Growth, steady-state, and time-resolved photoluminescence study of CdTe/MgCdTe double heterostructures on InSb substrates using molecular beam epitaxy

Michael J. DiNezza, Xin-Hao Zhao, Shi Liu, Alexander P. Kirk, and Yong-Hang Zhang

Citation: [Applied Physics Letters](#) **103**, 193901 (2013); doi: 10.1063/1.4828984

View online: <http://dx.doi.org/10.1063/1.4828984>

View Table of Contents: <http://scitation.aip.org/content/aip/journal/apl/103/19?ver=pdfcov>

Published by the [AIP Publishing](#)

Articles you may be interested in

[Minority carrier lifetime of lattice-matched CdZnTe alloy grown on InSb substrates using molecular beam epitaxy](#)
J. Vac. Sci. Technol. B **33**, 011207 (2015); 10.1116/1.4905289

[Determination of CdTe bulk carrier lifetime and interface recombination velocity of CdTe/MgCdTe double heterostructures grown by molecular beam epitaxy](#)
Appl. Phys. Lett. **105**, 252101 (2014); 10.1063/1.4904993

[Time-resolved and excitation-dependent photoluminescence study of CdTe/MgCdTe double heterostructures grown by molecular beam epitaxy](#)
J. Vac. Sci. Technol. B **32**, 040601 (2014); 10.1116/1.4878317

[CdSe/CdTe type-II superlattices grown on GaSb \(001\) substrates by molecular beam epitaxy](#)
Appl. Phys. Lett. **100**, 121908 (2012); 10.1063/1.3697676

[Steady-state and time-resolved photoluminescence from relaxed and strained GaN nanowires grown by catalyst-free molecular-beam epitaxy](#)
J. Appl. Phys. **103**, 124309 (2008); 10.1063/1.2940732

High-Voltage Amplifiers

- Voltage Range from $\pm 50\text{V}$ to $\pm 60\text{kV}$
- Current to 25A

Electrostatic Voltmeters

- Contacting & Non-contacting
- Sensitive to 1mV
- Measure to 20kV

ENABLING RESEARCH AND
INNOVATION IN DIELECTRICS,
ELECTROSTATICS,
MATERIALS, PLASMAS AND PIEZOS

www.trekinc.com

TREK, INC. 190 Walnut Street, Lockport, NY 14094 USA • Toll Free in USA 1-800-FOR-TREK • (t):716-438-7555 • (f):716-201-1804 • sales@trekinc.com

Growth, steady-state, and time-resolved photoluminescence study of CdTe/MgCdTe double heterostructures on InSb substrates using molecular beam epitaxy

Michael J. DiNezza,^{1,2} Xin-Hao Zhao,^{1,3} Shi Liu,^{1,2} Alexander P. Kirk,^{1,2} and Yong-Hang Zhang^{1,2,a)}

¹Center for Photonics Innovation, Arizona State University, Tempe, Arizona 85287, USA

²School of Electrical, Computer and Energy Engineering, Arizona State University, Tempe, Arizona 85287, USA

³School for Engineering of Matter, Transport and Energy, Arizona State University, Tempe, Arizona 85287, USA

(Received 4 July 2013; accepted 21 October 2013; published online 5 November 2013)

CdTe/MgCdTe double heterostructures (DHs) are grown on InSb substrates using molecular beam epitaxy and reveal strong photoluminescence with over double the intensity of a GaAs/AlGaAs DH with an identical layer structure design grown on GaAs. Time-resolved photoluminescence of the CdTe/MgCdTe DH gives a Shockley-Read-Hall recombination lifetime of 86 ns, which is more than one order of magnitude longer than that of typical polycrystalline CdTe films. These findings indicate that monocrystalline CdTe/MgCdTe DHs effectively reduce surface recombination, have limited nonradiative interface recombination, and are promising for solar cells that could reach power conversion efficiencies similar to that of GaAs. © 2013 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4828984>]

