Optical analogue for phase-sensitive measurements in quantum-transport experiments

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The Aharonov-Bohm effect has recently been employed to measure the quantum-mechanical phase of an electron propagating ballistically through a quantum dot (QD) contained in one arm of the interferometer [Schuster *et al.*, Nature (London) **385**, 417 (1997)]. Jauho and Wingreen [Phys. Rev. B **58**, 9619 (1998)] have analyzed this geometry under conditions where the QD is modulated harmonically in time as a way to distinguish unambiguously between coherent [e.g., photon-assisted transport] and incoherent processes [e.g., sequential resonant tunneling]. We show that an optical analogue of this experiment exists, namely, the propagation of light through a quantum well in the vicinity of an excitonic resonance subjected to a THz electric field. By carrying out an interferometric experiment, the phase of a transmitted (or reflected) optical beam at a sideband frequency—analogous to the electron's phase—can be measured. These phase shifts can be understood in terms of specific multi-THz-photon processes. The optical experiment affords fundamental insight not ordinarily accessible in transport experiments. [S0163-1829(99)16931-5]

Interference of a single particle propagating simultaneously over two paths is a fundamental manifestation of quantum-mechanical behavior. A textbook approach to this phenomenon is first to introduce the Young double-slit experiment.[1] Light which passes through a screen containing two slits interferes on another screen to produce fringes in the optical intensity. If the optical path lengths from each of the slits to a given point on the screen differ by an integer number of optical wavelengths, there is constructive interference and thus a bright fringe. If, however, the optical path lengths differ by an odd half-integer multiple of the wavelength, there is a dark fringe due to destructive interference.

Indeed, the similarity of quantum-mechanical and optical interferences is no accident; it is well known[2] that the electromagnetic wave equation and Schrödinger equation can be mapped to each other by $(\omega/c)n(\omega,\mathbf{r})\leftrightarrow\{(2m/\hbar^2)[E-V(\mathbf{r})]\}^{1/2}$, where ω is the electromagnetic frequency, c is the *in vacuo* speed of light, and $n(\omega,\mathbf{r})$ the frequency and spatially dependent refractive index for the electromagnetic case. For the quantum-mechanical case, we have m for the particle mass, $\hbar$ for the reduced Planck constant, E for the energy, and $V(\mathbf{r})$ for the spatially varying potential. The connection, though clearly instructive from a pedogogical point of view, may prove invaluable more widely since resonant-tunneling devices based on semiconductor hetero and split-gate structures introduce numerous practical experimental complications as well as make it extremely difficult to control a single elementary parameter of the device. The problems multiply when the potential is modulated in time. Most importantly to this study, the phases of the injected and detected particles cannot generally be controlled or measured without recourse to complex structures, whereas in the optical analogue both are straightforward. Moreover, an independent spectral resolution of the emerging particles is difficult for the electronic case but routine in optics. In other words, in transport experiments one usually integrates over all transport channels; the individual channels, however, contain information of fundamental importance concerning the roles of the various multiphoton processes contributing to the overall signal.

Thus optical analogues of single-particle quantum-transport problems provide a way to realize a number of effects, particularly in the time domain, that are otherwise difficult if not impossible to access. Already, considerable experimental work on superluminality and its relation to tunneling times have been carried out.[3]

These difficulties have hampered a detailed and direct study of the *quantum-mechanical phase* of particles undergoing quantum transport, in particular through time-dependent potentials. Most studies have been limited to a current-voltage curve which is closely related to the transmission probability summed over all transport channels as a function of the chemical potentials in the leads. Indeed, it is only quite recently[4,5] that a remarkable series of phase measurements in quantum-transport experiments have been carried out. In these studies, an Aharonov-Bohm interferometer is fabricated in a high-mobility two-dimensional electron gas in a split-gate geometry, and one of the arms contains a quantum dot (QD). This work has required the fabrication of state-of-the-art high mobility two-dimensional electron gases in submicron-patterned split-gate devices.

