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Magnetic-field suppression of THz charge oscillations in a double quantum well

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We have observed a suppression of the THz radiation emitted by charge oscillations photoexcited in the conduction band of GaAs/$Al_xGa_{1-x}As$ asymmetric double quantum wells in magnetic fields of order 1 T. Coherent charge oscillations are created by femtosecond excitation of a superposition of two states between which there is an electric dipole moment which oscillates in time as the wave packet evolves. The electric field radiated by this intraband polarization is coherently detected using an optically gated antenna. The nature of the suppression is qualitatively different for the two cases of magnetic field B parallel and perpendicular to the plane of the quantum wells. For B parallel the polarization dephasing rate increases with increasing field whilst the initial amplitude remains approximately constant. For B perpendicular the dephasing rate does not change significantly with increasing field but there is a reduction in the initial amplitude. The behavior in parallel field can be understood in a semiclassical picture in which electrons are deflected as they tunnel, thus destroying the phase coherence. The observation of amplitude suppression in a moderate perpendicular field may be partly associated with magnetic confinement but the effect is much larger than expected. The effect of magnetic field on the charge oscillation frequency is also discussed. [S0163-1829(98)52216-3]

Semiconductor heterostructures provide a versatile laboratory for the study of coherent wave packet dynamics. The asymmetric double quantum well, which provided the first observation of the spatial dynamics of a wave packet in a solid,[1] has been a particularly fruitful system to study. In this structure, the lowest energy states in the conduction bands of a narrow and wide quantum well can be tuned into resonance by an applied electric field in the absence of coupling. Coupling leads to hybridization of the two states and the levels then anticross at the ''resonant'' electric field where the energy gap has its minimum value, T. Carriers can be created in a coherent superposition of the two states by resonant excitation with a short laser pulse with spectral bandwidth greater than T. The time evolution of the wave packet involves an oscillatory motion of charge density between the two wells and consequently an oscillating electric dipole moment is established which has a maximum value at the resonant electric field where the electrons are delocalized over both wells. The holes remain largely localized in the wide well but the Coulomb interaction between electrons and holes is generally thought to play an important role in the dynamics. The time varying polarization has been observed as quantum beats in four-wave mixing[1] and time-resolved pump-probe transmission[2] and much more directly by the coherent detection of the emitted electromagnetic radiation.[3] The frequency of the dipole radiation is approximately given by $f=(T^2+\Delta^2)^{1/2}/h$, where Δ is the energy by which the uncoupled levels are detuned from resonance. The existence of the time-varying intraband polarization is a purely quantum mechanical effect relying on the phase coherence between the excited states. The lifetime of the coherent superposition state, equivalent to the polarization dephasing time, is typically a few picoseconds at low temperature. In this paper we describe experiments which extend the zero magnetic-field THz emission studies of Roskos *et al.*[3] to the case of moderate magnetic fields applied parallel and perpendicular to the quantum well plane.

The nominally undoped double quantum well structure used in our experiments is similar to that described in Ref. 3. It was grown by molecular-beam epitaxy and consists of ten pairs of GaAs wells with widths of 8.5 and 13 nm, separated by an $Al_{0.21}Ga_{0.79}As$ barrier of thickness 3 nm. The pairs of wells are spaced from each other by 20 nm barriers of the same composition and separated from the top surface and a 500-nm-thick n^+ GaAs back contact layer by 200 nm of $Al_{0.21}Ga_{0.79}As$. The substrate was semi-insulating GaAs. The alloy composition was checked by means of x-ray rocking curves. The critical layer thicknesses were verified by a comparison of the exciton energies obtained from low-temperature photoluminescence excitation (PLE) experiments with calculations of the energies using the methods described in Ref. 4. To vary Δ, an electric field could be applied to the wells by means of an 80-nm-thick, semitransparent indium tin oxide surface Schottky contact and an alloyed indium ohmic connection to the doped layer which was revealed by etching.

