Technical Note

High Spatial Resolution Cathodoluminescence.

Introduction

A common theme throughout the history of electron microscopy has been the pursuit of high spatial resolution results. This is true whether in the field of SEM or TEM, and is also common to a variety of advanced micro-characterisation techniques. With different techniques there are different factors to consider. This is especially true in the field of cathodoluminescence (CL) where high spatial resolution results are a key desirable feature, especially when comparing the technique to other luminescence characterisation techniques such as micro-photoluminescence (μPL), scanning near field optical luminescence (SNOM) and scanning tunnelling luminescence (STL) based techniques. The spatial resolution with cathodoluminescence is determined by many different factors which include the specimen characteristics, the microscope column and operating conditions, as well as the CL equipment and associated cryogen cooling apparatus. This technical note explains some of these factors from first principles and covers a variety of applications of high spatial resolution CL imaging, spectroscopy and mapping.

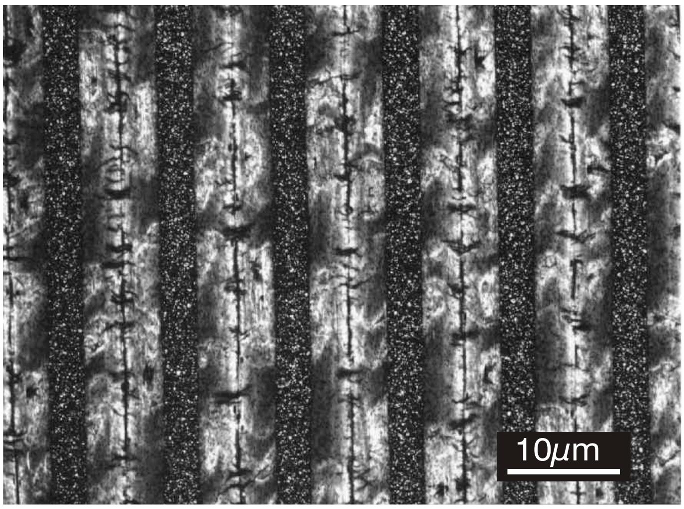

Figure 1. Panchromatic 2kV, 60pA beam current CL image of specially grown GaN material. [1]

Electron microscopy effectively bypasses the Abbé spatial resolution limit applied to conventional far field optical microscopy techniques, (e.g. SNOM, μPL) where the resolution is approximately limited to the

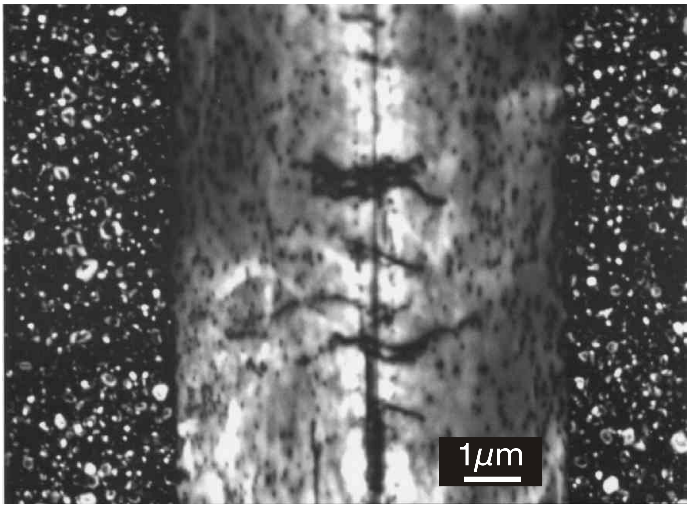

Figure 2. Higher magnification CL image of same GaN specimen showing dislocation defects in epitaxially overgrown layer. [1]

wavelength of the light used as an injection source. This resolution limit also applies to cathodoluminescence performed in an optical microscope in "flood gun" mode. Furthermore, an electron probe is extremely flexible due to the ease of scanning, focussing, and the large depth of field. This depth of field simplifies imaging specimens which are not flat, especially compared to near field techniques, where even small surface topography can limit the technique. The high vacuum of conventional electron microscopes can also provide an ideal environment for cryogenic cooling of specimens. In addition, modern environmental, or variable pressure SEMs are opening new fields of CL research, as the gaseous environment provides the means of studying specimens traditionally avoided with high vacuum microscopes at room temperature, e.g. food, biological and insulating specimens.

