Stimulated Raman Scattering in potassium nitrate, α-KNO_3

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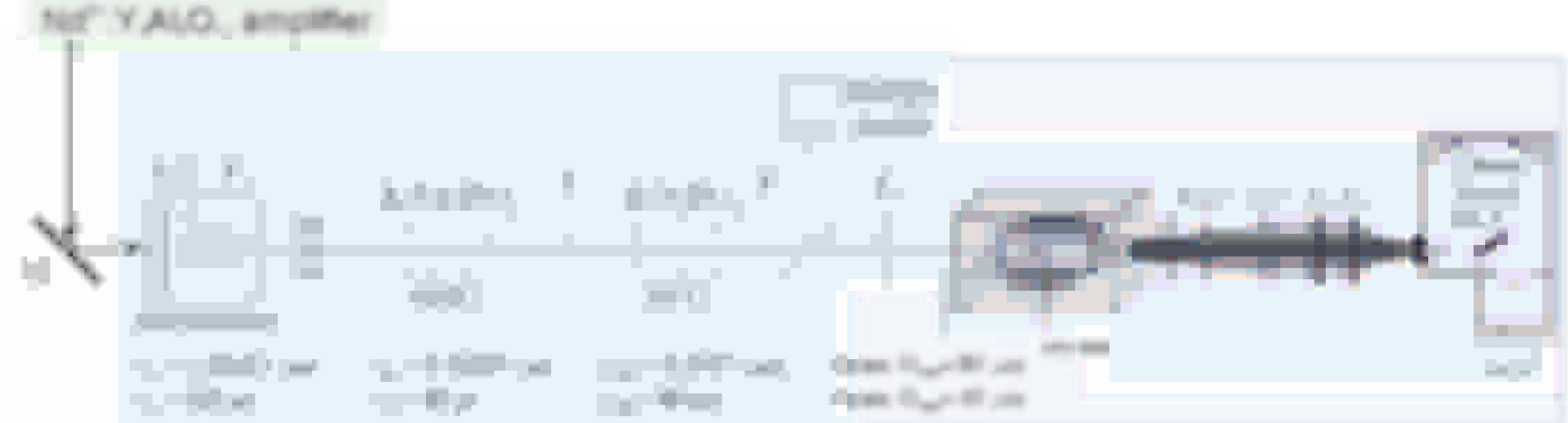
Introduction

The applicability of Stimulated Raman scattering (SRS), a $\chi^{(3)}$-based nonlinear optical process, for laser frequency shifting was demonstrated shortly after the invention of the laser [1] 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 [3], which gives rise to 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 using successfully grown single crystals of orthorhombic α-KNO_3. On the basis of refractive indices and their dispersion measured on these crystals phase matching properties for non-collinear RFWM-based anti-Stokes emission are analysed.

Stimulated Raman scattering and RFWM

- Measurement setup (with home-made laser) [5, 6]: (Raman-induced Four Wave Mixing)

