

## Measurement and Model of the Infrared Two-Photon Emission Spectrum of GaAs

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Two-photon emission from semiconductors was recently observed, but not fully interpreted. We develop a dressed-state model incorporating intraband scattering-related level broadening, yielding nondivergent emission rates. The spectrum calculations for high carrier concentrations including the time dependence of the screening buildup correspond well to our measured two-photon emission spectrum from GaAs.

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Two-photon emission (TPE) is a single electron transition accompanied by the emission of a photon pair. A possible second-order perturbation calculation of such TPE spectra requires summation over all system states, which span the virtual state. The energy of each individual photon of the pair is not determined, while the pair as a whole conserves the transition energy. The spectrum of TPE is therefore very broad and continuous [1]. TPE is of great importance for applications in astrophysics, contributing to the continuum radiation from planetary nebulae [1,2], and atomic physics due to the emitted spectrum dependence on the entire quantum level structure of the system [3]. TPE in condensed matter and specifically in semiconductors was only recently observed, exhibiting emission rates nearly 5 orders of magnitude weaker than one-photon processes and nonradiative transitions [4]. This phenomenon opens a wide range of possible applications including efficient electrically driven heralded single photon sources and entanglement sources [5,6]. Moreover, it is a crucial effect for future semiconductor two-photon amplifiers [7–9] and lasers [10] allowing ultrashort optical pulse generation due to the nonlinearity and the wide bandwidth of two-photon gain. The opposite process of semiconductor two-photon absorption (TPA) has been substantially investigated [11,12] and employed recently also in coherent-control applications [13–15]. In all TPA calculations, even in the nondegenerate case (two photons with different energies), the photon energies were considered to be near half the one-photon transition energy [16,17]. However, in spontaneous TPE and in ultrashort pulse stimulated TPE, very wideband continuum of photon energies must be included in the calculations.

In contrast to the discrete-level atomic systems, in semiconductors the energy levels comprise continuous bands corresponding to delocalized solid-state electron wave functions with definite crystal momentum, for which parity is not a good quantum number. Thus, the initial and final states of the electron transition should be included in a possible perturbative span of the intermediate state, as is shown in Fig. 1(a)—a situation which does not occur in discrete-level systems due to parity selection rules. This unique situation results in the initial and final states having

the largest contribution to the transition rates as intermediate states due to the small denominator of the matrix elements, where the electron initial-intermediate (or intermediate-final) state energy difference is close to zero

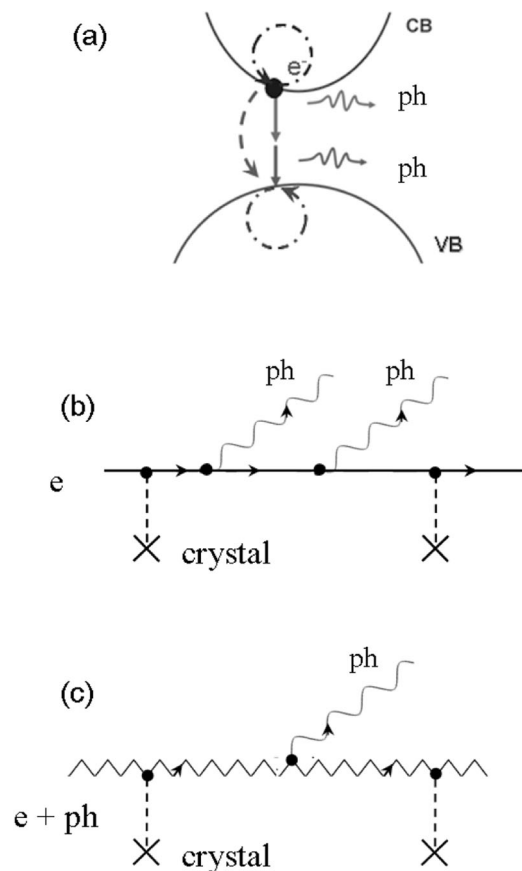


FIG. 1. Transitions diagrams (a) semiconductor TPE transition—the solid arrows indicate the actual transitions via virtual states, while the dash-dotted arrows indicate the contributions of the CB and VB intermediate states to the calculation. (b) A Feynman diagram of a second-order TPE with the crystal electron state described as an interaction with the static crystal potential, depicted by a straight line, while the photons are represented by wavy lines (c) Dressed-state first-order TPE. The dressed crystal electron state is represented by a zigzag line.

