

Differential Evanescent Light Intensity (DELI)

Principles, Validation, and Applications: A Review

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Abstract

The Differential Evanescent Light Intensity (DELI) method is a wide-field, label-free, non-contact optical nanoscopy technique designed for axial profiling of nanometer-thin material layers deposited on optical waveguide substrates. The technique exploits the evanescent field generated by total internal reflection (TIR) at the waveguide/air interface, which illuminates deposited nanostructures and produces far-field scattered light signals detectable in a wide-field optical microscope. The evanescent field intensity scales with the local layer thickness, enabling nanometric depth profiling across areas up to several hundred square millimeters. The physical basis of DELI is based on a multi-stage theoretical framework that spans TIR decay theory, phenomenological photon extraction models, and full-field electromagnetic simulations grounded in Maxwell's equations. Thickness reconstruction is performed pixel-wise through a discrete three-dimensional computational model, translating raw intensity maps into quantitative thickness distributions. Validation against atomic force microscopy (AFM), scanning electron microscopy (SEM), sputtering-rate calibration, and optical densitometry establishes an accuracy of 10–20% across diverse materials within a thickness range of 1–350 nm. The method is inherently diffraction-limited in lateral resolution and requires either a point calibration or the pre-tabulated materials optical evanescent constants to convert the relative response to absolute thickness values. To overcome the constraint of waveguide-type substrates, a complementary technique, Differential Reflected Light Scattering (DRLS) was subsequently developed, extending the method's applicability to a broader range of substrate classes. Together, DELI and DRLS form a versatile nanoscopy platform with promising future extensions, including multiwavelength spectroscopy, microfluidic integration, and advanced biological diagnostics.

Keywords: evanescent light, DELI, optical nanoscopy, total internal reflection, optical waveguide, nanoprobe, thin films, z-profiling, DRLS.

1. Introduction

The characterization of ultra-thin films and nanostructured surfaces is of high interest in modern materials science, photonics, and nanotechnology. Layers with thickness ranging up to a few hundred nanometers define the performance of semiconductor devices, metallic nanostructures, functional oxides, polymer coatings, and biological samples. While older established techniques for nanoscale surface profiling impose various operational constraints such as vacuum environments, substrate conductivity, in situ usage or slow throughput, the DELI method based on the evanescent phenomenon emerged explicitly to overcome these limitations. Developed systematically between 2007 and 2020 through a series of twenty publications [1–20], it has evolved from its fundamental proof-of-concept into a versatile profiling platform. Early investigations successfully established the technique’s physical foundations [1–3], exploring initial applications of environmental sensing [4] and investigation of morphology in semiconductors and polymers [5–10, 13]. Subsequent research significantly advanced the theoretical modeling of the evanescent field [11], facilitating analyses in metallic nanolayers [12, 17] and performing multi-material comparative studies [14, 15]. The method applicability was further expanded to include functional thin films such as zinc oxide [16] and titanium dioxide [18]. Maturation of the technique yielded a discrete, three-dimensional thickness model [19] and the development of a non-evanescent reflection-based variant, termed DRLS, used for profiling biological chromophore layers [20].

2. Physical Principles and Theoretical Framework

The physical foundation of the DELI technique rests on the phenomenon of Total Internal Reflection (TIR). When a light beam propagating within an optically dense waveguide medium of refractive index n_1 (such as glass) strikes an interface with a less dense medium of refractive index n_2 ($n_2 < n_1$) at an angle of incidence θ_i greater than or equal to the critical angle $\theta_c = \arcsin\left(\frac{n_2}{n_1}\right)$, no traveling light penetrates the second medium (n_2 is usually air). Instead, the beam undergoes TIR [1, 3, 12, 17, 21, 22]. To satisfy electromagnetic boundary conditions, a non-propagating, evanescent field forms on the n_2 side which decays exponentially with the perpendicular distance z from the interface according to the relation :

$$I(z) = I_0 e^{-z/d}$$

where I_0 is the optical field intensity at the waveguide surface, and d is a characteristic evanescent penetration depth. This penetration depth d of the evanescent wave is governed by the beam incident wavelength, the refractive indices of the media and the incident angle θ_i , [14, 17, 22]:

$$d = \frac{\lambda_{vac}}{2\pi\sqrt{n_1^2 \sin^2 \theta_i - n_2^2}}$$

where λ_{vac} is the vacuum wavelength of the light beam. For a standard glass waveguide with an index of 1.5 interfaced with air at incidence of the critical angle $\theta_c \approx 42^\circ$, the penetration depth evaluated for $\lambda_{vac} = 555 \text{ nm}$ is approximately $d \sim 79 \text{ nm}$, positioning the evanescent field within the operational range required for thin film profiling in the nanometer range [17]. The ability to tune this penetration depth by adjusting θ provides a direct control over the range of thickness range measurement and its axial resolution sensitivity.