The coherent THz spectroscopy experiments used a mode-locked Ti:sapphire laser which produces 65 fs duration pulses at the sample with the aid of extra-cavity dispersion compensation. The output of the laser was split into a 300 mW pump beam and a 20 mW probe beam. The p-polarized pump beam was weakly focused to illuminate $\sim 1.5\ mm^2$ of the sample at an angle of 45° to the surface normal. The sample was mounted in the variable temperature

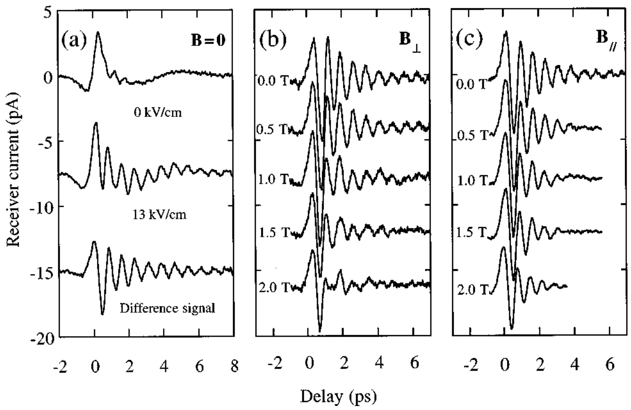

FIG. 1. Examples of THz emission spectra showing (a) the data subtraction procedure for background correction and (b), (c) the effect of perpendicular and parallel magnetic field on background corrected signals, respectively. The data has not been accurately corrected for propagation delay.

insert of a split-coil superconducting magnet in a helium gas atmosphere and maintained at 5 K. The time delayed probe beam was used to gate a 30 μm ion implanted silicon on sapphire photoconducting dipole antenna. This receiving antenna allowed the coherent detection of radiation emitted by the sample with a half-amplitude bandwidth extending from 0.1 to 2 THz.[5] The receiver current is proportional to the electric field $E(t)\propto d^2P/dt^2$ radiated into free space by the time-varying polarization, P, optically created in the sample. The signal-to-noise ratio in the experiment was improved by measuring the receiver current synchronously with 8 kHz modulation of the pump beam and by averaging with the aid of a repetitively scanned delay line. The THz beam emitted from the sample in the specular direction was coupled to the receiver using off-axis parabolic mirrors and a hyper-hemispherical silicon lens. The magnet has fused silica windows transverse to the bore and Mylar and high-resistivity silicon windows along the bore, thus allowing the introduction of visible radiation and the extraction of THz radiation with low loss and dispersion. Mirrors placed within the bore allow experiments with magnetic-field parallel, $B=(0,B_{\|},0)$, and perpendicular, $B=(0,0,B_{\perp})$, to the plane of the quantum wells. In both configurations the alignment accuracy was $\pm 2°$.

