

Terahertz sideband generation in quantum wells viewed as resonant photon tunneling through a time-dependent barrier: An exactly solvable model

D. S. Citrin* and W. Harshawardhan

Semiconductor Optics Theory Group, Department of Physics and Materials Research Center, Washington State University, Pullman, Washington 99164-2814

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The propagation of light through quantum wells in the vicinity of an excitonic resonance subjected to a THz electric field is considered. The treatment makes explicit the relationship between THz sideband generation and resonant photon tunneling through a time-dependent resonant-tunneling structure. A method for the coherent control of excitons in quantum wells is also suggested. [S0163-1829(99)11927-1]

The basis for much work on transport through harmonically varying time-dependent potentials dates back to the work of Tien and Gordon¹ on tunneling between superconducting films. Subsequent developments in semiconductor-heterostructure growth and split-gate devices largely account for the renaissance of interest in this area. A parallel development is the free-electron laser. The modulation via microwave and THz fields of transport properties of quantum-point contacts,² single-electron devices,³ and resonant-tunneling structures^{4,5} has been studied. Effort has also been put into observing THz sidebands (TS's) in the optical spectra of semiconductor quantum wells (QW's).⁶ Despite substantial theoretical progress in the area of tunneling through time-dependent potentials, experimental results have been scant. And apart from the common role of the low-frequency field (microwave, THz) in inducing multiphoton transitions in the system,⁷ an *explicit* elucidation of the connection between resonant tunneling through time-dependent potentials and TS generation (TSG) was hitherto unavailable.⁸ The connection though may prove valuable since resonant-tunneling devices based on semiconductor heterostructures introduce numerous complications prohibiting the controlled variation of a single parameter. These difficulties have hampered a detailed analysis of transport experiments through time-dependent potentials. Typically, experimental results have been modeled using Tien-Gordon theory which only captures the most basic effects.^{7,9} Recently, however, exciting work has been published showing that the quantum mechanical phase of electrons can be measured in *tour de force* transport experiments^{10,11} lending urgency to the exploitation of optical analogues where phase measurements are routine. Optical

analogues of time-dependent transport problems therefore hold out the intriguing prospect of realizing a number of effects that have been theoretically predicted. Already, considerable experimental work on superluminality and its impact on issues concerning tunneling times has been carried out.¹²

In this study we present a theoretical treatment of light propagation through a QW in which the exciton energy, linewidth, and oscillator strength are modulated with bandwidths in the 100-GHz to 5-THz range (henceforth denoted THz field), such as provided by excitonic Stark effects induced by a THz field $\mathbf{F}(t)$ polarized in the QW plane $\hat{\mathbf{x}}$ or the quantum-confined Stark effect with $\mathbf{F}(t)$ parallel to the growth direction $\hat{\mathbf{z}}$. The formalism employed makes *explicit* the close relationship between TSG and resonant tunneling of photons through the QW in the presence of a time-dependent dielectric constant. As is well known,¹³ the wave and Schrödinger equations can be mapped from one to the other via $(\omega/c)n(\omega, \mathbf{r}) \leftrightarrow \{(2m/\hbar^2)[E(\omega) - V(\mathbf{r})]\}^{1/2}$, respectively, where the symbols here have their usual meanings. Previous studies of TSG (Ref. 6) have avoided the details of the propagation effects which bring to the fore the close analogy between the two classes of phenomena.

We consider linear optical propagation although the THz field may be strong. Thus, we are concerned with the low-density regime in which excitation-induced dephasing and saturation may be neglected. Let an optical pulse [amplitude $I(t)$] be incident normally on the QW in the vicinity of an excitonic resonance. The reflected $R(t)$ and transmitted $T(t)$ optical fields are given by¹⁴

$$\begin{bmatrix} T(t) \\ 0 \end{bmatrix} = \begin{bmatrix} \int_{-\infty}^t dt' [\delta(t-t') + \chi(t,t')] & \int_{-\infty}^t dt' \chi(t,t') \\ -\int_{-\infty}^t dt' \chi(t,t') & \int_{-\infty}^t dt' [\delta(t-t') - \chi(t,t')] \end{bmatrix} \begin{bmatrix} I(t') \\ R(t') \end{bmatrix}, \quad (1)$$

with $\chi(t, t')$ the optical susceptibility (to be discussed below) and the integrals operate on everything to the right.¹⁴ We eliminate $R(t)$ from Eq. (1) to obtain

$$T(t) = I(t) + \int_{-\infty}^t dt' \chi(t, t') T(t'). \quad (2)$$

If $\dot{\mathbf{F}} = \mathbf{0}$, then $\chi(t, t') = \chi(t - t')$, and Eq. (2) is easily solved in the frequency domain; however, for $\dot{\mathbf{F}} \neq \mathbf{0}$, the susceptibility no longer depends simply on $t - t'$, but explicitly on the two time variables. Nevertheless, we shall see that even in this case, under certain conditions an explicit solution obtains.

