Coherent Plasma Oscillations in Bulk Semiconductors

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Coherent subpicosecond electron-hole charge oscillations that result from ballistic transport in the presence of a constant built-in field are observed in a bulk GaAs *p-i-n* sample by using differential electroabsorption techniques.

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Here, we report the first direct temporal observation of coherent plasma oscillations in a bulk semiconductor in the presence of a constant built-in field, and we confirm that these oscillations are the result of ballistic charge transport by demonstrating that they occur on a time scale that is short compared to the separately measured dephasing time. Even though plasmons have been observed and indirectly studied for many years via Raman spectroscopy [1], the direct observation of plasma oscillations in the time domain is significant because it allows the impulsive generation of large amplitude plasma oscillations that could not be realized in the Raman scattering experiments, and it raises the prospect of studying plasmons in the nonlinear regime. In addition, the study of the damping of coherent plasma oscillations is more likely to be relevant to the investigation of scattering processes in the context of transport than measurements of the dephasing of the coherent polarization [2] using four-wave mixing, where cold carriers (such as excitons) are typically studied and transport is not directly measured.

Specifically, in our experiment, electrons and holes are suddenly generated into bulk material in the presence of a constant field. For times short compared to the mean scattering time, the electrons and holes are initially accelerated in opposite directions by the presence of the constant field. As they move apart, a space-charge field develops as the result of the Coulomb attraction between the oppositely charged species. Until collisions dampen the oscillations, this restoring force causes the plasma to execute simple-harmonic motion at a frequency given by

$$\omega_p^2 = ne^2/\varepsilon m^* , \tag{1}$$

where ω_p is the plasma frequency, n is the carrier density, e is the electronic charge, ε is the dielectric constant, and m^* is the effective mass.

This is not the first attempt to observe the motion of particles in semiconductors in the absence of collisions. The prospect of studying ballistic transport has intrigued the solid-state community since it was first discussed by Shockley more than three decades ago [3]. While this idealized limit has never been fully realized [4], nearly ballistic peaks in the energy (momentum) distribution [5–14], velocity overshoot [15], and the nearly ballistic acceleration [16,17] of electrons have all been reported. These observations not withstanding, coherent plasma oscillations represent a new and important observation in the field of ultrafast transport because they represent the first clear demonstration of the self-consistent coherent coupling between the carrier transport and the dynamics of the electric field in a bulk material.

Coherent oscillatory electronic wave packets have been observed in coupled quantum wells [18], and Bloch oscillations have been observed in superlattices [19,20]; however, these are based on completely different mechanisms than the plasma oscillations described here. The former are based on quantum mechanical tunneling effects, and the latter are restricted to motion in minibands [19,20], where the charge motion is over the entire miniband and where oscillations are caused by Bragg reflections at the Brillouin zone edge. By contrast, the coherent plasma oscillations described here are limited to a small region of the Brillouin zone near the center (at the band edge), and the oscillations are produced by the restoring space-charge field (not by a reflection from the Brillouin zone edge).

Moreover, the coherent plasma reported here ballistically oscillates at THz frequencies. Terahertz radiation generated by transient space-charge fields (produced in semiconductors by the absorption of ultrashort optical pulses) has been the subject of considerable study [21]. Very recently, features have been observed [22] in the time-resolved studies of such THz emission that have been attributed to near ballistic acceleration and transport, but to our knowledge coherent charge oscillations have not been observed in such experiments.

Our measurements were performed in a GaAs vertical *p-i-n* structure that consists of a 400 nm intrinsic region sandwiched between 10 nm thick n and p regions doped to a density of 1.5×10^{17} cm^{-3}. The sample was cooled to 80 K to reduce phonon scattering and, thereby, to increase the mean time between collisions, and no external bias was applied. We estimate the built-in field across the intrinsic region to be 21 kV/cm at room temperature; however, a self-consistent calculation (that includes thermal generation, drift, diffusion, and recombination) indicates that it

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is reduced to ~ 10 kV/cm at 80 K because of the accumulation of spatially separated carriers due to the slowed recombination at the lower temperature. It is well known that the presence of a field of this magnitude will cause a significant alteration of the absorption properties near the band edge [23,24]. The change in the absorption properties of our sample upon application of this field is illustrated in Fig. 1, which shows the difference between the sample absorption when the built-in field is unscreened and when it is fully screened by photogenerated carriers.

