Tunneling assisted two-photon absorption: The nonlinear Franz-Keldysh effect

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The N-photon absorption coefficient of a direct gap semiconductor under the influence of a uniform electric field perpendicular to the layer and neglecting excitonic effects is calculated. For $N=2$ we recover the familiar results for the low-field regime. The system also shows the same universal behavior as for the $N=1$ case. Below the $E_g/2$ edge the absorption coefficient develops the exponential tail characteristic of $N=1$, but with a different photon energy and electric field dependency while above the $E_g/2$ edge Franz-Keldysh oscillations with strong damping features are present. An analytical expression for the tunneling assisted two-photon absorption coefficient is given. Our discussion gives suggestions for the use of the effect in the nonlinear regime to characterize the critical point of otherwise forbidden transitions in semiconductors.

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I. INTRODUCTION

Systematic studies on the effect of a dc electric field in the optical absorption of semiconductors in the nonlinear regime are scarce, and only few papers have touched on the contribution of strong electric fields on the nonlinear susceptibility, in particular in the imaginary part of the susceptibility that is related to the absorption coefficient. In this paper, we calculate the nonlinear absorption coefficient for the case $N \geqslant 1$ of a semiconductor in a strong dc electric field parallel to the propagation direction of the optical field, and give expressions for $N=2$ in the strong dc-field regime above and below the $E_g/2$ edge. We show that the Franz-Keldysh[1,2] effect possesses a universal behavior that depends on a modified energy gap given by E_g/N, where E_g is the energy gap of the semiconductor, and N in the number of photons absorbed in the process. We propose to extend the modulation spectroscopy technique to the nonlinear regime because of the universal behavior of the Franz-Keldysh effect

The effect of a dc electric field on the absorption coefficient of solids in the linear regime and particularly in semiconductors has been extensively studied.[3–8] The main characteristics of the effect includes the appearance of an exponential tail below the band edge reminiscent of the tunneling effect and the development of an oscillatory behavior (Franz-Keldysh oscillations) above the fundamental absorption edge E_g.[1,2] Later work noted that any full description of such an interband optical absorption should include the Coulomb interaction of the electron and hole.[9,10] The consequences of the interaction are an enhancement of the optical absorption above the optical band gap with no applied field, and a broadening and small shift to lower photon energy of the resonance with applied field. Although this shift is relatively small at low applied fields ($\leqslant 10\%$ of the exciton binding energy), the theory provides a relatively full description of electroabsorption, including excitonic effects, in bulk semiconductors. This complex phenomenon has been used to characterize the band structure and to identify critical points in semiconductors.[11,12] The technique, known as "modulation spectroscopy," uses the fact that the spectral shape of the differential reflectance is proportional to the third derivative of the primary spectrum with respect to the photon energy E.[13]

In the high-intensity optical field regime with no dc fields, multiphoton absorption has been studied extensively.[14,15] In particular, two- and three-photon absorption have received most of the attention for their role as a limiting factor in photonics applications, particularly in photonic switching. Several approaches have been used in the calculation of the nonlinear absorption coefficient using either perturbation theory or dressed state calculations. However, none of them have included the effect of dc fields in the nonlinear absorption even though some of them have mentioned the contribution of dc terms or quadrupole contributions to the nonlinear absorption that results in a dc and ac Stark effect, depending on the transitions involved[16] when nondegenerate two-photon processes are considered. This is not the case here, because our calculation addresses the fully degenerate case.

One of the many applications of two-photon absorption is in spectroscopy. The use of two-photon absorption for spectroscopy studies has advantages over the single-photon process because of the restriction on selection rules; e.g., when one photon absorption is forbidden by selection rules, a higher-order absorption process may be allowed.[17] Another advantage of two-photon absorption spectroscopy is the ability to study the properties truly characteristic of the crystalline volume because of the small values of the nonlinear absorption coefficient.

In Sec. II of this paper, we will present the formal theory of multiphoton electroabsorption in a bulk semiconductor, neglecting Coulomb interaction in a linearly polarized case, with an electric field applied in the direction parallel to the propagation of the optical field. In Sec. III, we will present illustrative examples for the $N=1$ and $N=2$ cases, and give analytical expressions for the low and high dc field regimes below and above the normalized E_g/N band gap for $N=2$. We will present our conclusions in Sec. IV.

