Journal of Optics A: Pure and Applied Optics

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To cite this article: P Ginzburg *et al* 2007 *J. Opt. A: Pure Appl. Opt.* **9** S350

IOP PUBLISHING JOURNAL OF OPTICS A: PURE AND APPLIED OPTICS

J. Opt. A: Pure Appl. Opt. **9** (2007) S350–S354 doi:10.1088/1464-4258/9/9/S11

Nonlinear optics with local phasematching by quantum-based meta-material

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Received 29 January 2007, accepted for publication 10 April 2007
Published 22 August 2007
Online at stacks.iop.org/JOptA/9/S350

Abstract
We propose a novel artificial medium, which locally provides both the optical nonlinearity and simultaneously the phasematching for the macroscopic build-up of the generated field. This is achieved by a special assembly of semiconductor quantum wells, which by careful design are both nonlinear and appropriately dispersive. A critical point for a practical realization of this meta-material is its wavelength tuning capability accomplished by applying bias voltage to the semiconductor structure, employing a voltage-controlled tunnelling-induced transparency mechanism. We theoretically demonstrate the quantum-based phasematched nonlinear meta-material principles using a specific structure comprised of GaN/AlN quantum wells providing the required near-infrared intersubband transitions. For this meta-material we estimate the enhanced second-order nonlinearity to be around 10^{-8} m V^{-1}, while the phasematching, tuned by a single-volt bias improves the nonlinear coherence length from 40 μm up to practically infinity.

Keywords: meta-material, nonlinear, quantum structure

(Some figures in this article are in colour only in the electronic version)

Nonlinear photonic devices, including frequency converters, electro-optic modulators, parametric amplifiers and oscillators, are of great importance for integrated photonics and optical communications. Two essential ingredients for an efficient macroscopic nonlinear photonic device are high values of nonlinearity and efficient phasematching in order to overcome the intrinsic material dispersion. In most of the nonlinear optics experiments phasematching is achieved by employing birefringent crystals to compensate the material dispersion [1], or alternatively by quasi-phasematching (QPM) in periodically poled ferroelectric crystals [2].

Semiconductors are excellent candidates for serving as nonlinear optical media, since they may have very high values of second-order nonlinear susceptibilities [3] and at the same time they are easily fabricated using mature technology which allows for the realization of miniature devices for integrated nonlinear photonics. Furthermore, semiconductor quantum structures can be designed to meet particular requirements of specific optical response and frequency range, which is a distinct advantage over bulk materials with a pre-determined response spectrum. However, the main limitation of bulk semiconductor materials for nonlinear optics applications is that they are mostly optically isotropic, which inhibits birefringent phasematching. Techniques to mitigate this issue were reported: using semiconductor waveguides modal phasematching has relatively low nonlinear efficiency due to the small mode overlap [4], whereas QPM techniques in semiconductors realized by epitaxial growth and wafer bonding are complex and usually result in large optical losses [3].

In this paper we propose a novel approach to mitigate the phasematching issue for highly nonlinear semiconductor materials in configurations which may allow the nonlinear operation even for quasi-continuous wave (CW) or long pulses. This is accomplished by assembling a multi-functional meta-material, composed of multiple quantum structures, that provides both the nonlinearity enhancement and the dispersion relations required for local phasematching. Usually phasematching techniques are realized by tailoring the global device properties, e.g. macroscopic periodic nonlinearity modulation (QPM), spatial field distribution and boundary conditions (e.g. modal dispersion). In our proposed method the phasematching is achieved on the atomic level by properly designing the meta-material susceptibility dispersion. We discuss the QW-based unit cell of the meta-material assembly providing the material properties, so that the overall nonlinear macroscopic interaction is significant. The optical susceptibilities of the proposed quantum-structure-based meta-material can be easily tuned by an applied voltage to match the design requirements, such as the specific wavelength of operation. The main ideas of this paper can be extended to lower dimensionality quantum structures (wires and dots). We start by rationalizing the use of intersubband transitions as the mechanism of interest, and then select a specific material system that is favourable to the visible–near-IR regime. Subsequently we discuss the design of the nonlinear features of the meta-material and subsequently detail the dispersion design to accommodate the phasematching requirements. Lastly, we discuss the tunability issues of the proposed medium.

