Picosecond time-resolved photoluminescence at detection wavelengths greater than 1500 nm

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We report what is to our knowledge the first application of high-efficiency InGaAs/InP photon-counting diode detectors in time-resolved photoluminescence measurements at wavelength greater than 1500 nm. When they were cooled to 77 K and used in conjunction with the time-correlated single-photon counting technique, the detectors were capable of an instrumental response of 230 ps and a noise equivalent power of 2×10^{-17} W $Hz^{-1/2}$. Preliminary measurement of a semiconductor heterostructure indicates sensitivity at photogenerated carrier densities as low as 10^{14} cm^{-3}. This development facilitates the detailed characterization of dominant recombination mechanisms in semiconductor optoelectronic materials and devices designed to operate in the third telecommunications spectral window.

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Time-resolved photoluminescence (TRPL) measurements can yield valuable information concerning electronic relaxation processes in semiconductor materials. When they are used with the time-correlated single-photon counting (TCSPC) technique,[1] semiconductor photon-counting diode detectors provide a means of measuring luminescence decays with high sensitivity and temporal resolution. Previously this approach had been used to measure decays with time constants as short as 10 ps at wavelengths up to 1000 nm and ~50 ps at wavelengths up to 1500 nm, with Si and Ge single-photon avalanche diode (SPAD) detectors, respectively.[2,3] TCSPC also offers the benefits of high sensitivity to low light levels and a dynamic range of several orders of magnitude, facilitating detailed characterization of switching between dominant mechanisms as the system relaxes.

The long-wavelength limit of sensitivity afforded by previous TCSPC measurements stopped short of the spectral region known as the third telecommunications window, from 1500 to 1600 nm, which corresponds to the absorption minimum in silica optical fibers and is the wavelength range within which most long-haul telecommunications devices are designed to operate. Time-resolved characterization of these devices, and the materials that they comprise, has therefore been possible only with pump–probe[4] or upconversion detection[5] techniques, which are much less sensitive to low light levels and thus require optical excitation at higher powers than would typically be used in device applications.

Recent developments in the production of low-noise InGaAs/InP separate absorption, grading, and multiplication layer avalanche photodiodes have now yielded a detector (Epitaxx Model EPM239) that exhibits good photon-counting characteristics when it is cooled to reduce thermal carrier generation in the absorption layer. We have incorporated this detector into an existing time-resolved photoluminescence microscope (an adaptation of the instrument described in Ref. 2) and performed what is to our knowledge the first high-sensitivity TRPL measurements of semiconductor multiple-quantum-well (MQW) heterostructures that emit at these wavelengths. In this Letter we report the resolution capabilities of the new measurements and assess their usefulness in materials and device characterization.

A schematic diagram of the TRPL apparatus is shown in Fig. 1. Pulsed optical excitation is provided by a passively Q-switched diode laser emitting at $\lambda = 1305$ nm with a pulse duration of 10 ps and an optical energy of ~4 pJ/pulse.[6] The excitation light is focused onto the sample, and the luminescence collected with the same lens, spectrally filtered and attenuated as necessary for the prevention of pulse pileup[1] and focused into a single-mode fiber. This fiber is coupled to the single-mode pigtail of the SPAD, which is situated in a liquid-nitrogen-cooled variable-temperature cryostat. Full details of the optical and electrical configurations of this instrument will be published elsewhere.[7]

Figure 2 shows an instrumental response characteristic at a detector temperature of 77 K and