II. NONLINEAR ELECTROABSORPTION

The general problem here is one in which a charged particle, in this case an electron initially bound in the valence band, experiences a transition to the conduction band

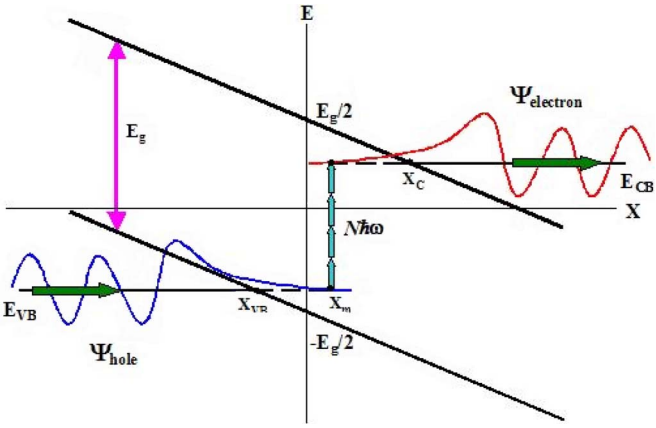

FIG. 1. (Color online) N-photon assisted tunneling in an electric field perpendicular to the layer. The sloping lines represent the valence- and conduction-band edges. A valence-band electron of energy E_{VB} tunnels from its turning point X_{VB} to X_m, absorbs N photons, and tunnels to X_{CB}, the turning point for a conduction electron of energy $E_{CB}=E_{VB}+N\hbar\omega$.

through the action of an intense plane-wave electromagnetic field as is shown in Fig. 1.

The S matrix to describe the transition can be written in general as

$$S_{fi} = \lim_{t\to-\infty} (\Psi_f^{(-)}, \Phi_i), \tag{1}$$

where $\Psi_f^{(-)}$ is the final state of the system containing the effect of the electromagnetic field and the crystal lattice potential, while Φ_i is the initial state of the unperturbed system with no field present. An electron in a solid in the effective-mass approximation has properties which resemble those of free electrons.[14,15,18] This is particularly true for conduction-band electrons, which are not strongly localized to the atomic sites of the crystal. Therefore it is assumed that after the transition the conduction electron can be represented by a Volkov-type wave function[19] whose energy will be completely determined by the electromagnetic (EM) field. We presume that the motion in the plane perpendicular to the EM-field propagation is still described by plane-wave approximation. Our theoretical treatment adopts Keldysh's view in this regard with one modification: the effect of the dc electric field on the hole and electron wave functions can be expressed as

$$\Psi_{k,n}^{c,v}(r,t) = U_k^{c,v} e^{ik\cdot\rho} \varphi_n(z) e^{-(i/\hbar)\int_0^t E_{c,v}(k)d\tau}, \tag{2}$$

where $U_k^{c,v}$ is the usual Bloch wave function which has the same periodicity as the lattice. As we mentioned before, the effect of the optical field is only to modify the energy of the electron in the final state of the system. It is worth mentioning that the dc-field direction only enters through the reduced mass in the direction of the dc field and will only affect the final result by a numerical factor but with the same functional form. The envelope function $\varphi_n(z)$ is given by

$$\varphi_n(z) = a_n^{c,v} Ai(\xi_{c,v}), \tag{3}$$

where

$$\xi_{c,v} = -\left(\frac{2m_{c,v}}{(e\hbar F)^2}\right)^{1/3} (E_{e,h} \mp eFz)$$

is a solution of the Schrödinger equation in the effective mass approximation,

$$\left[-\frac{\hbar^2}{2m_{c,v}}\frac{d^2}{dz} \pm eFz\right]\varphi_n(z) = E_{e,h}\varphi_n(z), \tag{4}$$

where $E_{e,h}$ are the energy of the electron and hole from their respective band edges, $Ai(z)$ are Airy functions,[20] m_c and m_v correspond respectively to the electron and hole effective masses, and the a's are normalization constants given by

$$(a_n^{c,v})^{-2} = \lim_{L\to\infty} \frac{1}{\pi}\sqrt{\frac{L}{f_{c,v}^{1/3}}}, \tag{5}$$

