Development of 3D Energy-Momentum Spectroscopy with High Wavelength- and Angle- Coverage and Resolutions using Cathodoluminescnce in the Scanning Electron Microscope

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Researchers are actively seeking new ways to direct and enhance the radiation or absorption of light-emitting and light-harvesting devices [1]. A promising approach is to utilize nanostructured surfaces or objects whose interactions with light may be modified through a change of their size, shape, or composition. Thus, the technique of cathodoluminescence (CL) in scanning- and transmission- electron microscopes has gained great interest due to the ability to excite optical processes in a wide range of materials with a sub-nanometer probe of (fast) electrons, correlating optical information with sample morphology, composition and crystal structure directly at the nanoscale [e.g. 2].

Light emissions are defined by their distributions in energy (wavelength), momentum (angular), and polarization. In most CL measurements, 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 the development of nano-structured optical devices. Previous work [e.g. 4] demonstrated momentum and energy-momentum spectroscopy in a CL setup. However, these experiments were limited to single, selected, wavelength or angles respectively making a complete analysis of the wavelength and angular distributions impractical. We demonstrate a new, highly parallelized, approach to capture energy-momentum spectra over >60% of the emission hemisphere using angular-resolved CL measurements: a nanometer-sized electron probe is used to excite optical transitions from a sample and the far-field radiation pattern collected by an off-axis parabolloidal mirror and coupled, via a spectrograph, to an array detector. The resultant 2D image reveals the distribution of light - with full wavelength resolution - at up to 400 unique angles (zenith and azimuthal) simultaneously (Figure 1); an energy-momentum spectrum with large angular-coverage is captured by scanning through angular space. We present apparatus capable of analyzing emitted (or reflected) light with high spatial, angular and wavelength resolutions (up to 10 nm, 1° and 0.1 nm respectively) over a wide angular range.

In this demonstration, 3D energy-filtered momentum spectra were reconstructed from samples using 30 energy-resolved momentum spectra captured in <150 s. A phosphor powder film (Figure 1) was found to emit light with wavelength spectra of identical form at all emission angles, however, light emitted from InGaN multi-quantum wells (MQWs) exhibited significant variance (Figure 2a and 2b). The MQWs were situated ~100 nm below the surface of a 4.9 m thick GaN film on sapphire substrate. The strong variance of the wavelength distribution as a function of angle could 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) assuming absorption and reflectivity coefficients of GaN on (0001) α-$Al_2 0_3$ from [4] and using Fresnel's equations and basic thin-film interference with a degree of coherence of 0.35.

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.

References:

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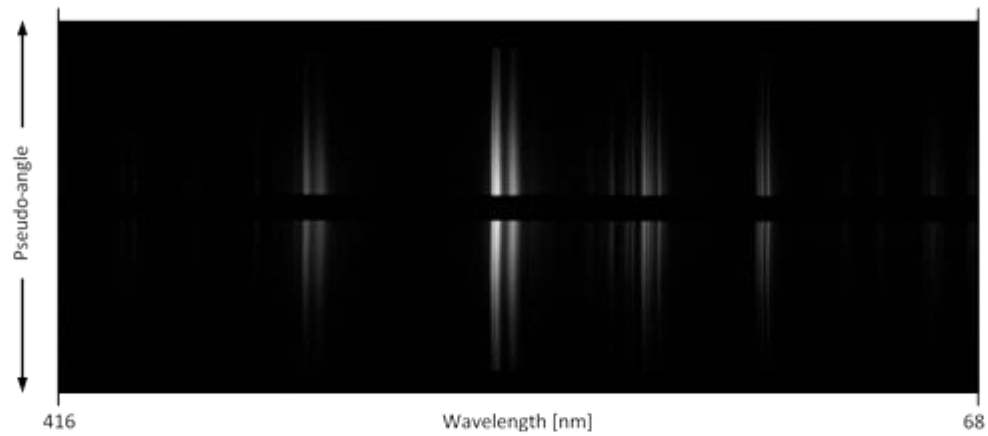

Figure 1. Energy-resolved momentum spectrum from a phosphor particle collected in a cathodoluminescence (CL) experimental setup. The angular distribution exhibits isotropic color (wavelength invariance) at all angles.

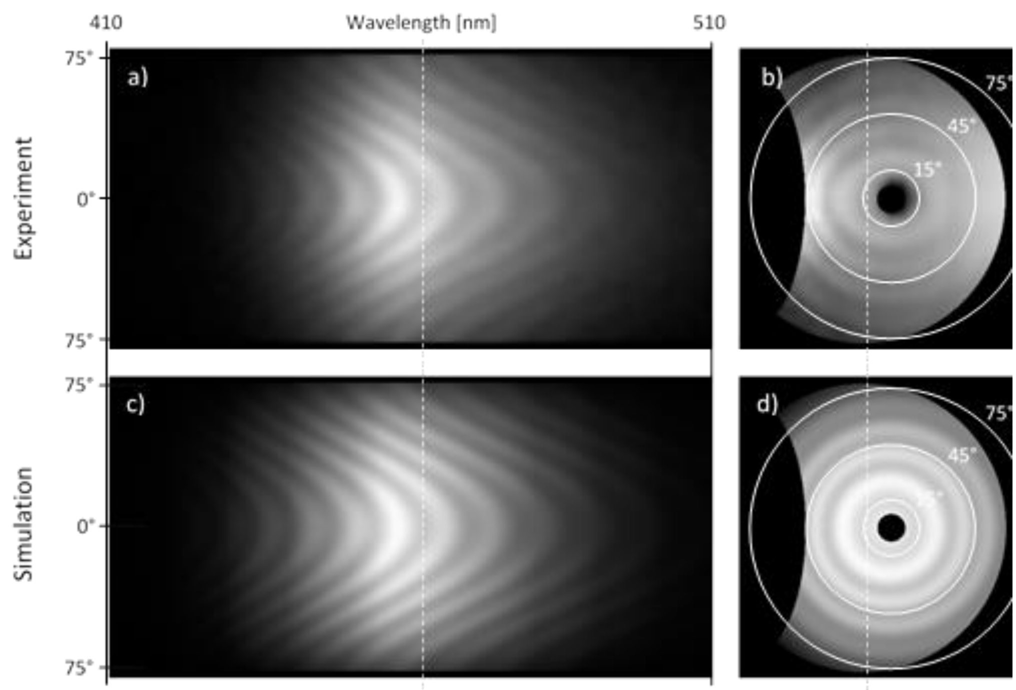

Figure 2. Slices extracted for the reconstructed 3D energy-momentum CL spectrum of InGaN multi-quantum wells in GaN film demonstrating strong angular dependence of the wavelength distribution: a) and c) are momentum-filtered energy spectra through the reconstruction at position indicated with the dotted 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.