# Fabrication of Highly Efficient Mid-Infrared Bragg Mirrors from IV-VI Semiconductors

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High reflectivity IV-VI semiconductor Bragg mirrors were grown by molecular beam epitaxy on BaF<sub>2</sub> (111) substrates. The  $\lambda/4$  layer pairs consisted of a low refractive index Pb<sub>1-x</sub>Eu<sub>x</sub>Te layer with  $x_{Eu} = 6$  % and a higher index pseudoalloy PbTe/Pb<sub>1-x</sub>Eu<sub>x</sub>Te superlattice with average  $x_{Eu}$  of 1 %. Mirrors with stop bands in the range between 4 and 6  $\mu$ m were obtained with reflectivities as high as 99%.

## 1. Introduction

Due to their favorable electronic band structure [1], lead salt (IV-VI) lasers have long dominated the field of mid- and far infrared band gap lasers in the 3 - 30  $\mu$ m region. The inherent wavelength tunability of these lasers have made them an ideal tool for high resolution infrared (IR) spectroscopy [2]. While these lasers are usually grown on lead salt substrates, (111) oriented BaF<sub>2</sub> has proven to be an excellent alternative as substrate material for lead salt heterostructures [3]. In comparison to lead salt substrates, BaF<sub>2</sub> exhibits a much higher thermal conductivity and mechanical hardness. This would allow significant improvements in heat dissipation during laser operation and facilitate device processing procedures. In the present work, we have explored the possibilities for molecular beam epitaxy (MBE) of highly efficient lead salt based mid-infrared Bragg mirror structures required for realization of vertical cavity surface emitting diode lasers in the 4 – 6  $\mu$ m spectral region. Such devices, grown on readily available BaF<sub>2</sub> substrates, have great potentials for reducing threshold currents and increasing the operation temperatures of IV-VI lasers.

## 2. The Design of Bragg Mirrors

The basic design for high reflectivity Bragg interference mirrors is the stacking of two alternating layers with different refractive index and with a thickness equal to a quarter optical wavelength. The mirror characteristics are then governed by the refractive index contrast and the number of  $\lambda/4$  pairs. In the present work we have focused on Pb<sub>1-x</sub>Eu<sub>x</sub>Te of different composition for realization of MIR Bragg mirrors that are compatible with PbTe as active material. For Pb<sub>1-x</sub>Eu<sub>x</sub>Te the refractive index below the fundamental absorption decreases with increasing Eu content and the energy gap Eg increases with dEg/dx = 3.5 eV [4]. Using Pb<sub>1-x</sub>Eu<sub>x</sub>Te layer pairs with alternating lower/higher Eu contents, Bragg mirrors without absorption of light emitted from a PbTe active region can be made. In order to keep the lattice mismatch as low as possible the Eu contents must be restricted to below about 10 %. Here, we have chosen a layer pair with Eu contents of 1 and 6 %. The 1 % layer was realized in the form of a short

period PbTe/Pb<sub>1-x</sub>Eu<sub>x</sub>Te (x = 6 %) superlattice pseudoalloy with 50 Å period and thickness ratio of 5:1.

For the mirror design, the optical properties of the individual layers were determined by FTIR transmission measurements of a thick  $Pb_{0.94}Eu_{0.06}Te$  reference layer and a 3  $\mu$ m pseudoalloy superlattice.



Fig. 1: Refractive index versus wavenumber at 300 K for two  $Pb_{1-x}Eu_xTe$  reference layers ( $x_{Eu} = 1$  and 6%). The arrows labeled #A and #B indicate the intended stop band positions for the Bragg mirror samples.



Fig. 2: Theoretical reflectivity at the stop band center (1570 cm<sup>-1</sup>) as a function of the number of  $\lambda/4$  pairs for Bragg mirrors of Pb<sub>1-x</sub>Eu<sub>x</sub>Te layers. (a) for incidence of the light from PbTe as active region, (b) incidence of light from air. The full and open symbols correspond to the different layering sequence where "2" corresponds to 6% Eu, and "1" to 1% Eu.

The optical constants (absorption constant  $\alpha(\omega)$  and refractive index  $n(\omega)$ ) were derived from the fit of the transmission spectra using a method described in Ref. [4]. As shown in Fig. 1, below E<sub>g</sub>, the refractive index is determined by a constant dielectric background  $\varepsilon_{\infty}$  and, close to E<sub>g</sub>, by the Kramers-Kronig contribution due to the changing absorption constant. The refractive index contrast therefore varies significantly with  $\omega$ and is largest close to the band gap of the 1 % pseudoalloy layer.

