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Observation of optical phonon instability induced by drifting electrons in semiconductor nanostructures

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We have experimentally proven the Cerenkov generation of optical phonons by drifting electrons in a semiconductor. We observe an instability of the polar optical phonons in nanoscale semiconductors that occurs when electrons are accelerated to very high velocities by intense electric fields. The instability is observed when the electron drift velocity is larger than the phase velocity of optical phonons and rather resembles a ‘‘sonic boom’’ for optical phonons. The effect is demonstrated in $p-i-n$ semiconductor nanostructures by using subpicosecond Raman spectroscopy. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1563730]

When electrons are accelerated by an electric field such that their drift velocity exceeds the sound velocity of the semiconductor, a large number of acoustic phonons are emitted coherently. This so-called ‘‘Cerenkov acoustoelectric effect’’ was predicted[1,2] and demonstrated[3–6] in the 1960s in semiconductors with large piezoelectricity[3] such as CdS, and multivalley crystals with electron–phonon interaction via the deformation potential.[5] A similar effect, but for optical phonons, was also predicted,[6–11] but has never been directly observed. This is in spite of the fact that optical phonons play such a major role in the energy relaxation of fast (hot) electrons in semiconductors.[12] If an optical phonon instability is induced by hot electrons during their transport, it will have enormous impact on the carrier dynamics in semiconductor devices; particularly, in nanostructure devices which inherently have large applied electric fields and electron drift velocities. In addition, the instability leads to amplification/generation of coherent optical phonons. It can be suggested that a number of applications will become possible on the basis of the electric methods of generation of coherent optical phonons.

In this work, we use subpicosecond Raman spectroscopy to study both the transient electron and the transient phonon dynamics in GaAs-based $p-i-n$ semiconductor nanostructures. An anomalous increase of the longitudinal optical (LO) phonon occupation number is observed when the applied electric field intensity is larger than 10 kV/cm. We attribute this anomaly in the LO phonon population to the amplification of LO phonons produced by electrons during their transient transport in the GaAs-based $p-i-n$ nanostructure.

The GaAs-based nanostructures used in this work consisted of an AlAs–GaAs–AlAs $p-i-n$ structure grown by molecular beam epitaxy on a (001)-oriented GaAs substrate. Details of the sample structure has been described elsewhere.[13] The sample was excited and probed by the outputs of two optical parametric amplifiers (OPA1 and OPA2) pumped by a common pulse from a Ti–sapphire amplifier system which is composed of the ultrastable Millennia/Tsunami short pulse oscillator and the Spitfire regenerative amplifier with the Merlin Nd:YLF pump laser. The output from OPA1 having a pulse width of about 600 fs (full width of half maximum) is chosen to operate at photon energy of $\hbar\omega_{\mathrm{pump}} \cong 1.65$ eV and is used to excited electron-hole pairs in GaAs-based $p-i-n$ semiconductor nanostructures, whereas, the output from OPA2 having the same pulse width is used to probe both the phonon populations and electron distributions with a variety of photon energies and wavevector transfers, as indicated below.

Details of our experimental technique have been reported in Ref. 14. All the data reported here were taken at $T \cong 10$ K and at zero time delay between the pump and probe pulses. The electron–hole pair density excited by the pump

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TABLE I. Measured electron drift velocity as a function of the applied electric field intensity for a GaAs-based $p-i-n$ nanostructure taken at $T = 10$ K.

E (kV/cm)	V_d ($\times 10^7$)	E (kV/cm)	V_d ($\times 10^7$)
1	1.65 ± 0.1	40	6.2 ± 0.5
2	2.30 ± 0.2	45	6.1 ± 0.5
3	2.62 ± 0.2	50	6.0 ± 0.5
4	2.95 ± 0.3	55	6.2 ± 0.5
5	3.01 ± 0.3	60	6.3 ± 0.5
7.5	3.80 ± 0.4	65	6.1 ± 0.5
10	5.5 ± 0.5	70	6.0 ± 0.5
12.5	5.6 ± 0.5	75	6.1 ± 0.5
15	5.5 ± 0.5	80	6.0 ± 0.5
20	5.7 ± 0.5	85	6.0 ± 0.5
25	5.8 ± 0.5	90	6.1 ± 0.5
30	6.0 ± 0.5	95	6.1 ± 0.5
35	6.1 ± 0.5	100	6.0 ± 0.5

pulse is $n \cong 2.5 \times 10^{17}$ cm^{-3}. The effective electric field intensity inside the laser-irradiated area was estimated by using the Franz–Keldysh effect similar to Ref. 15.

If $I_S(\omega_i)$, $I_{AS}(\omega_i - \omega_{LO})$ are the measured Raman scattering intensities for Stokes, anti-Stokes lines taken with photon energies $\hbar\omega_i$, $\hbar(\omega_i - \omega_{LO})$, respectively, then the population of nonequilibrium LO phonons $n(\omega_{LO})$ is obtained by,

$$n(\omega_{\rm LO}) = \left[\frac{I_s(\omega_i)}{I_{\rm AS}(\omega_i - \omega_{\rm LO})} - 1 \right]^{-1}$$

(see Refs. 16 and 17). The electron drift velocity as a function of the applied electric field intensity, deduced from the measured electron distributions, is listed in Table I.