Jauho and Wingreen[6] have suggested that an interferometric geometry incorporating a QD might also be of interest to investigate another class of quantum-transport phenomena, namely, photon-assisted transport (PAT). In PAT, a low-frequency electric field (typically 10 GHz to 10 THz) is used to modulate the energy of the confined level through which electrons propagate. The THz photons dress the confined QD level, leading to the formation of sidebands and photon replicas, through which the electron may tunnel. In most experiments performed to date, it is not possible to distinguish between coherent and incoherent processes such as PAT and sequential resonant tunneling (SRT), respectively. In other words, rather than tunneling coherently through the structure via PAT, an electron can tunnel into the QD, dephase, there absorb one or more THz photons, and then tunnel out. Thus, whereas PAT involves only virtual transitions (i.e., dressed states), SRT involves real transitions induced by the THz photons. Amplitude-sensitive measurements are ill suited to

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differentiate between the two; however, only PAT is coherent and thus is sensitive to the phase.[6]

In this study, we explore an optical analogue of the quantum phase measurement that yields fundamental insights into transport processes which in practice are experimentally inaccessible and therefore have not hitherto been considered theoretically. Moreover, optical experiments can be carried out under less stringent conditions and with more routine samples. At present a free-electron laser is required, although in the near future we expect that requisite tabletop solid-state THz sources will be available. The optical analogue of the quantum phase measurement is sensitive to coherent nonlinear optical/THz mixing rather than to incoherent sequential THz absorption and reëmission. For example, an experiment of this type would provide a deeper understanding of the THz sidebands observed on optical spectra obtained from THz-modulated quantum-well (QW) magnetoexcitons,[7] and help to understand why the sideband spectra are asymmetric with respect to the fundamental—an effect at least partly associated with SRT. That is, we consider the transmission T or reflection R of a monochromatic optical beam I at frequency ω through a QW in the vicinity of an exciton resonance. At the same time, the QW is illuminated by a strong cw THz electric field $\mathbf{F}(t)$. Due to the coherent interaction of the virtual excitons created by the optical beam with THz photons, light will emerge from the QW at new frequencies $\omega' = \omega + \mu\Omega$ (sidebands), where μ is an integer and Ω is the modulation frequency (or $\omega + 2\mu\Omega$ for an inversion symmetric system). These processes will be reflected in the phases of R and T with respect to that of I.

We consider linear *optical* propagation, although the THz field may be strong. This is the low-density regime in which excitation-induced dephasing and saturation may be neglected. We have shown elsewhere that the transmission and reflection coefficients can be mapped via $R \leftrightarrow T$ directly to the harmonically modulated quantum-mechanical tunneling problem.[8] First we define the problem under consideration, then we freely employ the results of Ref. 8.

Let an optical pulse with time-dependent electric-field amplitude $I(t)$ be incident normally (propagation in the $\hat{\mathbf{z}}$ direction) on the QW in the vicinity of an excitonic resonance. We denote the reflected and transmitted amplitudes by $R(t)$ and $T(t)$, respectively. The momentum of the THz photons is miniscule and therefore neglected. Thus both R and T propagate to all intents and purposes in the $\hat{\mathbf{z}}$ direction as well. We assume that $I(t)$ possesses spectral components only near, for example, the $1s$, heavy-hole, exciton resonance. If the THz field $\mathbf{F}(t)$ is absent, the excitonic resonance is described by its frequency ε with respect to the crystal-ground state (cgs), the radiative width Γ of the exciton to undergo spontaneous emission to the cgs, and the nonradiative contribution γ to the exciton linewidth. If the Hamiltonian governing the interband electronic excitations of the QW depends on time through $\mathbf{F}(t)$, but with a spectral content sufficiently below any low-frequency resonance, we can account for the effects of $\mathbf{F}(t)$ through a Stark shift, line broadening, and modification of the oscillator strength depending parametrically on time. (We will only treat the first two.) Ultrahigh-quality ZnSe or GaN QW's might therefore be the best candidates due to their high exciton binding energies.