Spatial Resolution in the SEM.

For scanning electron microscopes the spatial resolution in standard (secondary electron) imaging mode is determined by the spot size which is rastered across the specimen. This is because secondary electrons have very little energy and are only emitted from an area and depth a few nm from the incident spot. The incident spot size is determined by the spatial and energy profile of the electrons emitted by the electron gun, the quality of the column electron optics and the working distance. Remarkable progress has recently been made in field emission gun and column performance in terms of reducing spot sizes at both low and high kV, even whilst providing high incident beam currents.

For variable pressure and environmental SEMs, the profile of the central spot and surrounding "skirt" have been measured as a function of gas, pressure and working distance. This data shows that for many typical working conditions, the density of irradiance of the skirt region is negligible compared to that of the central, un-scattered central spot, and this also applies to CL.[2] The condition here is that no artefact is within the skirt region which luminescent very strongly compared to a weakly luminescing feature of interest at the spot centre.

The Generation Volume.

In a modern FESEM, the spot size and hence SE image resolution can be optimised to be a weak function of the kV. However, for imaging or mapping using CL or X rays, the spatial resolution is strongly influenced by the kV. This is due to the dimensions of the generation volume within which photons are generated. The volume is somewhat larger for CL than for X ray photons because of the smaller energy associated with CL transition, i.e. an incident electron gradually losing energy through a series of inelastic scattering events can still generate low energy events over a wider volume compared to higher energy events. The relationship is not linear. Rather, the CL generation volume dimensions scale approximately as $kV^{1.7}$. Monte-Carlo simulations are a useful tool for understanding the shape and extend of the generation volume. It should be noted that these do not take into account diffusion or drift after the carriers have been generated.

For most specimens and experimental conditions the simplest criterion for obtaining high spatial resolution CL is to optimise working conditions at low kV. These conditions will include consideration of the specimen, microscope and operating conditions, as well as CL detection apparatus. There are three main consequences to using low kV for CL.

1. Lower kV injects less energy; in semiconductors this represents fewer electron hole pairs. The creation of a single electron hole pair requires approximately three times the band gap energy. The end result is always specimen dependent, but the overall effect is usually a reduced CL intensity with lower kV for a given injection density.

2. The generation volume is closer to the surface. The surface region is where non-radiative recombination can dominate, i.e. where the CL is effectively killed. This surface "dead layer" can be due to polishing damage, surface recombination or surface electric fields that separate electron hole pairs.

3. A smaller generation volume necessarily dictates a higher injection density. For a focussed spot this has a considerable effect because at low kV, the same flux is injected into a much smaller 3-D volume. A higher injection density does not always mean a higher CL emission due to saturation effects, and there are other consequences as explained below.

Injection Density.

As stated above a lower kV leads to a much higher injection density for a given incident beam current and this is an important point to consider. Although a certain activation injection density is sometimes required for luminescence, in many cases a high injection density is best avoided. This is because it can cause local non-equilibrium conditions, unwanted beam induced changes to the specimen, luminescence saturation and quenching effects, and in extreme conditions on cryogenic specimens, heating effects. Hence optimum conditions for high spatial resolution CL often involve both low kV and low beam currents. For CL experiments, such low injection conditions dictate the most sensitive apparatus, and can mean that cryogenic temperatures should be employed for optimum results.

A very simple way of reducing the injection density for non-high resolution spectroscopic characterisation is to defocus the beam. This procedure can also be used for obtaining non-high resolution spectroscopic information from beam sensitive materials by reducing the injection density but keeping the overall injection rate the same.