The optical susceptibility $\chi(t, t')$ in the presence of $\mathbf{F}(t)$ can be treated as in Ref. 15 or solved for numerically,¹⁶ however, as we show, a simple model can be considered which leads to a closed-form solution of Eq. (2). To motivate a physically reasonable *ansatz* for $\chi(t, t')$, first consider the case $\dot{\mathbf{F}} = \mathbf{0}$. For a single resonance (e.g., retaining the $1s$ exciton, but neglecting the other bound states and continuum) we have¹⁴ $\chi(t - t') = \Gamma e^{-i(\varepsilon - i\gamma)(t - t')}$ where Γ is the exciton radiative width,¹⁷ ε is the frequency of the exciton measured from the crystal ground state, and γ is the nonradiative linewidth. The parameters ε , γ , and Γ are for $\dot{\mathbf{F}}(t) = \mathbf{0}$. All other interband resonances of the QW are neglected and the line shape is assumed for convenience to be Lorentzian. If the Hamiltonian governing the interband electronic excitations of the QW depends on time through a THz-frequency electric field, but with frequency content sufficiently below any intraband resonance, we can account for the effects of the THz field through an adiabatic Stark shift, line broadening, and modification of the oscillator strength. (For the parameters investigated here, ultrahigh-quality ZnSe or GaN QW's might be the best candidates due to their high exciton binding energies.) We can thus use the *ansatz* $\chi(t, t') = G(t)G^*(t')$ with $G(t) = \Gamma^{1/2}(t)e^{-i\int^t dt' [\varepsilon(t') - i\gamma(t')]}$ where $\varepsilon(t)$, $\Gamma(t)$, and $\gamma(t)$ all depend on time due to the influence of \mathbf{F} .¹⁸ The transfer-matrix method is a formulation of the scattering approach to such problems and is well known to follow from the tunneling Hamiltonian for the appropriate structure.¹⁸ The reflectivity and transmission coefficients are *equivalent* to the Green function of the appropriate tunneling Hamiltonian.

Equation (2) can be converted into a differential equation. The solution is

$$T(t) = I(t) + \int_{-\infty}^t dt' \chi(t, t') I(t') e^{-\int_{t'}^t dt'' \Gamma(t'')}. \quad (3)$$

This defines the response function $\delta(t - t') + \tilde{\chi}(t, t')$ with $\tilde{\chi}(t, t') = g(t)g^*(t')$, and

$$g(t) = \Gamma^{1/2}(t) e^{-i\int^t dt' \{\varepsilon(t') - i[\gamma(t') + \Gamma(t')]\}}.$$

Using $T = I + R$ [cf. Eq. (1)], we have $R(t) = \int_{-\infty}^t dt' \tilde{\chi}(t, t') I(t')$.

In the following we assume the THz field $\mathbf{F}(t) = \mathbf{F}_{dc} + \mathbf{F}_{ac} \cos \Omega t$ is applied to the QW with \mathbf{F}_{dc} (which may be zero) a dc bias and $\mathbf{F}_{dc} \parallel \mathbf{F}_{ac}$. We write the modulated parameters as $\varepsilon(t) = \varepsilon_0 + \varepsilon_1 \cos \zeta t$, $\gamma(t) = \gamma_0 + \gamma_1 \cos \zeta t$, and $\Gamma(t)$

