APPLIED PHYSICS LETTERS VOLUME 81, NUMBER 2 8 JULY 2002

Ultrafast spin dynamics in GaAs/GaSb/InAs heterostructures probed by second harmonic generation

Yu. D. Glinka,[a] T. V. Shahbazyan, I. E. Perakis, and N. H. Tolk
Department of Physics and Astronomy, Vanderbilt University, Nashville, Tennessee 37235

X. Liu, Y. Sasaki, and J. K. Furdyna
Department of Physics, University of Notre Dame, Notre Dame, Indiana 46556

(Received 3 April 2002; accepted for publication 17 May 2002)

We report the application of pump-probe second harmonic generation (SHG) to monitor spin dynamics in nonmagnetic semiconductor heterostructures. Spin-polarized electrons were selectively excited by a pump beam in the GaAs layer of GaAs/GaSb/InAs structures. However, the induced magnetization manifests itself through the SHG probe signal from the GaSb/InAs interface, thus indicating a spin-polarized electron transport. We find that the magnetization dynamics is governed by an interplay between the spin density evolution at the interfaces and the spin relaxation. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1494107]

Ultrafast spin-sensitive spectroscopy provides unique information about spin relaxation in semiconductor heterostructures as well as spin-polarized electron transport across interfaces. Knowledge of the processes governing spin dynamics is essential for designing novel multifunctional electronic and optoelectronic devices, including base components for quantum computing.[1] Among the wide variety of multilayer semiconductors, GaSb/InAs heterostructures are especially promising.[2]

The excitation of an ensemble of spins by a circularly polarized laser light tuned just above the band gap gives rise to a net magnetization. The typical time-resolved techniques, such as polarized photoluminescence spectroscopy,[3,4] pump-probe transmission/reflection,[5–8] and Faraday or Kerr rotation,[1,9] all rely on the *linear* response of the spin subsystem to a probing light, and are well suited for monitoring spin dynamics in the *bulk* of semiconductor structures. On the other hand, the *nonlinear* optical effects, such as second harmonic generation (SHG), are known to be highly sensitive to local magnetic fields occurring at magnetized surfaces and at interfaces in magnetic-semiconductor-based multilayers.[10] Therefore, the application of SHG in the pump-probe configuration is a promising method for studying the dynamics of optically excited spins at the semiconductor *interfaces*.

In this letter we report the application of pump-probe SHG technique to study the ultrafast spin dynamics in nonmagnetic heterostructures. Spin-polarized electrons were selectively excited in GaAs layer of GaAs/GaSb and GaAs/GaSb/InAs heterostructures. Only the GaAs/GaSb/InAs samples showed a significant induced magnetization indicating interlayer spin-polarized electron transport from GaAs to InAs. The dominant contribution to the magnetic-field-induced SHG signal results from high local density of spins accumulated at the semimetallic GaSb/InAs interface. Temperature dependence of induced SHG signals in the range from 4.3 to 300 K revealed two distinct mechanisms governing magnetization dynamics: the evolution of the local spin density at the interfaces and the spin relaxation.

We have investigated two heterostructures grown by molecular beam epitaxy: (1) GaAs(100 nm)/GaSb(400 nm) and (2) GaAs(100 nm)/GaSb(500 nm)/InAs(20 nm). 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 has passed through an optical delay stage. The pump beam was chopped at a frequency of 400 Hz and, after that, had the same average power. The overlap spot of the pump and probe beams on the sample was $\sim 100\ \mu$m in diameter. The pump beam was incident normally on the sample with either left- or right-handed circular polarization (σ^- or σ^+, respectively). The probe beam was linearly polarized (p or s), and was directed to the sample surface at the angle of 75°. The pump-induced SHG signal was monitored as a function of probe-to-pump delay times. Note that only the p linearly-polarized probe light contributes to the induced signal, in agreement with SHG measurements on magnetized surfaces.[11] 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 the pump-induced SHG signals taken on the GaAs/GaSb heterostructure (sample 1) at a temperature $T = 4.3$ K. No significant difference was observed between signals measured with σ^- or σ^+ polarized pump light [Figs. 1(a) and 1(b), respectively], indicating that the signal is due to the induced electric field at the interface. The interfacial electric fields caused by charge separation between photoexcited carriers are known to strongly enhance the SHG response.[12] The measured signal was fitted by a combined exponential rise/decay function. The signal intensity increases with a time constant of $\tau_{R1} \sim 2$ ps, followed by a decay with $\tau_D \sim 100$ ps. The induced signal completely disappears at room temperature [Fig. 1(a)].

