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Preparation and second-order nonlinearity of organic/inorganic hybrid materials doped with organic chromophore

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Abstract. A kind of hybrid material was made through sol-gel process with the PMMA, TEOS, and 4-[N]-ethyl-N-(2-hydroxyethyl) amino-4'-nitro-azobenzene (DR1). And the films of sol were prepared by using the spin-coated method. Through all optical poling, it could be found that there exists an optimum thickness for film. In addition, the higher of sample temperature was, the smaller intensity of SHG was. The experiment showed that the SiO_2 network was existent, and hybrid materials significantly improved orientation stability.

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Key words: thin film, all-optical poling, SHG, temperature

1 Introduction

Second-order nonlinear optical (NLO) materials have been extensively studied for applications in optical communication and electro-optic modulators due to their large optical nonlinearity, fast response and easy processing. In order to produce second-order effects and ordered arrangement of molecular in films, they need to be polarized by using such as electric-field poling and all-optical poling (AOP) [1]. In recent years, second harmonic generation (SHG) intensity has been optically induced in NLO polymer materials by all optical poling process [2–5],especially, study on thin films of azo compound has been acquired marked achievement [6, 7]. All-optical poling was described for the first time by Charra *et al.* as a novel method in 1993 [8]. This method is simple and possible to work at ambient temperature and an automatic molecular organization with a period satisfying the phase-matching conditions.

On the other hand, PMMA is one of the most commonly non-linear organic substrate materials, easy to be prepared and compatible with the chromophore. Under normal circumstances, it has no interaction with the chromophore molecules. But such material is restricted

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to the application of optical devices for its lower glass transition temperature and fast relaxation. In order to overcome these shortcomings, the organic-inorganic composite materials have been extensively studied [9–11]. Sol-gel technology provides a synthesis of organic-inorganic composite materials approach. The composite material does not require harsh conditions, and it has the advantages of organic and inorganic materials, such as maintaining the film-forming organic polymer, transparency, thermal stability, abrasion resistance.

In this paper, we prepared a new kind of second-order nonlinear optical hybrid material doped disperse red 1 (DR1) from sol-gel method. The second-order nonlinearity was achieved by all-optical poling.

2 Experiments

2.1 Materials and preparation

The molecular structures of 4-[N-(2-hydroxyethyl)-N-ethyl] amino-4'-nitroazobenzene (disperse red 1) and Polymethyl methacrylate (PMMA) were shown in Fig. 1. The organic-inorganic hybrid film was prepared as follows: 0.5 gram of PMMA was dissolved in THF on the mass ratio of 1:10, and then 0.5 ml of TEOS was put into the solution, stirring 1h. The hydrochloric acid (0.15 mol/l) was added to the solution while stirring. The molar ratio of TEOS and H_2O was 1:4. With the addition of hydrochloric acid, hydrolysis reaction took place, with the process of hydrolysis reaction, the solution became turbid, and accompanied by heat release. After a period of timečňwe continue to stir the solution to clarify once again. 0.02 grams DR1 was added to the above solution, this solution was sealed at room temperature for 3 days. The sol was spin coated on glass substrates, and the resultant gel films were dried at 40^oC for 24h. The samples were subject to all-optical poling and second harmonic generation measurements.

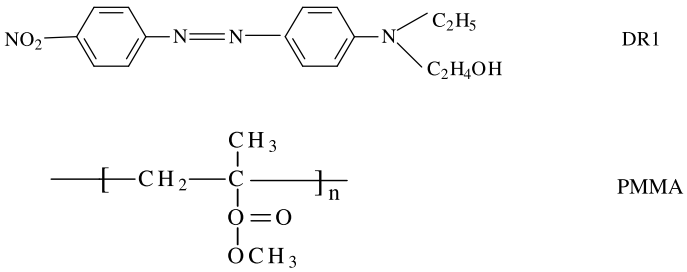

Figure 1: The molecular structure of DR1 and PMMA.

2.2 All-optical poling

The schematic diagram of the experimental setup is shown in Fig. 2. The laser beam was an Nd: YAG laser operating at wavelength of 1064 nm. The pulse-width and the repetition rate