Mechanism of THz Emission from Asymmetric Double Quantum Wells

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Impulsive interband optical excitation of the lowest two conduction subbands of a suitably engineered GaAs/AlGaAs double quantum well can lead to coherent THz emission. We demonstrate that, contrary to previous expectations, the dominant emission mechanism can involve the beating of either continuum or exciton states, depending on excitation conditions. The coherence of the continuum beats persists for several picoseconds even for excitation an optical phonon energy above the band edge. We attribute this to the small energy difference between the component eigenstates, which substantially reduces the number of relevant scattering events.

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The dynamics of coherently excited excitonic and continuum wave packets in semiconductors has been the subject of intense research over the last decade [1]. A model system for the study of both interband and intersubband optical coherence is the GaAs/AlGaAs asymmetric double quantum well (ADQW) [1–4]. In this system, it is possible to create a macroscopically coherent superposition of two different conduction subband states by femtosecond interband photoexcitation of sufficient bandwidth. The component eigenstates of a wave packet so created subsequently evolve at different rates leading to quantum beats and an oscillating electric dipole moment perpendicular to the plane of the quantum wells [2–4]. Such oscillations remain macroscopically coherent until the relative phase of the states is randomized by scattering or until the amplitude is suppressed by interference. The time evolution of the interband polarization, which involves wave packets formed from a superposition of electron-hole pair states, has been studied by four wave mixing [2,3]. The intersubband polarization, which mainly involves the coherence of electron states, has been indirectly studied by time resolved pump-prove transmission [2,3] and directly by detection of terahertz emission (TE) [4].

In the latter study the authors came to the conclusion that TE was associated with exciton beats. In later work [5] it was suggested that this was because the intersubband coherence of continuum electron states is short lived, as found for the interband case [6], and that TE would therefore be dominated by the supposedly longer lived coherence of exciton states. We use the term "continuum states" as an abbreviation for continuum edge exciton states in that there is no sense in which the Coulomb interaction can be switched off. The short interband coherence lifetime of such states is primarily due to the different dispersion of electron and hole bands, which leads to the creation of a macroscopic polarization comprised of an ensemble of oscillators with a frequency spread comparable with the laser bandwidth [7] and rapid loss of coherence by interference. In the intersubband case, the small dispersion of the gap in the conduction band leads to the creation of an ensemble of continuum oscillators with the same frequency in the absence of sample inhomogeneities. Decay of coherence then occurs by scattering, and both continuum and exciton states can thus separately contribute to the evolution of the macroscopic polarization on ps time scales. This picture is changed by the application of an in-plane magnetic field [8] and in the valence band [9], where significant dispersion of intersubband gaps is possible.

We report measurements made on a similar ADQW to those studied previously [2–4] in which wave packets made up of continuum states make an important contribution to the long lived part of the macroscopic polarization and tend to dominate the TE except when resonantly exciting the lowest exciton transitions. The observation of single particlelike behavior is significant in the light of recent experiment and theory on many particle interactions in superlattices which stress the importance of Coulomb correlations [10,11]. We also find that the relative phase of the wave packet component states is only weakly affected by scattering, as evidenced by the wide excitation energy and density ranges over which TE of several ps duration is observed. Recent studies of Bloch oscillations in superlattices [12–14] have in part revealed similar behavior, thus emphasizing the similarity of these two important model systems.

The active region of the ADQW sample [8] consists of ten pairs of undoped GaAs wells with widths of 8.5 and 13 nm, separated by a 3 nm thick $Al_{0.21}Ga_{0.79}As$ barrier. Each pair of wells is separated from its neighbors by 20 nm of $Al_{0.21}Ga_{0.79}As$. The energy gap between the lowest two single particle conduction band states $(e1, e2)$ can be tuned by an applied electric field and goes through a minimum, T, as the levels anticross at a "resonant" field F_0. The $1s$ exciton energies, as mapped out by photoluminescence excitation (PLE) spectroscopy, are shown in Fig. 1(a). The different binding energies of spatially direct

