16, 23, 29], along with the other well-known materials (e.g., β-BaB_2O_4 [17] or SiO_2 [21]), are not being used in

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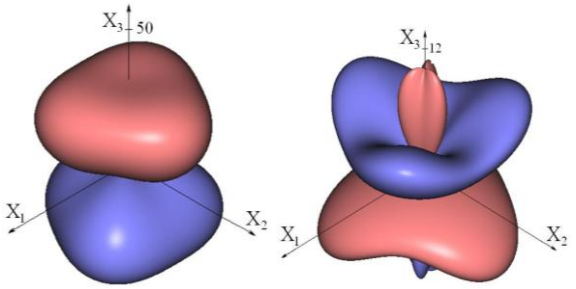
a proper manner, i.e. the corresponding sensitive crystalline elements for control of laser radiation could have, in principle, reached far higher performance.

As an example, the maximal electrically induced optical path difference in $LiNbO_3$ is almost 3 times larger (at $\theta = 54°$, $\varphi = 90°$ [23]) and the extreme acousto-optic figure of merit is 2.4 times higher (at $\theta = 60°$, $\varphi = 7°$; isotropic light diffraction [29]), as compared to the corresponding values typical for the standard geometries of direct crystal cuts typically used in optoelectronics. Indeed, the most efficient geometries corresponding to the global maximums of the piezo-optic effect (the angular spherical coordinates $\theta = 42°$, $\varphi = 30°$ and $\theta = 49°$, $\varphi = 30°$) provide almost 5 and 4 times higher efficiencies respectively for the lithium niobate [16] and the beta barium borate [17] used in piezo-optic cells and piezoelectric transducers.



a) b)

Fig. 1 – Indicative surfaces for the longitudinal (a) and transverse (b) geometries of EO effect in $LiNbO_3$ crystals (all the values are in pm/V) [23]

Notice that all the parameters cited are amplitude ones, while the energy consumption of any device is governed by the power-related parameters, with still higher gains. Rough estimates of potential improvement to the present capabilities based on our previous experience show that our approach used for the novel materials should prove high-payoff.

Summarizing, we suggest a novel optimization technology capable of providing notable increase in the NO and EO efficiencies for both new and well-known optical materials, which can be as large as order of magnitude. It is based upon the analysis of spatial anisotropy of the optical effects for the bulk, thin-film or nano-sized crystalline materials, relying on geometrical interpretation of the anisotropy of physical effects described by tensors of the ranks three (the EO Pockels and the three-wave NO effects) and four (four-wave NO parameters, including a nonlinear refractive index).

A tentative outline of such method includes accurate experimental measurements of the magnitudes and the signs of all the tensor components required [18, 19, 22, 27], along with the appropriate supplementary studies in need [27, 31–33]. The next step is constructing the indicative (see, e.g., [16, 17, 23, 29]) or the extreme (see [30]) surfaces of the tensors and their stereographic projections, which enable calculations of the extreme values of the effects under study, using all the known tensor components. The appropriate software (e.g., for constructing 3D surfaces, calculating their stereographic projections, etc.) has already been developed by the authors. Then the extremes of the EO or NO parameters and the corresponding optimal experimental geometries are being revealed, providing a basis for designing the devices with the improved performance.

Notice that all crystals, including cubic ones, should necessarily reveal anisotropy of higher-order optical effects and so in fact represent possible subjects of our analysis. However, one of the challenges here is that the three-wave NO phenomena are already 'improved' in the sense that the commonly used experimental geometries rely upon the phase-matching effect, the latter being geometrical 'improvement' based on the refractive-index anisotropy. Therefore we should thoroughly analyze both the conditions associated with the maximum NO coefficients and those linked with the phase matching. Of course, there is a risk that the above conditions can appear to be contradicting for some of the materials, thus partly reducing the gain. We will confine the objects of our investigation only to model materials and those of the greatest practical impact. These are advanced optoelectronic materials, including multifunctional dielectric materials (lithium niobate and borate-family crystals, KTP, etc.) and technologically important III-V and, partly, II-VI semiconductors (first of all, GaAs-related), which are the basis for modern electronics and optoelectronic integration. For the case of dielectric materials under test, one should additionally study the possible gain obtained while considering spatial anisotropy of their piezoelectric properties.

2.2 Fabrication and investigation of spatially uniform quantum dots for NO applications

Our second idea is to achieve a considerable gain in the third-order NO characteristics via production of much more spatially uniform nanoscale crystals (quantum dots – QDs) and studies for the influence of their size, shape, and density on those characteristics. At the telecom wavelength (1.55 μm), typical bulk semiconductors have poor NO characteristics (e.g., the NO refractive index for GaAs is 1.5×10^{-13} cm²/W [34, 35]) that falls far short of the strong optical Kerr effect (~ 10^{-10} cm²/W) needed to produce attractive index changes ~ 10^{-2} to 10^{-4} with the MW/cm² intensities. Yet, since these semiconductor materials are exactly what is used in today's electronic platforms, optical devices made of compatible materials are needed if they are to be integrated as active elements.

