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Next-Generation Photochemical Research Using Cathodoluminescence: Mapping Optical Properties and Resonances at the Nanoscale

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Abstract

This article explores the principles and diverse applications of CL, particularly in the analysis of inorganic compounds and nanophotonic structures. It examines various facets of CL, including aperture effects, transmission modes, resolution, momentum, scanning techniques, and modulation within CL imaging. The article underscores significant advancements in CL technology, highlighting its utility in characterizing nanostructures, plasmonic materials, and optoelectronic devices. The discussion emphasizes CL's role in mapping the Local Density of Optical States (LDOS) with high spatial and energy resolution, thereby advancing the understanding of light-matter interactions at the nanoscale. Emerging applications of CL in real-time monitoring of dynamic nanoscale processes are also explored, alongside its integration with advanced nanofabrication techniques. This integration facilitates enhanced characterization of quantum emitters and complex optical systems, pushing the boundaries of nanoscale optical research. Additionally, the article addresses the potential of CL to revolutionize optical sensing and imaging, offering transformative insights for future photonic device engineering. By leveraging its high spatial resolution and sensitivity to nanoscale phenomena, CL is positioned as a crucial tool in advancing both fundamental research and technological innovation. The review encapsulates how CL is evolving to meet the demands of cutting-edge applications, making it an indispensable technique in the ongoing exploration and manipulation of nanoscale optical properties and processes.

Keywords: Cathodoluminescence (CL), nanophotonic structures, local density of optical states (LDOS), quantum emitters, optical sensing

INTRODUCTION

Cathodoluminescence (CL) is a phenomenon wherein photons of potentially visible wavelengths are emitted when electrons strike a luminescent material, such as phosphors. This optical and electromagnetic process is analogous to the production of light in cathode ray tube (CRT) television screens, where an electron beam interacts with a phosphor-coated surface. Cathodoluminescence occurs as the reverse of the photoelectric effect, with photons being emitted following the interaction of an electron beam with a material. CL imaging, an advanced technique, captures the light emitted from materials under electron beam excitation, enabling precise intensity mapping. This technique is widely applied in various industries, particularly for studying

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inorganic compounds under scanning electron microscopes (SEM), including minerals, ceramics, and semiconductors. Despite its broad utility, CL is infrequently used for studying organic compounds.

Additionally, plasmonic cavities show a variety of modes, many of which are difficult to excite with other optical techniques and are "dark" to normally incident plane waves. However, despite their intrinsic interest, these modes have drawn attention because of their relatively high-quality factor and long lifetimes, which may serve as the foundation for new optical sensors with improved sensitivity and figure-of-merit compared to those that use "bright" dipole modes and emitters like quantum dots.. The interaction between the tip and the sample makes image interpretation difficult even though the technique offers better-than- diffraction-limited resolution. However, advancements have been made in the analysis of optical antenna modes using scanning probe techniques [1-5].

Cathodoluminescence

A common illustration is the production of light when an electron beam sweeps across the inner, phosphor-coated surface of a television screen that runs on a cathode ray tube. The opposite of the photoelectric effect, cathodoluminescence occurs when photons are irradiated, causing electron emission. With cathodoluminescence (CL) imaging, light emitted by an electron beam focused on a material is collected and measured. With this method, a single-pixel light detector is used to record the CL intensity for each beam position. While studying inorganic compounds under a scanning electron microscope, such as minerals, ceramics, and semiconductors, this method is frequently employed in numerous industries as a non-destructive approach. However, it is rarely used to study organic compounds.

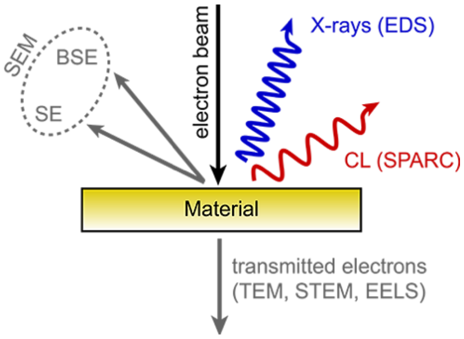

Figure 1. SEM Cathodoluminescence Imaging Microscopy.

