

Observation of two-photon emission from semiconductors

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Published online: 2 March 2008; doi:10.1038/nphoton.2008.28

Two-photon emission is a process in which electron transition between quantum levels occurs through the simultaneous emission of two photons. This phenomenon is important for astrophysics and atomic physics^{1,2}, and semiconductor two-photon emission was recently proposed as a compact source of entangled photons, essential for practical quantum information processing^{3–5}, and three orders of magnitude more efficient⁶ than the existing down-conversion schemes. Two-photon absorption in semiconductors has been extensively investigated^{7–11}; however, spontaneous semiconductor two-photon emission has not been observed, nor has it been fully analysed theoretically so far. We report the first experimental observations of two-photon emission from semiconductors and develop a corresponding theory. Spontaneous two-photon emission is demonstrated in optically pumped bulk GaAs and in electrically driven GaInP/AlGaInP quantum wells. Singly stimulated two-photon emission measurements demonstrate the theoretically predicted two-photon optical gain in semiconductors^{12–15}—a necessary ingredient for any realizations of future two-photon semiconductor lasers. A photon-coincidence experiment is presented to validate the simultaneity of the electrically driven GaInP/AlGaInP two-photon emission, limited only by the detector's temporal resolution.

Two-photon transitions are much weaker than the related first-order processes, so observations of multiphoton spontaneous decays have so far been restricted to a few atomic transition cases. In these instances the lowest-order transition is forbidden by selection rules¹⁶ or suppressed by a cavity¹⁷, and the two-photon spectrum is continuous and centred at about half the one-photon transition frequency^{1,18}. Semiconductors can be injected with very high charge-carrier densities, making even the weak second-order spontaneous processes measurable, and their two-photon emission (TPE) spectrum is expected to be determined by the photonic state density as well as by the carrier energy distribution.

Here we report the first experimental observation of spontaneous TPE from room-temperature semiconductors, excited both optically in bulk GaAs and electrically in GaInP/AlGaInP quantum wells (QWs) grown on a GaAs substrate. We develop a theoretical model for spontaneous TPE from semiconductors, validating the measured results. In additional experiments, the optically pumped bulk GaAs was singly stimulated to emit specific photon pairs by launching photons at a prescribed wavelength, yielding the co-emission of

complementary wavelength photons with the suppression of the rest of the spectrum. Similar results are shown for GaInP/AlGaInP QWs, and a photon-coincidence experiment verifies the simultaneity of the TPE.

In our first experiment, we optically pumped GaAs with a 100-mW continuous-wave 514-nm argon laser (photon energy, $\hbar\omega_{\text{pump}} \approx 1.73E_{\text{gap}}$ where E_{gap} is the energy gap between the electron levels), focused with a spot size of $\sim 30 \mu\text{m}$ (after filtering out spontaneous infrared (IR) emission) onto a $\sim 200\text{-}\mu\text{m}$ -thick GaAs sample (much thicker than the laser penetration depth), inducing a local carrier concentration of $\sim 1.2 \times 10^{18} \text{cm}^{-3}$, and the pump-induced local sample heating was estimated to be $\sim 330 \text{K}$. The pump laser was chopped at 236 Hz, and the collected emission from the sample in a transmission configuration was detected by a New-Focus IR femtowatt photoreceiver by means of a lock-in amplifier. The spectrum was obtained using an Acton-Research monochromator using a 1,600-nm blazed grating and $\sim 10\text{-nm}$ spectral resolution. As expected, the measured spectrum for the TPE was very wide (Fig. 1a), complying with the theory described below, and the overall collected optical power was $\sim 3 \text{nW}$.