Recently, thin-film polycrystalline CdTe solar cells have shown impressive improvements in device performance and market acceptance, only second to that of Si cells. Polycrystalline CdTe solar cells have demonstrated efficiencies over 19% under AM1.5G illumination at 298 K,¹ while monocrystalline GaAs solar cells have reached a single junction record efficiency of 28.8%,¹ which is close to the detailed balance limit of 33.2%. Our recent modeling results have shown that monocrystalline CdTe solar cells can potentially achieve efficiencies greater than 25% based on an 86 ns bulk Shockley-Read-Hall (SRH) carrier lifetime.² In order to understand the fundamental physics that limits the record efficiencies of polycrystalline and monocrystalline CdTe solar cells, it is highly desirable to have a model system in which the effects of various kinds of defects can be carefully studied in a controlled fashion. A double heterostructure (DH) with type-I band edge alignment provides a model system with reduced interface and surface recombination so that the middle CdTe region can be isolated and studied. However, few efforts have been undertaken to explore the use of monocrystalline CdTe and related DHs grown on lattice-matched substrates for such a purpose. In this letter, we report the molecular beam epitaxial growth and characterization of a CdTe/MgCdTe DH grown on an InSb substrate using X-ray diffraction (XRD), steady-state photoluminescence (PL) and time-resolved PL (TRPL) measurements. Also, the CdTe/MgCdTe DH is compared with a GaAs/AlGaAs DH grown on GaAs and a plain CdTe layer grown on InSb using steady-state PL measurements.

The layer structures of all three samples used in this study are shown in Fig. 1. The two DH samples A and B consist of a 1 μm thick undoped (middle) layer of either CdTe or

GaAs sandwiched between two 30 nm thick $\text{Mg}_{0.18}\text{Cd}_{0.82}\text{Te}$ or $\text{Al}_{0.30}\text{Ga}_{0.70}\text{As}$ barrier layers, respectively. Both CdTe/MgCdTe and GaAs/AlGaAs have a type-I band edge alignment, and the CdTe/MgCdTe heterojunction valence band offset is 30% of the difference in the bandgaps of the two materials.³ The composition of each barrier is chosen so that the conduction and valence band offsets are greater than several kT , so as to reduce the number of photogenerated carriers from reaching to the top surface and the bottom buffer layer. For sample A, the CdTe/ $\text{Mg}_{0.18}\text{Cd}_{0.82}\text{Te}$ heterojunction has conduction and valence band offsets of approximately 188 meV and 80 meV, respectively, assuming a MgTe bandgap of 3.0 eV.³ For sample B, the GaAs/ $\text{Al}_{0.30}\text{Ga}_{0.70}\text{As}$ barriers have conduction and valence band offsets of 237 meV and 138 meV, respectively. A 10 nm CdTe and GaAs layer is used to cap the MgCdTe and the AlGaAs top barrier layers, respectively, in order to prevent them from exposure to air.⁴ The cap layers have a relatively negligible impact on the PL intensity from the middle CdTe and GaAs layers from a comparison point of view. Sample C consists of an undoped 1.57 μm thick CdTe single layer grown on InSb. This sample is used to determine the effectiveness of the DH for photogenerated carrier confinement.

The growth is carried out using a dual-chamber VG V80H molecular beam epitaxy (MBE) system equipped with two separate III-V and II-VI growth chambers and an ultra-high vacuum (UHV) transfer chamber. The purity of all source materials is at least 7N, with the exception of Mg which is 6N. Effusion cells are used for the group-II materials, and a valved effusion cell is used for Te. The surface reconstruction during growth is monitored by reflection high energy electron diffraction (RHEED), and the substrate temperature is measured using 1550 nm and 950 nm infrared pyrometers for the InSb and GaAs wafers, respectively. RHEED oscillations observed during the growth of CdTe are

^{a)}Electronic mail: yhzhang@asu.edu

FIG. 1. Layer structures for the (a) CdTe/MgCdTe DH, (b) GaAs/AlGaAs DH, and (c) plain CdTe samples.

used to calibrate the 1:1 Cd/Te flux ratio based on the saturation of the growth rate with increasing Cd flux.⁵