The induced SHG signal from GaAs/GaSb/InAs samples is shown in Fig. 2(a). A new striking feature is a long-lived

---

[a] Electronic mail: yuri.d.glinka@vanderbilt.edu

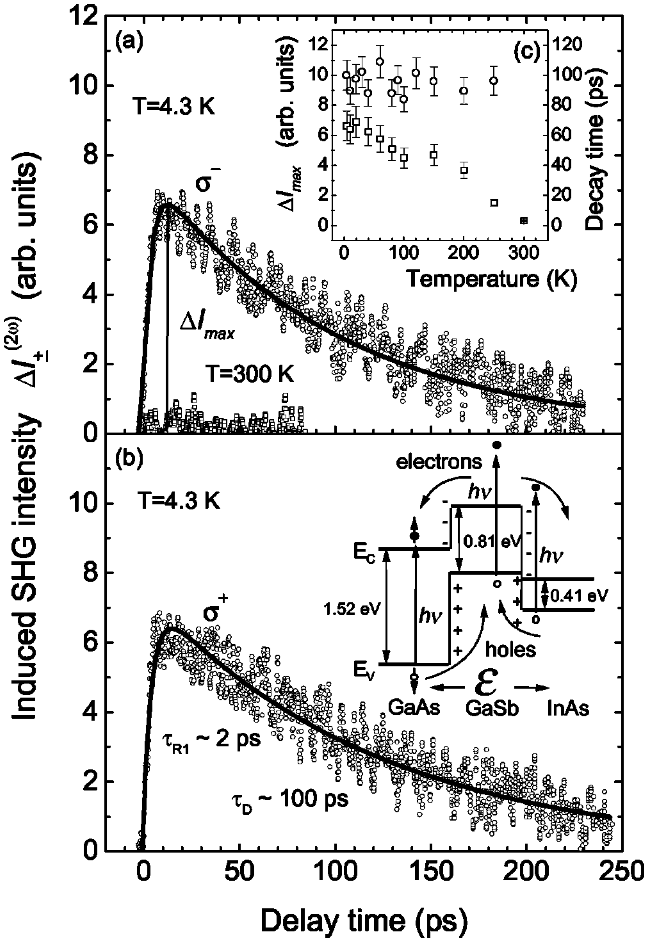

FIG. 1. Pump-induced SHG signal from GaAs/GaSb sample measured at 4.3 and 300 K for σ^- polarized (a) and σ^+ polarized (b) pump. The fit with exponential rise/decay functions is shown by solid curves. Inset (c): temperature dependence of maximum signal intensity, $\Delta I_{\max}$ (squares), and of decay-time constant, τ_D (circles). Initial band alignment for GaAs/GaSb/InAs heterostructure is schematically shown in (b).

$\tau_{R2} \sim 15$ ps rise-time component, which results in a shift of the signal peak towards longer times with respect to those for the GaAs/GaSb samples. Moreover, the SHG signal intensities for σ^- and σ^+ pump polarizations are different, indicating an induced magnetization, which we ascribe to the presence of spin-polarized electrons in the InAs layer. Since the spins were excited in the GaAs layer, this indicates an interlayer spin-polarized electron transport. Correspondingly, the long-lived $\tau_{R2} \sim 15$ ps rise-time component characterizes the rate of spin transfer to the InAs layer.

Retaining only linear terms in the induced electric field, $\epsilon(t)$, and magnetic field, $M(t)$, the nonlinear pump-probe polarization can be presented as[10,12]

$$P_{\pm}^{NL}(2\omega,t) = [\chi^{(2)} + \chi_e^{(3)}\epsilon(t) \pm \chi_m^{(3)}M(t)][E(\omega)]^2, \quad (1)$$

where $E(\omega)$ is the electric field component of the incident probe light, and $\chi^{(2)}$, $\chi_e^{(3)}$, and $\chi_m^{(3)}$ are the corresponding nonlinear susceptibilities. The alternate signs in Eq. (1) indicate two possible directions of the induced magnetic field normal to the interface. The magnetic- and electric-field-induced contributions were then extracted from the pump-induced SHG signal intensity, $\Delta I_{\pm}^{(2\omega)}(t)$, as

$$\Delta I_{-}^{(2\omega)} - \Delta I_{+}^{(2\omega)} \propto M(t), \quad \Delta I_{-}^{(2\omega)} + \Delta I_{+}^{(2\omega)} \propto \epsilon(t). \quad (2)$$

