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Citation for published version (APA):

DOI:
10.1364/OE.2.000462

Document status and date:
Published: 01/01/1998

Document Version:
Publisher’s PDF, also known as Version of Record (includes final page, issue and volume numbers)

Please check the document version of this publication:

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Surface emitted second-harmonic generation from a quasi-phase matched waveguide in an Al$_x$Ga$_{1-x}$As/Al$_2$O$_3$ microcavity

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Abstract: A nonlinear Al$_x$Ga$_{1-x}$As waveguide consisting of a quasi-phase matched heterostructure embedded in a microcavity has been designed and fabricated. The microcavity resonator is formed by Al$_2$O$_3$/Al$_{0.32}$Ga$_{0.68}$As multilayer mirrors located above and below the waveguide core. The cavity resonantly enhances the surface emitting second-harmonic generation. The SH conversion efficiency has been measured for wavelengths between $\lambda = 1525$ and 1575 nm. A simple waveguide loss measurement technique based on the SH autocorrelation of short optical pulses in a III-V waveguide is also demonstrated.

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OCIS codes: (190.2620) Frequency conversion; (190.4390) Nonlinear optics, integrated optics

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References and links

1. Introduction

Optical second-harmonic (SH) generation by counter-propagating beams in a nonlinear waveguide was first demonstrated almost two decades ago in LiNbO$_3$ waveguides [1]. In this geometry the second-harmonic light is radiated from the surface of the waveguide. Despite the low SH efficiency achieved in these prototype devices, some unique applications of the surface emitting geometry were evident. In particular, the use of nonlinear waveguide autocorrelation for short pulse measurement and signal processing was soon demonstrated [2,3]. More recently, optical spectrometers based on nonlinear waveguides have been demonstrated [4,5] and used in a frequency locking system for semiconductor lasers [6]. The SH power generated in the homogeneous LiNbO$_3$ and GaAs waveguides used in the early experiments was far too low for practical application because the nonlinear interaction is not phase matched. Conversion efficiencies are also limited because the interaction length for the SH and the pump light along the surface normal direction is limited to the waveguide thickness.

Two developments have increased the SH conversion efficiency of these waveguides. The first improvement was achieved by using a multilayer waveguide core consisting of alternating regions of high and low SH susceptibility, $\chi^{(2)}$ [7]. When the period of this multilayer structure corresponds to the wavelength of the SH light, the surface emitting conversion process is quasi-phase matched (QPM) and the efficiency can be increased by several orders of magnitude. Although the geometry differs from that of conventional co-propagating QPM waveguides [8], the principle is the same. QPM in surface emitting waveguides is usually implemented by using alternating layers of high and low Al concentration Al$_x$Ga$_{1-x}$As layers in the waveguide. The Al$_x$Ga$_{1-x}$As system not only has an extremely high second-order nonlinearity, but $\chi^{(2)}$ can be varied by up to an order of magnitude in the near infrared [9] simply by changing the Al concentration. The second technique to enhance the SH efficiency is to enclose the nonlinear waveguide core inside a microcavity resonator for the vertically emitted SH light [10]. The nonlinear interaction length is multiplied by the number of passes in the microcavity, which results in a much higher conversion efficiency at the cavity resonance wavelength.

This paper describes the integration of a quasi-phase matched (QPM) nonlinear waveguide with a microcavity resonator to combine the advantages of both techniques. Multilayer stacks of alternating Al$_x$Ga$_{1-x}$As and Al$_2$O$_3$ layers form the microcavity mirrors. In this work, the index of refraction of Al$_2$O$_3$ is taken to be $n = 1.6$ at both the fundamental ($\lambda=1550$ nm) and SH ($\lambda=775$ nm) wavelengths. This is approximately one half the index of Al$_x$Ga$_{1-x}$As at these wavelengths. Because the index difference between these two materials is so large, new optical devices can be fabricated which are not possible using conventional...
III-V epitaxy. For example, semiconductor waveguides containing Al₂O₃ layers can have an extremely large polarization birefringence [11]. This birefringence has recently been exploited to phase match frequency mixing processes in co-propagating geometries [12]. In our waveguides, the large index difference allows the fabrication of Al₂O₃ based mirrors using far fewer layers than conventional AlₓGa₁₋ₓAs mirror heterostructures. The large index difference also enables the pump modes to be tightly confined to the QPM waveguide core instead of spreading into the mirror regions, thereby increasing the SH conversion efficiency.

