Terahertz emission from $Ga_{1-x}In_xSb$

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(Received 24 March 2006; revised manuscript received 22 May 2006; published 17 August 2006)

We report an experimental study on terahertz (THz) emission from $Ga_{1-x}In_xSb$ with $0 \le x \le 1$. THz emission is excited by femtosecond near-infrared laser pulses. For this material system THz emission is maximized for an In mole fraction $x \approx 0.5$. The maximum in THz emission occurs as a result of carrier compensation ($N_A \approx N_B$) for this specific material composition. The THz emission from n-type InSb is twice as large than that from p-type GaSb. The THz emission from $Ga_{1-x}In_xSb$ is explained according to the photo-Dember model. The $Ga_{1-x}In_xSb$ material system enabled the study of the influence of carrier concentrations on the THz emission process in narrow band gap semiconductors. Our study demonstrates the existence of a compromise between the positive effect of high electron temperature provided by narrow band gap materials and the negative effect of a high intrinsic carrier concentration. This compromise dictates the extent to which the band gap in a semiconductor can be reduced in order to enhance the THz emission. This same analysis can be extended to explain why the THz emission from InSb is lower than that of InAs.

DOI: 10.1103/PhysRevB.74.075323 PACS number(s): 78.47.+p

INTRODUCTION

Excitation of semiconductor surfaces with femtosecond (fs) laser pulses at near-infrared (NIR) wavelengths is an important method to generate subpicosecond terahertz (THz) radiation pulses. This type of broadband THz-radiation source has enabled the development of time domain ultrafast THz spectroscopy and THz imaging for the otherwise difficult to access frequency range between 0.1 and 5 THz.[1] Many physical processes, such as rotational excitations in gases[2] and vibrations in many inorganic[3] and organic[4] crystalline structures, take place in this spectral region. Recent examples of major scientific breakthroughs include the ultrafast THz spectroscopy of single-base pair differences in femtomolar concentrations of DNA,[5] the observation of the temporal evolution of exciton formation in semiconductors[6] and understanding of carrier dynamics in high-temperature superconductors.[7] The development of bright, high bandwidth THz radiation sources is important in order to expand the applications. For this purpose it is necessary to understand the THz-emission process as determined by semiconductor properties.

Narrow direct band gap semiconductors such as InAs (E_g=0.35 eV),[8–16] and InN (E_g=0.68 eV) (Ref. 17) are particularly promising sources of optically excited THz radiation. Narrow band gap semiconductors are strong candidates for compact and lightweight time-domain THz spectroscopy and imaging systems powered by femtosecond fiber lasers with emission wavelengths at $\lambda = 1.55\ \mu m$ ($E \approx 0.8$ eV). Therefore, qualitative and quantitative understanding of THz emission from narrow direct band gap semiconductors has attracted great interest recently.

Initially it was assumed that the emission of a subpicosecond THz transient from the surface of a narrow band gap semiconductor upon incident of a femtosecond laser pulse is caused by acceleration of photocarriers in a surface depletion or accumulation field.[18,19] Surface field acceleration is the origin of femtosecond optically excited THz emission in semiconductors with wider band gaps such as GaAs or InP.[20] Subsequently, it was demonstrated that another mechanism, the built-up of a photo-Dember field, is also a strong source of femtosecond optically excited THz emission.[21] A photo-Dember field occurs as the result of different diffusion coefficients of electrons and holes. This effect is particularly pronounced in narrow band gap semiconductors because these materials are characterized by large differences between electron and hole mobilities, short optical absorption lengths, and large kinetic energies of electrons after photoexcitation. It has been demonstrated that the build-up of a photo-Dember field is the dominant mechanism of optically excited THz emission in n-type InSb (Ref. 8) p-type InSb, n- and p-type InAs (Refs. 10 and 13) and p-type GaSb.[22] Recently, it has been demonstrated that instantaneous polarization may contribute significantly to THz emission by p-type InAs with ⟨111⟩ surface orientation at large excitation fluence.[16]