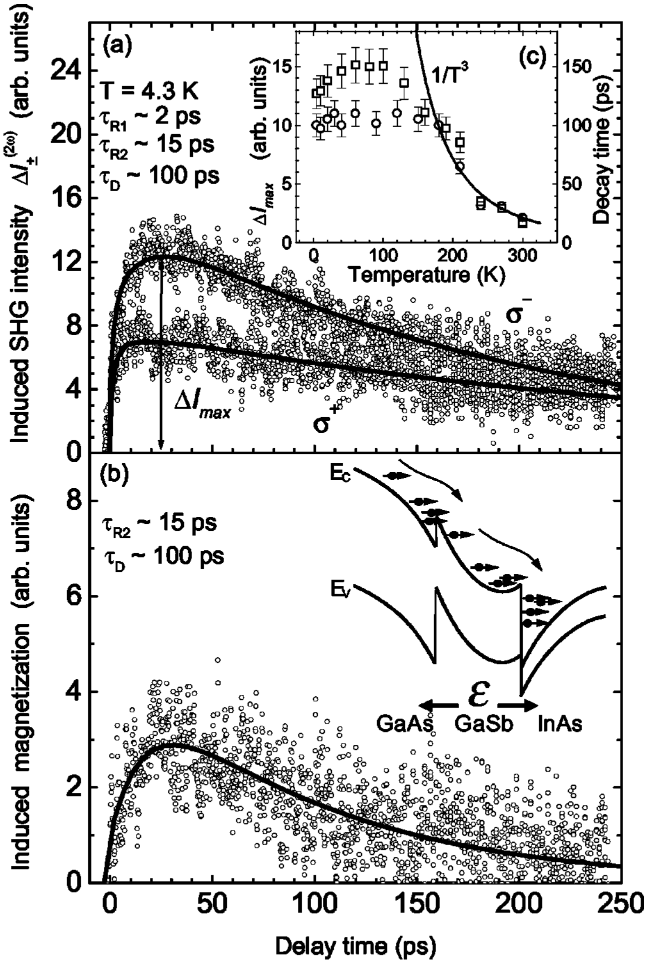

FIG. 2. Pump-induced SHG signal from GaAs/GaSb/InAs sample measured at 4.3 K with σ^+ and σ^- pump polarization (a) and extracted magnetization (b) obtained as a difference between signals in (a). The fit with exponential rise/decay functions is shown by solid curves. Inset (c): temperature dependence of maximum signal intensity, $\Delta I_{\max}$ (squares), and of decay-time constant, τ_D (circles). Band realignment due to interfacial electric fields for GaAs/GaSb/InAs heterostructure is schematically shown in (b).

Figure 2(b) shows the extracted induced magnetization for GaAs/GaSb/InAs heterostructure, whereas the extracted electric-field-induced signal closely follows that for the GaAs/GaSb sample.

Because the laser light was tuned just above the GaAs band gap, spin-polarized electrons excited in the smaller band gap GaSb and InAs layers are much more energetic (0.74 and 1.11 eV, respectively) and lose their spin polarization as they relax to lower energy states.[13] These unpolarized electrons accumulate in the GaAs and InAs regions while the holes are amassed in the GaSb layer. The resulting charge separation at the interfaces [inset in Fig. 1(b)] gives rise to the interfacial electric fields resulting in the initial growth of the SHG signal ($\sim$2 ps at 4.3 K). This rise time decreases to $\sim$300 fs for $T \sim 250$ K, matching the typical room-temperature values for carrier thermalization.[5–8] The induced electric fields at the interfaces bend the initial energy profile and lower the barrier at the GaAs/GaSb interface [inset in Fig. 2(b)]. A subsequent relaxation of the interfacial electric fields manifests itself as the $\tau_D \sim 100$ ps decay of the induced SHG signal.[14] The appearance of longer decay time in Fig. 2(a) originates from constant background due to a residual electric field at the GaSb/InAs interface. This background is present only in the GaAs/GaSb/InAs sample and reflects the semimetallic nature of the GaSb/InAs interface which does not fully ‘‘unbend’’ as the induced interfacial fields relax. The 100 ps decay-time constant (common for both samples) for interfacial electric fields is obtained after extracting this constant background. Importantly, the interfacial electric fields are known to be much stronger at the semimetallic GaSb/InAs interface as compared those at the GaAs/GaSb interface.[2] This leads to higher local spin density at the GaSb/InAs interface and, hence, to significant induced magnetization in the GaAs/GaSb/InAs sample.