One distinction between using electron beam to other optical injection techniques is that with the latter, the optical absorption of the surface material (which may not be the region of interest) is important to consider. With a PL experiment the depth absorption profile is determined by the absorption coefficient with respect to the wavelength of the stimulating photons. However, for an electron beam, the interaction volume is determined by the generation volume and hence by the kV and the atomic weight and density of the material.

Working Distance.

Many SEMs offer the best spatial resolution at a short working distance especially at low kV. This condition influences the design of the CL apparatus for achieving high spatial resolution results. Gatan uses a precision diamond turned, off-axis, parabolloidal mirror which offers peerless collection efficiency and optical coupling. As suitable flanges are close to the height of the pole piece, and the working distance is required to be ~1mm below the bottom of the CL mirror, this results in an optimum 11-13mm working distance with the standard mirror.

Alternatively, a Short Working Distance mirror can be used which is 6mm high. This design maintains optimal collection efficiency, but does give a reduced field of view, which may limit lower magnification work. This design therefore gives ~7.5mm as a minimum working distance. For scanning TEM based CL systems a 3mm mirror is used. Note; microscopes without fine Z control on the specimen stage pose problems in optimising CL conditions unless a standard thickness of sample is adhered to.

Cryogenic CL.

Cryogenic temperatures are achieved using liquid nitrogen or liquid helium as a cryogen. With nitrogen, a cooled gas is used to cool the specimen stage to approximately 85K and with helium, a small but continuous flow of the liquid is used to reach temperatures as low as 5K. Cryogenic temperatures often enhance the CL emission very significantly. The amount of the effect depends on the physics of the luminescence. For luminescence associated with rare earth, an "extrinsic" impurity for example, in geological specimens, there is little or no effect. However for semiconductor specimens, the effect of cooling with liquid helium can be orders of magnitude. This is also true of other geological specimens, where an optimum gain in luminescence can be achieved with cryogenic temperatures of approximately 120K.

The general explanation is that radiative recombination becomes more favoured as the temperature falls compared to competing non-radiative recombination events. In addition for reduced dimension materials, the quantum confinement becomes enhanced thus leading to significant differences in the radiative recombination probability. Hence a large increase in CL at cryogenic temperatures can enable high spatial resolution simply due to the fact that a signal can be measured at the low injection conditions. In addition, the enhanced luminescence can reduce the amount of time required to obtain results, and therefore reduce the beam flux on the specimen, thus avoiding beam damage.

The engineering choice of cooled gas for nitrogen and liquid cryogen for helium temperatures respectively is due to the need to eliminate vibration to the specimen, which would impede any high spatial resolution work with the SEM. Liquid Nitrogen is not a suitable cryogen for high spatial resolution results using continuous flow liquid Helium cold stages due to the effects of boiling introducing specimen vibration.

Specimen considerations.

The discussion above has dwelt entirely on the apparatus. However, a key consideration for high spatial resolution CL is the specimen configuration itself. The primary concern is that the region of interest in the specimen (taking into account any surface dead layer) has to be accessible to the generation volume. For plan view specimens it is important that the region of interest has to be at, or very close to the surface. For quantum well specimens with subsurface quantum wells this can mean a specially grown thin capping layer, e.g. <30nm, or with the growth of devices interrupted to allow chosen regions to be accessible. High resolution CL results cannot be achieved if the region of interest in a plan view specimen is buried relatively deep in the specimen.

An alternative approach for semiconductor specimens is to perform measurements in cross section. Although this can give the best high spatial resolution results, this approach is invariably destructive and necessarily probes a much small area of material for analysis. In recent published research, "Challenging the Spatial Resolution Limits of CL and EBIC", Dr Carl Norman of Toshiba Research Europe Ltd explains in detail the conditions required to obtain optimum results and presents data from a purpose grown MBE sample, some of which are reproduced here.[3] The GaAs/AlAs/AlGaAs sample was designed to contain seven sets of three quantum well/four barrier combinations. The thickness of the wells and barriers in each set were 10nm, 20nm, 30nm, 40nm, 50nm, 60nm, and 80nm. The image shown in figure 3(a) was recorded at 2kV and clearly shows the 30nm barriers at the left hand side to be resolved. The linescan was recorded at 1.5kV shows the 20nm quantum wells to be resolved.