[11]. Therefore, in TPE spectral calculations at vanishing photon energies these intermediate states result in infrared catastrophes causing spectrum distortions [4]. Related divergences, occurring in nearly cascaded atomic TPE with resonant intermediate states, were mitigated by the inclusion of a lifetime-related level broadening [1] which can be rigorously included by a proper derivation of the Fermi golden rule [18,19]. In solid-state physics the electron-photon interaction is usually not the strongest one, and various intraband carrier scattering mechanisms which determine the dephasing and the corresponding level broadening are included into the perturbative calculations rigorously via the imaginary self-energy approach [20].

We develop a nonperturbative approach to analyze the process of TPE in semiconductors. Furthermore, we validate experimentally that the corresponding spectrum in GaAs at room temperature does not exhibit any (physically originated) divergence-related distortions due to the zero-frequency resonance. The theoretical model is based on the solid-state dressed electron formalism, where the resulting matrix elements include energy broadening caused by scattering-related dephasing.

In a solid-state system with negligible intraband scattering, the calculated spectrum would be strongly distorted, and the calculated two-photon emission rate could exceed the one-photon emission rate, indicating a modeling problem. However, in most practical solid-state systems various scattering mechanisms induce significant level broadening, and spectrum calculations including the scattering-related level broadening yield correct physical results.

An elegant technique to avoid some of the unphysical divergences in quantum-field theory was proposed by Welton by treating the vacuum field fluctuations semiclassically to interpret phenomena such as the Lamb shift [21]. Our TPE model is based on a related approach, where one field dresses the electronic states while the spontaneous emission from the virtual state is given by a complete quantum-field description. Such a dressed-state method is useful in interpreting various other natural phenomena, such as optical nonlinearities [22] and tunneling [23].

In the standard second-order perturbation formalism, the transition amplitude [Fig. 1(b)] of the electrons from the conduction band (CB) to the valence band (VB) for specified crystal momenta  $\vec{k}_v, \vec{k}_c$  is given by

$$S^{(2)} = \left(\frac{1}{i\hbar}\right)^2 \sum_{\psi_n} \int_{-\infty}^{\infty} dt_1 \langle \psi_f | H_{\text{int}} | \psi_n \rangle \times \int_{-\infty}^{t_1} dt_2 \langle \psi_n | H_{\text{int}} | \psi_i \rangle, \quad (1)$$

with the interaction Hamiltonian  $H_{\text{int}} = -e/m_0(\hat{A} \cdot \hat{p})$ , where  $\hat{p}$  is the momentum operator,  $e$  the electron charge,  $m_0$  the free-space electron mass, and  $\hat{A}$  the vector potential operator for a lossless dielectric in the Coulomb gauge.  $\psi_i = |u_c e^{i\vec{k}_c \cdot \vec{r}}\rangle |0_1, 0_2\rangle$  and  $\psi_f = |u_v e^{i\vec{k}_v \cdot \vec{r}}\rangle |1_1, 1_2\rangle$  are the

initial and the final electron-radiation system states.  $u_v, u_c$  are the Bloch functions of the semiconductor in the VB and the CB, respectively. The calculation is performed by summation over all possible intermediate states  $\psi_n$  of the system; however, in the case of semiconductors only four intermediate states will be significant to the rate calculation with electron at the initial or the final state [Fig. 1(a)].

$$\begin{aligned} \psi_{n1} &= |u_c e^{i\vec{k}_c \cdot \vec{r}}\rangle |1_1, 0_2\rangle, & \psi_{n2} &= |u_c e^{i\vec{k}_c \cdot \vec{r}}\rangle |0_1, 1_2\rangle, \\ \psi_{n3} &= |u_v e^{i\vec{k}_v \cdot \vec{r}}\rangle |1_1, 0_2\rangle, & \psi_{n4} &= |u_v e^{i\vec{k}_v \cdot \vec{r}}\rangle |0_1, 1_2\rangle. \end{aligned} \quad (2)$$

Only one VB is considered here for simplicity, which is, however, sufficient to indicate the divergence problem. The spectral density of TPE in bulk semiconductor per unit frequency is  $W/\partial\omega_1 = \int dk |\vec{k}|^4 V_c M^2(\omega_1, E_{21})$ , where  $V_c$  is a crystal volume, and the unitless matrix element  $M$  is given by

$$\begin{aligned} M^2(\omega_1, E_{21}) &= \left(\frac{e}{m_0}\right)^4 \frac{p_{cv}^2 n_1 n_2}{2\pi^5 \epsilon_0^2 c^6} \left(\frac{m_0}{m_r}\right)^2 \\ &\times \left| \frac{1}{i\omega_1} + \frac{1}{i(E_{21}/\hbar - \omega_1)} \right|^2 \\ &\times f_2(|\vec{k}|)(1 - f_1(|\vec{k}|)) \omega_1 (E_{21}/\hbar - \omega_1), \end{aligned} \quad (3)$$

with  $f_2(|\vec{k}|)$  and  $(1 - f_1(|\vec{k}|))$  the population functions for CB and VB given by Fermi-Dirac distribution,  $m_r$  is the reduced mass,  $p_{cv}$  is the bulk material momentum matrix element,  $E_{21}$  is electron-hole energy separation.