where L is the slab thickness and

$$f_{c,v} = \frac{2em_{c,v}F}{\hbar^2},$$

where F is the dc electric field. The Hamiltonian of the electron in the optical field can be expressed as

$$H = \frac{1}{2m_o}\left(p + \frac{eA}{c}\right)^2 + V(r),$$

where p is the electron momentum, e is the charge of the electron, m_o is the free electron mass, A is the vector potential, and $V(r)$ is the potential energy of the electron in the solid. Therefore, the interaction Hamiltonian can be written as

$$H_{\text{int}} = -\frac{e}{2m_o c}(p\cdot A + A\cdot p) + \frac{e^2A^2}{2m_o c^2},$$

where the last term can be ignored because it is small compared to the other two terms. In the Coulomb gauge $\nabla\cdot A = 0$, and the first two terms commute, hence the interaction Hamiltonian for the system can be expressed in the dipole approximation as[21,22]

$$H_{\text{int}} = -\left(\frac{e}{m_o c}\right)A\cdot p; \quad A = \hat{z}A_o \cos\omega t. \tag{6}$$

It should be noted that the $(eA)^2$ term nominally in the H_{int} does not make any contributions to the transition rate in the dipole approximation because even though it contains the ordinary matrix element connecting the valence band and the conduction band, it must be remembered that it represents an electric quadrupole contribution to the transition rate and it would be inconsistent to include it with the pure dipole rates. Additionally it can be removed through a contact transformation.[17,18,23] The effect of the optical field is to alter the energy of the electrons and holes in the final and initial states, respectively. The energies for the hole in the valence band, and for the electron in the conduction band, are given by[19,22]

$$E_c = \frac{\hbar^2}{2m_c}k_\perp^2 + E_e - \frac{e\hbar}{m_c c}k\cdot A + E_g,$$

$$E_v = E_h - \frac{\hbar^2}{2m_v}k_\perp^2, \tag{7}$$

where A is the vector potential of the propagating optical field, and $k_\perp$ is the momentum of the electron and hole perpendicular to the dc field and optical field. The interaction term in Eq. (7) is obtained as follows: in the Coulomb gauge the scalar potential is zero and the electric field is given by $E=-\partial A/\partial t$. Therefore Eq. (7) can be transformed in the following way:

$$[p^2,x] = 2i\hbar p,$$

and using the above result, we get

$$\langle A|p|B\rangle = \left\langle A\left|\frac{im}{\hbar}[H_o,x]\right|B\right\rangle = \frac{im(E_A-E_B)}{\hbar}\langle A|x|B\rangle = -im\omega x_{AB},$$

and this is equivalent to the direct interaction Hamiltonian provided that we replace

$$-\frac{eA\cdot p}{mc} \rightarrow \frac{e}{c}x\cdot\frac{\partial A}{\partial t} = -ex\cdot E$$

Now we prove that the contribution from this term to the energy of the electron in the conduction band is equal to the $k\cdot A$ term in Eq. (6). The fundamental assumption in the Volkov approximation is that the optical field only modifies the final energy of the electrons in the conduction band. The energy of the system in first-order perturbation theory is

$$\langle nk|H_{\text{int}}|nk\rangle = -\frac{e}{2m}[2mA\cdot v_n(k)] = -eA\cdot v_n(k),$$

where $v_n(k)$ is the group velocity associated with the state k in the band n. For parabolic bands the group velocity is given by $v_n(k)=\hbar k/m_c$, and the contribution of the optical field to the final energy of the electron in the conduction band is

$$\langle nk|H_{\text{int}}|nk\rangle = -\frac{e\hbar}{m_c c}k\cdot A.$$

The transition rate can be expressed using Eqs. (2), (3), and (6) in Eq. (1),

$$S = -\frac{i}{\hbar}\int dtd^3x U_k^c(k)e^{-ik\cdot\rho}\varphi_n^*(z)e^{(i/\hbar)\int_0^t E_c(k)d\tau}A\cdot p \times\left(\frac{-e}{mc}\right)e^{ik'\cdot\rho}U_{k'}^v(k')\varphi_{n'}(z)e^{-(i/\hbar)\int_0^t E_v(k')d\tau}. \tag{8}$$

