Comment on "Generation of 10^{11} contrast 50 TW laser pulses"

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We argue for a different physical interpretation of the results given in the recent Letter by Chvykov *et al.* [Opt. Lett. **31**, 1456 (2006)] in which a double nonlinear crystal scheme for cross-polarized wave generation is analyzed. We discuss the most important factors that explain the origin of the two-crystal scheme's increased efficiency, namely, the Kerr lensing effect and a Gouy phase shift. The position and orientation of the second crystal relative to the first one are unambiguously defined; related effects are illustrated by already published works on the subject.

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In a recent paper [Opt. Lett. **31**, 1456 (2006)] Chvykov *et al.*[1] described an experiment in which high-contrast-ratio femtosecond pulses were obtained using tandem of $\chi^{(3)}$ nonlinear crystals. The maximum efficiency (22%) of cross-polarized wave (XPW) generation presented in Ref. 1 is explained by the authors as an independent azimuthally adjustment of the two crystals that is supposed to compensate for the "polarization rotation" in the first nonlinear crystal. We do not agree with this explanation, as polarization rotation does not contribute to the XPW generation process. The underlying physics is more intricate, as can be deduced from our previous experimental works[2–5] and theoretical investigations.[2] We already demonstrated and explained that the double-crystal scheme, patented in 2004,[6] allows one to increase the efficiency of the XPW process, which cannot be achieved in a single-crystal configuration either by increasing the nonlinear crystal length or by increasing the intensity.

In this Comment we prove that the azimuthal rotation of the crystals is not the decisive factor that influences the double-crystal scheme's efficiency. Adjustment of the optimal distance between the crystals was demonstrated in Ref. 2 to be of prime importance. Only the simultaneous adjustment of both angle and distance can lead to the achieved efficiencies. We suppose that the authors of Ref. 1 did not notice the optimal distance dependence because of the strong focusing ($f=10$ cm), and consequently of the small fundamental spot size, of a scheme where the optimal distance is comparable with the thickness of the BaF_2 crystals used in the experiment. We will assume, from the experimental details given in Ref. 1, that the distance between the two crystals is less than 1 mm. Indeed, the model described in Ref. 2 predicts a linear dependence of the optimal distance on the confocal parameter of the fundamental beam focused on the first crystal: the smaller is the confocal parameter of the fundamental beam, the smaller is the optimal distance (see Fig. 5 of Ref. 2). A simple scaling of our result,[2] where an optimal distance of 6.8 mm is obtained with $f=30$ cm, and assuming equivalent beam divergence, leads to the result that with $f=10$ cm the optimal distance should be 9 times smaller, say, the 0.75 mm value that was used as the assumed value for comparison with other experiments.

It is interesting to compare different available results to illustrate the role of the fundamental beam spot size in the plane of the first crystal. We present in Fig. 1 a compilation of published optimal distances[1–5] with two-crystal schemes for XPW generation obtained with different focusing (or different spot sizes) of the fundamental beam on the two-crystal setup. The deviation of the experimental points from the prediction of the model[2] (line with slope of 1) shown by the dashed line in the figure, is

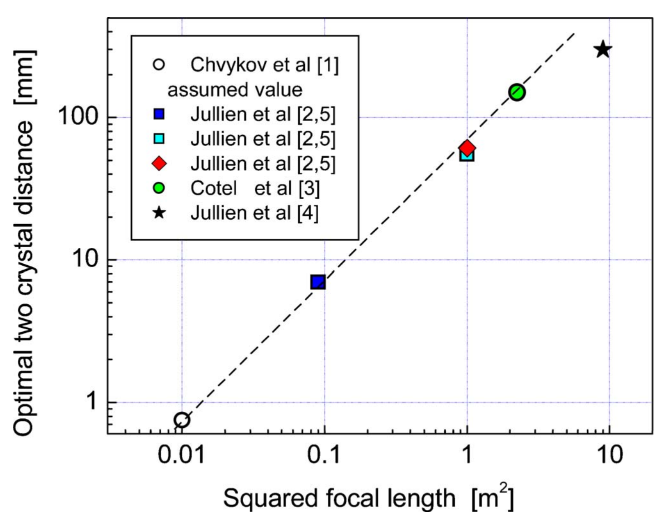

Fig. 1. (Color online) Summary of published optimal distances[1–5] with two-crystal schemes for XPW generation obtained with different focusing of the fundamental beam on the two-crystal setup. The dashed line corresponds to the theoretically predicted slope of 1.

