From the visible to the near infrared: revealing the optical response of J-aggregates thin films of cyanine dyes

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Abstract. J-aggregates thin films of cyanine dyes are very attractive organic nanomaterials formed by highly ordered assembly of supramolecular organic structures. These films show unique spectroscopic properties, making them promising candidates for integration into advanced electronic and photonic devices. Their spectra show very narrow resonances, resulting in spectral regions with high refractive index, comparable to semiconductors, and negative permittivity, similar to metals. This study focuses on the characterization of the optical properties of cyanine J-aggregates thin films, including refractive index and permittivity, in a wide spectral range from visible to near infrared wavelengths. Our findings highlight the potential of these structures for the development of sustainable photonic devices.

Cyanines are a well-established family of dyes known for their chemical stability, water solubility, and tuneable optical properties that extend from the visible to telecommunication wavelengths [1]. These characteristics make them highly promising for applications in photovoltaics [2], bioimaging [3], optical devices [4], and sensors [5]. Cyanine molecules can self-assemble into J-aggregates, a configuration in which the dye monomers stack head-to-tail. This unique arrangement induces strong interactions between transition dipoles that highly depend on the packing arrangement [6]. As a result, J-aggregates exhibit strong excitonic effects due to the formation of delocalized excitons, leading to very narrow, well-defined excitonic resonances [6,7], and red-shifted absorption spectra compared to their monomer counterparts. Furthermore, the ability to control their self-assembly in both solutions and on solid supports opens new perspective for creating multifunctional nanocomposites. These composites can combine light harvesting, wavelength conversion, superquenching, and optical nonlinear effects, offering efficient applications in various photonic and optoelectronic fields [6].

The presence of these narrow excitonic resonances plays an important role in altering the material's permittivity, resulting in negative values in the real part of the dielectric function at specific wavelengths. To further explore these optical properties, we performed

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ellipsometry measurements on J-aggregate thin films at room temperature using a variable-angle spectroscopic ellipsometer (VASE) from J.A. Woollam. Additionally, normal-incidence transmission spectra were recorded with the same instrument. The measured wavelength range extends from 300 nm to 1700 nm (0.7 to 4 eV), which covers the near-infrared (NIR) region relevant for telecommunications. This broad range allows for a more comprehensive analysis compared to previous studies [8,9], providing deeper insight into the material's optical behaviour across the visible and infrared regions.

Ellipsometric data provided the refractive index (n), extinction coefficient (k), and both real and imaginary components of the dielectric function (ε_1 and ε_2) for a series of J-aggregates thin films. The extended wavelength range was particularly important for analysing the refractive index in the NIR region, crucial for understanding the material's performance at longer wavelengths. We also analysed the Reststrahlen band, where the real part of dielectric function becomes negative, providing insight into IR interactions with electromagnetic waves.

Through the simultaneous analysis of spectroscopic ellipsometry measurements and transmission spectra, this in-depth study offers a comprehensive characterization of the optical properties of J-aggregate thin films, providing valuable insights for their potential applications in optoelectronics and telecommunications.

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