Aperture Descriptive conducted a probe study on the azimuthal trimer configurations of rectangular slots within a gold film positioned on a glass substrate. This research highlights how the spatial variation of emission across different wavelength bands and the changes in cathodoluminescence (CL) signals collected from specific sample locations provide valuable insights into the resonant modes, especially the sub-radiant modes, of these apertures. By comparing experimental results with electromagnetic simulations, it becomes possible to identify a Fabry-Pérot mode associated with these cavities, as well as resonances linked to the excitation of surface plasmon polaritons at the air-gold interface. The findings present compelling evidence regarding the excitation of dark or sub-radiant modes of both individual apertures and aperture ensembles. This study builds on early investigations inspired by advancements in radar and microwave technologies, focusing on how electromagnetic waves interact with apertures in metallic films. The research also involved examining diffraction patterns produced by small circular [6] apertures in a very thin, perfectly conducting film. Later, extended provisional expanded the series with additional terms to create a modal method to account for apertures in films of finite thickness. Respect studied simple shapes like cross-shaped and coaxial apertures as well as more complex ones like rectangular slots and circular apertures. Reports indicating that optical transmission through periodic arrangements of holes in a silver film outperformed that of randomly arranged apertures with the same hole-area fraction have sparked considerable interest in sub-wavelength metallic apertures. This enhancement is closely linked to the organized arrangement of the apertures.

However, theoretical and experimental investigations of isolated rectangular slots and coaxial apertures have demonstrated that these cavities exhibit unique localized resonances that are highly dependent on the geometry of the holes, including the thickness of the metal film. The sharp Fano resonances arising from the interference between two modes present promising applications in areas such as sensing, switching, and lasing, in addition to the previously mentioned excitations. These resonances have been studied in a variety of structures, including ring-disk dimers, plasmonic oligomers, nanorod dolmen, and thin film nano-gratings. Coaxial apertures and dolmen nanocavities are two examples of nanoholes where Fano resonances have also been seen. It has been shown that an array of double split-ring cavities can function as biosensors using terahertz Fano resonances. By utilizing the resonant properties of the subwavelength apertures, nanoholes, like nanoparticles, serve as fundamental building blocks in a variety of nanophotonic devices, such as color filters and compact polarizers. The high sensitivity of the resonant modes of apertures to the refractive index of the surrounding media enables the development of highly efficient ultra-compact chemical and biological sensors. This includes plasmonic electrochemical sensors and Surface-Enhanced Raman Scattering (SERS) sensors, all of which can benefit from this sensitivity.

Transmission

Cathodoluminescence (CL) is an effective method for examining a material's optical characteristics. In recent years, it has shown great success in revealing new physics in the fields of plasmonics and quantum emitters when combined with scanning transmission electron microscopy (STEM). Due to conceptual and technological constraints, the majority of these results were not even conceivable twenty years ago. This objective is to outline the most recent breakthroughs that eliminated these constraints as well as the fresh opportunities provided by the current STEM-CL methodology. The various STEM-CL operating modes and the technical details of STEM-CL instrumentation were described. STEM-CL is used to investigate the two primary classes of optical excitations: coherent (typically plasmons) and incoherent (typically light emission from quantum emitters). Descriptive issue discussed the physics of light production under electron beam irradiation as well as the physical foundation for interpreting STEM-CL experiments for these two main classes. Then, we contrast STEM-CL with its more well-known methods, including photoluminescence, electron energy-loss spectroscopy, and scanning electron microscope CL [7-8].

A well-known phenomenon is cathodoluminescence (CL), or the emission of light from a material as a result of an interaction with an electron. It has been used to characterize semiconductors and geological sciences with great success as a standard characterization technique. It has experienced a significant rebirth in the last fifteen years as a result of the creation of optically active nanomaterials and nanostructured materials. The fact that an electron beam can be made arbitrarily small in comparison to the nanometer scale, where the three primary phenomena driving the optical behaviors of nanomaterials or nanostructured materials occur, is undoubtedly related to this resurgence. In other words, these phenomena occur at scales that are hardly accessible through the use of conventional far-field diffraction limited optical methods. As a contrast, contemporary scanning electron microscopes (SEM) and scanning transmission electron microscopes (STEM) can now create incredibly small electron probes (less than 1 nm) and gain from improved light detection techniques. Although such tiny probes can result in extremely small excitation volumes, high spatial resolutions are not always the result [9-12].