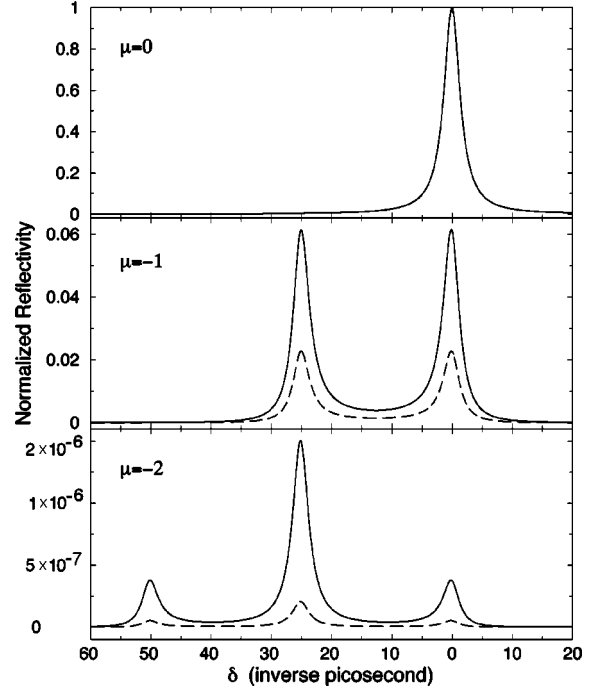


FIG. 1. Reflectivity $|R_\omega(\omega + \mu\zeta)|^2$ into harmonic μ normalized to peak reflectivity of the unmodulated fundamental $(\Gamma/\Gamma')^2 = 1.7\%$ as a function of detuning δ from exciton line center. The parameters are $\varepsilon_1 = 2.5$ (solid) and 1.52 ps^{-1} (dashed); in all cases $\gamma_0 = 1.52 \text{ ps}^{-1}$, $\Gamma = 0.1 \text{ ps}^{-1}$, and $\gamma_1 = 0$.

$= \Gamma_0 - \Gamma_1 \cos \zeta t$ where $\zeta = \Omega$ if $F_{dc} \gg F_{ac}$ or $\zeta = 2\Omega$ if $F_{dc} = 0$. Typical values may be found in Refs. 19. We set $\varepsilon_i = \varepsilon_i - i\gamma_i$ and $\tilde{\varepsilon}'_i = \tilde{\varepsilon}_i - i\Gamma_i$ with $i = 0, 1$. If $F_{dc} = 0$, Stark effects are quadratic and the parameters are modulated at frequency 2Ω . This leads to even TS's of Ω as is expected for a system with inversion symmetry. If $F_{dc} \neq 0$, the system lacks inversion symmetry and both even and odd TS's occur.

Henceforth, we assume $\Gamma_1 = 0$. Let $I(t) = e^{-i\omega t}$. Then

$$T(t) = T_\omega(t) = e^{-i\omega t} + \Gamma e^{-i\omega t} \int_{-\infty}^t dt' e^{-i(\tilde{\varepsilon}'_0 - \omega)(t - t')} e^{-i(\tilde{\varepsilon}'_1/\zeta)[\sin \zeta t - \sin \zeta t']}. \quad (4)$$

Set $\tau = t - t'$, $\bar{t} = (t + t')/2$, and $\sin \zeta t - \sin \zeta t' = 2 \sin(\zeta\tau/2) \cos \zeta\bar{t}$. Fourier transform $T_\omega(\omega')$ $= \int_0^{2\pi/\zeta} dt e^{i\omega' t} T_\omega(t)$ to obtain $T_\omega(\omega') = (2\pi/\zeta) [(\omega - \tilde{\varepsilon}'_0)/(\omega - \tilde{\varepsilon}'_0) \delta_{\omega, \omega'} + \mathcal{K}_\mu(\omega) \delta_{\omega - \omega', \mu\zeta}]$ with

$$\mathcal{K}_\mu(\omega) = 2i\Gamma\Delta \sum_{k=1}^{\infty} \frac{1}{\Delta^2 - (k\zeta/2)^2} \times J_{(k+\mu)/2} \left(\frac{\tilde{\varepsilon}'_1 k}{2\Delta} \right) J_{(k-\mu)/2} \left(\frac{\tilde{\varepsilon}'_1 k}{2\Delta} \right), \quad (5)$$

$\bar{\omega} = (\omega + \omega')/2$ the average of the incoming and outgoing frequencies, μ an integer, $\Delta = \Delta_\mu(\omega) = \bar{\omega} - \tilde{\varepsilon}'_0 = \omega - \mu\zeta/2 - \tilde{\varepsilon}'_0$ the complex detuning between the average frequency and the radiatively renormalized excitonic resonance, and

