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Ultrafast dynamics of interfacial electric fields in semiconductor heterostructures monitored by pump-probe second-harmonic generation

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We report measurements of the ultrafast dynamics of interfacial electric fields in semiconductor multilayers using pump-probe second-harmonic generation (SHG). A pump beam was tuned to excite carriers in all the layers in GaAs/GaSb and GaAs/GaSb/InAs heterostructures. The resulting carrier dynamics manifests itself via electric fields created by charge separation due to carrier redistribution at the interfaces. The evolution of interfacial fields is monitored by a probe beam through an eletric-field-induced SHG signal. We distinguish between several stages of dynamics originating from redistribution of carriers between the layers. We also find a strong enhancement of the induced electric field caused by hybridization of the conduction and valence bands at the GaSb/InAs interface. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1521573]

Knowledge of processes governing the carrier relaxation in multilayer semiconductor structures is essential for designing multifunctional high-speed electronic and optoelectronic devices.[1–3] Quantum confinement is known to significantly affect carrier thermalization dynamics in quantum wells[4] and quantum dots.[5] One can expect that the interface between different semiconductor layers will influence carrier dynamics even in nonquantum-confined structures. For example, during fast relaxation due to electron–electron ($\lesssim 200$ fs) and electron-phonon (~ 1 ps) scattering processes,[3–5] the photoexcited electrons and holes tend to accumulate in different layers. The resulting charge separation gives rise to interfacial electric fields which can change the initial band alignment and lead to interlayer transport phenomena.[6]

The techniques typically used for monitoring ultrafast carrier dynamics in semiconductors are pump-probe transmission or reflection spectroscopy.[1–5] These methods rely on the linear response of the electron-hole subsystem, excited by the pump pulse, to the probe light. As a result, they give an accurate account of carrier population dynamics occuring in the bulk, while being insensitive to electric fields near the interface. On the other hand, the second-harmonic generation (SHG) technique is known to be extremely sensitive to electric fields occurring at surfaces and interfaces.[7] This unique feature of SHG was employed to study long-time carrier dynamics at the silicon–oxide interface.[7,8]

In this letter we report measurements of the ultrafast dynamics of optically induced interfacial electric fields in semiconductor multilayers using SHG as a probe in time-resolved pump-probe spectroscopy. These measurements were performed on GaAs/GaSb and GaAs/GaSb/InAs heterostructures. The crucial advantage of our technique is the ability to simultaneously monitor the carrier dynamics at different interfaces of the same sample. In particular, by tracking the evolution of different interfacial fields contributing to the total electric-field-induced SHG (EFISHG) signal we are able to study the redistribution of carriers between the layers due to electron transport across interfaces.

Electronic properties of the GaSb/InAs heterojuction are very sensitive to the type of interface bonding. Since InAs and GaSb have no common atoms, two types of interfaces can be formed, GaAs bonds or InSb bonds.[9] A large difference in GaAs and InSb band structures result in a much stonger hybridization of InAs conduction and GaSb valence band states for the InSb interface type. The hybridization minigap was observed in an appearance of energy gaps in capacitance–voltage and quantum Hall measurements[10] as well as in a splitting of cyclotron resonance peak measured using far-infrared spectroscopy.[11] In our experiment, we find that band hybridization results in a strong enhancement of the EFISHG signal.