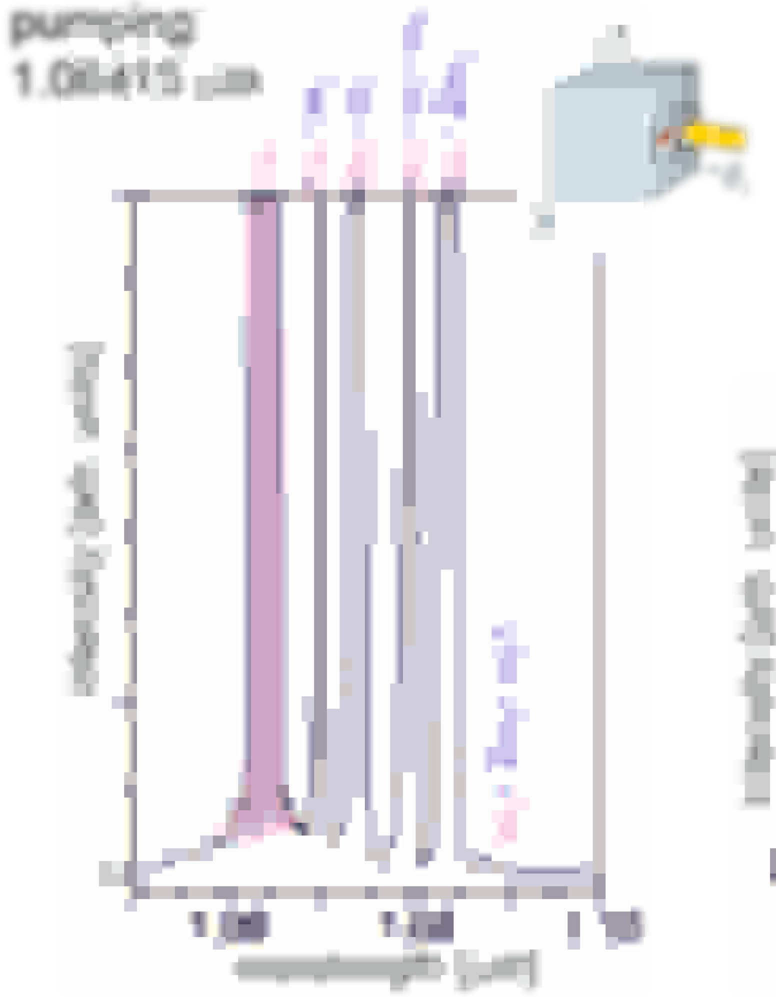
$Nd^{3+}:YAlO_3$ amplifier




Sensor spectral sensitivity

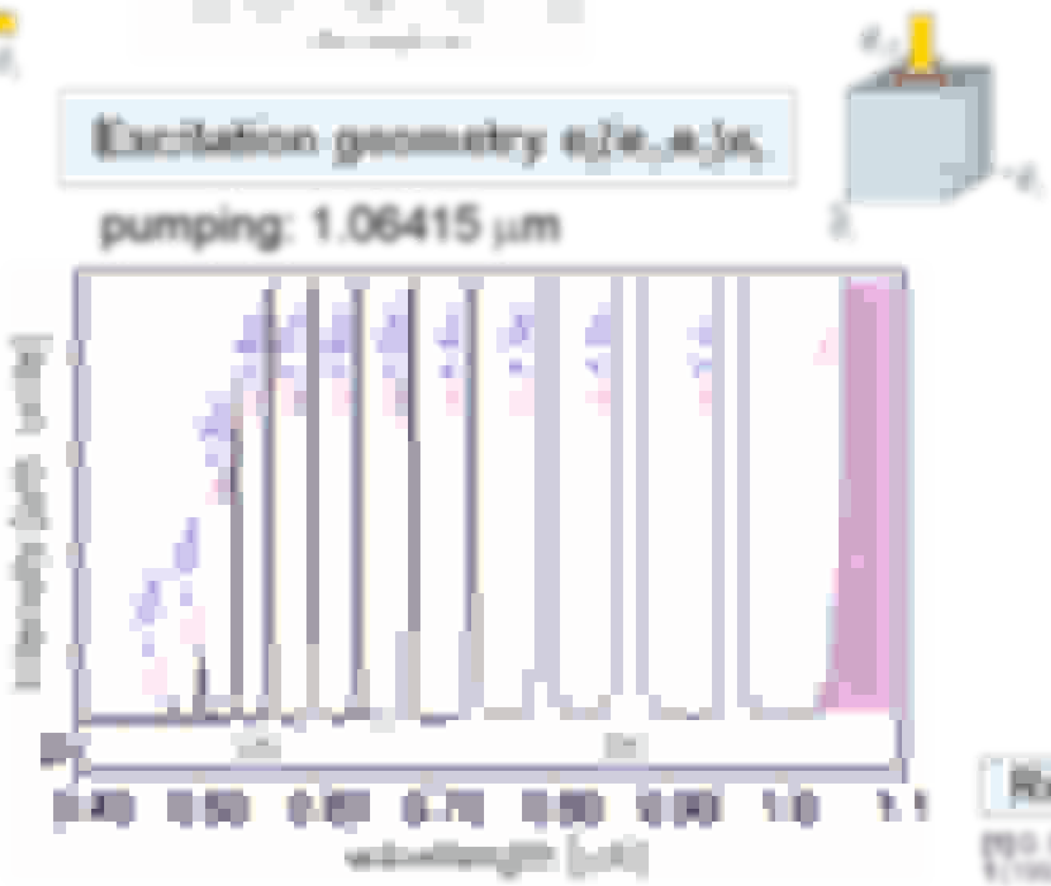
Excitation geometry

pumping 1.06415 µm



Excitation geometry

pumping: 1.06415 µm



Crystals of α-KNO_3

Crystal growth

Growth of α-KNO_3 by the **solution growth method**.

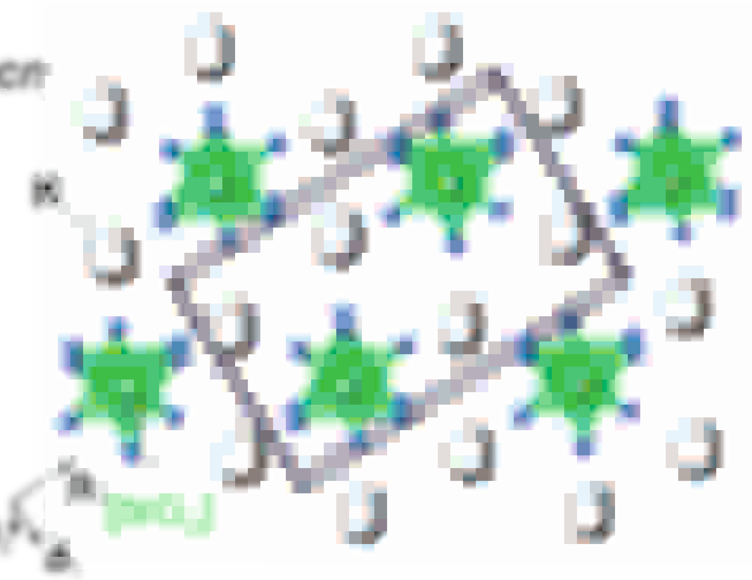
Best growth results:
Crystal growth from aqueous solution at 38°C by controlled evaporation of the solvent.
Growth duration ca. 15 weeks.

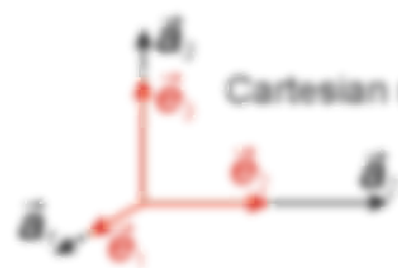
Typical morphology of α-KNO_3 crystals:
Pinacoids {001}, {010}, prisms {110}, {021} and dipyramid {111}

Crystal structure

- Aragonite-type structure, space group *Pmcn* (non-standard setting *bca*)
- Isolated coplanar $[NO_3]$ groups stacked along the a_1-axis
- K atoms nine-fold coordinated by oxygen

(Structure data taken from [4])