Theoretical calculations have shown [5, 6] that dipole moments associated with interband transitions are smaller than for intersubband transitions in specially designed quantum structures and, furthermore, the latter exhibits a much narrower frequency band. Some theoretical and experimental work has been performed in designing quantum structures such as asymmetric QWs in order to increase the dipole moments and to take advantage of multiple resonant intersubband transitions in the far and mid-infrared (IR) [5, 7]. Optical telecommunications and photonics applications, however, require near-IR wavelengths. The recently developed large-bandgap optoelectronic materials based on GaN and AlN compound semiconductor materials enable the preparation of quantum structures with high enough potential barriers, allowing near-IR intersubband transitions. Furthermore these structures exhibit high intrinsic asymmetry caused by substantial strain [8]. A prospect for visible and even deep UV intersubband generation may rely on semiconductor-oxide quantum structures prepared by standard fabrication techniques [9], with even higher bandgaps.

The engineered high dipole moments of the intersubband transitions, which enhance the optical nonlinearity, can be utilized for significant electrical susceptibility variation for material dispersion compensation as well. The expression for resonant second nonlinear susceptibility in intersubband single QW operation, according to a quantum-mechanical description, is [10]:

$$\chi^{(2)} = \frac{q^3 N \mu_{12}\mu_{23}\mu_{32}}{\varepsilon_0} \frac{1}{E_{21} - \hbar\omega + \mathrm{i}\hbar\gamma} \frac{1}{E_{31} - 2\hbar\omega + \mathrm{i}\hbar\gamma} \qquad (1)$$

where ε_0 is the vacuum electric permittivity, μ_{ij} is the dipole moment of the appropriate transition, q is the electron charge, N is the density of participating carriers, γ is the dephasing of the quantum coherence and ω is the pump angular frequency. The two Lorentzians in equation (1) correspond to the atomic transitions of the basic pump frequency and second harmonic. Generally, these Lorentzians have no considerable overlap and thus no significant second-order nonlinearity occurs. However, a properly designed QW structure would lead to a resonant condition

$$2E_{21} = E_{31} \qquad (2)$$

which drastically enhances the designed meta-material optical second-order nonlinearity by about two orders of magnitude.

The intersubband-transition-induced linear permittivity can be represented as an effective linear dielectric constant using the Lorentzian model [11]

$$\varepsilon = \varepsilon_{\mathrm{bac}} - \mathrm{i}\frac{Nq}{\varepsilon_0\hbar}\sum_{i<j}|\mu_{ij}|^2(\rho_{ii} - \rho_{jj})\frac{1}{\mathrm{i}(\omega - \omega_{ji}) - \gamma_{ij}}\Gamma \qquad (3)$$

where $\omega_{ji} = \omega_j - \omega_i$ is the resonant intersubband transition frequency, ρ_{ii} is the electron occupation density ($\leqslant 1$) of the ith level, reduced to the ground state for passive device and to the exit for active, $\varepsilon_{\mathrm{bac}}$ is the averaged permittivity of the background material and is the optical field confinement factor within the QW layer.

Theoretically, by choosing the appropriate parameters, it is possible to generate sufficient dispersion for narrow band phase velocity matching in the vicinity of the resonance, whereas ultra-short pulse operation would require additional design of group velocity matching, which is beyond the scope of this paper. For a given wavelength of operation the design sequence consists of the following steps. First, the QW dimensions and shape are chosen in order to resonantly match the wavelengths for the required nonlinear process and to fulfil the resonance condition (equation (2)). The QW potential shape is then determined in practical implementations by graded doping [5] or strain. Then, for a given QW configuration, the number of QW layers (with specific waveguide parameters) is determined for achieving the required field confinement factor—Γ from equation (1). The nonlinearity, to first order, is an extensive parameter—which is enhanced linearly by the number of quantum wells—provided that they are not mutually coupled and are spatially overlapping the region of field maximum. The interaction length of such devices, which employ regularly a waveguide structure, is of a few millimetres. Furthermore, the waveguide configuration allows substantially higher field intensities, which enable much higher enhancement of the effect.

In practical realizations a number of factors may limit the performance of the proposed scheme. The first is achieving a sufficiently high ground state electron density—either by strong electrical pump or alternatively by doping the QW layer. The outstanding advantage of some of the nitride compounds is the location of the room temperature Fermi level, lying within the conduction band contributing to very high electron populations up to carrier densities approaching those of conductors. The other crucial parameter is the dephasing process. The dephasing rate is a consequence of several physical processes—the most significant are the scattering due to interface roughness, QW width variation, phonon scattering, impurity scattering, alloy disorders, many-body effects [12] and subband dispersion [13]. While the phonon scattering may be significantly reduced by temperature manipulation, a further reduction of the dephasing rate is limited by technological issues, as well as the intrinsic material structure. The typical dephasing values taken from experimental data in the literature for our calculations are about a few millielectronvolts for cryogenic QWs and tens of millielectronvolts for room temperature devices significantly weakening the resonances. In order to regain the efficiency a relatively large number of the quantum structures must be applied, which for our specific example is translated to confinement of ∼10%.