The TPE rate calculated without any level broadening diverges linearly ( $\propto 1/\omega$ ) at low frequencies resulting in an infrared catastrophe [Eq. (3)], which may be avoided by incorporating level broadening through a complex-energy dephasing term [20]. In atomic discrete-level systems the dephasing can be simply attributed to the energy level lifetime [1]; however, for solid-state energy bands, a much richer variety of level-broadening intraband scattering mechanisms should be considered. In semiconductors, level broadening was calculated by the imaginary self-energy approach for phonon scattering [24] and for carrier-carrier scattering [25]. The dominant dephasing process is generally determined by the carrier density and temperature [26], and for high carrier concentrations required for significant TPE, carrier-carrier scattering is the most important process where the interaction strength is affected by the carrier density dependent screening [20].

We employ a dressed-state approach, where one of the interacting fields dresses the initial and final electronic states of the semiconductor—represented as Volkov states. This approach was successfully employed for description of multiphoton absorption in solids [17]; however, for emission processes this method has not been applied before. Moreover, the dressed-state approach is applicable in strong interaction scenarios such as spontaneous emission strongly coupled to a cavity and ultrafast pulse stimulated

emission, while in the weak-field limit the dressed-state calculation approaches the results of perturbative methods.

Volkov states  $\Psi(\vec{r}, t)$  are the exact solutions of the time-dependent Schrödinger equation for a single particle in an electromagnetic field, derived by applying a unitary transformation  $\Omega = \exp\{-\int_0^t \frac{e}{m} \vec{A}(\tau) d\tau \cdot \vec{\nabla}\}$  to the solution of the time-independent equation without external fields, where  $m$  and  $e$  are the particle's mass and charge. These states may be interpreted as plane waves in an oscillating frame, since  $\Omega$  represents a translation operator. The above derivations are correct in dipole approximation and neglecting the Stark effect. Volkov states [27] are therefore given by  $\Psi(\vec{r}, t) = u(\vec{k}, \vec{r}) e^{i\vec{k}(\vec{r} + \delta\vec{r})}$ , where  $u(\vec{k}, \vec{r})$  is the Bloch function, and the envelope phase is modulated by the classical trajectory of the electron oscillations with mean scattering time  $\tau$ . The phase factor of the exact Schrödinger equation solution for a quasifree particle in an electromagnetic field is equal to the semiclassical particle motion [28] (displacement of the oscillating frame):

$$\delta\vec{r}(t) = \left( \frac{e}{m_{\text{eff}} \omega} \frac{1}{\omega + i\tau^{-1}} \right) \sqrt{\frac{\hbar\omega}{2\varepsilon V}} \hat{\varepsilon}_k \cos(\omega t), \quad (4)$$

where  $m_{\text{eff}}$  is the effective mass of electrons within the solid,  $\varepsilon$  is the solid permittivity,  $\hat{\varepsilon}_k$  and  $\omega$  are the field polarization and angular frequency, and  $V$  is the effective quantization volume. The intraband electron motion determines the nonstationary dressed states comprising the virtual state in TPE, whereas the transition from the virtual state is described by a first-order perturbation term between the dressed states [Fig. 1(c)]

$$\begin{aligned} \Psi_i(\vec{r}, t) &= 1/\sqrt{V_c} |u_c(\vec{k}_c, \vec{r}) e^{-i\omega_c t} e^{i\vec{k}_c \vec{r}} e^{+i\eta_c \cos(\omega_2 t)}, 0) \\ \Psi_f(\vec{r}, t) &= 1/\sqrt{V_v} |u_v(\vec{k}_v, \vec{r}) e^{-i\omega_v t} e^{i\vec{k}_v \vec{r}} e^{+i\eta_v \cos(\omega_2 t)}, 1), \end{aligned} \quad (5)$$

