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We investigate the nonlinear optical response of a commercial extended-wavelength $In_{0.81}Ga_{0.19}As$ uncooled photodetector. Degenerate two-photon absorption in the mid-infrared range is observed using a quantum cascade laser emitting at $\lambda = 4.5\,\mu m$ as the excitation source. From the measured two-photon photocurrent signal, we extract a two-photon absorption coefficient $\beta^{(2)} = 0.6 \pm 0.2\,cm/MW$, in agreement with the theoretical value obtained from the E_g^{-3} scaling law. Considering the wide spectral range covered by extended-wavelength $In_xGa_{1-x}As$ alloys, this result holds promise for applications based on two-photon absorption for this family of materials at wavelengths between 1.8 and 5.6 μm. *Published by AIP Publishing.* https://doi.org/10.1063/1.5018619

The interaction between light and semiconductors with large optical nonlinearities enables multiphoton processes, rendering these materials suitable for a variety of photonic devices,[1] among them optical correlators[2,3] and switches[4,5] and quantum detectors.[6] Two-photon absorption (TPA) is a third-order nonlinear process in which the simultaneous absorption of a pair of photons excites an electron from a real state to a higher energy one.[7] TPA can be degenerate or non-degenerate—depending on whether equal or different input wavelengths are used—and is thus sensitive to the square or the product of the optical intensities, respectively. TPA can be used for a number of different applications, such as second-order autocorrelation, which allows one to estimate the duration of optical pulses and optical logic operations. Here, TPA has the advantage of not requiring phase matching and of producing a direct electrical response to the optical signal, in contrast to other nonlinear all-optical phenomena such as second harmonic generation.[8]

In semiconductors, the efficiency of a TPA process depends on the energy gap (E_g), and the photon energy must satisfy the requirement for TPA,[9] i.e., $E_g/2 \leq \hbar\omega < E_g$. It can be shown that the nonlinear coefficient of degenerate TPA scales as E_g^{-3}, a relation experimentally verified in a large number of semiconductors.[9,10] Therefore, narrow-bandgap semiconductors are generally the best choice for degenerate TPA. On the other hand, wide-bandgap semiconductors can exploit extreme non-degenerate TPA to enhance the nonlinear coefficient of the process.[11]

TPA has been characterized in the near-infrared range (0.75–3 μm) in several semiconductor materials,[12] such as Si, AlGaAs, and ZnSe, while in the long-wavelength infrared (8–15 μm), extensive measurements have been reported on InSb[13–15] and InAsSbP.[15] The latter, having cutoff wavelengths around 4.5 μm (InAsSbP) and 5.5 μm (InSb), potentially offers suitable solutions for two-photon detection also

in part of the mid-infrared range (3–8 μm)—a spectral region of high interest for applications such as spectroscopy and sensing—specifically between 4.5 and 8 μm. However, with the notable exception of two-photon GaAs/InGaAs quantum well infrared photodetectors,[16,17] which suffer from a narrow spectral range, there appears to be a lack of studies of possible materials for TPA in the shorter wavelength region of the mid-infrared range. Here, TPA could serve as a convenient method for pulse characterization,[8] as previously demonstrated using quantum cascade lasers[15] (QCLs) and free electron lasers.[16] In this work, we study TPA in a commercial photodiode (PD) based on extended wavelength $In_{0.81}Ga_{0.19}As$, having a threshold for one-photon absorption at 2.8 μm—much larger than that of standard $In_{0.53}Ga_{0.47}As$ (1.8 μm) lattice-matched to InP. This may potentially enable broadband TPA at wavelengths between 2.8 and 5.6 μm.

In order to study the nonlinear optical response of a semiconductor, the incident power density must be varied. This may be achieved by the Z-scan technique,[18] where the optical power is fixed and the spot size is changed by scanning the material through a well-defined beam pattern (such as a focus). As an alternative to the Z-scan, one may vary the optical power from the light source for a fixed spot size.[19] One can then measure the power of the beam transmitted through the material although this requires particular attention to interfering effects such as free carrier absorption in the material[20] and absorption in the substrate. More straightforwardly, in the case of a PD, the two-photon current (TPC) signal induced by TPA may be directly measured.