2. Theory

Waveguides for surface emitted second-harmonic generation (SESHG) are almost exclusively fabricated using the AlₓGa₁₋ₓAs material system. In this cubic zincblende material only the $\chi^{(2)}_{xyz}$ tensor element (and those with equivalent permutations of the indices) is non-zero. Here the coordinate system is aligned with the cubic unit cell axes. In waveguides grown on a standard (001) substrate, light propagation is usually along a (110) or equivalent direction. Therefore a counter-propagating transverse electric (TE) mode and transverse magnetic (TM) mode must interact to induce a radiating SH polarization. The induced SH polarization vector lies in the plane of the wafer and perpendicular to the direction of waveguide propagation, and has the form

$$\mathbf{P} = \chi^{(2)}_{xyz} \left[ E^+_{TM} E^-_{TE} + E^+_{TE} E^-_{TM} \right]$$

(1)

where $E^+_{TM,TE}$ and $E^-_{TM,TE}$ are the TM and TE components of the counter-propagating fundamental electric fields. We first consider a waveguide consisting of an AlₓGa₁₋ₓAs heterostructure core enclosed by two infinitesimally thin mirrors at $z = 0$ and $z = -D$, which form a vertical resonator. We assume that the spacing $D$ is chosen so that the cavity is in resonance at the SH wavelength. Neglecting the effect of interface reflections within the core, the SH intensity radiated from the waveguide has the form [13,14]

$$I_{\text{SH}} = \frac{E}{1-r^2} \left| \int_{-D}^{0} \frac{P(z)}{n(z)} \exp \left[ -i \int_{z}^{0} K(z') dz' \right] dz + r \int_{0}^{D} \frac{P(z)}{n(z)} \exp \left[ i \int_{0}^{z} K(z') dz' \right] dz \right|^2$$

(2)

where $E$ is a constant, $K(z)$ is the local wavevector of the SH light, and the z-axis is along the surface normal direction. Here $r$ and $t$ are the field reflectivity and transmission coefficients respectively. The two terms in Eq. (2) correspond to the two SH waves initially radiated towards the surface and towards the substrate. It is clear from Eq. (2) that the SH intensity will remain small unless the SH polarization varies spatially with a modulation wavevector equal to $K(z)$. In the AlₓGa₁₋ₓAs system this quasi-phase matching condition is easily implemented by growing alternating layers of high and low Al concentration with a period equal to the wavelength of the SH light. As the reflectivity of the cavity mirrors approaches 100%, Eq. (2) also predicts a large enhancement of the SH conversion efficiency as the $\left| \frac{1}{1-r^2} \right|$ term diverges. The resonant enhancement of the SESHG by the microcavity is related to the coherent nature of the SHG process. Since the coherence time of the emitter - the SH polarization - is very long compared to the cavity lifetime, increasing the cavity lifetime results in an increased coupling between the SH polarization and the electromagnetic field. This is in contrast to the case of spontaneous emission in...
microcavities at room temperature, where the emitters’ coherence time is usually shorter than the cavity lifetime.

