Generation of Ultrashort Electrical Pulses through Screening by Virtual Populations in Biased Quantum Wells

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We consider theoretically a semiconductor quantum well subject to an electrostatic field normal to the layer and a strong monochromatic laser field in the transparency region below the absorption edge. We show that ultrashort optical pulses generate ultrashort voltage pulses because the virtual electron-hole population induced by the laser field partially screens the applied static field. We predict that voltages >1 V could be generated in micron-thick multiple–quantum-well structures by use of low-power short-pulse lasers.

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In this Letter we show that photoexcitation of semiconductor quantum wells (QW's) well below the fundamental absorption edge generates coherently driven virtual populations of electron-hole (*e-h*) pairs that screen electrical fields applied to the QW. The screening generates transient electrical pulses and produces significant modification of the optical absorption as well as transient nonlinear optical effects. Because *no real population* of carriers is present in the QW, these processes last only as long as the optical field is applied. In the limit of dilute virtual populations, we derive universal results for the change in the static dielectric constant. We also present numerical results for GaAs QW's, demonstrating that these new effects should be readily observable and large enough to have important applications in optoelectronics.

It is now well understood that intense optical excitation of QW's below the optical absorption edge creates substantial virtual populations with measurable consequences. Recent experimental investigations revealed an ultrafast dynamical blue shift and a bleaching of the exciton resonances in QW's strongly photoexcited well below the absorption edge.[1,2] This "ac Stark effect" has been successfully explained as being due to anharmonic interactions of virtually created e and h among themselves and with the light field.[3] It was shown[3] that virtual *e-h* pairs coherently driven by an off-resonance laser field E_ω produce exactly the same physical effects as real ones, although they do not participate in any relaxation process. In particular, in order to describe the interaction of the virtual populations with a weak probe beam correctly, screening of the latter by *virtual* e and h had to be explicitly included in the "molecular-field" interaction Hamiltonian.[4,5] However, in the situation investigated in Refs. 1, 2, and 3 no static dipole is created.

Electroabsorption in QW's has also been extensively investigated.[6] In QW's biased by an electrostatic field normal to the plane of the layer, the e and h wave functions are strongly distorted. The positive and negative charges are pushed against opposite walls of the QW as illustrated in the inset of Fig. 1. This gives rise to large red shifts of the exciton resonances, an effect known as the quantum confined Stark effect (QCSE),[6] the field ionization of the excitons being inhibited by the potential barriers. Substantial charge separation can be obtained while keeping the tunneling time out of the QW significantly long, and hence retaining an abrupt absorp-

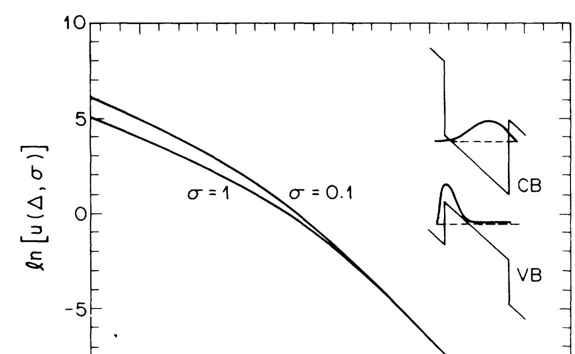

FIG. 1. Plot of the log of the universal function $U(\Delta,\sigma)$, which determines the change of dielectric constant, vs the log of the normalized detuning $\Delta=(E_g+W-\omega)/W$. Two curves are shown for electron-hole mass ratio $\sigma=m_e/m_h=0.1$ and 1. Inset: The electron and hole wave functions schematically, in the direction normal to the layer for a biased quantum well. (CB, conduction band; VB, valence band.)

tion edge even at high fields.[6]