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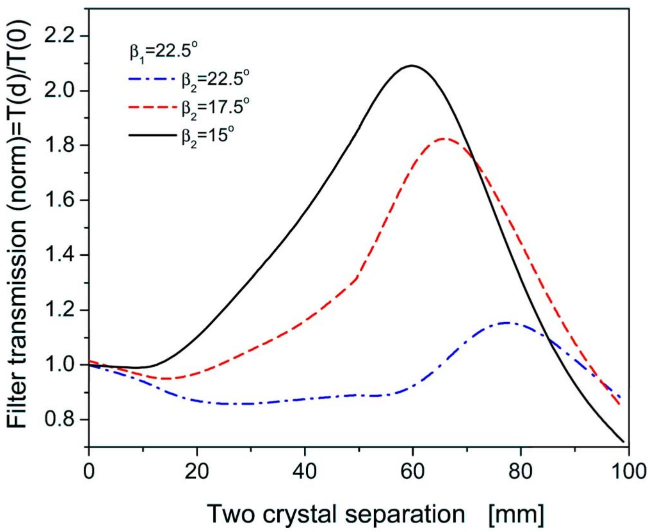

Fig. 2. (Color online) Theoretical dependences of the distance optimization curves calculated for different angles β_2 with the model given in Ref. 2 without time integration. The input beam has a radius of 150 μm (confocal parameter ~32 cm). Normalized input intensity $S=\gamma_0|A_0|^2L=2$.

due to uncertainties of the beam parameters for the various experiments.

To understand the relative roles of the optimization of the azimuthal angle β_2 of the second crystal and of the optimization of the distance between the crystals, we show in Fig. 2 the theoretical dependence of the distance optimization curves calculated for different angles β_2, using input beam radius $a=150$ μm (confocal parameter $ka^2n=32$ cm) and normalized input intensity $S=\gamma_0|A_0|^2L=2$,[2] a scheme that theoretically gives for a single 2 mm BaF_2 crystal a XPW generation efficiency of the order of 10%. It can clearly be seen from Fig. 2 that, when the confocal parameter is greater than the crystal thickness and the crystals are closely situated, optimization of the second crystal angle β_2 cannot lead to any serious increase of the efficiency. Adjustment of angle β_2 can lead to an improvement of the efficiency only when the crystals are located at a distance close to the optimal.

The authors of Ref. 1 claim that the process of XPW generation is a consequence of fundamental polarization rotation inside the nonlinear crystal, and the second crystal therefore has to be azimuthally rotated to compensate for this polarization rotation. This is not actually a good interpretation. As seen from the simplified model for nondepleted regime,[7] the XPW is shifted by $\pi/2$ with respect to the fundamental wave; this means that the field in the space between the crystals corresponds to an elliptically polarized light with the main axes lying in the polarization planes of the polarizers. A small rotation of this ellipse can arise from self-phase modulation of the fundamental beam, which results in a deviation of the phase shift between the fundamental wave and the XPW from the value $\pi/2$. Even at highest possible close to damaging intensity, the polarization rotation in BaF_2 in this process does not exceed 4°.[7] This small rotation cannot explain the observed efficiencies of XPW generation.

The reason for the increased efficiency due to azimuthal rotation of the second crystal (shown in Fig. 2) is that this rotation allows adjustment of the optimal phase between the fundamental and the generated XPWs toward better efficiency and constructive interference of the XPW signals generated in both crystals.

To summarize, the reasons for the improved efficiency at certain optimal distance between the two crystals are these:

1. Kerr lens refocusing of the fundamental beam in the space between the two crystals that leads to a smaller diameter in the plane of the second crystal. Furthermore, Kerr lensing is filtering the fundamental beam from its high spatial frequencies, creating a cleaner beam on the second crystal. Therefore XPW efficiency in the second crystal is improved.
2. Achievement of optimal phase shift between the fundamental wave and the XPW at the input of the second crystal. The change of the phase shift between these two waves is due to different accumulation of Gouy phase by propagation in the space between the crystals. The fundamental wave, which is more intense, is more strongly focused by the Kerr lens and consequently collects more Gouy phase shift. Recovering the optimal (or close to optimal) phase shift at the input of the second crystal leads to good phase matching between the generated XPW signals in the two crystals. They are in phase and can interfere constructively.
3. The possibility of independent optimization of angles β_1 and β_2, which is also used for obtaining the optimal phase shift between the fundamental wave and the XPW.

We believe that this Comment will be useful for understanding the physics of the increased efficiency in two-crystal schemes, a fact of prime importance for correct design and prediction of XPW filters for contrast improvements.

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