The waveguide described in this paper integrates both QPM and a microcavity in a single structure. The heterostructure must provide waveguide confinement of the fundamental light at $\lambda = 1550$ nm, quasi-phase match the SH generation process, and provide a resonant microcavity at the SH wavelength. For this SESHG waveguide, Eq. (2) is at best only a crude approximation since the effects of interface reflections and the distributed nature of the resonator mirrors are not included. Since the mirrors consist of multilayer stacks, both the SH cavity mode and fundamental waveguide modes will extend into the mirror structure itself. Furthermore, if the mirrors are composed of a nonlinear material such as Al$_x$Ga$_{1-x}$As, even the mirror layers will radiate SH light.

The design and modeling of our SESHG waveguides was performed numerically. The waveguide modes at $\lambda = 1550$ nm were calculated using a conventional transfer matrix algorithm. The SH light generated by the interaction of counter-propagating fundamental modes was calculated using a Green function formalism developed by Sipe [13] specifically for nonlinear optics in multilayer structures. The application of this formalism to SH generation in microcavity structures is also discussed in Beaulieu et al. [14]. The Green function method calculates the effect of multiple reflections at all interfaces, and hence the microcavity resonance is implicitly incorporated in the final result. Both the waveguide mode calculation and the microcavity calculation were combined into a single computer program that simultaneously optimized the waveguide and microcavity modes for maximum SH conversion efficiency. Measured values of $\chi^{(2)}$ at $\lambda = 1550$ nm for different Al concentrations are not available in the literature. To model this waveguide, we estimated $\chi^{(2)}$ values at different Al concentrations using an interpolation scheme based on existing data for GaAs and AlAs [15]. The $\chi^{(2)}$ values derived from this interpolation method are in good agreement with measurements carried out at $\lambda = 1064$ nm [9], and we assume that the values are therefore also approximately correct near $\lambda = 1550$ nm.

The calculated SH conversion efficiency of the SESHG waveguides is expressed in terms of the nonlinear cross-section $A_{nl}$ [14]. The radiated SH power is given by

$$P_{SH} = A_{nl} P^+ P^- \left( \frac{L}{w} \right)$$

where $P^+$ and $P^-$ are the powers of the counter-propagating pump beams, $L$ is the length of the waveguide in centimeters, and $w$ is the width of the waveguide in millimeters.

3. Waveguide design

The layer structure of nonlinear waveguide is shown in Fig. 1, along with the calculated waveguide mode intensity profile. The waveguide core consists of three 112 nm thick Al$_{0.32}$Ga$_{0.68}$As layers separated by 124 nm Al$_{0.8}$Ga$_{0.2}$As barrier layers. The layer thicknesses are exactly half the wavelength of the SH light in the material, so that the surface emitting SH conversion process is quasi-phase matched. The magnitude of $\chi^{(2)}$ at $\lambda = 1550$ nm in Al$_{0.32}$Ga$_{0.68}$As is expected to decrease with Al concentration. However to prevent absorption of the SH light, the Al concentration must be larger than $x = 0.32$. Using the estimation procedure described in Section 2, we find that $\chi^{(2)}$ is 2.5 times larger in Al$_{0.32}$Ga$_{0.68}$As (|$\chi^{(2)}$| = 0.82 x 10$^{-6}$ esu) than in Al$_{0.32}$Ga$_{0.68}$As (|$\chi^{(2)}$| = 0.32 x 10$^{-6}$ esu).

The mirrors forming the microcavity are formed from a two period quarter-wave stack of Al$_2$O$_3$ and Al$_{0.32}$Ga$_{0.68}$As layers. Each Al$_2$O$_3$ layer is 142 nm thick and each Al$_{0.32}$Ga$_{0.68}$As layer is 56 nm thick. The large index difference between these two materials means that even with only two periods a very high reflectivity is obtained at the SH wavelength of $\lambda = 775$ nm. Spacer layers of Al$_{0.3}$Ga$_{0.2}$As were inserted between the QPM
Fig. 1. SESHG waveguide layer structure showing the Al$_{0.8}$Ga$_{0.2}$As/Al$_{0.32}$Ga$_{0.68}$As QPM heterostructure core and the Al$_2$O$_3$/Al$_{0.32}$Ga$_{0.68}$As mirror layers. The dashed curve shows the calculated intensity profile of the fundamental TE mode.