Resolution

To determine the factors influencing the spatial resolution of coherent cathodoluminescence imaging spectroscopy (CL), a deep-subwavelength imaging technique, researchers investigated the nanoscale excitation of silver (Ag) nanocubes. Localized plasmons in 70 nm Ag cubes are coherently excited by the 10–30 keV electron beam at 2.4 and 3.1 eV. In the far-field, the secondary electron intensity is also collected along with the radiation from these plasmon modes. CL line scans across the nanocubes show exponentially fading tails away from the cube, revealing the evanescent coupling of the electron field to the resonant plasmon modes. The measured CL decay lengths are in the range of 8 nm to 12 nm and vary by only 13 nm from the calculated ones, which range from 10 keV to 30 keV. A statistical model of electron scattering inside the Ag nanocubes is used to examine and compare the secondary electron pictures with the CL data.

With a systematic error of less than 3 nm, the Ag nanocube edges are obtained from the CL line scans. The results show that CL precisely measures the electron-induced plasmon fields. An established method for assessing the properties of optical materials at the nanoscale is cathodoluminescence (CL) spectroscopy. A two-dimensional map of the emission spectra at each electron excitation site is produced in CL by raster-scanning a material with a high-energy (1–100 keV) electron beam [13].

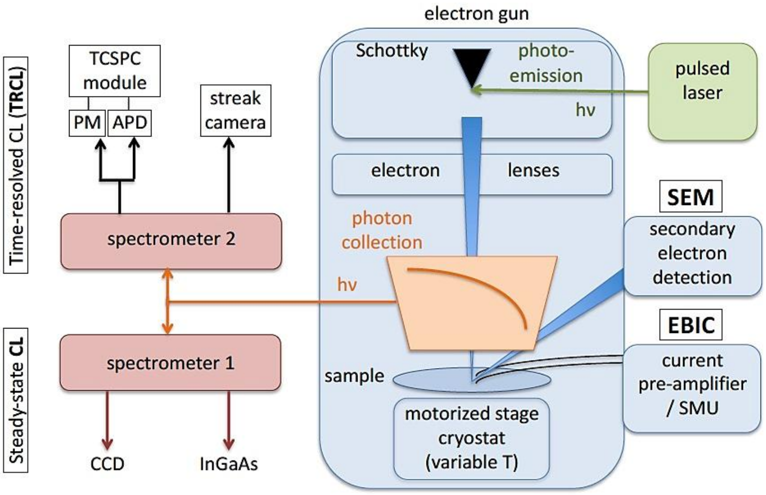

Figure 2. Cathodoluminescence (CL) and time- Resolution.

This method sheds light on optical properties at the subwavelength scale. The arrival of incoming electrons can cause incoherent emission processes like electron-hole recombination in semiconductors or color center emission in materials with wide bandgaps, which can lead to CL. On the other hand, in coherent CL excitation, the electron's time-varying electric fields cause a material to become directly polarized as it travels through it. The generated polarization radiates and is detected in the distant field. This coherent excitation is comparable to that of a laser beam that polarizes material and then scatters, except that the time-varying evanescent electron fields that link to the polarizable item only make one cycle in time [14]. The corresponding excitation energy spectrum spans a broad range from 0 to 40 eV, making high-energy electrons a valuable broadband source of excitation in the optical spectral range. Given that it offers a way to describe optical nanomaterials with three unique characteristics—nanoscale probe size, broadband Coherent CL has garnered significant attention in recent years because to its attosecond excitation pulse and excitation spectrum. Coherent CL has greatly facilitated a variety of activities, including measuring the local density of states in photonic crystals, hybridizing coupled nanoscale systems, and identifying optical modes in resonant plasmonic and dielectric nanostructures. A key subject that has not been sufficiently addressed up to this point is what essentially defines the spatial resolution of CL under realistic experimental conditions [15].

CONCLUSION

Cathodoluminescence (CL) has emerged as a transformative and highly versatile technique for probing the optical properties of materials with unprecedented precision at the nanoscale. By utilizing electron beams to excite luminescent materials, CL allows for the detailed investigation of phenomena that traditional optical methods cannot resolve. Its applications extend across a multitude of fields, including semiconductors, nanophotonics, and advanced material science, providing insights into the behavior and characteristics of materials at a level of detail previously unattainable. Recent advancements in CL technology, including enhanced imaging capabilities, improved resolution, and sophisticated momentum mapping techniques, have significantly broadened the scope and impact of this method. Innovations in modulation techniques have further refined the ability to analyze and interpret complex light-matter interactions, thereby enriching our understanding of material properties at the atomic and molecular levels.

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