Previous investigations of femtosecond optically excited THz emission by InSb and InAs (Refs. 8 and 10) are still insufficient to explain why InAs is a stronger THz emitter than InSb from the photo-Dember perspective. These investigations show that the THz emission from InSb is almost two orders of magnitude lower than that of InAs. From a quantitative point of view, there is the expectation that InSb would be a stronger THz emitter than InAs, because the band gap of InSb is lower than that of InAs providing higher kinetic energy to photoexcited electrons (Table I). Additionally the electron mobility in InSb is higher than the electron mobility in InAs (Table I). A higher probability of intervalley scattering of photoexcited electrons in InSb has been suggested[10] and experimentally identified[16] as one of the mechanism that limit THz emission in InSb compared to InAs (Table I).

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TABLE I. Summary of semiconductor parameters.

	GaSb	InSb	InAs
band gap (eV)	0.81[a]	0.24[a]	0.45[a]
electron mobility (cm^2/V s)	5000[b]	76000[c]	30000[c]
electron effective mass ($m_0=9.11\times10^{-31}$ kg)	$0.041m_0$[d]	$0.014m_0$[d]	$0.023m_0$[d]
electron scattering time (s)[e]	0.11×10^{-12}	0.59×10^{-12}	0.4×10^{-12}
hole mobility (cm^2/V s)	850[b]	800[c]	240[c]
intervalley scattering energy threshold ($\Gamma->L$) (eV)	0.084[d]	0.51[d]	0.73[d]

[a]Ref. 3.
[b]Ref. 27.
[c]Ref. 10.
[d]Ref. 26.
[e]Ref. 28.

In this report we investigate the influence of carrier concentrations on optically excited THz emission by narrow band gap semiconductors by studying THz emission of a $Ga_{1-x}In_xSb$ ingot with $0\leq x\leq 1$. This material system allows the uniform variation of the electron mobilities and the band gap from the GaSb values ($E_g=0.73$ eV) to the InSb values ($E_g=0.17$ eV). We demonstrate that the carrier concentration is a limiting parameter for photo-Dember field driven THz emission of a narrow band gap semiconductors. A trade-off exists between the positive effect of high kinetic energy of photoexcited electrons found in narrow band gap materials and the negative effect of a high intrinsic carrier concentration. This trade-off dictates the extent to which the band gap can be reduced in order to improve the photo-Dember field driven THz emission. This same analysis can be extended to explain why the THz emission from InSb is lower than that of InAs.

EXPERIMENTAL ARRANGEMENTS

A $Ga_{1-x}In_xSb$ ingot ($0.09<x<0.97$) was grown by the vertical Bridgman method.[23] A part of the ingot was sliced in wafers for dc Hall characterization and the results are shown in Fig. 1(a). In the low indium fraction region, the carrier concentration is 10^{17} cm^{-3}, and the sample is p type due to native acceptors in GaSb. In the high indium fraction region, the carrier concentration is 3×10^{16} cm^{-3}, and the sample is n type due to native donors in InSb. In other regions of the ingot where $0.09<x<0.97$, compensation occurs, and the carrier concentration is lower. The ingot has the lowest carrier concentration where the In mole fraction is about 50%.

Another section of the ingot was kept in one piece, with the surface extending from the region of low indium content ($x=0.09$) to the region of high indium content ($x=0.97$). The surface of the ingot was lapped and polished to achieve a high quality mirrorlike surface. For THz-emission measurements the ingot was mounted in a two-dimensional translation stage so that the surface of the ingot could be moved continuously through the laser excitation spot. This configuration made it possible to record quickly and with high spatial resolution the THz emission of a large area of the ingot.

