Fabrication and characterization of Teflon-bonded periodic GaAs structures for THz generation

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Abstract: A novel technique for low-temperature bonding of GaAs wafers using an interboundary Teflon film is developed. A fabricated stack of ten 25x25 mm² diced wafers demonstrated 75% transmission of a 10 μm CO₂ laser beam. Modeling of these Teflon-bonded (TB) structures as sequences of Fabry-Perot etalons gives a good agreement with transmission measurements. A 20x20 mm² quasi-phase matched structure of five wafers pumped by CO₂ laser lines generated the narrow-band THz radiation at a wavelength of 343 μm via a difference frequency mixing process.

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References and links

1. Introduction
Difference-frequency generation (DFG) in nonlinear crystals is the most efficient method of producing narrow-band, terahertz (THz) pulses tunable in the range of 0.3-3 THz. The use of the III-V semiconductors, such as GaP, InSb and GaAs, is advantageous in obtaining a highly efficient THz output, since their second-order nonlinear susceptibilities are several times larger than those of the commonly used electro-optic crystals LiNbO₃ and ZnTe. These crystals, being isotropic, require either noncollinear (vector) phase matching or quasi-phase matching. The former phasematching was recently used for generation of tunable THz pulses with kW to MW peak power in a GaAs crystal pumped by a two-wavelength CO₂ laser [1], and the latter method successfully produced 1mW average power at 2.8 THz in a diffusion-bonded GaAs device pumped by a 2 μm optical parametric oscillator [2]. Quasi-Phase
Matched (QPM) GaAs structures are particularly attractive for infrared nonlinear optics because the material has a relatively large nonlinear susceptibility and large-diameter GaAs crystals with a wide transparency range of 2-12 μm and 200-3000 μm are commercially available. Since the efficiency of THz wave generation is naturally small and limited for a pulse system by the surface damage threshold, scaling of both aperture and length of the OPM structure is required in order to achieve a high DFG output.

Current fabrication techniques for QPM GaAs structures include diffusion bonded (DB) GaAs [2,3], optically contacted (OC) GaAs [4] and epitaxially grown GaAs [5]. Epitaxial growth produces structures of high quality but limited aperture (~500 μm). Diffusion bonding can potentially result in a structure with a clear aperture of several cm²; however, high temperature (800 °C) bonding procedures at temperatures above thermal conversion of GaAs to p-type result in increased optical losses, especially in the THz range [6]. OC-GaAs solves this high-temperature bonding issue by using the Van der Waals forces that develop between smooth surfaces brought together in optical contact as a bonding mechanism. But homogeneous contact over the large area is hard to achieve via this method, making it challenging to use OC-GaAs for large aperture applications.

The use of a thin film as a bonding layer can overcome the abovementioned problems. A low-temperature, 300° bonding for a large aperture (20x20 mm²) GaAs-Si optoelectronic device have been demonstrated by using a 80 nm thick Au-Ge eutectic alloy between the wafers [7]. However, for nonlinear optical applications a metallic intermediate layer introduces formidable losses.

In this paper we present a novel technique for bonding GaAs wafers using an interboundary Teflon AF film to make a large-aperture IR and THz radiation transparent structure. The transmission properties of Teflon-bonded (TB) GaAs wafer stacks were characterized using 10 μm light. To demonstrate the potential of the Teflon-bonding technique, a QPM TB-GaAs structure manufactured using (110) GaAs wafers was fabricated and tested for THz DFG pumped by two CO2 laser lines.

2. Teflon bonding of GaAs wafers

2.1 Multiple Fabry-Perot etalon stack as a QPM structure

Quasi-phase matching is achieved by periodic modulation of the nonlinear susceptibility which compensates for the phase velocity mismatch between the interacting polarization waves, allowing efficient interactions in the absence of phase-velocity matching in an isotropic nonlinear material. The modulation of the nonlinear susceptibility is a sign reversal, with a period equal to twice the coherence length, \(L_c = \frac{\pi}{\Delta k}\), where \(\Delta k = (n_3 \omega_3 - n_2 \omega_2 - n_1 \omega_1)/c\) represents the net difference between the wave vectors, or the phase mismatch. For the specific process of DFG, the interacting frequencies are related by \(\omega_1 = \omega_3 - \omega_2\). If two CO2 laser beams participate in DFG in a GaAs crystal \(n_3 \approx n_2 \approx 3.28\) to produce the THz beam \(n_1 = 3.58\), the \(L_c\) is approximately 700 μm. This implies that readily available semiconductor industry wafers with a thickness equal to the coherence length can be periodically reversed and stacked together to form the QPM structure. Note that a typical industry tolerance on wafer thickness around 25 μm should not significantly affect a QPM structure operating in the THz range.

