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Second-order nonlinear optical properties of composite material of an azo-chromophore with a tricyanodiphenyl acceptor in a poly(styrene-*co*-methyl methacrylate) matrix

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ABSTRACT

The composite material of new synthesized 4-((4-(*N*,*N*-n-dibutylamino) phenyl)diazenyl)-biphenyl-2,3,4-tricarbonitrile (GAS dye) in commercial poly(styrene-*co*-methyl methacrylate) (PSMMA) was prepared, poled and its nonlinear optical properties compared with DR1 dye were studied. High thermal stability of the composite material was revealed, and the maximal concentration of the chromophore was found to reach ~20 wt%. The dipole moment, polarizability tensor, and first hyperpolarizability tensor of the investigated dyes were calculated by within the framework of the coupled perturbed density functional theory. A nanosecond second-harmonic generation Maker fringes technique was used which is capable of providing the magnitude of the second-order nonlinearity of optical materials at a wavelength of 1064 nm. For the tested GAS–PSMMA composite material, maximal coefficient d_{33} was found to be 50 pm/V. The nonlinear optical response, which was achieved here, shows possible usefulness of the GAS dye as a component for molecular design of nonlinear-optical materials with advanced characteristics.

1. Introduction

Researches on nonlinear-optical materials based on donor–acceptor organic chromophores are hot topics of modern science [1]. The use of chromophore–polymer films make it possible to create compact low-voltage electro-optic modulators for communication systems based on optical fibers or integrated optical planar waveguide components [2–4]. The optical communication systems on the base doped polymer or glass fibers are operating in the wide from the visible to infrared spectral ranges [5,6]. Developing chromophores with high second order nonlinear response in visible spectral region is very important for the broadband terahertz pulses generation by optical rectification, in particular with Ti-Sa laser radiation in 750–800 nm range [7]. The chromophores with absorption of λ_{max} ~500 nm have a very good relation between absorbance losses and nonlinear optical response in this region. In this regard, studies on the second-order nonlinear optical properties of film materials with appropriate donor–acceptor chromophores in the visible and near-infrared spectral region represent a currently relevant field of research.

The efficiency of the interaction of a modulating electric field with light propagating in nonlinear electro-optic materials depends on electro-optic coefficient r_{33} and is described by the following equation including a scalar product of the vector part of quadratic polarizability tensor β and dipole moment μ of a nonlinear chromophore [8]:

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