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Determination of CdTe bulk carrier lifetime and interface recombination velocity of CdTe/MgCdTe double heterostructures grown by molecular beam epitaxy

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The bulk Shockley-Read-Hall carrier lifetime of CdTe and interface recombination velocity at the CdTe/Mg_{0.24}Cd_{0.76}Te heterointerface are estimated to be around 0.5 μ s and $(4.7 \pm 0.4) \times 10^2$ cm/s, respectively, using time-resolved photoluminescence (PL) measurements. Four CdTe/MgCdTe double heterostructures (DHs) with varying CdTe layer thicknesses were grown on nearly lattice-matched InSb (001) substrates using molecular beam epitaxy. The longest lifetime of 179 ns is observed in the DH with a 2 μ m thick CdTe layer. It is also shown that the photon recycling effect has a strong influence on the bulk radiative lifetime, and the reabsorption process affects the measured PL spectrum shape and intensity. © 2014 AIP Publishing LLC.

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Reducing surface and interface recombination is important for minority carrier devices such as solar cells and infrared detectors. GaAs and CdTe (Ref. 1) are two popular materials for high efficiency solar cells. It has been found that many materials, such as AlGaAs and GaInP,^{2–6} provide sufficient carrier confinement to GaAs, which prevents carriers from reaching the top surface of the epilayers and thus effectively reduces the surface recombination rate by providing a heterojunction interface. The interface recombination velocity (IRV) of a high quality GaAs/Al_{0.5}Ga_{0.5}As interface has been demonstrated to be as low as 18 cm/s,³ whereas the recombination velocity of a GaAs free surface is on the order of 10⁷ cm/s.⁷ Similarly, the surface recombination velocity of CdTe was found to be on the order of 10⁵ cm/s.⁸ Research efforts of reducing CdTe surface recombination include using chemical passivation which reduces the surface recombination velocity down to 200 cm/s (Ref. 9) and using a CdS/CdTe heterojunction with an interface recombination velocity in the range of 10³ cm/s–10⁶ cm/s.^{10,11} It has been reported that MgCdTe and CdTe form a type-I band edge alignment,¹² suggesting that MgCdTe is good for electron and hole confinement and is expected to reduce the surface recombination rate of CdTe. Recently, we reported the growth, structural, and optical properties of CdTe/MgCdTe double heterostructures (DHs) grown on InSb (001) substrates by Molecular Beam Epitaxy (MBE).^{13,14} It was found that CdTe/MgCdTe DH samples show a three order of magnitude improvement in the photoluminescence (PL) intensity compared to plain CdTe layers grown on InSb, therefore, indicating qualitatively that the MgCdTe layers effectively confine carriers and that CdTe/MgCdTe heterointerface has a lower recombination velocity in comparison to the CdTe

surface. In this letter, we quantify the CdTe/MgCdTe interface recombination velocity by measuring the carrier lifetime of several CdTe/MgCdTe DHs with various CdTe layer thicknesses using time-resolved photoluminescence (TRPL). The bulk Shockley-Read-Hall (SRH) lifetime of CdTe is also extracted.

The CdTe/MgCdTe DHs are grown on closely lattice-matched 2-in. InSb (001) substrates using a dual-chamber MBE system. The system consists of a II-VI chamber and a III-V chamber inter-connected by an ultra-high vacuum preparation chamber. The InSb substrate is at first thermally deoxidized in the III-V chamber and a 500 nm InSb buffer layer is grown. After that the substrate is transferred through the preparation chamber to the II-VI chamber to grow a 500 nm CdTe buffer layer, followed by the growth of CdTe/MgCdTe DH with a 10 nm thick CdTe cap layer. Detailed growth conditions and sample structure were reported previously.¹³ To determine the interface recombination velocity, the CdTe middle layers in the DHs are designed with different thicknesses of 0.3 μ m, 0.5 μ m, 1 μ m, and 2 μ m. The 30 nm thick MgCdTe barrier layers have a Mg composition of 24%, as determined by high resolution X-ray diffraction (XRD) measurements. All the epilayers are undoped and the background doping level is estimated to be lower than 10¹⁵ cm⁻³ in the CdTe middle layer based on temperature dependent carrier lifetime measurements.