Creation of a free-standing stacked wafer QPM structure requires a bonding mechanism in order to cancel Fresnel losses. As stated above currently employed bonding methods [3-5] have limited application to creating large-aperture devices. At the UCLA Neptune Laboratory we have developed a low-temperature bonding technique for 20x20 mm² GaAs wafers using an interboundary Teflon film. Use of an intermidiate bonding layer allows a lower bonding temperature and homogeneous bonding over large areas. Figure 1 illustrates such a structure using GaAs wafers. To cancel Fresnel losses and therefore maximize transmission of incident beams, the intermediate layer must adhere to strict thickness requirements dictated by

\[
2n_l \cos \theta = m \lambda ,
\]
where $n_l$ is the layer refractive index, $\theta$ is the angle of incidence for the pump beam, $l$ is the thickness of the layer, $m=0,1,2,\ldots$, and $\lambda$ is the wavelength of the incident pump beam. If Eq. (1), which is the condition for constructive interference, does not hold true, cancellation of the pump beam may occur making DFG process inefficient. The device shown in Fig. 1 can be represented by a multiple-etalon stack and a well known Fabry-Perot formalism can be used to derive its transmission properties.

Fig. 1. A GaAs structure bonded by a Teflon intermediate layer.

The fractional transmitted intensity for a single etalon is given by [8]

$$ T = \frac{I}{I_i} = \frac{(tt')^2}{(1-rr')^2 + 4\sqrt{rr'} \sin^2 (\delta / 2)} $$

(2)

where $I$, and $I_i$ are the transmitted and incident intensities, respectively, $t,t'$, and $r,r'$ are the transmissivities and reflectivities, and $\delta=4\pi n_l l \cos \theta / \lambda$ is the path difference between different rays. This can be generalized to $s$ etalons as

$$ T_{\text{total}} = \frac{I_{\text{total}}}{I_i} = \prod_s \frac{(ts_s)^2}{(1-rs_s')^2 + 4\sqrt{rs_s} \sin^2 (\delta_s / 2)} $$

(3)

When a system is comprised of $s$ etalons of different thicknesses and refractive indices, the transmission profile is influenced by the dominant etalon. Simulations of such system showed that this etalon is one with the highest refractive index and largest thickness (a GaAs wafer in our case). Equation (3) indicates that optical losses can be minimized by tuning the incident angle $\theta$. Moreover, as more etalons added, the width of the transmission peaks becomes smaller, thus $s$ is a limiting factor in the transmission of a multilayer Fabry-Perot etalon. It is also important to note that the structure in Fig. 1 can only be used when the converted wavelength is much longer than the pump wavelength. In the case of the THz DFG it is true and the width of etalon transmission at the THz frequency is larger than that at the pump frequency.

2.2 Teflon bonding technology

To create a bonded structure using bonding layers, a very thin film of material is deposited homogeneously on GaAs wafers. The wafers are stacked with bonding layers between them.
The material is heated to the point of softening, and pressure is applied to the stack to ensure good contact. After cooling, the bonded stack can function as a free-standing device.

Ideally, the bonding material will have a low index of refraction, low absorption coefficient, the ability to be applied onto wafers as thin film, and potential for a low-temperature bonding procedure. Teflon AF (Dupont, Inc) meets all these criteria: it is a fluoropolymer that can form a low-viscosity solution at room temperature, allowing it to be easily spun on to any substrate to form a thin film. It has an amorphous structure that will not interfere with the transmission of incident beams. A low-glass transition temperature offers possibility of low-temperature bonding. Unfortunately there does not exist any published data on the absorption coefficient through both mid- and far-infrared wavelength, but as the Teflon AF would be ideally applied as a thin film (subwavelength thickness), there is a little need to consider absorption.

