Quantitative Cathodoluminescence Opens New Areas of Investigation in Semiconductor Research and Production

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Introduction

The increasing demand for new opto-electronics devices such as solar cells, laser diodes (LD), and high-brightness light-emitting diodes (HBLED), combined with the economic necessity to achieve lower energy consumption levels and higher device yields, is motivating researchers to develop new materials. The semiconductor industry is actively looking for alternatives to silicon, for example, to address new niche market applications in power devices. Constant efforts employed to reduce production costs are leading manufacturers to grow GaN on silicon substrate, creating new technical challenges, especially regarding the control of defect density on wafer. For all these reasons many studies are being initiated to improve understanding of the fundamental physical properties and behavior of compound semiconductor materials used in quantum wells, quantum dots and nanowire-like structures. Cathodoluminescence (CL) is a spectroscopy method that can generate reliable, quantitative, and stable data for research as well as prepare a basis for quality control during production.

CL refers to the light emitted by a material under electron irradiation. The most common example of this phenomenon is a television using a cathode ray tube. When combined with a scanning electron microscope (SEM), CL is a powerful spectroscopy technique with analytical spatial resolution down to the nanometer scale [1]. The multimode imaging capabilities of the SEM enable correlation of optical properties (via CL) with surface morphology at the nanometer scale [2]. For example, CL can reveal defects that are invisible to other detection methods [3, 4] and is also capable of probing a single selected nanostructure. Generated by high-energy electrons, CL is often the method of choice to study wide bandgap semiconductors such as diamond, boron nitride, or aluminium gallium nitride. In addition, the innovative CL system of Attolight can be upgraded to a time-resolved mode that yields time resolutions below 10 ps allowing the study of carrier dynamics in nanostructures. This technique has been used extensively [5–10] to study gallium nitride based nanostructures. The behavior of charge carriers in structures can be important for evaluating elementary components in quantum computers [11, 12], optical quantum gates [13], or single photon sources [14–16] for quantum cryptography [17].

Conventionally, CL is a challenging technique generally carried out using unwieldy laboratory-assembled imaging devices. A SEM is required along with a CL add-on device consisting of a light-collection mirror and a detection system. These components must be assembled on site, which can result in cumbersome and unstable alignment plus interfacing difficulties. This is particularly the case with cryogenic positioning stages. The collection mirror (parabolic or elliptic) has a very limited field of view (several micrometers). Thus, when the sample is moved slightly, most of the signal is lost and realignment is required. In addition, when light is not emitted exactly at the focal point of the mirror, the collection efficiency is reduced, resulting in limited sensitivity.

The Attolight company makes CL available in a simplified system that integrates CL with both light and electron microscopy. It features a hyperspectral imaging mode that reveals ultra-trace impurities and crystallographic defects not visible using other imaging modalities. At production sites, it is expected to become a major tool for detecting defective materials at an early stage of the manufacturing process thereby generating significant savings.

Methods and Materials

The Attolight system is an independent spectroscopy system in which an optical microscope is embedded within the electromagnetic objective lens of a SEM, matching both fields of view. The optical microscope visually positions the

