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OPTICAL PARAMETRIC OSCILLATORS OF BARIUMBORATE AND LITHIUMBORATE: NEW SOURCES FOR POWERFUL TUNABLE LASER RADIATION IN THE ULTRAVIOLET, VISIBLE AND NEAR INFRARED

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In summary these first experiments clearly demonstrate the advantages of the BBO and LBO-OPO's as tunable coherent light sources. The most attractive features are the high power capability, high conversion efficiency, and in particular the large tuning range. The results obtained so far are certainly very promising for the further development of these new tunable all solid-state sources, which might be in the near future the radiation source of choice for applications like pulsed laser spectroscopy.

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Optical Parametric Oscillators of Bariumborate and Lithiumborate: New Sources for Powerful Tunable Laser Radiation in the Ultraviolet, Visible and Near Infrared

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Since the first demonstration of an optical parametric oscillator (OPO) by Giordmaine and Miller[1] in 1965 the OPO has been subject to detailed theoretical and experimental investigations[2,3]. The OPO is considered as a source of powerful, broadly tunable coherent radiation. The development and the scientific application has been hampered, however, by the scarcity of nonlinear optical materials with suitable optical and mechanical properties.

Recently new nonlinear materials - Bariumborate (BBO) and Lithiumborate (LBO) - became available. With their unique properties (i.e. high nonlinearity, wide transparency range and high damage threshold) BBO and LBO should be very useful materials for an OPO. These OPO's combine the advantages of an all solid-state tunable source with a wide tuning range in the ultraviolet, visible and near infrared, high peak and average power and high conversion efficiency.

Pumped by the harmonics of the Nd-YAG laser, the BBO-OPO as well as the LBO-OPO generate tunable laser radiation in the spectral range between 300 and 3000 nm. Fig. 1 shows the calculated tuning regions of the signal and idler waves as function of the phase-matching angle. The OPO's are pumped by 532 nm, 355 nm and 266 nm Nd-YAG laser radiation.

In a first experimental demonstration a BBO OPO was pumped by the 532 nm second harmonic of a Nd:YAG laser[4]. The wavelength tuning (940-1220 nm) was limited by the used mirrors. The tuning range could be extended substantially into the UV and the near infrared (330 nm - 2550 nm) by pumping with the 355 nm third or the 266 nm fourth harmonic of a Nd:YAG laser or a 308 nm XeCl excimer laser. This was demonstrated in several experiments first reported in 1988 at the CLEO conference[5][6][7]. Details of these investigations have meanwhile been published[5][6][7].

In the experiments performed at the Stanford University[5] the BBO-OPO was pumped by single axial mode 355 nm third harmonic Nd-YAG laser radiation. The Nd-YAG pump laser with unstable resonator (Spectra Physics, Model DCR3) was injection seeded to obtain single-mode operation (Spectra Physics Model 6300 Injection Seeder). The doughnut shaped output beam was spatially filtered in the far field by a suitable pinhole and frequency tripled with KD*P. The 355 nm radiation with almost Gaussian intensity distribution (diameter d=2.8 nm) provided 6 ns long light pulses with an energy of 30 mJ/pulse and a repetition rate of 30 Hz.

The 3.2 cm long resonator consisted of two flat mirrors with a reflectivity of 98 percent (input mirror) and about 80 percent (output mirror). The transmission at 355 nm and at the idler wavelength exceeded 80 percent. The BBO