Grouping all the terms with spatial dependency, and simplifying it in the spirit of the slow varying envelope approximation, we obtain

$$I_{eh}(E_e,E_h) = \left(-\frac{eA_o}{mc}\right)P_{vc}\delta_{k_\perp k'_\perp}\int_{-\infty}^{\infty}\varphi_n^*(z)\varphi_{n'}(z)dz, \tag{9}$$

where P_{vc} is the interband momentum matrix element. We have made the assumption of a direct allowed transition with negligible photon momentum and an interband matrix element P_{vc} independent of k. The last integral can easily be evaluated using properties of the Airy functions[4] as

$$I_{eh}(E_e,E_h) = \left(-\frac{eA_o}{mc}\right)P_{vc}\delta_{k_\perp k'_\perp}\left(\frac{\hbar^2}{2MeF}\right)^{1/3}Ai \times\left\{-\left(\frac{2m_\mu eF}{eF}\right)^{1/3}\left[\frac{E_e+E_h}{eF}\right]\right\}a_n^c a_{n'}^v. \tag{10}$$

In the above equation, $M=m_c+m_v$, and m_μ the reduced mass given by

$$\frac{1}{m_\mu} = \frac{1}{m_c} + \frac{1}{m_v}.$$

Redefining the argument of the Airy function as δ_n, substituting Eq. (10) into Eq. (8) and with the aid of the series representation of the function $\cos(\omega t)e^{i\alpha\sin(\omega t)}$ in terms of Bessel functions, we get

$$S = -\frac{i}{\hbar}\left(-\frac{eA_o}{mc}\right)\left(\frac{m_c\omega c}{ek\cdot\hat{z}A_o}\right)\left(\frac{\hbar^2}{2MeF}\right)^{1/3}P_{vc} \times\int_{-\infty}^{\infty}e^{(i/\hbar)(E_{k_\perp}^c+E_{k'_\perp}^v+E_e-E_h+E_g)t}\delta_{k_\perp k'_\perp} \times\sum_{n=-\infty}^{n=\infty}nJ_n\left(\frac{ek\cdot\hat{z}A_o}{m_c\omega c}\right)e^{in\omega t}Ai(\delta_n)a_n^c a_{n'}^v dt. \tag{11}$$

Using the integral representation of the delta function and simplifying, we arrive at the final value for the S matrix,

$$S = \left(\frac{2\pi i P_{vc}m_c\omega}{mk\cdot\hat{z}}\right)\left(\frac{\hbar^2}{2MFe}\right)^{1/3}\sum_{-\infty}^{\infty}nJ_n\left(\frac{ek\cdot\hat{z}A_o}{m_c\omega c}\right) \times\delta(E_{k_\perp}^c + E_{k'_\perp}^v + E_e - E_h + E_g - n\hbar\omega)Ai(\delta_n)a_n^c a_{n'}^v. \tag{12}$$

Using the following property of the delta function:[21]

$$|\delta(E-E')|^2 = \frac{T}{2\pi\hbar}\delta(E-E').$$

The transition probability per unit time for the N-photon absorption can be calculated from the S matrix as

$$W^{(N)}(k) = \lim_{T\rightarrow\infty}\frac{1}{T}|S|^2 = \frac{|P_{vc}|^2}{2\pi\hbar}\left(\frac{2\pi m_c\omega N}{mk\cdot\hat{z}}\right)^2\left(\frac{\hbar^2}{2MFe}\right)^{2/3} \times\left|J_N\left(\frac{ek\cdot\hat{z}A_o}{m_c\omega c}\right)\right|^2[Ai(\delta_n)a_n^c a_{n'}^v]^2 \times\delta(E_{k_\perp}^c + E_{k'_\perp}^v + E_e - E_h + E_g - N\hbar\omega). \tag{13}$$