Bonding procedures were first developed and tested using (100) GaAs wafers made of semi-insulated, undoped GaAs with a specific resistivity of <1.2E8 (American Xtal). A layer of Teflon-amorphous fluoropolymer (Teflon-AF) solution with 1% concentration in a perfluorinated solvent (FC-75) was deposited on the GaAs wafer surface using a commercial spinner machine. Then the wafer was heated horizontally in a furnace to 160 °C in order to evaporate the solvent. Additionally, baking Teflon-AF above its glass transition temperature of 320 °C softened the Teflon layer and produced a smooth layer. By changing the revolving speed of the spinner from 1000 to 4000 rpm, the thickness of the solid layer was controlled in the 10 to 3 μm range, respectively. The Teflon coated samples were then cooled down to the room temperature and stacked face-to face, i.e., GaAs-Teflon/GaAs-Teflon/GaAs, in intimate physical contact between two quartz optical flats. The assembly was placed in the torque mechanical press and pressure sufficient for bow compensation was applied to assembly of stack. For bonding, the compressed assembly was annealed in the furnace for approximately one hour at 320 °C. Then the temperature was reduced to 100 °C and below that point was gradually decreased with a rate 3 °C per minute to room temperature. The average Teflon layer thickness in the stack of bonded GaAs wafers was in the range of 3-5 μm.

The optical properties of each TB-GaAs wafer stack fabricated according to this procedure were measured by using a CO₂ laser beam. The results are presented below.

2.3 Transmission measurements TB-GaAs stacks at 10.6 μm

A single longitudinal mode Gaussian beam generated by a TEA CO₂ laser was used for transmission measurements. An 8 mm beam was sent through a (100) GaAs wafer stack with a 25x25 mm² aperture. The stack was mounted on a rotation stage to allow variation of the incident angle of the 10.6 μm beam. The stack of two wafers demonstrated full cancellation of Fresnel losses and its transmission was the same as transmission of a single wafer. Figure 2 depicts transmission of the 10.6 μm beam through a stack of 5 and 6 wafers along with the simulated curves. The plot shows a good agreement between simulations done using Eq. (3) and measured transmission peaks location during the angle scan. However, the width (FWHM) of transmission peaks is larger in experiment than in simulations. This can be attributed to using a slightly diverging CO₂ laser beam for measurements whereas a plane wave approximation was used for simulations. A transmission of about 0.9 (0.85) was achieved for a stack of 5 (6) wafers at an optimal angle of 12.5 degrees. Transmission degraded slowly with increasing the number of wafers and reached 0.75 for a stack of ten wafers. Thus the etalon transmission pattern and high transmission show that this structure behaves as a stack of etalons. Therefore, the fabrication procedure developed succeeded in producing a thin Teflon layer that allowed cancellation of Fresnel losses and the pressure applied mostly compensated the inherent bow of the GaAs wafers.
Fig. 2. Transmission of 5- and 6-(100) GaAs wafer stacks as a function of the angle of incidence.

Figure 3 shows images of the incident Gaussian beam along with the beam emerging from 6- and 10-wafer stacks taken with a pyrocam (12.5x12.5 mm² pyroelectric detector, Spiricon, Inc). The profiles remain relatively Gaussian, however, demonstrating that the fabrication method is useful for large aperture structures. Losses of some peripheral rays occur due to the CO₂ laser beam divergence that clearly points out necessity to use a parallel pump beam for these multi-etalon structures.

Fig. 3. 12.5x12.5 mm images of the CO₂ laser beam transmitted through the TB-GaAs stacks.

3. THz generation in a QPM TB-GaAs pumped by CO₂ laser lines

GaAs is a 43m symmetry class crystal with the maximum nonlinear coefficient is for radiation polarized along a <111> direction. Therefore, the (110) wafers were chosen for the THz DFG because they provide the maximum effective electro-optic nonlinear coefficient of 50 pm/V for propagation normal to the input face [9]. Adjacent wafers were rotated by 180° to alternate the sign of the effective nonlinear coefficient. A pilot QPM TB-GaAs structure was made of a stack of up to five diced (110) GaAs wafers with an aperture of 20x20 mm². The average Teflon layer thickness was measured to be approximately 5 μm.

