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Invited Paper

Quantum Wells and Artificially Structured Materials for Non-Linear Optics

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ABSTRACT

Semiconductor lasers have been successful in covering only a limited portion of the desired spectral regions. New non-linear optical materials based upon semiconductor quantum wells and artificially structured materials greatly enhance the non-linear susceptibility and create possibilities for both new devices to reach these spectral regions and novel structures which can be integrated with the existing lasers. Second harmonic generation with asymmetric quantum wells and approaches for phase matching are described. The potential to extend these concepts into the visible spectral region are also discussed.

1. INTRODUCTION

Non-linear effects in electronics and optics are used to modulate or mix two signals, providing both sum and difference frequencies and d.c. rectification. A major application of such non linear effects has always been to reach portions of the frequency spectrum which are inaccessible by direct generation. Over the past two decades, we have been extremely successful in developing semiconductors lasers which cover the 0.65 to 1.6 μm wavelength region. These lasers are extremely efficient, reliable, and powers up to 10 watts can be generated in arrays. These lasers have provided the foundation for the technological advances in optical communications and storage. There are, however, many additional applications which require visible (0.3 - 0.65 μm) semiconductor lasers and laser sources in the 2-20 μm wavelength region. While there has been some progress in the development of semiconductor lasers for these two regions, it appears that the materials for the 0.65-1.6 μm wavelength region possess a particularly fortuitous set of properties that make high efficiency reliable lasers possible. Unfortunately, the currently known materials severely limit the potential for lasers in these other wavelength regions. The fundamental limitation for longer wavelength lasers appears to be Auger recombination, which competes with radiative recombination and severely reduces both efficiency and power. Longer wavelength semiconductor lasers have only been realized by operation at reduce temperatures. The limitations for visible lasers appear to be the relatively high resistivity and in ability to make good wide band gap P/N junctions in wide band gap II-VI compounds. A second problem may be even greater reliability problems than those troubling the short wavelength III-V lasers. In the 0.65 μm region, defect generation due to recombination of high energy photons significantly shortens the lifetime and reliability of these shorter wavelength lasers. These potential fundamental limitations, combined with the already highly developed 0.65 to 1.6 μm laser technology suggest that non linear optical elements could be combined with these existing semiconductor lasers to provide laser sources for these other important wavelength regions.

During the past two years, several breakthroughs have occurred which suggest that non-linear conversion from existing semiconductor lasers may be a more successful approach compared to direct generation with diode lasers from other materials for these other spectral regions. First, new laser structures based upon high finesse, vertical Fabry-Perot cavities created with quarter wavelength semiconductor mirrors have demonstrated that very high photon densities and optical fields can be achieved in a cavity with very low losses (1). Both of these conditions are necessary to realize high efficiency non-linear conversion Second, dramatic increases in non-linear optical susceptibility have been demonstrated with artificially structured quantum well materials (2,3). Finally, advances in localized epitaxy, in-situ lateral patterning and epitaxial regrowth suggest that we can achieve monolithic integration of highly efficient semiconductor lasers with artificially structured, non-linear optical materials to produce lasers over the entire 0.3 to 20 micron wavelength region. In this paper, we first review the physics of non-linear optical conversion and describe our work to increase the non-linear susceptibility using the Stark effect and compositionally asymmetric quantum wells in the AlGaAs system. This system provides subband transitions for second harmonic generation from 10 μm to 5 μm. We then describe approaches to achieve phase match with artificially structured materials and discuss the prospects for extending this approach into the visible region. A schematic diagram of such a non-linear quantum well structure is shown in Figure 1.

2. PHYSICS OF NON-LINEAR OPTICAL CONVERSION

Non-linear optics places rather stringent requirements on the properties of materials (4). First, the materials must be acentric (lacking inversion symmetry), they must be phase matchable and they must have low optical loss. There are relatively few naturally occurring materials which meet these criteria and those that do are often very difficult to grow, particularly of a useful size. These materials have also not been compatible with epitaxial growth technology for semiconductors, thus enabling monolithic integration of semiconductor lasers and non-linear optical structures. Over the past two or three years, non-linear optics has taken a dramatic turn from a search for new, naturally occurring materials which met the above criteria, to the synthesis of artificially structured, non-linear optical materials based upon the well known foundation of III-V materials technology. These materials can be optimized to produce a continuous variation in energy as well as materials which would be naturally integrable with semiconductor lasers because they are based upon the same materials foundation.