waveguide core and the microcavity mirrors. The 571 nm spacer thickness was determined by optimizing the calculated SH conversion efficiency as described in Section 2. Fig. 2 shows the calculated SH cross-section of this waveguide for wavelengths between $\lambda = 1525$ nm and 1575 nm. A peak $A_{nl}$ of $4.8 \times 10^{-5}$ W$^{-1}$ cm$^{-1}$mm is predicted at the microcavity resonance. The bandwidth of the QPM resonance is larger than 100 nm, and hence is not visible on the scale of Fig. 2. Fig. 2 also shows the SH efficiency when the microcavity mirrors are turned off. This was calculated by setting the index of refraction in the Al$_2$O$_3$ equal to that of the Al$_{0.32}$Ga$_{0.68}$As layers at the SH wavelength, while leaving all other parameters unchanged. This calculation demonstrates that the resonator enhances the SH conversion efficiency by 25 times over that of the QPM waveguide alone.

4. Experiment

The waveguide structure was grown by molecular beam epitaxy on a semi-insulating (001) GaAs substrate. Layers of AlAs were grown at the location of the Al$_2$O$_3$ layers in the final waveguide, and later converted to Al$_2$O$_3$ by wet oxidation. Reactive ion etching was used to define 3 $\mu$m wide ridge waveguides with 1.3 $\mu$m deep sidewalls. The ridges were located on top of a 100 $\mu$m wide mesa with 1.25 $\mu$m deep sidewalls. The waveguide and mesa sidewalls expose the AlAs mirror layers for subsequent lateral oxidation. The wafer was oxidized for 20 minutes at 400$^\circ$ C in a water vapour atmosphere obtained by bubbling a N$_2$ carrier gas through water at 95$^\circ$ C. The oxidation of the AlAs was observed to proceed.
lateral from the mesa edge and completely oxidize the AlAs mirror layers at the waveguide ridges. Waveguides of 2.4 mm in length were then cleaved out for testing.

A tunable colour centre laser was used as the pump light source in the SH generation experiment. The laser was operated in a synchronously pumped mode with a 50 ps pulse width and a 76 MHz repetition rate. The pump light was polarized at 45° to the plane of the waveguide surface normal, and coupled into the end facet of the waveguide using a 40× objective lens. This geometry ensured that TE and TM modes of equal intensity were propagating in the waveguide, as required by Eq. (1). The counter-propagating beam was provided by the reflections of the input beam off the end facet of the waveguide. The SH output beam emitted from the waveguide surface was filtered and detected by a cooled CCD array.

5. Results

Fig. 3 shows the variation of the measured SH intensity with wavelength between $\lambda =1530$ nm and $\lambda =1580$ nm. The shape of resonance peak $\lambda =1554$ in Fig. 3 is in close agreement with the predicted resonance from Fig. 2. The total SH power at the resonance peak was 3×10⁻¹⁰ W for an average pump power of 2.5 mW incident on the waveguide facet. We estimate a factor of two uncertainty in the SH power measurement, primarily due to uncertainty in the CCD array calibration. The estimated coupling efficiency into the waveguide was approximately 6%. When the duty cycle of the pulsed pump laser is taken into account, a conversion efficiency of 5×10⁻⁵ W⁻¹ is obtained. Using Eq. (3) and correcting for the reflection of the counter-propagating beams by the output facet ($R \sim 0.3$) gives the SH cross-section $A^{nl} = 2\times10^{-6}$. This is more than an order of magnitude smaller
than the model prediction shown in Fig. 2. Since the measured resonance line shape in Fig. 3 matches the theoretical $A_{nl}$ lineshape, the microcavity is operating as predicted and internal cavity losses are negligible. Therefore, we attribute the anomalously low $A_{nl}$ to waveguide losses at the fundamental wavelength.

