A new crystal for nonlinear optical generation in the mid-infrared

We have measured the phase matching directions for second harmonic generation and difference-frequency generation in the nonlinear crystal $CdSiP_2$, at infrared wavelengths up to 9.5 µm. By a simultaneous fit to all the angular data, the wavelength dependence of the ordinary and extraordinary principal refractive indices were refined over the entire transparency range of the crystal. This work shows the capability of $CdSiP_2$ for broadband generation of mid-infrared light.

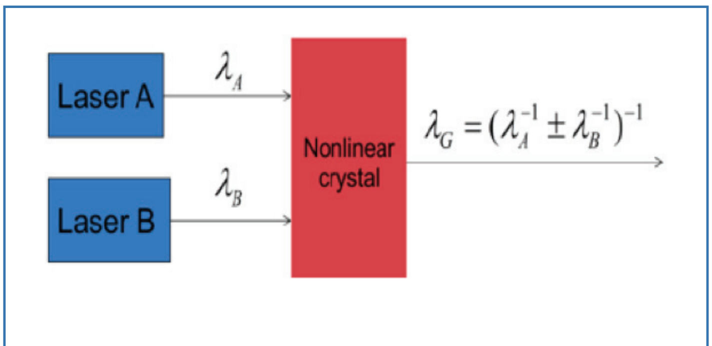

Figure 1 Scheme of sum-frequency generation and difference-frequency generation. λ_A and λ_B are the wavelengths of the incident beams; λ_G is the wavelength of the output wave generated by sum- or difference-frequency interactions

Generating light in the mid-infrared range beyond 5 µm remains an open problem due to the lack of suitable laser sources. Targeted fields of application are medicine, atmospheric telecommunication and spectroscopy. Crystals with strongly nonlinear optical properties appear attractive here because they can convert the wavelength of an incident laser beam into shorter or longer wavelengths via the processes of sum-frequency or difference-frequency generation respectively.

In this context CdSiP2 was recently identified as a promising crystal enabling efficient nonlinear generation near 6.2 µm when pumped by a Nd:YAG laser at 1.064 µm. $CdSiP_2$ is a semiconductor having the tetragonal symmetry, "chalcopyrite" crystal structure. Transparent between 0.56 µm and 10 µm, it exhibits one of the highest second order optical nonlinearities amongst infrared materials. We have performed the first complete measurements of the "phase-matching directions" in the crystal lattice of $CdSiP_2$, as a function of wavelength.

We did this for the two cases of second harmonic generation (SHG), which corresponds to a sum-frequency generation process where the wavelengths of the two incident beams are equal, and difference-frequency generation (DFG). These processes are shown in Fig. 1. The processes have maximum efficiency for propagation along crystal lattice directions where the incoming and outgoing waves have certain, precise, phase relations. The phase-matching condition is related to the dependence of the refractive index on propagation direction and frequency.

The phase-matching angles of $CdSiP_2$ were determined by using a highly accurate sphere cut from a single crystal at Néel Institute. This unique technique has the great advantage of allowing a laser beam to propagate undeviated in any crystal direction. Thus only one sample of the crystal is needed to do a complete and accurate angular variation study. The $CdSiP_2$ sphere was attached to a goniometric head as shown in figure 2. Its tetragonal axis was oriented horizontally with precision better than 0.05° in an automatic X-ray diffractometer before transfer of the goniometric head and sample to the optical bench.

With the sphere at the centre of an Euler circle, it could be rotated about its centre, the direction of the incident laser beams being kept fixed. A focusing lens located at the entrance side of the sphere ensures quasi-parallel propagation of the light beams inside the sample. The wavelength of the incident beams ranged between 0.4 µm and 10 µm. The phase-matching directions and associated conversion efficiencies were recorded for two kinds of processes: second harmonic generation by using as the only input the wavelength tunable beam of a parametric oscillator source; and difference frequency generation by mixing this tunable beam with an auxiliary (YAG laser beam) at 1.064 µm.

Rotating the sphere on itself, we identified the lattice directions for SHG or DFG phase-matching as being the directions where the associated frequency conversion efficiencies were maximum. By fitting all the measured phase-matching curves between 3 µm and 8 µm for single harmonic generation and over 6 µm and 9.5 µm for difference frequency generation, we refined the equations for the wavelength dispersion of the ordinary and extraordinary principal refractive indices. We could then calculate the tuning curve for mid-infrared generation in $CdSiP_2$ via a "parametric fluorescence" process with only a Cr2+:ZnSe laser at 2.4 µm as pump. This would generate simultaneously a super-continuum spreading from 3 µm to 8 µm.

Figure 2 The 5-mm-diameter $CdSiP_2$ sphere placed on the top of a high precision angular positioning stage.

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FURTHER READING

PHASE-MATCHING PROPERTIES AND REFINED SELLMEIER EQUATIONS OF THE NEW NONLINEAR INFRARED CRYSTAL $CDSIP_2$
V. Kemlin, P. Brand, B. Boulanger, P. Segonds, P. G. Schunemann, K. T. Zawilski, B. Ménaert, and J. Debray
Optics Letters 36, 1800-1802 (2011).