We have investigated four heterostructures grown by molecular beam epitaxy: GaAs/GaSb(20 nm); GaAs/GaSb(400 nm); GaAs/GaSb(500 nm)/InAs(20 nm), with an InSb interface between GaSb and InAs layers; and GaAs/GaSb(500 nm)/InAs(20 nm), with a GaAs interface between GaSb and InAs. In all samples, the thickness of GaAs layer was 100 nm. Samples were grown on semi-insulating (100) GaAs substrates. A pump-probe configuration with linearly polarized pump and probe beams was used in our measurements. The observed EFISHG signal was monitored as a function of probe-to-pump delay times. All optical measurements were carried out at 4.3 K. The initial beam of 150 fs pulses from a mode-locked Ti:Al_2O_3 laser (Mira 900) at the wavelength of 800 nm (1.55 eV) and a repetition rate of 76 MHz was split into pump and probe beams. The probe beam of 120 mW average power was channeled through an optical delay stage. The pump beam, after chopping at a frequency of 400 Hz, was of the same average power. The overlap spot of the pump and probe beams on the sample was 100 μm in diameter. The pump beam was incident normally on the sample with either p or s polarization. The probe beam (also

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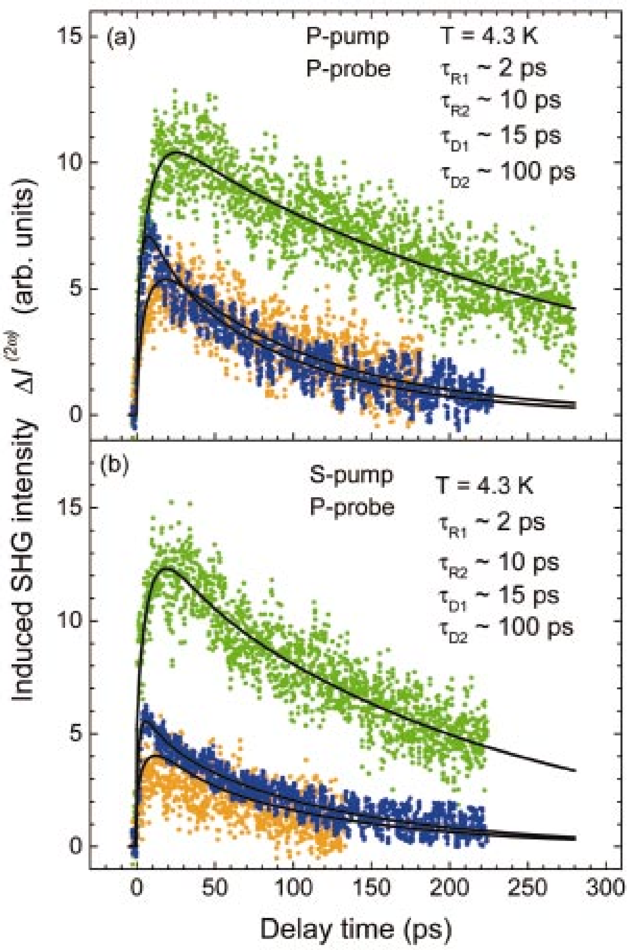

FIG. 1. (Color) EFISHG signals from GaAs/GaSb sample (blue) and GaAs/GaSb/InAs sample with InSb (green) and GaAs (orange) type interface measured with (a) p-polarized pump light and (b) s-polarized pump light. The fits with rise/decay exponential function are shown by solid lines.

p or s polarized) was directed to the sample surface at the angle of 75°. The SHG signal was optically separated from the reflected fundamental probe beam and measured by a photomultiplier tube through a ''lock-in'' amplifier triggered by the chopped pump pulses.

Figure 1 shows pump-induced SHG signals measured for the GaAs/GaSb(400 nm) heterostructure in comparison with those taken for the GaAs/GaSb/InAs with an InSb interface and with a GaAs interface. Note that the induced signal is observed only for the p-polarized probe beam and for both p and s pump polarizations. No pump-induced SHG signal was observed for the GaAs/GaSb sample with the thinner (20 nm) GaSb layer.

