Progress on All Solid State Deep-ultraviolet Laser with $KBe_2BO_3F_2$ Crystal

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General review for the progress on all solid state deep ultraviolet (DUV) laser with a $KBe_2BO_3F_2$ (KBBF) crystal was given in the paper. The frequency-conversion characteristics of KBBF crystal and the comparison between this crystal and other borate nonlinear optical crystals were simply described. A special prism coupling technique (PCT) for DUV laser generation and some excellent experimental results from the application of this crystal were commented. At last, we believe that the output power and conversion efficiency could be greatly increased in the future if the thicker KBBF crystal with good optical quality is grown and the application technique of solid state DUV laser is further developed.

Key Words: DUV, KBBF, Solid state laser, Nonlinear optics, SHG

1. Introduction

Deep ultraviolet (DUV) sources can be applied to many areas, such as semiconductor photolithography, micro-machining, laser spectroscopy, photoemission spectroscopy and photochemical synthesis.[1)] The DUV sources can be synchrotron light sources, gas-discharge sources or laser sources. Synchrotrons offer unparalleled flexibility in wavelength tenability (1-1000eV), but they cause low energy resolution (10 meV) and small photon flux (10^{12}-10^{13} photons/Second). Although gas-discharge sources offers better energy resolution (1.2 meV), it has drawbacks like large beam size (2~3 mm) and small photon flux (10^{12} photons/second). The DUV lasers, including the excimer laser, solid state laser and so on, are highly desirable for many applications. The excimer lasers can emit coherent light with a high average power output, however, their wavelength tunability and beam quality were poor. So, the compact, efficient solid-state lasers are more attractive, because of their narrower spectral bandwidth, better beam quality and easier maintenance etc. In the past, the solid state DUV lasers are generated by use of sum-frequency method, however, the need for two beams which are one short wavelength and the other long wavelength can be an inconvenience. So far, only $KBe_2BO_3F_2$ (KBBF) crystals can be used to achieve the DUV laser light through second harmonic generation (SHG). Similarly, the shorter wavelength can be also obtained by the sum-frequency generation (SFG) method in KBBF crystal. In the report, we made a general review for the progress on high power and high efficient DUV laser with a KBBF crystal.

2. Characteristics of KBBF Crystal

KBBF crystal [2)] was first synthesized by the former Soviet Union scientists in 1968. The crystal structure was re-determined by C. Chen, and the results showed that KBBF crystallizes in the space group R32 (not C2). KBBF is an excellent nonlinear optical (NLO) crystal with relatively larger SHG coefficients, wide transparent region from 152 to 3664 nm, moderate birefringence, out of hygroscopy, good optical uniformity with $\delta n \approx 10^{-4}$ /cm, high damage threshold as high as 4×10^{12}W/cm^2 at 532 nm with 7-ns pulse width and 10 Hz, high thermal conductivity of ～2.5W/mK, a wide band gap of ～8.2 eV. The melting point of KBBF is estimated to be above 1100C and it volatilizes severely and decomposes above 800C. Therefore, KBBF crystals is mainly grown from a flux melt, not hydrothermal method above 800 ^{0}C. Crystals showed a plate-like growth habit due to layered structure of KBBF. At present, KBBF can be grown with its thickness restricted to 3 mm only, the length along the z axis has not exceeded 2.5mm till now. Although the hydrothermal method below 750 ^{0}C condition can be used to grow the large size KBBF of ~10mm, the Phi degree was confusable at present. The plate-like crystals were still too thin to be cut along the phase-matching direction. A special prism coupling technique (PCT) [3)] was invented to solve the question in cutting the crystal. The invention was a sandwich structure and KBBF crystal is placed in between two prisms (UV fused silica or CaF_2 crystal). The two interfaces between the KBBF and CaF_2 (or fused silica) were brought into optical contact for reducing the interface losses of the laser beam, or the interfaces were filled with matching oil or deionized water. When the fundamental wave is input along the normal direction of the prism, the angle of refraction in KBBF is equal to the apex angle of the prism. The phase matching of the wavelength can be automatically achieved in KBBF when the special fundamental wave is input along the normal direction and the polarization direction of the fundamental wave is along the *a*

