

# Midinfrared Continuous-Wave Photoluminescence of Lead Salt Structures up to Temperatures of 190 °C

M. Böberl, W. Heiss, T. Schwarzl, K. Wiesauer and G. Springholz  
Institut für Halbleiter- und Festkörperphysik, Johannes Kepler Universität,  
A-4040 Linz, Austria

Continuous-wave photoluminescence in the midinfrared for PbSe/PbEuSe and PbTe/PbEuTe multiquantum well structures as well as for PbSe and PbTe bulklike structures, excited by a semiconductor laser diode, is investigated. All samples are grown by molecular-beam epitaxy on BaF<sub>2</sub>(111) substrates under the same growth conditions. Both for the Te-based systems and for the Se-based systems, it turns out that bulklike structures show photoluminescence up to higher temperatures than multiquantum well structures. In particular, emission spectra from PbTe/PbEuTe multiquantum wells are obtained up to temperatures of 200 K and from PbSe/PbEuSe multiquantum well structures up to 60 °C whereas for bulklike PbSe photoluminescence at temperatures as high as 190 °C is demonstrated.

## Introduction

Lead salt materials are of high interest for midinfrared optical coherent emitters and detectors applied for molecular spectroscopy. The IV–VI narrow gap semiconductors have a multi-valley band structure with band extrema at the L point of the Brillouin zone. Due to the favorable mirrorlike band structure, the nonradiative Auger recombination rate is reduced by one or two orders of magnitude below that of narrow gap III–V and II–VI materials [1], [2]. Therefore, the highest continuous-wave (cw) operation temperatures (223 K) [3] of midinfrared diode lasers are achieved by lead salt devices. In spite of the fact that the use of quantum wells (QWs) as laser active media usually allows one to reduce threshold currents [4], it is remarkable that the highest cw operation temperature for lead-salt diode lasers was achieved so far by separate confinement buried heterostructure devices with bulklike active regions [3]. Recently, a comparative study of optically pumped vertical-cavity surface-emitting PbTe lasers (VCSELs) containing active regions of different dimensionality also showed no advantageous operation of QW devices over that containing a bulklike active region [5]. To rule out that the superior emission of bulklike lead-salt materials is caused by the high excitation required for laser emission, we study in this work the spontaneous emission of lead-salt structures under moderate optical excitation using a cw InGaAs pump laser diode. The results of these photoluminescence (PL) experiments performed for PbTe/Pb<sub>1-x</sub>Eu<sub>x</sub>Te and PbSe/Pb<sub>1-x</sub>Eu<sub>x</sub>Se multiple QWs (MQWs) as well as for thick bulklike PbSe and PbTe epilayers confirm the higher operation temperature achieved for bulklike samples, in contrast to observations in other semiconductor material systems [6]. In particular, for PbSe, strong luminescence is demonstrated even far above room temperature, namely, up to 190 °C.

## Experimental

The PbSe and PbTe epilayers, as well as the PbSe/PbEuSe and PbTe/PbEuTe MQW structures, were grown by molecular-beam epitaxy (MBE) on (111)-oriented BaF<sub>2</sub> sub-

strates. The 50 period PbTe/PbEuTe MQW structures have QW thicknesses between 54 Å and 120 Å and Pb<sub>0.90</sub>Eu<sub>0.1</sub>Te barrier layer thicknesses between 240 Å and 300 Å, and were grown on a 1.5–2 µm thick buffer layer of Pb<sub>0.90</sub>Eu<sub>0.1</sub>Te. For the three investigated samples, S1, S2, and S3, the QW thickness is 53, 80, and 120 Å, respectively. The 30 period PbSe/PbEuSe MQW sample has a well layer thickness of 100 Å and a Pb<sub>0.93</sub>Eu<sub>0.07</sub>Se barrier thickness of 400 Å and was grown on a 3 µm thick buffer layer of Pb<sub>0.93</sub>Eu<sub>0.07</sub>Se. The bulklike PbSe and PbTe samples have a layer thickness of 3 µm and were fabricated with and without excess group-VI flux during MBE growth. For the PL investigations, the samples were mounted in a He flow cryostat for measurements below room temperature or on a heating element for above room-temperature measurements. In both cases, the temperature was measured close to the sample with either a Si diode or a thermocouple within an accuracy of 5 K. PL experiments were performed using a 970 nm InGaAs pump laser diode with an output power of about 150 mW. The laser was focused on the sample to a spot size of about 1 mm<sup>2</sup> under an angle of about 45°. The luminescence from the sample was analyzed using a grating monochromator and recorded by an InSb detector.

## Results

In detail, the highest cw emission temperature from the PbTe/PbEuTe MQW structures is about 200 K. In contrast, for bulk-like PbTe epilayers, cw PL emission is obtained even above room temperature, as is seen in Fig. 1 (a).