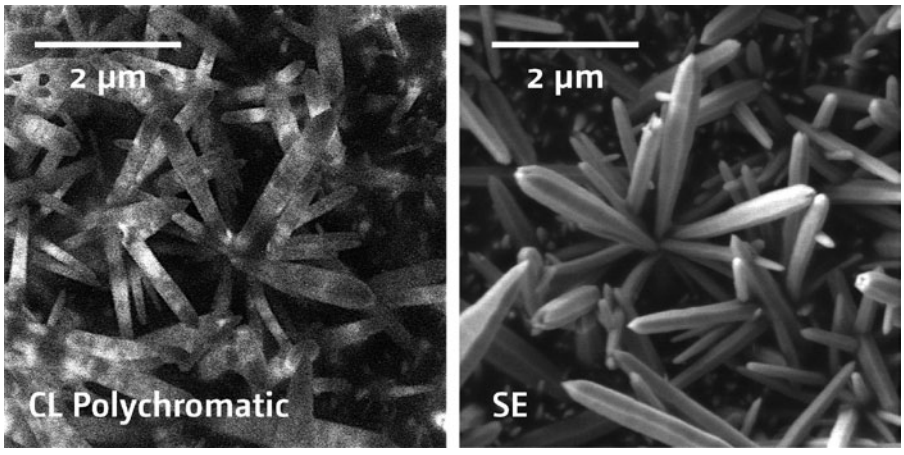

Figure 1: ZnO nano-structures. The acquisition of a CL micrograph (left) does not affect the detection of secondary electrons (right). Copyright 2012 Attolight AG.

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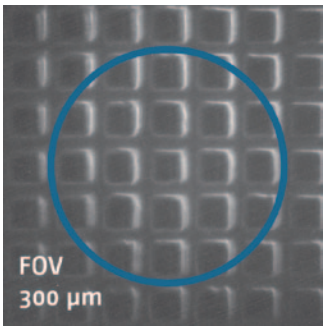

Figure 2: Attolight optical microscope features constant resolution and photon collection efficiency over a field of view of 300 µm. Copyright 2012 Attolight AG.

system so that no other optical alignment is required. This innovative design is optimized to achieve superior CL imaging without compromising SEM performance: secondary electrons are not obstructed by the collection optics (Figure 1). Integrated light optics allows consistent high-efficiency light collection over the entire field of view, and, thanks to a numerical aperture of 0.72 (f/0.5), the average collection efficiency over the 300 µm field of view is up to 100 times larger than for a conventional CL apparatus (Figure 2).

Quantitative measurements are not possible with the traditional parabolic mirror approach because the field of view is limited to a few microns and the optical image is plagued by blur and vignetting. However, with the Attolight design, quantitative CL measurements can be performed and data can be compared from one sample to another. In addition, the light optical system is achromatic from the ultraviolet to the infrared spectral range, allowing a wide range of applications.

For advanced applications requiring a deep understanding of the properties of a material or to boost the CL emission efficiency, an optional cryogenic nano-positioning stage can control the specimen temperature from 15 to 300 K. This facilitates the measurement of weak luminescent samples, providing low drift and low vibration. A proprietary mechanical design with 6 degrees of freedom ensures arbitrary sample positioning. The system also features, without degrading the spatial resolution, a time-resolved option for lifetime and charge-carrier dynamics measurements in which picosecond electron pulses are generated to excite the specimen.

Determination of Threading Dislocation Density in a GaN Template

The Attolight hybrid CL system is particularly efficient for characterizing defects in semiconductors. As an example, we display in the left-hand side of Figure 3 a secondary electron micrograph of the surface of a defective GaN template, taken with Attolight's proprietary SEM. The image reveals particles at the surface, but no threading dislocation (TD) is observed.

However, CL can detect these TDs. When an electron beam is focused onto a sample, it generates charge carriers, which recombine either radiatively (light emission) or non-radiatively (heat emission). The Attolight CL embedded optics inside the column of the SEM are optimized to collect the emitted light, the latter being detected by a photomultiplier tube (UV-visible). On the CL micrograph in Figure 3 (right), TDs appear as dark spots because these crystal defects act as efficient non-radiative recombination centers. This polychromatic CL micrograph reveals a high density of dark spots, which correspond to TDs. Simply counting the number of dark spots per unit area allows determination of the TD density (TDD), which in the present case is of the order of $3 \times 10^7/cm^2$.

Acquisition of both SEM and CL images allows for precise determination of the local density of TDs emerging at the surface of a GaN sample. Depending on the acceleration voltage, which is typically set between 3 and 10 kV, the impinging electron beam generates charge carriers in a volume with a spatial extension between 10 nm and 1 µm. Threading dislocations therefore can be potentially detected in a specimen down to a depth of 1 µm. Future developments can be envisaged to automate the measurement of TDD through image processing for large-scale manufacturing control purposes.