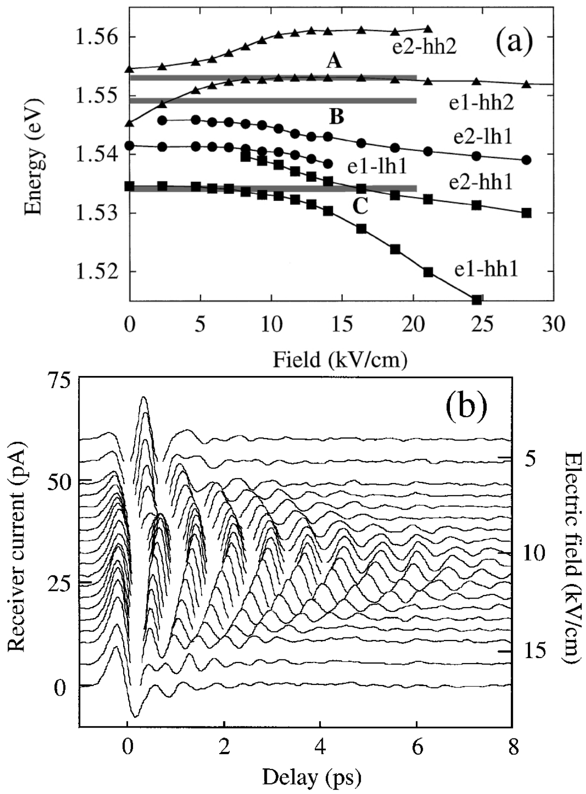

FIG. 1. (a) Measured exciton energies. Horizontal bars indicate center excitation energies for TE measurements. (b) TE signals for 1.549 eV excitation (bar B).

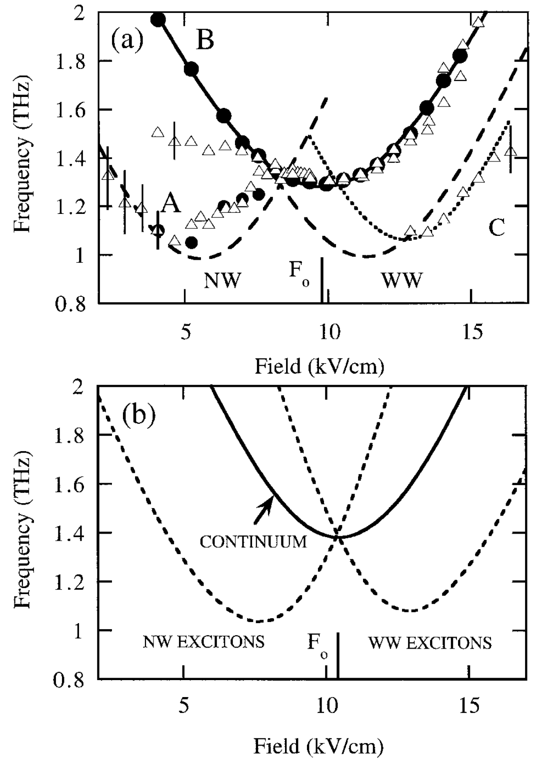

FIG. 2. (a) Differences between $e1$-$hh1$, $e2$-$hh1$ (WW) and $e1$-$hh2$, $e2$-$hh2$ (NW) exciton frequencies at excitation densities of 2×10^6 cm^{-2} (dashes) and 5×10^9 cm^{-2} (dots). Symbols show TE frequency at excitation energies of 1.553 eV (squares, A), 1.549 eV (circles, B), and 1.534 eV (triangles, C). Solid curve is fit to data B of the model discussed in the text. (b) Calculated $1s$ exciton (dashed curves) and continuum (solid curve) beat frequencies.

and indirect excitons lead to separate anticrossing fields of 12 and 5.5 kV/cm for the $e1$-$hh1$, $e2$-$hh1$ wide well (WW) and $e1$-$hh2$, $e2$-$hh2$ narrow well (NW) excitons, respectively. In our notation $hh1$, $hh2$ denote the lowest energy heavy hole states in the double well.

TE was excited by 8 meV bandwidth ($>T$), 160 fs laser pulses, and coupled to a 2 THz bandwidth photoconducting receiver. Traces showing the time dependence of the receiver current, proportional to the transient electric field radiated by the sample, under 1.549 eV excitation are shown in Fig. 1(b). Ringing arising from coherent charge oscillations in the two lowest conduction subbands [4,8] is seen following an initial transient arising from an instantaneous polarization adiabatically created in the wells [15]. Although weak hh-lh beats with a frequency of 1.65 THz are observable in TE at flat band, charge oscillations in the valence band are suppressed by strong localization of holes in the wider well at the fields of interest. Unless otherwise stated, TE measurements were made with the sample at 5 K and with a photoexcited electron-hole pair density $n_s = 4(1) \times 10^9$ cm^{-2} per double well per pulse, which places our experiments in the low density limit $n_s a_0^2 \sim 0.01$ where a_0 is a typical exciton radius.

