Waveguide-type Optical Devices Using Poled Thin Film Doped with Organic Nonlinear Materials

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Organic second-order nonlinear optical materials have attracted much attention for application to efficient second-harmonic generation (SHG) and high-speed optical modulation and switching, because of its large nonlinearity and high-speed response. In particular, dye-doped poled polymers have the advantages of mechanical strength and ease of film formation necessary for application to waveguide optical device. In this thesis, first a waveguide modulator having a poled polymer film doped with mixed nonlinear organic materials 2-methyl-4-nitroaniline (MNA) and p-nitroaniline (pNA) was presented. Although the mixtures of plural organic materials have stimulated much interest in increasing the nonlinearity, it is difficult to grow large crystals. It is shown that the measured electro-optic coefficient becomes the maximum value $r_{33} = 0.46$ pm/V at a mixing ratio of MNA/pNA=40 which is 1.8 times as large as the MNA only doped film. However, the poled polymer has one problem: the nonlinearity decreases with time because of the orientation relaxation of nonlinear molecules. Hence, sol-gel-processed silica films doped with azo dye 4-[*N*-ethyl-*N*-(2-hydroxyethyl)]amino-4¹ -nitroazobenzene (DR1) in higher concentration were prepared at a high baking temperature with corona-discharge poling of high voltage. A large SHG coefficient of 153 pm/V was obtained, and the value was unvaried for 700 h with no relaxation. Moreover, a waveguide SHG device with DR1-doped sol-gel-processed film was fabricated for the first time and the Cherenkov-type phase-matched SHG was observed successfully.