To validate unambiguously that the observed luminescence is indeed TPE, and to dismiss any possibility of inhomogeneously broadened emission from mid-gap levels, we measured singly stimulated TPE by launching a specific wavelength into the optically pumped GaAs. As a result of this excitation, a complementary TPE-wavelength peak appeared. In all the stimulated TPE experiments only the carrier density was modulated for lock-in detection; the stimulating lasers were not modulated, and the observed signals were proportional to the pump power. Stimulation with a 0.2-mW 1,630-nm (0.761 eV) laser resulted in a peak at 1,451 nm (0.854 eV), whereas a 1,600-nm (0.775 eV) laser yielded a peak at 1,476 nm (0.84 eV) (Fig. 1a). In both cases the photon energies are complementary about the centre of the spontaneous emission at 0.81 eV within an error of $\sim 3 \text{meV}$, as expected owing to energy conservation. Changing the stimulation wavelength to 1,570 nm (0.79 eV) caused the stimulating and complementary peaks to merge, approaching the degenerate case. The peaks appeared to merge for any stimulation energy closer than 30 meV to the spontaneous TPE centre. Furthermore, during this excitation, the rest of the spontaneous emission spectrum was suppressed (Fig. 1), clearly demonstrating that the wideband emission is homogeneously broadened, and the strongest effect of the stimulation on the TPE spectrum was observed in the direction collinear with the stimulation. In TPE, the wideband

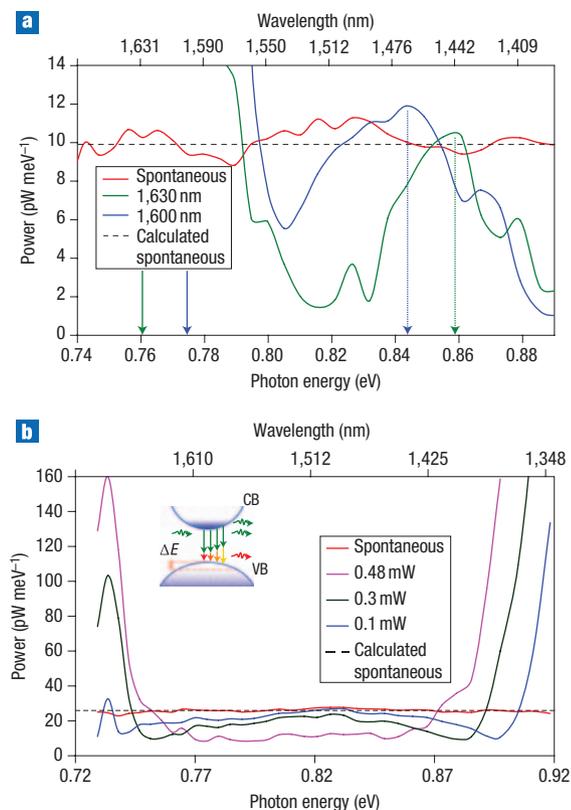


Figure 1 Bulk GaAs TPE measurements and calculations with optical pumping by a 514-nm Ar laser. **a**, Spontaneous and singly stimulated TPE, using a 100-mW pump, at different wavelengths and with 0.2-mW stimulation power. The solid arrows indicate the stimulating photon energies and the dotted arrows indicate the theoretical stimulated photon central energies. **b**, Spontaneous and singly stimulated TPE, using a 180-mW pump, with various stimulation powers at 1,310 nm. The inset is a schematic description of singly stimulated TPE energy broadening in bulk semiconductor.

homogeneous broadening stems from the very short lifetime of the intermediate virtual state, in contrast to an inhomogeneously broadened emission from any possible deep levels within the bandgap¹⁹.

In a third experiment, we performed singly stimulated TPE measurements to study the dependence of the complementary wavelength peak on the stimulating power. To increase the dynamic range of the measurement the pump power was increased to 180 mW, resulting in $\sim 2 \times 10^{18} \text{ cm}^{-3}$ carrier concentration and a 0.84-eV TPE centre. Stimulation with a 1,310-nm (0.946 eV) laser yielded a complementary peak at 1,691 nm (0.733 eV), and the observed power dependence was shown to be linear (Fig. 1b), as expected from theory, where the complementary peak width ΔE is related to the carrier-energy distribution (Fig. 1b, inset).