The measured transient nonequilibrium LO phonon occupation number as a function of the applied electric field for a GaAs-based $p-i-n$ nanostructure is shown in Fig. 1 for two phonon wave vectors as indicated. The nonequilibrium LO phonon occupation number with wave vector $q \cong 6.44 \times 10^5$ cm^{-1} gently increases as the electric field intensity increases to $\cong 10$ kV/cm, and then flattens out. This can be reasonably well explained by ensemble Monte Carlo calculations that include conventional electron–phonon scattering.[18] In contrast, the nonequilibrium LO phonon occupation number with wave vector $q \cong 1.096 \times 10^6$ cm^{-1} exhibits a very different behavior. The nonequilibrium phonon population first increases smoothly as the applied electric field intensity increases up to ≈ 7.5 kV/cm. It increases very sharply to a maximum at $E \cong 10$ kV/cm, then, decreases slowly to an almost constant value at an electric field intensity of 60 kV/cm and higher. Two aspects are worthwhile pointing out: first, the observed phonon occupation for the phonon wave vector at $q \cong 1.096 \times 10^6$ cm^{-1} is significantly smaller than that at $q \cong 6.44 \times 10^5$ cm^{-1} for electric field intensity up to $E = 7.5$ kV/cm; second, the phonon occupation is much larger for the phonon wave vector at $q \cong 1.096 \times 10^6$ cm^{-1} than at $q \cong 6.44 \times 10^5$ cm^{-1} for electric field intensity $E \geqslant 10$ kV/cm. The former can be very well understood by considering the $1/q^2$ dependence of the Fröhlich interaction[19] for the emission of LO phonons by energetic electrons. However, the latter is contrary to this expectation. For electric field intensity $E \geqslant 10$ kV/cm, the LO phonon occupation for $q \cong 1.096 \times 10^6$ cm^{-1} increases by a factor of about 3 over that for $q \cong 6.44 \times 10^5$ cm^{-1}, instead of the expected decrease by a factor of about 3 (if the $1/q^2$ dependence is assumed).

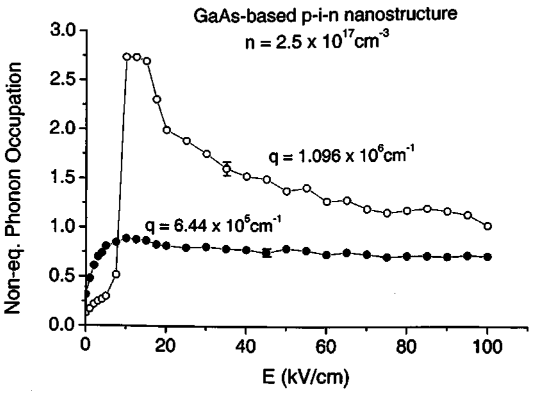

FIG. 1. Measured nonequilibrium LO phonon occupation as a function of the applied electric field intensity for GaAs-based $p-i-n$ nanostructures for phonon wave vectors $q \cong 6.44 \times 10^5$ cm^{-1} and $q \cong 1.096 \times 10^6$ cm^{-1}, respectively. The injected electron–hole pair density is $n \cong 2.5 \times 10^{17}$ cm^{-3}. The abrupt increase of the nonequilibrium phonon population at $E = 10$ kV/cm is attributed to the detection of amplification of GaAs LO phonons.

The effects of intervalley scattering will significantly reduce the number of electrons in the Γ valley after about 1 ps (Refs. 20–23) and the intra-X or intra-L valley electron relaxation emits much larger wave-vector LO phonons than $q \cong 1.096 \times 10^6$ cm^{-1} as a result of their much heavier electron mass; therefore, the effects of intervalley scattering processes cannot account for our observed results. The penetration depths for the probe laser at $q \cong 1.096 \times 10^6$ cm^{-1} and $q \cong 6.44 \times 10^5$ cm^{-1} are about 900 and 5000 Å, respectively. The influence of the carrier excitation at the cap GaAs layer (50 Å) and the p-type AlAs layer (100 Å) is, therefore, minimal. Under the application of an electric field, electrons acquire a significant drift velocity, and tend to escape from the probing volume of the laser. However, due to the consideration of the penetration depth, this effect will tend to reduce the measured LO phonon occupation more for the phonon wave vector at $q \cong 1.096 \times 10^6$ cm^{-1} than for the phonon wave vector at $q \cong 6.44 \times 10^5$ cm^{-1}; therefore, this cannot explain our experimental results. We attribute the anomalous increase of the phonon-occupation observed for electric field intensity $E \geqslant 10$ kV/cm to the amplification of LO phonons in the GaAs-based $p-i-n$ nanostructure during the electron's transport from the p to n regions.