Figure 2(a) shows the variation of TE frequency with field for three different excitation energies [A–C in Fig. 1(a)]. The behavior is different for excitation resonant and nonresonant with the lowest energy WW and NW heavy hole exciton anticrossings. The nonresonant behavior is typified by 1.549 eV excitation which lies in the continua of the $e1$-$hh1$, $e2$-$hh1$ WW excitons [Fig. 1(b); data B in Fig. 2(a)]. The dominant spectral component of the TE shows an approximately parabolic dependence of the frequency, f, on field, centered around a field $F_0 \sim 10$ kV/cm. The solid curve in Fig. 2(a) is a fit to the equation $f = \sqrt{\beta^2(F - F_0)^2 + T^2}$ appropriate for uncorrelated electrons and small detuning [2]. β is the constant of proportionality between the frequency separation of the uncoupled electron states in the WW and NW and the field F. The dephasing time has a maximum value of 3 ps near F_0, and is smaller at higher and lower applied field, primarily because of inhomogeneities in the internal electric field. Although continuum edges were not observed in the PLE spectra, the values of T and F_0 observed in TE are consistent with calculations [16] for the beating of continuum states. They are, however, inconsistent with expectations for $1s$ heavy hole [PLE data in Fig. 2(a) and calculations in Fig. 2(b)] or light-hole (not shown) excitons.

In contrast, TE traces obtained for excitation resonant with the lowest energy heavy hole excitons in the NW and

WW at their respective anticrossing fields [data A and C in Fig. 2(a)] exhibit frequency spectra dominated by excitons. This is particularly clear when comparing data A in Fig. 2(a) for resonant excitation of the NW at 1.553 eV with the PLE data (dashed curve in Fig. 2(a)). At this excitation energy no evidence of continuum beating is seen for fields below $\sim$7 kV/cm. Changing the excitation energy by $\pm$5 meV resulted in approximately equal spectral weights of exciton and continuum beats at the same electric field. A dephasing time of $\sim$0.6 ps is observed for NW exciton beating near 5.5 kV/cm. This is smaller than the 2 ps dephasing time for continuum beating at the same electric field obtained with nonresonant excitation, possibly because of dissipation associated with the degeneracy of exciton and continuum excitations (Fano resonance). For resonant excitation of the wide well at 1.534 eV [data C in Fig. 2(a)] spectral components at two frequencies consistent with simultaneous WW exciton and continuum state beating were observed. In this case the spectral weight of the continuum contribution at 12 kV/cm was about 0.3 of the exciton contribution, and the respective dephasing times had values of $\sim$2 ps and 3 ps. This greater similarity of dephasing times might be associated with the fact that the lower WW exciton does not exhibit Fano resonance.

Our results apparently differ from a previous TE study [4] where no distinct variation in frequency was observed, probably because of differences in sample structure, field homogeneity, and excitation conditions. In particular, the wider wells previously used lead to a calculated difference in emission frequencies for exciton and continuum beating at their respective resonance fields 3 times smaller than in our experiments. When combined with the larger excitation bandwidth of 14 meV and the excitation energy previously used, this would make separating out and identifying the different contributions much more difficult.

To investigate the effect of excitation density, we performed measurements at different pump powers. PLE spectra were measured for $n_s = 2 \times 10^6$ and 5×10^9 cm^{-2}. At the higher carrier density, which is similar to that used in the TE measurements, both F_0 and the energy separation between $e1$-$hh1$ and $e2$-$hh1$ excitons are only 5% larger than at the lower density [Fig. 2(a)], thus demonstrating that screening of both excitons and electric field are small effects. Figure 3 shows the effect of varying n_s on the frequency and dephasing time of the TE and confirms that the experiments are indeed performed in the low density limit suggested by the PLE measurements. The TE frequency increases by $\sim$5% as n_s is increased from 10^8 to 10^{11} cm^{-2}, but decreases above 10^{11} cm^{-2}. Account was taken of the small reduction in internal electric field due to screening by adjusting the applied bias for minimum TE frequency at each excitation density. The initial increase with n_s can be qualitatively explained by Coulomb renormalization of the tunnel gap [17] and the decrease at large n_s by dissipation [3,18]. The reduction in dephasing time above 10^{10} cm^{-2} is consistent with estimated electron-electron scattering and radiation damping rates [19]. The charge oscillations in the ADQW are much less affected by carrier-carrier scattering than in the superlattice case where a dramatic decrease in dephasing time is observed [13] on increasing the carrier density from 2×10^9 cm^{-2} to 2×10^{10} cm^{-2}. Although screening plays a less important role in 2D compared with 3D [20], suggesting a reduction in scattering in the superlattice case, we believe that the scattering events most effective in destroying intersubband coherence are those perpendicular to the quantum well planes which affect the wave packet eigenstates differently. These are greatly restricted by momentum conservation in the ADQW compared with the superlattice.