The transition rate (pert unit of volume) can be expressed as

$$W^{(N)} = \frac{1}{(2\pi)^2}\sum_n \sum_{n'} \int dk_{\parallel} W^{(N)}(k), \tag{14}$$

where we are summing over the electron-hole energy system. We can change the summation over the electron-hole energy to an integral over the density of states using the fact that

$$n = \frac{2}{3\pi\hbar}(2m_{cv}eF)^{1/2}\left[\frac{L}{2} + \frac{E_{cv,n}}{eF}\right]^{3/2} + \frac{1}{4}, \tag{15}$$

so that the density of states in energy becomes

$$\frac{dn}{dE_{cv,n}} \cong \frac{1}{\pi\hbar}\left[\frac{m_{cv}L}{eF}\right]^{1/2} \tag{16}$$

as $L \to \infty$, and the transition rate becomes

$$W^{(N)} = \frac{1}{(2\pi)^2}|P_{vc}|^2 N^2 \left(\frac{2\pi m_c \omega}{m}\right)^2 \left(\frac{\pi^2}{2\pi\hbar}\right)\frac{(f_c f_v)^{1/6}}{\hbar^2}\left(\frac{m_c m_v}{(eF)^2}\right)^{1/2} \times \left(\frac{\hbar^2}{2MeF}\right)^{1/3} \int dk_{\parallel} \frac{(eF)}{(k\cdot\hat{z})^2}\left|J_N\left(\frac{ek\cdot\hat{z}A_o}{m_c\omega c}\right)\right|^2 \times \left|Ai\left[\left(\frac{2m_\mu eF}{\hbar^2(eF)^3}\right)^{1/3}\left(\frac{\hbar^2 k_{\parallel}^2}{2m_\mu} + E_g - N\hbar\omega\right)\right]\right|^2. \tag{17}$$

To simplify the above expression, we used the lowest order MacLaurin expansion term for the Bessel function $J_n(x) \approx x^n/2^n n!$ for the case where $x \ll 1$. The amplitude of the vector potential is related to the intensity in cgs units as $A_o^2 = 8\pi cI/n\omega^2$, where n is the linear index of refraction and I is the irradiance of the optical field. In a solid, the electron momentum is of the order of $k \approx \pi/a_o$ where a_o is the unit cell dimension $a_o \sim 10^{-10}$ m, $\omega = 1.77 \times 10^{15}$ s^{-1}, and using $n = 2.34$ for CdS. Using the previous values for $x \ll 1$, we find that the approximation is valid for optical field intensities $I \ll 9.9 \times 10^4$ W/cm^2. At this power level, it is practically possible to observe nonlinear effects as this corresponds to a typical Nd:YAG (yttrium aluminum garnet) laser operating at $\lambda = 1.06$ μm, with a pulse width of 50 ps, repetition rate of the order of 100 MHz with average power levels of less that 10 mW. The N-photon absorption coefficient can be expressed finally as

$$\beta^{(N)}(\omega) = \left(\frac{2N\hbar\omega}{I^N}\right)W^{(N)}, \tag{18}$$

where I is the intensity of the optical field. Using the above formalism, we find after a long calculation the N-photon absorption coefficient in the presence of a dc electric field to be

$$\beta^{(N)} = \frac{\alpha_b f^{1/3}}{2^{2N-1}\pi}\left[\frac{8\pi e^2 E_\mu}{nm_c^2 c\omega^4}\right]^{N-1}\left[\frac{N(2N-3)!!}{[(N-1)!]^3}\right]\left(\frac{m_\mu}{\hbar^2}\right)^N \times \int_{\varepsilon_o^{(N)}}^{\infty}(\varepsilon - \varepsilon_o^{(N)})^{N-1}|Ai(\varepsilon)|^2 d\varepsilon, \tag{19}$$

where $E_\mu = (\hbar^2 e^2 F^2/2m_\mu)^{1/3}$ is the characteristic energy of the dc electric field, m_μ is the reduced mass, n is the semiconductor index of refraction, and f, α_b, and $\varepsilon_o^{(N)}$ are given by

$$f = \frac{2eFm_\mu}{\hbar^2},$$

$$\alpha_b = \frac{8\pi^2}{nc}\frac{|P_{vc}|^2 e^2}{m_o^2\omega},$$

$$\varepsilon_o^{(N)} = \frac{E_g - N\hbar\omega}{E_\mu}.$$

Equation (19) is the final result, the Nth-order absorption coefficient, which is given as a integral equation over the Airy function convoluted with an energy argument that depends strongly on the characteristic energy of the system E_μ. One thing that should be mentioned is the fact that the effect of the electric field in the N-photon absorption coefficient is to replace the delta function obtained in the zero-field expression by a function of unit area but of finite width. As Aspnes[12] points out, the electric field averages or convolutes the zero-field structure in the absorption coefficient and, of course, in the dielectric constant.