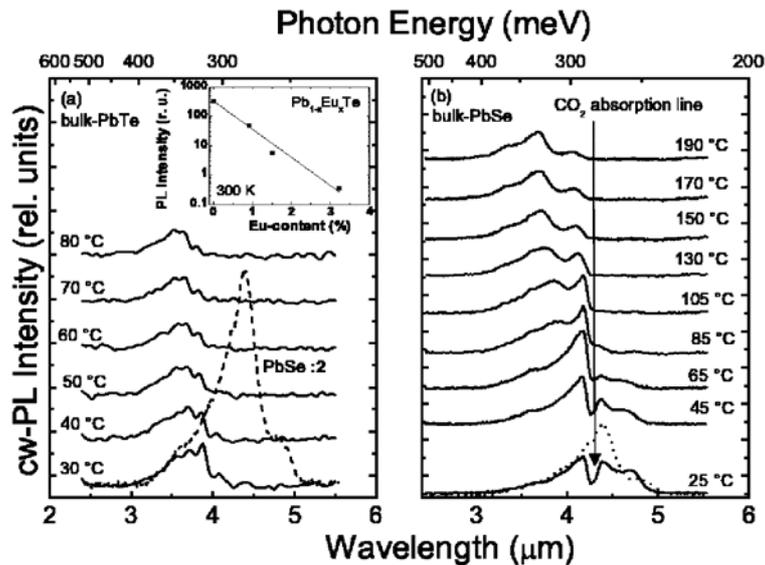


Fig. 1: (a) Temperature dependent cw PL spectra of a bulklike PbTe epilayer on BaF<sub>2</sub>(111). The inset shows the integrated room temperature PL intensity emitted from Pb<sub>1-x</sub>Eu<sub>x</sub>Te epilayers as a function of the Eu-content  $x$ . (b) PL spectra of various temperatures of the PbSe epilayer. The dashed line at 25 °C shows the PL spectrum measured in an evacuated sample chamber.

Even at 80 °C, strong PL emission between 300 meV and 400 meV is observed, indicating a higher PL efficiency for bulk-like PbTe as compared to the QWs. This surprising observation is attributed to the increased nonradiative carrier recombination in the QWs due to intermixing of Eu at the heterointerfaces, as evidenced by x-ray and secondary ion mass spectroscopy experiments [7]. This interpretation is further supported by the observation of a very rapid drop in the PL efficiency of bulk-like Pb<sub>1-x</sub>Eu<sub>x</sub>Te epi-

layers with increasing Eu content demonstrated in the inset of Fig. 1 (a), with an intensity decrease by a factor of 100 already for Eu concentrations as low as 1.5 %. We attribute this to the intermixing between the localized Eu 4f states and the valence band, leading to reduced interband matrix elements. The 4f levels are located close to the valence-band maximum and for Eu contents above 7 % the Eu 4f level energies are even above the PbTe-like valence-band maximum [8].

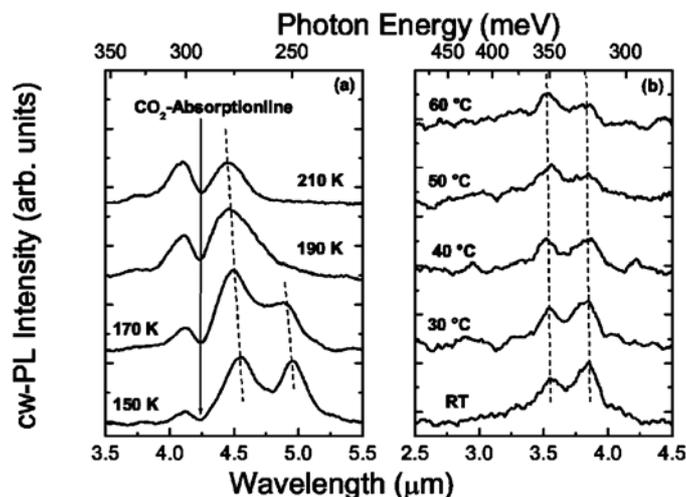


Fig. 2: Temperature dependent PL spectra of a PbSe/Pb<sub>0.93</sub>Eu<sub>0.07</sub>Se MQW structure with a well width of 100 Å.