When nanoparticles or continuous nanolayers are deposited onto the waveguide surface, they locally perturb the TIR condition. The deposited material couples with the evanescent field, extracting photons from the waveguide via scattering or evanescent tunneling in direct proportion to both the local layer thickness and the intrinsic optical properties of the material [1, 3, 17, 23, 24]. This extracted light propagates into the far-field and is captured by a conventional optical zooming microscope positioned above the waveguide, generating a spatial profile map of the evanescent light emission intensity. To quantify this phenomenon, the DELI photon extraction model relies on the assumption that deposited nanoparticles scatter evanescent photons in proportion to their volume number density N_a . This leads to a differential equation for the scattered evanescent field intensity I_z at any distance in the z -direction:

$$\frac{dI_z}{dz} = \gamma I_0 e^{-2z/d_m}$$

where I_0 is the base incident evanescent field intensity at the waveguide/film interface ($z=0$) and $I_0 e^{-2z/d_m}$ represents the decaying evanescent field intensity for $z > 0$. d_m is the characteristic penetration depth of the field into the deposited nanomaterial, and γ is the material's specific extraction parameter given by the product of the particles concentration and the effective evanescent scattering cross-section σ_{ev} :

$$\gamma = N_a \sigma_{ev}$$

Integrating the differential equation along the layer thickness h , yields the normalized detected intensity η :

$$\eta(h) = \frac{I_z(h)}{I_0} = \frac{1}{2} \gamma d_m (1 - e^{-2h/d_m})$$

where η represents also the total photon extraction efficiency of the deposited film. In the ultra-thin regime, layers are significantly thinner than the characteristic penetration depth ($h \ll d_m$), and since the γ and d_m product is usually of the order of $\gamma d_m \sim 1$, $\eta(h)$ can be well approximated in the end by a linear proportion with respect to the layer thickness h [1,3,17]:

$$\eta(h) \approx (1 - e^{-\gamma h}) \sim \gamma h$$

To rigorously validate this phenomenological extraction mechanism, full-field electromagnetic simulations were performed using the FullWAVE [25] Maxwell equation solver [8, 11]. Simulations of the scattered evanescent optical field from

amorphous selenium (a-Se) and polyethylene (PE) nanoparticles demonstrate that optical scattering by the nanoparticles, acting as electric dipoles, is the primary mechanism of evanescent light extraction [8, 26]. We found that while the volume number density N_a increases from elements like indium to palladium, the effective scattering cross-section σ_{ev} decreases with increasing molecular mass. This indicates that lighter-atom materials usually function as intrinsically stronger evanescent scatterers per molecule [17].

For materials exhibiting strong optical absorption, such as metallic layers [12], the photon extraction efficiency $\eta(h)$ is modified by the bulk absorption constant α , and producing then a competing interaction with γ :

$$\eta(h) = \frac{\gamma}{\gamma + \alpha} (1 - e^{-(\gamma + \alpha)h})$$

This relationship exhibits a definitive peak at a thickness h_{max} given by:

$$h_{max} = \frac{1}{\gamma} \ln \left(1 + \frac{\gamma}{\alpha} \right)$$

beyond which optical absorption dominates the scattering process, delimiting the functional dynamic range of DELI for absorbing metals. For highly transparent dielectric materials [6, 13], where α approaches zero, the extraction efficiency monotonically increases with h giving :

$$\eta(h) \approx 1 - e^{-\gamma h}$$

3. Instrumentation, Processing, and the DRLS Evolution

The experimental setup of DELI uses operational simplicity, functioning entirely at ambient laboratory conditions without requiring vacuum chambers, probe tips, or high-precision laser alignments [1, 3, 14, 17]. The core concept relies on a planar waveguide, typically a standard borosilicate glass slide or UV-transparent fused silica quartz, which simultaneously functions as both the deposition substrate and the light-guiding medium [5-8,11,16,18]. Light from a broadband visible halogen lamp is coupled directly into the polished edge of the waveguide, propagating along the substrate to generate a continuous evanescent field across the entire illuminated surface [5, 6, 8-18]. The evanescent light scattered upward by the surface nanostructures is captured by a conventional zooming stereomicroscope equipped with a CCD or CMOS camera matched to the spectral peak of the source.

