Physical mechanism of coherent acoustic phonons
generation and detection in GaAs semiconductor

P Babilotte, E Morozov, P Ruello, D Mounier, M Edely, J-M Breteau, A Bulou and V Gusev
Laboratoire de Physique de l’Etat Condensé, UMR 6087 CNRS-Université du Maine, Avenue O. Messiaen 72085 Le Mans, France.
E-mail: pascal.ruello@univ-lemans.fr

Abstract. We first describe the picosecond acoustic interferometry study of GaAs with two-colors pump-probe laser pulses. The dependence of the generation process on the pump wavelength and the detection process on the probe wavelength both can cause the shift in the phase of the Brillouin signal. Secondly, in order to distinguish the short high frequency wide-band acoustic pulse from low frequency Brillouin contribution, we accomplished experiments with (100)GaAs semiconductor coated by a transparent and photoelastically inactive thin film, serving a delay line for the acoustic pulse. Even with highly penetrating pump light ($\xi \approx 680\, \text{nm}$), short acoustic disturbances of $\approx 7\, \text{ps}$ of duration have been registered.

1. Introduction
Knowledge of the fundamental processes of laser/matter interaction is essential to pave the way for generation of acoustic phonons with tunable characteristics such as wavelength, magnitude and polarization. One of the challenges is to get very high frequency coherent acoustic phonons with wavelength of the order of a few nanometers suitable to probe nanoscaled systems. First strategy consists in using nanometric metallic thermoelastic transducer [1, 2]. Secondly, the high frequency phonons can be tuned by the semiconductors superlattices period [3]. An other possibility, not dependent on a geometric factor, lies on the use of rapid intrinsic electron-hole dynamics in semiconductors [2]. Such approach requires the understanding of laser-matter interaction in the region of acoustic phonons emission. In particular, the relationship between acoustic emission and intervalley optical excitation, photoexcited electron-hole plasma recombination and carriers diffusion must be described. In this paper we report two experimental studies. The first one concerns the study of the influence of the pump and probe energy onto the phase of the Brillouin signal in GaAs. That phase provides information on the stages of the acoustic phonons generation driven by electron-hole plasma dynamics but also on detection mechanism involving for example inter-band process. Since the study of semi-infinite GaAs semiconductor in reflection geometry often provides essentially Brillouin signal due to large photoelastic effect, and for that reason can hide higher frequency phonons signal, we have worked, in a second experiment, with a photoelastically inactive and pump and probe transparent delay line (ZnO) deposited onto GaAs. In that experiment, and by choosing proper energy of probe, it has been possible to demonstrate the existence of a short acoustic pulse of around $7\, \text{ps}$ of duration even with deep penetration of pump light ($\xi \approx 680\, \text{nm}$).
2. Experimental part
A classical pump-probe set-up was used to monitor transient reflectivity spectra. A tunable (720-880nm) mode-locked Ti:Sapphire fs laser was employed with pulse repetition rate of 76MHz. The pump beam is chopped at high frequency with an acousto-optic modulator. The probe is delayed relative to the pump with a delay-line mirror. The signal is processed with a high band pass lock-in amplifier. A non-linear crystal of BBO is also employed for SHG. For the study of Brillouin phase we have worked with wafers of GaAs with two different dopings: a low \((10^{16} \text{cm}^{-3})\) \((E_g = 1.43\text{eV})\) and high \((10^{18} \text{cm}^{-3})\) \((E_g = 1.45\text{eV})\) doping. For the second experiment ZnO \((E_g = 3.4\text{eV})\) is deposited by reactive radio frequency magnetron sputtering onto the highly doped GaAs wafer. A Raman investigation performed over a depth of 52nm in GaAs, using \(\lambda_{cw} = 457.9\text{nm}\) as excitation radiation, within 0.7 \(\text{cm}^{-1}\) resolution, did not show any spectral change, that could probe for stress or defect, between free and ZnO coated GaAs.

3. Results and discussion
3.1. Phase of Brillouin signal
The measured transient reflectivity of the single color pump-probe experiment (red-red) is given in Fig.1. The Brillouin phase \(d\phi_E\) (Fig.2) was determined with mean least squares procedure, assuming that each maximum of oscillation has a \(i^{th}\) position in time determined by \(t_E(i) = T_E [d\phi_E/2\pi + i]\), where \(T_E\) is the Brillouin period for probe energy \(E\). A jump of the phase appears around the band gaps.