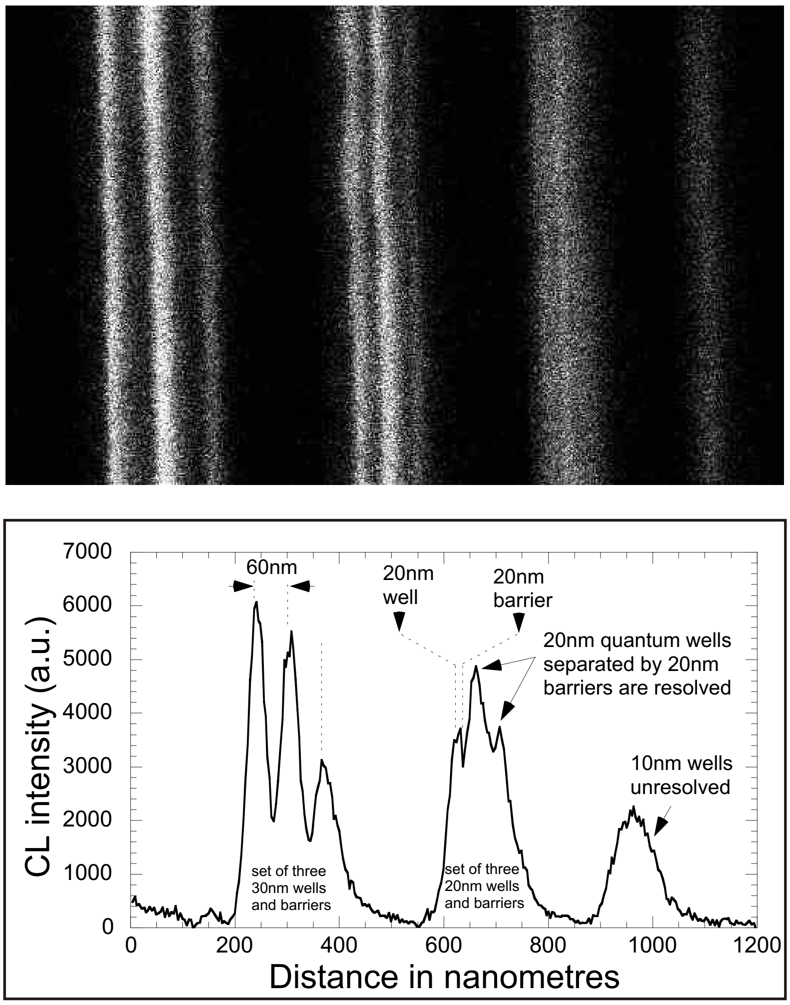

Figure 3. (a) 2kV CL image and (b) 1.5kV CL linescan of a cleaved sample cross-section containing a 30nm, 20nm and 10nm well/barrier sets.

Porous or low atomic weight specimens.

High spatial resolution results will usually be more difficult to achieve in low density or very porous materials. This is because the generation volume is determined by the atomic weight and density of the interaction volume. Such materials therefore dictate the lowest possible accelerating voltage and sensitivity of detection to obtain optimum results.

Topography.

Luminescent specimens which show surface structure, whether in the form of growth structures, etch pits or porous etching structures, asperities, or device structures will often show enhanced spatial resolution because generation, which also includes differences in back scattered electrons, will be influenced by the topography.

Absorption.

If a specimen which emits both short and long wavelength (high and low energy respectively) photons uniformly throughout the generation volume, the short wavelength photons reaching the detector will be more predominantly from the near surface region compared to longer wavelength photons. This is because higher energy photons are more likely to be reabsorbed compared to long wavelength photons. Hence this may lead to the curious effect of the long wavelength signal giving an image of lower resolution, thus suggesting greater homogeneity in the specimen compared to the short wavelength signal. This effect has been noted in GaN specimens where some debate existed as to the depth location and homogeneity of the longer wavelength "yellow-band" compared to the near band edge signal.[4] This effect is reduced where the generation volume is small and shallow so that the differences in re-absorption of different wavelength photons are minimized.