The energy band structure for a quantum well is shown in Figure 2. It is readily apparent that there are two types of energy gaps which can be used for optical transitions; those from the valence band to the conduction band, resulting in relatively large energy differences, and those between the subbands in either the conduction band or valence band which result from quantum confinement and which have relatively small energy differences. These variations in energy gaps enable us to pick materials and structures in which the energy gaps cover the range from 0.01 to 2.0 eV (120 μm to 0.62 μm)

The non-linear susceptibility is similar to the refractive index in that there is a strong enhancement where there is an abrupt change in the absorption coefficient, however, it is spread over a much broader spectral region than the absorption edge. It is thus important to first create a resonant enhancement in the non-linear susceptibility and second, to utilize this enhancement below the resonance energy such that the ratio of susceptibility to absorption is maximum. Because of the wide choices over barrier heights and widths and quantum well widths, there is a great deal of optimization possible in the quantum well design which improves the susceptibility resonance, particularly for a specific application (ie. second harmonic generation, mixing, parametric amplification, etc.)

The energy band dispession relations can be expressed as:

$$E_n(k_\perp) = E_{0n} + \frac{\hbar^2 k_\perp^2}{2m_n} + \alpha_n k_\perp^4$$

where E_{0n} is the subband energy, h is the Plank constant, m_n is the carrier effective mass, $k_\perp$ is momentum in the direction$\perp$ to the quantum well confinement and α_n is a non parabolic band parameter. The intersubband transitions are particularly interesting because they are nearly parallel in $k_\perp$,thus the energy difference between any two subbands remains constant over all values of $k_\perp$. The joint density of states for both the interband and intersubband transitions are shown in Figure 3, because of the parallel nature of the subbands, the joint density of states becomes a delta function at the energy difference between the bands, ΔE_{12}. These intersubband resonances are of particular interest because the intrinsic semiconductor materials from which the quantum wells are constructed have no intrinsic optical absorption at the energy, ΔE_{12}. Near such a resonance, the second order non-linear susceptibility is (4):

$$\chi^{(2)}_{zzz} \propto \frac{z_{12}\, z_{23}\, z_{31}}{\left(\omega - \Omega_{21} + i\Gamma_{21}\right)\left(2\omega - \Omega_{31} + i\Gamma_{31}\right)} + \dots$$

where the z_{ij} are dipole matrix elements, ω is the fundamental frequency, Ω_{ij} are the resonant energies of the intersubband transitions and Γ_{ij} are the line width broadening factors. In a system with a symmetric potential, such as that of Figure 2, the dipole transition between two states with the same parity (ie. 1-3) is zero, resulting in a zero $\chi^{(2)}$. It is thus essential to create a non-symmetric potential in the quantum well. This can be done by applying an external bias voltage or using an asymmetrical quantum well structure created by compositional grading or compositional steps which do not possess a center of symmetry. If such an asymmetry is established and there is only a single intersubband resonance, where $\omega=\Omega_{31}/2$, then the susceptibility becomes:

$$\chi^{(2)} \propto \frac{z_{12}\, z_{23}\, z_{31}}{\left(\frac{\Omega_{31}}{2} - \Omega_{21}\right)\Gamma_{31}} + \frac{z_{31}^{2}\left(z_{33} - z_{11}\right)}{\frac{\Omega_{31}}{2}\,\Gamma_{31}}$$

If three subbands can be created with equal energy separations between subbands 1-2 and 2-3, such that $\omega=\Omega_{12}=\Omega_{31}/2$, then the susceptibility becomes:

$$\chi^{(2)} \propto \frac{z_{12}\, z_{23}\, z_{31}}{\Gamma_{21}\, \Gamma_{31}}$$

and an additional enhancement in the susceptibility can be realized as long as the dipole matrix element, z_{13} is again non-zero. Since the intersubband energies go as n^2 in the unperturbed square well, it is very difficult to achieve a double resonance when the symmetry is broken via an applied field to a nominally square well. On the other hand, such a design of uniformly spaced levels is possible in quantum wells with compositional steps. In the following section, we describe experimental results demonstrating the resonant enhancement of the second order non-linear susceptibility by both application of an external field to a symmetric square well and by design of an asymmetric quantum well with a compositional step.