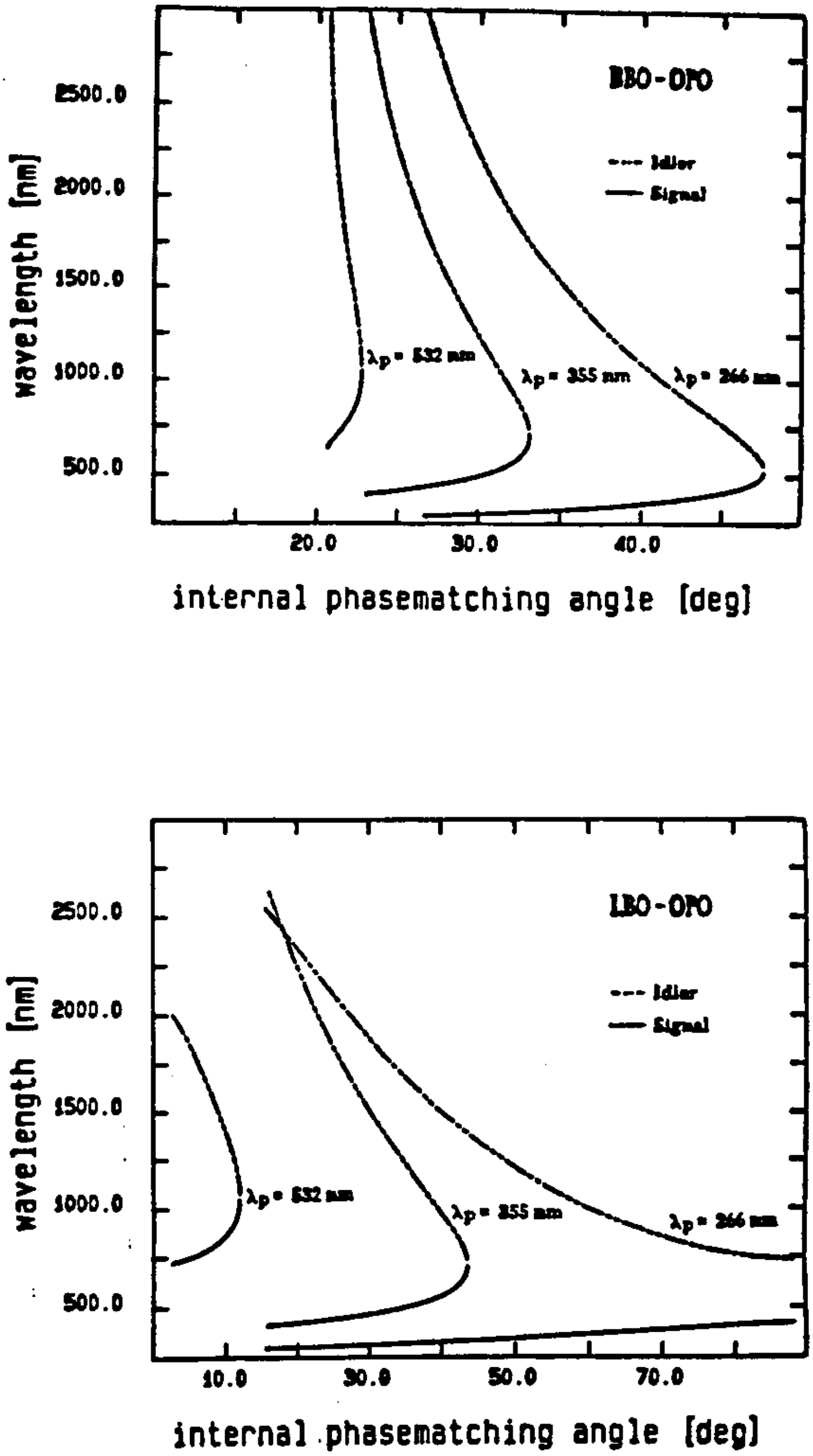

Fig.1: Wavelength of the signal and idler wave of the BBO-OPO and LBO-OPO as function of the phase-matching angle. The OPO's are pumped by the second, third or fourth harmonic of a Nd-YAG laser.

crystals (size 6 x 6 x 12 mm^3) were cut at an angle of 25 and 35 degrees. Fig. 2 displays the measured wavelengths of the signal and idler radiation as function of the type I phasematching angle. The estimated experimental accuracy of each angle and wavelength measurement was about ±1 degree and ±1 nm, respectively. Within these uncertainties the measured values were in good agreement with those calculated from the Sellmeier formula reported by Kato [8]. The observed tuning range extended from 412 nm to 710 nm (signal wave) and 710 nm to 2.6 µm (idler wave). This operating range corresponds almost to the maximum possible tuning range which is limited by the increasing absorption of BBO at wavelengths larger than 2.5 µm [9]. As shown in Fig. 2 the measured phasematching angle varies between 24.5 and 33.2 degrees.

Besides the wavelength tuning, parameters like threshold power, conversion efficiency and the spectral width of the generated light are of special importance.

Fig.3 displays the energy density at threshold (J_o) measured as function of wavelength with an experimental set up similiar to the one described in ref. 5. The value of J_o depends - as expected - on the mirror reflectivity . The minimum values of J_o increase with decreasing wavelength from J_o=0.12 J/cm² to J_o=0.19 J/cm². The corresponding pulse energy is 5-7 mJ. The power density of 20 to 40 MW/cm² is well below the BBO damage threshold which is expected to be several GW/cm². Calculated values of J_o - also shown in Fig.3 - are larger by a factor of about 2. This difference between the theoretical and experimental results might indicate that the previously measured nonlinear coefficent of BBO is low[10].

Fig.4 displays the measured energy conversion efficiencies as function of the ratio J_p/J_o, where J_p is the energy density of the pump radiation. With a 12 mm long crystal the conversion efficiency is close to 25 percent. For an 8 mm long crystal J_o is larger by a factor of 2.2. At a ratio of J_p/J_o=1.8 - which is limited by the available energy E_p=24mJ of the laser pulse - the conversion efficiency is less than 8 percent. The efficiency should increase with crystal length and larger values of J_p/J_o. However, walk-off between the pump and the generated radiation limits the useful BBO crystal length to about 25 mm.

