Terahertz Generation from Electric-Optic Side-Chain Polymer Films

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***Abstract*— We investigated terahertz (THz) emission properties of the electric-optic (EO) side-chain polymer films. We evaluated the THz electric field intensity from the EO polymer film by comparing the results with that of a ZnTe crystal. We also characterized the material properties of the EO polymer with respect to the THz generation.**

I. INTRODUCTION

POLYMERIC electric-optic (EO) materials [1-7] have been attracted much attention as the material for ultra-high-speed optical modulators. On the other hand, the EO polymers also have considerable potential for terahertz (THz) sources and detectors [7]. The EO coefficient of the EO polymer (r_{33} > ~100 pm/V) is much larger than that of inorganic and organic nonlinear optical (NLO) materials such as lithium niobate ($LiNbO_3$), zinc telluride (ZnTe), and DAST. This large EO coefficient enables highly efficient THz generation via optical rectification. In addition, because the amorphous EO polymers have no absorption loss due to lattice vibrations that the crystalline NLO materials have, the EO polymers can intrinsically generate broadband THz waves. Furthermore, the EO polymers are suitable for the fabrication of the optical devices using microfabrication processes. These excellent characteristics make the EO polymer the ideal candidate as the material for fabricating highly efficient THz generating and detecting devices.

Until now, we have developed poly(methyl methacrylate) (PMMA)-based EO side-chain polymers with NLO chromophores that exhibit excellent EO property [2-6]. In the present paper, we prepared thin films of the EO side-chain polymer and investigated the THz emission properties of the films. We also characterized the material properties of the EO polymer by using time-domain THz spectroscopy and far-infrared spectroscopy.

II. RESULTS

The EO polymer was coated on ITO coated glass substrates, and the film was poled vertically to the substrate by applying a voltage (100 -120 V/μm). The top electrode on the EO polymer film was removed after the poling process. For the experiment of the THz generation, a regeneratively amplified femtosecond laser (800 nm, ~100 fs, 1 kHz) with a beam diameter of 2 mm was used as the pump and probe light, and the THz electric field was detected by using EO sampling with a 0.5-mm thick <110> ZnTe crystal. The incident angle of the laser beam for the EO polymer samples was 45°, and the beam was irradiated from the back side of the EO polymer/ITO coated glass substrates.

Figure 1(a) shows the representative result of the temporal waveform of a THz pulse generated from the EO polymer film whose EO coefficient (r_{33}) is 83 pm/V at the wavelength of 1308 nm. To evaluate the THz electric field intensity from the EO polymer film, we compared the intensity with that from a ZnTe crystal. Figure 1(b) shows the peak intensities of the THz electric field from a 2.6-μm thick EO polymer and a 0.1-mm thick <110> ZnTe crystal. The THz peak intensities from these two samples were almost equivalent at the pump powers of 4-16 μJ/pulse. However, because the pump wavelength (800 nm) is close to the absorption peak of the EO polymer film, whose optical density is ~6 (2.6 μm), the penetration depth of the pump light is limited (~0.4 μm). Therefore, the effective thickness of the EO polymer is less than the film thickness (2.6 μm).