Typical THz emission results revealing the oscillatory polarization associated with the lowest two conduction subbands, and obtained with 804 nm wavelength, 10 nm bandwidth excitation are shown in Fig. 1. The traces in Figs. 1(b) and 1(c) show the difference in signal (receiver current) between zero electric field (flat band) and a field of ~ 14 kV/cm, close to the resonant field of 13 kV/cm,[6] as a function of delay for magnetic fields perpendicular and parallel to the quantum well planes. The reason for the data subtraction is that several largely bias-independent effects in the substrate otherwise complicate the THz signal from the quantum wells, as shown by the zero magnetic field results in Fig. 1(a). This figure shows traces at zero electric field, at the resonant electric field, and the difference signal. The flat band trace shows a single cycle-like transient near-zero delay together with a slow oscillation, both of which arise from the photoexcitation of carriers in the high field region of the substrate near the interface with the n^+ layer. The substrate signal contains contributions from the creation of an instantaneous polarization and the subsequent acceleration of carriers. In the presence of a magnetic field the time-varying polarization associated with cyclotron orbital motion of electrons photoexcited in the substrate can also be observed.[7] Immediately following the initial transient in Fig. 1(a) are a few weak oscillations with a frequency of 1.65 THz. This is approximately equal to that of the $n=1$ heavy-hole–light-hole (hh-lh) splitting in the PLE spectrum which leads us to believe that these oscillations originate in the beating of coherently excited hh and lh valence band states. An electric dipole moment connects the states for finite in-plane wave vector. The THz signal from such valence band oscillations does not contribute significantly to the much stronger signal that we measure near the resonant electric field, because the hh-lh oscillations increase only slightly in amplitude with increasing electric field.[8] This picture is confirmed by reflective electro-optic sampling measurements of the hh-lh oscillations at the same fluence and electric field as the THz emission experiments, which shows that their frequency is 0.50 ± 0.03 THz higher than the oscillations in Figs. 1(b) and 1(c), and that they have a very different electric field dependence. The relative weakness of the valence band oscillations compared with those in the conduction band is due to the strong localization of holes in the wide well.[3]

The traces of receiver current $I(t)$ versus delay, excluding the first cycle where there is a significant bias-dependent contribution to the emission from the instantaneous polarization created in the wells,[3] were fitted to the function

$$I(t)=I_0 e^{-t/\tau}\cos(2\pi f t). \qquad (1)$$

At the resonant electric field and in zero magnetic field the oscillation frequency f was 1.38 ± 0.02 THz and the intraband dephasing time τ was 2.5 ps. Assuming an average absorption coefficient of $5000\ \mathrm{cm}^{-1}$ over the spectral bandwidth of the excitation pulse, the excited carrier density (electrons+holes) within a pair of wells was approximately $1.2\times 10^{10}\ \mathrm{cm}^{-2}$. We found that τ was insensitive to carrier density in the range $8\times 10^{8}\ \mathrm{cm}^{-2}$ to $4\times 10^{10}\ \mathrm{cm}^{-2}$. The experiments in magnetic field were performed with the quantum wells slightly detuned (by 0.06 THz) from resonance at an electric field where the amplitude was a maximum but τ had a somewhat smaller zero magnetic field value of $\tau_0 = 1.75$ ps. We believe that the variation of dephasing time with bias may be due to sample inhomogeneities such as well and barrier thickness fluctuations.

The variation in THz amplitude, frequency, and dephasing time deduced from fitting Eq. (1) to the data obtained in the range 0–2 T [Figs. 1(b) and 1(c)] are shown in Figs. 2–4. Above 2 T the shape of the signal changes and fitting to a simple damped oscillator function becomes unrealistic for $B_{\|}$ whilst for $B_{\perp}$ the signal is too weak to fit reliably, so that we have only presented data at lower fields. The variation of emission frequency with electric field was checked at several values of magnetic field to verify that the resonant electric field did not vary significantly with magnetic field.

First of all we discuss the $B_{\perp}$ case. Between 0 and 2 T the most significant findings are that the initial oscillation amplitude, I_0 in Eq. (1), decreases by a factor of ~ 3 whilst τ remains approximately constant. Over the same field range

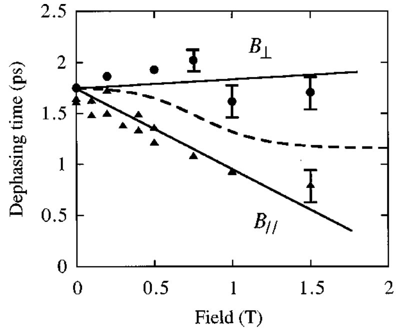

FIG. 2. Measured dephasing time of dipole oscillations [τ in Eq. (1)] as a function of perpendicular (circles) and parallel (triangles) magnetic field. Continuous lines are guides to the eye, dashed line is a theoretical prediction for parallel field as described in the text.

