Control of Stimulated Polariton Scattering in Periodically Structured KTiOPO$_4$

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Quasi-phase-matching (QPM) in ferroelectrics is based on periodical structuring of the second-order nonlinearity by locally inverting the sign of the $\chi^{(2)}$ susceptibility. Due to symmetry reasons this sign inversion does not affect pure odd-order susceptibilities such as $\chi^{(1)}$ (refractive index) or $\chi^{(3)}$. (Kerr effect). The corollary of this simplified picture is that stimulated Raman scattering (SRS), which is also an odd-order effect, should not be affected by the spatial structuring of the ferroelectric domains. In this work we will show that this is not the case and that structuring the ferroelectric does indeed present an opportunity to control the SRS process.

In some ferroelectrics the crystal bonds responsible for the permanent microscopic and bulk dipole moment are also the main contributors to the second-order nonlinear response of the material. For example, in KTP crystals one finds alternating long and short titanyl (Ti-O) bonds along the polar *z*-axis rendering the largest second-order coefficient d$_{33}$. Vibrations of the same titanyl bonds give rise to the strongest Raman scattering lines at a frequency shift of 8 THz and 21 THz. From this simple qualitative discussion one should conjecture that it should be possible to modify SRS properties by periodic poling of ferroelectrics since the same bonds are associated with the strongest SRS lines and the sign of $\chi^{(2)}$.

The SRS in KTP is governed by phonon-polariton scattering. The scattering angles and frequency shifts are determined by the energy conservation $\omega_p = \omega_s + \omega_{pol}$ and momentum conservation, $\vec{k}_p = \vec{k}_s + \vec{k}_{pol}$ where the subscripts *p*, *s*, *pol* signify pump, Stokes and polariton waves. The Stokes angle α can be obtained from the momentum conservation, $\Delta k^2 = k_{pol}^2 = k_p^2 + k_s^2 - 2k_p k_s \cos\alpha$.
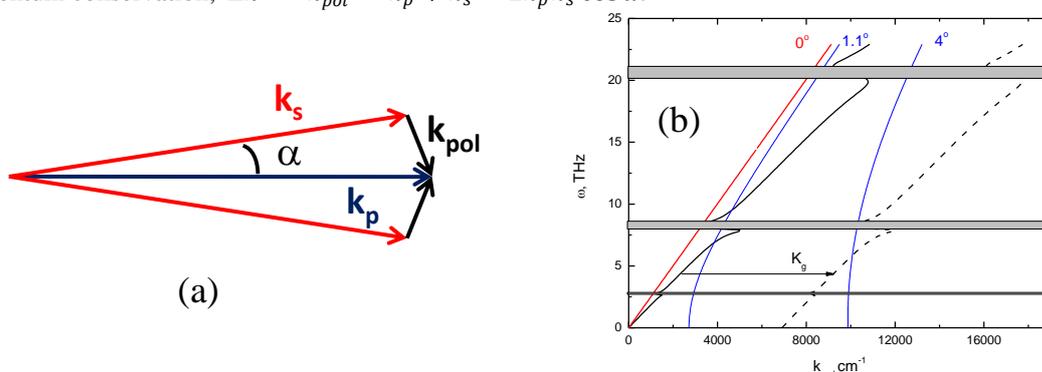
Fig. 1 a) Phase matching of forward polariton SRS, b) Polariton dispersion in KTP (black solid line) and PPKTP with QPM grating period of 9.1 µm (black dashed line). Color lines show phase-mismatch **k**$_p$-**k**$_s$ for different noncollinear angles α.

Shown in the Fig.1b by a solid black line is an estimate of the polariton dispersion in KTP calculated by taking into account the three strongest A1 symmetry phonon modes [1]. The colored lines show the dependence of Δk as a function of Stokes detuning for several noncollinear angles α. α=1.1° corresponds approximately to the polariton wave-front propagating at an angle θ=40° with respect to the pump, α=4° corresponds to θ=80°. The shaded regions in the Fig. 1b represent polariton gaps characterized by strong absorption where excitations cannot propagate. The intersection between the Δk line and the polariton dispersion curve would give the frequency of the polariton wave. When the material is periodically poled the momentum-conservation relation becomes $\vec{k}_p - \vec{k}_s = \vec{K}_g + \vec{k}_{pol}$ and hence the polariton dispersion curve is shifted as shown in Fig. 1b. Intersection with the Δk line still occurs, but now the pump and polariton propagate at almost right angles, resulting in a very small overlap between the waves and hence a conversion efficiency that approaches zero.

It has been experimentally verified in many samples that when the pump propagates in the single-domain part at intensities 2-times above SRS threshold, about 40 % of the pump is converted into SRS. If the pump instead propagates through a periodically poled region in the same crystal the SRS is completely suppressed.

References

[1] G. E. Kugel, F, Bréhat, B. Wyncke, M. D. Fontatna, G. Marnier, C. C. Nedelec, and J. Mangin, "The vibrational spectrum of KTiOPO$_4$ single crystal studied by Raman and infrared reflectivity spectroscopy, " J. Phys. C **21**, 5565-5583 (1988).