Image processing relies on a standardized computational pipeline. Initially, a differential background subtraction is performed using a reference image of the bare waveguide to eliminate fixed-pattern noise and illumination non-uniformities. This critical differential step provides an inherently dark background, yielding high-contrast images where brightness directly correlates with material thickness. The image is then analyzed using the Normalized Integrated Optical Density (NIOD) metric

[1,3,4,9,10,12,14-17,19,20], defined as the spatial average of the optical density $D(x, y)$ over the sampled area S , which can be equivalently expressed in terms of the density distribution $H(D)$ as:

$$\text{NIOD} = \frac{1}{S} \iint_S D(x, y) \, ds = \frac{1}{S} \int_0^\infty D \cdot H(D) dD$$

To evaluate the relative and then the absolute surface nanoprofile thicknesses of the various samples from the evanescent captured images, the mean NIOD is recorded experimentally from the evanescent optical density of images from the deposited zones.

Quantitative thickness values h_i are then retrieved via a straightforward single or multi point calibration against an independent reference measurement h_{ref} according to the ratio:

$$\frac{h_i}{h_{ref}} \approx \frac{\text{NIOD}_i}{\text{NIOD}_{ref}}$$

This procedure has proven highly robust across dielectric, semiconducting, and metallic materials [1, 3, 9, 17].

A significant evolutionary step in the technique's instrumentation occurred with the introduction of Differential Reflected Light Scattering (DRLS) [20]. To overcome the standard DELI's requirement for waveguide substrates, DRLS illuminates the sample from above using an oblique light source beam situated near the Brewster angle [27]. This configuration suppresses specular reflection, mimicking the dark-field conditions of standard DELI while enabling thickness profiling across diverse material classes without the need for waveguide coupling. The mathematical model describing the z-directed backscattered intensity $I_z(x, y, z)$ in DRLS is formally similar to the DELI extraction model and its parameters:

$$\frac{\partial I_z}{\partial z} = \gamma[(1 - R)I_0 e^{-\alpha z} - I_z(x, y, z)]$$

where R is the upper surface reflectance and I_0 is the upper incident beam intensity [20]. Integration yields the detected intensity equation:

$$I_z(x, y) = (1 - R)I_0 \frac{\gamma}{\gamma - \alpha} (e^{-\alpha h} - e^{-\gamma h})$$

This equation indicates that the photon extraction model is governed here similarly by the nanolayer optical scattering rather than waveguide geometries, thereby useful for the extension of the method across various substrates, not necessarily waveguides [20]. γ is here the same material's specific extraction parameter and α its optical absorption constant.

4. Materials Investigated and Systematic Findings

DELI exhibits good versatility across a broad spectrum of deposition methodologies and material classes, profiling features from highly absorbing metals to completely transparent biological layers.

Amorphous selenium served as the foundational material system due to its controllable photodeposition process under UV-Visible irradiation [28]. Using KrF and ArF pulsed excimer lasers alongside continuous-wave Xenon lamps, DELI profiled a-Se layers ranging from 5 nm up to 340 nm [1, 2, 3, 5, 6]. The real-time nature of the deposition technique allowed direct observation of morphological evolution, tracking the growth rate and the transition from isolated spherical nanoparticles to the interconnected and final continuous films morphology as a function of incident fluence [5, 6].

Insulating dielectrics, polyethylene (PE) and polypropylene (PP) nanolayers deposited via matrix-assisted pulsed laser evaporation (MAPLE) and melt deposition were also profiled [7, 9, 10, 13]. DELI proved capable of differentiating between pure polymer matrices and those doped with amidon or cellofiber by variations in their evanescent contrast signatures [7]. For ultra-thin MAPLE-deposited PE layers, the effective photon-material interaction depth d_{eff} was determined to be approximately 130 nm. This depth allows for accurately capturing the large lateral surface peak periodicities and the spatial heterogeneity characteristic of complex polymer aggregations [13]. Further extending the analysis to wide-bandgap functional oxides, both zinc oxide and titanium dioxide thin films deposited by pulsed laser deposition were evaluated, demonstrating DELI's sensitivity for the deposition temperature induced variations in nanostructure crystallinity and density [16, 18].

