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Influence of Intrinsic Phonon Modes in Nonlinear Optical Crystals on the Performance of Ultrafast Frequency Conversion Devices

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Abstract: We demonstrate that coherently-excited Raman-/IR-active phonons in certain nonlinear optical crystals (KTiOPO₄, KTiOAsO₄) significantly affect the operation of ultrashort-pulse pumped optical parametric amplifiers. The photon-phonon coupling in these materials leads to non-instantaneous, pump-dependent 2nd-order susceptibility.

OCIS codes: (190.4410) Nonlinear optics, parametric processes; (190.7110) Ultrafast nonlinear optics

Nonlinear optical (NLO) frequency conversion devices such as harmonic generators, optical parametric oscillators/amplifiers (OPO/OPA) etc., have become indispensable in the industrial and research applications to generate tunable laser beams at frequencies that are outside of the spectra of typical laser gain media. One of the fundamental reasons behind the reliable operation of NLO laser sources is the instantaneous nature of the second-order nonlinear optical susceptibility $\chi^{(2)}$ that drives the three-photon mixing processes in these devices. Contrary to the operation of lasers with active-gain media (where the time-dependent accumulation of population-inversion is necessary), in NLO devices the electrons in the crystal lattice respond instantaneously to the oscillations of the interacting electromagnetic waves as long as the resonant transitions within the NLO medium are not involved in the frequency-conversion process. The latter allows carrying out various wave-mixing schemes in NLO crystals practically without a spectral gap over the corresponding crystal's transmission range, for as long as the phasematching condition is achieved.

In this work, we demonstrate that in certain NLO materials the effective $\chi^{(2)}$ may become largely modulated by intrinsic vibrational modes of the NLO material to such extent that it cannot be considered as instantaneous anymore (Fig. 1). Namely, we investigate the effects that arise when a typical femtosecond optical parametric amplifier (OPA) based on materials such as KTiOPO₄ (KTP) or KTiOAsO₄ (KTA) is pumped by broadband, ultrashort pulses (~50 fs duration) that are shorter than the characteristic oscillation period of the Raman- and IR-active phonon modes, e.g. the bending and stretching vibrations of the TiO₆ structural unit (~235 and ~700 cm⁻¹; ~7 and ~20 THz respectively) [1]. Such conditions create the possibility for the coherent impulsive stimulated Raman scattering (ISRS) excitation of the phonons, even though the pump pulse (~800-nm) is out of the resonance with any

Fig. 1. Characterization of the photon-phonon coupling in the ~50-fs pumped KTP-OPA in the frequency domain. (a) Exemplary pair of signal-idler spectra (black), measured at ~56 μJ pump pulse energy, together with a fit of the signal spectral profile (red) as well as the deduced parametric gain profile (magenta). “[0]” denotes the position of the signal based on the idler and ~795-nm pump; Δ, the red-shift of the signal spectrum from [0] to the signal center-of-mass frequency. The two bold arrows indicate the spectral positions of the “intrinsic” signal modes that oscillate out of phase with the effective nonlinearity $\chi_{eff}^{(2)}$ (and thus appear as “negative” peaks on top of the effective parametric gain profile). (b) Pump pulse energy dependence of the OPA output P_{Δ} for KTP (black squares) and BBO (blue circles) crystals, together with fits to the model $P_{\Delta} \propto [1 + \sinh^2(\text{const} E_p \chi_{eff}^{(2)})]$, where $\chi_{eff}^{(2)} = \chi_0^{(2)} + e^{i\theta} \chi_0^{(3)} E_p$, and $E_p = \text{const} (\text{Pump energy})^{1/2}$ [2].

vibrational modes of the crystal, and its frequency is much lower than the material's bandgap. This work provides the interpretation for our last year's report on the observation of considerable frequency red-shifts, in the corresponding KTP- and KTA-OPAs, of the [signal+idler] frequency from that expected based on pump (appearing as a "deviation" from the energy conservation principle, Fig. 1, a) [3]. The characterization of this phenomenon both in frequency- and time-domains [2] shows that the effective nonlinearity driving the OPA process can be modeled as $\chi_{eff}^{(2)} = \chi_0^{(2)} + e^{i\varphi} \chi_0^{(3)} E_p$, where $\chi_0^{(2)}$ and $\chi_0^{(3)}$ are the NLO material's intrinsic nonlinear susceptibilities, E_p is the pump wave electric field, and φ is the phase lag between the electronic and lattice oscillations.

Fig. 2. Seed-pump delay dependence of the KTP- (red) and BBO- (blue) OPA output. The double arrows indicate FWHM=75 fs thus corresponding to <50-fs pump pulsewidth. Inset: proposed model for the temporal modulation of the NLO crystal response due to the coherent impulsive excitation of lattice phonons [2].

The experimental optical layout is that of a common white-light seeded femtosecond OPA and is described in more detail elsewhere [2, 3]. In the frequency domain, the photon-phonon coupling in KTP and KTA crystals appears as the considerable red-shift Δ of the signal spectrum from its position based on the frequency difference between the pump and idler waves (Fig. 1, a) [2]. Due to the involvement of the phonon modes into the overall OPA process, the parametric gain profile becomes red-shifted by a value close to Δ (magenta line in Fig. 1, a) which correlates with the frequencies of the strongest phonon lines in the Raman spectra [2]. The signal waves whose amplification is driven by $\chi_{eff}^{(2)}$ oscillate out of phase with the "intrinsic" signal amplified by the instantaneous (electronic) susceptibility $\chi_0^{(2)}$ thus appearing as out-of-phase (negative) peak(s) on top of the effective parametric gain profile (Fig. 1, a; the double-"dip" feature is due to the double-peak structure of the pump scattered on the coherently-excited phonon-polariton modes [2]). Such modulation of the $\chi_0^{(2)}$ electronic nonlinearity by the resonant response of the NLO crystal (phonon-driven $\chi_0^{(3)} E_p$) is supported by the analysis of the pump intensity dependence of the OPA output (Fig. 1, b). Since for KTP and KTA, $\chi_0^{(3)} \approx 10^{-21} \text{ m}^2/\text{V}^2$, in the conditions of the applied pump peak intensities (corresponding to $E_p \approx 2 \cdot 10^9 \text{ V/m}$ or higher), the absolute values of $\chi_0^{(2)}$ and $\chi_0^{(3)} E_p$ become comparable. Note that for BBO-OPA the pump intensity dependence of the output can be fit by fixing $\chi_0^{(3)} = 0$ (Fig. 1, b; blue) which is supported by much weaker third-order nonlinearity in this material. Furthermore, the non-instantaneous nature of $\chi_{eff}^{(2)}$ in the KTP- and KTA-crystals with ISRS-excited phonons becomes apparent in the time-domain characterization of the respective OPAs (Fig. 2). The ultrashort pump pulse impulsively creates the transient oscillating grating of the lattice vibrations that lags behind it; within the phonon coherence lifetime τ_{coher} , the NLO crystal becomes transiently "periodically-poled" with the effective wave-vector $k_{vibr} = 2\pi n/c T_{vibr}$, where T_{vibr} is the phonon oscillation period. Such modulation leads to the temporal evolution of the parametric gain in KTP (red in Fig. 2) and KTA, while BBO shows practically the instantaneous response of $\chi^{(2)}$ -nonlinearity (Fig. 2, blue).

We will present additional data supporting the photon-phonon coupling mechanism in KTP- and KTA-OPA and will discuss the implications of the observed phenomena for the operation of NLO frequency conversion devices, as well as for ultrashort terahertz- (THz-) pulse generation in titanyl crystals.

4. References

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