For many applications narrowband operation is highly desirable. Pumping with a single-axial mode laser source[5] the output contained typically 6 axial modes of the 3.2 cm long OPO resonator indicating a linewidth of about 23 GHz. Single mode operation was achieved at 532 nm or 1,06 µm (and at the corresponding idler and signal wavelength) by injection seeding with light of the second harmonic or the fundamental of the single-mode Nd-YAG pump laser. The injection seeding not only provided single-mode operation but also reduced the oscillator build-up time from 4 to 2.2 nsec. Because of the reduced build-up time the pulse length increased from 2.5 nsec to 4 nsec. Simultaneously the OPO threshold power decreased by a factor of 3.

In addition to the systems mentioned so far, synchronously pumped BBO-OPO devices have been investigated. With the second harmonic of a mode-locked pulsed Nd-YAG laser [11] or

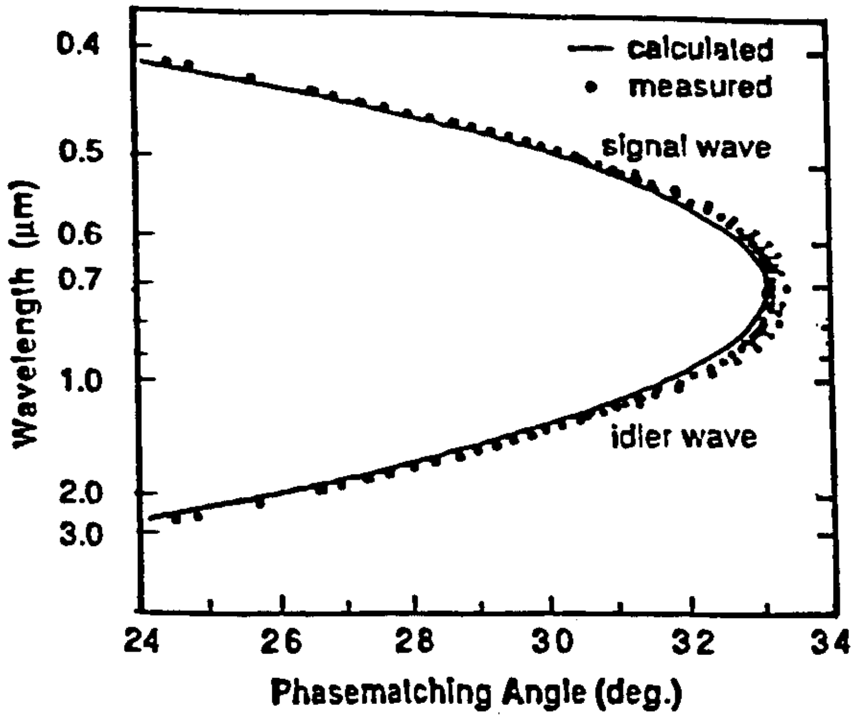

Fig.2: Measured and calculated wavelength of the signal and idler wave as function of phasematching angle of the BBO optical parametric oscillator (Ref.5).