A schematic of the experimental setup used for our study is shown in Fig. 1(a). The PD is a commercial extended-wavelength $In_{0.81}Ga_{0.19}As$ photovoltaic detector (Teledyne Judson Technologies) operating without thermoelectric cooling and without applied bias, having an active window size of 250 μm diameter. The 50% responsivity cutoff wavelength of the PD is 2.6 μm. The PD is mounted on an XYZ micropositioning stage. The light source used for
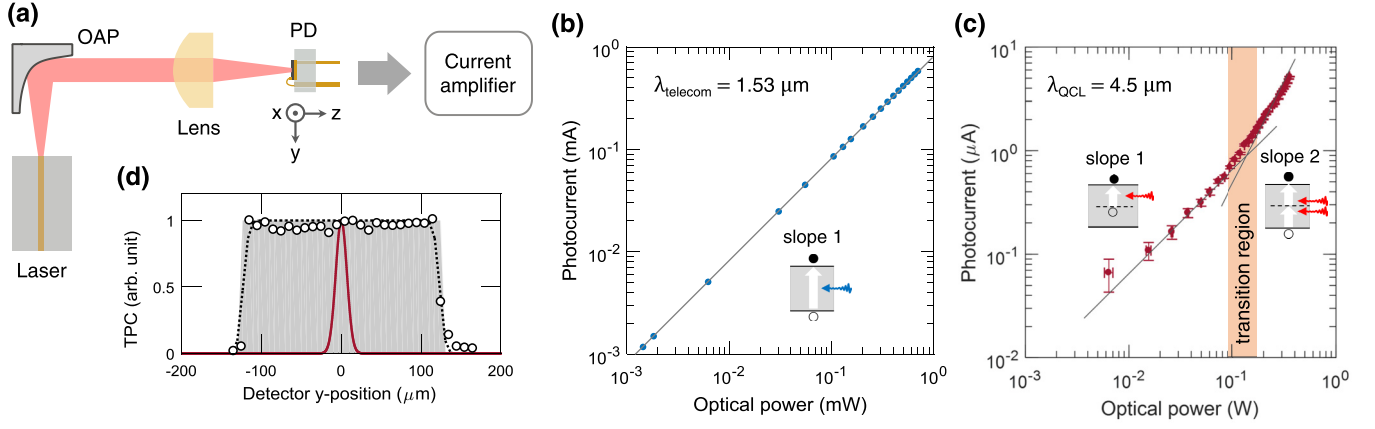
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FIG. 1. (a) Schematic of the experimental setup used for the characterization of two-photon absorption in the extended-wavelength $In_{0.81}Ga_{0.19}As$ detector. OAP: off-axis parabolic mirror and PD: photodiode. (b) One-photon response of the PD for the above-bandgap excitation using a near-infrared laser. (c) Photocurrent response of the PD to sub-bandgap excitation using a quantum cascade laser. Two regions exhibiting a linear and a quadratic response are observed. Lines with slopes 1 and 2 are plotted in (b) and (c). (d) Experimental measurement of the two-photon current (TPC) signal of the PD (circles) as a function of the scan position along the y-axis. The optical power of the quantum cascade laser is 350 mW. Using the size of the detector active window (shaded area), the experimental data are fitted with a model simulating the overlap of the laser spot (assumed to be Gaussian) with the detector active window as a function of the y-position. From the best fit (dashed line), the beam profile is obtained (red line).

TPA characterization is a Fabry-Perot QCL operating under continuous wave (CW) electrical injection and emitting at a central wavelength of $4.5\,\mu m$. Depending on the level of current injection, the QCL may operate in a single or multi-mode regime.[21] However, due to its small bandwidth ($\Delta\lambda/\lambda = 3.6\%$), we can consider it as an excitation source of nearly degenerate TPA. Its optical output is expected to be quasi-CW.[15,22] The QCL is operated using a low-noise current driver (Wavelength Electronics QCL LAB 1500), while its temperature is regulated by a thermoelectric controller (Wavelength Electronics TC5). The QCL output is collimated with an off-axis parabolic mirror (15 mm focal length, 12.7 mm diameter) and focused using a plano-convex, anti-reflection coated ZnSe lens (38 mm focal length and diameter). The photocurrent induced in the PD is measured directly with a low-noise current amplifier (Keithley Multimeter 2000)—no sophisticated means of detection (e.g., lock-in detection) is required, a testament to the accessibility of TPA in this material. The experiments presented here have been repeated on distinct but similar devices (both PD and QCL), leading to the same behavior.