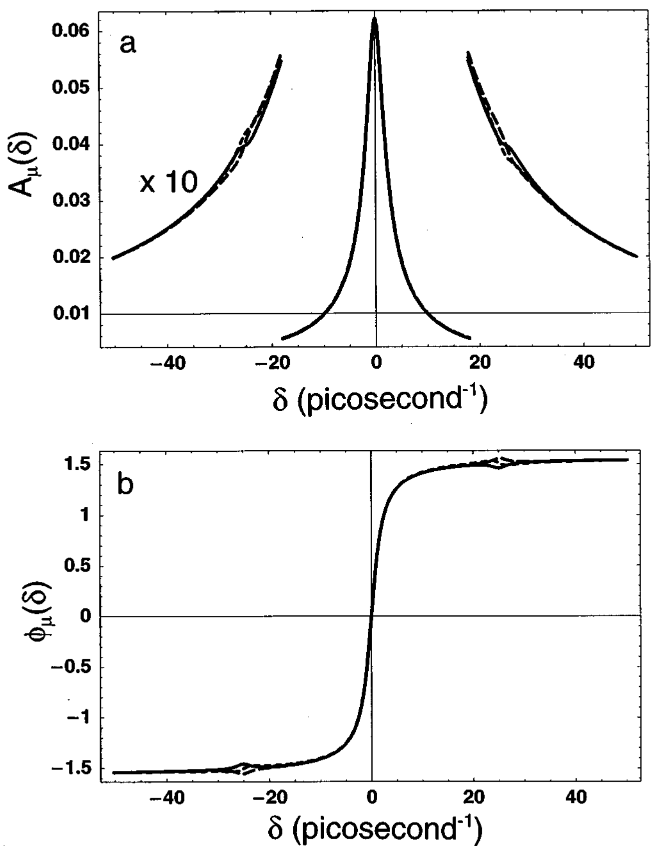

FIG. 1. Amplitude (a) and phase (b) into the fundamental ($\mu = 0$) as functions of detuning δ from the exciton line center for $\theta = 0$ (solid line), $\pi/4$ (dotted line), and $\pi/2$ (dashed line).

The quantum-confined Stark effect with $\mathbf{F}(t)$ parallel to $\hat{\mathbf{z}}$ or the excitonic Stark effect with $\mathbf{F}(t)$ parallel to the in-plane direction $\hat{\mathbf{x}}$ provides the harmonic modulation of the optical properties associated with the exciton. In particular, for $\mathbf{F}\|\hat{\mathbf{z}}$ the main effect is to modulate the exciton energy, while for $\mathbf{F}\|\hat{\mathbf{x}}$ the dominant effect is the modulation of the exciton linewidth due to exciton ionization. Below we investigate the impact on the phase resulting from modulating the exciton center frequency or the linewidth.

In the following we assume $\mathbf{F}(t)$ is of the form $\mathbf{F}_{\rm dc} + \mathbf{F}_{\rm ac}\cos(\Omega t + \alpha_\Omega)$, where $\mathbf{F}_{\rm dc}$ (which may be zero) is a dc bias and α_Ω is the phase of $\mathbf{F}$. For the cases treated in this study, we take $\mathbf{F}_{\rm dc}\|\mathbf{F}_{\rm ac}$. This dc offset permits the lifting of inversion symmetry, thus leading to the appearance of odd- as well as even-order THz sidebands in the optical spectra. We assume that the modulated QW parameters in the presence of the THz field may be written as $\varepsilon(t) = \varepsilon_0 + \varepsilon_1\cos(\zeta t + \alpha)$ and $\gamma(t) = \gamma_0 + \gamma_1\cos(\zeta t + \alpha)$, where $\zeta = \Omega$ if $F_{\rm dc} \gg F_{\rm ac}$ or $\zeta = 2\Omega$ if $F_{\rm dc} = 0$. Typical values of the parameters may be found in Refs. 9; α is a phase related to α_Ω. For brevity, we put $\tilde{\varepsilon}_i = \varepsilon_i - i\gamma_i$ ($i = 0$ and 1) and $\tilde{\varepsilon}_0' = \tilde{\varepsilon}_0 - i\Gamma$.