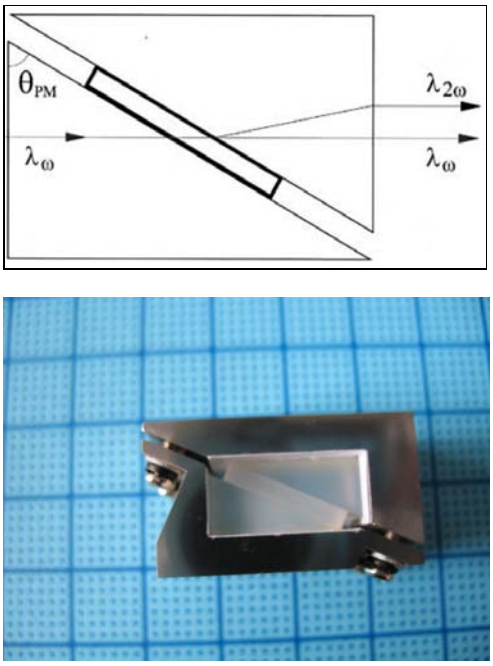

Fig.1 Scheme of the KBBF-PCT and the optical contact prism-coupled setup.

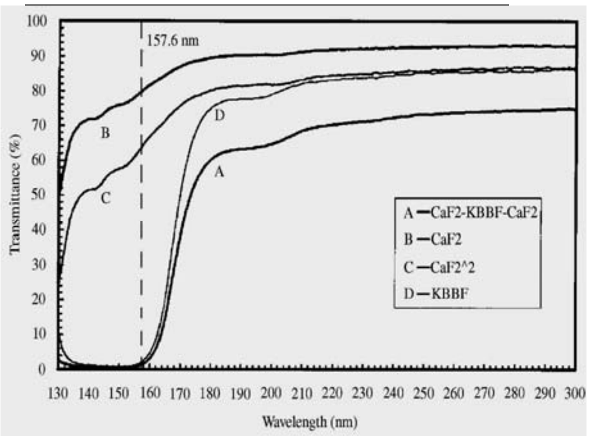

Fig.2 Curves A, B, C, and D represent the transmission spectra of KBBF crystal plus two CaF_2 prisms, a single CaF_2 prism, two CaF_2 prisms, and a single KBBF crystal, respectively

axis of KBBF, if the apex angle of the prism is equal to the phase-matching angle of KBBF at a special wavelength. It can be seen that this crystal can produce very short wavelength from second to harmonic output from the above narration. The KBBF-PCT setup and the transmittance of KBBF device were shown in Fig. 1 and 2, respectively.

3. All Solid State DUV Laser Generation

3.1 Phase Matching of KBBF and Other Borate Crystals [3)]

So far, only KBBF crystal can be use to obtain the DUV laser below 200nm by the SHG generation from Fig 3. In

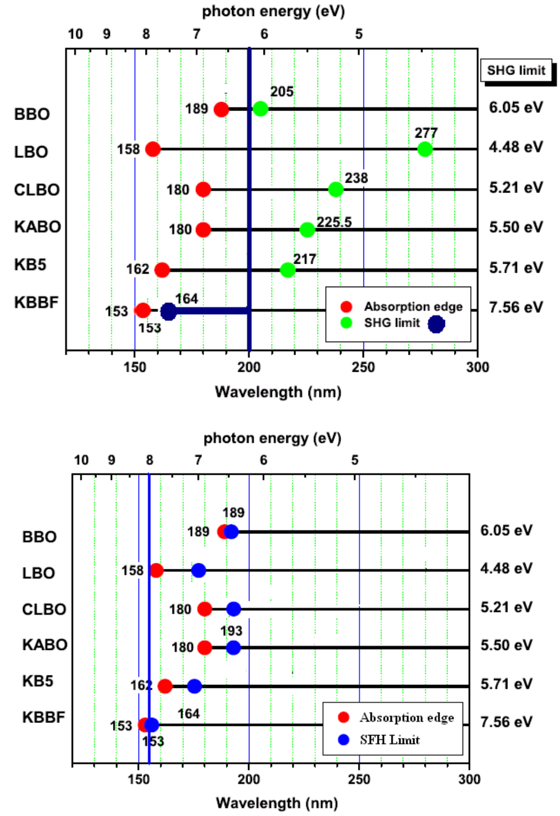

Fig.3 SHG and SFG limit for some typical borate crystals

addition, although some crystals such as LBO, BBO, CLBO, CBO, SBBO, TBO, KABO, BABO, KB5 etc, also can generate the DUV through SFG technique, at present, the obtained wavelength is shortest in DUV region by KBBF SFG technique.