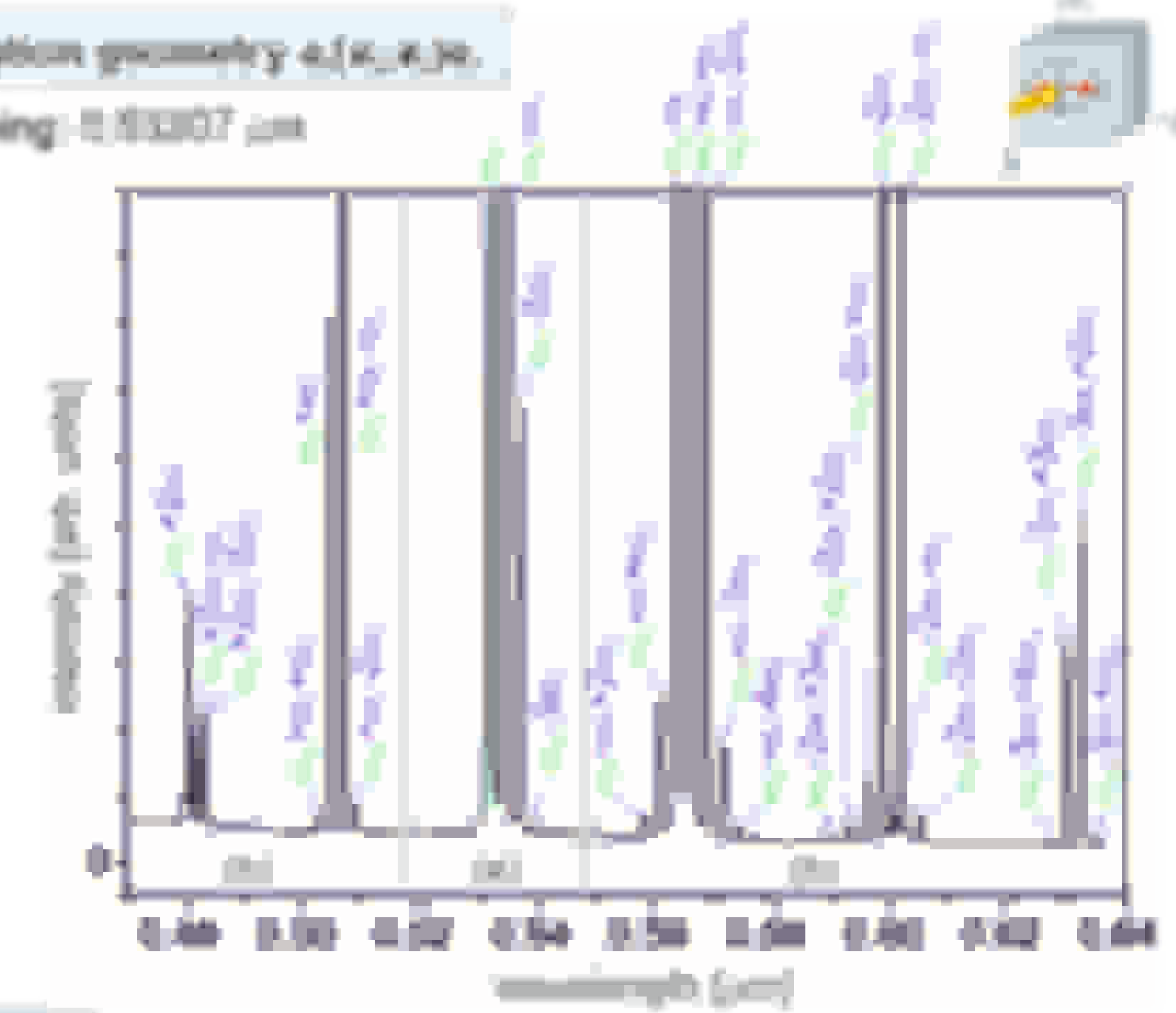




SRS-promoting vibration modes (at 300 K):
$\omega_1 \approx 1050\ cm^{-1}$ ($A_g(\nu_1)$ of $[NO_3]$) $\omega_2 \approx 53\ cm^{-1}$ $\omega_3 \approx 83\ cm^{-1}$ (lattice modes)
(assignment according to spontaneous Raman spectroscopy investigations [7, 8])

