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Asymmetry in the excitonic Wannier-Stark ladder: A mechanism for the stimulated emission of terahertz radiation

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Using an excitonic basis, we calculate the coherent intraband response of a photoexcited undoped semiconductor superlattice in combined along-axis static and terahertz electric fields. We find that the terahertz field drives the amplitude of the excitonic Bloch oscillations, yielding an intraband polarization that continues to oscillate at times much longer than the intraband dephasing time—an effect which is totally absent for noninteracting electron-hole pairs. Furthermore, these driven, Bloch-oscillating excitons are found to produce stimulated emission at terahertz frequencies as long as the Bloch oscillation amplitude is driven.

The concept of Bloch oscillations (BO) was first introduced by Bloch over 70 years ago.[1] The basic idea is that, if an electron in a periodic potential is subjected to a static electric field, the electronic wavepacket will oscillate spatially at the so-called BO frequency, $\omega_B \equiv edE_{dc}/\hbar$, where e is the magnitude of the charge on an electron, E_{dc} is the magnitude of the static electric field, and d is the period of the potential. These oscillations are simply the quantum beating of the Wannier Stark ladder (WSL) eigenstates.[2] Because the WSL state energies are equally spaced with energies $E_p^o = E_0^o + p\hbar\omega_B$ (where p is an integer), only harmonics of the Bloch frequency enter into the temporal evolution of the wavepacket centroid.

There has been considerable work on Bloch-oscillating excitons in a static field,[3–9] on excitons in a THz field,[10] and even on the optical response of a superlattice in combined static and THz fields.[11,12] There are also a number of papers treating the motion of electrons in doped superlattices in combined static and THz fields.[13] The ultimate goal of much of this work is to understand how to prolong the oscillations, and perhaps even create a THz laser. One might imagine that the electrons in the WSL could be stimulated by the THz field to emit many photons as they are driven down the ladder by the THz field. A similar effect, with the inclusion of scattering is, in fact, the basis of the quantum cascade laser.[14] Yet a serious flaw in this scenario was pointed out a number of years ago by Bastard and Ferreira.[15] They noted that photon absorption and emission are equally probable from an electron in a WSL state; hence, in a single-particle model, there will be no net stimulated emission or absorption of THz radiation. Although THz radiation emitted by Bloch-oscillating excitons has been experimentally observed,[4] this emission only occurs for times on the order of the intraband dephasing time, when the *initial* intraband polarization created by the optical pulse is still ringing. Thus far there has been no experimental evidence of stimulated emission, which appears to be consistent with the above theoretical prediction.

In this paper we present the results of the first calculation of the coherent temporal response of the *intraband* polarization of an undoped semiconductor superlattice in *combined static and THz along-axis electric fields* when excited via an ultrashort optical pulse. We find that the inclusion of the electron-hole Coulomb interaction breaks the emission/absorption symmetry in the WSL, with the remarkable effect that *the excitonic population can be stimulated to coherently emit photons for times much longer than the intraband dephasing time,* an effect that is totally absent when the electron-hole (e-h) Coulomb interaction is neglected.

We employ the quasi-Bosonic treatment of Hawton and Nelson.[16] This formalism is very similar to the dynamically controlled truncation (DCT) formalism of Axt and Stahl,[17] and thus contains the exciton-exciton correlations that have been shown to be essential in the calculation of P_{intra}.[6,18] We work within the envelope function approximation, and neglect band nonparabolicities and valence-band mixing. Furthermore, we limit ourselves to the inclusion of only the first superlattice minibands from both the conduction and valence bands. We write the Hamiltonian in terms of *excitonic* creation and annihilation operators, $B_\nu^\dagger$ and B_ν which create and destroy excitons in the νth state, respectively.[16] The excitonic states are the e-h eigenstates of the system in the presence of the superlattice potential, the e-h Coulomb interaction, *and the static electric field* (E_{dc}). The envelope functions of these states are denoted by $\psi^\nu(z_e, z_h, \rho)$, where z_e (z_h) is the position of the electron (hole) along the superlattice growth direction and ρ is the e-h separation in the transverse planes.