3. SECOND HARMONIC GENERATION IN ASYMMETRIC QUANTUM WELLS

3.1 Molecular beam epitaxial quantum well structures

Two types of GaAs/AlGaAs quantum well structures were grown and characterized to investigate the resonance enhancement realized by intersubband transitions for second harmonic generation. All of the the structures were grown by molecular beam epitaxy (MBE) in a Varian GEN II MBE system. The samples were grown under typical MBE growth conditions with a growth rate of 1 μm/hr, substrate temperature of 580°C and As/Ga beam equivalent pressure ratio of 20. All layers are either undoped or doped N-type with silicon.

The first set of samples were grown on (100) oriented N+ GaAs substrates to facilitate backside ohmic contact for application of a bias potential to the otherwise symmetric quantum wells. The structures are shown schematically in Figure 4. They consist of a 0.5 μm, 2E17 cm^{-3} doped GaAs buffer, a 500 Å, 1E18 cm^{-3} GaAs contact layer, a 50 period multi-quantum well (MQW) structure with each period consisting of $Al_{0.5}Ga_{0.5}As$ barriers of 309 Å with the center 132 Å doped at 6E17 cm^{-3} and 92 Å undoped GaAs wells. The MQW is followed by another 500 Å, 1E18 cm^{-3} GaAs contact layer, 0.75 μm, 2E17 cm^{-3} GaAs layer and final 0.5 μm, 2E18 cm^{-3} GaAs contact layer.

The second set of MBE structures were grown on (100) oriented, semi-insulating GaAs substrates. The structures consist of a 1 μm, 1E18 cm-3 GaAs buffer layer, a 500 Å undoped $Al_{0.55}Ga_{0.45}As$ buffer layer, a 50 period, asymmetric MQW structure, followed by another 500 Å undoped $Al_{0.55}Ga_{0.45}As$ buffer layer and a top 0.5 μm, 1E18 cm^{-3} GaAs contact layer. The MQW structure consists of barriers of 80 Å of undoped $Al_{0.55}Ga_{0.45}As$, followed by 10 Å of 3E18 cm^{-3} GaAs and 80 Å of undoped $Al_{0.55}Ga_{0.45}As$. The asymmetric quantum wells consist of 35 Å of undoped GaAs and

60 Å of undoped $Al_{0.21}Ga_{0.79}As$. The thin (10 Å) GaAs layers in the barriers are to produce doping in the barriers and thus free electrons to populate the lowest subband of the quantum well without creating D-X centers in the nominally $Al_{0.55}Ga_{0.45}As$ barriers. A schematic band diagram for these asymmetric MQWs is shown in Figure 5. The calculated energies for the three subbands, which will be used in the second harmonic generation measurement described in the next section, are also shown.

3.2 Second harmonic generation in a biased MQWs

Optical absorption measurements were made in a single pass Brewster's angle configuration with a Digilab FTS40 Fourier-transform infrared spectrometer. Measurements on the first set of samples described above (5) were made at 77°K in order to minimize thermionic emission current over the barriers and thermal destruction of the samples. The Ω_{12} transition was observed at ~110 meV with zero applied bias. Applied biases up to 36 kV/cm were applied, but in no case were we able to observe an Ω_{13} transition in the absorption spectra. Experimental values of $\chi^{(2)}$ were obtained from second harmonic generation (SHG) measurements using a grating tuned, Q-switched CO_2 laser. The laser output was typically a train of 200 ns pulses with a peak power of 500 W at a 100 Hz repetition rate. The laser was incident on the the sample at Brewster's angle and focused to a nominal 50 μm diameter spot. The generated second harmonic output was detected with an InSb photovoltaic detector. The absolute value of $\chi^{(2)}$ was extracted by comparison to a GaAs sample measured in the same apparatus. The measured SHG power at several wavelengths vs. applied bias is shown in Fig. 6. The SHG exhibits a strong resonance in the vicinity of 10.7 μm, close to the observed Ω_{12} absorption resonance. A most important result of this experiment is that the sign of $\chi^{(2)}$ reverses with applied bias. This will be discussed further in the following section concerning phase matching techniques. The calculated value of $\chi^{(2)}$ vs. energy is shown in Fig. 7. both experimental values and calculated values are shown. The $\chi^{(2)}$ calculation uses a value of Ω_{13}= 252 meV, which was calculated from the well dimensions and applied field of 36 kV/cm (6). The maximum value of $\chi^{(2)}$ exhibits an enhancement of 70 X compared to that of bulk GaAs.While this is an impressive enhancement of c(2), the requirement of 77°K operation is a severe impediment to implementation in practical laser systems.