For THz DFG in a QPM TB-GaAs the CO₂ laser lines had to be chosen carefully. Different lines had different transmission peak locations due to etalon effects. As was discussed in Sec. 2.1, the transmission profile of a multiple etalon system is defined by a GaAs wafer, which is an etalon with the highest refractive index and largest thickness. In Fig. 4 we present typical transmission measurements in a single (110) GaAs wafer for the 10.3 μm (the 10R(16) transition) and 10.6 μm (the 10P(20) transition) lines of the CO₂ laser. As apparent at 11.5° transmission for both lines is 0.75. It should be noted that maximum transmission on any of these lines was around 0.85. This transmission behavior for a single (110) wafer was different compared to close to 100% transmission in a (100) GaAs wafer.
described earlier. This is mainly due to a nonparallelism of the (110) wafers used in the experiment, as a result etalon transmission losses for the wedge could not be compensated by the angle tuning. The maximum transmission a stack of two wafers was about 0.82, which dropped to 0.42 for the stack of five wafers tuned to 11.5 degrees. Also almost flat angle dependence of transmission for the pilot TB-GaAs structure was observed indicating a very low finesse of the etalon.

A dual-beam TEA CO$_2$ laser described elsewhere [1] was utilized for THz DFG measurements. The laser was tuned to the 10.6 and 10.3 μm lines and two optical beams were combined and sent collinearly into the TB-GaAs wafer stack. A THz frequency of 0.87 THz, or a wavelength of 343 μm, was produced with this pair of lines. THz radiation was collected by an off-axis parabolic mirror, separated from the pump radiation by a slab of Teflon, and detected by a Golay cell. Incident intensities for a 5 mm diameter laser beam were around 10-15 MW/cm$^2$ for each line. Figure 5 shows generated THz intensity versus number of bonded wafers as well as theoretical calculations made in a plane wave approximation. A five wafer stack produced about 0.7 μJ of THz radiation, or approximately 9 W/cm$^2$. When the measured transmission losses are taken into account, the calculated values (represented by triangles in Fig. 5) are in a good agreement with the experimental data. The calculation used an estimated wafer thickness 730 μm. This demonstrates that there is in fact a length-squared dependence
dependence of generated THz power as expected from a nonlinear DFG process. To
demonstrate potential of the QPM TB-GaAs, we present a theory curve (solid curve in Fig. 5)
assuming an intensity of 10 MW/cm² for each pump line and transmission losses
encoding to a single wafer. If transmission is improved by using wafers with a
nonparallelism better than 30 angle seconds, this TB-GaAs structures can generate
significantly higher THz power. Additional increase in the THz conversion efficiency may be
potentially obtained due to the enhancement of the 10-μm electrical field inside the GaAs
structure for close to ideal etalon conditions. In the pilot (110) GaAs wafer structure these
etalon related effects were weak and not measurable.

4. Summary
This paper describes the development of a new low-temperature wafer bonding technology
using Teflon AF as an interboundary layer. A bonding procedure was developed for GaAs
wafers and applied for fabrication of large-aperture stacks of up to ten 25x25 mm² (100) GaAs
wafers and up to five 20x20 mm² (110) GaAs wafers. A use of the Teflon intermediate layer
minimizes the pressure applied to the surface for bow compensation and requires temperatures
much below the transition point for a semi-insulated GaAs. It opens possibility of fabricating
very large aperture (up to 10 cm in diameter) TB structures.

Free standing TB-GaAs stacks were optically characterized by using a CO₂ laser beam.
Transmission of 100 % for a 2-wafer stack and 75 % for a 10-wafer stack were observed for
(100) GaAs wafers. The bonded stacks were modeled as a multiple Fabry-Perot etalon
structure and its optical behavior was simulated. Measurements of transmission were in very
good agreement with simulations.

A pilot large-aperture QPM device for THz DFG was made from (110) GaAs wafers
bonded according to the procedure developed. Approximately 1 μJ of 343 μm radiation was
obtained via difference frequency mixing of the CO₂ laser lines in the TB-GaAs structure.
Simple, drop-off QPM devices based on TB GaAs and InSb structures pumped with a large
aperture CO₂ laser beam can provide a practically useful high-power THz source for a variety
of applications.

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