The Hamiltonian takes the form $H = H_0 + H_I + H_C$, where H_0 is the single-exciton Hamiltonian for superlattice excitons in the presence of a static electric field, H_I contains the interaction of the excitons with the external optical and THz fields, and H_C contains the exciton-exciton Coulomb interactions. The single-exciton Hamiltonian is identical to that employed in an earlier work;[19] in second quantized form, it is given by $H_0 = \Sigma_\nu E_\nu B_\nu^\dagger B_\nu$, where E_ν are the energies of the WSL excitons.

The interaction Hamiltonian takes the form $H_I = -V\mathbf{E}_{ac}\cdot\mathbf{P}$, where V is the volume of the superlattice, $\mathbf{E}_{ac} = \mathbf{E}_{opt} + \mathbf{E}_{THz}$ is the sum of the applied optical plus THz fields, and $\mathbf{P}$ is the total polarization *operator*,

$$\mathbf{P}=\frac{1}{V}\sum_{\nu}\,[\mathbf{M}_\nu B_\nu^\dagger+\mathbf{M}_\nu^* B_\nu]+\frac{1}{V}\sum_{\nu\mu}\,\mathbf{G}_{\nu\mu}B_\nu^\dagger B_\mu, \qquad (1)$$

where the first term is the interband polarization and the second term is the intraband polarization. The interband dipole matrix element of the νth excitonic state is $\mathbf{M}_\nu \equiv \mathbf{M}_o\sqrt{AN_z}\int dz\,\psi^{\nu *}(z,z,0)$, where $\mathbf{M}_o$ is the bulk interband dipole matrix element, A is the transverse area, and N_z is the number of superlattice sites. The intraband dipole matrix element is given by $\mathbf{G}_{\nu\mu}\equiv\langle\psi^\nu|-e(\mathbf{r}_e-\mathbf{r}_h)|\psi^\mu\rangle$. Finally, the Coulomb Hamiltonian takes the form $H_C=\Sigma_{\{\nu_i\}}V^{\nu_1,\nu_2}_{\nu_3,\nu_4}B^\dagger_{\nu_4}B^\dagger_{\nu_3}B_{\nu_2}B_{\nu_1}$, where $V^{\nu_1,\nu_2}_{\nu_3,\nu_4}$ describes the interaction of two excitons initially in states with quantum numbers ν_1 and ν_2, which are scattered into states ν_3 and ν_4 via the Coulomb interaction.[20]

Using this Hamiltonian we derive the equations of motion for the interband and intraband correlation functions. We note that only excitonic states with zero center-of-mass momentum are included in the calculation, as these are the only ones which couple to the optical field. The intraband polarization is the result of the relative motion of the electrons and holes. As has been discussed earlier,[16,17] a DCT approach such as is employed here gives results which are exact to the appropriate order of the optical field. To first order in the optical field, the dynamical equation of motion for $\langle B_\nu^\dagger\rangle$ (the interband correlation function) is

$$i\hbar\frac{d\langle B_\nu^\dagger\rangle}{dt}=\left[E_\nu-\frac{i\hbar}{T_{2inter}}\right]\langle B_\nu^\dagger\rangle+\mathbf{E}_{opt}\cdot\mathbf{M}_\nu +\mathbf{E}_{THz}\cdot\sum_\beta\,\mathbf{G}_{\nu\beta}\langle B_\beta^\dagger\rangle, \qquad (2)$$

where interband dephasing has been included phenomenologically through the interband dephasing time constant, T_{2inter}. The dynamical equation of motion for $\langle B_\mu^\dagger B_\nu\rangle$ (the intraband correlation function) to second order in the optical field is

$$i\hbar\frac{d\langle B_\mu^\dagger B_\nu\rangle}{dt}=-\left[E_\nu-E_\mu+\frac{i\hbar}{T_{\mu\nu}}\right]\langle B_\mu^\dagger B_\nu\rangle+\mathbf{E}_{opt}\cdot[\mathbf{M}_\mu^*\langle B_\nu\rangle -\mathbf{M}_\nu\langle B_\mu^\dagger\rangle]+\mathbf{E}_{THz}\sum_\beta\,[\mathbf{G}^*_{\mu\beta}\langle B_\beta^\dagger B_\nu\rangle -\mathbf{G}_{\nu\beta}\langle B_\mu^\dagger B_\beta\rangle]. \qquad (3)$$

In this expression, we define the phenomenological time constants $T_{\mu\nu}$ such that if $\mu=\nu$ then $T_{\mu\nu}=T_1$ (excitonic population decay time), and if $\mu\neq\nu$, then $T_{\mu\nu}=T_{2intra}$ (intraband dephasing time). This separation of the phenomenological dephasing and population decay constants is only possible because we employ an excitonic basis; such a simple separation is not possible if one uses the more common noninteracting e-h basis.[6] We note that to second order in the optical field, the exciton-exciton interaction does not appear in the dynamic equations.

