THz pulses from the creation of polarized electron-hole pairs in biased quantum wells

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(Received 3 June 1992; accepted for publication 24 August 1992)

We report generation of coherent terahertz electromagnetic transients from GaAs/Al\textsubscript{0.3}Ga\textsubscript{0.7}As quantum wells in perpendicular fields. Although, at low temperature, the quantum well barriers suppress the transport current perpendicular to the layers by at least two orders of magnitude compared to bulk, we observe terahertz signals that are comparable in strength to those generated from bulk GaAs surfaces. This directly proves that the field-induced polarization of photoexcited electron-hole pairs is an important mechanism for the generation of terahertz radiation at semiconductor surfaces.

It has been known for some time that terahertz electrical transients can be generated from the surface of many semiconductors, such as Si, GaAs, and In\textsubscript{P}, when the surface is illuminated with a femtosecond laser pulse.\textsuperscript{1,2} The effect was originally explained by the acceleration of the photoexcited carriers within the surface depletion field of the semiconductor. The resulting time varying current $J(t)$ radiates an electromagnetic transient $E\sim \partial J/\partial t$ that consists of a single cycle of the electric field and contains frequency components up to a few terahertz. Consistent with this explanation in terms of current transport are the observed dependence of the amplitude and polarity of the radiated field on the strength and polarity of the built-in field,\textsuperscript{1,2} and also the observed amplitude increase with increasing carrier mobility as inferred from temperature-dependent measurements.

Recently, a further mechanism in those experiments was proposed in terms of optical rectification and the field-induced second-order nonlinear susceptibility $\chi^{(2)}$, that is equally well capable of explaining many of the above observations made in the generation of THz pulses from semiconductor surfaces.\textsuperscript{3} The latter explanation does not involve current transport but instead is based on the photoexcitation of electrons and holes in the depletion field into states in which they are already polarized. This creates a polarization $P$ that grows with the integrated pulse energy, and hence radiates an electrical transient according to $E\sim \partial^2 P/\partial t^2$. Evidence for this mechanism has been obtained from an orientation dependence in the efficiency of the generation of THz pulses when the semiconductor wafer is rotated around its surface normal, which is not predicted in the current transport picture.\textsuperscript{3} Also, we recently observed a broadband THz signal in an experiment involving coherent charge oscillations in a coupled quantum well system and attributed this transient to the instantaneous $e$-$h$ polarization after optical excitation.\textsuperscript{4}

Although in bulk semiconductors a separation of both mechanisms is not easy, we can clearly prove the importance of the creation of polarized $e$-$h$ pairs in the generation of THz pulses in single quantum wells. In quantum wells, current transport at low temperatures is strongly inhibited in the direction perpendicular to the layers, whereas the creation of polarized electron-hole pairs is not. In Fig. 1 we schematically draw the electron and hole envelope wave functions in a biased quantum well. The wave functions of the electron and hole are spatially displaced, resulting in a dipole moment $\mu$ after optical excitation. Since the barriers strongly inhibit current transport, we expect the generation of THz radiation to be completely dominated by the creation of polarized electron-hole pairs. We would like to point out that unlike the transport current model, the duration of the electromagnetic transient generated in this process should be limited by the pulse duration only and not by the acceleration of carriers.

The quantum well sample consists of 82 periods of 145 Å thick GaAs wells, separated by 102 Å Al\textsubscript{0.3}Ga\textsubscript{0.7}As barriers on a Si substrate. An electric field perpendicular to the quantum wells can be applied through a semi-transparent chromium Schottky contact on top. The electric field in the sample is calibrated by measuring the shift of the exciton peaks from the quantum confined Stark effect (QCSE).\textsuperscript{5} The sample is mounted in a continuous flow liquid helium cryostat and kept at a temperature of 77 K to suppress thermionic emission that might otherwise contribute to current transport. An argon-ion pumped self-mode locked Ti: Sapphire laser delivers tunable pulses around 800 nm with a 100 fs duration and an energy of 1.5 nJ per pulse. Mode-locking of the laser is resonantly started using a weak saturable absorber in a dye jet containing a mixture of the dyes HITCI and IR-140.\textsuperscript{6} Part of the experimental setup is shown in Fig. 2. The laser beam is split in two. One beam with an average power of roughly 50 mW is focused onto the quantum well sample under a 45° angle of incidence with its polarization parallel to the plane of incidence. The spot size at the sample is roughly 1 mm. The generated THz radiation emerges from the sample in the same direction as the partially reflected laser beam and leaves the cryostat through a high-resistivity silicon window. A pair of off-axis parabolic mirrors collimates and then focuses the radiation onto a 50 μm photoconducting dipole antenna\textsuperscript{7,8} with a silicon hyperhemispherical substrate lens. The second laser beam is sent through a variable delay and is used to gate the dipole antenna. By measuring the photocurrent from the antenna versus the delay between the gating pulse and the THz signal, we measure the electric field $E(t)$ (both phase and amplitude) of the
FIG. 1. Schematic diagram of the envelope wave functions of the electron and hole in a biased quantum well. The electric field shifts the electron and hole wave functions to opposite sides of the well, creating a dipole moment $\mu$.