Electrons and holes with minimal kinetic energy were photogenerated throughout the intrinsic region by 100 fs pump pulses from a mode-locked Ti:saphire laser that was tuned to the GaAs band edge. The temporal evolution of the change in the sample absorption $\Delta\alpha$ was then monitored by measuring the transmission of a time-delayed probe pulse. For times short compared to the mean scattering (or dephasing) time, at least two processes would be expected to contribute to the change in the absorption coefficient. One is the bleaching associated with phase-space filling, and the other is the change in the near-band-edge electroabsorption as the ballistic charge oscillations screen the built-in p-i-n field.

In order to isolate and monitor the changes in the electroabsorption associated with the ballistic charge oscillations, we made two essential changes to the standard pump-probe geometry. First, in order to accentuate the small, but rapidly varying, changes in the probe transmission associated with oscillating electroabsorption, we arranged to measure the time derivative of the differential transmission:

$$\frac{\partial(\Delta T)}{\partial\tau} = \frac{\partial}{\partial\tau}[T(\text{pump}) - T(\text{no pump})], \qquad (2)$$

where T(pump) and T(no pump) are the probe transmission with and without, respectively, the pump present. The differential transmission was extracted by chopping the pump and detecting the probe using a lock-in amplifier, and the time derivative was provided by continuously scanning the probe delay and carefully ac coupling the lock-in to a digital oscilloscope. Second, to enhance the sensitivity of our measurements to changes in the near-band-edge electroabsorption, we placed a spectral band-pass filter before the probe detector. The 8 meV bandwidth and the center frequency of the filter were chosen to ensure that only a single peak in the change in the electroabsorption was monitored, as indicated in Fig. 1.

Representative measurements of the time derivative of the change in transmission measured using this simple technique are shown in Fig. 2(a) for several pump fluences. Several features are worth noting. First, notice that clear oscillations are observed over a range of fluences. These oscillations decay in a time period of a few hundred of femtoseconds, consistent with the expected mean time between collisions at this temperature in GaAs. Finally, the frequency of oscillation clearly increases with increasing fluence, as illustrated in Fig. 2(b), where the

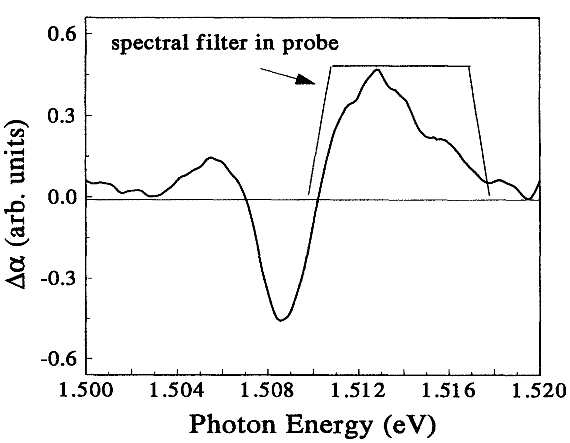

FIG. 1. The change in the absorption coefficient of GaAs at band edge at 80 K induced by the built-in p-i-n field. The rectangle indicates the spectral region monitored by the probe.