Metallic nanolayers presented unique optical challenges successfully resolved by the phenomenological model. For electron-beam evaporated indium, the high optical absorption causes the DELI signal to peak at a specific threshold thickness, experimentally determined as $h_{\text{max}} \approx 20$ nm, beyond which the metal bulk absorption exceeds surface scattering [12]. Similarly, sputtered palladium layers were profiled in the shallow 1 to 10 nm thickness range [17]. Utilizing the discrete 3D model, the palladium thickness maps exhibited a good agreement with independent Atomic Force Microscopy (AFM) profile measurements [19]. Materials analyses combining data from indium, amorphous gold, palladium, selenium, and polyethylene established that the effective penetration depth d_{eff} increases monotonically from metals to semiconductors to insulators. This provides a suitable baseline of predictive parameters for characterizing other as-yet untested similar classes materials [12, 14, 15, 17].

The more recent transition to biological applications utilized the DRLS geometry to obtain surface profiles of photodeposited chlorophyll-a (Chl-a) and anthocyanin biochromophores on polymethyl-methacrylate (PMMA) substrates [20]. By analyzing the scattering light response, the experimental extraction parameter for Chl-a on PMMA substrates was established as $\gamma = 0.974 \times 10^5 \text{ cm}^{-1}$ closely matching the scattering parameters observed for synthetic PE [20]. This confirms the technique's applicability

also to soft organic structures and thus opening pathways for biofilm and point of care diagnostics in other organic materials as well.

5. Comparative Analysis with Established Nanoscopy Techniques

Comparing DELI with other nano-microscopy techniques helps clarify its place within this field. Although its lateral resolution is diffraction-limited to about 250–300 nm and cannot reveal the atomic-scale details seen by AFM or SEM, DELI offers clear advantages in speed and field coverage. Also, unlike serial scanning methods, contact-probe techniques, or electron microscopy, which requires vacuum operation, DELI enables non-destructive, wide-field fast speed z-profiling over hundreds of square millimeters at ambient conditions [1, 6, 9, 12, 14, 17]. In contrast to Total Internal Reflection Fluorescence (TIRF) microscopy, DELI depends exclusively on the intrinsic optical response of the deposited layer, and is label free [1, 6, 12, 17]. Its main limitation, however, is the need for at least one calibration point to convert the relative NIOD values into absolute thickness values. Nevertheless, when the tabulated values of γ and α are known for a given material, thickness profiles can be obtained directly from the theory without external calibration. A further limitation though is the signal saturation observed for highly absorptive metallic films when thickness exceeds the effective interaction depth of the evanescent field [12, 17, 19].

The demonstrated ability of DELI and DRLS to profile a wide range of layers, supported by the underlying extraction models, opens several paths for further development. Replacing broadband halogen illumination with tunable narrowband sources could enable multi-wavelength, depth-sectioned imaging, and allowing complex layered materials to be distinguished by their optical signatures. In parallel, integrating microfluidic systems directly onto the waveguide could enable real-time monitoring of dissolution, electrodeposition, and molecular adsorption in liquid environments [4]. Finally, overcoming the lateral diffraction limit may be approached through machine-learning-based image inversion trained on paired DELI and AFM data, potentially improving lateral resolution without altering the simplicity of the optical hardware.

DELI operates as a far-field, non-contact optical technique, enabling simultaneous wide-field vertical profiling over areas of hundreds of square millimeters without disturbing the native sample morphology. In contrast, AFM, despite achieving sub-nanometer resolution in both lateral and axial dimensions, is fundamentally limited by slow serial scanning, restricted fields of view, and reliance on physical probe contact [2, 6, 8, 9, 13, 19]. This proximity can also risk altering or damaging ultra-thin, soft, or weakly adhered nanostructures.

When compared to SEM and Transmission Electron Microscopy (TEM), DELI offers also distinct operational advantages for continuous process monitoring. Electron microscopy demands high-vacuum environments and frequently requires destructive sample preparation, including focused ion beam milling or the application of conductive coatings [1, 2, 3, 12, 20]. These preparatory steps can artificially inflate the

apparent thickness of sub-20 nm layers or completely obscure the delicate phase morphologies of the native material. DELI and its DRLS variant operate entirely under ambient atmospheric conditions without any sample modification, preserving the authentic structural state of the deposited layers.

DELI relies purely on the intrinsic optical scattering and absorption cross-sections of the deposited materials, making it a fully label-free technique [1, 6, 12, 17]. In contrast, TIRF microscopy, although it shares the evanescent illumination geometry and achieves similar axial selectivity within the 100–200 nm range above a substrate [29], fundamentally depends on fluorescent labeling. This requirement limits TIRF applicability to intrinsic inorganic systems such as amorphous selenium, zinc oxide, titanium dioxide, indium, and palladium, unless artificial tags are introduced, potentially altering the native morphology. DELI also distinguishes itself from optical ellipsometry, which, while providing highly accurate area-averaged thickness measurements, lacks lateral spatial resolution. By comparison, DELI generates high-contrast lateral maps of thickness variations, enabling visualization of spatial heterogeneities, localized agglomerations, and non-uniform coverage that ellipsometry would average out [9, 14, 17].

