PHOTON ECHO IN DIRECT GAP SEMICONDUCTOR

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Using statistical quantum electrodynamics techniques, we investigate photon echo in intrinsic direct transition semiconductor materials. Possible applications of this effect in ultrashort time measurements and optical digital computation are discussed.

In a recent paper [1] the results of photon echo in gases were derived by use of the techniques of statistical quantum electrodynamics. In this note the dynamics of photon echo in a direct gap semiconductor are investigated, where photon absorption is vertical and is accompanied by the generation of an electron–hole pair. The present work is prompted by the recent availability [2] of pulses of width equal to or smaller than the recombination and dephasing times associated with ultrafast semiconductor interband transitions. Photon echo measurements would contribute to a better understanding of semiconductor relaxation processes and to the operation of semiconductors as ultrafast signal processing devices.

In this work, we summarize the results for photon echo in two level atom systems, compute the vertex function for the direct gap semiconductor, the effects on the vertex function of a photonic pulse of arbitrary duration, which result leads to an expression for the form factor, a representation of the wavefunction with a specific history, and the two-pulse photon echo results. Finally, we propose possible applications of the ultrashort photon echo effects in direct gap semiconductor material.

In a two-level atom systems where the energy of the ground state is $\epsilon_p^{\mathrm{g}} = p^2/2m$, that of the excited state is $\epsilon_p^{\mathrm{e}} = \Omega_0 + p^2/2m$ and the transition between the two states is dipolar, the integrated hamiltonian density is given, assuming that the states of the atom and the photon are expanded in plane waves, by

$$\int \mathrm{d}^3x\, \mathrm{d}t\, \mathcal{H}_{\mathrm{int}}(x, t) = N(\widetilde{p} \cdot \hat{\varepsilon}) a^{+}_{\mathrm{e},p+k} a_{\mathrm{g},p} \alpha_k \delta[(p+k)^2/2m - p^2/2m + \Omega_0 - \omega] + \mathrm{h.c.}\,, \quad (1)$$

where $\hat{\varepsilon}$ is the photon polarization, N is a normalization constant, $a^{+}_{\mathrm{e},p}$ is the creation operator for an excited atom of momentum p, the index g refers to the ground state, a_k^{+} is the creation operator for a photon of momentum k and $\omega = |k|$, and p is the transition dipole moment.

If the incoming photonic field is a gaussian pulse of finite duration and a central frequency ω, and with the representation

$$E_{\mathrm{pulse}} = E(t - \hat{k} \cdot x) \exp(-\mathrm{i}\omega t + \mathrm{i}k \cdot x)\,, \quad (2)$$

where

$$E(t) = E_0 \exp[-\tfrac{1}{2}(t/\tau_{\mathrm{c}})^2]\,, \quad \widetilde{E}(u) = \int_{-\infty}^{\infty} \mathrm{d}t\, E(t)\, \mathrm{e}^{-\mathrm{i}ut}\,,$$

then the vertex function is given by:

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$$P(p,k) = (\tilde{p} \cdot \hat{\varepsilon}) a^{+}_{\mathrm{e},p+k} a_{\mathrm{g},p} \alpha_k \widetilde{E}(\omega - \omega_{p,k}) , \tag{3}$$

where

$$\omega_{p,k} = (p+k)^2/2m - p^2/2m + \Omega_0 .$$

The ensemble averaged ground state atom wavefunction is given by

$$\langle \psi_\mathrm{g}(x,t) \rangle = (2\pi)^{-3/2} \int \mathrm{d}^3 p\, A(p) \exp[\mathrm{i} p \cdot x - \mathrm{i}(p^2/2m)t] \,|1\rangle , \tag{4}$$

and that of an excited state, which has been excited by a photon of momentum k_1, in the short pulse limit, is given by:

$$\langle \psi_\mathrm{e}(x,t) \rangle = (2\pi)^{-3/2} \int \mathrm{d}^3 p\, A(p) \exp\{\mathrm{i}(p+k_1) \cdot x - \mathrm{i}[(p+k_1)^2/2m]t - \mathrm{i}\Omega_0 t - \gamma t\} \,|2\rangle , \tag{5}$$

where $A(p) = [f(p)]^{1/2}$, $f(p)$ is the Maxwell distribution, and $\gamma = (2T)^{-1}$ where T is the lifetime of the excited state (dephasing time).

