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**Optical nonlinearities and excited state dynamics of self-assembled cobalt  
phthalocyanine multilayer films**

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**Abstract:** An ultrathin composite films containing cobalt phthalocyanine (CoPc) and poly(diallyldimethyl ammonium chloride) (PDDA) were fabricated through layer-by-layer assembly technique. The resulting films were characterized by UV-Vis spectroscopy and atomic force microscopy. Their third-order nonlinear optical properties and photo-excitation dynamics were investigated with 20 ps laser pulses at 532 nm. The results demonstrate that the ultrathin films have notable third order optical nonlinearities ( $\chi^{(3)} \sim 4.7 \times 10^{-9}$  (esu)) and fast excited-state relaxation ( $\tau \sim 67 \pm 6$  ps), indicating their potential applications in future nonlinear optical devices.

**Key words:** Organic; Thin films; Optical materials and properties

## 1. Introduction

Nonlinear optical (NLO) materials have attracted considerable research attention in recent years due to their potential applications in optical switching, laser protection, etc [1]. Phthalocyanines (Pc) and their derivatives have been subjected to intensive studies on nonlinear optics because of their architectural flexibility, chemical stability and large NLO properties [2,3]. For practical applications, it is essential to obtain solid materials, like thin films, in which the NLO properties of the chromophores are preserved [4-7]. Therefore, the NLO properties of Pc films have proved very fruitful

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