The measured signals were fitted by a combined exponential rise/decay function. The pump-induced SHG signal for GaAs/GaSb heterostructure is described by a single rise-time constant, $\tau_{R_1} \simeq 2(\pm 1)$ ps, and two decay-time constants, $\tau_{D_1} \simeq 15(\pm 3)$ ps and $\tau_{D_2} \simeq 100(\pm 10)$ ps. In contrast, for GaAs/GaSb/InAs heterostructures, there are two stages in the induced signal rise. The additional slow-rising component in both samples has rise-time constant $\tau_{R_2} \simeq 10$ (± 2) ps. The fast rise-time, τ_{R_1}, and long decay-time, τ_{D_2}, constants for GaAs/GaSb/InAs and GaAs/GaSb samples are similar. Note here that all of the observed time constants are much larger than the characteristic time of electron–electron interactions in such structures ($\lesssim 200$ fs) so that deviations from the relaxation-time approximation are small.[1,2]

The intensity of pump-induced SHG signals measured for GaAs/GaSb/InAs heterostructures shows strong dependence on the interface type between GaSb and InAs layers. For the sample with InSb type interface, the signal amplitude is significantly larger than that for GaAs/GaSb structure, while for the sample with GaAs type interface, the signal is comparable to that for GaAs/GaSb. Moreover, despite similar decay-time constants τ_{D_2} for both GaAs/GaSb/InAs samples, there is considerable long-time (>250 ps) constant background in the case of InSb type interface while it is significantly smaller for GaAs type.

An apparent presence of several stages in the evolution of the measured SHG signal indicates the rather complicated dynamics of interfacial electric fields originating from a redistribution of carriers between the interfaces. The induced local electric fields, $\mathcal{E}_1(t)$ and $\mathcal{E}_2(t)$ (subscripts 1 and 2 refer to GaAs/GaSb and GaSb/InAs interfaces, respectively), depend on the number of carriers as well as on their spatial distribution near each interface at a given time. Retaining only linear terms in $\mathcal{E}_i(t)$, the nonlinear polarization can be written as[7,8]

$$P^{NL}(2\omega,t)=[\chi^{(2)}+\chi_1^{(3)}\mathcal{E}_1(t)+\chi_2^{(3)}\mathcal{E}_2(t)][E(\omega)]^2, \quad (1)$$

where $E(\omega)$ is the electric field component of the incident probe light, $\chi^{(2)}$ is the second-order bulk susceptibility, and $\chi_i^{(3)}$ are the third-order susceptibilities at the interfaces. The measured EFISHG intensity, $\Delta I^{(2\omega)}=I^{(2\omega)}-I_0^{(2\omega)}$, which is obtained by subtracting the bulk contribution $I_0^{(2\omega)}=|\chi^{(2)}|^2|E(\omega)|^4$ from the total intensity, $I^{(2\omega)}=|P^{NL}(2\omega)|^2$, has the following form [after neglecting higher-order nonlinear in $\mathcal{E}_i(t)$ terms]:

$$\Delta I^{(2\omega)} \propto \chi_1^{(3)}\mathcal{E}_1(t)+\chi_2^{(3)}\mathcal{E}_2(t). \quad (2)$$

We attribute the observed several stages in the dynamics of the interfacial electric fields to an interplay between relaxation of carriers and their transport across the heterostructures. Since carriers are excited in all the heterostructure layers, electrons with high excess energies relax to the lower-energy conduction band states in the GaAs and InAs layers, while the holes are accumulated in the GaSb layer (Fig. 2). The resulting charge separation leads to a rise of interfacial fields which manifests itself in the initial growth of the EFISHG signal characterized by fast rise-time constant τ_{R_1} ~ 2 ps. Note here that in the GaAs/GaSb(20 nm) sample, the carriers are accumulated predominantly in the GaAs layer, so the interfacial field is weak and the corresponding EFISHG signal is undetectable. In all other samples, the majority of carriers are excited in the thicker GaSb layer leading to significant concentration of holes in that layer. The induced interfacial fields bend the initial energy profile, thus lowering the barrier at the GaAs/GaSb interface (Fig. 2) and resulting in electron transport across the interface. As negative charges in GaAs transfer through the barrier, the electric field at the GaAs/GaSb interface decreases. For GaAs/GaSb heterostructure, such a decrease shows up in a fast decay of the EFISHG signal with decay-time constant $\tau_{D_1} \sim 15$ ps (Fig. 1). The