In a two-photon echo experiment, the first excitation is described by (k_1, t_1) and the second excitation is described by (k_2, t_2) where k refers to the incoming photon momentum and t to the time of collision of an infinitely narrow pulse with the system. The echo amplitude is proportional to the matrix element:

$$M = \langle \psi_{\mathrm{g,g,e}} | \tilde{p} \exp(\mathrm{i} k_{\mathrm{echo}} \cdot x) | \psi_{\mathrm{g,e,g}} \rangle , \tag{6}$$

where the indices refer to the history of that particular component of the wavefunction as it develops with time. The first comma refers to time t_1 and the second comma refers to time t_2 as previously defined. In the same infinitely short pulse approximation, the above wavefunctions can be written as:

$$\psi_{\mathrm{g,g,e}} \propto \exp(\mathrm{i}\chi_1) \int \mathrm{d}^3 p\, A(p) \exp[\mathrm{i}(p+k_2) \cdot x] \exp[-\mathrm{i}\epsilon^{\mathrm{g}}_p t_2 - \mathrm{i}\epsilon^{\mathrm{e}}_{p+k_2}(t-t_2) - \gamma(t-t_2)] \,|2\rangle , \tag{7}$$

and

$$\psi_{\mathrm{g,e,g}} \propto \exp(\mathrm{i}\chi_2) \int \mathrm{d}^3 p\, A(p) \exp[\mathrm{i}(p+k_1-k_2) \cdot x]$$

$$\times \exp[-\mathrm{i}\epsilon^{\mathrm{g}}_p t_1 - \mathrm{i}\epsilon^{\mathrm{e}}_{p+k_1}(t_2-t_1) - \mathrm{i}\epsilon^{\mathrm{g}}_{p+k_1-k_2}(t-t_2)] \exp[-\gamma(t_2-t_1)] \,|1\rangle , \tag{8}$$

where $\chi_{1,2}$ is a phase factor that is p independent. Integrating M over x, one obtains for the echo momentum:

$$k_{\mathrm{echo}} = 2k_2 - k_1 , \tag{9}$$

and the echo amplitude is proportional to

$$E_{\mathrm{echo}} \propto \exp(\mathrm{i}\chi) \int \mathrm{d}^3 p\, f(p) \exp[-\gamma(t-t_1)] \exp\{(\mathrm{i}/m) p \cdot [2k_2(t-t_2) - k_1(t-t_1)]\} . \tag{10}$$

This integration over p is maximum when the phase of the oscillating term is zero, i.e.

$$2k_2 \cdot p(t-t_2) = k_1 \cdot p(t-t_1) , \tag{11}$$

which for $k_2 \sim k_1$ reduces to $2(t-t_2) = t - t_1$. Using the notation $k_1 = k\hat{e}_\parallel$, $k_2 = k\hat{e}_\parallel + \beta k\hat{e}_\perp \approx k\hat{e}_\parallel + \theta k\hat{e}_\perp$, and $t_2 - t_1 = \tau$, the echo intensity is given by

$$I = I_0 \exp(-t/T) \exp[-(2k^2 q_0^2 \beta^2/m^2)(t-\tau)^2] \exp[-(k^2 q_0^2/2m^2)(t-2\tau)^2] , \tag{12}$$

indicating that the echo occurs at $t = 2\tau$, and its intensity is exponentially decreasing with τ and the square of the angle between k_1 and k_2. This result was obtained and tested in ref. [3] over a range of 12 decades. If the pulse is of finite width the q_0^2 in the last exponent is changed to q_3^2 where $q_3^{-2} = q_0^{-2} + \frac{3}{2} q_{1/e}^{-2}$ and $q_{1/e} = m(k\tau_{1/e})^{-1}$.

In an intrinsic semiconductor with spherical energy surfaces having extrema located in the center of a Brillouin zone, the effective mass approximation [4] gives for the eigenfunctions and eigenvalues the following forms:

$$\epsilon_p^c = E_g + p^2/2m_c^* , \quad \epsilon_p^v = -p^2/2m_v^* , \quad \psi_p^c(x,t) = L^{-3/2} \exp(ip \cdot x - i\epsilon_p^c t) , \quad \psi_p^v(x,t) = L^{-3/2} \exp(ip \cdot x - i\epsilon_p^v t) , \tag{13}$$

where the superscripts c and v refer respectively to the conduction and valence bands and L is the unit cell dimension.