With the observations described above, the measured IR luminescence was validated as TPE from bulk GaAs. These stimulated TPE experiments also demonstrated two-photon optical gain in semiconductors, which has been proposed theoretically in a degenerate case for two-photon semiconductor amplifiers^{12,14,15} and two-photon semiconductor lasers¹³. Two-photon gain and lasing were observed previously only in discrete-level atomic systems^{20,21}, exhibiting interesting nonlinear behaviour. Usually, two-photon lasers require seed optical power to achieve threshold. Given the nonlinearity of two-photon gain,

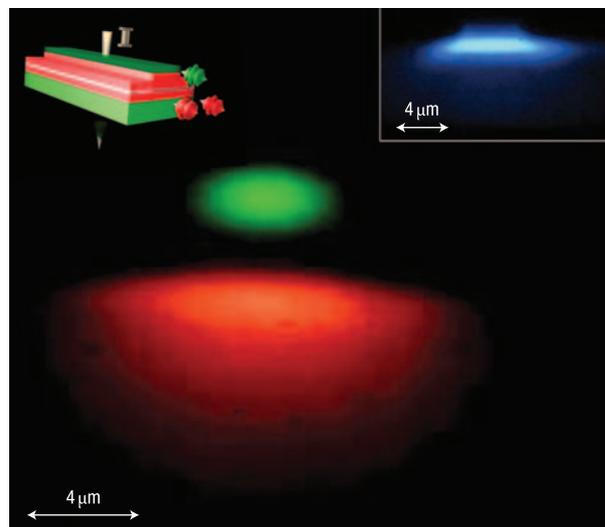


Figure 2 Experimental false-colour IR emission imaging of the facet of the GaInP/AlGaInP QWs waveguide. The upper lobe (green) is the ~ 1.4 -eV one-photon emission from the GaAs cap layer and the lower lobe (red) is the wide-band TPE from GaInP/AlGaInP QWs at ~ 200 mA injection current. The inset is a snapshot of the visible emission from the same structure at ~ 10 mA injection current.

despite the fact that one-photon oscillator strength is much larger, at high seed intensities two-photon gain can become the dominant process. The unwanted one-photon lasing can also be prevented by designing cavities for the two-photon wavelengths only, which do not support any one-photon wavelength modes, by considering the material dispersion or using specially designed reflectors. Using this effect in semiconductor materials may result in novel integrated-photonics sources, allowing ultrashort pulse generation¹⁵, and nonlinear devices for optical switching and digital logic²².

In a fourth experiment, we made a further step towards the implementation of the effect as a device. In this electrically driven TPE experiment we used semiconductor samples consisting of four periods of compressively strained 50-Å $\text{Ga}_{0.45}\text{In}_{0.55}\text{P}$ QWs separated by 55 Å $(\text{Al}_{0.5}\text{Ga}_{0.5})_{0.51}\text{In}_{0.49}\text{P}$ barriers, ~ 1.1 - μm AlGaInP cladding and a heavily doped 300-nm GaAs cap layer. Lateral light confinement was achieved using a 4- μm -wide ridge waveguide, realized by etching techniques, to enhance the emitted photons' collection efficiency. The structure was electrically pumped below the lasing threshold, which was raised to above 200 mA by applying an antireflection coating onto the facets to enable higher carrier population inversion, while the samples were kept at 300 K by closed-loop thermoelectric cooling. Spatial distribution of the wide-band IR TPE intensity was centred near the QWs, whereas the one-photon ~ 1.4 -eV narrow-band emission was located in the GaAs-cap-layer region (Fig. 2). Carrier population was modulated by the injection current at 316 Hz for lock-in detection through a 500-nm blazed grating monochromator. The measured spectrum for the TPE was very wide, as expected, and centred at 0.98 eV, in good agreement with the calculations (Fig. 3a) using the theoretical model described below. The overall TPE measured optical power was ~ 30 nW, which is five orders of magnitude weaker than the measured one-photon emission (Fig. 3a, inset), corresponding to TPE rates much higher than TPA at a single-photon level owing to a continuum of photonic vacuum states in TPE.