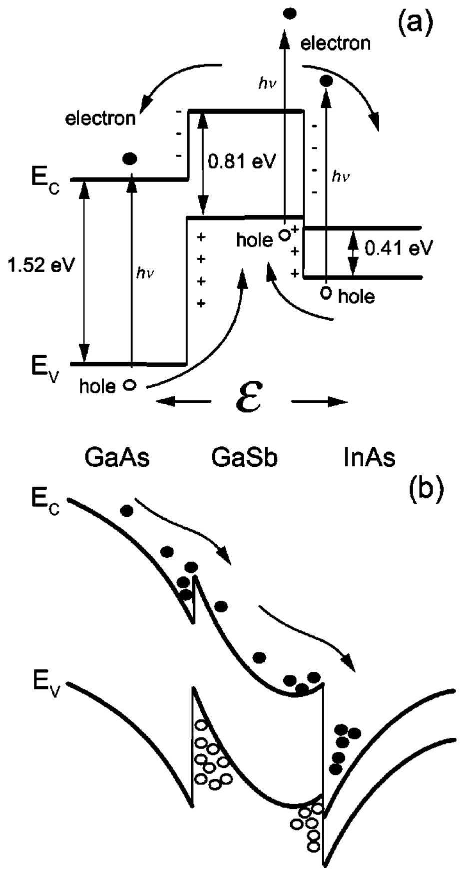

FIG. 2. The initial band alignment for GaAs/GaSb/InAs heterostructure (a) and its realignment due to induced interfacial electric fields (b).

subsequent relaxation of the interfacial electric field, characterized by long decay-time constant $\tau_{D_2}\sim 100$ ps, is due to carrier migration away from the interface.

For GaAs/GaSb/InAs heterostructures, the situation is completely different. In this case, the electrons crossing the GaAs/GaSb interface accumulate in the InAs layer. The resulting increase in the GaSb/InAs interfacial field manifests itself as the additional rise component of the EFISHG signal with rise-time constant $\tau_{R_2}\sim 10$ ps. Note that this arrival time is comparable to the departure time τ_{D_1} in the GaAs/GaSb sample. A subsequent relaxation of the interfacial electric fields is characterized by similar $\tau_{D_2}\sim 100$ ps decay-time constant.

A striking feature observed for GaAs/GaSb/InAs heterostructures is a significantly stronger EFISHG signal for InSb type interface between GaSb and InAs layers than that for GaAs type. We attribute this difference to a much stronger hybridization of InAs conduction and GaSb valence bands for the GaSb/InAs interface with InSb bonds.[9] The strength of the coupling across that interface is known to be directly related to the overlap integral between the band envelope functions.[11] On the other hand, this coupling determines the magnitude of the interfacial electric field. The large band overlap for the GaSb/InAs interface with InSb bonds leads to a stronger EFISHG signal as compared to that for weekly hybridized GaAs type interface. In fact, the induced signals for GaAs/GaSb/InAs with GaAs type interface and for GaAs/GaSb samples are comparable (Fig. 1). Note that the stronger carrier confinement leads to a larger residual electric field in the sample with InSb type interface, which is observed as a long-time constant background in the EFISHG signal.

In summary, we have studied the ultrafast dynamics of interfacial electric fields in GaAs/GaSb and GaAs/GaSb/InAs heterostructures using a pump-probe SHG technique. We observed a complicated evolution of the interfacial fields originating from the redistribution of carriers between the interfaces. We also found a strong enhancement of the SHG signal caused by an interband mixing at the GaSb/InAs interface. The ability of the EFISHG signal to monitor spatially separated regions makes pump-probe SHG a unique tool for studying relaxation and transport phenomena in multilayer semiconductor structures.

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