The birefringence of LBO is only about 0.045, which is too small to achieve deep-UV harmonic generation. The same situation also occurs in the other borate crystals of the LBO family such as CBO and CLBO. It is known that even though CBO, CLBO KB5 can achieve DUV output, two different wavelength beams must be included as input, which is obviously not convenient for practical applications. From Fig. 3, we immediately knew that the capability of BBO to produce the DUV coherent light below 200 nm was limited by the absorption edge (189 nm). The SBBO structure has macroscopic order but microscopic disorder, which could be a reason why the optical uniformity of SBBO is very poor. As a result, the phase-matching angles of the crystal at the different wavelengths cannot be determined accurately till now, which heavily limits its applications in various frequency-conversion devices. TBO crystal seems to have the same structure problem as SBBO. Conversely, the space structure of KABO can be determined exactly, so high optical quality crystals of KABO in large size can be grown. KABO can also achieve fourth and fifth-harmonic generation of a Nd:YAG laser and 193-nm wavelength output with sum-frequency generation. In addition, this crystal is not hygroscopic and has good mechanical properties for cutting, polishing, and coating. Therefore, KABO is a good potential UV and deep-UV NLO crystal, and further research is deserved. The BABO crystal should have the same optical properties as KABO, but a crystal large enough have not been yet obtained to measure its linear and nonlinear optical properties.

3.2 DUV Laser Generation in Typical Borate Crystals[3)]

LBO was used to generate the DUV in the range of 172.7-187 nm by phase-matched sum-frequency mixing of the Ti:sapphire's (1kHz 0.3mJ, 740-850 nm**,** 150-200fs) fourth harmonic (ω+3ω) and a parametrically generated infrared pulse which was generate by an optical parametric generator/amplifier scheme with the same Ti:sapphire laser as the pump source.[4)] A similar experimental setup as LBO was used to generate the DUV from 172 down to 166nm in a KB5 crystal.[5)] In addition, the KB5 was the first borate crystal to achieve 194-nm coherent light output with the sum-frequency mixing method (792nm+515nm→194nm). 177.4nm DUV laser in KB5 was provided by the harmonic generation of a Nd:YAG laser (ω+5ω). BBO crystal was the first borate crystal which obtained the 193nm effective output by the SFG of a linearly polarized picosecond KrF excimer laser at 248.5nm and a a tunable dye laser at 950–800 nm. Recently, The fourth-harmonic generation (ω+3ω) of a Ti:sapphire laser in BBO was used to obtained 22-mW power output at 193.5 nm. Using the same method in CBO, 193.3nm was obtained the effective output. Also, CBO was used to generate 193-nm coherent light output through a sum-frequency mixing process (2.0*um*+213nm to193 nm), meanwhile, over 5mJ/pulse energy has been obtained. CLBO crystal was used to generate 193nm output though an eighth-harmonic generation (ω+7ω process) of an Er-doped fiber amplifier system. Thereinto, the 2ω, 3ω, and 4ω harmonics were produced in LBO crystal, BBO was used to SFG (3ω+ 4ω) for 7ω generation. KABO crystal can generate 193 nm with a SFG procedure (1064 nm+236 nm to 193.2 nm).

3.3 DUV Generation in KBBF Crystal

In order to produce wavelengths below 200 nm, only KBBF so far has enough birefringence and sufficient transparency range to achieve directly fourth or fifth-harmonic generation of a Ti: sapphire laser. It also can be used to generate the fourth harmonic of the entire tunable spectral region of the Ti: sapphire laser. The crystal can further achieve sixth-harmonic generation of a Nd:YVO4 (or Nd:YAG) laser (3ω+3ω procedure) to produce 177.3-nm coherent light output. The shortest SHG wavelength of KBBF is 162 nm and the shortest theoretical SHG wavelength obtained experimentally so far is 170 nm.[6)] In addition, at present, the shortest wavelength has been obtained by SFG in KBBF crystal.