III. ONE- AND TWO-PHOTON ABSORPTION

In this section we will use the above equation for $N=1$ and $N=2$. We use these two cases because the result is well known in the zero-field limit. Our objective is to show that in the zero-field limit the results should be equal. We will show for the case of two photons that the system shows the same general behavior of the linear Franz-Keldysh effect. This characteristic gives the effect an universal character. This universal character will be used to study some mathematical consequences in future publications.

To verify the validity of Eq. (19), we calculate the electroabsorption coefficient for $N=1$. In this particular case, we get

$$\beta^{(1)}(\omega) = \frac{\alpha_b}{2\pi}f^{1/3}\left(\frac{m_\mu}{\hbar^2}\right)\int_{\varepsilon_0}^{\infty}|Ai(\varepsilon)|^2 d\varepsilon. \tag{20}$$

Using integral properties of the Airy functions,[24] this becomes

$$\beta^{(1)} = \frac{\alpha_b m_\mu f^{1/3}}{2\pi\hbar^2}\{-\varepsilon_0^{(1)}Ai^2(\varepsilon_o^{(1)}) + [Ai'(\varepsilon_o^{(1)})]^2\}. \tag{21}$$

This is the well-known result for the linear Franz-Keldysh effect. Equation (21) possesses two limits. One is below the band edge and large field limit, in which case Eq. (21) gives the well-known result for Zener tunneling. The appearance of an exponential tail can be understood as a photon-assisted field-induced tunneling. Above the band edge and for low field limits, the free carrier absorption coefficient is recovered.

Next we take the case $N=2$. In this case, Eq. (19) becomes

$$\beta^{(2)} = \frac{2\alpha_b f^{1/3}}{8\pi}\left[\frac{8\pi e^2 E_\mu}{n m_c^2 c \omega^4}\right]\left[\frac{m_\mu}{\hbar^2}\right]^2 \int_{\varepsilon_o^{(2)}}^{\infty} (\varepsilon - \varepsilon_o^{(2)})|Ai(\varepsilon)|^2 d\varepsilon. \tag{22}$$

The integral can be expressed as two integrals with solutions given by[24]

$$\int x|Ai(x)|^2 dx = \frac{1}{3}[Ai'(x)Ai(x) - x|Ai'(x)|^2 + x^2|Ai(x)|^2],$$

$$\int |Ai(x)|^2 dx = x|Ai(x)|^2 - |Ai'(x)|^2. \tag{23}$$

Expressing the dipole matrix elements in terms of E_g obtained by Ashkinadze *et al.*,[25] $P_{vc}^2/m_o^2 = 3E_g/4m_\mu$, based on a Hartree-Fock calculation, we find that the two-photon absorption coefficient for a direct-gap semiconductor in the presence of a strong dc electric field is given by

$$\beta^{(2)}(\omega, F) = \left(\frac{5\pi}{2\sqrt{3}}\right)\frac{K(E_p E_g E_\mu)^{1/2}}{n^2(2\hbar\omega)^5}\{2[\varepsilon_o^2 Ai^2(\varepsilon_o) - \varepsilon_o|Ai'(\varepsilon_o)|^2] - Ai(\varepsilon_o)Ai'(\varepsilon_o)\}, \tag{24}$$

where K is a material-independent constant,

$$K = \frac{2^9\pi}{5}\frac{e^4}{\sqrt{m_o c^2}},$$

which has a value of $K=1940$, $E_p = 2|P_{vc}|^2/m_o \approx 21$ eV for most semiconductors, and $\varepsilon_o = (E_g - 2\hbar\omega)/E_\mu$. Using the asymptotic expansion for the Airy function in the limit of large negative and positive arguments given by Ref. 20,