A simple way to evaluate the criterion of the optical phonon instability is to use a macroscopic approximation, which is based on well-known equations for the optical displacement **u**, the polarization **P**, and the field **E**.[24]

The criterion of optical phonon instability is then given by[25]

$$\Gamma(q) \equiv \frac{2\pi e^2 n_e \omega_{\rm LO}}{\hbar q^3} \left(\frac{1}{\varepsilon_\infty} - \frac{1}{\varepsilon_0} \right) \left[f\left(\frac{\omega_{\rm LO}}{q} + \frac{\hbar q}{2m_e^*} \right) - f\left(\frac{\omega_{\rm LO}}{q} - \frac{\hbar q}{2m_e^*} \right) \right] > \gamma, \tag{1}$$

where $\omega_{\rm LO} \equiv \omega_{\rm TO}\sqrt{\varepsilon_0/\varepsilon_\infty}$; e, m_e^* are the charge and effective mass of an electron, respectively; n_e is the electron density; $\hbar = h/2\pi$, h is Planck's constant; and f is the electron distri-

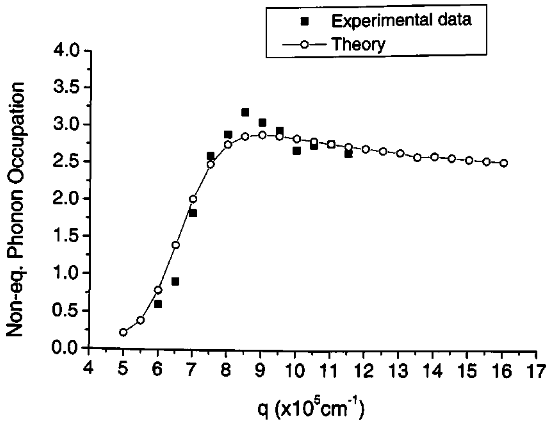

FIG. 2. Comparison of the measured nonequilibrium LO phonon occupation with the theory of phonon amplification. The data were taken at an applied electric field intensity of $E = 10$ kV/cm.

bution function. ε_0, ε_∞ are the static and high frequency dielectric constants, respectively; ω_{TO} is the angular frequency of the transverse optical phonons; γ is the phonon damping constant. Here, since the lifetime of LO phonons for GaAs at $T = 10$ K is about 9 ps,[26,27] we have $\gamma \cong 1.1 \times 10^{11}\ \mathrm{s}^{-1}$. Because the phonon occupation is proportional to the square of the amplitude of the phonon wave, the measured phonon occupation is expected to be proportional to $\exp[2\Gamma(q)\tau]$,[25] where τ is the time interval during which the intensity of the phonon wave grows.

Figure 2 shows the measured LO phonon intensity as a function of the phonon wave vector at an applied electric field intensity of $E = 10$ kV/cm. The best fit between the theory and experimental data is obtained when $\tau = 500$ fs. This value is consistent with the fact that the electron velocity overshoot phenomenon lasts for about 1 ps in GaAs.[28]

Therefore, the experimental results shown in Fig. 1 can be interpreted as follows: The LO phonon occupation increases as the applied electric field intensity increases for up to $E = 7.5$ kV/cm, primarily as a result of relaxation of energetic electrons by emitting LO phonons. The measured phonon occupation roughly follows $1/q^2$ dependence, as expected from the Fröhlich interaction. As the electric field intensity increases to and beyond $E = 10$ kV/cm, the electron drift velocity increases with the applied electric field intensity (as indicated in Table I), the phonon occupation with the wave vector at $q \cong 6.44 \times 10^5\ \mathrm{cm}^{-1}$ decreases slightly because of the effect of intervalley scattering processes and the drifting away of electrons from the probing volume. On the other hand, phonon occupation having wave vector $q \cong 1.096 \times 10^6\ \mathrm{cm}^{-1}$ increases very rapidly for $E = 10$ kV/cm due to the amplification of optical phonons, the electron drift velocity increases with the applied electric field intensity (as indicated in Table I), the phonon occupation then decreases with further increase of electric field intensity due to the effect of intervalley scattering and the drifting away of electrons from the probing volume.

We have also carried out similar experiments in an $\mathrm{In}_{0.53}\mathrm{Ga}_{0.47}\mathrm{As}$-based $p-i-n$ nanostructure (which are not shown here). The optical phonon instability for GaAs-like optical phonons is observed. Therefore, our results confirm that optical phonon instability is a universal phenomenon for polar semiconductors.

In semiconductor nanostructure devices, electron transport is primarily governed by electron velocity overshoot effects;[18,28] as a result, electron drift velocity usually can be as high as 10^8 cm/s. This extremely high electron drift velocity means that optical phonon modes will be amplified for a wide range of the phonon wave vectors. The phonons with the smallest wave vectors from this wave-vector interval can be probed by Raman spectroscopy, as demonstrated in this work. Since the strength of the Fröhlich interaction is inversely proportional to the square of the phonon wave vector for nanostructures of size $\geqslant 500$ Å in which quantum confinement effects are minimal, our experimental results indicate that instability/amplification of LO phonons will have an enormous impact on the carrier dynamics and relaxation in semiconductor nanostructure devices.

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