In 2007, our lab [7)] employ a frequency-tripled Nd:YVO4 laser with a passive-mode-locking technique based on a saturable Bragg reflector as the pumping source of a 2.1mm thick KBBF-PCT with a phase matching angle of 68.6 degree, the high power sixth harmonic generation (3ω+3ω)of an Nd:YVO4 laser is obtained at 177.3nm. For the input power of 3.5 W, the maximum output power is 12.9mW in a hermetic chamber filled with N_2, and this is the highest output power ever obtained in deep ultraviolet region by means of the direct harmonic generation. Moreover, the output power is not saturated yet and higher power should be obtained if more power density at 355 nm is applied. The experimental setup and the power curve are shown in Fig. 4. Also, in our lab,[7)] the fourth harmonic generation of a Ti: sapphire laser system (150fs) at wavelength 200 nm with a high conversion

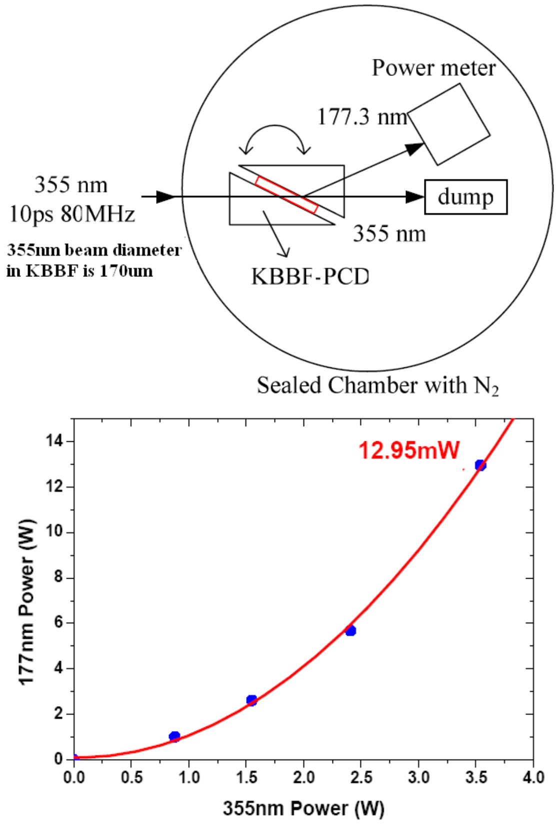

Fig.4 The experiment setup for 12.9mW at 177.3nm and the output power and in power curse.

efficiency of 26.1% has been also obtained using the KBBF-PCT with a KBBF size of 10.5×6×2.3mm[3] and a phase matching angle of 55 degree. The maximum output at 200nm is 10.7mW when the input power at 400nm is 40.9mW. This is the highest conversion efficiency ever obtained in deep ultraviolet region with KBBF-CaF_2 prism-coupled device. Meantime, the conversion efficiency is not saturated yet and higher conversion efficiency could be obtained if higher power intensity at 400 nm is applied. During the experiment, no damage is found in either KBBF or CaF_2 prisms. The conversion efficiency curve is shown in Fig. 5.

Our lab[7)] has developed a ns widely tunable t DUV laser in the wavelength range from 175 to 210nm by the fourth harmonic generation of Ti:Sapphire laser with KBBF crystal.

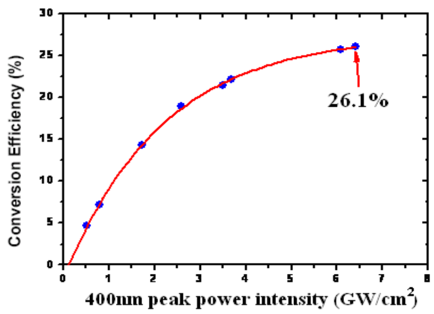

Fig.5 The conversion efficiency curve.