If a monochromatic wave $I(t) = e^{-i\omega t}$ is incident, we have shown from a scattering approach[8] that the transmitted field at time t is

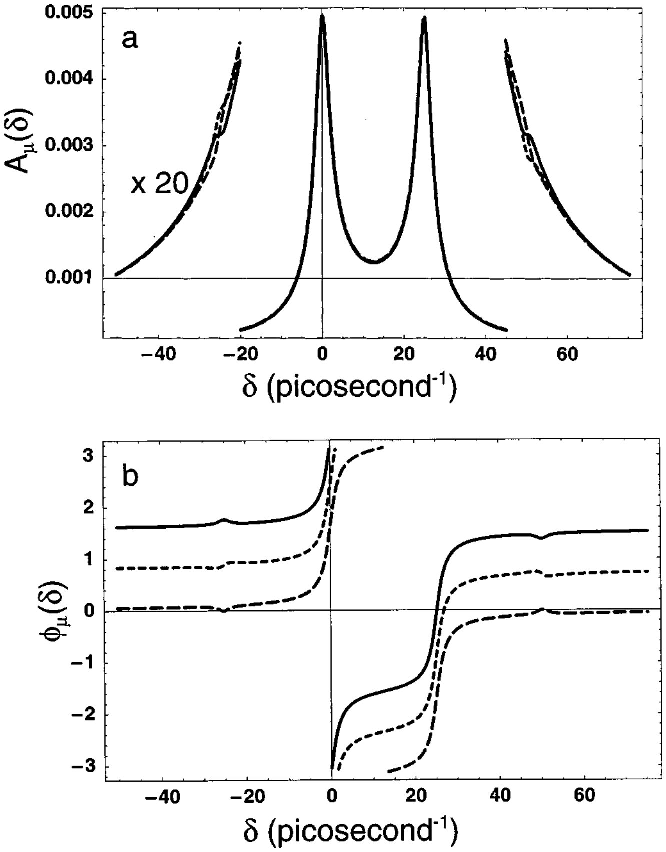

FIG. 2. Amplitude (a) and phase (b) into the first sideband ($\mu=1$) as functions of detuning δ from the exciton line center for $\theta=0$ (solid line), $\pi/4$ (dotted line), and $\pi/2$ (dashed line).