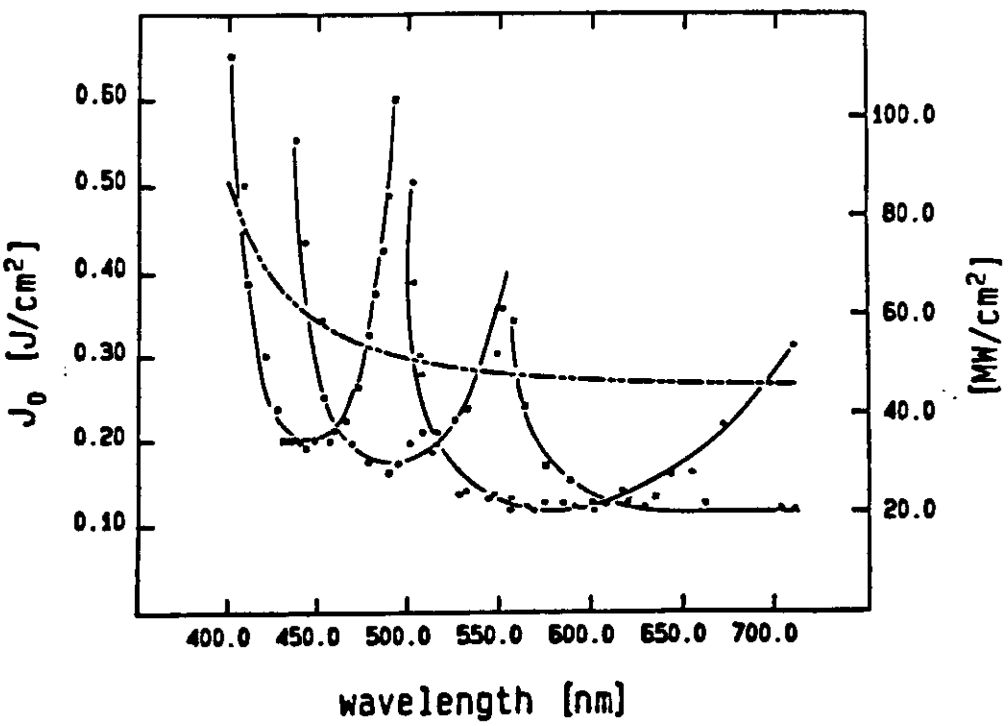

Fig.3: Energy density J_o at threshold measured for four sets of mirrors. The beam diameter is 2.5 mm. The theoretical values of J_o are calculated for a mirror reflectivity of 70 percent.

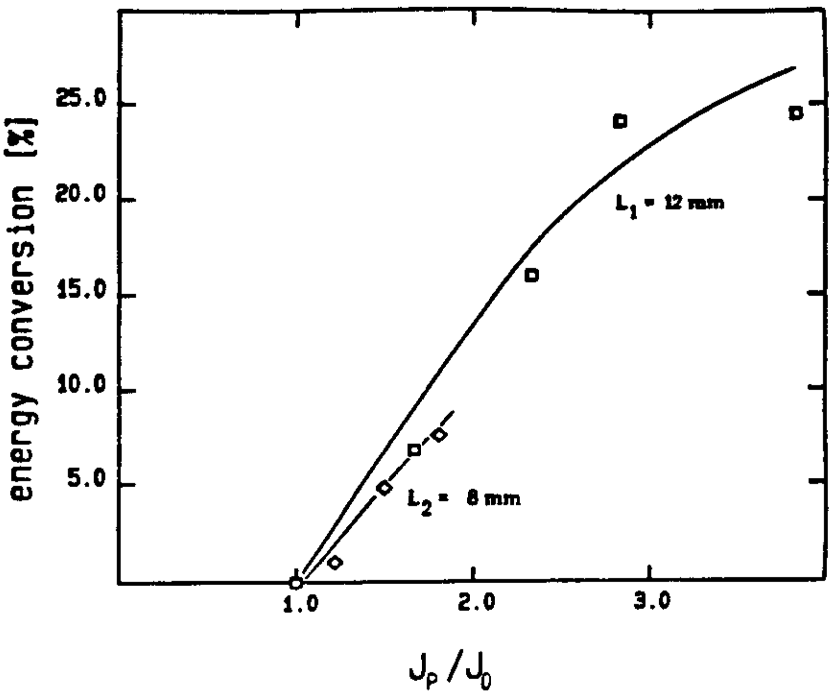

Fig.4: Total energy conversion measured as function of the ratio J_p/J_o. J_p is the energy density of the pump radiation. The length of the crystals are L_1= 12 mm and L_2=8 mm. The corresponding values of J_o are J_{o1} = 0.12 J/cm^2 and J_{o2} = 0.26 J/cm^2. The wavelength of the signal wave is 620 nm.

the third-harmonic of a Nd-YAP system[12] the BBO-OPO produced 75 psec long light pulses tunable in the regions of 680 nm to 2.4 µm [11] or pulses of about 20 psec duration at 406 nm to 3.17 µm [12], respectively.

In addition to BBO LBO crystals are now available in sufficiently large size and with high optical quality. As seen in Fig. 1 the LBO-OPO should be tunable in the same wavelength range as the BBO-OPO. In LBO the effective nonlinearity is smaller, however. At 710 nm, for example, d_{eff}(LBO)=0.39 d_{eff}(BBO). The values of J_o of the LBO-BBO should thus be larger by a factor of 6.7. The values of the ratio of J_o(BBO)/J_o(LBO) obtained in first measurements with 12 mm long crystals at the wavelengths of 630 nm, 560 nm and 440 nm provide ratios of 4.7, 5.2, 3.2, respectively [13]. These measurements confirm the higher energy density at threshold of the LBO-OPO. The results are in agreement with the theoretical predictions.

In LBO the tuning rate is considerably smaller compared to BBO. Wavelength tuning in the range of 410-2600 nm requires, for example, a change of the type I phase-matching angle $\Delta\Phi$ =28°. In the UV tuning from 300 nm to 400 nm even requires a change by $\Delta\Phi$ =55°.

The possible advantages of the LBO-OPO have still to be investigated in particular in respect to narrow bandwidth operation and short pulse generation.