$$\lim_{x\to\infty} Ai(-x) = \frac{1}{\sqrt{\pi}x^{1/4}}\sin\left(\frac{2}{3}x^{3/2} + \frac{\pi}{4}\right),$$

$$\lim_{x\to\infty} Ai'(-x) = \frac{x^{1/4}}{\sqrt{\pi}}\cos\left(\frac{2}{3}x^{3/2} + \frac{\pi}{4}\right),$$

$$\lim_{x\to\infty} Ai(x) = \frac{1}{2\sqrt{\pi}x^{1/4}}e^{-(2/3)x^{3/2}}\left(1 - \frac{3c_1}{2x^{3/2}}\right),$$

$$\lim_{x\to\infty} Ai'(x) = \frac{x^{1/4}}{2\sqrt{\pi}}e^{-(2/3)x^{3/2}}\left(1 - \frac{21c_1}{10x^{3/2}}\right), \tag{25}$$

where $c_1 = 15/216$. Using Eq. (10), we recover the familiar two-photon absorption coefficient in the absence of the dc field[19] for large negative arguments ($2\eta\omega \gg E_g$), given by

$$\beta^{(2)}(\omega) = \left(\frac{5}{\sqrt{3}}\right)K\frac{(E_p E_g)^{1/2}}{n^2(2\hbar\omega)^5}(2\hbar\omega - E_g)^{3/2}. \tag{26}$$

In the large positive limit for Eq. (24), the energy of the photon is below the two-photon absorption edge, and $\beta^{(2)}$ reduces to

$$\beta^{(2)}(\omega, F) = \left(\frac{5\sqrt{3}}{3\cdot 2^7}\right)\frac{K(E_p E_g)^{1/2}}{n^2(2\hbar\omega)^5}\frac{E_\mu^2}{(E_g - 2\hbar\omega)^{3/2}} \times e^{-4/3(E_g - 2\hbar\omega/E_\mu)^{3/2}}. \tag{27}$$

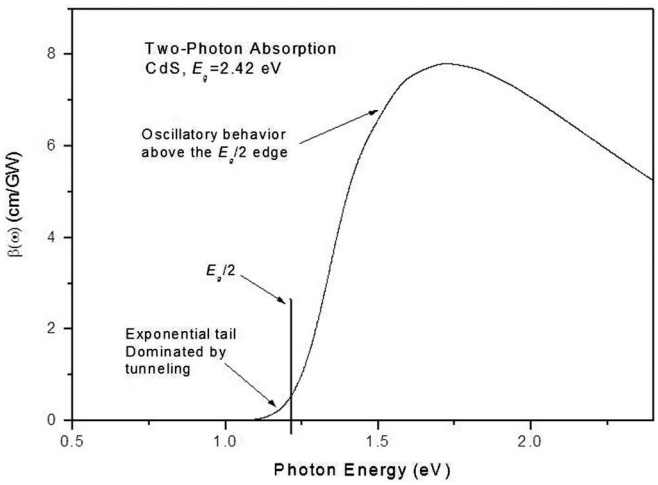

FIG. 2. Two-photon absorption as a function of photon energy for CdS for a dc electric field of $F = 2.0\times 10^6$ V/cm.

Equations (24) and (27) are the main results of this paper. We call Eq. (27) *two-photon assisted field-induced tunneling*, since it is reminiscent of the tunneling effect. However, there are major differences between the two effects. For example, the energy argument in the denominator of Eq. (27) has a 3/2-power dependency as compared to the single-photon case where the energy has a linear dependency. Also, the prefactor of the exponential for the case of single-photon absorption depends linearly on the electric field, while in the two-photon case the dependency in the prefactor is to the power 4/3 as can be seen by the definition of E_μ. However, the exponential dependency of the absorption coefficient for the single as well as the two-photon case shows the same behavior as a function of the electric field. This can be understood in the sense that the exponential behavior depends strictly on the overlap of the electron and hole wave function, which is completely determined by the dc electric field. Above the band edge, the oscillatory behavior is less severe for the two-photon process, and the oscillations decay faster compared to the one-photon case as is shown in Fig. 3.

