

# Ultrafast reflectivity changes in photoexcited GaAs Schottky diodes

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The transient reflectivity of GaAs Schottky diodes is measured by femtosecond time resolved pump-probe experiments. The measured reflectivity for photon energies near the band gap reveals transient quasio oscillatory behavior with frequencies up to 5.5 THz. The changes of the reflectivity are due to extremely fast changes of the carrier density within the depletion layer. We interpret the observed oscillatory signal as coherent plasma oscillations. Ensemble Monte Carlo simulations for the scenario agree well with the observed plasmon frequencies. © 1996 American Institute of Physics. [S0003-6951(96)00520-7]

Ultrafast charge transport processes after optical excitation in semiconductors have been a subject of comprehensive investigation since femtosecond laser-pulse techniques have been established.<sup>1</sup> By pulsed excitation and time-resolved spectroscopy, coherent oscillations such as coherent tunneling,<sup>2</sup> Bloch oscillations,<sup>3-5</sup> and coherent phonons<sup>6</sup> have been observed in semiconductors. Coherent oscillations on the femtosecond time scale are important due to promising applications as a source of coherent THz radiation.<sup>7</sup> Furthermore, coherent oscillations reveal the physical behavior of the excited system immediately after photoexcitation, and before the phase information of the exciting laser pulse has been destroyed by various scattering processes.

A new mechanism for coherent oscillations in semiconductors is based on the optical excitation of *coherent plasmons*. Coherent plasma oscillations can be generated by injection of electron-hole pairs into a region of high electric field which acts as a driving force on the electrons and holes. Recent experiments<sup>8</sup> show that in the ballistic time regime ( $t < 500$  fs) a photoexcited cold electron-hole plasma in a GaAs *pin* diode oscillates with frequency close to the plasma frequency,  $\omega_p^2 = n_e \cdot e^2 / m^* \cdot \epsilon \cdot \epsilon_0$  ( $n_e$ ... electron density,  $m^*$ ... effective electron mass). In addition, it has been shown that the damping time of the plasmons is correlated with the momentum relaxation time for single electrons. In 2D systems<sup>9</sup> coherent plasma oscillations have recently been observed by detecting the emitted THz radiation. In this work, we report time resolved reflectivity pump-probe experiments in the space charge region of GaAs Schottky diodes. Injection of cold electron-hole pairs with ultrashort laser pulses leads to an oscillatory behavior of the electron density within the space charge region. We interpret the measured reflectivity signals as coherent plasma oscillations. Ensemble Monte Carlo simulations confirm that under our experimental conditions coherent oscillations of the electron density and the electric field within the depletion region are present.

In our experiments, we use Schottky diodes consisting of (100) oriented *n*-GaAs with a 1.4  $\mu\text{m}$  thick *n*-doped layer (Si doped,  $N_D = 3 \times 10^{16} \text{ cm}^{-3}$ ) grown on a semi-insulating

GaAs substrate and a 10 nm thick semitransparent Cr layer on the surface of the *n*-GaAs sample. The sample is kept at a temperature of  $\sim 100$  K in order to operate near the maximum of the electron mobility. The calculated width of the depletion region for this Schottky diode<sup>10</sup> is about 180 nm and the corresponding maximum electric field is about 70 kV/cm.

We performed time-resolved reflectivity experiments using a Kerr lens mode-locked tunable Ti:sapphire laser with a 76 MHz repetition rate. The laser pulses ( $\sim 90$  fs) are split into an intense pump and a weak probe pulse which are linear polarized perpendicular to each other. The probe beam is polarized parallel to the  $\langle 110 \rangle$  crystal direction. A shaker periodically delays the probe beam with a frequency of 1 Hz; the pump beam is chopped with a frequency of 4 kHz. The focus of the probe beam on the sample is kept smaller than that of the pump beam which has a diameter of about 60  $\mu\text{m}$ . The photon energy of the laser pulses is tuned around the band-gap energy ( $\sim 1.5$  eV). A large area silicon *pin* photodiode followed by a lock-in amplifier and a signal averager records the reflected probe beam intensity.

