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 CdSiP₂, 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 CdSiP₂ for broadband generation of mid-infrared light.

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. CdSiP₂, 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 CdSiP₂, 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 CdSiP₂, were determined by using a highly accurate sphere cut from a single crystal at Neel Institute. This unique technique has the great advantage of allowing a laser beam to propagate undisturbed in any crystal direction. Thus only one sample of the crystal is needed to do a complete and accurate angular variation study. The CdSiP₂ sphere was attached to a goniometric strob at as shown in Fig. 2. Its tetragonal axis was orientated 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 (Nd: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 CdSiP₂, via a “parametric fluorescence” process with only a Cd₃-In₂ laser at 2.4 μm as pump. This would generate simultaneously a super-continuum spreading from 3 μm to 8 μm.

We have recently taken the first steps in this direction by attaching an NV centre on the vibrating extremity of a SIC nanowire. In order to obtain mechanical control over the nanowire, it was attached at the apex of a tungsten tip, which was positioned on a piezo-electric crystal. The NV fluorescence was collected through a high numerical-aperture microscope objective on ultrasensitive avalanche photodiodes (see Fig. 1). The electronic structure of the NV defect exhibits an optical transition in the visible, which constitutes a stable single photon source. By recording the emitted photon rate, we observed the effect of the resonator’s motion across the optical spot of the microscope. More precisely, with an autocorrelator optical setup, we observed the influence of the nanoresonator vibrations (hence its mechanical motion used in this experiment) on the collected photon statistics.

A single spin magnetically coupled to a nanomechanical oscillator

The intense experimental efforts of several groups worldwide during the last six years have very recently culminated by demonstrating what the scientific community considered unrealistic only 10 years ago: cooling a macroscopic mechanical oscillator down to its quantum ground state of motion. Experiments going beyond ground state cooling and aiming at generating non-classical states of motion require an actual engineering of the quantum mechanical state of the oscillator. This can best be achieved by coupling the ultracold oscillator to a second quan tum state through which one can interact with the oscillator.

The combination of these two components defines a “hybrid” mechanical system. The goal of our research is to investigate these novel hybrid quantum systems consisting of a nano-mechanical oscillator and a fluorescent Nitrogen-Vacancy (NV) defect centre in a diamond nanomembrane. In the work presented here, our system is at room temperature, but we plan to do experiments with ultracold reservoirs in the future. The electronic spin state of the defect represents a unique quantum system for our purpose: it features ultralong coherence times and can be read out and manipulated with optical and microwave fields. The ultimate goal of the project is to enter the quantum regime of a hybrid nano-mechanical system, where the spin and the oscillator dynamics are entangled, and to investigate phenomena at the interface between the classical and quantum worlds.

Furthermore, the NV defect ground state is a spin triplet state with an ultralong coherence time that can be manipulated with microwave fields. We achieved magnetic coupling of the NV defect spin state to the oscillator’s position via the combination of an external strong magnetic field gradient and the Zeeman splitting of the energy levels of the NV centre. More precisely, we immerse the system in a field gradient of 100 kHz/m (by approaching a structured permanent magnet). At the nanoresonator’s mechanical transition into motion, the NV centre sweeps through the magnetic field gradient, thereby experiencing large variations in the magnetic field. Th us, the energy levels of the NV centre, which depend on the magnetic field through the Zeeman effect, are subject to oscillations as well, and consequently are coupled to the nanoresonator motion.

The NV spin energy levels were probed by scanning a microwave field across the transition energy, while having the nanoresonator vibrating. By combining the microwave spectroscopy and a simultaneous optical read-out, a clear splitting in the spin energy spectrum, induced by the nanomotion, was observed in the fluorescence spectrum. In reverse, this coupling intrinsically generates a spin dependent force, whose magnitude is quantized and depends on the NV defect’s spin state. This represents the key ingredient for mapping the quantum state of the spin onto the nanoresonator and thus creating non-classical states of motion. The possibility of generating spin-dependent forces combined with groundstate cooling of mechanical reservoirs will in the future allow us to perform quantum physics experiments with nano-mechanical oscillators.