For comparison, we have also investigated the cw PL emission behavior of the PbSe/PbEuSe material system. Figure 2 shows the observed cw-emission spectra of a 30 period PbSe/PbEuSe MQW structure with 10 nm well width below and above room temperature (Figs. 2 (a) and 2 (b), respectively). The dip in the low-temperature spectra of Fig. 2 (a) at  $\lambda = 4.23 \mu\text{m}$  is due to the strong absorption of atmospheric CO<sub>2</sub> in the open path of the PL setup. With respect to the bulk PbSe band gap, the QW emission is blueshifted by about 50 meV due to the quantum size effect. In contrast to the PbTe MQWs, cw emission from the PbSe MQW sample is obtained even up to 60 °C (see Fig. 2 (b)), which is comparable with the reported value of PbSe/PbSrSe MQWs [9]. For pure PbSe bulk-like reference layers, the corresponding above room-temperature cw-emission spectra are shown in Fig. 1 (b). Clearly, a strong band-gap cw emission is found even at temperatures up to 190 °C. This is again higher than that obtained for the PbSe/PbEuSe MQWs, which we also attribute to increased nonradiative interface recombination. Even more interesting, however, is the fact that the PbSe cw emission is also much stronger than that observed for the PbTe epilayers (Fig. 1 (a)) persisting also to much higher temperatures. Since the PbSe emission spectra are strongly distorted by the strong absorption band of CO<sub>2</sub> in the optical path of the measurement setup, we have also measured the PL emission in an evacuated sample chamber. As is indicated by the dashed line in Fig. 1 (b), the PL emission obtained such is even stronger (increased by about a factor of 2) and shows a much more Gaussian-type line shape. It is centered at about 282 meV (4.3  $\mu\text{m}$ ), corresponding to the PbSe band gap at 30 °C as determined from FTIR transmission measurements on this sample.

For a direct comparison of the PbTe and PbSe emission, the measured room-temperature PbSe PL spectrum is plotted as a dotted line on top of the PbTe data in Fig. 1 (a) on the same intensity scale. Clearly, the PbSe emission is by about a factor of 20 stronger than the PbTe emission. We can exclude that this marked difference is due to differences in sample quality. This leads to the important conclusion that the

higher PL efficiency of PbSe is really an intrinsic effect. It can be explained by the higher band edge density of states of PbSe as compared to PbTe, but also by the significantly lower nonradiative Auger recombination rate of PbSe as previously reported by Klann *et al.* [2]. The latter is closely related to the factor of 5 smaller effective mass anisotropy of the conduction and valence bands of PbSe as compared to PbTe.

## Conclusion

To conclude, our systematic PL investigations have clearly demonstrated above room-temperature cw emission in the midinfrared spectral region from various lead salt structures. The comparison of MQW and bulk-like samples revealed a significantly higher cw-emission efficiency of the bulk-like samples. In particular, for PbSe epilayers, strong PL emission was demonstrated up to a temperature as high as 190 °C, whereas the highest emission temperature of MQWs was only about 60 °C. This explains the fact that until now no superior performance of lead-salt QW lasers could be achieved as compared to their bulk-like counterpart (see, e.g., Ref. 5). On the other hand, the high spontaneous cw-emission temperatures observed, in particular, for the PbSe system clearly demonstrates that there are still great potentials for significant improvements of lead salt midinfrared lasers.

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## References

- [1] P. C. Findlay, *et al.*: “Auger recombination dynamics of lead salts under picosecond free-electron-laser excitation”, *Phys. Rev. B*, Vol. 58, 1998, p. 12908.
- [2] R. Klann, *et al.*: “Fast recombination processes in lead chalcogenide semiconductors studied via transient ...”, *J. Appl. Phys.*, Vol. 77, 1995, p. 277.
- [3] Z. Feit, M. McDonald, R. J. Woods, V. Archambault, and P. Mak: “Low threshold PbEuSeTe/PbTe separate confinement buried heterostructure diode laser”, *Appl. Phys. Lett.*, Vol. 68, 1996, p. 738.
- [4] Z. Alferov, in *Nano-Optoelectronics: Concepts, Physics, and Devices*, edited by M. Grundmann (Springer, Berlin, 2002), p. 4.
- [5] J. Fürst, *et al.*: “Midinfrared IV-VI vertical-cavity surface-emitting lasers with zero-, two- and three-dimensional systems ...”, *Appl. Phys. Lett.*, Vol. 81, 2002, p. 208.
- [6] G. Biwa, H. Yaguchi, K. Onabe, and Y. Shiraki: “PL and PLE spectroscopy of GaPAsN/GaP lattice-matched MQW structures”, *J. Cryst. Growth*, Vol. 195, 1998, p. 574.
- [7] G. Springholz, A. Holzinger, H. Krenn, H. Clemens, G. Bauer, H. Böttner, P. Norton, and M. Maier: “Interdiffusion in PbEuSe/PbSe multi-quantum-well structures”, *J. Cryst. Growth*, Vol. 113, 1991, p. 593.
- [8] M. Iida, T. Shimizu, H. Enomoto, and H. Ozaki: “Experimental Studies in the Electronic Structure of PbEuTe,” *Jpn. J. Appl. Phys.*, Part 1, Vol. 32, 1993, p. 4449.
- [9] P. J. McCann, K. Namjou, and X. M. Fang: “Above-room-temperature continuous-wave mid-infrared photoluminescence from PbSe/PbSrSe quantum wells”, *Appl. Phys. Lett.* Vol. 75, 1999, p. 3608.