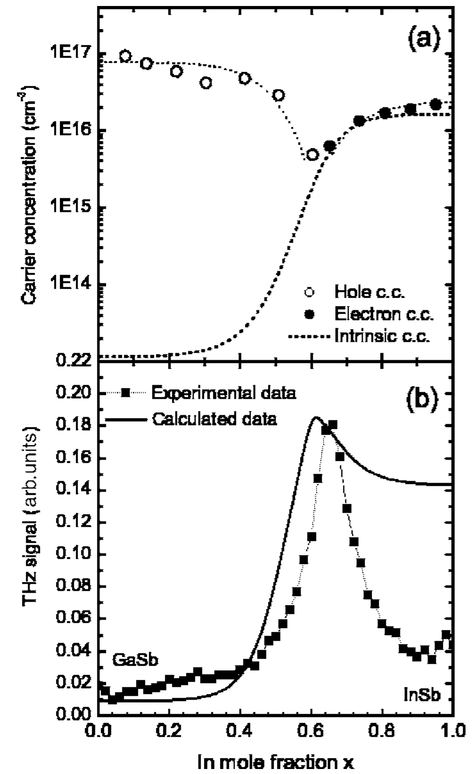

FIG. 1. (a) Electron and hole concentrations of the $Ga_{1-x}In_xSb$ samples (circles). The indium mole fraction x defines the carrier concentration because of native defect compensation. (b) THz emission amplitude from the $Ga_{1-x}In_xSb$ ingot (squares) vs In mole fraction x. THz emission amplitude is maximum where the In fraction is close to $x\approx0.5$ because native acceptors in GaSb compensate native donors in InSb. Experimental THz field amplitude (squares) and calculated THz emission from the photo-Dember model (solid line). THz emission is maximized for lowest carrier concentration.

The time-domain THz emission measurement setup that was used for this work is an optical pump-probe arrangement.[24] A commercial diode-pumped titanium-sapphire (Ti:S) laser delivers pulses with a duration from of 130 fs at a wavelength of 800 nm. The repetition rate is 82 MHz, the maximum average power is 700 mW. The laser beam is split into a pump beam and a probe beam using an uncoated glass as beamsplitter. The power ratio is typically 95% on the pump beam and 5% on the probe beam. Emission from unbiased semiconductor surfaces is achieved by exposing the surface to the pump light beam. The pump beam is slightly focused at the semiconductor surface, and with an optical absorption length of 200 nm and a laser spot size of 1 mm^2 the injected carrier density is approximately 10^{17} cm^{-3}. Measurements were performed setting the angle between the laser beam and the surface's normal to the Brewster's angle in order to minimize the Fresnel's losses at

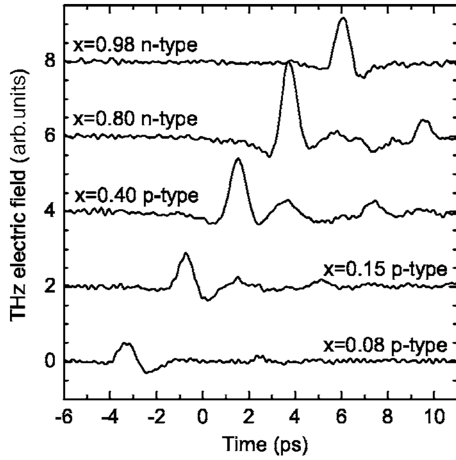

FIG. 2. THz transients of the $Ga_{1-x}In_xSb$ ingot for various In mole fractions x. No polarity reversal is observed when the majority carrier concentration chances from p to n type.

the semiconductor surface and therefore maximize the amount of photocarriers injected in the material. The pulsed THz radiation is detected through electro-optic sampling[25] using a ⟨110⟩ ZnTe crystal of 1 mm thickness.

RESULTS AND DISCUSSIONS

The experiment with the $Ga_{1-x}In_xSb$ ingot resulted in THz emission from all scanned regions, ranging from GaSb (x=0.09) to InSb (x=0.97). Figure 1(b) shows the indium content and the THz emission amplitude as a function of the In mole fraction. The THz emission is maximized when the indium mole fraction is close to x=0.5. In this position the native p-type defects in GaSb compensate the native n-type defects in InSb, therefore the carrier concentration is the lowest [Fig. 1(a)].

