

Section 5: Functional materials and devices

# Nonlinear dielectric response of poled amorphous polymer dipole glasses

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## Abstract

Temperature-dependent spectra of the linear, second- and third-order nonlinear dielectric permittivities are reported for an amorphous polymethylmethacrylate/Disperse Red 1 guest–host polymer and a poly(styrene maleic anhydride)-Disperse Red 1 side-chain polymer glass. Both polymer systems contain Disperse Red 1 chromophores, a very strong molecular dipole. In guest–host polymers with low dye loading, the dipole density and dipole moment of the chromophores can be determined from the linear and third-order nonlinear dielectric relaxation strength, associated with the micro-Brownian motion of the chromophore dipoles. The second-order nonlinear dielectric permittivity is non-vanishing in the glassy state only in poled polymers. Contributions to the second-order dielectric permittivity arise from piezoelectricity and from the elasto-optical and electronic electro-optical Pockels effect. In poled polymer dipole glasses with nonlinear optically active chromophores, the electronic electro-optical response is the dominant source for the second-order nonlinear dielectric permittivity. Therefore, electrical current versus voltage measurements enable a measurement of the electro-optical Pockels effect in poled polymer chromophore dipole glasses.

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## 1. Introduction

Broadband dielectric relaxation spectroscopy is widely used for the investigation of conduction processes and molecular motions in soft matter physics [1]. An extension to the nonlinear regime provides additional information, for instance on dielectric relaxation processes associated with the micro-Brownian motion of dipoles in the non-crystalline segments of polar polymers [2,3], or on ferroelectric phase transitions in ferroelectric polymers [4–8]. The second-

order nonlinear dielectric permittivity is non-vanishing only in non-centrosymmetric materials and provides information on the degree of poling in polar polymers [5,8].

In this contribution, we report on the nonlinear dielectric response of poled polymeric dipole glasses with strong chromophore molecular dipoles. Such materials are considered for applications in photonic devices, like electro-optical modulators, switches, etc. [9]. It will be shown that the second-order nonlinear dielectric permittivity in such polymers is caused by the piezoelectric and electronic electro-optical effect of the poled polymer glass. Thus electrical measurements enable the measurement of piezoelectric and electronic electro-optical coefficients of polar amorphous polymers at low frequencies.

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