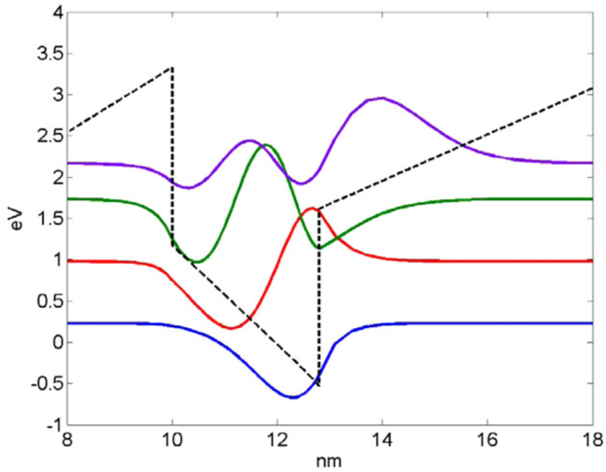

Figure 1. Electron energy level structure in a single 2.8 nm wide GaN/AlN QW. The solid lines are the electron wavefunctions and the dashed line is the QW potential structure.

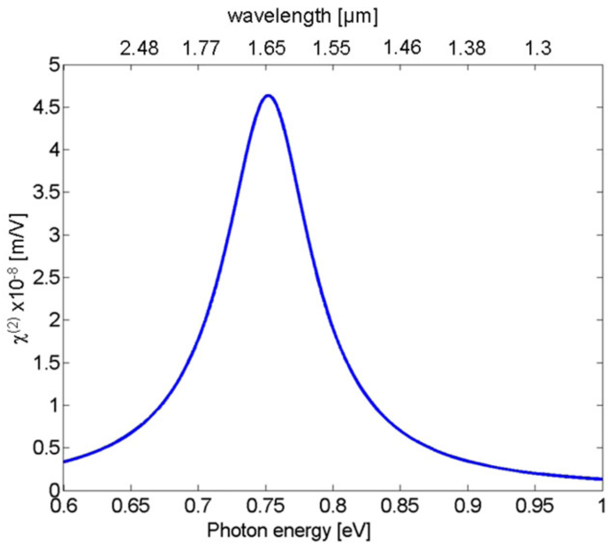

Figure 2. Second-order optical nonlinearity $\chi^{(2)}$ photon energy (wavelength) dependence in a single 2.8 nm wide GaN/AlN QW.

The dispersion of bulk nitrides were studied experimentally [14] based on the following empirical model [15]:

$$\varepsilon(\lambda) = 1 + \frac{A_0\lambda^2}{\lambda^2 - \lambda_0^2} \tag{4}$$

where A_0 and λ_0 are the material-dependent constants, that may be calculated strictly from the basic bandgap and appropriated constants for a specific material family. The background susceptibility of the composite material can be calculated according to the Maxwell–Garnett approximation as follows:

$$\varepsilon_{\text{alloy}}(\lambda) = \Gamma\varepsilon_{\text{GaN}}(\lambda) + (1-\Gamma)\varepsilon_{\text{AlN}}(\lambda) \tag{5}$$

where Γ is the optical field confinement. Generally for near-IR semiconductor nonlinear processes about $\sim$1% of index mismatch compensation is required. For the specific example of 10% GaN/AlN QW-based material for second harmonic generation process at 0.7 eV pump photon energy, the refractive index difference is 0.4%.

For a symmetrical QW potential the resonant condition (equation (2)) would not be fulfilled. However, in the case of GaN/AN QW, the wurtzite crystal structure results in a very high intrinsic strain, totally breaking the inversion symmetry of the QW confining potential (figure 1). The special shape of the strained QW potential allows the selection of the appropriate parameters to fulfil the resonant condition (equation (2)), similar to the case of QW layer graded doping [5]. Following this principle, we found by finite element calculations the optimal QW width to be 2.8 nm, and calculated the appropriate band structure and conduction band electron wavefunctions (figure 1). Calculations of the second-order nonlinearity (equation (1)) result in a Lorentzian $\chi^{(2)}$ wavelength dependence (figure 2) with a single maximum corresponding to the resonance condition (equation (2)). The optimized QW configuration leads to nonlinearity enhancement by about two orders of magnitude ($\chi^{(2)} \sim 10^{-8}$ m V^{-1}) compared to the recently measured values in similar structures [16]. Due to the energy level detuning this nonlinearity enhancement has a finite but relatively broad bandwidth of $>$100 nm.