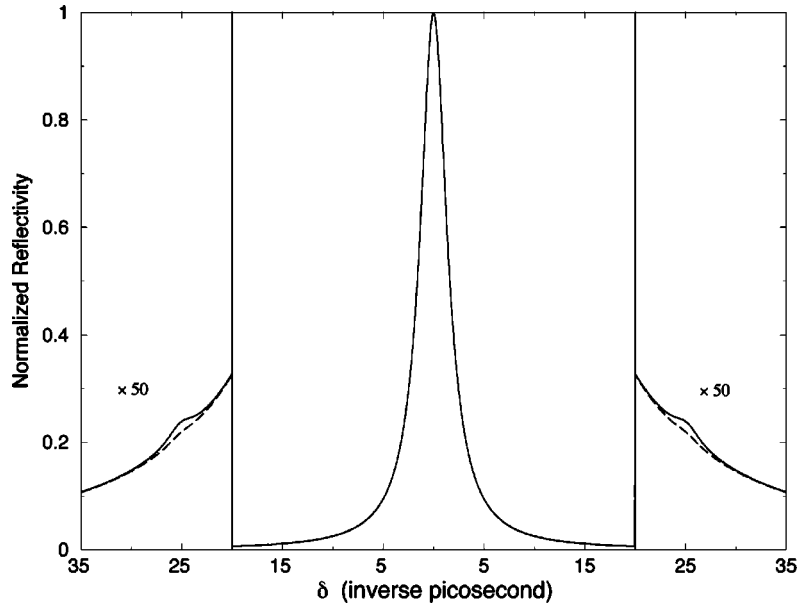


FIG. 2. Total reflected probability $R_{\text{tot}}(\varepsilon)$ for the same parameters as Fig. 1 (solid). Note that for clarity, the region $|\delta| > 20 \text{ ps}^{-1}$ is multiplied by a factor of 50.

$\sum_{k=1}^{\infty}$ the sum restricted to integers k of the same parity as μ . $R_{\omega}(\omega')$ is obtained from the previous formula by replacing $(\omega - \tilde{\varepsilon}_0)/(\omega - \tilde{\varepsilon}'_0)\delta_{\omega, \omega'}$ by $i\Gamma/(\omega - \tilde{\varepsilon}'_0)\delta_{\omega, \omega'}$. The first term leads to the unmodulated result,¹⁴ $\varepsilon_1, \gamma_1 = 0$. The other term gives the TS's as well as the modified transmission of the fundamental. Finally, for $\tilde{\varepsilon}_1 = \varepsilon_1$ ($\gamma_1 = 0$), direct integration gives for the total reflected probability per unit time

$$R_{\text{tot}}(\omega) = (\zeta/2\pi) \int_0^{2\pi/\zeta} dt |R_{\omega}(t)|^2 = (\zeta/2\pi) \times (\Gamma/\Gamma') \text{Re} R_{\omega}(\omega) \quad (\text{optical theorem}) \quad \text{where } \Gamma' = \Gamma + \gamma_0.$$

The foregoing expressions for R and T are compact; however, it is useful to reexpress the Kapteyn series for $\mathcal{K}_{\mu}(\omega)$ in Eq. (5) as a power series in $z = \tilde{\varepsilon}_1/\zeta$ (Ref. 20) to identify the various multi-THz-photon processes that contribute to a given TS:

$$\mathcal{K}_{\mu}(\omega) + i \frac{\Gamma}{\Delta} \delta_{\mu,0} = \begin{cases} i\Gamma \Delta \sum_{n=|\mu|/2}^{\infty} \left(\frac{\tilde{\varepsilon}_1}{2}\right)^{2n} \binom{2n}{|\mu|} \prod_{j=0}^n [\Delta^2 - (j\zeta)^2]^{-1}, & \mu \text{ even} \\ i\Gamma \sum_{n=(|\mu|+1)/2}^{\infty} \left(\frac{\tilde{\varepsilon}_1}{2}\right)^{2n-1} \binom{2n-1}{|\mu|} \prod_{j=1}^n \left[\Delta^2 - \left(\frac{2j-1}{2}\zeta\right)^2\right]^{-1}, & \mu \text{ odd.} \end{cases} \quad (6)$$

Series (5) and (6) converge in $|ze^{\sqrt{1-z^2}}/(1+\sqrt{1-z^2})| < 1$ and for arbitrary $2\Delta/\zeta$ not an even (odd) integer.²⁰ The terms to a given order in $\tilde{\varepsilon}_1$ summed over μ are the nonlinear susceptibilities. For fixed μ , the term of degree p in $\tilde{\varepsilon}_1$ describes p th-order multi-THz-photon processes contributing to the TS. This in turn allows one to write the quantum mechanical transmission amplitude as a sum of nonlinear susceptibilities, as is commonly carried out in optics. The expansion (6) in terms of susceptibilities applies *mutatis mutandis* to the transport case, and to our knowledge was hitherto unknown.