The temperature dependence of the peak intensity for the GaAs/GaSb sample exhibits a sharp decrease in the range of 4.3–100 K, while for the GaAs/GaSb/InAs sample, the signal first grows and then stabilizes [Figs. 1(c) and 2(c)]. We attribute this behavior to thermally activated electrons in GaAs overcoming the interfacial barrier. In the GaAs/GaSb/InAs sample, the initial signal increase is attributed to the arrival of additional spins at the GaSb/InAs interface. Note that electrons with uncompensated spin in GaAs are activated first since they occupy states with higher energies. Subsequent signal stabilization in the range from 50 to 100 K is due to the competing process involving a decrease in the interfacial electric field as the unpolarized electrons begin to pass through the barrier. Further intensity decrease in the range from 100 to 300 K indicates a weakening of the interfacial fields as the electron wave functions become more extended, effectively reducing the carrier density at the interface. Note that the induced signal in GaAs/GaSb samples shows a plateau in the range from 100 to 170 K, which we attribute to the thermal activation of electrons previously trapped at the impurity centers in bulk GaAs.

The temperature dependence of decay-time constants is similar for both samples in the range from 4.3 to 180 K [Figs. 1(c) and 2(c)] staying at $\tau_D \sim 100$ ps. At higher temperatures, however, τ_D decreases as T^{-3} for the GaAs/GaSb/InAs sample, while remaining unchanged for the GaAs/GaSb sample. The T^{-3} dependence is consistent with Dyakonov–Perel (DP) mechanism,[13] and the measured room-temperature value $\tau_D \simeq 20$ ps matches that in InAs measured previously using pump-probe spectroscopy.[8] Note, however, that the DP mechanism is expected to dominate for temperatures down to ~ 50 K. The regime change at $T \simeq 180$ K indicates a crossover to interface dominated dynamics. As the electric field at the GaSb/InAs interface relaxes, the time evolution of spin density follows that of charge density, leading to a reduction of the magnetic-field-induced SHG signal.

In summary, pump-probe SHG measurements for nonmagnetic GaAs/GaSb/InAs semiconductor heterostructures revealed interlayer spin-polarized electron transport. We found that the optically induced magnetization dynamics in such structures originates from two distinct sources: one of them related to the evolution of the local spin density at the interfaces, and the other one arising from the spin relaxation. The extreme sensitivity of the SHG to the interfacial fields, which allowed us to distinguish between these two mechanisms, makes it a unique tool for studying the spin and carrier dynamics in multilayer semiconductors.

This work was supported by ONR and by the DARPA/SPINS Program.

[1] S. A. Wolf, D. D. Awschalom, R. A. Buhrman, J. M. Daughton, S. von Molnar, M. L. Roukes, A. Y. Chtchelkanova, and D. M. Treger, Science **294**, 1488 (2001).

[2] I. Vurgaftman, J. R. Meyer, and L. R. Ram-Mohan, J. Appl. Phys. **89**, 5815 (2001).

[3] J. F. Smyth, D. A. Tulchinsky, D. D. Awschalom, N. Samarth, H. Luo, and J. K. Furdyna, Phys. Rev. Lett. **71**, 601 (1993).

[4] A. P. Heberle, W. W. Ruhle, and K. Ploog, Phys. Rev. Lett. **72**, 3887 (1994).

[5] A. Tackeuchi, O. Wada, and Y. Nishikawa, Appl. Phys. Lett. **70**, 1131 (1997).

[6] Y. Ohno, R. Terauchi, T. Adachi, F. Matsukura, and H. Ohno, Phys. Rev. Lett. **83**, 4196 (1999).

[7] K. C. Hall, S. W. Leonard, H. M. van Driel, A. R. Kost, E. Selvig, and D. H. Chow, Appl. Phys. Lett. **75**, 3665 (1999).

[8] T. F. Boggess, J. T. Olesberg, C. Yu, M. E. Flatte, and W. H. Lau, Appl. Phys. Lett. **77**, 1333 (2000).

[9] I. Malajovich, J. J. Berry, N. Samarth, and D. D. Awschalom, Phys. Rev. Lett. **84**, 1015 (2000).

[10] K. H. Bennemann, J. Magn. Magn. Mater. **200**, 679 (1999).

[11] J. Reif, J. C. Zink, C. M. Schneider, and J. Kirschner, Phys. Rev. Lett. **67**, 2878 (1991).

[12] Yu. D. Glinka, W. Wang, S. K. Singh, Z. Marka, S. N. Rashkeev, Y. Rogachyova, R. Albridge, S. T. Pantelides, N. H. Tolk, and G. Lukovsky, Phys. Rev. B **65**, 193103 (2002).

[13] M. I. Dyakonov and V. I. Perel, in *Optical Orientation*, edited by F. Meyer and B. P. Zakharchenya (North-Holland, Amsterdam, 1984), p. 11; G. E. Pikus and A. N. Titkov, *ibid.* p. 73.

[14] The dynamics of the interfacial electric fields was studied using linearly polarized pump beam and will be published elsewhere.