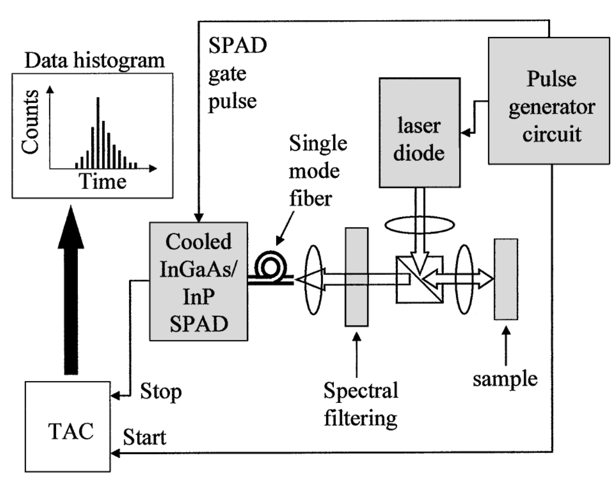

Fig. 1. Schematic diagram of the TRPL apparatus. This is a standard TCSPC setup but with a cooled InGaAs/InP SPAD detector, which is sensitive to wavelengths as long as 1600 nm. TAC, time-to amplitude converter.

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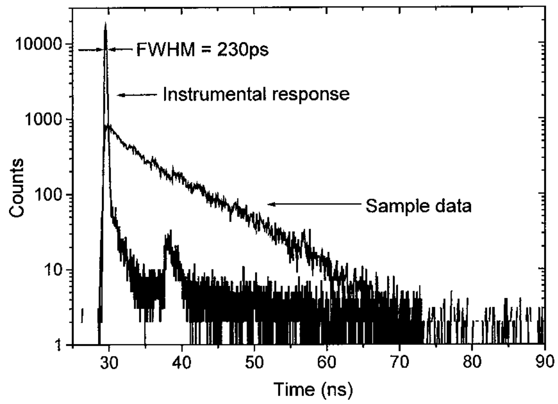

Fig. 2. Semilogarithmic plot of the instrumental response and sample data for the TRPL measurements. The sample data were measured from an InGaAsP MQW p–i–n-doped structure at room temperature and at a luminescence wavelength of 1540 nm.

a sample data set measured from an InGaAsP MQW p–i–n-doped heterostructure emitting at $\lambda_{PL} = 1540$ nm at room temperature. For measurement of the instrumental response characteristic, a mirror was situated in place of the sample and no spectral filtering was used, so the attenuated excitation pulse was measured directly. The FWHM of the resultant histogram peak, at a detector temperature of 77 K, is 230 ps. This temporal width is determined primarily by the timing jitter of the SPAD and is seen to increase slightly to 270 ps at an incidence wavelength of 1550 nm. Iterative reconvolution of photoluminescence decay data by use of the instrumental response can permit resolution of lifetimes as short as one fifth of the FWHM, or ~60 ps. The secondary peak in the instrumental response is simply a result of backreflection in the optical fiber used to collect the luminescence signal and could be eliminated by the use of angled fiber ends.

Now let us consider the sensitivity of the measurements. Photodetector sensitivity is commonly indicated by the noise equivalent power (NEP), which represents the incident optical power required in order that the signal-to-noise ratio equal unity. For a single-photon detector the NEP is given by

$$\mathrm{NEP} = \frac{\hbar\omega}{\mathrm{DE}}\sqrt{2N_D}\,, \qquad (1)$$

where $\hbar\omega$ is the incident photon energy, DE is the detection efficiency, and N_D is the dark-count rate of the detector. The square root is a result of the Poissonian nature of the temporally uncorrelated dark counts.