Depending on whether \mathbf{F} is polarized in the $\hat{\mathbf{x}}$ or $\hat{\mathbf{z}}$ direction, the dominant effect will differ; for $\mathbf{F} \parallel \hat{\mathbf{x}}$, the time dependence of $\gamma(t)$ will dominate through the ionization rate, while if $\mathbf{F} \parallel \hat{\mathbf{z}}$, $\varepsilon(t)$ will play a larger role.¹⁹ Of course, $\varepsilon(t)$,

$\gamma(t)$, and $\Gamma(t)$ are closely interlinked (see comments in Ref. 5), but for our purposes we can treat them as parameters determined by a microscopic theory external to our model.

To illustrate the different types of behaviors expected in these cases, we present results for the normalized reflectivity $(\Gamma'/\Gamma)^2 |R_{\omega}(\omega + \mu\zeta)|^2$ [$(\Gamma/\Gamma')^2 = 0.017$ is the peak unmodulated reflectivity] of a high-quality QW in Fig. 1. We take $\gamma_1 = 0$ and consider the two cases 2.5 (solid) and 1.52 ps^{-1} (dashed) for $\mu = 0, 1$, and 2. The results are plotted as a function of $\delta = \text{Re} \Delta$. [In all cases, $\Gamma = 0.1 \text{ ps}^{-1}$, $\gamma_0 = 1.52 \text{ ps}^{-1}$ ($\hbar\gamma_0 = 1 \text{ meV}$), and $\zeta/2\pi = 4 \text{ THz}$.] This gives the reflected probability into the μ th TS as a function of incident frequency. The normalized reflectivity for $\mu = 1$ is in the few % range while that of $\mu = 2$ in the $10^{-4}\%$ range for the parameters. Figure 2 shows $R_{\text{tot}}(\omega)$ for the same parameters. The relative maximum strength of the first TS with

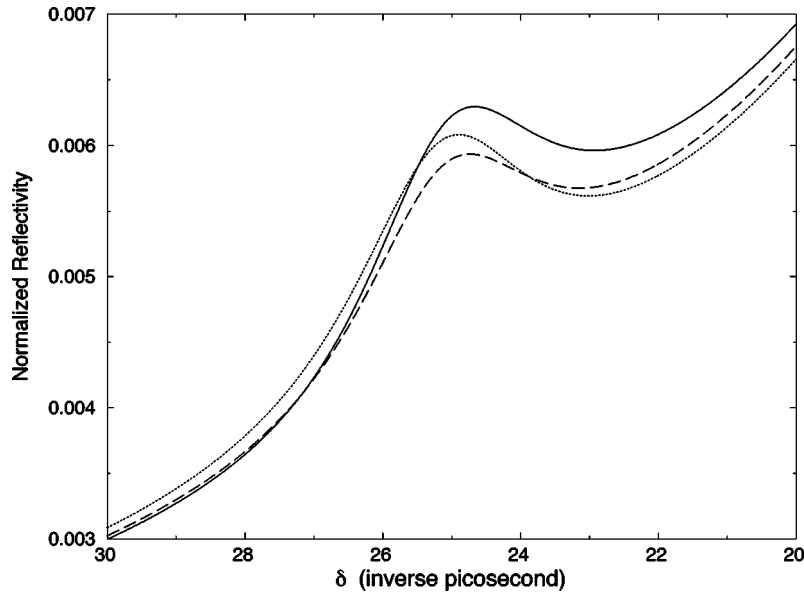


FIG. 3. Total reflected probability for $\tilde{\varepsilon}_1 = \sqrt{2}\pi \text{ ps}^{-1}$ (dotted), $(1+i)\pi \text{ ps}^{-1}$ (dashed), and $i\sqrt{2}\pi \text{ ps}^{-1}$ (solid). The parameters are otherwise those of Fig. 1.

respect to the unmodulated response is $\sim 14.5\%$ for $\varepsilon_1 = 2.5 \text{ ps}^{-1}$. Figure 3 shows $R_{\text{tot}}(\omega)$ for $\tilde{\varepsilon}_1 = \sqrt{2}\pi \text{ ps}^{-1}$ (dotted), $(1+i)\pi \text{ ps}^{-1}$ (dashed), and $i\sqrt{2}\pi \text{ ps}^{-1}$ (solid) with the other parameters the same as in Fig. 1 to show the effect of modulating the line center versus the linewidth. In all cases $|\tilde{\varepsilon}_1| = \sqrt{2}\pi \text{ ps}^{-1}$.