The sample is excited roughly 4 nm above the band edge, near the $lh$ and the $hh$ excitons. Because of the near-bandgap excitation, we create excitons (rather than uncorrelated electron-hole pairs) in our experiment. Although the $lh$-$hh$ splitting (8.1 meV at flatband) is within our excitation bandwidth, we do not observe any effects from the simultaneous excitation of the $lh$ and $hh$ excitons, because of the broad exciton linewidths in the present sample. Care is taken to keep the laser detuning from the band edge constant as the band edge is shifted by the QCSE. Also mounted in the cryostat is a sample of semi-insulating (LEC-grown) bulk GaAs. We can measure the THz emission from this sample under conditions that are identical to those of the MQW. This allows us to compare the relative amplitudes of the THz emission from the MQW and the GaAs bulk sample.

The detected THz waveforms as a function of field in the sample are plotted in Fig. 3. We can see that for decreasing positive field strengths the signal decreases until it becomes almost zero when the built-in field is nearly canceled. This suggests that contributions to the THz signal that are not field-induced are small. For negative field strengths, the amplitude increases again but this time the polarity of the signal is reversed. However, the shape of the electromagnetic transients does not change significantly in the displayed field range. We note that the speed of these signals is presumably limited by the finite response time of the photoconducting dipole antenna, having a 3 dB bandwidth of roughly 1.5 THz.

We can eliminate transient transport currents as the source of THz radiation in our data by the following arguments: There are two contributions to the transport current in a quantum well—thermionic emission and tunneling through the barriers. Carrier sweepout times from quantum wells have been measured in a GaAs/Al$_{0.3}$Ga$_{0.7}$As quantum well structure with 65 Å barriers. They deduced a tunneling time of a few hundred picoseconds for carrier tunneling at field strengths comparable to ours. This implies that transport current by carrier tunneling in our sample (which has thicker and higher barriers) will be at least 100 times lower than in bulk GaAs. They also showed that below 100 K, thermionic emission from quantum wells, with a barrier height comparable to ours, is negligible compared to the tunneling current. At a sample temperature of 77 K we can therefore safely assume that current transport by thermionic emission is not a significant source of THz radiation in our experiment. We obtain additional evidence that current transport is not responsible for our measured signals from the Fourier spectra of the measured signals. In Ref. 12 the Fourier spectra of the THz signals from a silicon $p$-$i$-$n$ structure showed an upward frequency shift with increasing field strength, consistent with the current transport model. In Fig. 4 we plot the Fourier transforms of time-domain signals at three different field strengths. Within the accuracy of our measurements we do not observe a frequency shift.

The above arguments confirm that transport current in our quantum wells is at least 100 times lower than in bulk GaAs. However, the THz signal amplitude at 15 kV/cm is down by only a factor of two compared to the measured signal amplitude generated in bulk GaAs at the same temperature. We therefore conclude that the generation of...
THz pulses from quantum wells is caused by the creation of polarized electron-hole pairs only.

The dependence of the generated peak amplitude of the THz radiation on the applied electric field is shown in the inset of Fig. 3. Initially, the amplitude rises linearly with field, but shows saturation for higher fields. There are two reasons for this: First, the barriers prevent the dipole moment from growing linearly with electric field. Second, the oscillator strength of the transition decreases as the electron and hole wave functions are pulled apart. We have modeled both effects by calculating the field-induced polarization \( P \sim |\langle \phi_1 | \phi_h \rangle|^2 - |\langle \phi_h | z | \phi_e \rangle - \langle \phi_e | z | \phi_h \rangle| \), with \( \langle \phi_e | \phi_h \rangle \) being the transition matrix element and the term within brackets being the field-induced dipole. The solid line in the inset of Fig. 3 shows the results of this calculation with the proportionality constant adjusted to give the best fit to the data. With a laser spot size of 1 mm and a pulse energy of 260 pJ, the estimated carrier density is \( 2.0 \times 10^9 \) cm\(^{-2} \) for each quantum well. As a function of laser intensity, the amplitude of the THz pulse shows a linear dependence, indicating that space charge fields from the polarized photoexcited carriers are much smaller than the externally applied field.

It is interesting to point out the relation between the generation of THz radiation from polarized electron-hole pairs and the QCSE. For moderate electric fields \( F \), the electron-hole dipole moment \( \mu \) is proportional to the field: \( \mu \sim F \). Hence, the Hamiltonian \( \mu \cdot F \) of a dipole in an electric field \( F \) leads to a quadratic energy shift of the exciton—this is the QCSE. It is the same field-induced dipole moment \( \mu \) that is also responsible for the THz radiation after optical excitation with an ultrashort laser pulse.

In conclusion, we have observed THz radiation from a GaAs/Al\( _{0.3} \)Ga\( _{0.7} \)As MQW sample at 77 K after optical excitation with an ultrashort laser pulse. Because of the AlGaAs barriers, transport current in the MQW is strongly suppressed and the only mechanism is the creation of field-polarized electron-hole pairs. However, the measured THz signals are comparable in strength to those generated from the surface of bulk GaAs. This proves the importance of the optical rectification mechanism at semiconductor surfaces based on the creation of polarized electron-hole pairs.

10. Because transport current is strongly inhibited by the Al\( _{0.3} \)Ga\( _{0.7} \)As barriers at 10 K, we can reach flatband and even apply moderately large forward bias (negative) fields.