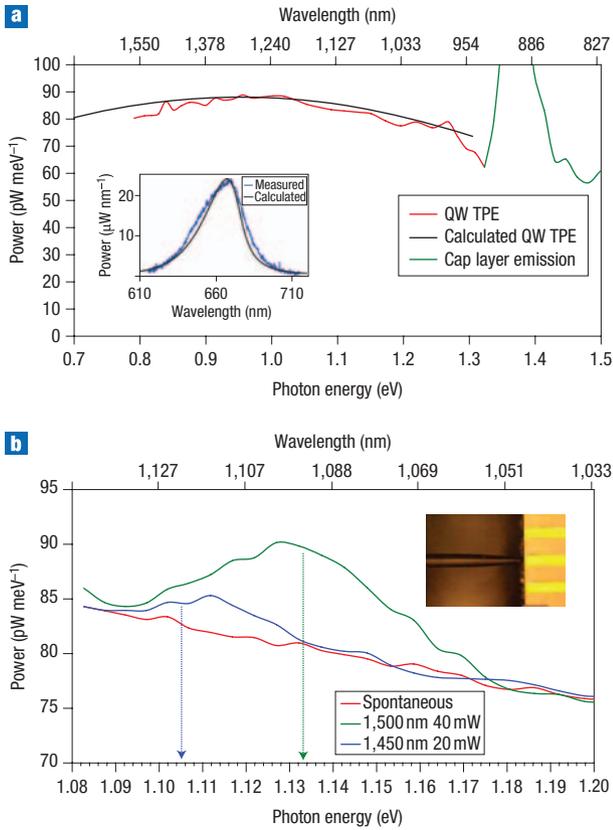


Figure 3 Measured and calculated IR emission spectrum from GaInP/AlGaInP QWs at ~ 200 mA injection current. **a**, Spontaneous TPE. The narrow-band ~ 1.4 -eV peak (green) is the one-photon emission from the GaAs cap layer and the wide-band TPE (red) is from the GaInP/AlGaInP QWs. The inset shows the measured and calculated one-photon emission spectrum from GaInP/AlGaInP QWs at ~ 200 -mA injection current. **b**, Singly stimulated TPE measurement from GaInP/AlGaInP QWs with different stimulation wavelengths and powers. The dotted arrows indicate the theoretical stimulated photon central energies.

The fifth experiment was aimed at observing singly stimulated TPE in electrically pumped GaInP/AlGaInP QWs. The stimulating laser was facet-coupled into the waveguide with a mode area of $\sim 5 \mu\text{m}^2$ by a polarization-maintaining lensed fibre (Fig. 3b, inset). The measurements were taken using a 500-nm blazed grating monochromator with a resolution of 5 nm. Stimulation with a 1,500-nm (0.826 eV) laser resulted in a complementary peak at 1,100 nm (1.127 eV), and 1,450-nm (0.855 eV) stimulation yielded a peak at 1,117 nm (1.11 eV) (Fig. 3b). Both cases are complementary around the centre of TPE at 0.98 eV according to the theory, within an error of 4 meV. The width of the complementary peak is close to the one-photon-emission spectral width, as expected (Fig. 3a, inset); however, the effect of stimulation is much smaller in QWs than in bulk GaAs as a result of the very small overlap of the optical mode with the QWs ($< 10^{-2}$).