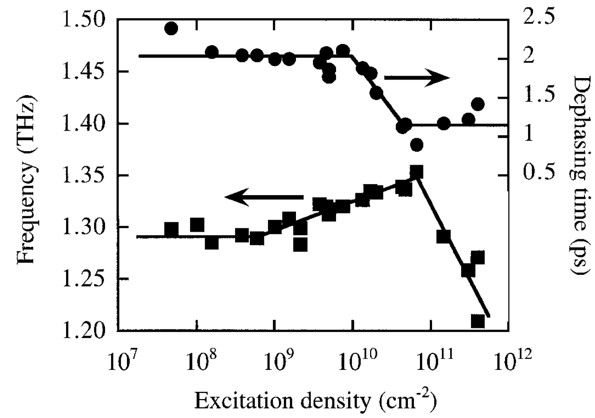

FIG. 3. Variation of dephasing time (circles) and emission frequency (squares) with excitation density for excitation at 1.549 eV.

Further evidence for the involvement of continuum states comes from the behavior of the TE as a function of excitation energy; see Fig. 4. At 10 kV/cm, the initial TE amplitude is not strongly resonant with the lowest energy exciton transitions, but instead peaks approximately

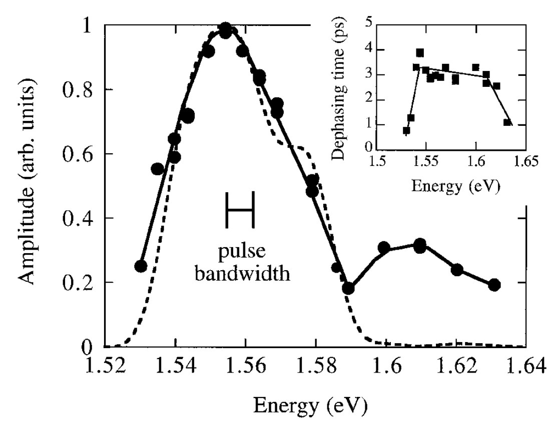

FIG. 4. Dependence of initial amplitude of TE on excitation energy at 10 kV/cm. Dotted curve shows calculation for excitation of zone center $e1$, $e2$ continuum states as discussed in the text. The inset shows variation of dephasing time.

20 meV above the lowest subband edge and is still measurable 100 meV above. Data at fields of 5.5 kV/cm and 12 kV/cm show ~8 meV wide peaks in the amplitude of exciton beating centered around 1.553 and 1.534 eV, respectively, as well as a similar persistence of continuum TE to large excess energies. The dephasing time of the continuum TE at 10 kV/cm is approximately independent of excitation energy over a range of almost 100 meV (Fig. 4, inset). The emission frequency and resonant field also change little with excitation energy between 1.53 and 1.63 eV ($\Delta f < 0.06$ THz, $\Delta F_0 < 0.5$ kV/cm). This is consistent with the excitation of higher lying $e1$, $e2$-hhn and $e1$, $e2$-lhm continuum states (n, m denote subband indices), but inconsistent with the dominant involvement of excitons when the frequencies and resonant fields should vary with excitation energy. The variation of TE amplitude with excitation energy, calculated by summing over the product of interband and intersubband matrix elements for continuum states and taking into account the laser pulse spectrum, is also shown in Fig. 4. The agreement with experiment is good below 1.59 eV, but because of the negligible size of the interband matrix elements for $n > 3$, $m > 2$, the observation of TE at higher excitation energies requires the involvement of sizable in-plane wave vector $e1$, $e2$ continuum states and possibly closer to band edge $e3$ states which subsequently relax to $e1$, $e2$. Carriers with sufficiently large excess energies can lose energy by longitudinal optical phonon emission in a few hundred fs [21]. The onset of this emission is suggested by a phase shift in the TE [22] for excitation near 1.59 eV. Another energy relaxation mechanism which proceeds on 100 fs time scales is intraband carrier thermalization via electron-electron scattering [23]. Carriers relaxing by either process will only contribute to long lived TE if the relative phase of the component eigenstates is insensitive to many body interactions. We believe that the small scattering rate of continuum wave packets in the ADQW is a property of the close energy level spacing of the eigenstates. Physically, the in-plane variation of eigenstates is sufficiently similar that they are almost equally affected by in-plane scattering events, thus preserving the relative phase [24]. Behavior similar in some ways has previously been observed in heavy-light hole quantum beating [7,25], Bloch oscillations in superlattices [14,22], and the long lived coherence of optically excited cyclotron emission [26].

In conclusion, we find that the field and excitation energy dependence of the TE from an ADQW is consistent with the beating of continuum exciton states except under conditions where excitons are resonantly created and that the intersubband coherence is relatively insensitive to scattering. Recent quantum kinetic theories predict that the electron-hole interaction should play an important role in determining the dynamics of both the interband and intersubband polarizations [10,11]. Our results do not contradict such theories in which the TE frequency is given by a difference of frequencies in the linear absorption spectrum because this includes both exciton and continuum states, but more theoretical work is needed to understand the relative strengths of the different contributions. Factors to be considered include the small exciton linewidths relative to the spectrally broad excitation, the larger continuum state intersubband dipole moment, and dissipation associated with coupling of excitons to continuum excitations.

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