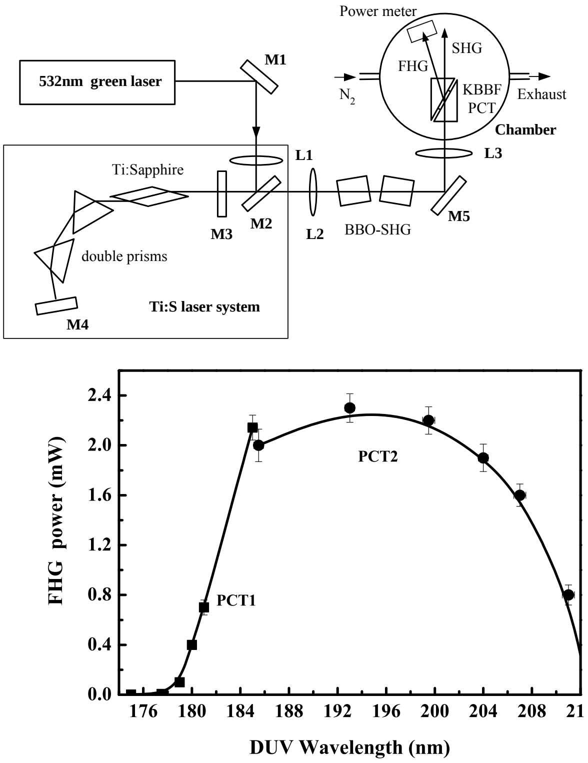

Fig.6 The experiment setup for tunable DUV laser and the tunable wavelength curve.

The highest output power is 2.23mW at 193nm and the power of the DUV laser is more than 2mW from 185nm to 200nm. It is the first demonstration of mW-level ns continuously tunable DUV all-solid-state laser in such a wide wavelength range. The experimental setup and tunable curve are shown in Fig. 6. The output power of Ti:Sapphire laser is more than 3W during the whole tuning range from 690 to 840nm. At the maximum output power at 780nm, the beam quality factor is M^2~3. The bandwidth (FWHM) is less than 2nm and the pulse width (FWHM) is 24ns. Tunable UV output power higher than 1.5W in most tuning range is achieved by this walk-off compensated double BBO configuration. Phase-matching angle of KBBF in type-I SHG changes from 68.3^o to 48.9^o with fundamental wavelength from 345 to 420nm. Two KBBF PCT devices with thicknesses of 0.65mm and 0.69mm along Z axis are used in the wide tunable DUV light generation for the large difference of phase matching angles. One PCT (PCT1) device with KBBF cut at θ=66.4°is best suited for frequency doubling of 354.7nm beam to 177.3nm. The other KBBF PCT device (PCT2) with KBBF cut at θ=56.4°is optimized for producing 193nm light.

Cooperation with Tokyo university[8)], an important progress, over 350mW DUV laser at 193nm and a stable power of 150mW are obtained by KBBF crystal, and this is a highest result at 193nm by use of the borate crystals. The KBBF–CaF_2 PCT device was also used to generate the fifth harmonic (from 157 to 160 nm) of a Ti:sapphire laser system at 1-kHz repetition rate and 16-ns pulse width at 800–785 nm wavelength range. This process was achieved by

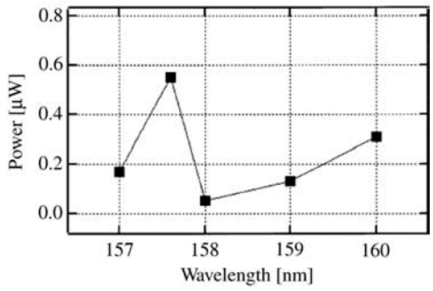

Fig.7 The tunable curve.

sum-frequency mixing of the fourth harmonic with the fundamental (4ω+ω→5ω). The 5*ω* spectral width is estimated to be 0.01 pm and the pulse width was 9.7 ns at 157.6 nm. The output power decreased around 158–159 nm, as shown in Fig. 7, but the reason for this is not clear.

4. Conclusions and Acknowledgements

The DUV laser generation in our lab and Tokyo university has obtained great progress in KBBF crystal. The merits of KBBF have been demonstrated through these good results. At last, we believe that the output power and conversion efficiency can be greatly increased in the future if the thicker KBBF crystal with good optical quality is grown and the application technique of KBBF crystal is further developed. In addition, BPO crystal[9)] whose characteristics are currently still under investigation has a large transparent range from 130 to 4300nm and its birefringence index is 0.0045. It may be a promising NLO material candidate as DUV laser generation.

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