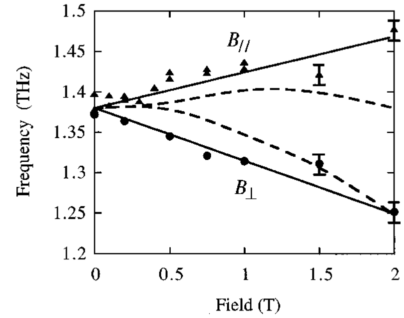

FIG. 4. Measured frequency of THz emission [f in Eq. (1)] as a function of perpendicular (circles) and parallel (triangles) magnetic field. Continuous lines are guides to the eye, dashed lines are theoretical predictions for parallel field (upper curve) and perpendicular field (lower curve).

the oscillation frequency decreases by 0.12 THz. In arriving at an understanding of the behavior of the oscillator dephasing time, it helps to realize that the important interactions are those which affect the transverse motion of the states in each well differently and are therefore a function of coordinates both parallel and perpendicular to the quantum well plane.[9] To first order this is not true of the interaction of free carriers or excitons with a perpendicular field which has no effect on the motion between wells in a collisionless approximation. In contrast, the interband exciton dephasing rate in quantum wells generally decreases with increasing $B_\perp$ for two reasons. Firstly, the increased binding energy leads to a smaller diameter and therefore a smaller scattering cross section,[10] and secondly the ''exchange'' interaction between electron and hole becomes a less important dephasing mechanism as the exciton spin degeneracy is removed.[11] There is no evidence of a significant decrease in dephasing rate in our experiments, emphasizing the difference between intraband and interband scattering processes.[12]

The amplitude of the THz oscillations depends principally on the product of matrix elements $\langle 1|3\rangle\langle 2|3\rangle\langle 1|z|2\rangle$, where $|1\rangle$, $|2\rangle$ are the conduction band wave functions, $|3\rangle$ is the wave function of the heavy hole in the wide well, and z is the coordinate perpendicular to the quantum well planes.[13] The first two matrix elements determine the interband optical excitation efficiency and are expected to exhibit a small increase with $B_\perp$ reflecting the increase in exciton binding energy with magnetic confinement.[14] The third is the intraband dipole moment which is expected to show a small decrease

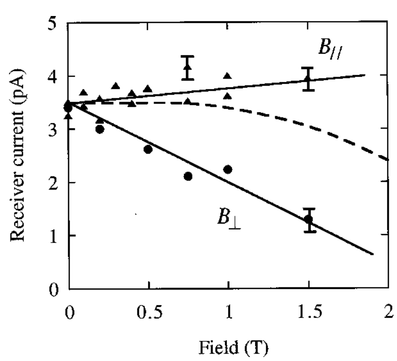

FIG. 3. Initial amplitude of THz emission as measured by receiver current [I_0 in Eq. (1)] as a function of perpendicular (circles) and parallel (triangles) magnetic field. Continuous lines are guides to the eye, dashed line is a theoretical prediction for parallel field.

with increasing $B_\perp$ as the increase in exciton binding energy leads to a reduction in the spatial extent of the exciton in all three dimensions. In a single particle model a reduction in amplitude is additionally expected when the cyclotron energy becomes comparable with the spectral bandwidth of the laser pulse and the generation of carriers able to participate in the coherent dynamics is reduced, but this only occurs at fields an order of magnitude higher than those in our experiments and can be neglected here. In order for magnetic confinement to be a significant effect the magnetic length $(\hbar/e\,B_\perp)^{1/2}$ needs to be small compared with the exciton diameter which requires $B_\perp > 1.5$ T. Calculations[14] show that the exciton binding energy only increases by $\sim 10\%$ between 0 and 2 T, which taken together with the opposing changes in the interband and intraband matrix elements, suggests that the initial amplitude should exhibit only a small variation of order a few percent. We cannot presently account for the observed factor of 3 change in initial amplitude.

