Study of nonlinear optical phenomena in classical and quantum regime

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1. INTRODUCTION

We performed different interesting studies on nonlinear optical phenomena in the last year. From the point of view of materials fabrication, optically nonlinear AlN/GaN samples were grown despite the large lattice mismatch of the two different nitrides. An experimental set-up has been developed, which allows delay-time experiments in PC media; moreover, by using Quasi Normal Mode theory, a theoretical analysis was applied to allow the coherent control of the stimulated emission inside a one-dimensional PC.

2. THEORETICAL ANALYSIS OF STIMULATED EMISSION INSIDE 1-D PBG

A theoretical analysis was applied to allow the coherent control of the stimulated emission inside a one-dimensional photonic crystals. The Quasi Normal Mode (QNM) theory was adopted in order to discuss the quantum problem of an atom embedded inside a one dimensional (1D) Photonic Band Gap (PBG) cavity pumped by two counter-propagating laser-beams. The novel is that the decay-time of the exited atom in the cavity depends on the position of the atom inside the cavity itself and can be controlled by the phase-difference of the two pumping laser-beams. Such a system might therefore be relevant for a single-atom, phase-sensitive, optical memory device on atomic scale.

The behaviour of small systems coupled to dissipative reservoirs represents a central theme in many physical contexts. In quantum optics the problem consists of a two-level excited atom decaying in open space to its ground state through an allowed electric dipole transition. Spontaneous emission depends not only on the properties of the excited atomic system but also on the nature of the environment to which the system is optically coupled. It is possible to control the rate of spontaneous emission by altering the density of electromagnetic modes near the resonant frequency. An important situation arose when it was realized that it is possible to create environments in which the spectrum of the electromagnetic field exhibits gaps in frequency like in PBG materials. The photon density of states (DOS) is the fundamental feature that determines the behaviour of the system ‘atom-field’. Strong modification in the DOS can be effected by means of photonic crystals. Gaponenko et al [1] have reported on modifications of the emission processes of dye molecules embedded in a three-dimensional solid-state photonic crystal exhibiting a stop band in the visible range. Our approach uses a realistic model for the photonic crystal, as a finite cavity with discontinuities in the refractive index, so our approach improves the results previously obtained in literature. The e.m. field is quantized in terms of the QNMs in the 1D-PBG and the atom is modelled as a two levels system. In the electric dipole approximation, the atom is assumed to be weakly coupled to just one of the QNMs. In the free space, the two counter-propagating pumps are tuned at the frequency $\omega$ and prepared in the quantum state $|\psi_0\rangle = |\psi(t = 0)\rangle$. The initial state $|\psi_0\rangle$ coincides with a coherent state, while the QNMs are a discrete set of couples $[\omega_n, f_n^N(x)]$ and form an orthogonal basis only inside the open cavity according to complex metrics.

FIG. 1: (a): Decay-time at the low-frequency band-edge. (b): Decay-time at the high-frequency band-edge.
After some algebra, it results that \( \tau_n^{(B)}(x_0) \approx \frac{\tau_n^{(A)}(x_0)}{\sqrt{1 + (-1)^n \cos \Delta \varphi}} \) where \( A \) refers to the case of spontaneous emission while \( B \) refers to the stimulated one. This verifies that the decay-time of the dipole depends from the position of the dipole inside the cavity, and can be controlled by the phase-difference of the two laser-beams. After performing calculation, the decay-time for stimulated emission and the dwell-time for the two laser-beams are compared on different scales as functions of the phase-difference between the two laser-beams. In the low-frequency (high-frequency) band-edge, the decay-time ratio is rising (slopes down) and so the dwell-time ratio is zero when the phase-difference of the two laser-beams is increasing from \( \Delta \varphi = 0 \) to \( \Delta \varphi = \pi \); in fact, the decay-time [see Figs. 1.(a) and 1.(b)] tends to the maximum when the laser-beams are going opposite of phase. Then, in the low-frequency (high-frequency) band-edge, the decay-time ratio tends to infinity and so the dwell-time ratio is null when the phase-difference of the two laser-beams is \( \Delta \varphi = \pi \) (\( \Delta \varphi = 0 \)); in fact, the dipole is not coupled to the QNM corresponding to the low-frequency (high-frequency) band-edge when the two laser-beams are (opposite in phase) in phase.

Since the decay-time depends from the position of the dipole inside the cavity, and can be controlled by the phase-difference of the two laser-beams, such a system might therefore be relevant for a single-atom, phase-sensitive, optical memory device on atomic scale.