Local Lifetime Measurements in ZnO Nano Belts

Time-resolved cathodoluminescence (TRCL) can provide reliable evidence of high crystalline quality in isolated ZnO nano-objects [18].

Figure 4 shows continuous CL experiments carried out on a ZnO nano belt. Hyperspectral CL mapping shows that the CL intensity fluctuates along the length of the structure, with a dramatic reduction of the signal where the wire is slightly bent. In addition, when scanning along the axis of the nano belt, the emission peak wavelength shifts by approximately 2 nm revealing local strain fluctuations.

Photoluminescence experiments could not provide such a nanometer-scale characterization of ZnO nano belt emission properties. The spatial resolution of photoluminescence, limited typically to few microns, is therefore insufficient to rate the quality of single nanostructures. Unfortunately, continuous-wave CL alone does not reveal the respective variations of charge carrier non-radiative and radiative recombination lifetimes at the origin of the emission intensity fluctuations observed

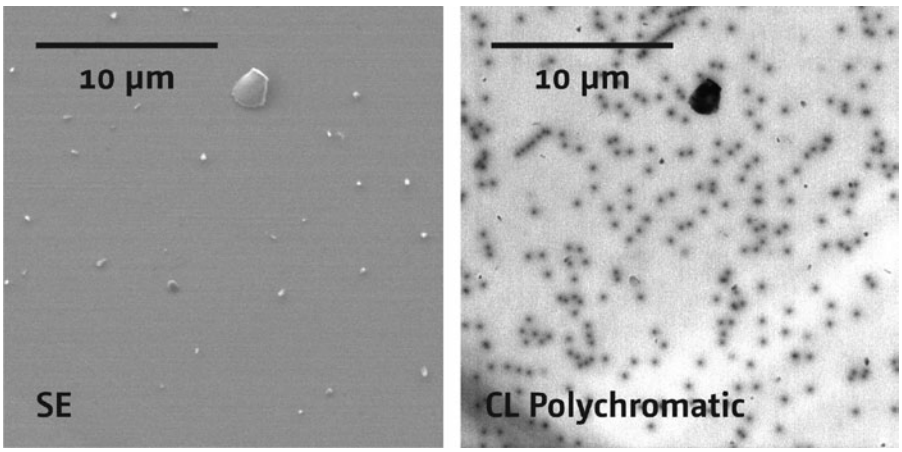

Figure 3: Left: SEM image taken on a polished GaN layer. Right: cathodoluminescence image showing threading dislocations in the GaN layer; dislocations appear as dark spots because of non-radiative recombination in their vicinity. On the secondary electron image (left) taken of the same region, no threading dislocations can be identified. Copyright 2012 Attolight AG.

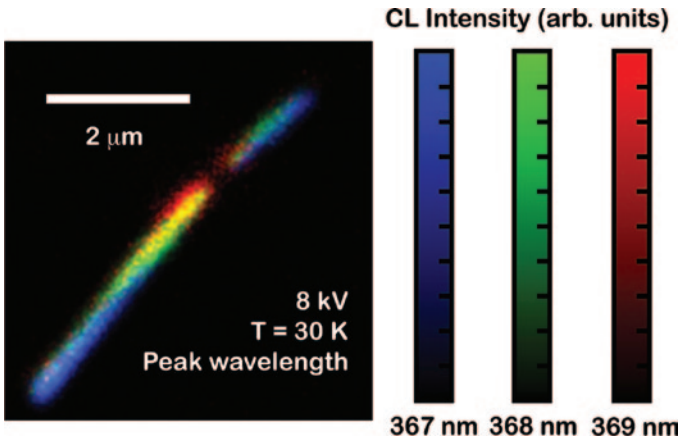

Figure 4: Continuous CL on a ZnO nano belt at T = 30 K. A drastic drop in CL intensity is observed where the nano belt is slightly bent. Compared to the extremities of the nanostructure, the emission is redshifted when the excitation is delivered in the vicinity of the region where CL intensity drops.

in Figure 4. Still, this knowledge is indispensable to understand what causes local intensity drops along the nano-belt axis.