6. Quantitative Validation, Physical Insights, and DELI Constraints

The quantitative reliability of DELI is supported by systematic validation against multiple independent reference techniques, consistently demonstrating absolute thickness retrieval accuracies of 10–20% across the 1–340 nm dynamic range [6]. For amorphous selenium, AFM and SEM cross-sections of photodeposited regions directly corroborated the DELI-derived thicknesses. The integrated optical density increased monotonically with laser fluence up to the expected saturation point, capturing the transition from isolated nanoparticles to continuous films [6]. Polyethylene nanolayers deposited via matrix-assisted pulsed laser evaporation showed similar agreement, with AFM-based validation yielding consistent extraction parameters at absolute thicknesses of 83, 100, and 173 nm [13].

In the strongly absorbing metallic regime, DELI's phenomenological model was cross-validated using real-time quartz crystal monitors during electron-beam evaporation of indium. The measurements confirmed the theoretically predicted signal peak near the ~20 nm interaction depth, beyond which absorption suppresses the recoverable scattering signal [12]. Sputtered palladium nanolayers were evaluated using both sputtering-rate equations and AFM topography, and a discrete 3D computational reconstruction achieved pixel-level agreement with AFM within 15% [17, 19]. For wide-bandgap functional oxides, optical spectrometry further supported DELI's accuracy, confirming its sensitivity to deposition-temperature-dependent density and crystallinity variations in zinc oxide and titanium dioxide [16, 18].

Beyond static profiling, DELI has also proven effective for real-time monitoring of thin-film growth kinetics. By sequential imaging without altering the experimental configuration, it enabled continuous extraction of a-Se growth rates, revealing the

progressive shadowing of the waveguide surface during extended photodeposition [1, 3, 6]. A broader physical insight emerging from all studied materials is the trend in effective evanescent penetration depth. It increases from highly absorbing metals (~20–70 nm for indium and palladium), to semiconductors (~100 nm for amorphous selenium and zinc oxide), and reaches its maximum in transparent dielectrics (~130 nm for PE) [12, 15, 17]. Within metallic systems, the effective scattering cross-section also decreases with increasing atomic mass, indicating that lighter elements provide a higher density of scattering dipoles per unit volume [17].

Despite these strengths, the method must be understood within its physical limits. DELI remains diffraction-limited laterally, making it effective for nanoscopic vertical profiling but unable to resolve atomic-scale lateral features. It also requires transparent waveguide substrates capable of sustaining total internal reflection, though this restriction is partially addressed by the DRLS variant [20].

7. Conclusions and Future Research Trajectories

The systematic thirteen-year development of the DELI framework has firmly established it as a practical, non-contact, and label-free optical nanoscopy system. By combining large-area field-of-view imaging with nanometric axial sensitivity, DELI offers an inexpensive and robust solution for thin-film profiling under ambient conditions. A key evolutionary step was the mathematical unification of the DELI extraction model with the DRLS model, demonstrating that z-directed nanoparticle scattering can be exploited independently of specific waveguide geometries and no longer requires substrate-coupled waveguide designs [20].

Looking ahead, the introduction of tunable narrowband spectroscopic sources offers a promising direction. Transitioning from broadband halogen source illumination to multiwavelength excitation will enable more advanced depth-sectioned vertical profiling. Because the evanescent penetration depth depends on wavelength, sweeping the source spectral range would allow reconstruction of vertical concentration gradients and spectral discrimination of heterogeneous materials on a single substrate, effectively extending DELI into a chemo-morphological diagnostic tool.

Integrating custom microfluidic cells directly on the substrate would further allow continuous, real-time monitoring of liquid-phase processes. Building on initial environmental corrosion studies [4], such integration would enable in-situ observation of molecular adsorption, dissolution kinetics, and electrodeposition, without the technical challenges associated with liquid-mode AFM. Operating without vacuum conditions or fluorescent staining, DRLS may well be suited also for rapid detection of chromophores, protein monolayers, cell membranes, and early-stage biofilm formation [20]. In parallel, recent investigations of biological chromophores highlight an additional pathway toward medical diagnostics [30,31].

Finally, modern deep-learning inversion methods trained on the paired DELI–AFM dataset accumulated throughout this series may offer a computational route to surpass

the diffraction limits, improving the effective resolving power in nanometric profiling without altering the underlying optical hardware.

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