The growth procedure starts with thermal deoxidation of III-V substrates under a group-V overpressure, followed by the growth of a 500 nm thick III-V buffer layer. Typical growth conditions are used for all III-V layers,⁶ and streaky RHEED reconstructions are observed. Samples A and C are transferred directly to the II-VI growth chamber under UHV after the InSb buffer layer growth. Immediately prior to CdTe growth, InSb surfaces are exposed to a Cd flux for several minutes to prevent the formation of a group III-VI alloy at the interface. A CdTe buffer layer is then grown using an initial Cd/Te flux ratio of 3.5 in order to further prevent the formation of In₃Te₂ at the interface.⁷ After 2 min of growth, the flux ratio is then reduced to 1.5. Upon initiation of CdTe growth on InSb, the RHEED pattern becomes slightly hazy as the surface reconstruction transitions from InSb to CdTe. After 10 min of growth the pattern becomes streaky, as shown in Fig. 2. Both (2 × 1) and c(2 × 2) RHEED reconstructions are observed which indicate a Cd-rich growth condition.⁸ The substrate temperature for samples A and C are 265 °C and 280 °C, respectively, and the growth rate for both samples is 9.6 nm/min as determined by RHEED oscillations. The Cd and Te fluxes are kept constant during the MgCdTe layer growth, since Mg has a larger sticking coefficient and will displace the excess Cd.⁹

The structural properties of sample A are investigated using high-resolution XRD measurements. An ω -2 θ scan is taken in the vicinity of the (004) diffraction peak, and reciprocal space maps (RSMs) are taken in the vicinity of the (004) and (115) diffraction peaks. A narrow diffraction peak with a FWHM of 19 arc sec is observed for CdTe (004)

FIG. 2. RHEED pattern along the [011] direction recorded 10 min after the start of CdTe growth on InSb. Half order reconstructions are also seen along the [001] direction, giving a (2 × 1) + c(2 × 2) pattern, which indicates a Cd-rich surface.

diffraction, as shown in Fig. 3. Pendellösung fringes of the CdTe layer are clearly observed, indicating smooth DH interfaces. A weak and broad MgCdTe peak is also observed, which is expected for the thin barrier layers. Interference fringes for MgCdTe give a thickness of approximately 30 nm, which is in excellent agreement with the designed thickness. The (115) RSM in Fig. 4 shows diffraction peaks that are aligned parallel to the vertical axis, and a similar pattern is observed for the (004) RSM. The (004) RSM indicates that there is no tilt present in the crystal structure, while the (115) RSM indicates that the in-plane lattice constants of CdTe and MgCdTe are matched to InSb. Therefore, the CdTe and MgCdTe layers are coherently strained. The composition of the MgCdTe barrier layer is determined using the biaxial strain relationship

$$\frac{a_z - a_o}{a_o} = -\frac{2C_{12}}{C_{11}} \times \frac{a_x - a_o}{a_o}, \quad (1)$$

where a_z is the measured lattice constant of MgCdTe in the growth direction determined using the Bragg equation and the (004) ω -2 θ scan, a_o is the lattice constant of unstrained MgCdTe, C_{11} and C_{12} are elastic stiffness coefficients of

FIG. 3. XRD ω -2 θ scan of the CdTe/MgCdTe double-heterostructure sample taken in the vicinity of the (004) diffraction peak.

FIG. 4. RSM of the CdTe/MgCdTe double heterostructure in the vicinity of the (115) peak with contours plotted on a logarithmic scale. A similar RSM is observed in the vicinity of the (004) peak. The alignment of the InSb, CdTe, and MgCdTe diffraction peaks parallel to the vertical axis indicates that all epilayers are coherently strained to the InSb substrate.

MgCdTe, and a_x is the in-plane lattice constant of MgCdTe which is equal to that of bulk InSb. The values of C_{11} and C_{12} for $\text{Mg}_{0.18}\text{Cd}_{0.82}\text{Te}$ are $5.34 \times 10^{11} \text{ dyn/cm}^2$ and $3.68 \times 10^{11} \text{ dyn/cm}^2$, respectively, which are obtained using linear interpolation of the elastic stiffness coefficients for CdTe and MgTe.¹⁰ Also, it is assumed that MgTe has a lattice constant of $6.42 \text{ \AA}$.¹¹ Based on the XRD scans and parameters above, the Mg composition is determined to be $18\% \pm 1\%$.

