Third-order Nonlinear Optical Properties and Crystal Structures of *N*-(2-Nitrobenzalidene)-2,4-dimethylaniline and *N*-(3-Nitrobenzalidene)-2,4-dimethylaniline

Aslı Karakaş[a], Hüseyin Ünver[b], and Ayhan Elmalı[c]

[a] Department of Physics, Faculty of Arts and Sciences, Selçuk University, TR-42049 Campus, Konya, Turkey
[b] Department of Physics, Faculty of Sciences, Ankara University, Tandoğan, TR-06100 Ankara, Turkey
[c] Department of Engineering Physics, Faculty of Engineering, Ankara University, Tandoğan, TR-06100 Ankara, Turkey

Reprint requests to Dr. Hüseyin Ünver. Fax: +903122232395. E-mail: unver@science.ankara.edu.tr

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N-(2-nitrobenzalidene)-2,4-dimethylaniline (**1**) and *N*-(3-nitrobenzalidene)-2,4-dimethylaniline (**2**) have been synthesized and characterized by X-ray diffraction analysis. Linear optical characteristics have been evaluated theoretically using the configuration interaction (CI) method. The maximum one-photon absorption (OPA) wavelengths of the studied compounds are shorter than 450 nm, giving rise to good optical transparency in the visible and near IR regions. To provide an insight into the third-order nonlinear optical (NLO) behavior of the title molecules, both dispersion-free (static) and frequency-dependent (dynamic) linear polarizabilities (α) and second hyperpolarizabilities (γ) at λ = 825 – 1125 nm and 1050 – 1600 nm wavelength ranges have been computed using the time-dependent Hartree-Fock (TDHF) method. The *ab initio* computational results on (hyper)polarizabilities reveal that both compounds exhibit second hyperpolarizabilities with non-zero values, implying microscopic third-order NLO behavior.

Key words: Third-order Optical Nonlinearity, One-photon Absorption, First Hyperpolarizability, Second Hyperpolarizability, Configuration Interaction