We begin by characterizing one-photon absorption in the PD. In this case, we use a near-infrared laser (DFB diode, Mitsubishi FU-68PDF-5) emitting at $1.53\,\mu m$ as the optical source. The measured response [Fig. 1(b)] exhibits a linear behavior, as expected, with a responsivity of 0.8 A/W. Next, we characterize higher order optical nonlinearities in the device using the QCL for sub-bandgap excitation. As can be seen in Fig. 1(c), two main regions can be observed, separated by a transition region. The first exhibits a linear photo-current response to incident optical power, while the second is quadratic. The identification of the microscopic mechanism giving origin to the linear response is beyond the scope of this work. It may originate from the excitation of deep defects in the gap[18,19] and has a relatively low quantum efficiency of 2.8×10^{-6}, although considerably larger than those measured in GaAs[19] and Si[18] PDs (in the order of 10^{-8} and 10^{-9}, respectively). The quadratic response corresponds to

TPA. By analyzing the response in this region, we can extract the TPC coefficient of the device as[18]

$$\gamma_{TPC} = \frac{I}{e} \times \frac{A}{P^2}, \tag{1}$$

where I is the photocurrent, P is the incident optical power on the PD, and A is the spot size. Equation (1) assumes a spot size smaller than the detector active window and a constant optical intensity over the beam area and across the space-charge layer of the PD (non-depletion approximation). The spot size can be obtained by measuring the TPC signal while scanning the detector along the y-axis [Fig. 1(d)]. Assuming a Gaussian profile of the laser and given the size of the detector window, the beam profile at the focal spot can be numerically deconvolved, yielding a $1/e^2$ diameter of $28 \pm 4\,\mu m$. From Eq. (1), we deduce $\gamma_{TPC} = (1.8 \pm 0.5) \times 10^9$ cm^2 W^{-2} s^{-1}. In order to extract the TPA coefficient of the material, a device-independent parameter, we use the following relation:[18]

$$\beta^{(2)} = \frac{\gamma_{TPC} \times 2\hbar\omega}{\eta_{coll} \times w}, \tag{2}$$

where η_{coll} is the carrier collection efficiency of the device and w is the width of the absorber layer.[23] From Eq. (2), we obtain $\beta^{(2)} = 0.6 \pm 0.2$ cm/MW. A theoretical value for the nonlinear coefficient of extended-wavelength $In_{0.81}Ga_{0.19}As$ can be predicted using the theoretical scaling law of $\beta^{(2)}$ with E_g^{-3} given by[9,10,14]

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

where $K^{(2)} = 3.10$ eV$^{5/2}$ cm/MW is a material-independent constant that can be empirically obtained by fitting TPA in different semiconductors,[9,10] E_p is the Kane energy[24] and possesses a value of ≈ 20 eV in most direct gap semiconductors,[10] and n is the linear refractive index. Using the material

parameters,[25] $n = 3.5$ and $E_g = 0.44 \, \text{eV}$, we obtain a theoretical value of $\beta^{(2)} = 0.54 \, \text{cm/MW}$ for excitation at $\lambda = 4.5 \, \mu\text{m}$, close to the experimental determination reported here. Thanks to the bandgap tunability that can be achieved with $\text{In}_x\text{Ga}_{1-x}\text{As}$ alloys, this family of materials holds promise for TPA detectors covering a wide wavelength region in the mid-infrared, between 1.8 and $5.6 \, \mu\text{m}$, which could be potentially tailored for specific applications in this range.

In summary, we have studied the nonlinear optical response of a commercial extended-wavelength $\text{In}_{0.81}\text{Ga}_{0.19}\text{As}$ photodetector. By using a QCL as a sub-bandgap excitation source emitting moderate optical power at $\lambda = 4.5 \, \mu\text{m}$, we observed a TPA response of the detector. The TPA coefficient of the material extracted from the two-photon photocurrent measurement is $\beta^{(2)} = 0.6 \pm 0.2 \, \text{cm/MW}$, in agreement with the theoretical value obtained using the E_g^{-3} scaling law. In the future, we envision a waveguide geometry[26,27] for the photodetector, designed to increase the beam propagation length in the absorbing material while preserving a high power density due to optical confinement. Assuming a waveguide length of the order of 1 mm ($\approx 1000 \times$ longer than the length of the absorber layer in the regular absorption geometry studied in this work), an optimized coupling efficiency ($\geq 30\%$), and negligible free carrier absorption, we expect that the efficiency of two-photon current generation in the detector could be enhanced by at least two orders of magnitude by means of such a scheme.

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