Excitation geometry

pumping: 0.53207 µm



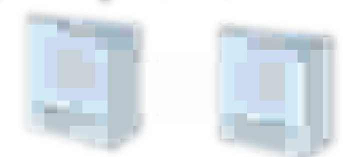
Linear optical properties

Non-polarized transmission spectrum

Plates (100), (110) and (001) of ~3 mm thickness

Refractive indices and their dispersion

- Normal incidence using two prisms

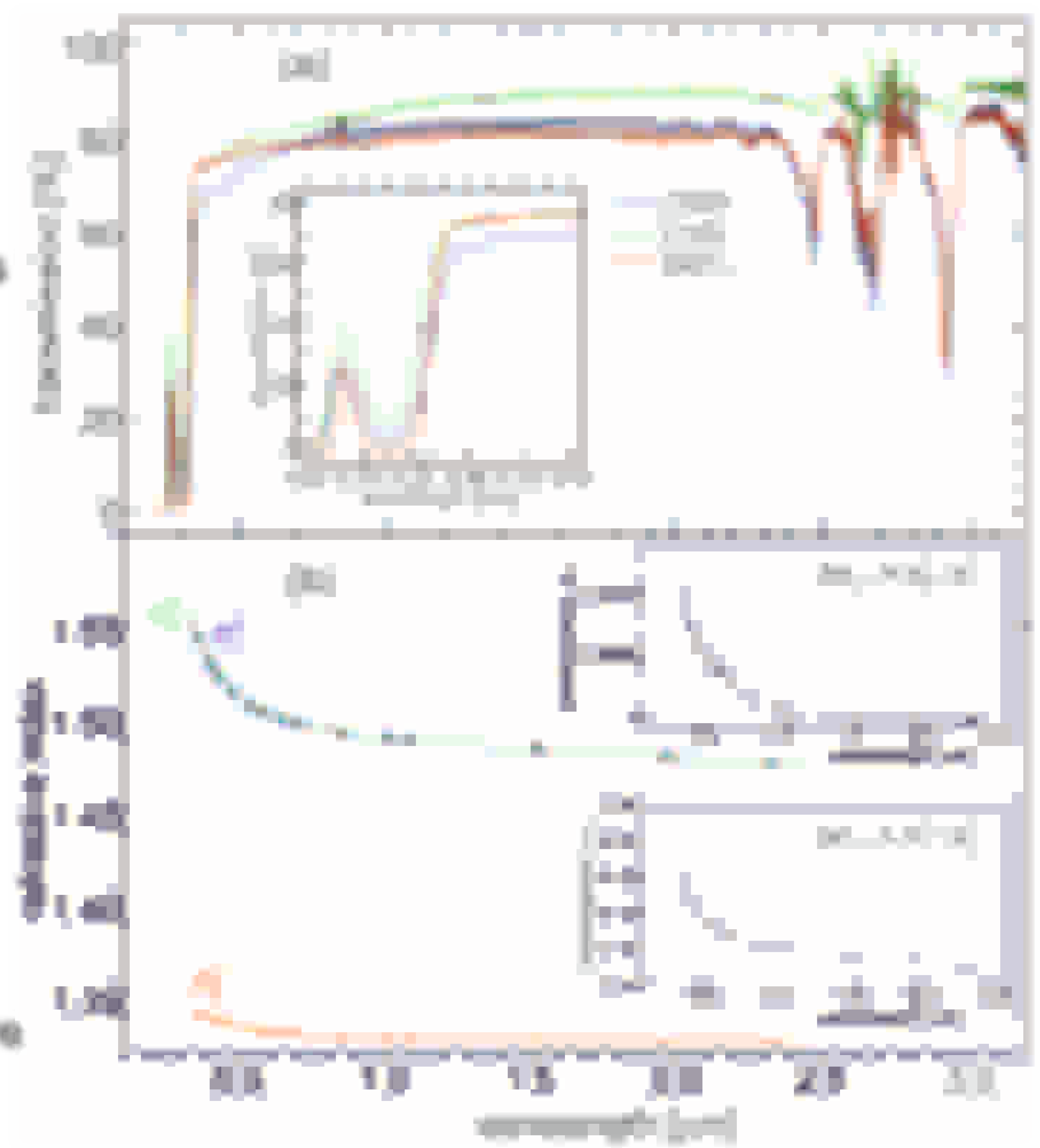


- 14 discrete wavelengths in the range 0.365 - 2.325 µm

➡

- transmission range ~0.33 µm - ~2.35 µm
- small birefringence $\Delta n_{21} = |n_2 - n_1|$
- strong birefringence $\Delta n_{31} = |n_3 - n_1|$ and $\Delta n_{32} = |n_3 - n_2|$

➡ optical character pseudo uniaxial negative



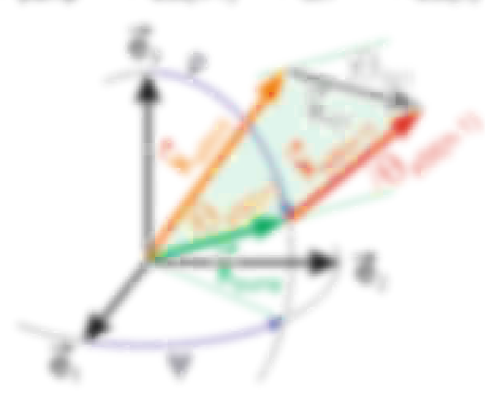
RFWM phase matching