In Fig. 1, the relative change of the reflectivity is plotted as a function of the time delay between pump and probe pulse. The central wavelength of the laser pulses (spectral width  $\sim 12$  nm) was tuned from  $\lambda_c = 835$  nm to  $\lambda_c = 813$  nm. The average power of the pump pulse is about 200 mW while that of the probe pulse is about 100 times lower. In the range from  $\lambda_c = 835$  nm to  $\lambda_c = 820$  nm all signals exhibit a fast initial rise of the reflectivity during the absorption of the pump pulse. Depending on the central wavelength of the laser pulse, a minimum and a second maximum occur followed by a decrease to a quasistationary level on a picosecond time scale. The position of the second maximum nears the first with decreasing wavelength. At  $\lambda_c = 813$  nm the second maximum is only indicated as a shoulder appearing on the decay of the reflectivity signal. At wavelengths shorter than  $\lambda_c = 813$  nm (not plotted) oscillations are no longer observed.

The large reflectivity change in the measured signal is caused by the photoexcitation of the system. Injection of free electron-hole pairs into the space-charge region of the Schottky depletion layer leads to a change of the occupation

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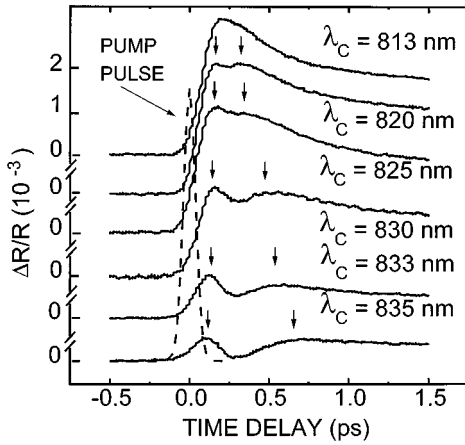


FIG. 1. Relative change of the reflectivity of the GaAs Schottky diode ( $T \sim 100$  K) as a function of the delay between pump and probe pulse. The wavelengths specify the center of the spectral distribution ( $\Delta\lambda \sim 12$  nm). The pump pulse is centered around 0 ps.

density (carrier distribution function) of the conduction band and valence band in the semiconductor. As a consequence, the absorption coefficient decreases due to band filling.<sup>11</sup> According to the Kramers–Kronig relations, the refractive index changes in the spectral range of the excitation and the probe pulse. For the case of normal incidence the change of the reflectivity  $\Delta R$  is related to the change of the refractive index  $\Delta n$  by:

$$\Delta R = \frac{(n-1)^2}{(n+1)^2} \cdot \frac{4 \cdot \Delta n}{n^2-1}, \quad (2)$$

provided that  $\Delta n \ll n-1$ . The contributions of the other physical effects that modify the reflectivity (electromodulation,<sup>12</sup> band-gap renormalization, free-carrier absorption,<sup>11</sup> and the electro-optic effect<sup>13</sup>) are smaller. Therefore, the reflectivity directly probes the local carrier density below the surface.

Theoretical, ensemble Monte Carlo simulations show the dynamics of the excited system below the semiconductor surface. Figure 2 shows the electric field 70 nm below the surface as a function of time after excitation for three different carrier densities. The calculated system is for a Schottky diode with the parameters used in our experiment. Excitation of electron-hole pairs occurs within a penetration depth of  $\sim 1.5 \mu\text{m}$  at the  $\Gamma$  point with 100 fs FWHM pulses. The results of the space- and time-dependent Monte Carlo simulations, taking into account carrier-phonon interaction, impurity scattering, but without the Pauli exclusion principle and carrier-carrier scattering demonstrate that under the conditions of our experiment coherent oscillations of the electron density and the electric field within the surface depletion region are present. The oscillation frequency is close to the plasma frequency and increases with increasing excitation density.

The dynamical behavior of the calculated field and carrier density is due to the generation of electron-hole pairs in the space charge region of the Schottky depletion layer. Because of the vertical electric field the charge carriers are separated within the Schottky depletion region. The corresponding electric field grows in sign opposite to the built-in

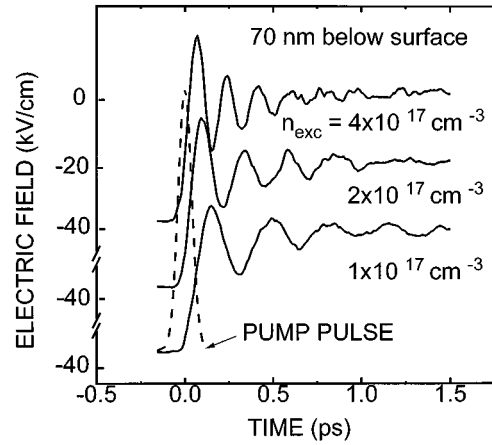


FIG. 2. Results of ensemble Monte Carlo simulation: Electric field as a function of time after the excitation pulse. The pump pulse is centered around 0 ps, the electric field is plotted at a distance of 70 nm below the semiconductor chromium junction for three different excitation densities.

