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Peer Review

Review of: "Terahertz-Induced Second-Harmonic Generation in Quantum Paraelectrics: Hot-Phonon Effect"

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This manuscript presents a new theoretical explanation of the slow-relaxing and non-oscillating SHG signal observed by various authors after THz pumping of the soft mode in the incipient ferroelectric KTaO3. The authors come to the reasonable conclusion that the THz-induced non-oscillatory SHG signal is a consequence of the induced nonequilibrium hot-phonon effect and is not associated with the breaking of the inversion center typical for the ferroelectric phase. This is a very nice paper, but I need to clarify a few things before accepting it and suggest some changes:

1. Fig. 1 shows a single pulse over time, but the spectrum of the pumping pulse is important for explaining the temperature dependence of SHG. The pulse usually has a maximum intensity at 0.7 or 0.9 THz. The observed THz-induced SHG signal has a maximum at 20 K, but the soft mode should have a frequency of 0.6 THz at this temperature. Optimal pumping of the soft mode should therefore occur sometime between 20 and 30 K. Please add the frequency dependence of the THz pulse to Fig. 1.
2. Fig. 2c shows the obtained temperature dependence of the soft mode frequency and its damping, but these do not match the published experimental values. References 22 and 24 deal not with KTaO3 but with SrTiO3, where the soft mode frequencies are different. So they should be removed on page 7. Ref. 23 shows the soft mode frequencies in KTaO3 from Raman scattering in an electric field (the soft mode is normally Raman inactive), and there its frequency could be affected by the field. More accurate soft-mode values have been published based on hyper-Raman spectra (H. Vogt. Refined treatment of the model of linearly coupled anharmonic oscillators and its application to the temperature dependence of the zone-center soft-mode frequencies of KTaO3 and SrTiO3. Phys Rev B 51, 8046-8059 (1995)) and IR spectra (S. Glinšek, et al. Lattice dynamics

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and broad-band dielectric properties of the KTaO3 ceramics. J Appl Phys 111 (2012)). Their soft mode values are in agreement but lower than those in your Fig. 2c. If the published soft mode frequencies are well and if the pumping pulse has a maximum intensity at 0.9 THz, a maximum SHG signal above 30 K would be observed. Can you comment on this?

3. The low-frequency peak in Fig. 2b, visible at 0.4 THz and labeled as a collective vector mode, is not satisfactorily explained. What is it? Is it the so-called central mode visible near ferroelectric transitions? The central peak has never been observed in incipient ferroelectrics, only in displacive ferroelectrics.
4. Page 10: You write that "the THz-induced SHG response in quantum paraelectrics is a near-equilibrium response." How can I understand this if it is time-dependent?
5. I highly appreciate the SHG measurements on ferroelectric KTaO3, showing a completely different THz-induced SHG response. This proves that the previous explanation of the non-oscillating SHG signal comes not from polar nanoclusters but from the hot-phonon effect. However, there is no mention at all of how a ferroelectric crystal differs from a paralelectric one. How many impurities or dopants does it have, and which ones? What is its Tc when the measurement is made at 77 K? On page 12, you cite reference 1, but this refers to SrTiO3 and not ferroelectric KTaO3. It is therefore an incorrect reference. I request a more detailed description of the ferroelectric sample, at least giving its Tc. Furthermore, Fig. 3 lacks a description of the x-axis (Delay time in ps).

If the authors answer my questions satisfactorily and correct some deficiencies, I will agree to accept the paper for publication.

Declarations

Potential competing interests: No potential competing interests to declare.