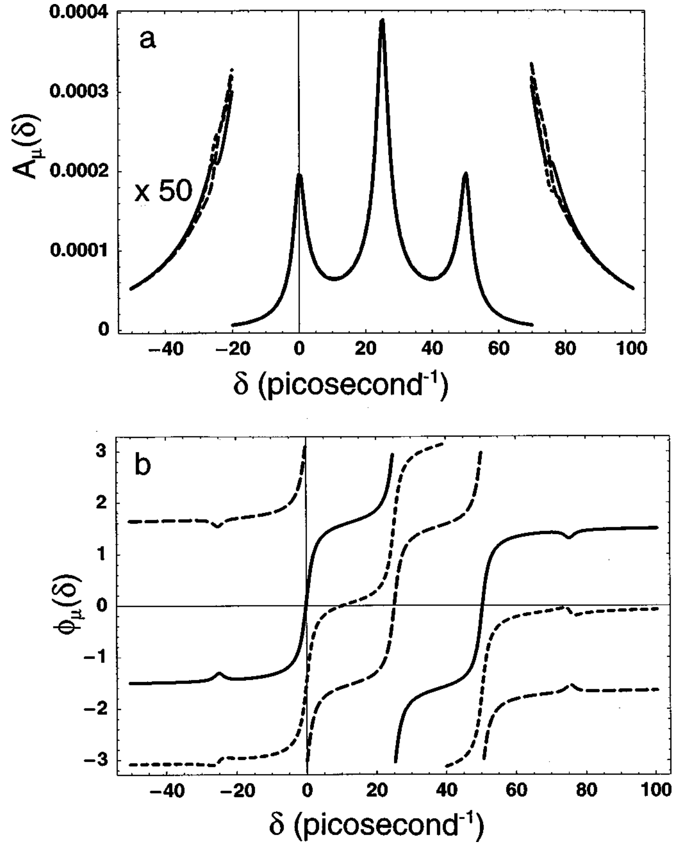

FIG. 3. Amplitude (a) and phase (b) into the second sideband ($\mu=2$) as functions of detuning δ from the exciton line center for $\theta=0$ (solid line), $\pi/4$ (dotted line), and $\pi/2$ (dashed line).

$$T(t,\omega)=e^{-i\omega t}+\Gamma e^{-i\omega t}\int_{-\infty}^{t}dt'\,e^{-i(\tilde{\varepsilon}_0'-\omega)(t-t')}\times\exp\left(-i\frac{\tilde{\varepsilon}_1}{\zeta}[\sin(\zeta t+\alpha)-\sin(\zeta t'+\alpha)]\right)\quad(1)$$

and $R=T-I$. (This result and all others carry over to the quantum-mechanical double-barrier problem via the mapping $R\leftrightarrow T$.) We make the substitutions $\tau=t-t'$ and $\overline{t}=[(t+t')/2]+\zeta$, employ the identity $\sin(\zeta t+\alpha)-\sin(\zeta t'+\alpha)=2\sin(\zeta\tau/2)\cos\zeta\overline{t}$, and perform the Fourier transform $T(\omega',\omega)=\int_0^{2\pi/\zeta}dt\,e^{i\omega' t}T(t,\omega)$ to obtain the transmitted amplitude at frequency ω',

$$T(\omega',\omega)=(2\pi/\zeta)[(\omega-\tilde{\varepsilon}_0)/(\omega-\tilde{\varepsilon}_0')\delta_{\omega,\omega'}+\mathcal{K}_\mu(\omega)\delta_{\omega-\omega',\mu\zeta}],$$

with

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

Here $\overline{\omega}=(\omega+\omega')/2$ is the average of the incoming and outgoing frequencies, μ is an integer denoting the sideband order, $\Delta=\Delta_\mu(\omega)=\overline{\omega}-\tilde{\varepsilon}_0'=\omega-(\mu\zeta/2)-\tilde{\varepsilon}_0'$ is the complex detuning between the average frequency and the radiatively renormalized excitonic resonance, and $\Sigma_{k=1}^{\infty\prime}$ is the sum restricted to integers k of the same parity as μ. $R(\omega',\omega)$ is obtained from $T(\omega',\omega)$ 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 in R and T leads to the unmodulated result,[10] $\varepsilon_1,\gamma_1=0$. The other term gives the sidebands 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 $R_{\rm tot}(\omega)=(\zeta/2\pi)\int_0^{2\pi/\zeta}dt|R(t,\omega)|^2=(\zeta/2\pi)(\Gamma/\Gamma'){\rm Re}R(\omega,\omega)$ (optical theorem), where $\Gamma'=\Gamma+\gamma_0$.

Optical theorems are standard in quantum transport.[11] The coherent transport associated with individual channels, however, is rarely considered, and in our model appears naturally in R or T associated with the various sidebands. Note that although the expressions for R and T are valid for modulation of either the exciton frequency or width, the optical theorem is only valid for modulation of the width. Thus our treatment allows for a more thorough investigation of parameter space than one based solely on the total transmitted or reflected probability.