In a collisionless approximation the oscillation frequency is independent of $B_\perp$ in a single particle model although scattering processes can modify the oscillation frequency[15] and these may depend on $B_\perp$ but we have no way to estimate this effect. We can, however, estimate the effect of electron-hole correlation by calculating the exciton diamagnetic shift, $\Delta E_\perp$, using variational techniques[4] with

$$\Delta E_\perp = e^2 B_\perp^2 \langle r^2\rangle / 8\mu, \tag{2}$$

where r is the in-plane electron-hole separation and μ is the reduced exciton mass. The shift of the $e1$-hh1 exciton (i.e., the exciton formed from $n=1$ electron and heavy hole states) is larger than that of $e2$-hh1. This leads to a decrease in THz emission frequency with increasing field which, as shown in Fig. 4, agrees with the measurements reasonably well.

In contrast to perpendicular magnetic field, a parallel field decreases the dephasing time by a factor of ~ 2 over the range 0 to 2 T, whilst the initial amplitude changes by at most 25%. The oscillation frequency increases by about 0.05 THz over the same range. In this field configuration the magnetic and spatial quantization are coupled which gives rise to an additional dephasing mechanism. In a semiclassical picture, $B_\parallel$ deflects the motion of the electrons as they tunnel and drives the coupled states out of resonance so that coherent phenomena are suppressed. This effect is also responsible for the quenching of the resistance resonance observed in coupled two-dimensional electron gases.[16] Starting from a three-level model of a single heavy-hole valence band and two conduction bands, Vasko and Raichev[17] have developed a theory for the charge oscillations which neglects Coulomb interactions and scattering. Raichev[18] has extended the model, which is based on solving the equation of motion of the conduction band density matrix in a momentum representation, to include the effects of a parallel magnetic field. We have approximately included the zero field dephasing by incorporating a phenomenological relaxation time $\tau_0 = 1.75$ ps. If the pump-pulse duration is considerably smaller than the oscillation period (which is fulfilled in our experiments), the oscillating part of the electron polarization perpendicular to the layers, $P_z(t)$, is given by

$$P_z(t) \propto \sum_{\underline{p}} \int_{-\infty}^{t} dt' \left(\frac{T}{\hbar \omega_p} \right)^2 G_z(\underline{p},t') e^{-t/\tau_0} \cos \omega_p (t-t'), \tag{3}$$

where G_Z is the electron generation function, and

$$\hbar \omega_p = \sqrt{T^2 + (\Delta - eB_\parallel s p_x/m)^2}. \tag{4}$$

The sum over $\underline{p}$ in Eq. (3) takes into account the momentum distribution of the electrons which are optically injected with a range of transverse momentum $\sim \sqrt{\mu \hbar/\tau_l}$ where τ_l is the duration of the laser pulse. The term proportional to $B_\parallel$ in Eq. (4) reflects the momentum change $\delta p_x = eB_{\parallel s}$ experienced by an electron tunneling the distance s between well centers. The generation function G_z is given by

$$G_z = w(t) \int_{-\infty}^{0} dt' w(t+t') \cos \varepsilon_p t', \tag{5}$$

where $\varepsilon_p = (p_x^2 + p_y^2)/(2\mu\hbar)$ and $w(t)$ describes the envelope of the optical excitation pulse. We have calculated the behavior of the radiated electric field $E(t) \propto \partial^2 P_z/\partial t^2$ for the parameters of our sample and assuming a sech^2 dependence for $w(t)$. Significant suppression of the dipole oscillations occurs when the energy shift $p_x \delta p_x/m$ is comparable with T which requires $B_\parallel \sim 1.5$ T. $E(t)$ is found to have a similar functional form to Eq. (1) for $B_\parallel < 2$ T and exhibits a dephasing time which decreases with increasing $B_\parallel$ and an amplitude only weakly dependent on $B_\parallel$ over the range 0–2 T, in qualitative agreement with experiment. The results of the calculation, scaled to agree with experiment at $B_\parallel = 0$, are shown in Figs. 2 and 3.