The distribution function for the electrons is given by the Fermi–Dirac distribution $f_e(E, T)$, and that of the holes by $f_p(E, T) = 1 - f(E, T)$. At room temperature the FD distribution reduces to the Maxwell distribution.

In the effective mass approximation, the hamiltonian in the presence of an electromagnetic field is given by:

$$H = (p - eA)^2/2m_v^* . \tag{14}$$

To first order in A, the interaction hamiltonian is given in the Lorentz gauge $\nabla \cdot A = 0$ by

$$H_{int} = -A \cdot j , \tag{15}$$

where

$$j = -(ie/m_v^*) \nabla .$$

The plane wave representation for the vector potential A is given by

$$A(x, t) = A_0 \exp[i(\omega t - k \cdot x)] , \tag{16}$$

and the interaction hamiltonian is given by

$$H_{int} = (ie/m_v^*) \exp[i(\omega t - k \cdot x)](A_0 \cdot \nabla) , \tag{17}$$

its matrix element between a state p_1 and in the valence band and a state p_2 in the conduction band is given by

$$\int \psi_{p_1}^{v*}(x,t) H_{int} \psi_{p_2}^c(x,t)\, d\tau = -(e/m_v^*)(A_0 \cdot p_2) \exp[i(\epsilon_{p_1}^v - \epsilon_{p_2}^c + \omega)t]\, \delta_{p_1+k,p_2} . \tag{18}$$

Given that $\langle p_1^2 \rangle \sim (2m^*kT)$ and $k_1 \sim E_g$, at $T = 300$ K, $m^* \sim 10^{-27}$ g and $E_g \sim 1$ eV, $p \sim 10^7$ cm^{-1} and $k \sim 6 \times 10^4$ cm^{-1} i.e. $k \ll p_1$ we obtain $p_1 \sim p_2$. The time phase factor reduces to

$$\epsilon_{p_1}^v - \epsilon_{p_2}^c + \omega \sim -p^2/2m_v^* - p^2/2m_c^* - E_g + \omega \sim \omega - E_g - p^2/2m_{red}^* , \tag{19}$$

where

$$m_{red}^* = m_c^* m_v^*/(m_c^* + m_v^*) .$$

The probability of transition in a unit of time corresponding to the above matrix element is

$$W(p_1, v; p_2, c) = \delta_{p_1,p_2} (2\pi e^2/m_v^{*2})(A_0 \cdot p_2)^2 \delta(\omega - E_g - p^2/2m_{red}^*) , \tag{20}$$

and the number of photons absorbed in a unit of time t is

$$\rho = \iint f(\epsilon_{p_1}^v) f_p(\epsilon_{p_2}^c) W(p_1, v; p_2, c)\, dp_1\, dp_2 , \tag{21}$$

which leads in the low temperature limit to an absorption coefficient (5):

$$\alpha(\omega) = (2/3\pi^3)(e^2/\omega)[(2m_{red}^*)^{5/2}/m_v^{*2} n](\omega - E_g)^{3/2} . \tag{22}$$

We have expressed the photons flux by $(A_0 \omega^2/8\pi n)$, where n is the background index of refraction. The above expression for the absorption coefficient refers to the so-called forbidden transition, the expression for $\alpha(\omega)$ corresponding to the allowed transition is

$$\alpha(\omega) = (2e^2/\pi^3 m_e^2)(\omega n)^{-1} |P_{nn'}(0)|^2 (2m^*_{red})^{3/2} (\omega - E_g)^{1/2} \,, \tag{23}$$

where

$$P_{nn'}(p_2) = \int \varphi^*_{n,p_2} [\hat{\varepsilon} \cdot (-\mathrm{i}\nabla + p_2)] \varphi_{n',p_2} \,,$$

$\hat{\varepsilon}$ is the photon polarization and the Bloch function is $\psi_{n,p} = \exp(\mathrm{i}p \cdot x)\phi_{n,p}(x)$.