Surface dead layers.

"Dead" layers are where non-radiative recombination dominates over radiative recombination. When these exist at the surface, high spatial resolution

CL results are limited. This is because insufficient signal is generated in the dead layer at low kV. A dead layer is either due to surface damage associated with polishing, or oxide layers. In semiconductors, there is the additional possibility of surface electric fields which reduce the probability of radiative recombination. To maximise high spatial resolution CL, surface effects that kill luminescence, or cause astigmatism should be minimised. Achieving this aim will necessarily involve considerations of both the specimen and the vacuum environment of the microscope. It is best to assume that standard SEM and FESEM vacuums contain hydrocarbons, unless efforts are made to clean them, and maintain this cleanliness. As the ultimate solution of a UHV environment is considered excessive, it is worth noting that much can be achieved in a well maintained SEM, by using careful sample preparation and cryogenic anti-contaminators. In the future greater emphasis may be placed on in-situ, or chamber mounted specimen preparation equipment.

For geological specimens standard polishing techniques for optical microscopy, which do not eliminate fine scale surface abrasions, may limit conditions to greater than 10kV. However, more careful polishing techniques to give a mirror finish can be employed which allow lower kV results giving sub micron resolution.

The generation volume can be restricted in size by using thin film specimens. As well as a useful technique in the SEM, this is the principle behind using a scanning TEM to obtain very high spatial resolution CL. The upper limit to the spatial resolution will be specimen and kV dependent because a certain critical specimen depth will be required to produce measurable CL emission.

If the specimen is too thin, then non-radiative, surface recombination from both sides of the specimen may dominate, rather than radiative recombination. The light levels are necessarily low because of the small volume being stimulated and because of the relatively proximity of the surfaces which kill radiative recombination. In addition, excitons can have low radiative recombination probabilities as their separation can be similar to the dimensions of the film.

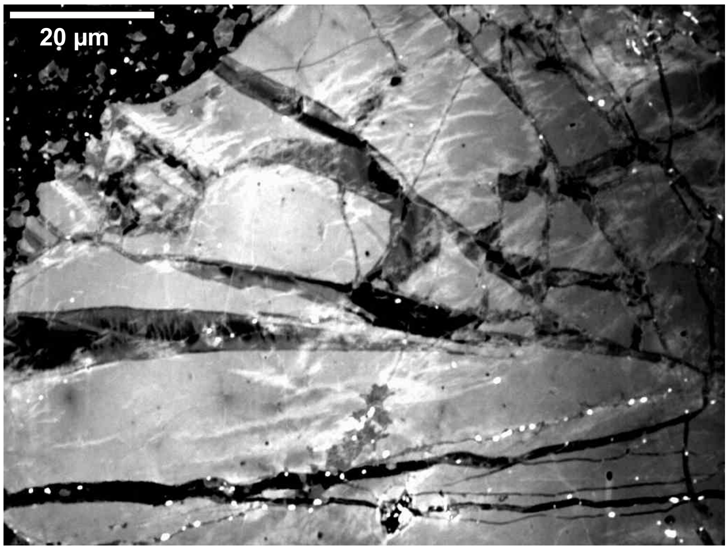

Figure 4. Panchromatic 6kV CL image of fractured and healed quartz.