In previous experiments we demonstrated that the THz emission from p-type GaSb (unintentionally doped) is dominated by the photo-Dember emission mechanism.[22] Recent studies on n-type and p-type InSb (Ref. 10) have shown that the THz electric field polarity from samples of opposite carrier type do not exhibit reversal, and therefore the THz emission in n-type InSb is also attributed to the photo-Dember mechanism. Further, the surface field contribution in InSb is expected to be small because the depletion field is weak due to the narrow band gap character of InSb.[10] Some recorded THz field transients are shown in Fig. 2. We see that all transients have similar shape and similar polarity. From the observation that no polarity reversal occurs along the entire length of the $Ga_{1-x}In_xSb$ the THz emission mechanism is attributed to the photo-Dember effect. We have not regarded optical rectification as a possible mechanism of THz radiation because we did not observe that the strength of THz emission depends on the angle between polarization of the excitation laser beam and crystallographic orientation of the $Ga_{1-x}In_xSb$ surface.

The THz emission of n-type InSb ($x \approx 1$) is only twice as large than THz emission by p-type GaSb ($x \approx 0$). This is a surprisingly low difference considering that the ratio of the electron and hole mobilities of InSb is larger than that of GaSb and that the probability of intervalley scattering of photoexcited electrons is significantly lower in InSb than in GaSb.

Next, the THz emission was calculated for the $Ga_{1-x}In_xSb$ material system within the photo-Dember model [Eq. (1)]. As derived previously,[22] the magnitude of the emitted THz radiation in the far field E_{far} depends on the electron and hole concentrations n_b and p_b, the ratio of electron and hole mobilities $b=\mu_n/\mu_h$ and the electron temperature T_e. Furthermore, in Eq. (1), S is the laser focal spot size, τ is the laser pulse duration, r is the distance in the far field, and Δn the density of photocarriers:

$$E_{\text{far}} = \frac{S}{4\pi\varepsilon_o c^2 \tau r}\frac{k_B T_e \mu_n}{(1+b)}\left[\frac{b(n_b-p_b)}{1+b} \times \ln\left(1+\frac{\Delta n(1+b)}{p_b+bn_b}\right)-\Delta n\right]. \tag{1}$$

Equation (1) represents a low-frequency limit. The application of Eq. (1) in our analysis is justified because the shortest timescales probed in our experiments [~0.8 ps (Fig. 2)] are longer than the inverse of the scattering rate that determine the mobilities (~0.59 ps for InSb and 0.11 ps for GaSb, Table I).

The majority carrier concentrations n_b and p_b in $Ga_{1-x}In_xSb$ as a function of the In mole fraction x were obtained from the Hall data from extracted samples as shown in Fig. 1(a). The band gap values E_g as a function of the In mole fraction x were calculated from the following expressions;[23]

$$E_g = 0.18x + 0.7(1-x) - 0.415x(1-x). \tag{2}$$

The minority carrier concentrations in $Ga_{1-x}In_xSb$ as a function of the In mole fraction x were calculated from the intrinsic carrier concentration n_i. The intrinsic carrier concentrations were calculated from the electron (m_n^*) and hole effective mass (m_h^*) according to the following expressions:[23]

$$m_e^* = (0.041 - 0.0362x + 0.0092x^2)m_0, \tag{3}$$

$$m_h^* = (0.82 - 0.39x)m_0, \tag{4}$$

$$N_{c,v} = 2\left(\frac{2\pi m_{n,p}^* k_B T}{h^2}\right)^{3/2}, \tag{5}$$

$$n_i = \sqrt{N_c N_v}e^{-E_g/2k_BT}. \tag{6}$$

N_c and N_v are the density of states in the conduction band and in the valence band, respectively. The electron temperature was estimated to be $T_e=[k_B(h\nu-E_g)/3]\times(\Delta n/n_b)$, and the photoinjected carrier concentration is assumed to be constant and equal to $\Delta n = 2.5\times10^{17}$ cm^{-3}. The calculations are shown in Fig. 1(b) (solid line). The calculated electric field amplitude follows the experimental field amplitudes (squares) well. The THz emission maximum at an In mole fraction $x \approx 0.5$ is due to the logarithmic term in Eq. (1), which maximizes the radiated field at the lowest carrier concentrations. The small disagreement in THz field amplitude