Figure 2 shows a plot of Eq. (24) for the case of CdS using an electric field of the order of 2.0×10^6 V/cm. As can be seen, below the $E_g/2$ edge, the exponential tail is predominant, while above $E_g/2$ the oscillatory behavior is less pronounced as in the $N=1$ case. In Fig. 3 we plot the electro-optic function, defined as the differential absorption in the presence and in the absence of the electric field, and we can see that the damping of the oscillations is more severe compared to the $N=1$ case. Figure 4 shows the dependence of the electroabsorption coefficient as a function of the electric field, and we can see that as the electric field increases the intensity of the oscillation also increases, but with a longer period.

IV. CONCLUSIONS

In conclusion, we have shown that the N-photon absorption coefficient in the presence of a dc electric field shows the same features as the single-photon process: exponential

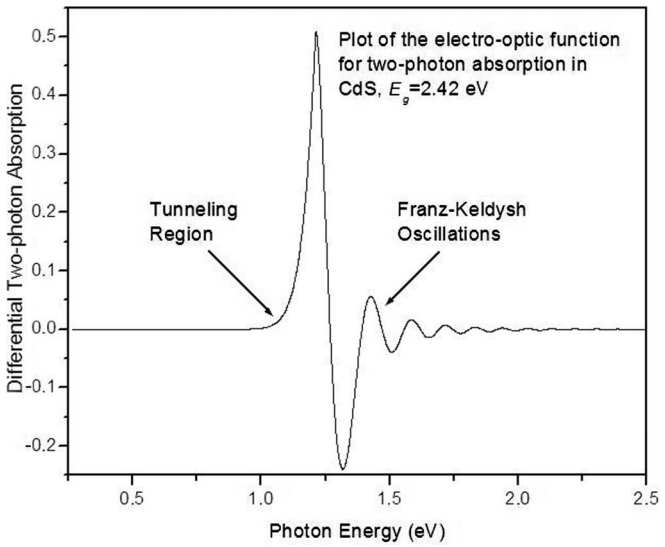

FIG. 3. Plot of the electro-optic function defined as $\Delta\beta^{(2)}\times(\omega,F)=[\beta^{(2)}(\omega,F)-\beta^{(2)}(\omega,F=0)/\beta^{(2)}(\omega,F=0)]$, for CdS in a $F=2.0\times10^6$ V/cm dc electric field.

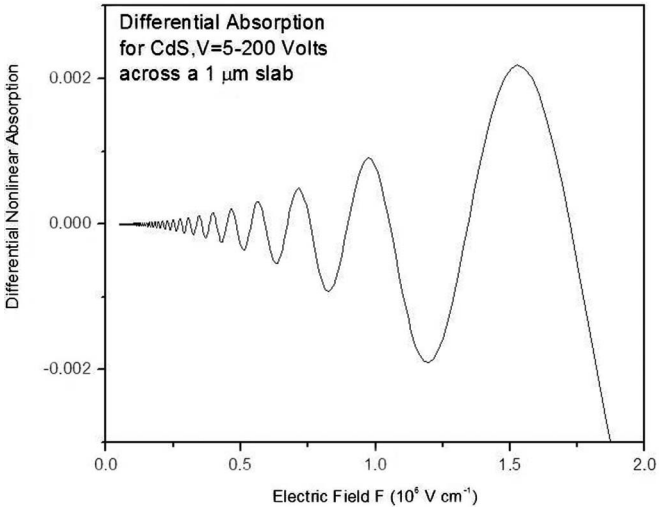

FIG. 4. Differential absorption as a function of electric field for CdS with $V=5-200$ V across a 1 μm thickness sample.

decay below the E_g/N band edge, where $N=1$ is the fundamental edge, and oscillatory behavior above the E_g/N band edge. This scalability leads us to believe that there is a deep connection between linear modulation spectroscopy and nonlinear spectroscopy. For example, Aspnes's[7] results relating the third derivative of the fundamental absorption in the low-field limit and large-energy broadening to the differential absorption modulation should be applied here. One aspect we are still exploring is the deep similarity of the third derivative of the nonlinear two-photon absorption in the presence of the dc field [Eq. (9)] plotted as a function of photon energy shifted by $\hbar\omega$ and the differential one-photon absorption behavior.[26] This may have important implications for nonlinear spectroscopy. Experiments are under way to test the theory.

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