Hybrid Terahertz-Wave Source with Ultrawideband Tunability utilizing Organic DAST and BNA Crystals

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Abstract: Crystal quality of a new nonlinear organic material BNA was much enhanced successfully. A novel hybrid THz-wave source with extremely ultrawideband frequency tunability was proposed using new BNA and established DAST crystals.

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1. Introduction

The developments of coherent monochromatic terahertz (THz)-wave sources with agile and ultrawideband frequency tunabilitiy are indispensable to boost THz-wave researches and applications [1-2]. Difference frequency generation (DFG) scheme satisfying collinear phase matching is unique and sophisticated method to generate monochromatic THz-wave with high efficiency and ultrawideband tunability. In particular, nonlinear optical crystals made from organic materials are more suitable to meet the collinear phase matching condition because refractive indexes of them at THz-wave and optical regions are widely comparable. Besides, they possess larger second-order optical nonlinearity compared with that of conventional inorganic crystals.

DFG-THz source with ultrawideband frequency tunability by using an organic 4-Dimethylamino-N-Methyl-4-Stilbazolium Tosylate (DAST) crystal was already established [3-4]. Tuneing frequency range of current DAST-DFG is roughly from 1.5 to 30 THz and it cannot cover below 1.5 THz due to large absorptions by phonon vibrations and phase mismatchings. Another DFG-THz source by using a new organic crystal N-Benzyl-2-methyl-4-nitroaniline (BNA) have been developed [5-6] to supplement such dips in THz-wave spectrum of the DAST-DFG.

In this work, drastic improvement of crystal quality of BNA is described firstly. Then, a concept of compact hybrid DFG-THz source using DAST and BNA crystals is proposed. The hybrid DFG-THz source can cover extremely wideband frequency range from sub-THz to 30 THz and can be pumped by single dual BiB_3O_6 optical parametric oscillator (BiBO-OPO).

2. High quality BNA crystal

The crucial issues to be resolved in using organic materials for nonlinear optical devices are scarce crystal quality and size. In general, growing large and high quality organic crystal is more difficult because the intermolecular force of organic materials is relative weak and three dimensional structure of them are quite complex. A method to grow large and high quality DAST crystal has already been well developed since the DAST crystal has long history. On the other hand, vertical Bridgman method was known as only technique for BNA growth. However, crystal qualities of BNA grown by Bridgman method are not so good because it imposes strong stress on the crystal during the growing process. We have succeeded in growing high quality BNA crystal by applying ethanol solution growth method instead of Bridgman method. By optimizing several key factors such as concentration of BNA saturated solution, cooling rate and temperature, high quality single BNA crystals with area of roughly 100 mm^2 and thickness of 3 mm have been obtained successfully at the present.

In order to evaluate crystal qualities of BNAs, X-ray diffraction analysis was conducted. Fig.1 shows the so-called X-ray rocking curve from the (020) lattice plane in BNA single crystals. The vertical axis represents normalized diffracted X-ray count number and the horizontal axis indicates slight displacement of incident X-ray angle from the legitimate incident angle which just fills Bragg's diffraction condition. Linewidth of the rocking curve reflects on the orientation regularity of lattice planes and better orientation regularity without distortion and/or lattice defects results in narrower linewidth. The linewidth of BNA crystal grown by our solution method was drastically reduced to about one-fifth compared with that grown by past Bridgman method. This tiny linewidth is comparable with that of silicon wafer which is well known as the inorganic single crystal with the best quality in the market. Besides, enhancement of damage threshold level due to pump laser was also confirmed. These results signify that quality of BNA crystal was much improved successfully by applying solution growth method.

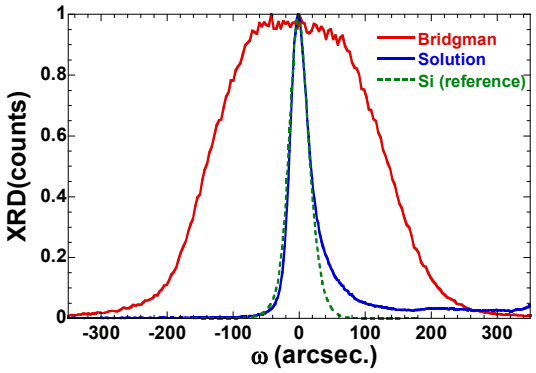

Fig.1 Rocking curve of BNA crystals

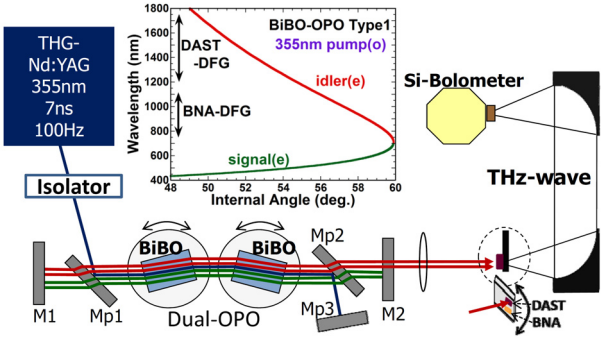

Fig.2 Hybrid DFG-THz system pumped by dual BiBO-OPO

3. Hybrid terahertz-wave source with ultrawideband tunability

Although each DAST- and BNA-DFG system possesses ultrawideband frequency tunability, they have several intensity dips in the spectrum depending on their phonon absorptions and phase mismatchings. In order to compensate these spectrum dips, a hybrid DFG-THz source combined DAST and BNA have been developed. Since optimum pump wavelengths for DAST- and BNA-DFG are different [3-6], a new dual OPO which can generate near-infrared lights with wideband tunability have been developed by employing BiBO crystals. Fig. 2 shows whole view of the sophisticated hybrid DFG system pumped by only one dual BiBO-OPO. The original pump source is frequency tripled Nd:YAG laser with wavelength of 355 nm, pulse duration of 7 ns and repetition rate of 100 Hz. Two BiBO crystals are pumped by the 355 nm beam under the condition of type 1 phase matching for OPO. Here, to free the OPO cavity mirrors from the requirements of complicate reflection and transmission coatings for intense 355 nm pump beam, two steering mirrors (Mp1 and Mp2) set at Brewster's angle with respect to the cavity axis are inserted into the cavity [7]. Calculated wavelengths for signal and idler waves are also shown in Fig. 2 as a function of internal propagation angles. Since BiBO crystals are mounted on galvano scanners, oscillation wavelengths can be widely and agilely controlled by angle tuning with time of 1ms. Two signal waves are confined between two specially coated cavity mirrors (M1 and M2) and two idler waves are extracted from the M2 as pump lights for both of DAST- and BNA-DFG. After focusing two pump idler lights, they are injected into DAST or BNA crystal. Since the DAST and BNA crystals are also mounted on a galvano scanner, we can switch the crystals in synchronization with two pump wavelengths of dual BiBO-OPO depending on the desired THz frequency so as to compensate each weak radiation region. Since BNA can cover below 1 THz region [5], monochromatic coherent THz-wave source with extremely ultrawideband tunability beyond double-digit from sub-THz to 30 THz will be realized.

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