TRPL measurements are carried out using a time-correlated single photon counting system as reported previously.¹⁴ The excitation source is a pulsed Ti:Sapphire laser operating at 750 nm wavelength with 0.8 MHz repetition rate. The laser power is 2 mW and the beam radius is about 1 mm. It is estimated that for samples of different thickness the initial excited carrier density is on the order of 10¹⁵ cm⁻³. Steady-state PL spectra are measured using a

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spectrometer equipped with a photomultiplier tube. A 532 nm diode pumped solid-state laser is used as an excitation source. The laser power is set to 0.92 mW and the beam radius is 0.54 mm.

PL decay measurements can be used to measure the carrier lifetime of a sample. However, this lifetime can be affected by carrier diffusion, surface recombination, etc. The use of DHs simplifies the carrier lifetime model. In our case, the MgCdTe barrier layers confine the carriers inside the middle CdTe layer and it is reasonable to assume that the excess carriers distribute uniformly in CdTe due to the long diffusion length of minority carriers. The effective carrier lifetime τ_{eff} of a CdTe/MgCdTe DH sample can then be expressed using the following equation:¹⁵

$$\frac{1}{\tau_{eff}} = \frac{1}{\tau_{bulk}} + \frac{1}{\tau_{interface}} = \frac{1}{\tau_{bulk}} + \frac{2S}{d}, \quad (1)$$

where τ_{bulk} is the bulk carrier lifetime, $\tau_{interface}$ is the interface recombination lifetime, S is the interface recombination velocity, and d is the thickness of the sample. The above equation is valid when S is relatively small¹⁵ and the diffusion length of minority carriers is much longer than the middle layer thickness.

Fig. 1 shows the room temperature PL decays of the CdTe/MgCdTe DHs with different CdTe middle layer thicknesses, where the initial PL intensity has been normalized. The carrier lifetime is determined by fitting near the tail of the decay curve. It is found that the carrier lifetimes vary across the wafer, which is probably due to non-uniformity in the substrate temperature and beam flux distribution during MBE growth, and the lifetimes shown here are measured near the center of the wafer. It is also found that the carrier lifetimes of CdTe/MgCdTe DHs with a thin 10 nm cap layer gradually degrade with time, and the lifetime measurements were carried out within days after the samples were taken out of the MBE chamber. Fig. 1 shows that the thinner samples have shorter decay times, suggesting a non-zero recombination rate at the CdTe/MgCdTe interface. The longest lifetime measured at room temperature is 179 ns for the sample with a 2 μm thick middle layer.

FIG. 1. Time-resolved photoluminescence decay of CdTe/MgCdTe double heterostructures with different CdTe middle layer thicknesses. The thinner the layer, the shorter the lifetime, indicating that interface recombination lifetime plays an important role in the effective carrier lifetime.

Traditionally the bulk carrier lifetime is treated as thickness independent at low injection levels. However, the bulk carrier lifetime can vary with thickness of the sample, as it consists of both SRH and radiative lifetime and the latter is related to photon recycling factor γ as shown below²

$$\frac{1}{\tau_{bulk}} = \frac{1}{\tau_{SRH}} + \frac{1}{\tau_{rad}} = \frac{1}{\tau_{SRH}} + (1 - \gamma)BN_{doping}, \quad (2)$$