Since pump and probe energy change simultaneously the generation and detection processes are intimately correlated. Nevertheless, the following calculation shows that phase jump is more likely associated to a detection and not to generation process. The acoustic deformation spectrum \(U(w)\) driven only by deformation potential (more efficient process than thermoelasticity [4]) is determined by electron-hole plasma dynamics and is given by Eq.1 [2]

\[
U(w) \sim \frac{(-iw)^3 m_D w_D}{w_D (w_R - iw) + w^2} \left[ \frac{w_D}{w^2 + m_D^2 w_D^2} + \frac{1}{\sqrt{w_R - iw} (m_D w_D^{1/2} + \sqrt{w_R - iw})} \right]. \tag{1}
\]

where \(w_D = \frac{c^2}{D}\), \(w_R = \frac{1}{\tau_R}\) and \(m_D = \frac{a D}{C_\alpha} = \frac{\xi}{E_D^2 D}\). \(\xi = 1/\alpha\) is the optical depth penetration of the pump light. \(D\) is the carriers ambipolar diffusion coefficient, \(C_\alpha\) the longitudinal sound speed and \(\tau_R\) the volumic radiative recombination time. \(w\) was the probe energy dependent Brillouin frequency. A first theoretical calculation of \(U(w)\) has been performed for single color

**Figure 1.** Single color (red-red) transient reflectivity measured in GaAs (low doping) for various energies.

**Figure 2.** Brillouin phase obtained for single (a) and two colors (b) pump-probe experiments.
pump-probe experiment. All parameters required for that calculation are known: $\alpha$ can be found in Ref. [5], $D \approx 3.10^{-4}m^2/s$ and longitudinal sound speed ($C_a = 4800m/s$) are found in Ref. [4]. A rapid estimation shows that $\tau_R$ does not play a significant role since it exceeds 1ns in our range of photoexcitation ($max 10^{19}cm^{-3}$)[6]. In Fig.3a the magnitude of the acoustic deformation increases above the band gap as expected. The phase exhibits a small jump of less than 1 degree when crossing the band gap energy. This calculation suggests that the $\pi$ jump of the measured phase has its origin in the detection process. It is known that the modulated reflectivity of GaAs is given by $\Delta R/R \approx \frac{\delta \ln(R)}{\delta \varepsilon_1} \Delta \varepsilon_1$ for probe energy close to $E_g$ [7]. Following classical expression of real dielectric constant close to an interband transition [9, 8], it is possible to show that $\Delta \varepsilon_1 = \frac{\delta \varepsilon_1}{\delta E} \frac{\delta E}{\Delta Q} \Delta Q$ exhibits a change of the sign when the probe is tuned in the range of interband transition due to variations of $\frac{\delta \varepsilon_1}{\delta E}$ (see upper inset of Fig.1, Q is the normal coordinate). Such explanation seems to be reasonable since that jump shifts in the same way as band gap of GaAs caused by doping. Nevertheless, two sign modifications would be expected according to the upper inset of Fig.1 but was not observed in the range of investigation.

| $E_{\text{pump}}$ | $E_{\text{probe}}$ | $\Delta \varepsilon_1$ |
|------------------|------------------|------------------|
| 3000 eV          | 3000 eV          | 0.000            |
| 3050 eV          | 3050 eV          | 0.004            |
| 3100 eV          | 3100 eV          | 0.008            |
| 3150 eV          | 3150 eV          | 0.016            |
| 3200 eV          | 3200 eV          | 0.020            |
| 3250 eV          | 3250 eV          | 0.024            |

Figure 3. The theoretical calculations of the magnitude $U$ and of the phase of the single frequency $w$ acoustic deformation are given for three cases described in text.

In the two colors pump-probe experiment a jump of the Brillouin phase is also observed (see Fig.2b) consistently with detection process discussed above. The phase shift exhibits an additional large linear energy dependence which is not understood yet. Using Eq.1 a jump of the Brillouin phase due to generation processes is theoretically predicted if the carrier ambipolar diffusion drastically decreases when electrons are promoted directly in the L-valley where it is well known that the effective mass can be thirty times larger than that of the electron in the $\Gamma$ valley [8] (Fig.3b and Fig.3c). However, additional experimental verifications of this hypothesis are necessary. For that calculation, the parameter $\alpha$ is tabulated in Ref.[5] while we have kept the radiative recombination time at 1ns. The origin of that expected singularity lies on the fact that the $U(w)$ magnitude and its phase usually depend on the difference between $w$ (Brillouin frequency) and a characteristic frequency dependent on the properties of the excited media. Below L-edge ($E_{\text{pump}} = 2.82eV$, $\alpha = 2.8.10^7 m^{-1}$), the photo-excited carriers remain in the $\Gamma$-valley ($D \approx 3.10^{-4}m^2/s$) and the characteristic frequency $w_D/2\pi$ and the parameter $m_D$ become $\approx 12GH\text{z}$ and $1.9$, while above L-edge ($E_{\text{pump}} = 3.1eV$, $\alpha = 6.7.10^7 m^{-1}$) they become $\approx 360GH\text{z}$ and 0.13 respectively. As a consequence, such drastic change modifies the phase of $U(w)$.