Non-collinear phase matching for four-wave-mixing process

Phase matching conditions:
- first anti-Stokes generation: $2\vec{k}_{pump} = \vec{k}_{S1} + \vec{k}_{AS1}$
- higher (n^{th}) anti-Stokes generation: $\vec{k}_{pump} + \vec{k}_{AS(n-1)} = \vec{k}_{S1} + \vec{k}_{ASn}$

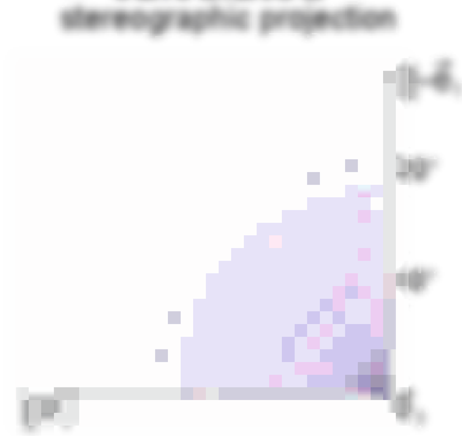
➡ **Conical anti-Stokes emission**

Example:
Pumping wavelength $\lambda_{pump} = 1.064\ \mu m$, $\vec{k}_{pump} \parallel \vec{e}_1$, $\omega_1 \approx 1050\ cm^{-1}$

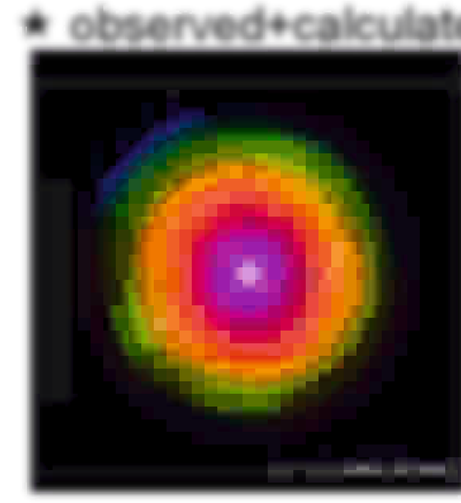


→ Anti-Stokes emission angles θ_{ASn}

- calculated (stereographic projection)



- observed+calculated



➡ slightly elliptical anti-Stokes emission cones

References