At 77 K the InGaAs/InP SPAD used in this research displays a normalized dark-count rate of $\sim 100\ \mathrm{s}^{-1}$ and a detection efficiency of 8%, giving a NEP at an incident photon wavelength of 1550 nm of approximately $2 \times 10^{-17}\ \mathrm{W\ Hz}^{-1/2}$. This value is approximately an order of magnitude lower than was previously recorded with similar devices.[8] The NEP in the form given in Eq. (1), however, applies to a temporally uncorrelated optical signal. In TRPL measurements, the signal is limited to a few excess carrier lifetimes after the excitation pulse, and we may gate the detector accordingly. A better indication of the sensitivity of these measurements is obtained by consideration of the probability that a dark count will occur inside such a gate. For instance, a 100-ns gate might be used to record a photoluminescence decay with a time constant of the order of 10 ns, such as is observed in the sample data in Fig. 2. At a dark-count rate of $100\ \mathrm{s}^{-1}$ the probability of such an event during the gate is 10^{-5}. By considering the optical collection efficiency of our setup and the photoluminescence efficiency of the semiconductor heterostructure that is measured,[9] we may now estimate the excess carrier density required in the heterostructure to generate a signal count probability that is equal to the dark-count probability. Whereas the latter definition of measurement sensitivity is still simplistic and does not make full use of the temporal correlation that is present in the signal,[10] it is nonetheless a useful indicator and for this situation yields a value for the minimum excess carrier density of the order of $10^{14}\ \mathrm{cm}^{-3}$. By comparison, previous studies were limited by their sensitivity to carrier densities greater than $\sim 10^{17}\ \mathrm{cm}^{-3}$.[4,5]

Support for this estimate of the measurement sensitivity is provided by an independent estimate of the peak excess carrier density photoexcited in the InGaAsP MQW structure. Given an excitation spot diameter of 30 μm, and assuming an absorption coefficient of $10^4\ \mathrm{cm}^{-1}$ at 1305 nm, we calculate an initial carrier density in each quantum well of $10^{16}\ \mathrm{cm}^{-3}$. Noting that the count-rate axis scales with the square of the excess carrier density, we therefore estimate the lowest carrier density detected to be $3 \times 10^{14}\ \mathrm{cm}^{-3}$. As no effort has been made in these measurements to maximize sensitivity, and indeed the temporally uncorrelated background is not yet visible in the data, this density value merely serves as experimental verification that routine access to such low carrier densities is possible.

The accessible dynamic range also sets these measurements apart from other TRPL techniques. Figure 2 shows a signal-to-background ratio of approximately 35 dB in the instrumental response and of 30 dB in the sample data (the average number of background counts per channel, seen toward the right of the figure, is approximately 1), permitting accurate determination of the carrier density dependence of the luminescence decay. In the sample data shown, a single exponential decay is observed throughout this entire range, indicating that the recombination dynamics are dictated by a single carrier type in this device structure for carrier densities from 10^{16} to $3 \times 10^{14}\ \mathrm{cm}^{-3}$ (The small oscillations of ~3-ns period observed on the sample decay are not genuine TRPL features but a result of electromagnetic pickup in the detection circuitry.) Characterization over such a wide range of carrier densities is not usually possible with less-sensitive TRPL techniques, which typically exhibit a dynamic range of less than 10 dB.[4,5]

The long-wavelength spectral limit of these measurements, determined by the absorption edge of InGaAs (lattice matched to InP) at 77 K, is approximately 1600 nm. The short-wavelength limit corresponds approximately to the absorption edge of the InP layers, at $\lambda \approx 900$ nm, beyond which the timing jitter of the detector increases substantially as a result of absorption in the p-type contact. This wavelength is short enough to overlap the long-wavelength range of silicon homojunction SPADs and results therefore in complete coverage of the wavelength range 400 nm $< \lambda <$ 1600 nm for high-sensitivity TCSPC measurements.

The temporal resolution and sensitivity to low excess carrier densities described in this Letter indicate that TRPL that uses TCSPC with InGaAs/InP SPAD detectors can be of great benefit in characterizing optoelectronic materials and devices that are designed to operate at wavelengths in the third telecommunications window, beyond the reach of Si and Ge homojunction detectors. In addition to measurement of bulk and heterostructure samples, our results indicate sufficient sensitivity for the detection of individual emissions from a single dipole such as is observed in a quantum dot or dye molecule and as such represent a substantial milestone toward the development of new quantum optical components such as triggered single-photon sources for operation in this spectral region.

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10. For a full treatment, See Ref. 7.