The foregoing expressions for R and T are compact; however, it is instructive to reexpress the Kapteyn series for $\mathcal{K}_\mu(\omega)$ in Eq. (2) as a power series in $z=\tilde{\varepsilon}_1/\zeta$,[12] in order to identify the various multi-THz-photon processes that contribute to a given sideband. One obtains

$$\mathcal{K}_\mu(\omega)+i\frac{\Gamma}{\Delta}\delta_{\mu,0}=\begin{cases} i\dfrac{\Gamma\Delta}{\zeta^2}e^{-i\mu\alpha}\displaystyle\sum_{n=|\mu|/2}^{\infty}\left(\frac{\tilde{\varepsilon}_1}{2\zeta}\right)^{2n}\binom{2n}{n-|\mu|/2}\prod_{j=0}^{n}[(\Delta/\zeta)^2-j^2]^{-1}, & \mu \text{ even},\\ i\dfrac{\Gamma}{\zeta}e^{-i\mu\alpha}\displaystyle\sum_{n=|\mu|+1/2}^{\infty}\left(\frac{\tilde{\varepsilon}_1}{2\zeta}\right)^{2n-1}\binom{2n-1}{n-(|\mu|+1)/2}\prod_{j=1}^{n}\left[(\Delta/\zeta)^2-\left(\frac{2j-1}{2}\right)^2\right]^{-1}, & \mu \text{ odd}.\end{cases} \tag{3}$$

Series (2) and (3) converge in $|ze^{\sqrt{1-z^2}}/(1+\sqrt{1-z^2})|<1$ (in particular within the circle $|z|<0.6627434\ldots$) and for arbitrary $2\Delta/\zeta$ not an even (odd) integer.[12] The terms to a given order in $\tilde{\varepsilon}_1$ summed over μ are the nonlinear susceptibilities. For a fixed μ, the term to a given order in $\tilde{\varepsilon}_1$ describes multi-THz-photon processes of that order contributing to the sideband. 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. Expansion (3) in terms of susceptibilities applies *mutatis mutandis* to the transport case, and to our knowledge was hitherto unknown. As we see below, it provides the key to understanding the phase shifts of the sidebands, and, by analogy, the phase shifts in transport experiments as predicted in Ref. 6.

In the following, we consider the phase of the reflected sideband at frequency ω' as a function of the detuning $\delta=\omega-\varepsilon_0$ between the incident frequency and the exciton line center. As typical values for a very high-quality QW, we take $\gamma_0=1.52\ \text{ps}^{-1}$ (1 meV), $\Gamma=0.1\ \text{ps}^{-1}$, and $\zeta=8\pi\ \text{ps}^{-1}$ ($\zeta/2\pi=4$ THz). For definiteness, we choose $\alpha=0$. We consider the effect of modulating the exciton linewidth as well as the frequency. Previously,[8] we found that this has minimal effect on the amplitude of the sidebands. Our focus here is on the phase which is a unique signature of the coherent, as opposed to the incoherent, nonlinear optical process leading to sidebands.

Figures 1–3 show (a) $A_\mu(\delta)=|R(\omega+\mu\zeta,\omega)|$ and (b) $\varphi_\mu(\delta)=\arg R(\omega+\mu\zeta,\omega)$ for the fundamental $\mu=0$, first $\mu=1$, and second sideband $\mu=2$ as a function of detuning and θ ($=0$, solid line; $\pi/4$, dotted line; and $\pi/2$, dashed line) where $\tilde{\varepsilon}_1=\eta e^{-i\theta}$ and $\eta=4\ \text{ps}^{-1}$. The gross features—the large peaks in $A_\mu(\delta)$ and the jumps in $\varphi_\mu(\delta)$—are due to the first term of Eq. (3); the behavior of the weak replicas is governed by higher-order terms. The total phase shift is thus $\sim\mu\pi$, as δ is swept from below to above the resonance. The θ dependence of $\varphi_0(\delta)$ is thus weak since it is dominated by the leading (i.e., unmodulated) $\tilde{\varepsilon}_1$-independent term. The replicas display a nontrivial θ dependence due to the interference between the lowest- and next-order terms in Eq. (3), and in principle could serve as a sensitive diagnostic of θ. More dramatic, however, is the θ dependence of the overall phase shift of the sideband, which again is determined by the leading term of Eq. (3); clearly, the phase of the sideband is quite sensitive to whether the exciton energy, the linewidth, or some combination is modulated by the THz field. We reiterate that approaches relying on the optical theorem are inadequate to investigate anything but $\theta=0$; and moreover, they miss all the detail associated with the individual channels (sidebands). The sidebands, not the replicas present in the fundamental, contain the clearest signatures of the coherent processes. Moreover, the power series (3) rather than the Kapteyn series (2) as usually given provides the essential insight into the phase shifts.

