Optical parametric oscillation in random polycrystalline $\chi^{(2)}$ medium

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Abstract: We demonstrate the first OPO based on random phase matching. The OPO was based on ZnSe ceramic pumped by 62-fs, λ=2.35-μm Cr:ZnS laser pulses, had 90-mW pump threshold and produced an ultra-broad spectrum spanning 3-7.5 μm.

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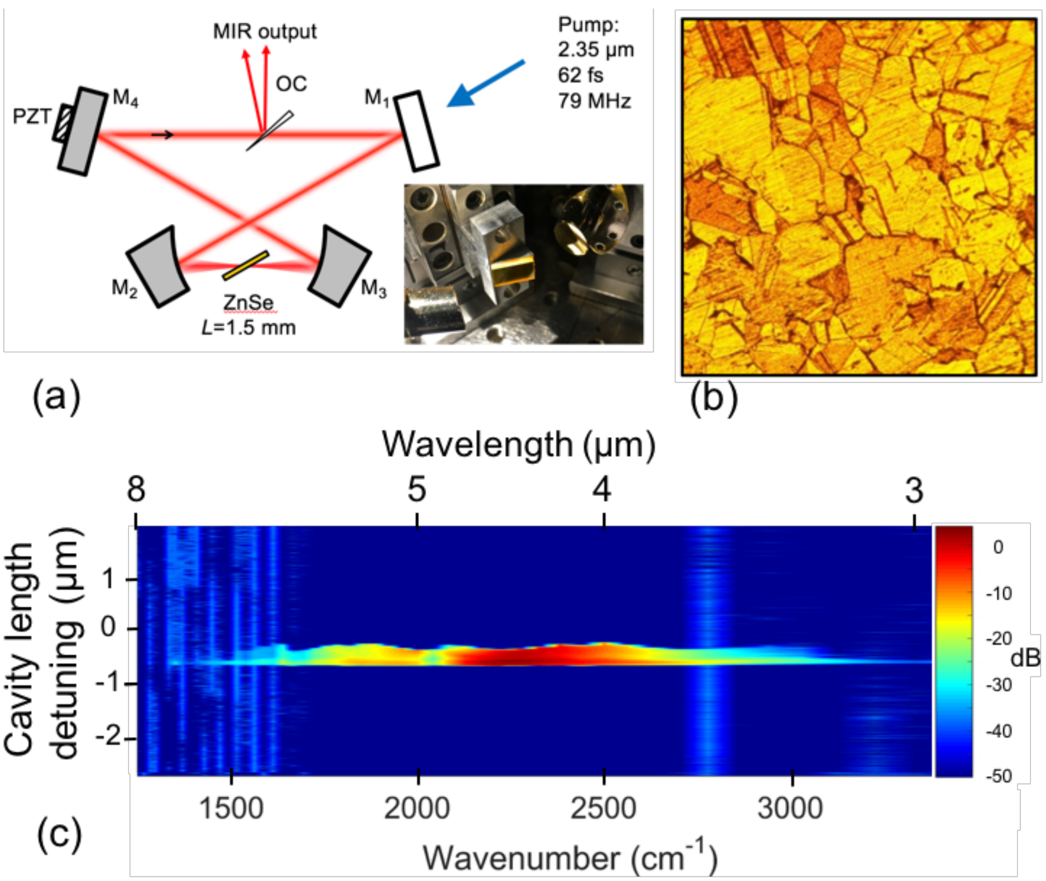
It has commonly been assumed that perfectly regular crystals are paramount for the operation of frequency converters based on quadratic nonlinearity, such as optical parametric oscillators (OPOs). The necessary phase-matching condition is typically achieved either by angular phase matching, or quasi-phase matching through flipping crystalline domains. Here we show that a disordered material consisting of randomly-oriented domains, with the nonlinear wave-coupling coefficient arbitrarily varying between its maximum (d_{NL}) and minimum ($-d_{NL}$) values, can perform as an OPO gain material.



(a) Schematic of the RPM OPO. M1, incoupling dielectric mirror; M2-M4, gold-coated mirrors; OC, an OPO in-coupler mirror; PZT, piezo-actuator for cavity-length control. Inset: OPO 'engine' including the ZnSe ceramic sample located between two parabolic mirrors. (b) 500 x 500 µm cross section of our chemically etched ZnSe ceramic sample. (c) OPO output spectrum vs. cavity-length detuning showing a continuous spectral span of 3–7.5 µm.