A well-known approach is making use of low-dimensional semiconductors. Quantum confinement in one or more dimensions (quantum wells, wires or QDs) of the conduction-band electrons can change the NO response of a semiconductor in a number of ways [36–39]. In spite of this, the expectations of dramatically large enhancement in the NO coefficients for the QDs have not been clearly and consistently demonstrated. Besides of the overlap integral and the dipole transition strength, in practice there can be many factors that additionally affect the optical response of the QDs (the binding energy of excitons, thermal energy and influence of temperature, line-widths and non-radiative lifetime, coherent interaction length, availability or absence of resonant coupling of the laser beam to the excitons in all of QDs, fill-factor, surface and interface states, etc. [40–44]). The results reported in the literature vary noticeably and even comparison of the nonlinearities of low-dimensional

structures with those of the bulk is potentially not a simple question. Even if the comparison were correct, a further issue of importance is the question whether the enhancement is really due to confinement of the wave function (at least as a dominant mechanism) or due to, e.g., the effect of interfaces.

As a consequence, our idea is focused on the fundamental issues related to the impact of geometrical factors at the nanoscale on the NO properties. Here the efforts should be aimed at the role of size, shape, and density of nanoscale crystals on enhancing the NO parameters. Moreover, it seems interesting to survey the possibilities for influence of crystallographic orientation of the substrates on the properties of QDs (see Subsection 2.1). To succeed in these directions, however, we should at first solve a technological problem, a demand for highly uniform nanostructures under test. The usual approach to fabricate QDs is a Stranski–Krastanov growth mode [45, 46], where the deposited material has a slightly larger lattice constant than the substrate (e.g., InAs is deposited on a substrate of GaAs or AlAs). As one might expect, the size, shape, and position of the self-assembled 3D islands would depend on the growth parameters and statistical variations of the substrate surface, all of which lead to a distribution of 3D islands. Unfortunately, the QDs though coherent are typically observed to be significantly non-uniform in size, shape, and position [47–51], while achieving control to better than 10 to 20 per cent remains a considerable challenge, in spite of that the relevant improvement can yield really creative NO materials.

As a solution, we suggest delivering an order-of-magnitude improvement in the homogeneity of QDs. The growth technique called as a 'directed droplet epitaxy' (see Fig. 2) places uniform droplets of indium at precise locations before they are crystallized into the InAs QDs. This droplet delivery technique relies on the parameters like temperature, voltage, current, and other things we can control, rather than stochastic processes.

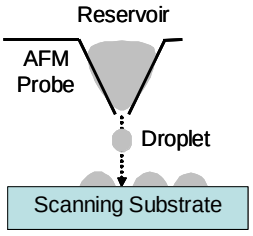


Fig. 2 – Illustration of directed droplet epitaxy

In other words, our idea is based on:

(1) our preliminary experience with the droplet epitaxy (we have certainly gained a good understanding of the role of various parameters for this growth mode);

(2) our experience with the STM and AFM scanning probe systems, allowing us to explore new droplet growth approaches that depend on scanning techniques to deposit droplets, and to evaluate the progress towards homogeneity;

(3) our preliminary studies and those by the other authors on controlling the deposited droplet volume and position [52–54]. Every bit of data we have taken or examined encourages us to investigate the possibility of depositing uniform Ga or In droplets.

Hence, we expect improving in the homogeneity of formation of the QDs by an order-of-magnitude (the homogeneity of size, shape and positioning up to 1–2 per cent). The feasibility of this idea is supported by the preliminary studies using a test probe-tip, which shows a droplet height uniformity of 2 per cent. Using these QDs of different sizes, shapes, and densities, one can systematically determine their role on enhancement of the NO coefficients.

3. DISCUSSION AND CONCLUSIONS

In this work we have focused on breakthrough improvements of the efficiency of optoelectronic devices for NO and EO control of laser radiation. We expect to achieve this owing to far better performance of active solid-state crystalline elements of the NO and EO cells, which are made of specialized dielectrics and III-V semiconductors, comprising both bulk- and nano-structures. The principal ideas of our approach are as follows:

(1) Enhancement of the NO and EO interactions in both bulk and nano-structured materials for optoelectronics, basing on novel techniques for 3D spatial analysis of the crucial parameters of those optical effects.

(2) Enhancement of the NO parameters of III-V semiconductors, achieved due to an order-of-magnitude improvement in the spatial homogeneity of quantum dots (QDs) and quantitative control of the QD characteristics.

Implementation of our main ideas should result in both new understanding of different underlying physical phenomena and factors that control NO and EO parameters and important technologies that ensure a breakthrough in the efficiency of NO and EO cells.

In particular, applying of our techniques would provide significant decrease in the half-wave voltage for the EO devices and higher frequency conversion efficiency, therefore yielding essentially lower power consumption of the devices at the industry scale. There is another important benefit expected. Very often, the needed parameters of the devices are reached at the risk of exceeding radiation damage threshold of a given material. At the same time, implementation of our ideas should enable use of crystalline materials under lower light intensities, with no losses in the performance.

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