The exciton WSL energies and envelope functions are obtained using the approach of Dignam and Sipe.[19] We only consider here the $1s$ excitons,[21] and thus expand the eigenstates in terms of two-well $1s$ exciton states, $\Phi_m^{1s}(z_e,z_h,\rho)$, wherein the electron and hole are localized in wells m sites apart. In this basis the wave functions of the excitonic states with zero center-of-mass motion are given by

$$\psi^\mu(z_e,z_h,\rho)=\frac{1}{\sqrt{N_z}}\sum_{n,m}\,C_m^\mu\Phi_m^{1s}(z_e-nd,z_h-nd,\rho),$$

where the C_m^μ are calculated by solving the eigenvalue problem in the two-well basis.[19]

Once Eqs. (2) and (3) are solved, P_{intra}, to second order in the optical field, is given simply by the z component of the expectation value of the intraband portion of Eq. (1). We perform the calculation using two different models: the excitonic model (EX) employing $1s$ excitonic states, and the noninteracting (NI) model, in which the e-h Coulomb interaction is effectively removed by increasing the dielectric constant by more than a factor of 1000.

The system we are modeling is a GaAs/$Ga_{0.7}Al_{0.3}As$ superlattice with 67 Å wells and 17 Å barriers—a structure that has been studied in a number of recent experimental and theoretical papers on excitonic BO.[7,8,11] The calculations are all performed with a static electric field of $E_{dc}=15$ kV/cm. The THz field takes the form $E_{\mathrm{THZ}}=F\sin(\omega_T t+\phi)$, where the time origin is at optical pulse center. Unless otherwise stated, we shall take $\omega_T=\omega_B$, i.e., the THz field is resonant with the NI transitions. We set $T_1=\infty$ (Ref. 22) and take the sample to be optically excited by a 100 fs Gaussian pulse centered on the state with an intraband dipole moment which is closest to $-ed$ (the WSL $p=-1$ state). We employ an interband dephasing time of $10/\omega_B$ (0.52 ps) and intraband dephasing time of $20/\omega_B$ (1.04 ps) (Ref. 23)

We present in Fig. 1 the temporal evolution of the intraband polarization, for both the NI and EX models. To simplify the discussion, we break P_{intra} into two components: $P_{intra}=P_{THz}+P_{SV}$, where P_{THz} is the component oscillat-

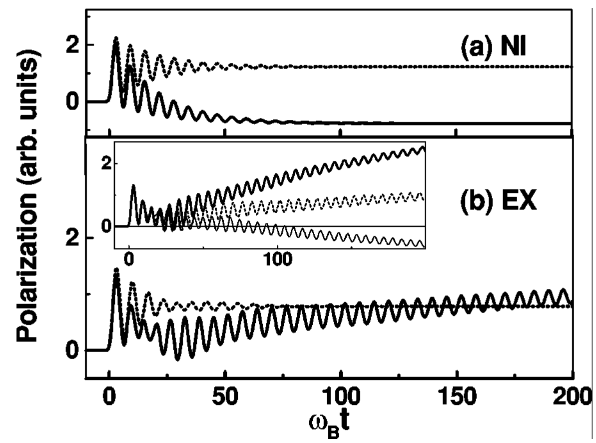

FIG. 1. The intraband polarization as a function of time for the NI (a) and EX (b) models for an interband dephasing time of $T_{2inter}=10/\omega_B$, an intraband dephasing time of $T_{2intra}=20/\omega_B$, a THz phase of $\phi=0$, THz frequency of $\omega_T=\omega_B$, and no population decay. The dotted lines give the results for no THz field while the solid lines are the results for a THz field of $F=2.5$ kV/cm. The inset shows the EX results under that same conditions as in 2(b) but with THz field frequencies of $\hbar\omega_T=E_0-E_{-1}$ (bold), $\omega_T=\omega_B$ (dash), and $\hbar\omega_T=E_{+1}-E_0$ (thin).

