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Energy relaxation in IR laser excited Hg\textsubscript{1-x}Cd\textsubscript{x}Te

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Abstract. IR laser excitation of Hg\textsubscript{1-x}Cd\textsubscript{x}Te by low-fluence femtosecond and high fluence microsecond pulses was explored for the technologically important alloy fractions $x \sim 0.2$ and $x \sim 0.28$. We have used first principles (LAPW) electronic structure calculations and finite element modelling, supported by Monte Carlo simulation for the description of femtosecond pulse carrier relaxation and the transport parameters. Laser wavelengths considered were 6.4 - 10.6 $\mu$m for $x \sim 0.2$ and 3.8 - 4.8 $\mu$m for $x \sim 0.28$, with an incident 1 microsecond pulse fluence of 2 J/cm$^2$. Many energy transfer mechanisms are invoked due to the long timescales of the microsecond pulses, and a main challenge is therefore to elucidate how these interplay in situations away from thermal equilibrium. Mechanisms studied include one- and two-photon absorption (OPA and TPA) across the band gap, inter-valence band absorption (IVA) between light- and heavy hole bands, electron-hole recombination/impact ionization, band gap renormalisation, intra-band free carrier absorption (FCA), excess carrier temperatures, non-equilibrium phonon generation, and refractive index changes. In the high fluence case, lattice temperatures evolve considerably during the laser pulse in response to the heated carriers. The chosen photon energies lie just above the band gap at the starting lattice temperature of 77 K, and nonlinear effects therefore dominate as the material heats up and the band gap begins to exceed the photon energy. Because of the low photon energy we must rely on Auger recombination, inter-valence band absorption and free carrier absorption to heat the carrier plasma. Although some Hg\textsubscript{1-x}Cd\textsubscript{x}Te material parameters are now relatively well known, existing data for many of the processes are inadequate for cases far away from thermal equilibrium. Furthermore, the role of Auger recombination in relation to non-intrinsic recombination has been a matter of debate lately. In this respect, information from experiments can only be interpreted correctly if the intrinsic behaviour of the material is well known, and we have therefore assumed intrinsic Hg\textsubscript{1-x}Cd\textsubscript{x}Te for the present study.

1. Introduction

Excitation by low-fluence, ultrashort laser pulses is a central technique for extracting information about non-equilibrium carrier dynamics in semiconductors. Using photon energies far exceeding the fundamental band gap, such experiments can provide a very direct insight into carrier relaxation and hot phonon generation phenomena by isolating these processes from subsequent effects that occur on longer timescales. Hot, non-equilibrium optical phonons are then usually created due to the sudden, simultaneous relaxation of a large amount of photoexcited carriers entering the conduction band far above the conduction band edge. After the ultrashort laser pulse has passed and the short lived hot optical phonons have decayed, the overall lattice temperature is left almost unchanged. Energy transfer
due to long, high fluence laser pulses on the other hand, increases the lattice temperature and involves many additional physical effects. In particular, if the photon energy of the incident radiation just exceeds the band gap, absorption can be highly nonlinear and depend strongly on wavelength, radiation intensity, carrier temperatures, and lattice temperatures. Using a long photon wavelength would also allow intra-band free carrier (FCA) and inter-valence band (IVA) absorption between the light and heavy hole bands to be studied. The high fluence in such experiments can be sufficient to raise the overall lattice temperature by several hundred K. Although the total number of carriers generated in the conduction band can be comparable to that of a short pulse experiment, the majority of carriers are well thermalized at carrier temperatures which lie much closer to the overall lattice temperature. The difference between carrier and lattice temperatures will often be smaller than 100 K. Nevertheless, such small carrier excess temperatures can have a major impact on processes occurring near the band gap. For example, a slightly elevated carrier temperature can strongly reduce the saturation or "bleaching" of one-photon absorption across the band gap, or affect other long-timescale parameters, like carrier lifetimes and impact ionization rates.

2. Simulation Results and Discussion

Short pulse Monte Carlo (MC) simulation results (electrons only) shown in figure 1 are for the material Hg$_{0.72}$Cd$_{0.28}$Te excited at $\lambda = 1064$ nm assuming an initial lattice temperature of 300 K. Due to the small effective mass of the conduction band, the electrons receive most of the photonic energy. They enter the conduction band with an initial energy corresponding to $\sim 6000$ K. Delayed cooling at higher carrier densities is clearly seen, and this effect is attributed to the creation of hot phonons. The experiments carried out by Pelouch et al. [1] apparently did not show any strong hot phonon effect, but the presence of multiple relaxation time constants was seen. We plan to analyze this experiment more thoroughly in a future study, when further upgrades to our MC simulator have been introduced.