with  $\eta_{c,v} = \{e/[m_{c,v} \omega_2 (\omega_2 + i\tau_{c,v}^{-1})]\} \sqrt{\hbar\omega_2/2\varepsilon V} \hat{\varepsilon}_2 \cdot \vec{k}_{c,v}$ . The dressed-state transition matrix element  $S_d = \frac{2\pi}{\hbar} (\hat{\varepsilon}_1 \cdot \vec{p}_{cv}) (e/m_0 \sqrt{\hbar/2\varepsilon_1 V \omega_1}) J_1(\eta_c + \eta_v)$ , where  $J_1$  is the first-order Bessel function. The final expression [Eq. (6)] is derived by taking the weak-field limit—namely the first term in Taylor expansion—and by including a symmetrical Feynman diagram term [Fig. 1(c)], where the first and second photon frequencies are interchanged resulting in

$$\begin{aligned} S_d &= \frac{\pi}{2} \left( \frac{e}{m_0} \right)^2 \frac{m_0 (\hat{\varepsilon}_1 \cdot \vec{p}_{cv}) (\hat{\varepsilon}_2 \cdot \vec{k}_i)}{\varepsilon_0 n_1 n_2 V} \frac{1}{\sqrt{\omega_1 \omega_2}} \left( \frac{1/m_c}{\omega_1 + \frac{i}{\tau_c(\omega_1)}} \right. \\ &\quad \left. + \frac{1/m_c}{\omega_2 + \frac{i}{\tau_c(\omega_2)}} + \frac{1/m_v}{\omega_1 + \frac{i}{\tau_v(\omega_1)}} + \frac{1/m_v}{\omega_2 + \frac{i}{\tau_v(\omega_2)}} \right), \end{aligned} \quad (6)$$

where  $\tau_c, \tau_v$  are the relaxation times in the CB and VB. The relaxation times are in general dispersive—dependent on the excitation frequency, for both phonon related scat-

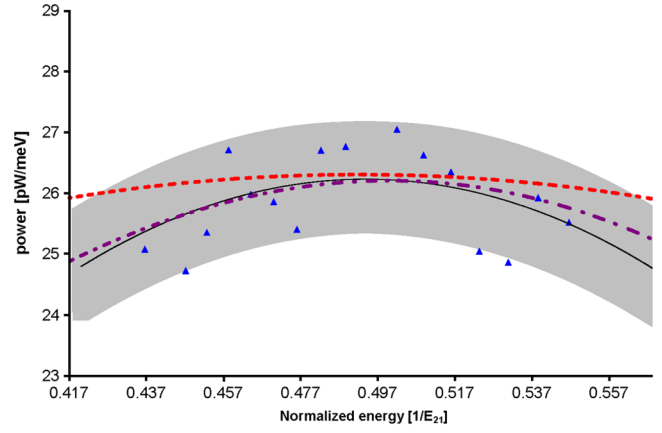


FIG. 2 (color online). TPE spectrum versus energy normalized by the average carrier energy. Experimental measurements are marked by blue triangles, solid black line is the experimental quadratic fit, while the  $1\sigma$  variance is denoted by the grayed area; calculated spectrum by the dressed-state method with constant carrier dephasing time of  $<100$  fsec (dashed red line), and calculated spectrum with the frequency-dependent carrier dephasing (dash-dotted purple line).

tering [29] as well as for carrier-carrier interaction [30]. In the specific case of room-temperature GaAs at high carrier injection of  $2 \times 10^{18} \text{ cm}^{-3}$ , the scattering rates were shown experimentally to be determined by carrier-carrier scattering [31]. The carrier-carrier scattering rates were previously reported to be faster than  $1/(100 \text{ fs})$ , including temporal dependence of the screening buildup determined by the plasma frequency, in agreement with theory [30]. The consideration of frequency dependence of relaxation constants included in our model yields a significant correction to the TPE spectrum shape, conforming to our measurements (Fig. 2). The correct frequency-dependent level-broadening term, which was previously approximated by constants [13], appears in the resulting rates.

The experimental measurements of TPE spectrum from optically pumped bulk GaAs at room temperature (for details of the measurement procedure, see configuration in [4]) are in good agreement with the calculated spectrum (Fig. 2) within  $\sim 10\%$  error when the dispersive electron-electron scattering dephasing mechanism is included. The broad measured TPE spectrum is symmetric about half of the average transition energy  $E_{21}$ . The carrier concentration levels used in the experiments and calculation  $\sim 2 \times 10^{18} \text{ cm}^{-3}$  determine the scattering rate and its dispersion.

In conclusion, we have developed a dressed-state theory of semiconductor TPE which is in good agreement with the observed emission spectrum. Furthermore, we have included more accurately the frequency-dependent dephasing mechanisms which are significant in shaping the related spectrum, as confirmed by the experimental measurement of the spectrum. This theory may be useful for description of other various delocalized-electron phenomena.

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