3.3 Second harmonic generation in asymmetric MQWs

Optical absorption and second harmonic generation measurements were made on the second set of samples (asymmetric MQWs) described in the section above (2). The measurements were performed in the same manner as described above for the biased samples, except no bias was applied and the measurements were made at room temperature. The optical absorption spectrum is shown in Fig. 8. The Ω_{12} and Ω_{13} transition energies are 127.6 meV and 228.2 meV, respectively, with FWHM of 11.6 meV and 16.6 meV, respectively. This is the first clear observation of the forbidden Ω_{13} transition in absorption. These values are reasonably close to the calculated values based upon the structure (2).

The measured SHG power vs. incident angle at two wavelengths is shown in Fig. 9. The strong increase at 10.74 μm is expected for the energies of the two resonances realized in this asymmetric MQW structure. The measured and theoretical values of $\chi^{(2)}$ vs. energy are shown in Fig.10. This is an enhancement of a factor of 320 in $\chi^{(2)}$ compared to bulk GaAs. Since the agreement between the calculated and experimental transition energies was very good, we assumed that the errors between the experimental and theoretical integrated absorption fraction were due to a difference between the effective carrier density and the intended dopant density. We forced agreement between experiment and theory in the integrated absorption fraction by adjusting the effective sheet carrier density and then used this carrier density in the calculation of $\chi^{(2)}$.

In addition to an enhancement of $\chi^{(2)}$, it is desirable to control the sign of $\chi^{(2)}$ where waveguide structures are desired and phasematching is necessary. Various structures to achieve this are discussed in the following section. In the previous section, we demonstrated control of the sign and magnitude of $\chi^{(2)}$ through application of an applied bias. It is clear that the sign of $\chi^{(2)}$ can also be reversed by reversing the order of the MBE growth of the two layers comprising the asymmetric quantum wells, however, this is relatively difficult because patterning, etching and MBE regrowth of alternate sign reversed regions is required. If the losses in the structure are low, a longer phase matching structure can be used and one simply alternates regions with and without a strong non-linear susceptibility. Since the intersubband resonant enhancement of $\chi^{(2)}$ is due to a transition of free carriers from the lowest subband to higher subbands, one can eliminate the enhancement of $\chi^{(2)}$ by simply eliminating the free carriers. This is relatively easy to do in GaAs/AlGaAs structures because the bandgaps are sufficiently large, that if traps are created in the material by proton or ion implantation, all the free carriers are trapped and the material becomes semi-insulating. This technique is widely used for electrical isolation of GaAs devices and integrated circuits. This technique was demonstrated by first removing the the heavily doped GaAs cap layer and then proton bombarding the exposed structure to generate a number of traps which exceeded the doping density of the structure. After proton bombardment, the structure showed no resolvable intersubband absorption. The measured SHG power at 10.74 µm as a function of inciden angle is shown in Fig. 11 for both the proton bombarded and non-bombarded regions. The proton bombarded regions are indistinguishable from the blank substrate, thus indicating that the bombardmen has completely eliminated the non-linear susceptibility due to the quantum wells.