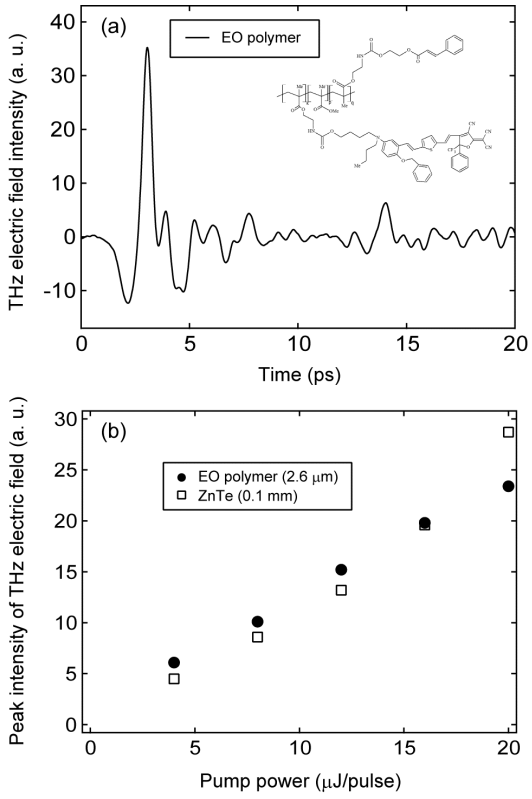

Fig. 1. (a) Representative result of the temporal waveform of a terahertz (THz) pulse generated from an EO polymer film. (b) Comparison of the peak intensities of the THz electric field emitted from a 2.6-μm thick EO polymer film and a 0.1-mm thick <110> ZnTe crystal.

To characterize the material properties of the EO polymer, we measured the spectra of the absorption coefficient and refractive index. Figure 2(a) shows the absorption coefficient of the EO polymer from THz to far-infrared region. The absorption peak around 11 THz is attributed to the intramolecular vibrations of the PMMA backbone. On the other hand, the absorptions around 5 THz and over 15 THz are

attributed to the intra- or intermolecular vibrations of the NLO chromophores. Because the inorganic NLO materials such as $LiNbO_3$ and ZnTe show strong absorption in the high frequency region (>2-4.5 THz) due to the lattice vibrations, the result suggests that the EO polymer can generate broadband THz waves with small absorption loss. Figure 2(b) shows the refractive index of the EO polymer in THz region. The refractive index of the EO polymer is around 1.7 in the THz region, and the value is almost the same as that (~1.7) in the infrared light wavelength (1550 nm), which can be used as the alternative pump wavelength for the THz generation. The very small difference in the refractive indexes between the infrared light and the THz wave suggests that the EO polymer can achieve efficient phase matching for the THz generation.

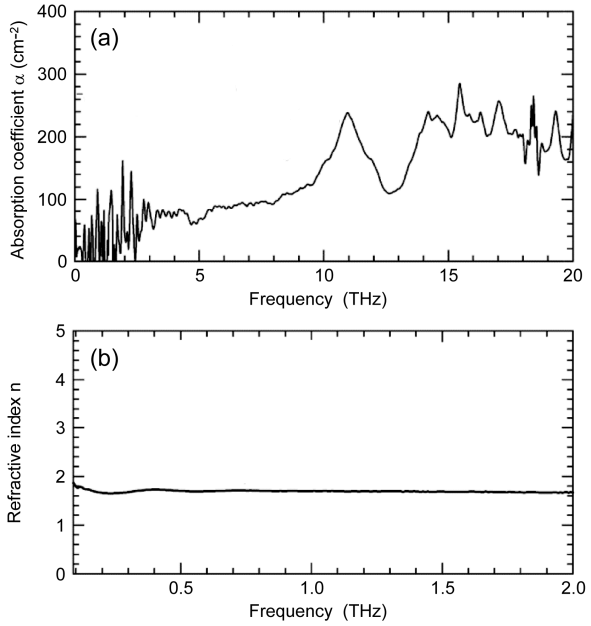

Fig. 2. Absorption coefficient (a) and refractive index (b) of an EO polymer film in far-infrared and THz region. The spectra were respectively obtained by far-infrared spectroscopy and time-domain THz spectroscopy.

III. SUMMARY

We investigated the THz emission properties of the thin films of the EO side-chain polymer. The THz electric field intensity from the 2.6-μm thick EO polymer was almost equivalent to that from the 0.1-mm thick <110> ZnTe crystal with the pump wavelength of 800 nm. We also characterized the material properties of the EO polymer and revealed that the EO polymer had small absorption loss in the THz region and very small difference in the refractive indexes between the pump infrared light and the THz wave. The results indicate that the EO polymer is a promising material for the highly efficient and broadband THz generating and detecting devices.

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