3.2. Short acoustic pulse in ZnO-GaAs

The experimental transient signals obtained in ZnO-GaAs for three configurations (Fig.4) are given in Fig.5. Large similarities are observed for configuration (1) and (2). The signal is composed of two main parts. The first one concerns signal between zero time delay and time of 60ps. The low frequency oscillations are unambiguously attributed to Brillouin scattering in
GaAs. At the time \( t_2 = 60 \text{ps} \), oscillations exhibit a jump in magnitude but with remaining value of the period for \( t > 60 \text{ps} \). That event is due to the coming back to the ZnO-GaAs interface of the phonons which have propagated in ZnO thin film \( (V_L(\text{ZnO}) = 6096 \text{m/s}) \). The increase of the Brillouin magnitude is due to interferometric effect and changes as a function of ZnO thickness. These two spectra reveal essentially Brillouin oscillations in GaAs. Nevertheless, at \( t_1 \approx 30 \text{ps} \) (conf. 2) and \( t_2 \approx 60 \text{ps} \) (conf. 1), some pulse-like disturbances (\( \approx 7 \text{ps} \)) are superimposed onto Brillouin signal.

![Figure 4](image)

**Figure 4.** Geometry of the system with characteristics of the three configurations.

The existence of high frequency acoustic phonons is clearly confirmed by the detected pulse \( (\tau_a \approx 7 \text{ps}) \) obtained with the configuration 3 where optical depth penetration of probe beam \( (13 \text{nm}) \) is suitable to detect short pulse and to prevent large contribution of Brillouin signal. The magnitude of that acoustic pulse increases linearly with photo-excited carriers density (maximum of \( 10^{19} \text{ cm}^{-3}\)) and neither variation of the shape nor of the duration was observed disregarding non-linear carriers recombination process as driving mechanism of short pulse generation[1].

In the classical approach of optoacoustics, the acoustic pulse duration scales with \( \Delta t = \xi/C_a \). In the configuration 3, that characteristic time would be around 100ps. The measured pulse echo is an order of magnitude shorter. The duration of 7ps indicates that the process occurs over a spacial characteristic length of \( l = \tau_a \times C_a \approx 33 \text{nm} \). This shows that the short acoustic pulse detected here has its origin in an intrinsic phenomenon taking place in the ZnO-GaAs interface. First analysis of the pulse shape and first calculation indicate that the acoustic pulse detected here could come from a rapid surface recombination of carriers. A rough estimation of the diffusion length of carrier over the pulse duration \( \tau_a \) provides \( l_D \approx 45 \text{nm} \) \( (D \approx 3.10^{-4} \text{m}^2/\text{s} [4]) \). That characteristic diffusion length compares pretty well with the other characteristic length of 33nm deduced above. That provides estimate for the surface recombination velocity \( S \approx l_D/\tau_a \approx 6.4.10^5 \text{cm/s} \) which is a reasonable value for GaAs [10].

**References**

[1] Gusev V and Karabutov A 1993 *Laser Optoacoustics* AIP New York

[2] Akhmanov S and Gusev V 1992 *Sov. Phys. Uspekhi* 35 153

[3] Yamamoto A, Mishina T, Masumoto Y and Nakayama M 1994 *Phys. Rev. Lett.* 73 740

[4] Wright O B, Perrin B, Matsuda O and Gusev V E 2001 *Phys. Rev. B* 64 081202(R)

[5] Aspnes D E and Studna A A 1983 *Phys. Rev. B* 27 985

[6] McLean D G, Roe M G, D’Souza A I and Wigen P E 1986 *Appl. Phys. Lett.* 48 (15) 992

[7] Seraphin B O and Botella N 1966 *Phys. Rev.* 145 628

[8] Yu P Y, Cardona M 2001 *Fundamentals of Semiconductors, Springer* Third Edition

[9] Bartels A, Dekorsy T, Kurz H and Köhler K 1999 *Phys. Rev. Lett.* 82 1044

[10] Beck S M and Wessel J 1987 *Appl. Phys. Lett.* 50(3) 149