In the long pulse length case, Auger lifetimes are important for calculating carrier temperatures, since this process conserves the energy of the recombination within the carrier system. Interestingly, these lifetimes have become a matter of debate lately. Some of the measured lifetimes in Hg$_{1-x}$Cd$_x$Te and similar materials are very short [2] while others are very long [3]. Theoretical predictions using simplified band models suggest relatively short Auger lifetimes [4], whereas results from more elaborate models [5] predict much longer lifetimes. For an update on possible causes for this (and e.g. the suspected contribution from non-intrinsic recombination mechanisms in the lifetime measurements) we recommend references [6] and [7]. Here we have taken an average between the conventional short estimates used in a previous work on Hg$_{0.72}$Cd$_{0.28}$Te [8] and the longer estimates of Krishnamurty [5]. Effectively, this increases the Auger lifetime by a factor of 5-6 compared to [8]. Uncertainties of this magnitude are usually considered 'normal'. We further generalized the expressions to accommodate different carrier and lattice temperatures. High fluence simulations were carried out for two undoped 10 $\mu$m thick films of Hg$_{0.72}$Cd$_{0.28}$Te and Hg$_{0.8}$Cd$_{0.2}$Te, in ideal thermal contact with a highly conductive underlying substrate with a temperature of 77 K before illumination. The samples were subjected to an incident radiation intensity of 2 MW/cm$^2$ with a laser pulse length of 1 microsecond. As expected, the introduced changes resulted in much smaller initial excess carrier temperatures than in the previous work [8], but more importantly, the physics now becomes more diversified. Obviously, any extraction of data from non-equilibrium laser experiments must be performed with utmost care, since controlling the state variables of the material is essential for an accurate measurement. With lower carrier temperatures, carrier migration to higher valleys becomes less important. Still, a much needed paper displaying the energy differences between e.g. the $\Gamma$ valley and the L valley has recently been published [9]. In figure 1 we show our corresponding gap-corrected results using first principles LAPW [10] electronic structure calculations. Figure 2 shows the temporal evolution of physical variables at the surface during the 1 $\mu$s laser pulse, for two wavelengths in each material. Initially the photon energy exceeds the band gap by approximately 0.1 eV and 0.05 eV for the respective short and long wavelengths incident on each material. As the band gaps increase with
**Figure 1.** Energy relaxation after $\lambda = 1064$ nm femtosecond laser pulse excitation, $x = 0.28$ (upper graph), and calculated band structures for Hg$_{0.72}$Cd$_{0.28}$Te and Hg$_{0.72}$Cd$_{0.28}$Te respectively, at 77 K (lower graphs).

**Figure 2.** Temporal evolution of some key variables at the surface during a 1 $\mu$s laser pulse. Two laser wavelengths for each material, Hg$_{0.72}$Cd$_{0.28}$Te and Hg$_{0.8}$Cd$_{0.2}$Te. The melting point of Hg$_{1.0}$Cd$_{0.5}$Te is 1000 K.
temperature, one-photon absorption (OPA) is weakened and the other energy transfer processes begin to dominate. In fact, only in Hg$_{0.72}$Cd$_{0.28}$Te at a laser wavelength of 3.8 $\mu$m does OPA have a very prominent role. Here, carrier excess temperatures are high for a short period at the beginning of the pulse, and are driven by OPA combined with Auger recombination. After termination of regular OPA the lattice temperature increases more slowly, resulting in a 'kink' in the lattice temperature curve. For the other cases, OPA is severely bleached, and in Hg$_{0.8}$Cd$_{0.2}$Te, with its narrow conduction band, carriers generated by two-photon absorption (TPA) slightly reverses OPA at the 10.6 $\mu$m laser wavelength, resulting in an OPA absorption coefficient fluctuating around zero (not shown in figure 2).

3. Conclusions

Energy relaxation in two representative, intrinsic Hg$_{1-x}$Cd$_x$Te alloys has been investigated. Specifically, we have investigated the low gap material Hg$_{0.8}$Cd$_{0.2}$Te and the medium gap material Hg$_{0.72}$Cd$_{0.28}$Te. MC simulation of short pulse electronic relaxation shows reduced cooling due to the generation of hot phonons, but final conclusions have to await a more comprehensive study. For the long pulse length case, we have demonstrated a need for non-equilibrium rather than equilibrium parameters for the absorption and recombination mechanisms. This is necessary in order to determine excess carrier temperatures more accurately as a function of the applied laser intensity. In one case, high initial carrier excess temperatures obviously played an important role by dispersing the carriers over a large range of energy states, thereby reducing saturation (bleaching) of one-photon absorption across the band gap. Furthermore, we emphasized the importance of knowing the true intrinsic Hg$_{1-x}$Cd$_x$Te behaviour when interpreting experimental results, and presented band plots displaying the energy differences between conduction band valleys.

References

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