A comparison of the room-temperature steady-state PL of all three samples is shown in Fig. 5. The pump laser wavelength is chosen to be 532 nm , since the difference between the absorption coefficients of CdTe and GaAs at this wavelength is less than 6% .¹² The laser excitation power density for samples A and B is 0.10 W/cm^2 , while sample C needed a stronger excitation of 2.1 W/cm^2 to increase the signal to noise ratio. As shown in the figure, sample A has over double the PL intensity as sample B. The strong PL intensity from sample A is attributed to both improved carrier confinement provided by the CdTe/MgCdTe DH and the use of a lattice matched substrate to obtain high quality monocrystalline material. The effectiveness of the carrier confinement is evident from the relatively low PL intensity of sample C, which gives over three orders of magnitude lower PL intensity compared with sample A when measured with the same excitation density. This comparison shows that the MgCdTe barriers have a low interface state density and effectively reduce both surface recombination and transport of photogenerated carriers to the narrow gap (0.17 eV) InSb buffer layer. Further improvements in CdTe material quality with respect to GaAs are expected by optimizing the growth temperature and flux ratios.

The photoluminescence decay of sample A shown in Fig. 6 is obtained using time-correlated single photon counting TRPL measurements. A 530 nm pulsed laser operating at

FIG. 5. Room-temperature PL spectra of the (a) CdTe/MgCdTe and (b) GaAs/AlGaAs DHs grown on InSb and GaAs, respectively, as well as a (c) plain CdTe layer grown on InSb. A higher excitation density is used for the plain CdTe layer to improve the signal-to-noise ratio, and the curve is magnified by a factor of 15. The PL intensity of sample A is over three orders of magnitude stronger compared with sample C when measured under the same excitation power density.

an average power density of 0.14 W/cm^2 is used. The pulse width is 6 ps , and the repetition rate is 2 MHz . These conditions give an initial carrier concentration of less than $1 \times 10^{15} \text{ cm}^{-3}$, which is determined by assuming a normal incidence reflection loss of 27% and a cap layer absorption loss of 7% at 530 nm .¹² A spectrometer with 16 nm resolution is set to a detection wavelength of 820 nm . Fig. 6 shows a decay time of 86 ns when fitted using a single exponential decay model in the range of 90 ns – 400 ns . The absence of a second decay time constant in this region suggests that the 86 ns decay time is the SRH carrier lifetime of the middle

FIG. 6. Time-resolved photoluminescence decay of the CdTe/MgCdTe double heterostructure grown on InSb at room temperature. The initial carrier concentration is less than $1 \times 10^{15} \text{ cm}^{-3}$. A Shockley-Read-Hall lifetime of 86 ns is extracted using a single exponential decay model.

CdTe layer. Furthermore, photon recycling is negligible because the InSb substrate absorbs luminescent photons and SRH recombination likely dominates in this sample. This carrier lifetime is much longer than that of polycrystalline CdTe solar cells, which is typically less than 6 ns.¹³ It should be noted that as-deposited polycrystalline CdTe film has a carrier lifetime of only 0.087 ns, and several processing techniques are needed to improve the material quality.¹³ The carrier lifetime of sample A is also longer than the current record of 66 ns reported for bulk single-crystal CdTe measured using a sub-bandgap two-photon method at 850 nm.¹⁴ This method selectively excites the bulk material several hundred microns below the surface, therefore surface recombination is negligible and a fair comparison can be made to our sample.

In summary, CdTe/MgCdTe DHs grown on InSb substrates show a SRH lifetime of 86 ns and over twice the PL intensity as an identical GaAs/AlGaAs structure grown on GaAs. Such a fair comparison is enabled by the use of identical DH layer designs grown on lattice-matched substrates to provide strong photogenerated carrier confinement and to reduce the misfit dislocation density, respectively. Coherently strained high quality MgCdTe barrier layers with relatively low interface recombination are evident from the over three order of magnitude improvement in PL intensity for CdTe/MgCdTe DHs compared with a plain CdTe 1.57 μm thick layer grown on InSb. The strong PL intensity and long PL decay time show the promise of monocrystalline CdTe DHs for high efficiency solar cell applications. Furthermore, this CdTe/MgCdTe DH sample design provides a model system in which the effects of various defects in CdTe layers can be investigated in a controlled fashion.