For the mirror design we calculated the reflectivity of various mirror structures using the transfer matrix method and the experimental  $\alpha(\omega)$  and  $n(\omega)$  values as input parameters. As shown in Fig. 2, we find that for incidence of light from an optically denser medium (e.g. PbTe) 18 mirror pairs are required to achieve a reflectivity of 95 % and 28 pairs for R above 98 % at a wavelength of 5  $\mu$ m. For incidence of light from air, due to the additional phase shift, the stacking sequence of the mirror pair has to be reversed.

## 3. Results

The PbTe/Pb<sub>1-x</sub>Eu<sub>x</sub>Te multilayers were deposited on cleaved BaF<sub>2</sub> (111) by molecular beam epitaxy. The ternary composition was determined by the PbTe to Eu beam flux ratio, and an excess Te<sub>2</sub> flux was used to retain the correct stoichiometry of the ternary. All flux rates were calibrated with a quartz crystal microbalance that could be moved into the substrate position. To minimize the growth time for the thick multilayer stacks, growth rates of about 2  $\mu$ m/h were used.

In the following we show the results for a Bragg mirror designed to match the 77 K PbTe band gap (217 meV or 1750 cm<sup>-1</sup>). The number of  $\lambda/4$  pairs was 32. From Fig. 1, the refractive index contrast is 6 %, which is comparable to that for Bragg mirrors of III-V materials [5].



Fig. 3: (a) Measured (dots) and calculated (full line) transmission spectra for a 32 pairs Bragg mirror at 77 K with a stop band at  $v_m = 1740 \text{ cm}^{-1}$  (emission wavenumber of PbTe at 77 K), (b) Cross sectional scanning electron micrograph of a 20 pair Bragg mirror mesa structure. The cleavage edge was selectively etched to reveal the layers with different Eu composition.

Due to the pseudoalloy superlattices the required total number of individual layers is 3230 and the total thickness of the Bragg mirror is 16 µm. The sequence of the  $\lambda/4$  pairs, starting with the 6 % Pb<sub>1-x</sub>Eu<sub>x</sub>Te on BaF<sub>2</sub>, was chosen to yield a high reflectivity for incidence of the light from air. The structural properties of the Bragg mirrors were determined by high resolution x-ray diffraction and scanning electron microscopy (SEM). The right hand side of Fig. 3 shows a cross sectional SEM micrograph of a 20 pair Bragg mirror sample with mesa structure, demonstrating the high lateral homogeneity and smooth interface structure of the samples. The cleavage edge was selectively etched to reveal the  $\lambda/4$  layers with different Eu composition using a CH<sub>4</sub>/H<sub>2</sub> plasma etch. The dark layers correspond to the  $\lambda/4$  layers with the lower average Eu content, but the short period superlattice structure of this pseudoalloy layer can be resolved only by x-ray diffraction.

The FTIR transmission spectra of the 32 pair Bragg mirror sample measured at 77 K is shown in Fig. 3 (a). The main features are (1) the Fabry-Perot interference fringes due to the multiple reflections at the sample surface and the layer/BaF<sub>2</sub> interface, (2) the stop band of low transmittance of the Bragg mirror at  $v_m = 1740$  cm<sup>-1</sup>, and (3) the high frequency cut off at around 2150 cm<sup>-1</sup> due to the absorption edge of the 1 %  $Pb_{1-x}Eu_xTe$ pseudo-alloy. The spacing of the fringes corresponds to the inverse optical thickness of the multilayer stack. The large refractive index jump at the  $BaF_2$  interface also enhances the Bragg reflectivity of the samples. The transmission in the stop band region is below 0.5 %, which indicates a mirror reflectivity of better than 99 % (negligible absorption losses in the stop band region). Bragg mirrors with 20  $\lambda/4$  pairs typically exhibit a larger stop band transmission with corresponding reflectivity of about 93 %. Both values agree very well with our calculations. In spite of the large total thicknesses and large number of layers we have obtained excellent control and reproducibility of the layer thicknesses and composition. This opens promising perspectives for fabrication and applications of IV-VI mid-infrared resonant cavity light emitting diodes and vertical cavity surface emitting laser devices.

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