4. NON-LINEAR WAVEGUIDE DEVICES

In addition to creating a strong non-linear susceptibility, useful application of non-linea conversion requires phase matching between the various frequency components in the process (ie fundamental and second harmonic for SHG).Very few materials provide phase match at even a singl desired frequency, let alone over a broad spectral region. A solution to this problem was first describe by Armstrong, et al. (7) and is known as quasi-phasematching. Structures using quasi-phasematchin are shown schematically in Fig. 12. The distance L_c is the critical length over which the fundamenta and second harmonic shift phase by 180°. If as shown in the top figure, the sign of $\chi^{(2)}$ is reverse then the sign of the second harmonic is reversed and the two are effectively back in phase so that powe is continuously transferred from the fundamental to the second harmonic. In a case where reversing th sign of $\chi^{(2)}$ is either difficult or impossible, an alternative is to create only regions with strong $\chi^{(2)}$, b which are separated by regions of small or zero $\chi^{(2)}$ with a dimension of $2L_C$. This is shown in th lower part of Fig. 12. The first approach would require structures like those described in section 3. where the sign of $\chi^{(2)}$ is reversed by bias polarity or the asymmetric structures described in section 3. where multiple MBE growths are utilized to reverse the sign of $\chi^{(2)}$. The second approach is far easier to fabricate and would utilize the proton bombardment approach described in section 3.3 to create non-active regions between the regions of strong non-linearity with the same sign of $\chi^{(2)}$.

One of the difficult technical problems with waveguide structures is coupling the light into and out of the waveguide. In order to get very high electric field strengths in the waveguide and hence higher conversion efficiency, the guide thickness is of the order of the fundamental wavelength. It is thus difficult to align and focus more than a small fraction of the incident laser into the waveguide. One means of overcoming this is to utilize grating couplers on the surface of the waveguide such that the incident laser does not have to be aligned and focused to the guide, but only a much broader area grating. Such a grating coupled waveguide structure is shown schematically in Fig. 13. This structure shows not only the grating, but a structure for alternating the sign of $\chi^{(2)}$.

While these structures look quite promising for producing semiconductor based lasers in the 4 to 20 μm region, they will not work for short wavelength visible lasers. The key problem for visible lasers is that while a very strong $\chi^{(2)}$ can be created using the interband (ie. valence to conduction band) transitions, the material is very strongly absorbing to the generated second harmonic. Normandin, et al (8) and Vakhshoori, et al (9) have suggested approaches to use the intrinsic non-linearity of GaAs near the bandedge to generate visible light from counter propagating fundamental waves such that it is coupled vertically out of the top of the waveguide and thus propagates only through a very thin region of strongly absorbing GaAs. Enhancements to these concepts using MQWs to increase the non-linearity and highly reflective quarterwave semiconductor mirrors to trap the fundamental and increase the optical field strength could make these concepts useful for generation of visible lasers down to 0.35 μm.

5. CONCLUSIONS

Multi quantum well structures offer the potential to greatly enhance the non-linear optical susceptibility compared to bulk materials. These structures also permit design optimization for different spectral regions and the potential to quasi-phasematch by either reversal of the sign of $\chi^{(2)}$, or destruction of $\chi^{(2)}$ by proton bombardment. Both of these concepts have been demonstrated in the mid-infrared spectral region which is accessible to AlGaAs/GaAs MQWs. New structures have been suggested which require waveguide structures and different materials, but which could provide laser sources across the entire 0.35-20 μm spectral region. There remains a considerable amount of work to demonstrate useful visible optical conversion by these approaches, however, the combination of high power semiconductor lasers, highly reflective semiconductor mirrors, resonance enhancement of the non-linear susceptibility in MQWs and in-situ processing advances to fabricate and integrate the various components, suggests that such visible lasers may be realizable. The realization of reliable, efficient, low cost semiconductor lasers over the entire visible spectral region would indeed have a revolutionary impact on optical display, scanning, copying, storage and communications technologies.

6. ACKNOWLEDGEMENTS

The authors would like to acknowledge the valuable contributions of current and former graduate students, Alex Harwitt, Ben Yoo and Gideon Yoffe for the results described in this paper. They would also like to thank Tony Ma, Jim Adkisson, Darrell Schlom, Bob Lodenkamper, Bartev Vartanian and Eric Lim for MBE growth and helpful discussions on design and processing. This work was supported by ONR/DARPA by contract N00014-84-K-0077, ONR/SDIO by contract N00014-89-K-0067 and JSEP by N00014-84-K-0327.

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