The authors would like to thank Bas A. Korevaar and Aharon Yakimov at General Electric Global Research Center and Su Lin at ASU for assistance with TRPL measurements. This work was partially supported by AFOSR (Grant No. FA9550-12-1-0444), Science Foundation Arizona (Grant No.

SRG 0339-08), and NSF (Grant No. 1002114). M.J.D. was supported by the National Science Foundation Graduate Research Fellowship (Grant No. DGE-0802261) and A.P.K. is supported by the Bisgrove Postdoctoral Scholars program. The authors gratefully acknowledge the use of facilities in the LeRoy Eyring Center for Solid State Science at Arizona State University.

¹M. A. Green, K. Emery, Y. Hishikawa, W. Warta, and E. D. Dunlop, *Prog. Photovoltaics* **21**, 827 (2013).

²A. P. Kirk, M. J. DiNezza, S. Liu, X.-H. Zhao, and Y.-H. Zhang, in *Proceedings of the 39th IEEE Photovoltaic Specialists Conference, Tampa, Florida, USA*, 16–21 June, 2013.

³A. Waag, F. Fischer, Th. Litz, B. Kuhn-Heinrich, U. Zehnder, W. Ossau, W. Spahn, H. Heinke, and G. Landwehr, *J. Cryst. Growth* **138**, 155 (1994).

⁴A. Waag, H. Heinke, S. Scholl, C. R. Becker, and G. Landwehr, *J. Cryst. Growth* **131**, 607 (1993).

⁵Th. Litz, Th. Behr, D. Hommel, A. Waag, and G. Landwehr, *J. Appl. Phys.* **72**, 3492 (1992).

⁶The growth conditions for the GaAs/AlGaAs sample are very similar to the ones reported in the following paper without the use of Sb as a surfactant during growth. J.-B. Wang, D. Ding, S. R. Johnson, S.-Q. Yu, and Y.-H. Zhang, *Phys. Status Solidi B* **244**, 2740 (2007).

⁷T. D. Golding, M. Martinka, and J. H. Dinan, *J. Appl. Phys.* **64**, 1873 (1988).

⁸S. Tatarenko, F. Bassani, J. C. Klein, K. Saminadayar, J. Cibert, and V. H. Etgens, *J. Vac. Sci. Technol. A* **12**, 140 (1994).

⁹G. Mackh, M. Hilpert, D. R. Yakovlev, W. Ossau, H. Heinke, T. Litz, F. Fischer, A. Waag, G. Landwehr, R. Hellmann, and E. O. Göbel, *Phys. Rev. B* **50**, 14069 (1994).

¹⁰S. Adachi, *Handbook on Physical Properties of Semiconductors: II-VI Compound Semiconductors* (Kluwer Academic Publishers, Boston, MA, 2004), Vol. III, pp. 54 and 367.

¹¹J. M. Hartmann, J. Cibert, F. Kany, H. Mariette, M. Charleux, P. Alleysson, R. Langer, and G. Feuillet, *J. Appl. Phys.* **80**, 6257 (1996).

¹²S. Adachi, *Optical Constants of Crystalline and Amorphous Semiconductors-Numerical Data and Graphical Information* (Kluwer Academic Publishers, Boston, MA, 1999), pp. 222 and 540.

¹³D. Kuciauskas, J. N. Duenow, A. Kanevce, J. V. Li, M. R. Young, P. Dippo, and D. H. Levi, in *Proceedings of the 38th IEEE Photovoltaic Specialists Conference, Austin, Texas, USA, 3–8 June*, (Institute of Electrical and Electronics Engineers, 2012), pp. 001721–001726.

¹⁴D. Kuciauskas, A. Kanevce, J. M. Burst, J. N. Duenow, R. Dhere, D. S. Albin, D. H. Levi, and R. K. Ahrenkiel, *IEEE J. Photovoltaics* **3**, 1319 (2013).