Energy-momentum spectroscopy by wavelength- and angularly-resolved Cathodoluminescence

M. Bertilson, D.J Stowe, T Worsley, T. Franklin, L. Muray, M. Hsueh, J.A Hunt
Gatan Inc., Pleasanton, CA,
dstowe@gatan.com

Characterization of the optical properties of materials with spatial resolution better than the diffraction limit of light is required to further our understanding of light-matter interactions in nanostructured materials and devices. In recent years, the technique of cathodoluminescence (CL) – performed in the scanning- and transmission- electron microscopes – has gained great interest to study optical properties at the nanoscale due to the ability to excite optical processes with a sub-nanometer probe of (fast) electrons.

Light emission is defined by its distribution in energy (wavelength), momentum (angular), and polarization. Typically, researchers maximize the signal in one distribution by integrating over or filtering out the other two thereby eliminating information in the other two distributions; an issue that has become important in nanophotonics. Previous work, e.g. Yamamoto[1], demonstrated momentum and energy-momentum spectra in a CL setup. However, these experiments are limited to a single (selected) wavelength or angle respectively making a complete analysis of the wavelength and angular distributions impractical.

We demonstrate energy-momentum spectroscopy in a CL setup collected in a highly parallelized manner with high spatial, angular and wavelength resolutions (up to 10 nm, 1° and 0.1 nm respectively). In this demonstration, complete energy-filtered momentum spectra were reconstructed from samples using 30 energy-resolved momentum spectra; complete wavelength and angular distributions were acquired in <150 s. A 10 μm thick phosphor powder film (Figure 1) was found to emit light with wavelength spectra of identical form at all angles, however, light emitted from InGaN multi-quantum wells (optical source in high-brightness blue light emitting diodes) exhibited significant variance (Figure 2a and 2b). This can be ascribed to interference in the far field between light emitted from the excitation point and light reflected at the rear surface of the sample; simulation was found to agree with experiment (Figure 2c and 2d).

As researchers seek new ways to direct and enhance the radiation from light emitting and harvesting devices, such streamlined methods to collect energy-momentum spectra, as demonstrated here, will become increasingly important.

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[1] N Yamamoto, The Transmission Electron Microscope, Ed. K Maaz, (In Tech, 2012)

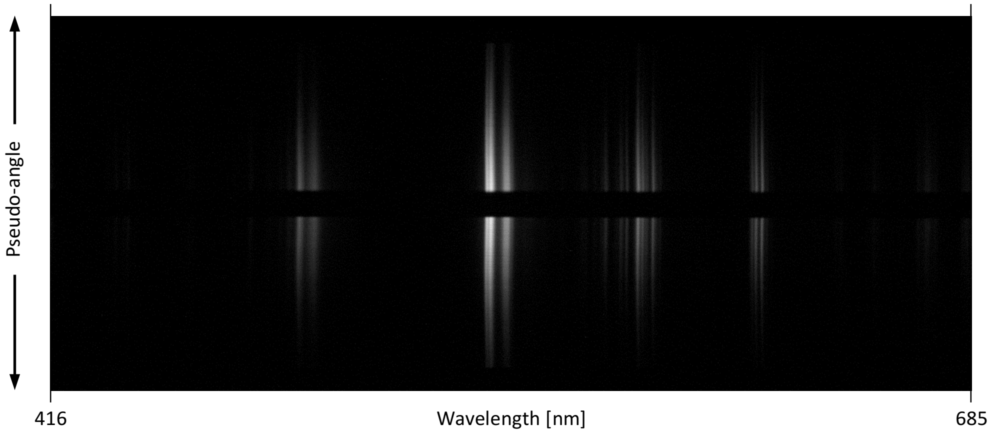

Figure 1: Energy-resolved momentum spectrum from a phosphor particle collected in a cathodoluminescence (CL) experimental setup: the focused electron beam of a scanning electron microscope is used to form a sub-nanometer probe to excite *d*-to-*f* shell optical transitions from a phosphor particle ~2 µm in diameter. The far field radiation pattern is collected by an off-axis parabolloidal mirror and coupled, via a spectrograph, to an array detector. The resultant 2D data set reveals the distribution of light - with full wavelength resolution - at 400 unique angles (zenith and azimuthal) simultaneously; the complete energy-momentum spectrum may be captured by scanning through the angular space. The angular distribution was found to exhibit isotropic color (wavelength invariance) at all angles.

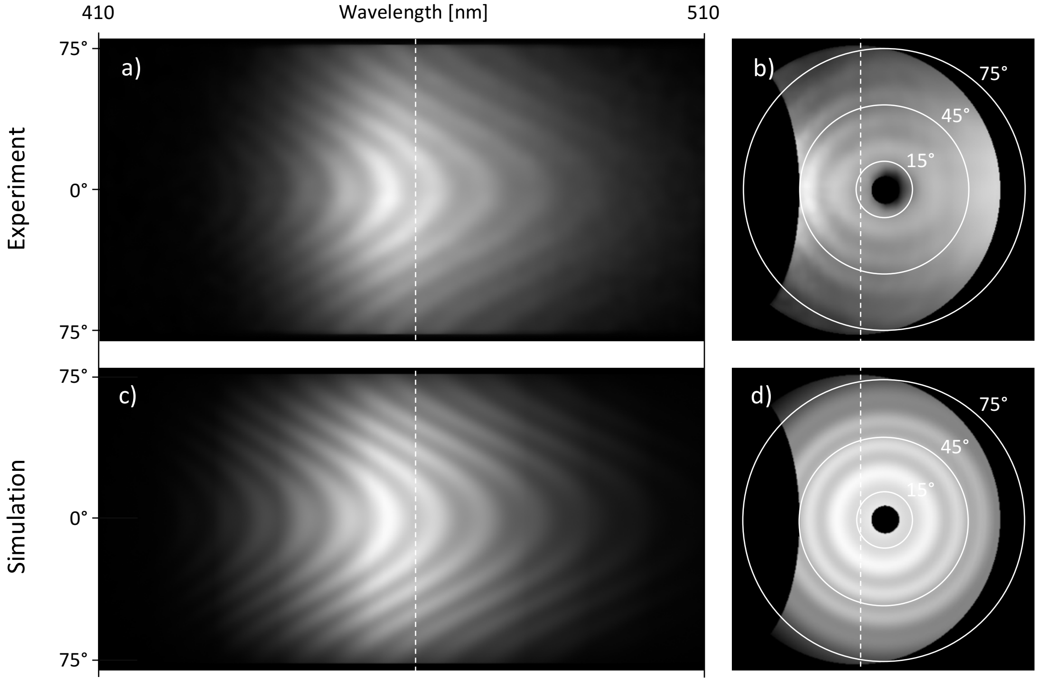

Figure 2: Reconstructed 3D energy-momentum CL spectrum of InGaN multi-quantum wells in GaN film demonstrating strong angular dependence of the wavelength distribution: a) and c) are momentum-filtered slices through the reconstruction at position indicated with the dotted line in b) and d); b) and d) are energy-filtered momentum spectra from the same data sets respectively displaying the angular distribution of light of wavelength at ~462 nm. The InGaN multi-quantum wells are situated ~100 nm below the surface of a 4.9 µm thick GaN film on a sapphire substrate. Simulated data was calculated from *A. S. Barker Jr. and M. Ilegems. Infrared lattice vibrations and free-electron dispersion in GaN. Phys. Rev. B 7, 743-750 (1973)* assuming an isotropic source distribution. The interference pattern was then calculated using Fresnel's equations and basic thin-film interference with a degree of coherence of 0.35.