In developing semiconductor TPE theory, a fundamental difference between two-photon processes in semiconductors and in discrete-level atomic systems that must be considered is the solid-state delocalized electron wavefunctions with definite crystal momentum, in contrast with the localized atomic states. The rate calculations for TPE require summing over a complete set of the

system states¹. The continuum of delocalized electron states in every band of the semiconductor introduces a vast number of close intermediate states, making the semiconductor two-photon process more efficient. Furthermore, using intraband transitions as part of the two-photon second-order calculations results in a wave-vector (k -) dependence of the matrix element, unlike for the interband transitions for parabolic bands. The spontaneous TPE spectrum from a semiconductor can be calculated by methods similar to those used for atomic systems²³ by a second-order time-dependent perturbation term using the $H = -(e/m_0)\mathbf{A} \cdot \mathbf{p}$ electron–radiation interaction Hamiltonian in the dipole approximation, where \mathbf{p} is the momentum operator, e is the electron charge, m_0 the free-space electron mass, and \mathbf{A} the vector potential operator for a lossless dielectric in Coulomb gauge given by

$$\mathbf{A}(r, t) = \sum_{\mathbf{q}} \sqrt{\frac{\hbar}{2n_q^2 \epsilon_0 \omega_q V_c}} \hat{\epsilon}_q \left(\hat{a}_q^+ e^{i\omega_q t - i\mathbf{q} \cdot \mathbf{r}} + \hat{a}_q e^{-i\omega_q t + i\mathbf{q} \cdot \mathbf{r}} \right) \quad (1)$$

where \hbar is the reduced planck's constant, $\hat{\epsilon}_q$ is photon polarization, ϵ_0 is vacuum permittivity, \mathbf{q} is the photon wavevector, ω_q is the photon radial frequency, V_c is the field quantization volume, \hat{a}_q and \hat{a}_q^+ are the field annihilation and creation operators, and n_q is the material refractive index at ω_q . Using the photon state density per unit frequency of $\rho_p(\omega_i) = V_c \omega_i^2 n_i^3 d\Omega_i / (2\pi)^3 c^3$ for each of the two photons (where the subscript 'i' indicates either photon 1 or photon 2) emitted into a solid angle $d\Omega_i$ (where c is speed of light in a vacuum), the TPE rate in the bulk semiconductor and in QWs is calculated per unit frequency as

$$\begin{aligned} \left. \frac{W}{\partial \omega_1} \right|_{\text{Bulk}} &= \int dk |\mathbf{k}|^4 V_{\text{Bulk}} M^2(\omega_1, E_{21}) \\ \left. \frac{W}{\partial \omega_1} \right|_{\text{QW}} &= \int dk |\mathbf{k}|^3 \pi \langle |\phi_c| \phi_v \rangle^2 S_{\text{QW}} M^2(\omega_1, E_{21}) \end{aligned} \quad (2)$$

where the dimensionless matrix element M is calculated considering the initial and final states as intermediate states. This is similar to semiconductor TPA calculations⁹, where

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

and m_r is the reduced electron mass, p_{cv} is the bulk material momentum matrix element⁸, E_{21} is the electron–hole energy separation, \mathbf{k} is the corresponding electron crystal momentum, V_{Bulk} is the bulk semiconductor sample volume, S_{QW} is the QW sample area, f_1 and f_2 are the conduction band (CB) and valence band (VB) Fermi–Dirac population functions, ϕ_c and ϕ_v are CB and VB QW ground-state envelope functions, and Γ is a phenomenologically included transition dephasing^{24,25}.

TPA calculations performed without this dephasing term were confirmed by experiments^{7,8}. Nevertheless, all non-degenerate TPA calculations^{10,11} were done far from the extreme case of $\hbar\omega_1 \rightarrow 0$ and $\hbar\omega_2 \rightarrow E_{21}$, where one-photon absorption is dominant. In spontaneous TPE calculations, however, a complete continuum of frequencies must be considered, and in the absence of the dephasing term Γ , the matrix element divergence for $\omega_1 \rightarrow 0$ or