between photo-Dember effect calculations and the experimental data in the Ga rich region may be due to the fact that in this region the hole carrier concentration is four orders of magnitude higher than the intrinsic carrier concentration [Fig. 1(a)], and therefore the estimate of electron carrier concentration from the intrinsic carrier concentration may be not sufficiently precise to fully reproduce the experiment. Experiment and theory qualitatively agree for $Ga_{1-x}In_xSb$ with $x>0.5$. However, calculations quantitatively deviate from experiments in this composition range. We attribute the quantitative discrepancy to the fact that our model does not include the effects of carrier-carrier scattering on the number of photocarriers available for THz emission.

In the InSb region the electron carrier concentration is of the same order of magnitude as the intrinsic carrier concentration, as shown in Fig. 1(a). The intrinsic carrier concentrations in the indium rich region are high because the band gap of InSb (0.18 eV for $x=0.97$) is small and as a result the factor $\exp(-E_g/2k_BT)$ is much larger than for GaSb. Previous analysis of the THz emission within the photo-Dember model suggested that materials with a high electron temperature after optical excitation and low probability of intervalley scattering are strong THz emitters.[8,10,16] Additionally, it was suggested[8,10] that a large electron mobility and a large difference between electron mobility and hole mobility would be beneficial for photo-Dember field driven THz emission. InSb has a higher electron mobility, smaller band gap and a lower probability of intervalley scattering than GaSb. The reason why THz emission from InSb is not significantly higher than that of GaSb is that the intrinsic carrier concentration in InSb, governed by the $\exp(-E_g/2k_BT)$ factor, is by 3 orders of magnitude higher than in GaSb. This high intrinsic carrier concentration dominates the carrier concentration in *n*-InSb and results in a negative effect on the THz emission process and limits the extent to which the THz emission efficiency can be enhanced by reducing the band gap.

The observation that the intrinsic carrier concentration of a material limits efficacy of photo-Dember field driven THz emission can also be extended to explain the difference in THz emission from InSb and InAs. Previous low temperature experiments on THz emission from InSb and InAs have shown that at room temperature the THz emission from InSb is lower than from InAs by an order of magnitude.[8,10] Higher probability of intervalley scattering in InSb compared to InAs has been identified as one of limiting factors.[16] However, as temperature decreases, THz emission from InSb increases faster than that of InAs, being InSb a stronger THz emitter than InAs at 20 K. This effect was explained[8] by the proportionality between the photo-Dember field and the electron mobility, and the fact that the mobility in InSb increases faster at low temperature than that of InAs. The mobility argument is insufficient to explain entirely this observation. Another reason for the enhanced THz emission of InSb at low temperature is that as the temperature decreases, the intrinsic carrier concentration is reduced, and this reduction has a positive effect on the photo-Dember emission mechanism.

CONCLUSIONS

Femtosecond optically excited THz emission from $Ga_{1-x}In_xSb$ was explained according to the photo-Dember model. For this material system THz emission is maximized for an In mole fraction $x\approx 0.5$. The maximum in THz emission occurs as a result of carrier compensation ($N_A\approx N_B$) for this specific material composition. The THz emission by *n*-type InSb is twice as large than that by *p*-type GaSb.

The $Ga_{1-x}In_xSb$ material system enabled the study of the influence of carrier concentrations on the THz emission process in narrow band gap semiconductors. Our study demonstrates the existence of a compromise between the positive effect of high electron temperature provided by narrow band gap materials and the negative effect of a high intrinsic carrier concentration. This compromise dictates the extent to which the band gap can be reduced in order to enhance the THz emission. This same analysis can be extended to explain why the THz emission from InSb is lower than that of InAs.

ACKNOWLEDGMENTS

This material is based upon work supported by the National Science foundation under Grant No. 0333314 and National Science Foundation (NSF) Faculty Early Career Development Award No. ECS-0093706.

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