

Low temperature Photoluminescence (PL) spectroscopy on Quantum Dots

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BACKGROUND

This article provides an overview of the Photoluminescence application, its associated experimental problems and how they can be overcome.

The basic elements of a PL spectroscopy set-up consist in an optical source, a spectrophotometer and a detector. (see Figure 1). The excitation laser beam is focused on the sample with a lens of typically $f=20-30$ cm focal length. The emitted light can be collected by a single lens ($2f/2f$ geometry) or by two lenses F1 and F2. The advantage of this latter geometry is to allow the use of a short focal length for F1 in order to improve the collection efficiency of the emitted light. Moreover, F2 can be chosen in order to match the optical aperture of the spectrometer, avoiding unnecessary supplementary losses.

PROBLEMS ASSOCIATED WITH PHOTOLUMINESCENCE APPLICATIONS

The main difficulty arises from the poor collection of the emitted radiation. Generally, the PL signal is emitted over a 4π solid angle in free space (spontaneous emission). For a lens F1, with diameter D and focal length f_1 , the solid angle of collection is $2\pi(1-\cos\theta)$ with $\tan\theta=D/2f_1$. For $D=50\text{mm}$ and $f_1=80\text{mm}$, only 2.3% of the emitted light is collected.

The picture is even worse if we examine the extraction efficiency of the PL signal from semiconductor heterostructures. These materials are characterized by a high index of refraction, typically of the order of 3. For a localized emitter embedded in a semiconductor medium such as GaAs ($n_{sc}=3.46$), total internal reflection at the semiconductor/air interface occurs for an angle of 17° . This means that all photons reaching the semiconductor surface with an incident angle greater than 17° with respect to the normal will be reflected inwards and will not be detected. The internal solid angle corresponding to the photons that will be transmitted outside the semiconductor material is equal to $2\pi(1-\cos 17^\circ) \approx \pi/n_{sc}^2$. Taking into account the reflection coefficient at the semiconductor/air interface, we find that only 1.5% of the emitted photons leave the sample and are emitted over a 2π external solid angle.

QUANTUM DOT APPLICATIONS

In semiconductor physics, a quantum dot is a small region of a semiconductor material buried in another one with a larger bandgap. Such a nanostructure has a size ranging from a few nanometers to a few hundreds of nanometers. Charge carriers are confined in three dimensions on a scale that is comparable to the electron's De Broglie wavelength, thus leading to quantized energy levels, much like an atom.

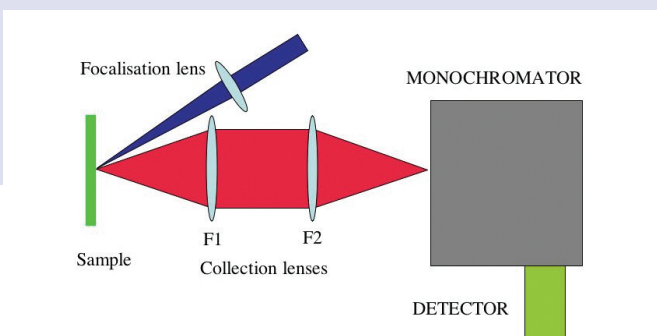


FIGURE 1

Schematic diagram of a photoluminescence spectroscopy experiment

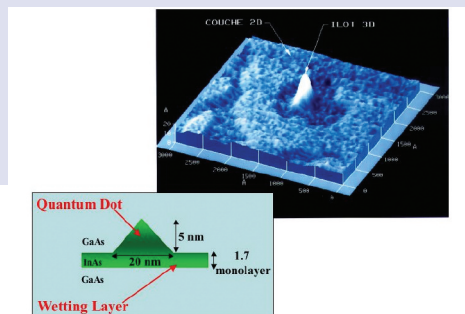


FIGURE 2

(Top) Atomic Force Microscopy (AFM) image of an uncovered InAs pyramid on top of a thin InAs layer. (Bottom) Schematic representation of the InAs quantum dot above its wetting layer. J. M. Gérard et al, J. Cryst. Growth 150, 351 (1995).

For this reason, quantum dots are often called «artificial atoms» or «macroatoms». Single-quantum dot devices attract much attention in the field of semiconductor nanostructures because of their potential for the storage of single electrons, emission of single photons, and manipulation of single qubits for quantum information processing. The current interest in these nanostructures is fundamentally related to the discretization of the electron energy spectrum due to the three-dimensional quantum confinement. However, contrarily to the atomic case, a semiconductor QD can interact with its environment through electron-phonon, electron–electron interactions. The suppression of these processes to get an isolated macroatom appears as a major challenge in semiconductor nanotechnology.