ing near the Bloch frequency, while P_{SV} is a slowly varying component. Superscripts NI and EX will be used to distinguish between results from the two models. We consider first, in Fig. 1(a), the NI results. When $F=0$ (dotted line), we see that P_{THz}^{NI} contains the normal BO motion damped over a time T_{2intra}, while P_{SV}^{NI} adds an approximately dc offset to P_{intra}^{NI}. This offset is due to the permanent dipole moment of the WSL states when $p\neq 0$. When $F=2.5$ kV/cm (solid line), P_{THz}^{NI} is essentially unchanged, indicating that the THz field does not drive the oscillation amplitude. However, P_{SV}^{NI} increases with time at a rate proportional to the amplitude of P_{THz}^{NI}. This results in a driven quasi-dc component to the current for times less than the intraband dephasing time. The current arises from the spatial separation of the electron and hole in a given pair due to the THz field. For the example shown, the electron is being driven up the ladder in the positive z direction relative to the hole. This quasi-dc current is essentially due to photon assisted hopping, as has been noted before.[13] It can be understood in a simple semiclassical model as arising from the work the THz field does on the electrons and holes, with the phase of the THz field relative to the BO phase determining the sign of the work done. However, in contrast with a driven harmonic oscillation, this energy does not go into increasing the amplitude of the BO, rather it goes into moving the electron up or down in the potential of the static electric field. The reason that the amplitude of the BO is not driven (and hence there is no long-lasting dc current) lies in the equal energy spacings and in the equal intraband dipole matrix elements for up and down inter-WSL transitions, i.e., $G_{\nu,\nu+n}=G_{\nu,\nu-n}$. Combined, these two factors lead to equal probability for up as for down transitions.[15]

In Fig. 1(b) we present the EX results under the same conditions employed in Fig. 1(a). In the absence of a THz field the polarization undergoes oscillations similar to those found in the NI case, but one can just detect frequency beating effects due to the unequal spacing of the excitonic WSL energy levels.[2] The really surprising results occur when the THz field is applied. For times $t<T_{2intra}$ the polarization is driven down, as it was in the NI model. However, in contrast to the NI results, P_{THz}^{EX} does not die out when $t>T_{2intra}$: *the terahertz field is driving the amplitude of the excitonic Bloch oscillations.* In addition, for $t>T_{2intra}$, the relative phase of the BO and the THz field becomes essentially locked, with the BO occurring at ω_B despite the fact that the excitonic Bloch frequencies are not precisely ω_B. Because of this fixed relative phase, the work done on the system by the THz field is always of the same sign, and the rate of change in P_{SV}^{EX} with time is essentially proportional to the instantaneous amplitude of the BO.

The differences between the NI and EX results arise from the breaking of the up/down symmetry in the WSL via the *e-h* Coulomb interaction. This interaction strongly affects the intraband dipole matrix elements and the energy spacing, as can be seen in the inset to Fig. 2. The matrix elements favor downward transitions ($G_{\nu,\nu-1}>G_{\nu,\nu+1}$) from the $\nu=-1,0,+1$ states, and upward transitions from all other states. We find that unequal energy spacing is the most important factor in producing the driven BO, but that the asym-

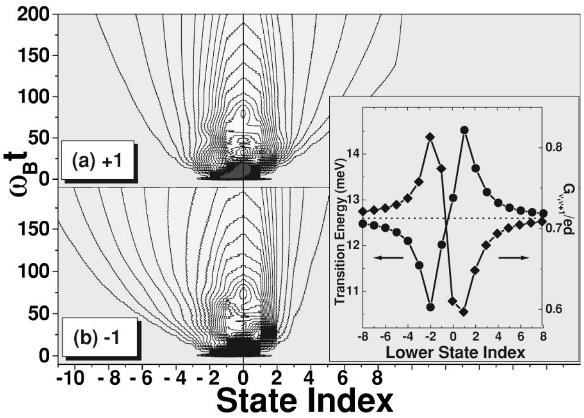

FIG. 2. Contour plots of the excitonic populations in each of the excitonic WSL states as a function of time for THz field frequency of (a) $\hbar\omega_T=E_0-E_{+1}$ and (b) $\hbar\omega_T=E_{-1}-E_0$. All other parameters are identical to those used in 1(b). The inset shows the excitonic energy level spacing $E_{\nu+1}-E_\nu$ (diamonds) and the intraband dipole matrix elements $G_{\nu,\nu+1}$, (circles) as a function of the state index, ν.

metry in both the matrix elements and the energy spacing combine to determine the direction in which the polarization is driven.