Waveguide loss at $\lambda = 1554$ nm was determined from the SH intensity pattern at the waveguide surface generated by a 76 MHz mode-locked train of 8 picosecond pump pulses. As before, the laser beam was coupled into the waveguide at 45° to the surface normal to generate both TM and TE waveguide modes. The surface of the waveguide was imaged using the cooled CCD array. Fig. 4 shows the SH intensity profile along the waveguide ridge. The high frequency beat pattern of the SH intensity in Fig. 4 arises from the TE-TM birefringence of the waveguide at the pump wavelength. The broad peak at the left-hand side of Fig. 4 is due to the interaction of the incident pulse with its counter-propagating reflection from the output facet. The much smaller peak at the right is due to the interaction of the pulse reflected from the output facet interacting with its second reflection from the input facet. The ratio of the two SH peak amplitudes is proportional to the square of the waveguide loss per pass multiplied by the facet reflectivity. A comparison the two peaks in Fig. 4 yields an estimated internal waveguide loss of 13 dB/cm. The SH conversion efficiency scales as the square of the waveguide loss per pass, so this measured loss predicts that $A_{nl}$ should be reduced by an order of magnitude. High waveguide losses in the near infrared have been observed in other waveguides incorporating Al$_2$O$_3$ layers. We attribute the loss to scattering by roughness at the Al$_2$O$_3$/Al$_{0.8}$Ga$_{0.2}$As interface, and absorption by mid-gap defect states in the Al$_{0.8}$Ga$_{0.2}$As. These defects may be associated...
with excess As atoms that have migrated from the AlAs to the AlGaAs during oxidation [16].

The loss measurement technique described here is itself of some interest. All III-V waveguides have a zincblende crystal structure, and therefore a second-order susceptibility tensor of the same form as Al x Ga 1-x As based materials. It is therefore possible to use this SH autocorrelation technique to measure internal loss in virtually any III-V waveguide. This loss measurement requires a source of optical pulses several times shorter than the length of the waveguide being tested, and a sufficiently sensitive imaging system. In this lab, a cooled CCD array camera with an effective noise floor of 10^{-16} J per pixel can easily obtain images of the surface emitted second-harmonic light generated in homogeneous GaAs and InP based waveguides by pump beams with average powers of a few milliwatts. Loss measurements carried out using this technique are non-destructive, and do not require a tunable light source as do loss measurements obtained using the Fabry-Perot transmission oscillations of the waveguide cavity.

6. Summary

A nonlinear waveguide with a quasi-phase matching multilayer core and a microcavity resonator has been designed for surface emitting SH generation at a pump wavelength of \( \lambda = 1550 \) nm. The resonator structure is formed from two Al_{2}O_{3}/Al_{0.32}Ga_{0.68}As multilayer mirror stacks above and below the waveguide core. The SH cross-section of \( \chi^{(2)} = 4.8 \times 10^{-5} \) W^{-1}cm^{-1}mm has been calculated at the resonance wavelength. The microcavity resonator enhances the SH conversion efficiency of the waveguide by a factor of 25. This waveguide structure was fabricated and tested. The line shape of the measured SH resonant peak agreed closely with the calculated line shape of the microcavity resonator. However the

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**Fig. 4.** The SH intensity profile along the SESHG waveguide ridge when pumped by 8 ps long pulses at \( \lambda = 1554 \) nm. The output facet is at left and the input facet is at the right. The SH intensity to the right of the dashed line has been scaled up by a factor of ten.
experimental $A^{nl}$ value at resonance was more than an order of magnitude smaller than predicted by theory. The anomalously low $A^{nl}$ is consistent with the measured internal waveguide loss. Finally, we have demonstrated a simple waveguide loss measurement technique based on the SH intensity autocorrelation pattern generated by short optical pulses in the waveguide. This technique is generally applicable to any III-V waveguide with a bulk second-order susceptibility.