The case where a QW biased along its normal (z) is strongly photoexcited in the transparency region below the gap corresponds to a combination of the situations described in the previous two paragraphs. The virtual electrons are then spatially separated from the virtual holes in the z direction, and thus the optical field induces a macroscopic static polarization $P_0 \simeq -N_1 e \int z[\phi_{e1}^2(z) - \phi_{h1}^2(z)]dz$, where $\phi_{e(h)1}$ is the e (h) ground-state wave function and N_1 is the corresponding virtual-pair density.[3] This polarization induces an electrostatic field $\delta F = 4\pi P_0/\epsilon_0$ that tends to screen the external field F and lasts only as long as E_ω (ϵ_0 is the static dielectric constant). This field is *real* and will produce the same effects as any other applied field. In particular, ultrashort optical pulses will generate ultrashort voltage pulses. This transient electrostatic field, by opposing the applied field, will "undo" part of the QCSE and thus produce an apparent blue shift of the exciton resonances that is additional to the ac Stark shift, and this is also associated with additional optical nonlinearities. In the following, we will derive from first principles a theory of these effects.

For the sake of clarity, we take the simplest model, assuming infinite barriers and neglecting excitonic and other Coulomb effects. Our final results can be easily generalized, and some of these effects can be included semiempirically. The QCSE reduces then to the quantum confined Franz-Keldysh effects (QCFKE),[7] and the basic Hamiltonian in the rotating frame is

$$H = \sum_{n,k,s} (\epsilon_{enk} c_{nks}^\dagger c_{nks} + \epsilon_{hnk} d_{nks}^\dagger d_{nks}) + P_{\text{intra}} F - (P_{\text{inter}} E_\omega^* + \text{H.c.})/2, \quad (1)$$

where

$$P_{\text{intra}} = -e \sum_{n,m,k,s} \langle n|z|m\rangle (c_{nks}^\dagger c_{mks} - d_{nks}^\dagger d_{mks}),$$

and

$$P_{\text{inter}} = -e \sum_{n,k,s} r_{vc} d_{nks} c_{nks}$$

are respectively the (dc) intraband and (ac) interband polarization operators. $c_{nks}^\dagger$ (c_{nks}) and $d_{nks}^\dagger$ (d_{nks}) are creation (annihilation) operators for electrons and holes in subband n with momentum k parallel to the layer and spin s, respectively. $\epsilon_{ink} = (E_g - \omega)/2 + (k^2 + n^2\pi^2/L^2)/2m_i$, $i = e,h$, are the corresponding kinetic energies ($\hbar = 1$), L is the QW thickness, and m_e and m_h are the electron and hole effective masses. Diagonalization of the first two terms of the Hamiltonian [Eq. (1)] yields the eigenstates and energies of a biased QW,[6,7] while diagonalization of the first and last term yields the electron and hole states dressed by an optical field.[3]

Screening by coherent e-h pairs has been extensively studied in the context of exciton Bose condensation[8]; in leading order in the virtual-pair density $N \simeq |er_{vc}E_\omega|^2$, we find for the relative change in the static dielectric constant

$$\frac{\delta\epsilon_0}{\epsilon_0} = \frac{2\pi e^2 |er_{vc}E_\omega|^2}{\epsilon_0 AL} \sum_{n,m,k,s} \frac{(\langle n|z|m\rangle)^2}{\epsilon_{enk} + \epsilon_{hmk}} \left[\frac{1}{\epsilon_{enk} + \epsilon_{hnk}} - \frac{1}{\epsilon_{emk} + \epsilon_{hmk}}\right]^2, \quad (2)$$

where A is the QW area. The induced electrostatic field is $\delta F = -(\delta\epsilon_0/\epsilon_0)F$.