We turn our attention to time-domain effects. Specifically, we describe a new technique for coherent control of excitons in QW's using phase-locked pulse pairs. Previous schemes^{21,22} utilize the relative phase between two identical but time-delayed optical pulses coincident with a narrow optical resonance in a QW. If the two pulses are in phase, the interband polarizations excited by each are in phase, and as a result ideally four times the population of excitons associated with a single pulse are generated; if, however, the pulses are π out of phase, the interference is destructive and the QW is coherently depopulated of excitons following the passage of the second pulse. Or at least that is the idea; dephasing, nonlinearities, and inhomogeneous broadening all conspire to make this effect less marked than one might hope.²² In the scheme proposed here, two identical phase-locked time-delayed pulses are also used, but rather than modulate the relative phase of the two pulses, we introduce a phase shift in the polarization induced by the first optical pulse by a half-cycle THz pulse incident on the QW between the two optical pulses. Set $I(t) = I_0(t+t_0/2) + I_0(t-t_0/2)$ where $I_0(t) = A(t)e^{-i\varepsilon_0 t}$; $A(t)$ is the slowly varying envelope of a single constituent pulse of duration $\tau_0 \ll t_0 \ll \gamma_0^{-1}$, the time delay between pulses. Also let $\varepsilon(t) = \varepsilon_0 + \varepsilon_1 B(t)$ and $\gamma(t) = \gamma_0$. $B(t)$ is a peaked function of time centered at $t=0$ whose duration is much less than t_0 . For example, $B(t)$ might be due to a photoconductively generated half-cycle THz pulse propagating down a transmission structure deposited on the QW. For $t \gg t_0$,

$$\begin{aligned} T(t) &= \Gamma \int_{-\infty}^{\infty} dt' e^{-i\tilde{\varepsilon}_0'(t-t')} e^{-i\varepsilon_1 \int_t^t dt'' B(t'')} I(t') \\ &= \Gamma e^{-i\tilde{\varepsilon}_0' t} [e^{-i\varepsilon_1 \int_{-\infty}^{\infty} dt'' B(t'')} + 1] \int_{-\infty}^{\infty} dt' A(t') \quad (7) \end{aligned}$$

and $R(t) = T(t)$. If the THz pulse is chosen so that $\varepsilon_1 \int_{-\infty}^{\infty} dt'' B(t'') = (2n+1)\pi$, destructive interference between the polarizations excited by the two optical pulses occurs and thus coherent depopulation of the excitons ensues; if the action integral is $2n\pi$, then constructive interference ensues.

As an example, if the half-cycle THz pulse is of 0.5 ps duration, we need $\hbar\varepsilon_1 \approx 2 \text{ meV}$ to achieve a π phase shift. This is feasible using available THz techniques, though carrying out this experiment may be difficult. One needs very high-quality QW's at low temperatures to ensure narrow excitonic lines. In addition, the THz transmission structure should be designed with $\mathbf{F}(t) \parallel \hat{\mathbf{z}}$ to suppress modulation of the dephasing; however, this can be compensated for by decreasing the amplitude of the second optical pulse in the sequence. Finally, the desirable effect will be degraded if there is temporal overlap of $B(t)$ and $A(t \pm t_0/2)$, thus requiring clean optical and THz wave forms. We note that the present scheme in no way circumvents the central difficulties with coherent depopulation of the exciton level, namely, inhomogeneous broadening and excitation-induced dephasing.

To conclude, we present an exactly solvable model for TSG by and photon propagation through THz-modulated QW's. Because of the mapping between the electromagnetic wave equation and the Schrödinger equation, light propagation through THz-modulated QW's will be a fruitful way to explore resonant tunneling through time-dependent potentials. We believe the area of optical analogues of time-dependent quantum mechanical transport problems—as pioneered by the work reviewed in Ref. 12—is in its infancy,

and in particular, exploitation of optical pulse propagation through QW's shows great promise to advance the field materially. In particular, we have explored the effects of modulating the exciton linewidth and energy on the TS's appearing in the optical spectra. We have also proposed an

alternative scheme for carrying out two-pulse coherent control of excitons in QW's.

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*Electronic address: citrin@wsu.edu

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