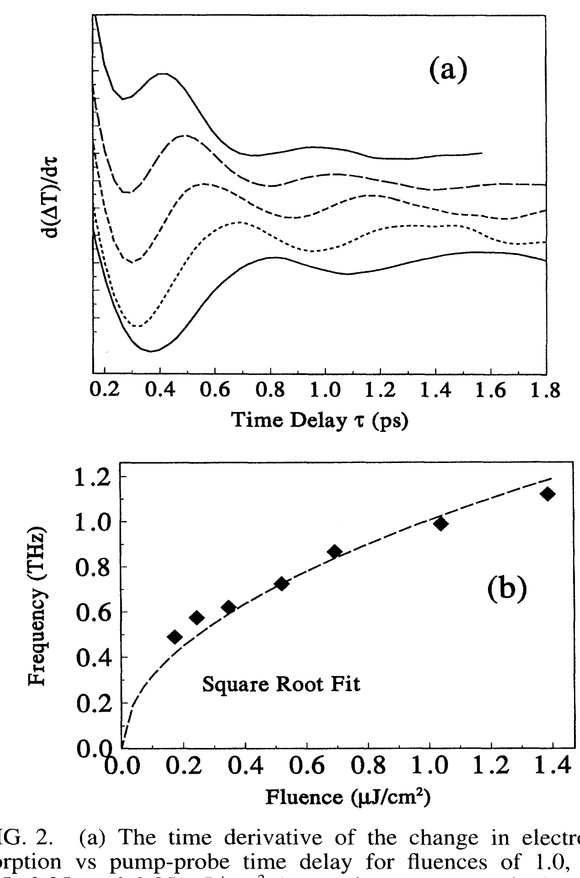

FIG. 2. (a) The time derivative of the change in electroabsorption vs pump-probe time delay for fluences of 1.0, 0.7, 0.5, 0.35, and 0.25 μJ/cm^2 (top to bottom, respectively), and (b) the frequency of the oscillating electric field (as extracted from the electro-absorption measurements) as a function of incident optical fluence. The dashed curve is a least squares fit by a square-root function.

oscillation frequency is plotted for a range of fluences. In obtaining these data, we verified that the optical absorption was linear over this range of fluences, and therefore this figure illustrates that the frequency of oscillation has a square-root dependence on carrier density, consistent with the plasma frequency given by Eq. (1). (The approximate carrier density in number/cm^3 can be obtained by multiplying the fluence in μJ/cm^2 by 1.1×10^{16}.) In fact, by estimating the carrier densities, we find that the measured frequencies deviate from the calculated plasma frequencies $\omega_p/2\pi$ less than 25% for the entire fluence range. In making these comparisons, we have taken into account that the electroabsorption is sensitive to the magnitude of the electric field, and therefore that it oscillates at twice the frequency of the oscillating space-charge field.

Also, from an elementary simple-harmonic model for the ballistic charge oscillations, we can estimate the amplitude of the oscillations and the maximum kinetic energy gained in these experiments as

$$x_{\max} = 2\varepsilon E_B/ne\,, \tag{3}$$

$$E_{\max}^{\text{kin}} = \varepsilon E_B^2/2n\,, \tag{4}$$

respectively, where E_B is the built-in field before the pump is incident. The results (shown in Fig. 3) indicate that, for low fluences, the carriers move ~200 nm and gain more than 10 meV in energy before collisions disrupt their motion. In making these estimates, it was necessary to include the effects of carrier accumulation from pulse to pulse at the laser repetition rate of 2.5 MHz used here. Because of this accumulation, we do not know the quasi-steady-state built-in field E_B precisely, but, from more extensive pump-probe measurements at longer delays and from self-consistent numerical calculations that include generation, recombination, drift, and diffusion, we estimate it to be reduced to roughly 3.5 kV/cm. The curves shown in Fig. 3 were based on this value.