To further investigate this behavior, in the inset to Fig. 1(b) we plot P_{intra}^{EX} for three different frequencies of the THz field: $\hbar\omega_T=E_0-E_{-1}$, $\omega_T=\omega_B$, and $\hbar\omega_T=E_{+1}-E_0$. As can be seen, for $t\gg T_{2intra}$, the rate of change of P_{SV}^{EX} is strongly dependent on the THz frequency. Because $G_{0,+1}\simeq 0.79G_{0,-1}$, the $0\rightarrow+1$ transition acts as an effective cap on the transitions unless the THz field is tuned to this frequency. We find that the polarization increases whenever the THz frequency is appreciably below this frequency and the laser is centered between the $\nu=-2$ and $\nu=+2$ states.

In Fig. 2 we present a contour plot of the populations in the different excitonic WSL states as a function of time for the excitation laser centered on the $\nu=-1$ state. For a THz field resonant on the $0\rightarrow-1$ transition [Fig. 2(b)] the populations are preferentially driven down the ladder. This is seen in the large asymmetry in the population density about the $\nu=-1$ state on which the laser was centered. The effective blocking of the $0\rightarrow+1$ transition allows a population inversion to be set up for all of the states with $\nu<0$. In addition, the *populations are driven a long way down the ladder*: at times of about $200/\omega_B$, the population of the $\nu=-10$ state is at least 20% of the population in the $\nu=0$ or $\nu=-1$ states. In Fig. 2(a) we present the same data when the THz field is resonant on the $0\rightarrow+1$ transition. In this case, the population distribution remains approximately symmetric at all times, which is consistent with the small change of P_{SV}^{EX} found in this case in the inset to Fig. 1(b). The THz polarization decays to zero only after the electron and hole are pulled far enough apart by the THz field such that the Coulomb interaction between them becomes negligible. Under these conditions, the energy spacings and dipole matrix elements between states are essentially equal and we obtain the NI result of no driving of the polarization. The intraband polarization will also decay due to population relaxation which has not been included in this calculation.[22] The monotonic increase in P_{SV}^{EX}, shown in Fig. 1(b), is the direct signature of gain in the THz field; this can be verified by a simple calculation of $d/dtP_{intra}\cdot\mathbf{E}_{THz}$. As an example, for an exciton density of 5 x 10^9 cm^{-2} per period, gain coefficient exceeding 10 mm^{-1} are achievable for times greater than 25 ps. Furthermore, we find that there is a wide range of central laser frequencies for which P_{SV}^{EX} always *increases* with time, independent of the phase of the THz field. The only restriction on the applied static field for the appearance of gain is that the excitonic binding energy be less than, but not too much less than, the WSL energy spacing.

We have demonstrated that a THz field coherently drives both the Bloch-oscillating and slowly varying components of the excitonic intraband polarization for times much longer than the intraband dephasing time. Our results show that these effects are totally absent from a model that does not include the e-h Coulomb interaction. The important implication of these results is that there should be gain in the THz field in the excitonic system as long as there is a non-negligible excitonic population near the $\nu=-1$ state. Future directions include adding excited in-plane states in the excitonic basis, and introducing exciton-phonon interactions such that a more complete picture of the photocurrent and population scattering and decay can be obtained.

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[20] $V_{\nu_1,\nu_2}^{\nu_3,\nu_4}$ is related to $W_{n_3,n_4,0}^{n_1,n_2,0}$ given in Eq. (21) of Ref. 16.

[21] It has been shown (Ref. 2) that the effect of the unbound e-h pairs on the BO dipole is very small when the laser is centered on or below the $\nu=-1$ excitonic transition.

[22] The population decay time T_1 models across-gap recombination and scattering out of the 1s states. It is generally expected to be considerably larger than the dephasing times and so we set $T_1=\infty$. If T_1 is finite, then the intraband polarization will decay over a time on the order of $T_1 \gg T_{2intra}$.

[23] We have chosen the values of T_{2inter} and T_{2intra} to be in approximate agreement with the experimental results of Ref. 18. The precise value of T_{2inter}, however, has almost no effect on the calculated intraband polarization.