Time-resolved measurements have therefore been carried out to investigate the spatial dependence of charge carriers radiative and non-radiative lifetimes. The general equations below (Equations 1 and 2) give the time evolution of the CL intensity $I_X(t)$:

$$I_X(t) \propto \frac{1}{\tau_r} \exp\left[-t\left/\left(\frac{1}{\tau_r}+\frac{1}{\tau_{nr}}\right)^{-1}\right.\right] \quad (1)$$

$$I_X(0) \propto \frac{1}{\tau_r} \quad (2)$$

where τ_r and τ_{nr} are the charge carriers radiative and non-radiative lifetimes, respectively. Equation 2 clearly shows that at t = 0 (that is, at the beginning of the decay) the CL intensity is inversely proportional to the radiative lifetime, τ_r. Measurements at different locations on the nano belt show that the CL intensity at t = 0 stays constant (Figure 5), leading to the conclusion that τ_r does not vary along the nano belt. In other words, the CL intensity drop observed in Figure 4 is only due to a local decrease of the non-radiative lifetime. In agreement, the CL decay taken where the nano belt is bent (green curve in Fig. 5) is the one exhibiting the fastest decay.

By measuring the variation in CL decay time along the nano-belt axis (Figure 6), and assuming that the internal quantum efficiency η is given by

$$\eta = \frac{\tau_{nr}}{\tau_r + \tau_{nr}} \quad (3)$$

it is possible to trace the evolution of η along the nano belt axis and to characterize locally the crystalline quality of the material.

Conclusion

The new CL method presented here is a powerful tool able to provide a deeper understanding into the quality of semiconductor devices. TRCL has proven to be an efficient

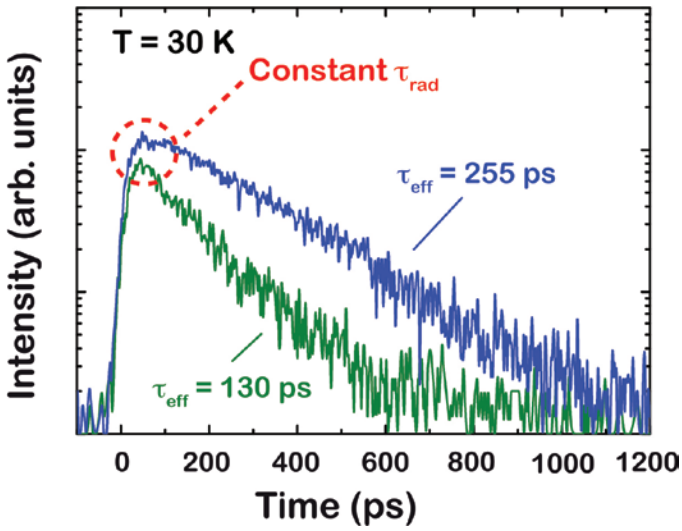

Figure 5: TRCL on a ZnO nano belt. The curves correspond to two different excitation spots along the nano-belt axis. The constant emission intensity at zero delay shows that the charge carrier radiative lifetime is constant along the nanostructure.