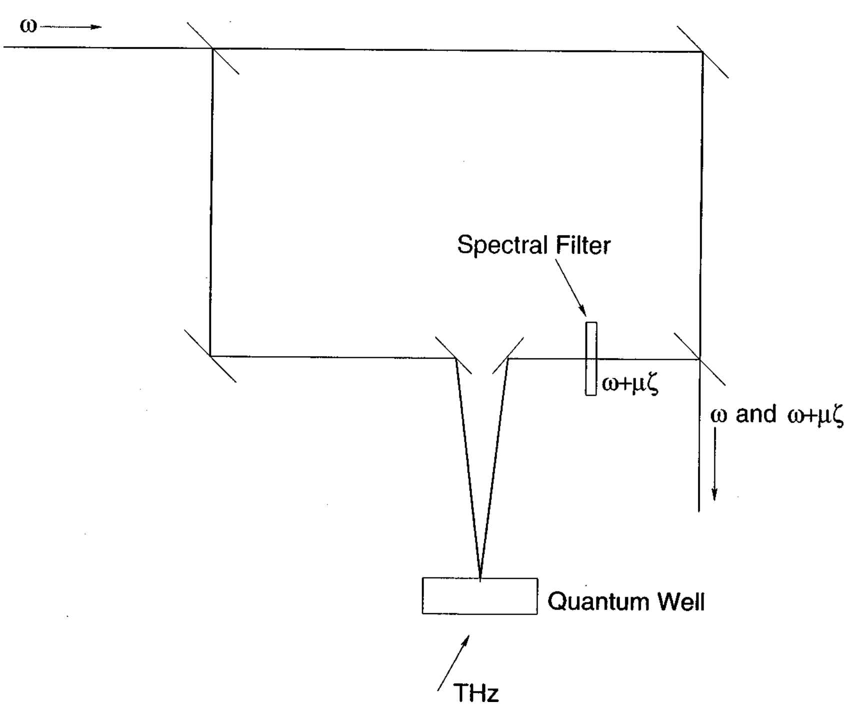

FIG. 4. Interferometer used to measure the phase shift of the THz sideband.

How might these phase shifts be measured? Shown in Fig. 4 is a possible setup consisting of a Mach-Zehnder interferometer in which the THz-modulated QW lies in one arm.[13] A beam I at frequency ω is incident. The reflected beam R from the QW is spectrally filtered to leave a single THz sideband $\omega+\mu\zeta$ propagating through the interferometer. The beams at ω and $\omega+\mu\zeta$ are allowed to interfere producing beats. The phase as a function of ω of the beats with respect to the optical phase of I then gives $\varphi_\mu(\delta)$.

To conclude, we have explored an optical analogue for time-dependent quantum-transport phenomena that allows for an unambiguous differentiation between coherent and incoherent multiphoton processes. In particular, we have shown that the phase of the sidebands, and analogously the phase of the transmitted electrons via the various transport channels, contains a clear signature of whether the modulation is of the energy, the linewidth, or some combination of the two. The treatment presented here is of a single THz-modulated excitonic resonance, though more complicated systems including resonant THz dressing of two exciton levels[8] and realistic band structure[14] can also be included. 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 quantum transport through time-dependent potentials. We believe the area of optical analogues of time-dependent quantum-transport problems—as pioneered by the work reviewed in Ref. 3—is in its infancy, and in particular, exploitation of optical pulse propagation through QW's shows great promise to advance the field materially.

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