The vertex function associated with the impulse function given by eq. (2) is then given, in the approximation of eq. (19), by

$$P(p_1, k) = \int \mathrm{d}t\, (-e/m_v^*) p_2 \cdot A_0 \exp(\mathrm{i}\omega t) \exp[-\tfrac{1}{2}(t/\tau_c)^2] \exp[\mathrm{i}(\epsilon^v_{p_1} - \epsilon^c_{p_2})t]\, \delta_{p_1,p_2}$$

$$= -(e/m_v^*)(p_2 \cdot A_0) \widetilde{E}(y) \delta_{p_1,p_2} \,, \tag{24}$$

where

$$y = -\omega + p_1^2/2m^*_{red} + E_g$$

and

$$\widetilde{E}(y) = (2\pi\tau^2_{1/e})^{1/2} \exp(-y^2 \tau^2_{1/e}/2) \,.$$

(Notice that at resonance $\omega = E_g$, y is quadratic in p while the corresponding dependence for a two-level system is linear in p.) To understand the physical meaning of the form factor, notice that to a pulse centered at ω and of duration τ, there corresponds in the Fourier domain a band of width $\sim\tau^{-1}$. The form factor measures the effectiveness of the interaction of each frequency component, its magnitude is non-zero only within the energy band corresponding to τ^{-1}, as specifically exhibited in eq. (24). The above considerations also lead to bounds for the width and energy of a pulse that is suitable for pumping a direct gap intrinsic semiconductor. The condition for the availability of empty states in the conduction band can be written as:

$$2 \int_{E_g}^{E_g+\Delta E} N(E)\, \mathrm{d}E > n(T) \,, \tag{25}$$

where $N(E)$ is the density of states in the conduction band, $n(T)$ is the number of thermally excited electrons in this band, and ΔE is the energy sub-band in the conduction band corresponding to τ^{-1}. This condition translates into

$$\tau < (kT)^{-1} \exp(E_g/3kT) \tag{26}$$

at room temperature and $E_g \sim 1$ eV, $\tau < 1$ ns (this condition will be superseded in photon echo experiment by the recombination and dephasing times $\sim 10^{-12}$ s). The minimum energy for the pulse is given by:

$$\mathcal{E}(\text{pulse}) > N_c \exp(-E_g/2kT)\, \omega \Delta v \,, \tag{27}$$

where $N_c = 2(2\pi m_c^* kT)^{3/2}$, ω is the photon frequency and Δv is the effective volume in cc of light–semiconductor interaction. (Typical number is $\mathcal{E}$ (pulse) $> [10^{-10}(\Delta v)]$ J.)

Next, let us examine the two-photon echo experiment. Denote the wavefunctions corresponding in the two-level atom system to $\psi_{g,g,e}$ and $\psi_{g,e,g}$ by respectively $\psi_{v,v,c}$ and $\psi_{v,c,v}$. In these wavefunctions, the quantities corresponding to ϵ^g_p and ϵ^e_p are respectively ϵ^v_p and ϵ^c_p, and the lifetime corresponds to the electron–hole recombination time. From the x integration we obtain the wave-vectors phase matching condition:

$$k_{echo} = 2k_2 - k_1 \,. \tag{28}$$

The p-dependent phase factor of the matrix element can be written as

$$\chi(p) = ap^2 + bp \cdot k_1 + cp \cdot k_2 , \tag{29}$$

where

$$a = (1/2m^*_{\text{red}})(t - 2t_2 + t_1) , \quad b = -(t_2/m^*_{\text{red}} - t_1/m^*_{\text{c}} - t/m^*_{\text{v}}) , \quad c = (t - t_2)(1/m^*_{\text{c}} - 1/m^*_{\text{v}}) .$$

This phase factor is zero for

$$k_1 \sim k_2 \quad \text{and} \quad t = (2t_2 - t_1) , \tag{30}$$

i.e., the echo amplitude is maximum for $t = 2\tau$. The echo amplitude can be specifically obtained by choosing a system of axes, with the z-component corresponding to the perpendicular to the plane (k_1, k_2), and the x-axis to k_1. In this coordinate system, the p-dependent phase factor is given by:

$$\chi(p) = ap^2 + dp_{\parallel}k\hat{e}_{\parallel} + fp_{\perp}\beta k\hat{e}_{\perp} , \tag{31}$$

where

$$k_1 = k\hat{e}_{\parallel} , \quad k_2 = k\hat{e}_{\parallel} + \beta k\hat{e}_{\perp} , \quad d = (1/m^*_{\text{c}})(t + t_1 - 2t_2) , \quad f = (t - t_2)(1/m^*_{\text{c}} - 1/m^*_{\text{v}}) .$$

The echo intensity is

$$I = I_0 \exp(-t/T) G^2(t - 2\tau) \exp[-\eta\theta^2(t - \tau)^2] , \tag{32}$$

where $G(t - 2\tau)$ is a function peaked at $t = 2\tau$, θ is the angle between k_1 and k_2, and η is a function of the effective masses and temperature. The echo as indicated in eq. (28) is emitted at an angle 2θ.