To verify that our oscillations were indeed associated with the built-in field, we also repeated the measurements described above on an undoped electric-field-free sample under otherwise identical conditions. No oscillatory behavior was observed. In addition, to verify that our oscillations were the result of ballistic charge motion, we measured the dephasing of the coherent macroscopic polarization induced by the pump in the same undoped sample by using transient degenerate four-wave-mixing (FWM) techniques [2]. Specifically, for the FWM measurements, two 100 fs pulses with propagation vectors $\mathbf{k}_1$ and $\mathbf{k}_2$ were incident on the sample, and the temporally integrated FWM signal diffracted in the $2\mathbf{k}_2$-$\mathbf{k}_1$ direction was measured as a function of time delay τ_{12} between the two pulses. The results are shown in Fig. 4 for selected fluences. Notice that the quasiexponential decay times range from 250 fs for the lowest fluence to ~100 fs for the highest. Relating these measured decays in the FWM signal to the dephasing times is known to be complicated by whether the system is homogeneously or inhomogeneously broadened and by many body effects [2,25]. Nevertheless, by spectrally resolving the temporally integrated FWM signals shown in Fig. 4 using the techniques described in Ref. [26], we estimate that the dephasing times of the carriers are in the range of 400–500 fs for the fluences shown. By comparison, the decay times extracted from the oscillations shown in Fig. 2(a) vary from 300 to 500 fs (±100 fs). If one assumes that the same collisions that dephase the macroscopic polarization also dampen the plasma oscillations, then the excellent match between the dephasing times measured in the FWM experiments and the damping times of the oscillations confirms that our observed transport is indeed in the ballistic regime.

To exclude the possibility that the oscillations are the result of intervalley scattering between the Γ and L valleys, we note that the maximum energy gained

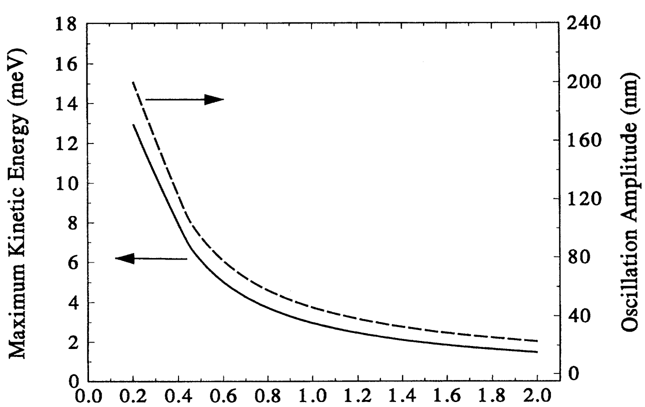

FIG. 3. The estimated amplitude of the ballistic oscillations (dashed line) and the maximum kinetic energy (solid line) acquired per carrier as a function of incident fluence.

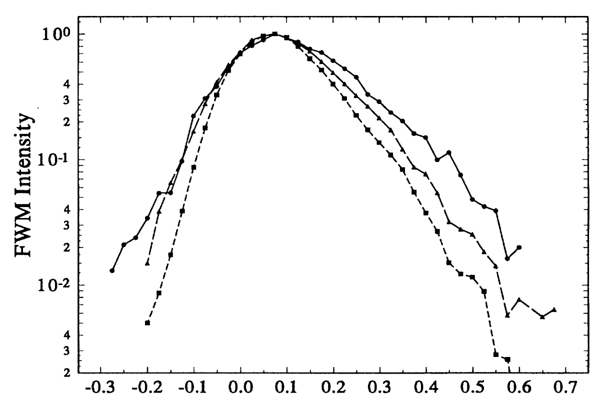

FIG. 4. The time-integrated degenerate-four-wave-mixing signals in an undoped, electric-field-free bulk GaAs sample for pump fluences of 0.29, 1.2, and 2.9 μJ/cm^2 (top to bottom).

per carrier (as shown in Fig. 3) is of the order of 15 meV, which is much lower than the ~300 meV needed for intervalley transfer (and for Gunn oscillations [27]). Finally, to exclude the possibility that the oscillations in transmission observed here have a similar origin to the oscillations in reflectivity reported recently [28], we note that the latter have been attributed to the abrupt production of coherent phonons by the injection of electrons and holes by a femtosecond pulse. Consequently, in the latter, the amplitude of the oscillations in reflectivity is determined by the density of injected carriers and the frequency by the optical phonon energy. In our experiments, the frequency is determined by the number of carriers injected, and the observed frequencies (ranging from ~0.5 to 1.1 THz) are clearly distinct from the optical phonon frequency in GaAs (~8.8 THz). Consequently, we conclude that we have observed coherent oscillations of electrons and holes in a bulk semiconductor and that the charge oscillations are the result of ballistic transport that occurs on a subpicosecond time scale.

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