3. SECOND ORDER NONLINEARITY OF AlN AND GaN FILMS

We investigated the second order optical nonlinearity of aluminium nitride and gallium nitride films grown by sputtering technique and by MOCVD on different substrates. Second-harmonic generation both from single films and multilayer structures was measured as a function of the incidence angle. Measurements were performed by means of the rotational Maker fringes technique for different polarization configurations, thus allowing the determination of the \( \chi^{(2)} \)-components of the second order susceptibility at the fundamental wavelength of 1064 nm. Preliminary linear optical characterization of the films was carried out by spectrophotometric optical reflectance measurements, thus the dispersion laws for both ordinary and extraordinary refractive index were retrieved.

III-Nitride semiconductors (such as GaN, AlN and their alloys) are meeting increasing interest for the fabrication of optoelectronic devices operating in the visible and near ultraviolet ranges, due to their wide band gap at room temperature. Their non-centro symmetric crystalline structure gives rise to second order nonlinear optical response (via the third rank \( \chi^{(2)} \) tensor) that can be enhanced in heterostructures like AlN/GaN multilayer stacks. Nevertheless, the AlN/GaN material system still represents a challenging task, due to the low contrast in refractive indices (\( n_{\text{AlN}} = 2 \), \( n_{\text{GaN}} = 2.4 \)) that is achieved together with a very large lattice mismatch (\( \Delta a/a \approx 2.5\% \)), above the conventional criteria for coherent and pseudomorphic heteroepitaxy. We had already investigated and presented the characterization of second harmonic generation in AlGaN, GaN and AlxGa1-xN/GaN multiple quantum well structures. These structures were grown on sapphire substrates by metal-organic chemical vapour deposition (MOCVD) [2]. In this work we studied samples of AlN, GaN and AlN/GaN multilayer, grown by metal-organic chemical vapor deposition (MOCVD) on HT-AlN buffer layer. The structural characterization, carried out by AFM and SEM measurements, evidenced the arising of strain effects in the AlN/GaN systems. Meanwhile we performed the second harmonic generation experiments in order to evidence the effect of the strain defects on the nonlinear process efficiency. Our aim is the realization of a distributed Bragg reflectors (DBR) structure able to enhance the confinement of the electromagnetic field at the pump and/or harmonic wave frequencies, thus resulting in a strong enhancement of the nonlinear optical response than to strong field localization. Specifically, we investigated the primary cell of the proposed DBR structure, composed by three AlN/GaN/AlN alternating thin layers, deposited on sapphire substrate. The sample structure is reported in Fig. 2(a), while the second harmonic generated signal is shown in Fig. 2(b).

We have succeeded in the growth of AlN/GaN samples, despite the large lattice mismatch of the two different nitrides. The primary cell for a possible multilayer structure has been realized and its efficiency for second order nonlinear process has been investigated.
4. PICOSECOND SET-UP FOR DELAY LINES

An experimental set-up has been developed which allows the measurement of delay-time produced by the transit of light pulses (4 ps) in an active medium embedded in a PC microcavity.

A delay line is a device that generates at its output a delayed replica of the input pulse with an externally controlled delay time $\tau$. The ideal delay line must satisfy the following requirements: not distortion of the input pulse (a transmission function with a flat spectrum is needed), the delay time must be tunable, and the optical dimension of the device must be as short as possible.

Different solid state solutions have been proposed, but it is not easy to realize tuneable devices for ultrashort (100 fs – 10 ps) pulses that are distortion free, since the frequency band of the pulse is in this case very broad [3]. Our set-up is developed in order to measure the delay time produced by a vertical cavity InAlGaAs multi-quantum-wells laser diode working at the telecommunication wavelength of 1550 nm. The source of the set-up is an OPA emitting at the wavelength of 1550 nm with a repetition rate of 1 KHz and pulse duration of 4 ps. As shown in Fig. 3 the pulses impinge from the input facet of the laser diode and are delayed by a factor that depends on the effective gain of the laser. In fact, if the laser diode is electrically pumped with a current above the lasing threshold $\approx 0.7$ mA, the positive gain of the light simulates a complex refractive index that changes the phase $\Phi$ of the input beam producing a delay $\tau = \left. \frac{d\Phi}{d\omega} \right|_{\lambda = \lambda_0} = -\frac{\lambda_0^2}{2\pi c} \left. \frac{d\Phi}{d\lambda} \right|_{\lambda = \lambda_0}$. The delay can be controlled by changing the pumping current.

The expected delay time is of the same order of the pulse duration (4 ps).

The set-up consists in two arms: a reference arm and a measurement arm. The light originated from then source is divided in the two arms and then recombined on a nonlinear crystal impinging with non collinear direction. By regulating the micrometric translation stage (MMT) it is possible to adjust the differences in the optical path in the two arms of the set-up. When the two pulses arrive on the crystal in the same time a second harmonic signal is generated along the central direction verifying that the two arms have the same propagation time. By measuring the translation to be applied on the reference arm respect at the case when the laser diode is under threshold, it is possible to measure the delay time produced by the devices.
FIG. 3: Scheme of the set-up for the ps delay time measurements.