Potential applications of the above photon echo effect might be found in ultrashort time measurements and optical digital computation [6]. Specifically, (1) to measure time differences between pulses in the range 10^{-14}–10^{-13} s such as in differential measurements due to a variation of the index of refraction of a medium, rotation of a medium, etc., it is sufficient to measure the intensity of the echo generated by the incoming two pulses, $I \propto \exp(-2\tau/T)$. This technique applied, for example, to measuring the time delay between two pulses propagating in two similar (1 km) optical fibers and whereby one of the fibers is subjected to an external field that changes its index of refraction, could detect variation in the index of refraction of the order of one part in 10^9. (2) In optical digital computation, the following parameters are key in determining the suitability of a specific system: fast decay of the component excitation, low energy requirements and an inherent algorithm in the physical process under consideration. In a potential system built around the photon echo effect in semiconductors, (i) the decay time 10^{-12} s i.e. the possibility of 10^{12} operations per second (ii) the power dissipation (from eq. 27) is (for $\Delta v \sim 10^{-5}$ cm^3) 10^{12} s^{-1} $\times$ 10^{-15} J $\sim$ 1 mW (iii) the wave vectors phase matching condition $k_{\text{echo}} = 2k_2 - k_1$ provide a suitable multiplication table, specifically, if the input signals (pulses) are incoming along two directions such that $(k_1, k_2) = \theta$, the echo is outgoing at an angle 2θ and no echo appears unless both signals are present i.e. (0,0) = 0, (1,0) = 0, (0,1) = 0 and (1,1) = 1. The *not* operation is performed by an optical switch. The physical configuration for the *not* logic element is as follows: a reference pulse and the input signal pulse are incident on the switch. In the presence of the input signal, the reference pulse is absorbed, otherwise the reference pulse goes through. If the input signal to this device is the echo signal $(A \cap B)$ the output signal will then be $(\overline{A \cap B})$ i.e. the NAND logical function, which is a canonical operation from which all other logical functions can be obtained, is realized. A possible physical realization of the *not* optical switch compatible with the above decay times and energy requirements is the 2γ-switch [7]. Absorption in this switch occurs via a two-photon process. For intensities larger than a critical value, the number of photons transmitted is equal to the number of photons in the reference pulse minus the number of photons in the echo pulse. Given that the echo pulse intensity is lower than the reference pulse intensity, for maximum contrast it is necessary to amplify the echo pulse, a possible scheme is passing this photon through a semiconductor amplifier [8] (modelled after the dye amplifiers). To construct a simple memory device from NAND functions one cross couples two NAND gates to form a set–reset flip–flop as shown in fig. 1. In normal

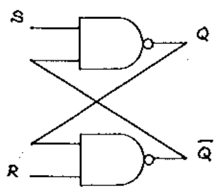

Fig. 1. Set–reset flip–flop circuit.

electronic logic this device has as its quiescent state an input of $R = 1$ and $S = 1$. An input of $S = 0$ (while R is still 1) will cause Q to be set to 1 and similarly an input of $R = 0$ (while $S = 1$) will cause $\bar{Q}$ to be set to 1. $R = S = 0$ is unstable, and thus is an unallowed state. The NAND gate formed by the photon echo effect together with the optical switch requires for its operation several sequential events namely the incidence of the two input pulses to form the AND function, the resultant echo pulse must then propagate through the amplifier, following which the reference pulse and the amplified echo are incident on the switch. Finally time must pass so that the switch may recover (1 ps). The total time associated with the complete sequence is of the order of 1 ps (which corresponds to an optical path length of 0.3 mm). Therefore, in order that an event propagates to the other NAND gate of this flip–flop, allowance for travel time over the optical path length must be incorporated into the design. But, this suggest that the entire flip–flop can be made with only *one* physical NAND gate with an optical feedback path length of the order of 1 ns (i.e. 30 cm) and allow the single gate to function half the time on the upper gate and half the time on the lower gate of the flip–flop. Thus with the above numbers, 500 bits could be held with a single gate configuration. Naturally since we are dealing with optical phenomena in finite size aperture, many bits can be processed in parallel.

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