To understand the change in oscillation frequency with $B_\parallel$ we have again used a perturbative approach, this time with a cruder approximation for the exciton diamagnetic shift:[19]

$$\Delta E_\parallel = e^2 B_\parallel^2 \langle r \rangle \sqrt{\langle z^2 \rangle}/8\mu. \tag{6}$$

In addition to this shift there is an opposing contribution to the change in oscillation frequency from the modified dispersion curves in parallel field.[18] The calculations, taking both effects into account, are compared with experiment in Fig. 4, where reasonable qualitative agreement with experiment is obtained.

In summary, we have investigated the effect of moderate magnetic fields on the coherent charge dynamics of an asymmetric double quantum well. The behavior is qualitatively different for the two cases of magnetic field applied parallel and perpendicular to the plane of the quantum wells. For parallel field the intraband dephasing rate deduced from the time dependence of the emitted THz radiation increases with increasing field, whilst the initial amplitude shows little variation. This can be understood qualitatively in a semiclassical picture in which the electrons are deflected by the field as they tunnel, thus suppressing coherent oscillations. For perpendicular field the intraband dephasing rate remains approximately constant but the initial oscillation amplitude is reduced. The decrease in amplitude appears too large to be accounted for by magnetic confinement and is not presently understood. The magnetic-field variation of the THz emission frequency has also been measured and compared with exciton models which very approximately describe the behavior. In both magnetic-field geometries the net effect of moderate magnetic fields is a suppression of the coherent charge oscillations in the conduction band. We have recently observed a similar suppression of hh-lh quantum beats in wide quantum wells suggesting that this is a general phenomenon. Hopefully this work will stimulate further theoretical work on coherent intraband phenomena.

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[1] K. Leo *et al.*, Phys. Rev. Lett. **66**, 201 (1991).
[2] K. Leo *et al.*, Semicond. Sci. Technol. **7**, B394 (1992).
[3] H. G. Roskos *et al.*, Phys. Rev. Lett. **68**, 2216 (1992).
[4] S. R. Andrews *et al.*, Phys. Rev. B **37**, 8198 (1988).
[5] M. van Exter and D. Grischkowsky, IEEE Trans. Microwave Theory Tech. **38**, 1684 (1990).
[6] We observe an approximately parabolic dependence of emission frequency on electric field which allows the resonant electric field to be identified. This behavior is different from that reported in Ref. 3.
[7] D. Some and A. V. Nurmikko, Phys. Rev. B **50**, 5783 (1994).
[8] P. C. M. Planken *et al.*, Phys. Rev. Lett. **69**, 3800 (1992).
[9] S. Luryi, IEEE J. Quantum Electron. **27**, 54 (1990).
[10] T. Rappen *et al.*, Appl. Phys. Lett. **63**, 1222 (1993).
[11] R. E. Worsley *et al.*, Phys. Rev. Lett. **76**, 3224 (1996).
[12] P. Leisching *et al.*, Phys. Rev. B **51**, 18 015 (1995).
[13] S. C. Luo *et al.*, IEEE J. Quantum Electron. **30**, 1478 (1994).
[14] R. L. Greene and K. K. Bajaj, Phys. Rev. B **31**, 6498 (1985).
[15] F. T. Vasko and O. E. Raichev, JETP **81**, 1146 (1995).
[16] Y. Berk *et al.*, Phys. Rev. B **51**, 2604 (1995).
[17] F. T. Vasko and O. E. Raichev, Phys. Rev. B **51**, 16 965 (1995).
[18] O. E. Raichev, J. Appl. Phys. **80**, 924 (1996).
[19] M. Sugawara *et al.*, Phys. Rev. B **55**, 13 155 (1997).