where the photon recycling factor γ is defined as the percentage of photons created by radiative recombination that are reabsorbed within the sample.¹⁶ For CdTe, the absorption coefficient near the band edge is on the order of 10^4 cm^{-1} resulting in a short absorption length of the photons. The value of γ for CdTe middle layer is calculated using the ray-tracing method¹⁷ as shown in Fig. 2 and it increases as a function of CdTe layer thickness. Thus, the radiative lifetime increases with increasing CdTe layer thickness. The material radiative recombination coefficient B was determined previously from excitation-dependent PL measurements.¹⁴ It is calculated from Eq. (2) that the radiative lifetimes for the 0.3 μm , 0.5 μm , 1 μm , and 2 μm thick DH samples are 0.7 μs , 0.9 μs , 1.6 μs , and 3.1 μs , respectively, by assuming a doping concentration of 10^{15} cm^{-3} . These values are much longer than the measured effective carrier lifetime for each sample. Hence, it is reasonable to assume that radiative lifetime does not affect the effective carrier lifetime at room temperature. This assumption is further supported by temperature-dependent and excitation-dependent PL measurement, which show that non-radiative recombination dominates at room temperature and under low injection levels.¹⁴ Therefore, the measured lifetime is only related to the SRH bulk carrier lifetime and the interface recombination lifetime as shown below

$$\frac{1}{\tau_{eff}} \approx \frac{1}{\tau_{SRH}} + \frac{1}{\tau_{interface}} = \frac{1}{\tau_{SRH}} + \frac{2S}{d}. \quad (3)$$

High-resolution XRD measurements show that all the CdTe layers in the studied samples are coherently strained even when the thickness reaches 2 μm . Thus, we can assume that the bulk SRH carrier lifetime is the same for CdTe DHs with different CdTe layer thicknesses. By linearly fitting $1/\tau_{eff}$ versus $2/d$, both the bulk SRH lifetime and the interface

FIG. 2. Calculated photon recycling factor for the CdTe middle layer of the double heterostructure using ray-tracing method.

recombination velocity can be extracted. Figure 3 shows the carrier lifetime as a function of CdTe layer thickness and the fitted curve of Eq. (3). Based on this fitting, the interface recombination velocity at the CdTe/MgCdTe interfaces and the bulk SRH lifetime of CdTe are extracted to be $(4.7 \pm 0.4) \times 10^2$ cm/s and $0.5 \mu\text{s}$, respectively. This interface recombination velocity is much smaller than that of a free CdTe surface and comparable to that of a typical GaAs/AlGaAs interface, suggesting that MgCdTe is an excellent barrier layer for CdTe based solar cells. The long bulk SRH carrier lifetime indicates that the CdTe epilayer grown on InSb substrates is of high quality, which is in agreement with the low defect densities of 10^4 cm^{-2} measured using confocal PL mapping.¹⁸ If it is assumed that the effective carrier lifetime of the $0.3 \mu\text{m}$ sample is limited only by interface recombination, an upper limit of the interface recombination velocity can be obtained by using $\tau_{\text{eff}} \approx \tau_{\text{interface}} = d/2S$. Using $d = 0.3 \mu\text{m}$ and $\tau_{\text{eff}} = 31 \text{ ns}$, the upper limit of S is determined to be 484 cm/s , which is close to the value obtained by the linear fitting. Therefore, the recombination process in the sample with $0.3 \mu\text{m}$ thickness is dominated by interface recombination, and the fitting provides a relatively accurate measurement of S . However, it should be noted that the extracted bulk SRH lifetime is very sensitive to this fitting method and $0.5 \mu\text{s}$ is only a rough estimation.

PL spectra of the DH samples with different thicknesses are measured under the same conditions. As shown in Fig. 4, the PL peak shifts to longer wavelengths when the CdTe layer is thicker, which is an indication of photon reabsorption. The photons generated deep inside the CdTe layer can be reabsorbed before escaping the front surface of the CdTe layer. As the absorption coefficient of longer wavelength photons is smaller than that of shorter wavelength photons, the probability for longer wavelength photons to be reabsorbed is lower and thus the measured PL spectra shape changes and the PL peak shifts to a longer wavelength. Kuciauskas *et al.* reported a similar effect on single crystalline CdTe using subbandgap two-photon excitation PL measurements. The measured PL peak moves significantly to longer wavelengths when the excitation region is a few mm below the surface of the sample.⁸

FIG. 3. Effective carrier lifetime τ_{eff} as a function of sample thickness d . The IRV and the bulk Shockley–Read–Hall carrier lifetime are extracted to be $(4.7 \pm 0.4) \times 10^2$ cm/s and $0.5 \mu\text{s}$, respectively.