Equation (2) is nothing but $-\langle 4\pi P_{\text{intra}}\rangle/(\epsilon_0 ALF)$ and comprises the four elementary polarization bubbles that can be constructed from normal and anomalous e and h propagators.[8] It can be recast in the form

$$\delta\epsilon_0/\epsilon_0 = (|er_{vc}E_\omega|^2/4R^2)(R/W)^{5/2} U(\Delta = (E_g + W - \omega)/W, \sigma = m_e/m_h), \quad (3)$$

where $W = \pi^2/(2mL^2)$ is the confinement energy, and $R = e^2/(2\epsilon_0 a_0) = (2ma_0^2)^{-1}$ the excitonic Rydberg, m being the reduced e-h mass. The universal function U is plotted in Fig. 1 as a function of the dimensionless detuning Δ from the lowest QW transition. Clearly, there exist two different regimes. For detunings much larger than the confinement energy, $\Delta \gg 1$, U varies as $\Delta^{-5/2}/4$, so that $\delta\epsilon_0/\epsilon_0$ becomes *independent* of the QW thickness. This shows nicely that nonresonant nonlinearities are rather insensitive to size quantization, because they involve sums over complete sets of states which, because of the conservation of total oscillator strength, are of course not affected by any change in quantization. Such sum rules are also implicit in the QCSE and QCFKE models that we use.[7,9] Thus, for large detunings, $\delta\epsilon_0/\epsilon_0$ reduces to its bulk value

$$\frac{\delta\epsilon_0}{\epsilon_0} = \frac{2\pi e^2 |er_v cE_\omega|^2}{\epsilon_0 AL} \sum_{k_z,k,s} \frac{(k_z/m)^2}{[E_g + (k^2 + k_z^2)/2m - \omega]^5}. \quad (4)$$

On the other hand, for small detunings, $\Delta \ll 1$, only the (most resonant) QW ground state contributes and Eqs. (3) and (2) reduce to

$$U(\Delta,\sigma) = (1+\sigma)(1+\sigma^{-1})(15-\pi^2)/(6\pi\Delta)$$

and

$$\delta\epsilon_0 = 4\pi N_1(\alpha_{e1}+\alpha_{h1}),$$

where

$$N_1 = (|er_{vc}E_\omega|^2/4AL)\sum_{k,s}[E_g+W+k^2/2m-\omega]^{-2}$$

is the density of virtual $n=1$ e and h, and

$$\alpha_{i1} = 2e^2\sum_n(\langle n|z|1\rangle)^2/(\epsilon_{ink}-\epsilon_{i1k}),$$

$i=e,h$, is their polarizability.

We now examine some consequences of this theory. The approximations for which we have presented results here are equivalent to retaining nonlinear susceptibilities up to order $\chi^{(3)}$, although the theory is not restricted to this. It can be shown from Eq. (2) that, for a given detuning below the zero-field optical absorption edge, the $\chi^{(3)}$ (and consequently the change in field, δF) is larger for larger layer thickness L, hence appearing to favor thick layers or even bulk materials. The problem, however, is that for thicker layers progressively less applied field is required to induce real absorption at the below-gap optical frequency because of the QCSE/QCFKE red shift of the lowest transition.[7] Such real absorption would create a photovoltage that would generally persist long after the ultrashort light pulse was over, reducing the usefulness of the effect. It is clear that the generated change in field cannot exceed the applied field. Hence, thin layers are required for the generation of substantial fields without incurring real populations.

Within the $\chi^{(3)}$ approximation, this effect can be regarded as field-induced optical rectification from the inversion of the QCSE/QCFKE-induced quadratic electrorefraction $\chi^{(3)}(-\omega,\omega,0,0)$.[10] By the permutation symmetries of the nonlinear susceptibility,[11] there must be an equal $\chi^{(3)}(0,0,-\omega,\omega)$. In fact, *exactly* the same expression [Eq. (2)] can be obtained by calculating the leading changes in the refractive index due to an applied dc field F, i.e., the QCFKE.

We can estimate the size of the effect for 100-Å GaAs quantum wells (with infinitely high barriers). We use $m_e=0.067m_0$ and $m_h=0.45m_0$ (where m_0 is the free-electron mass), $\epsilon_0=12.35$, giving $R=4.2$ meV and $W=64.8$ meV, and take $|er_{vc}|^2=230$ eV Å^3 (corresponding to a momentum matrix element $P^2=29$ eV). We calculate $\chi^{(3)}(0,0,-\omega,\omega)\simeq1.2\times10^{-9}$ esu for a detuning $E_g+W-\omega$ of $10R$ (42 meV) below the zero-field energy of the lowest interband transition. Such a detuning is sufficiently large that this would be usable even at room temperature without large real absorption. For a multiple-well sample with equal well and barrier thicknesses, the average $\chi^{(3)}$ is half of this.