We design the required dispersion compensation by calculating the combined optical response of the bulk material with the embedded strained QW structure by solving a one-dimensional Schrödinger equation using the finite element method.

According to the calculations of the specific example, the local quasi-CW phasematching condition is achieved for a meta-material with QW layers comprising about 10% of the total meta-material volume (figure 3). This can be easily realized in waveguide structures similar to those of QW lasers, which can have field confinement factors higher than 5%. However, once implemented, such a material will perform only for the predetermined narrow wavelength band without any possibility of external control. A number of different mechanisms may be employed to control the material dispersion of quantum systems, such as an additional optical field following the incoherent version of electromagnetically induced transparency [17] or voltage controllable tunnelling induced transparency (VCTIT) [18]. The latter configuration is more feasible due to the simpler material composition and control. The basic concept of VCTIT is controlling the resonant tunnelling between quantum low-dimensional structures by an inverse bias voltage, applied to the structure. The bias modifies the compound quantum system (e.g. coupled QWs) and causes a change in the coupling strength, which is translated into significant modification in the meta-material susceptibility [18].

Constructing a VCTIT system from the strained coupled QWs under discussion is not straightforward due to the complicated effect of the strain on the coupled system energy levels, and thus detailed quantum engineering is required. For properly chosen coupled QWs with thickness of 2.8 and 0.8 nm separated by a 0.7 nm barrier, the band structure has semi-degenerate ground states (figure 4), manifesting the electron tunnelling between the wells with no applied bias, which can be reduced or turned off by an applied voltage, allowing the

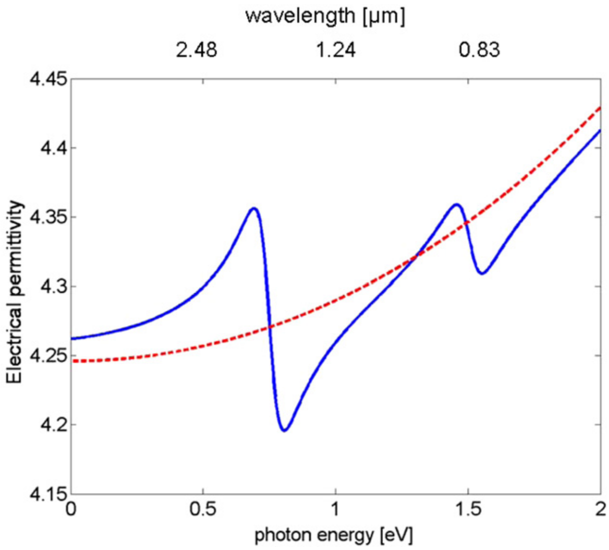

Figure 3. Single 2.8 nm wide GaN/AlN QW dispersion curves: solid line is the meta-material dispersion with phasematching for 0.7 and 1.4 eV photon energies, and the dashed line is the bulk semiconductor background dispersion.

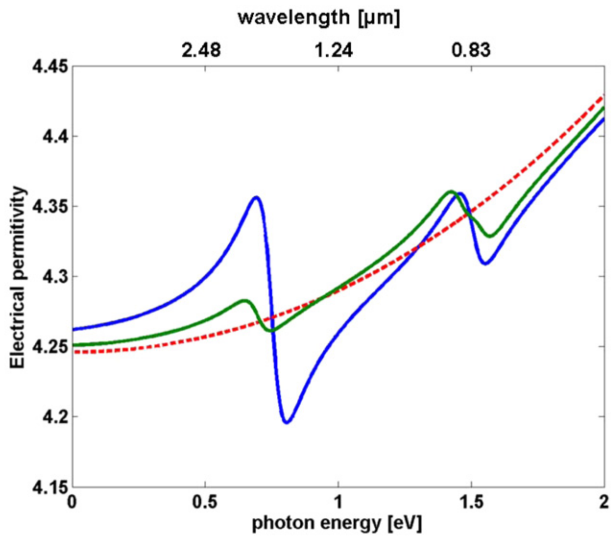

Figure 5. Coupled 2.8 and 0.8 nm GaN QWs separated by a 0.7 nm AlN barrier meta-material dispersion curves: turned-on phasematching for 0.7 and 1.4 eV photon energies (blue) and turned-off phasematching (green). The dashed line is the bulk semiconductor background dispersion.