FIG. 4. Photoluminescence spectra of CdTe/MgCdTe double heterostructures with different CdTe layer thicknesses. The inset figure shows that the photoluminescence peak position changes with the CdTe layer thickness, indicating photon reabsorption effect.

Fig. 4 also shows that PL intensity is a function of CdTe layer thickness. The PL intensity increases with thickness up to $1 \mu\text{m}$, then decreases. This finding can be explained as follows. In steady state PL measurements, the generation rate G is equal to the recombination rate R inside the CdTe layer. G is related to the laser power density P , the thickness of the sample d , and the photon energy E_{photon} as shown in Eq. (4). The recombination rate R is related to the excess carrier densities Δn and the effective carrier lifetime τ_{eff} as shown in Eq. (5). Using Eqs. (4) and (5), the excess carrier densities Δn can be obtained. PL intensity under low excitation is proportional to the net radiative recombination rate, which takes into account the photon recycling factor, radiative recombination coefficient, background doping N_{doping} , excess carrier density, and the thickness of the sample, as shown in Eq. (6)

$$G = \frac{P}{dE_{\text{photon}}}, \quad (4)$$

$$R = \frac{\Delta n}{\tau_{\text{eff}}}, \quad (5)$$

$$PL \propto (1 - \gamma)BN_{\text{doping}}\Delta n d = (1 - \gamma)\tau_{\text{eff}}BN_{\text{doping}}P/E_{\text{photon}}. \quad (6)$$

Therefore, under low excitation, PL intensity is simply proportional to $(1 - \gamma)\tau_{\text{eff}}$. The term $(1 - \gamma)$ is defined as the photon extraction factor, which is the percentage of radiatively generated photons that emit into the free space.¹⁶ The effective carrier lifetime can be calculated using the above thickness dependent carrier lifetime fitting results. Fig. 5 shows the measured PL intensity plotted together with the calculated $(1 - \gamma)\tau_{\text{eff}}$ curve. It is observed that the measured PL intensity varies with the curve as predicted by the theory. On one hand, with a thicker layer, the effective carrier lifetime increases as a result of smaller interface recombination rate and thus more excess carriers are generated during steady state PL measurements. On the other hand, the photon reabsorption is enhanced in thicker layer samples and those photons generated by radiative recombination are more likely to be reabsorbed before escaping the CdTe layer. Therefore, the PL intensity is observed to follow the trend of

FIG. 5. Comparison between measured photoluminescence intensity and $(1 - \gamma)\tau_{\text{eff}}$ as a function of sample thickness.

$(1 - \gamma)\tau_{\text{eff}}$ and appears to exhibit a peak around a thickness of $1 \mu\text{m}$ for the CdTe layer.

In summary, long bulk SRH lifetime and low interface recombination velocity have been demonstrated in the CdTe/MgCdTe DHs grown by MBE. The bulk SRH carrier lifetime is approximately $0.5 \mu\text{s}$, which shows the high quality of the epitaxial CdTe layer on InSb. The longest lifetime observed is 179 ns for a DH sample with a $2 \mu\text{m}$ thick CdTe layer. The interface recombination velocity is estimated to be $(4.7 \pm 0.4) \times 10^2 \text{ cm/s}$ from the effective carrier lifetimes of the samples with different CdTe middle layer thicknesses. It indicates that MgCdTe is a promising barrier layer material for solar cell applications. The photon recycling effect is discussed and it has a strong influence on the radiative lifetime; however, the radiative lifetime does not play a significant role in these samples since the lifetime is dominated by interface and bulk SRH recombination. The PL spectra of different samples show that the peak shifts due to the photon reabsorption and the PL intensity of the samples varies as a function of $(1 - \gamma)\tau_{\text{eff}}$.

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