SINGLE QUANTUM DOT PHOTOLUMINESCENCE SPECTROSCOPY

Single quantum dot (QD) spectroscopy is performed by microphotoluminescence measurements in the far field in a confocal geometry (see Fig. 3). The sample is mounted on the cold finger of a continuous flow helium cryostat for measurements down to 5 K. The **Microstat**[®]HiRes cryostat from Oxford Instruments offers short working distances (3 to 7 mm) allowing the use of a high magnification microscope objective (x36, 8.6 mm working distance) with a 0.5 numerical aperture. The excitation beam, provided by a tunable cw Ti:sapphire laser, is focused on the sample with a spot size of approximately 1 μm , accurately positioned using X–Y piezoelectric stages moving the microscope objective with a precision of $\pm 0.05 \mu\text{m}$.

Such a spatial resolution does not allow single dot spectroscopy. In fact, typical QD densities are in the order of 10^{10} - 10^{11} cm^{-2} . Therefore, from 100 to 1000 dots of different sizes are present under the laser spot and the PL spectrum is non homogeneously broadened. In order to isolate single QDs, the sample is processed on the surface with submicron mesa patterns. Another solution is to use submicron metallic optical masks. The PL signal is collected by the same microscope objective as for the excitation. A pinhole located in an image plane allows for spatial resolution and increases the signal-to-noise ratio. The size of the pinhole is determined by the magnification of the collection optics (microscope + lens). For our set-up, the magnification is about 100 so that a 100 μm pinhole selects a 1 μm area on the sample surface.

FIGURE 3

Microphotoluminescence experimental set-up. On the right side, view of the cryostat and the microscope objective

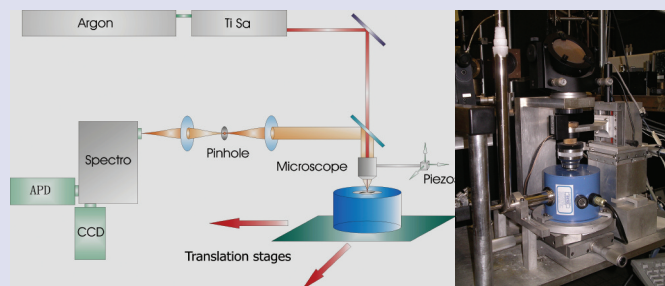
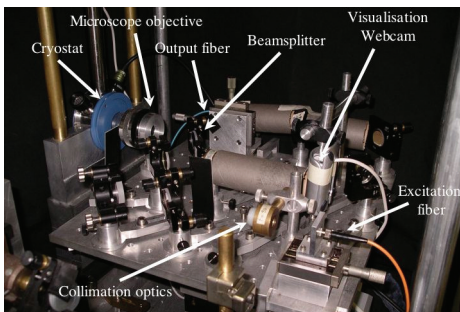


FIGURE 4

Microphotoluminescence set-up using optical fibers



A more versatile version of a microphotoluminescence set-up uses fiber optics as shown in Figure 4. The excitation laser is coupled to an optical fiber. The output of the fiber is then collimated and sent through a beam splitter to the microscope objective. The emission light is collected by the same microscope objective and then focused on the output fiber. Since the core of this latter is 200 μm , the pinhole is no longer necessary. The optical cryostat is fixed on the optical table and all the excitation and detection optics, placed on a small table, can be moved in the three spatial directions with a 100nm precision. Such a set-up enables the excitation source or the detection unit to be changed very easily.

For the InAs/GaAs system a typical value for the PL recombination time is of the order of 1 ns. This means that, under cw excitation, when the emission from the fundamental transition of the QD is saturated, about 10^9 photons are emitted per unit time ($\approx 8 \cdot 10^7$ for pulsed excitation at a 80 Mhz repetition rate). The values of the extraction and collection efficiencies detailed above first reduce this number by approximately three orders of magnitude. Furthermore, the transmission of the detection part of the set-up including microscope objective, lenses, beam splitter, grating spectrometer is about 10%. In such conditions, at saturation, only 10^5 photons reach the optical detector per unit time. Finally, measurements are often performed at excitation powers one or two orders of magnitude below saturation thus giving emission rates of 10^3 - 10^4 photon/s.