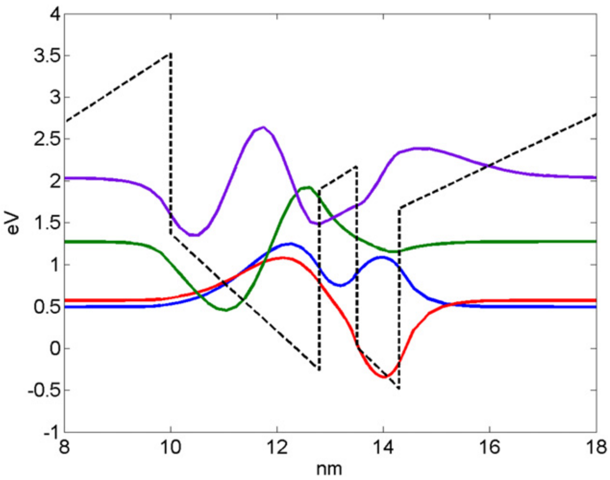

Figure 4. Electron energy level structure in coupled 2.8 and 0.8 nm GaN QWs separated by a 0.7 nm AlN barrier. The solid lines are the electron wavefunctions and the dashed line is the QW potential structure.

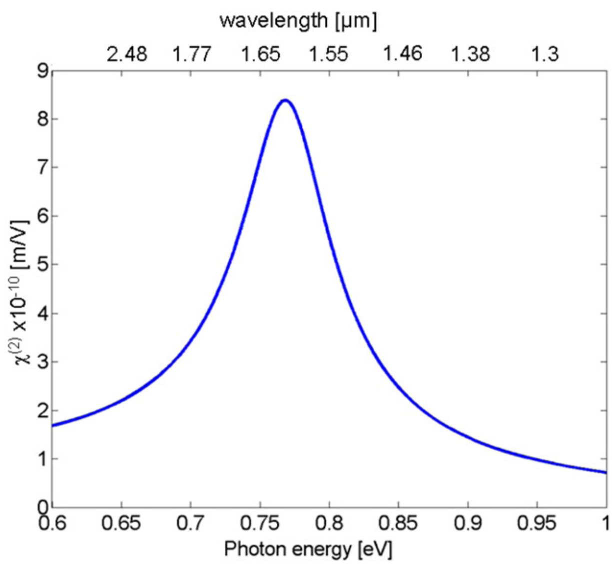

Figure 6. Second-order optical nonlinearity $\chi^{(2)}$ photon energy dependence in coupled 2.8 and 0.8 nm GaN QWs separated by a 0.7 nm AlN barrier.

meta-material dispersion tunability (figure 5). The results here are given for the extreme values of the tuning range to provide a measure for the tuning capabilities. Even at these points (total coupling and total decoupling) the main effect is on the wavelength where the phasematching is achieved and only a second-order influence on the nonlinearity.

According to our calculations the second-order nonlinear phase matching for quasi monochromatic operation can be tuned by the applied voltage to exhibit nonlinear coherence length from $\sim$40 μm (non-phasematched) without applied bias, up to total phasematching with virtually infinite coherence length for a bias of the order of a single volt. When no group velocity matching is required the dispersion curve slope has no effect on the narrow band phasematching and thus only the value of the refractive index determines the phasematched wavelengths. Therefore the same effect allows it to exhibit wavelength tunability of $\sim$50 nm range (the resonance width), regardless of the dispersion curve slope. The tunable dispersion compensation in the discussed structure is well within the nonlinearity enhancement range of $\sim$100 nm (figure 6).

In conclusion we propose a novel meta-material to overcome the phasematching issue combined with highly nonlinear semiconductor materials by assembling a carefully designed multiple quantum structure meta-material providing both the nonlinearity enhancement and the dispersion relations required for phasematching. The optical properties of the proposed meta-material are easily controlled by an applied voltage according to the design requirements. We theoretically demonstrate the quantum-structure nonlinear meta-material principles on a specific example of GaN/AlN QWs. According to our calculations the second-order nonlinearity phasematching can be tuned over a 50 nm wavelength range by applying a low bias voltage in the order of 1 V, making such meta-materials applicable in real-world nonlinear integrated photonics. The demonstrated general approach may be also applied to semiconductor-over-isolator quantum structures to achieve deep UV coherent light sources.

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