Stimulated Raman Scattering in potassium nitrate, α-KNO_3

Oliver Lux[1,2], Hans-Joachim Eichler[1], Alexander A. Kaminskii[3], Ladislav Bohatý[4], Petra Becker[4]

[1]Institute of Optics and Atomic Physics, Technical University of Berlin, Berlin, Germany, [2]German Aerospace Center (DLR), Institute of Atmospheric Physics, Oberpfaffenhofen, Germany, [3]Institute of Crystallography, Federal Scientific Center "Crystallography and Photonics", Russian Academy of Sciences, Moscow, Russia, [4]Institute of Geology and Mineralogy, Sect. Crystallography, University of Cologne, Cologne, Germany, ladislav.bohaty@uni-koeln.de, petra.becker@uni-koeln.de

Stimulated Raman scattering (SRS), a $\chi^{(3)}$-based nonlinear optical process, was discovered in bulk media in 1963 [1] shortly after the invention of the laser. In the following, the applicability of SRS for laser frequency shifting was demonstrated and triggered an intensive search for suitable crystalline media. Among others, nitrates were investigated and in particular cubic $Ba(NO_3)_2$ turned out to be an efficient material for the construction of crystalline Raman lasers [2]. For all nitrate crystals investigated so far, the singular SRS-promoting vibration mode is the fully symmetric vibration of the $[NO_3]$ group with energies of about 1050 cm^{-1} [3], resulting in relatively large Raman frequency shifts of tens to hundreds of nanometers depending on the fundamental (or pump) laser wavelength. While for the alkali nitrates $NaNO_3$ and $CsNO_3$ SRS data are available [3], surprisingly for KNO_3 no information about SRS is known so far in literature.

In this work, SRS and Raman-induced four wave mixing (RFWM) processes were studied for orthorhombic α-KNO_3. Large single crystals of optical quality of α-KNO_3 were grown at 310 K from aqueous solution by controlled slow evaporation of the solvent. The size of the crystals allowed the preparation of samples with faces perpendicular to all orthorhombic main axes and typical dimensions of 10 – 20 mm, which enabled single-pass SRS experiments with sufficient nonlinear-lasing efficiency.

SRS experiments were performed with single-wavelength picosecond laser excitation in the near IR (λ_{f1} = 1.06415 μm, 80 ps) and in the visible spectral range (λ_{f2} = 0.53207 μm, 60 ps) for the three orthorhombic main directions of α-KNO_3. In potassium nitrate, as similarly in other nitrate crystals investigated so far, SRS arises from the vibration mode with $\omega_{SRS} \approx 1050$ cm^{-1}, that is related to the fully symmetric $A_g(\nu_1)$ vibration of the $[NO_3]$ structural units. Interaction of the incident infrared and green laser radiation with this mode via SRS and RFWM gives rise to the generation of more than ten new emission lines ranging from the ultra-violet to the near-infrared spectral range. Unlike other studied nitrates, two lattice modes in the energy region of 50 – 100 cm^{-1} were found to be SRS-active in α-KNO_3 as well. Here, an analogy can be drawn to the isomorphic carbonate compound aragonite ($CaCO_3$), where the detected SRS-promoting modes also correspond to three vibration modes: the totally symmetric A_{1g} breathing mode of the $[CO_3]$ units and two lattice modes in the energy range 150-200 cm^{-1} [4].

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[4] Kaminskii AA, Rhee H, Lux O, Eichler HJ, Koltashev VV, Kleinschrodt R, Bohatý L, Becker P. Stimulated Raman scattering spectroscopy and $\chi^{(3)}$-nonlinear lasing effects in single crystals of aragonite (orthorhombic $CaCO_3$). Laser Phys. Lett., 9, 259-284 (2012)