The emission being in the mid-infrared (900 nm-1 μm) photomultiplier tubes cannot be used, their cathode sensitivity falling dramatically in this spectral region. The

detection could be performed by avalanche photodiodes (APD) but their typical photosensitivities are of the order of 30-50 A/W (Si APDs). At most, our signal would be at least 8 orders of magnitude below the dark current. Ultra-low noise detecting systems are thus required. For example, in our single QD micro-PL set-up, the signal detection is performed, after spectral filtering by a 32 cm monochromator, by a low noise Si-based photon counting module (Perkin-Elmer SPCM-AQR-16). Such a detector may provide less than 25 dark counts per second with a quantum efficiency in the 10-30% range. At saturation (excitation density of approximately $10 \text{ kW}\cdot\text{cm}^{-2}$ for a laser tuned in resonance with the wetting layer absorption band) the expected signal-to-noise ratio is about 1,000 and falls down to 10 for low excitation power experiments.

Typical PL and photoluminescence excitation (PLE) spectra of single quantum dots are shown in Figure 5. The spectral resolution is approximately 150 μeV in PL spectra, and 30 μeV in PLE spectroscopy where the laser spectral width gives the spectral resolution.

On the left side (lower panel), at low power excitation (light blue curve) the PL emission consists in a narrow resolution-limited line which corresponds to the fundamental transition (S_e - S_h) of the QD. When the excitation power is increased, the intensity of this line increases linearly, and then saturates. At that point recombination lines related to excitonic complexes (bi-excitons, charged excitons...) or higher energy transitions can be observed. For example, on the high-power spectrum (blue curve), a bi-excitonic line appears 5 meV below the fundamental transition, and the PL line from

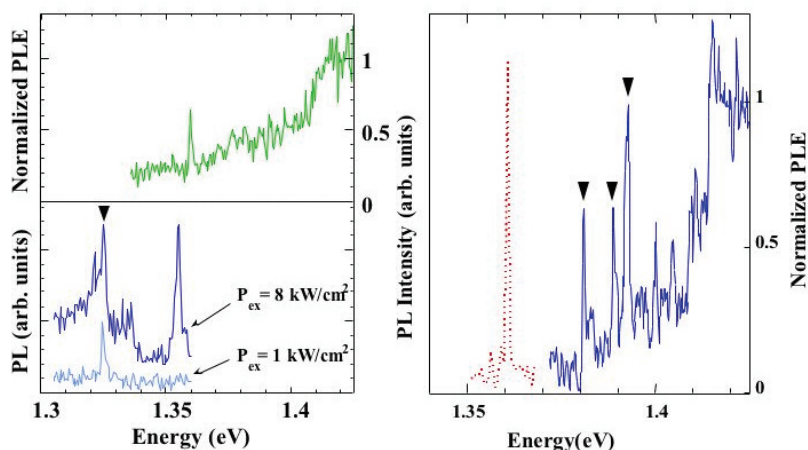


FIGURE 5

(Left panel) Photoluminescence (blue curves) and photoluminescence excitation (green curve) spectra of a single quantum dot. (Right panel) Photoluminescence (dashed) and photoluminescence excitation (solid line) spectra of a single quantum dot.

the P_e - P_h transition becomes observable approximately 30 meV above. This latter one is also present in the PLE spectrum (green curve, left panel), superimposed to the quasi-continuum related to the mixed transitions involving a confined electron and a delocalized hole or vice versa. On the right panel of Figure 5, the PL and PLE spectra of a single QD with different size and geometry are depicted. Here a few excited transitions are now observable. The absorption-onset of the wetting layer band occurs at the same energy for both QDs (1.42 eV).

In order to record a complete spectrum (PL or PLE), the acquisition time is a few seconds per point so a complete PL or PLE spectrum takes about twenty minutes. This requires a high mechanical stability of the whole setup. In particular, the thermal drift of the cryostat has to be minimized. In fact due to the spot size, the signal arising from a single dot disappears for a sample drift of about 1 μm . The **Microstat**[®]HiResII cryostat is ideal when long acquisition time measurements are required, as its stable design enables thermal drift as low as 0.15 μm per hour, at 4.2 K (after stabilization). This enables a complete spectra to be collected, without having to realign the optics.

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Microstat[®]Hires II

The **Microstat**HiresII is a high resolution helium continuous flow cryostat, which benefits from extremely low sample vibration (less than 20 nm) and sample drift at both constant and changing temperatures. This cryostat provides a wide temperature range from 2.7 K to 500 K and a very short working distance (as low as 2.2 mm).