electrostatic field. For the conditions of our measurements the resulting electric field changes its sign driving the carriers back until the total field strength again reverses in sign. Under the condition of weak scattering (ballistic regime) the electrons are expected to oscillate in real space. The oscillation frequency is precisely the plasma frequency with the photoexcited electron concentration. One can estimate an upper limit for the momentum relaxation time  $\tau_m$  of single electrons as  $\tau_m = m^* \cdot \mu / e$ , using the electron mobility  $\mu$  for a certain doping concentration. A comparison of  $\tau_m$  with the plasma frequency  $\omega_p$  results in a damping of the plasma oscillations to an amplitude of  $1/e$  within roughly one oscillation period.

The oscillation periods of the experimentally observed reflectivity oscillations are shown in Fig. 3. The plotted oscillation periods are the time  $\tau_p$  between the two maxima in the experimental curves (Fig. 1) from  $\lambda_C = 835$  nm to  $\lambda_C = 813$  nm. As we will show in the following, the main effect of changing the excitation wavelength is a change of the photoexcited carrier density. In order to compare the measured oscillation periods with the theoretical plasma frequency ( $\omega_p = 2 \cdot \pi / \tau_p$ ), the excitation density is determined

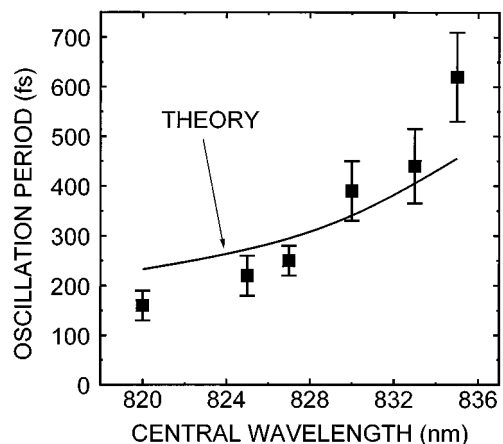


FIG. 3. Comparison of the measured and calculated oscillation period as a function of the central wavelength of the spectrum.

by taking into account the considerable decrease of the absorption coefficient due to band filling effects<sup>11</sup> and an exponential decay of the joint density-of-states (DOS) into the band gap<sup>14,15</sup> due to potential energy fluctuations. The photoexcited carrier density at shorter excitation wavelengths is higher due to the strongly increasing joint DOS towards higher energies. The theoretically calculated plasma frequencies yield the same order of magnitude and show the same qualitative dependence on the excitation wavelength. *Therefore, we interpret the observed oscillations as coherent plasma oscillations of the photoexcited electrons.*

The differences between theory and experiment in Fig. 3 probably arise from neglecting excitonic and impurity effects in the estimate of the carrier density at the band gap, as well as from the uncertainty in the determination of the precise band filling. Up to about 30 meV above the band gap the joint DOS is the main limiting factor for the excitation density because of absorption saturation.

The reflectivity signals in Fig. 1 also show a varying damping behavior. The higher the excitation energy the less pronounced are the oscillations. Ultrafast carrier thermalization and strong acceleration of the photoexcited electron-hole pairs lead to considerable occupation of energy levels above the LO-phonon energy (36 meV). This gives rise to efficient electron-phonon scattering which is mainly responsible for the short momentum relaxation time. The electron-phonon scattering time is  $\sim 180$  fs, close to the plasma oscillation period for the carrier concentrations in our experiment. At high excitation energies ( $\lambda_c > 813$  nm) a considerable fraction of electrons is already generated with an excess energy more than the LO-phonon energy. In addition, due to the larger joint DOS the number of the photoexcited electron-hole pairs is in a range where the electron-hole scattering rate is comparable to the electron-phonon scattering rate. Both increased scattering rates lead to a shortening of the momentum relaxation time and to a stronger damping at high carrier energies.

The stronger damping observed in the experiments (Fig. 1) as compared to the theoretical simulations (Fig. 2) is partly due to electron-hole scattering which is not taken into account in the theory.