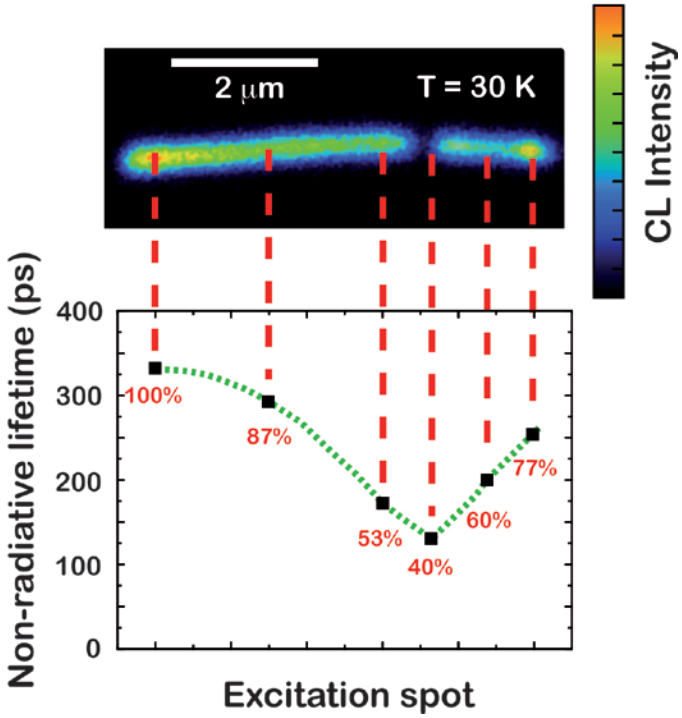

Figure 6: Top: Time-integrated CL mapping of a ZnO nano belt at 30 K. The signal has been spectrally integrated between 3.34 and 3.40 eV. Below: TRCL reveals non-radiative lifetime variations along the length of the structure. Where the structure is bent, we can clearly observe a steep decrease of the non-radiative recombination time, leading to a reduction of the CL emission intensity. From Equation 3, it is then possible to extract the internal radiative efficiency along the length of the nano belt (shown in red).

method to probe the dynamics of charge carriers, where photoluminescence appears to be ineffective because of the lack of spatial resolution.

Although most of our experience is so far in semiconductor applications, we think that our method can also open new possibilities in geology and biology. The fact that the system

can measure low signals, with better resolution, over a large field of view, all without any alignment, will certainly open new areas of investigations.

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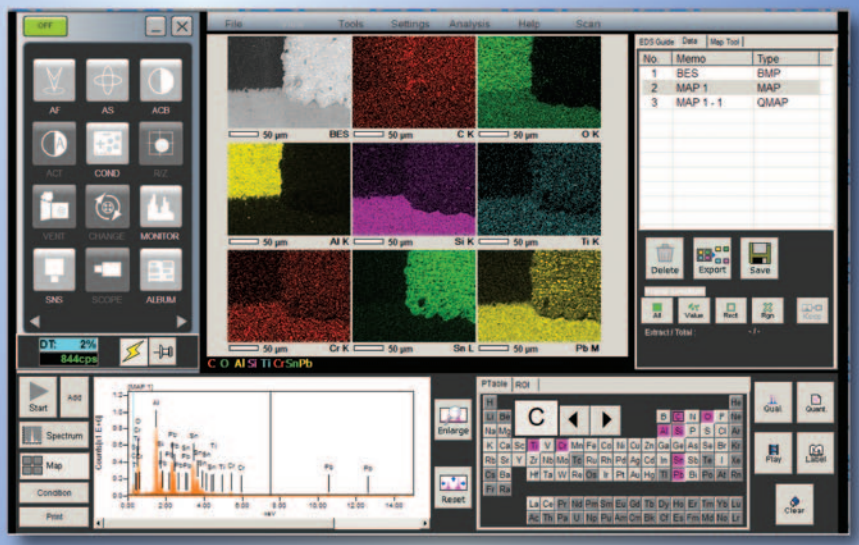
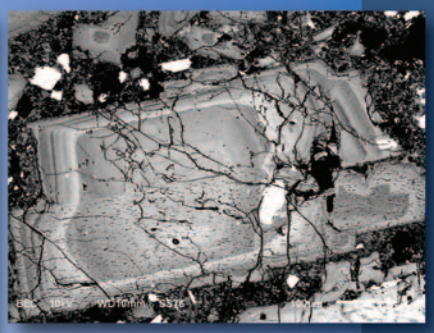
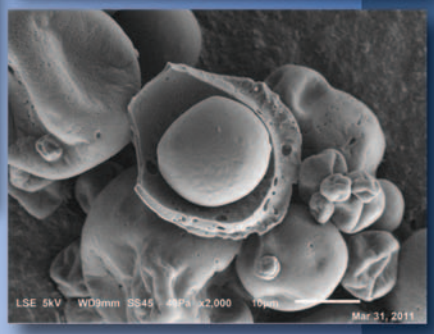
Defining a New Class of SEM







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