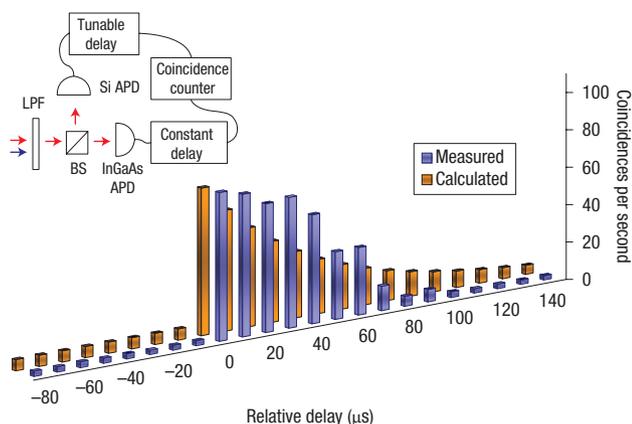


Figure 4 Calculated and measured photon coincidences in electrically pumped GaInP/AlGaInP QWs TPE versus relative delay between detectors. The inset is a schematic of the set-up including silicon and InGaAs avalanche photodiodes (APD), beamsplitter (BS) and a low-pass filter (LPF) blocking the one-photon emission.

($E_{21}/\hbar - \omega_1$) $\rightarrow 0$ (equation 3) causes an IR catastrophe similar to those occurring in zero-frequency nonlinear optics calculations²⁶.

TPE spectra from bulk GaAs at 330 K for the discussed injection levels were calculated according to the described model, using the CB and VB as the intermediate states, neglecting other intermediate bands owing to their energy remoteness^{8,27} using the Joyce–Dixon approximation²⁸ and including bandgap shrinkage effects²⁹ (Fig. 1), where the transition dephasing Γ is determined mainly by the carrier decoherence time—less than 100 fs for such injection levels. The TPE spectrum for GaInP/AlGaInP QWs was also calculated for the given current density (Fig. 3a). According to our model, the ratio of spontaneous TPE versus one-photon emission is in general dependent on the injection levels because the TPE matrix element is k -dependent but one-photon emission is not (for parabolic bands). However, for the specific injection levels considered in GaInP/AlGaInP QWs, TPE is five orders of magnitude weaker than one-photon emission. This ratio is in good agreement with the experiments presented.

In the sixth and last experiment, the photon emission simultaneity was demonstrated by probing the coincidences in TPE from GaInP/AlGaInP QWs. In semiconductors, TPE coincidence measurements are more involved than in typical atomic systems as a result of the relatively low photon energies. For GaInP/AlGaInP QW TPE, at least one InGaAs-based photon counter (PC) must be used. Unlike silicon PCs, InGaAs-based PCs must be operated in gated mode owing to high dark-count rates, and they suffer from significant afterpulsing³⁰ (~ 50 μ s trapped-carrier lifetime at ~ 200 K). In our GaInP/AlGaInP QW TPE coincidence experiment, one silicon PC (Perkin Elmer) and one InGaAs PC (Princeton Lightwave), cooled to ~ 200 K, were used. The sample was driven by 10-ns current pulses at a repetition rate of 100 kHz, which were synchronized with the InGaAs PC gating. Pulse magnitude was set to $\sim 1,000$ PC counts s^{-1} . For this pulsed operation, delays between PCs' outputs that differed from an integer number of periods (10 μ s) resulted in vanishing correlations. Zero relative delay yielded near 10% coincidences owing to the optical component losses. Negative integer-period number relative delay (silicon output

before InGaAs) resulted in $\sim 0.5\%$ coincidences, close to the calculated accidental background level (Fig. 4). Positive integer-period number relative delay (silicon output after InGaAs) resulted in coincidence rates decaying nearly exponentially with ~ 50 μ s time constant owing to the InGaAs PC afterpulsing.

In conclusion, we have demonstrated experimentally and analysed theoretically TPE from semiconductors, which has not been studied before. Spontaneous and singly stimulated TPE was demonstrated in optically pumped GaAs and in current-driven GaInP/AlGaInP QWs. Our theoretical calculations validate the experimental observations. The overall TPE is relatively strong at ~ 30 nW in GaInP/AlGaInP QWs. Such high TPE intensities may help in exploiting this fundamental physical phenomenon for practical uses in both science and engineering, and the demonstrated photon coincidences are crucial for quantum applications such as photon entanglement.

Received 3 July 2007; accepted 22 January 2008; published 2 March 2008.

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