TECHNICAL NOTE

TRCL is a High-Resolution Technique

Given the context of developing nanosciences, there is currently a growing interest in obtaining local information about light-emitting phenomena in condensed matter. In particular, dynamical information such as the luminescence decay time is of major importance to analyse the competition between radiative and nonradiative processes in materials and nanostructures designed to serve in optoelectronic devices.

However, while it is easy to measure the photoluminescence decay dynamics with a spatial resolution of a few hundreds of squared micrometers, achieving such measurements at the scale of a few tens of nanometers is still challenging.
Therefore, in most instances, TR-PL experiments only provide information that is integrated over a number of light-emitting nano-objects and over a number of competitive processes: radiative processes themselves, carrier transfer among different radiative or nonradiative centers, nonradiative carrier recombination...

In the case of individual nano-objects or of materials comprising a variety of light-emitting sub-structures, such integrated information is not detailed enough and leads to conjectural analyses of the recorded data.

On the other hand, thanks to a strongly localized excitation, TRCL permits to isolate (spatially and spectrally) the different light sources and to analyse their dynamical characteristics separately.

Interest of Temperature-dependent TRCL

Like for other spectroscopic techniques the possibility to use cryogenic temperatures has several beneficial effects.

First of all, by hindering thermally-enhanced nonradiative recombinations, low temperatures increase drastically the signal-to-noise ratio. For the same reason, the CL decay dynamics measured at low-temperatures are closer (if not exactly identical) to the purely radiative dynamics.
Indeed, the effective decay time, τ_{CL}, is related to the radiative (τ_R) and nonradiative (τ_{NR}) lifetimes by: $\tau_{CL} = (\tau_R^{-1} + \tau_{NR}^{-1})^{-1}$. In other words, when the sample is placed at low temperatures, the nonradiative lifetime becomes so long that the effective lifetime is close to the radiative lifetime. The latter is a precious piece of information, especially in the study of low-dimensional semiconductor systems such as quantum wells or quantum dots.

The second favourable effect of low temperatures is the reduction of emission linewidths. Whenever several optical transitions, corresponding to various recombination mechanisms, are close in energy, this linewidth reduction permits to separate these mechanisms. In CL, this point is quite often very profitable because the different mechanisms can correspond to different localizations in space. In TRCL, the temporal behaviours pertaining to these different mechanisms can be studied separately.

The third advantage of low temperatures is that it reduces the diffusion of carriers across the sample and generally favours optical recombination of localised excitons. Consequently, the CL images obtained are sharper: the resolution, specially in monochromatic detection mode, is enhanced.

There is also clearly an interest for experiments conducted under variable temperature conditions. This is particularly true when dealing with carrier diffusion/capture processes that can be studied by analysing the CL rise-times. Their behaviour when T is changed, along with the appropriate modelling, provides crucial information on diffusion coefficients, energy transfer mechanisms, activation energies...

TRCL is an Excitation Spectroscopy Technique

Contrary to the intuitive approach, cathodoluminescence, when seen as a mapping method, does not show, strictly speaking, where the emitted light comes from. In fact, it rather correlates, with very high resolution, the 2D coor-

dinates of the excitation spot with the emission of light by the sample, wherever this emission really takes place.
In other words, in CL, the high resolution is a unique characteristic of the excitation, not of the emission.

In many cases, especially in low-quality samples, where the carrier diffusion lengths are small, the
above remark is of little relevance: the CL emission indeed takes place where the electronic excitation has been

delivered, at least within the accuracy limited by the size of the generation volume and by the carrier diffusion lengths. Typically, a resolution of a few hundred nanometers is reached.

But in high-quality samples, the situation can be totally different. For example, for the complex pyramidal quantum systems of Ref. [1], wherever the excitation spot is placed on the pyramid, the quantum dot at the tip of the pyramid is efficiently alimented by scattered carriers and it efficiently emits light at a characteristic wavelength. Therefore the CL image shows an entirely "white" pyramid although it is quite clear that light does not come from the entire pyramid, but rather from a zone that does not exceed a few nanometers at the tip of the pyramid.
In this case of high-quality samples, the high diffusion length paradoxically destroys the spatial resolution of the CL experiment.

Luckily, "time is on our side": the temporal resolution provided by TRCL allows measuring the rising dynamics of luminescence signals at a given wavelength, as a function of the position of the excitation spot. It therefore provides an unprecedented insight into dynamical phenomena such as carrier diffusion and capture. In the above example, the rise-time of the CL from the tip-quantum dot is a growing function of the distance between the excitation spot and the tip and a simple model yields important characteristics such as diffusion lengths.

In summary, for the study of high-quality samples where the carrier diffusion lengths are large, the time-resolution is a necessary ingredient.
By careful analysis of luminescence rise-times under variable conditions of temperature and of distance from the emitting object, TRCL permits to characterize the ambipolar transport of electron-hole pairs (or excitons), which the other techniques (μPL – SNOM) cannot achieve.

[1] M. Merano *et al.*, Nature **438**, 479 (2005)

About the author

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Pierre Lefebvre is a senior researcher in Physics specialized in optical properties of semiconductors, nanosciences and nanotechnology. He was the founder and first director of the Center of Competence in Nanosciences for the South-West of France. Interested in testing and promoting new concepts based on nanophysics for novel applications in optics, information technologies, solar energy.