We can estimate the importance of excitonic effects and obtain an independent estimate of the $\chi^{(3)}$ from semiempirical quadratic electrorefraction calculations that are based on Kramers-Kronig transformation of real QW electroabsorption spectra.[10] At 10^5 V/cm and 42 meV below the zero-field exciton peak, the calculated d^2n/dF^2 for equal well and barrier widths is $\simeq7.2\times10^{-13}$ cm^2/V^2 [$\chi^{(3)}(-\omega,\omega,0,0)\simeq1.8\times10^{-8}$ esu]. With our field definitions, the permutation relation gives $\chi^{(3)}(0,0,-\omega,\omega)=\frac{1}{2}\chi^{(3)}(-\omega,\omega,0,0)\simeq9\times10^{-9}$ esu, implying an enhancement of $\simeq15$, which would make the actual effect correspondingly larger than our simplified first-principles estimate above. Although there are contributions from light holes and the actual wave functions are somewhat larger in the real well than in our idealized infinite-barrier case, we estimate that the majority of this enhancement comes from the excitonic effects.

As an illustration, consider for example a multiple-quantum-well structure 1 μm thick consisting of fifty 100-Å GaAs wells separated by 100-Å barriers and contained in the intrinsic region of a *p-i-n* diode reverse biased to 10 V to give a field $F\simeq10^5$ V/cm. For an incident intensity of 10 GW/cm^2 at a photon energy 42 meV below the zero-field absorption edge, the voltage change generated would be -2.2 V. Such an optical intensity could be generated by an unamplified short-pulse dye laser producing $\simeq100$ pJ, $\simeq100$ fs optical pulses focused to a 10-μm^2 area. This calculation is near the limit of validity of the present model, as the calculated field change is a substantial fraction of the applied static field, and the generated virtual-pair density ($N\simeq5\times10^{17}$ cm^{-3} within the well) is sufficiently large that other many-body effects may become important.

This effect can be viewed as a photodetector working with virtual rather than real electron-hole pairs. An extension of this is to use the resulting electric field to change the optical properties of quantum-well material through the QCSE to make an all-optical device. This is the general principle that underlies the self-electro-optic effect device (SEED) for real transitions.[12] An analogous "virtual SEED" process would be for example, a field-induced four-wave mixing in which two interfering beams generate an electric field "grating" that in turn generates a refractive index grating through QCSE electrorefraction, and hence allows scattering of a third beam off the grating to generate a fourth beam. It can readily be shown that this effect is given by $\chi^{(3)}_{\text{effective}}(-\omega',\omega',-\omega,\omega)=-8\pi\chi^{(3)}(-\omega',\omega',0,0)\times\chi^{(3)}(0,0,-\omega,\omega)F^2/\epsilon_0$; this is $\simeq7\times10^{-11}$ esu for the semiempirical $\chi^{(3)}$ above at 10^5 V/cm and 42 meV detuning for both ω and ω'.

The effects discussed here could also be obtained in quantum-well materials with internally rather than externally generated field. Recently, it has been proposed that fields $\simeq10^5$ V/cm that would result in a built-in QCSE would occur naturally in quantum wells in certain strained-layer superlattices[13] through piezoelectric effects and the built-in strain.

In conclusion, we predict that ultrafast electrical pulses can be generated in biased QW's by short, intense light pulses in the transparent spectral region. We expect substantial voltage using low-power laser sources. Importantly, these voltages could be generated directly inside micron-thick semiconductor structures, offering new opportunities in high-speed electrical measurements and optoelectronic devices.

Note added.—Since the original submission of this manuscript, we have learned from Professor M. Yamanishi that he has independently proposed a similar mechanism (see preceding Letter[14]).

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