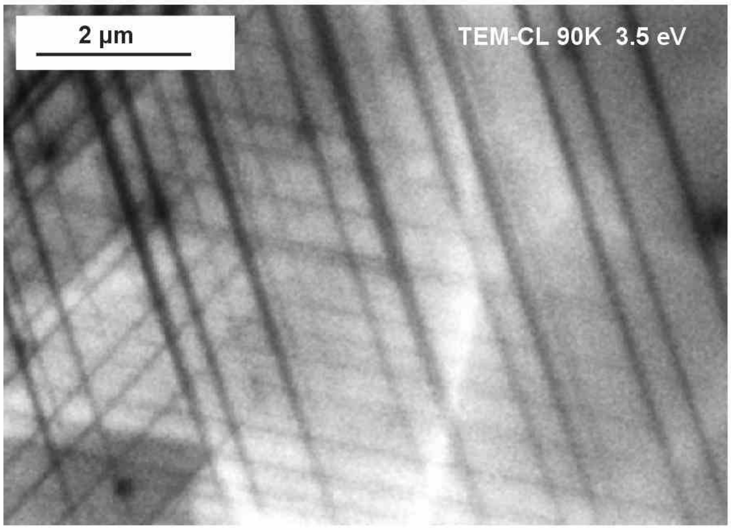

Figure 5. Monochromatic CL image from STEM CL system recorded using 170kV and 90K. Image shows dislocation slip bands. [5]

Carrier Diffusion and Drift.

Carrier diffusion occurs in a semiconductor in direct response to the perturbation of the equilibrium distribution associated with the injected beam. Drift only occurs in response to a local electric field. Diffusion and drift tend to smear out the sharpness of the generation distribution as modelled in Monte-Carlo simulations as carriers have a finite time to move about prior to recombination. Although this is often a small effect compared to considerations of the kV in determining the size of the generation volume, the size of the diffusion effect naturally depends on whether the diffusion length is longer or shorter than the dimensions of the generation volume. In general, the shorter the diffusion length the better,and this is a prime reason why materials such as Gallium Nitride allow good, high spatial resolution results.

Drift associated with electric fields can limit spatial resolution as well as causing the curious effect of luminescence being emitted from areas "remote" from the electron beam spot. As CL is recorded synchronous with the beam scanning, the image may not record strictly where the light is emitted.

Conclusions.

CL performed in an SEM or TEM offers opto-electronic characterisation with a flexible probe which is not restricted by Abbé diffraction limits, or near field restrictions. The spatial resolution is determined principally by the size of the generation volume. Other influences are associated with specimen characteristics, e.g. specimen configuration, diffusion lengths, emission and absorption characteristics, and surface effects. For best results using low injection conditions, optimal collection and detection apparatus is equally important as specimen characteristic. There are strong advantages in using a field emission microscope. However as the generation volume is important, good results can be obtained with care using traditional tungsten or LaB_6 guns which are optimised to focus to a small spot size at low kV.

Results recorded at less than 5kV in semiconductor specimens can show spatial resolution less than 100nm. Very low kV, topography or other specimen enhancements can reduce this figure to approximately 30nm. However, it is important to remember that although the spatial resolution may not reach quantum dimensions, spectrally resolved cathodoluminescence can detect variations down to atomic dimensions. For example in quantum wells, a slight change in alloy composition, or well thickness by one monolayer can be detected due to quantum confinement effects. In addition, electrically active impurities can be detected at concentrations many orders of magnitude beneath that which can be detected using X-ray techniques.

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References and Acknowledgements.

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2. Work reported by B. Thiel, Cavendish Laboratory, University of Cambridge.

3. C.E. Norman, Proc. BIAMS 2000, Solid State Phenomena Vols 78-79, 19-25, (2000), Scitec Pub Ltd, eds. Tomokage and Sekiguchi.

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5. TEM CL image courtesy of M. Albrecht and H.P. Strunk, University of Erlangen, working in collaboration with J. Weyher, I. Grzegory, S. Porowski, UNIPRESS, Polish Academy of Sciences, Warsaw, Poland and T. Wosinski, Institute of Physics, Polish Academy of Sciences, Warsaw, Poland.

Cathodoluminescence equipment from Gatan.

The note explains the reasons why low injection conditions are required to obtain optimum high spatial resolution results. The MonoCL3 (including ParaCL and XiCLone) and PanaCL systems from Gatan are designed with this principal in mind, where detection sensitivity is paramount, and system configuration to an intended microscope and application is standard. This is achieved with high precision optics, direct optical coupling to a chamber mounted monochromator (for MonoCL), and optimum sensitivity detectors.

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