Another feature which is observed in the measured reflectivity signals is a broadening of the second maximum compared to the first one. This effect could be a consequence of a decrease of the carrier density. Although the holes have negligible oscillation amplitudes (due to their large effective mass) they influence the electron oscillation by their space charge. At the metal-semiconductor junction the holes can diffuse very fast into the chromium since the Fermi energy in the metal is far above the edge of the valence band. A further reason for the broadening of the second maximum could be an increase of the effective electron mass. The electrons gen-

erated close to the surface are highly accelerated. Without scattering these electrons can occupy energy levels from which they are scattered into the *L* and *X* valleys where their effective masses are much higher and their mobilities much lower than in the  $\Gamma$  valley.

We are aware that the transient reflectivity signals in our experiment resemble some of the data in highly doped *p*-GaAs,<sup>16</sup> explained as a result of the time dependent carrier distribution. Further investigations are clearly necessary in order to obtain a full understanding of the transient optical properties around the band gap after photoexcitation.<sup>17</sup>

In summary, we have presented a time resolved study of the transient reflectivity changes in GaAs Schottky diodes. Our experimental results are in good quantitative agreement with theoretical predictions of coherent plasma oscillations, driven by the ballistic dynamics of photoexcited carriers in the surface space charge field. We will continue these experiments in systems with larger momentum relaxation times. This also includes experiments in 2D systems<sup>9</sup> where the electron mobility can be much higher than in 3D systems.

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<sup>1</sup>For a review, see *Hot Carriers in Semiconductor Nanostructures*, edited by J. Shah (Academic, New York, 1992), Chap. IV, and *Ultrashort Laser Pulses: Generation and Applications*, 2nd ed., edited by W. Kaiser (Springer, Berlin, 1993).

<sup>2</sup>K. Leo, J. Shah, E. O. Göbel, T. C. Damen, S. Schmitt-Rink, W. Schäfer, and K. Köhler, *Phys. Rev. Lett.* **66**, 201 (1991).

<sup>3</sup>J. Feldmann, K. Leo, J. Shah, D. A. B. Miller, J. E. Cunningham, T. Meier, G. von Plessen, A. Schulze, P. Thomas, and S. Schmitt-Rink, *Phys. Rev. B* **46**, 7252 (1992).

<sup>4</sup>K. Leo, P. Haring Bolivar, F. Brüggemann, R. Schwedler, and K. Köhler, *Solid State Commun.* **84**, 943 (1992).

<sup>5</sup>C. Waschke, H. G. Roskos, R. Schwedler, K. Leo, H. Kurz, and K. Köhler, *Phys. Rev. Lett.* **70**, 3319 (1992).

<sup>6</sup>G. C. Cho, W. Kütt, and H. Kurz, *Phys. Rev. Lett.* **65**, 764 (1990).

<sup>7</sup>H. G. Roskos, M. C. Nuss, J. Shah, K. Leo, D. A. B. Miller, A. M. Fox, S. Schmitt-Rink, and K. Köhler, *Phys. Rev. Lett.* **68**, 2216 (1992).

<sup>8</sup>W. Sha, A. L. Smirl, and W. F. Tseng, *Phys. Rev. Lett.* **74**, 4273 (1995).

<sup>9</sup>M. Vossebürger, H. G. Roskos, F. Wolter, C. Waschke, and H. Kurz, *J. Opt. Soc. Am. B* **13** (1996) (in press).

<sup>10</sup>*Semiconductor Surfaces and Interfaces*, edited by W. Mönch (Springer, Berlin, 1993).

<sup>11</sup>B. R. Bennet, R. A. Soref, and J. A. del Alamo, *IEEE J. Quantum Electron.* **26**, 113 (1990).

<sup>12</sup>H. Shen and M. Dutta, *J. Appl. Phys.* **78**, 2151 (1995).

<sup>13</sup>L. Min and R. J. D. Miller, *Appl. Phys. Lett.* **56**, 524 (1990).

<sup>14</sup>B. I. Halperin and M. Lax, *Phys. Rev.* **148**, 722 (1966).

<sup>15</sup>H. C. Casey, Jr. and F. Stern, *J. Appl. Phys.* **47**, 631 (1976).

<sup>16</sup>A. Davidson, R. C. Compton, F. Wise, D. Mars, and J. Miller, *J. Appl. Phys.* **76**, 2255 (1994).

<sup>17</sup>G. C. Cho, H. J. Bakker, T. Dekorsy, and H. Kurz, *Phys. Rev. B* **53**, 6904 (1996).