Our approach is based on the phenomenon known as random (or 'stochastic') phase matching (RPM) [1,2]. RPM in disordered $\chi^{(2)}$ materials can be described in terms of random walk ('drunken sailor walk') theory that also accounts for such phenomena as diffusion and heat transfer. RPM eliminates the need for orientation patterning and, most importantly, enables 3-wave interactions with extremely large bandwidths. A broadband and flat response in RPM is the result of phase randomization due to arbitrary distribution of crystalline domains, which eliminates destructive interference. The price to pay, however, is a slow growth of the output signal since the output intensity in RPM scales linearly with the sample length ($\sim LL_{coh}$ in the optimal case), as opposed to the quadratic dependence ($\sim L^2$) for perfect phase or quasi-phase matching. (Here L is the physical length and L_{coh} is the coherence length for a 3-wave interaction.)

ZnSe, a cubic-symmetry semiconductor with a bandgap of 2.7 eV, is a perfect candidate for RPM because of its outstanding transparency range (0.55 to 20 µm), relatively high 2nd order nonlinearity ($d_{14} \approx 20$ pm/V), high optical damage threshold, and good mechanical properties. A proof of principle experiments on mid-IR difference frequency generation via RPM in ZnSe ceramics have been carried out using nanosecond pulses [2]. Although in continuous-wave and nanosecond regimes RPM is much less efficient, than the conventional phase- (or quasi-phase) matched process, our finding is that RPM is very well suited for nonlinear conversion when few-optical-cycle pump pulses are used. For example, efficient RPM intracavity frequency doubling, tripling and quadrupling of femtosecond 2.35-µm laser pulses have been recently demonstrated [3].

Here we demonstrate the world's first OPO based on RPM. The key to success was (i) using pump pulses with long enough center wavelength (2.35 µm), which corresponds to a large OPO coherence length ($L_{coh} \sim 100$µm for ZnSe) and (ii) processing commercial ZnSe ceramic samples, via high-temperature annealing, to increase the average grain size to be close to L_{coh}, – an optimal condition for RPM [2].

The OPO was synchronously pumped by a Kerr-lens mode-locked Cr:ZnS laser with the center wavelength 2.35 µm, 62-fs pulse duration, 650-mW average power, and 79-MHz repetition rate. The bow-tie ring OPO cavity (Fig. 1) was composed of an in-coupling dielectric mirror M1 with high transmission (>85%) for the 2.35-µm pump and high reflection (>95%) for 3-8 µm, two gold-coated parabolic mirrors (M2 and M3) with 30° off axis angle and 30 mm apex radius, five gold-coated flat mirrors (for simplicity only M4 was shown in Fig. 1; the other four mirrors were used for folding the beams to reduce the footprint). An uncoated plane-parallel polished L=1.5 mm ZnSe ceramic sample placed at Brewster angle acted as gain medium. A 0.3-mm-thick ZnSe wedge was used inside the cavity for (variable) outcoupling the OPO radiation [4]. The OPO operated in a doubly resonant frequency-divide-by-2 mode at degeneracy. In addition to lowering the pump threshold, this arrangement provides other advantages [5]: (i) phase- and frequency locking to the pump laser – a prerequisite for creating precision mid-IR frequency combs, and (ii) the possibility of achieving extremely broadband spectrum due to negligible group velocity dispersion of ZnSe (ZnSe has zero GVD crossing at 4.8 µm, i.e. near OPO degeneracy). The OPO threshold was measured to be 90 mW in terms of the average pump power and the measured output spectrum spanned 3 -7.5 µm (-40 dB level) and was centered at 4.7-µm degeneracy wavelength (Fig. 1c). At the maximum pump, the OPO average power was ~20 mW, with the OPO outcoupling not being optimized. The pump depletion was as high as 79%, which indicates that one can obtain high, approaching 100%, conversion efficiency from such a device.

In conclusion, we demonstrate an OPO based on random phase matching in a disordered polycrystal, ZnSe ceramic. This is the first $\chi^{(2)}$ OPO that utilizes disordered material and the first OPO based on ZnSe. Thanks to a very broad gain bandwidth of RPM based frequency conversion devices, RPM can be used for generating few-cycle mid-IR pulses as well as multi-octave frequency combs. More generally, RPM in ZnSe and similar polycrystalline ceramic materials, such as ZnS, ZnTe, ZnO, CdSe, GaP, GaN, opens up new avenues in nonlinear optics.

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