LETTER

Terahertz-wave generation from surface phonons at forbidden frequencies of lithium niobate

Jun-ichi Shikata^{1, a)}, Seigo Ohno^{2, 3}, and Hiroaki Minamide³

Abstract This paper describes terahertz (THz)-wave generation within forbidden bands in polar crystals, focusing on the A_1 phonon modes in lithium niobate. This material exhibits two negative-permittivity frequency ranges at 7.4–12.7 THz and 18.8–25.6 THz for the lowest and highest A_1 modes, respectively. Exploiting the finite-difference time-domain simulations, we demonstrate that both the surface phonon modes can be radiative with a structured grating. Fourier analyses of the radiative fields reveal the relevant peaks in the spectrum as well as the dispersion relations. Our results provide a novel method for coherent THz-wave sources at unexplored THz frequencies.

Keywords: terahertz-wave generation, surface phonon, lithium niobate, negative permittivity, A_1 mode

Classification: Electromagnetic theory

1. Introduction

Recently, a variety of compact coherent terahertz (THz)-wave sources have been developed based on photonics and ultrafast electronics. These sources include THz-wave generators using nonlinear optics [1, 2, 3, 4, 5, 6, 7, 8, 9], photoconductive antennas [10, 11, 12], THz quantum cascade lasers [13, 14, 15], TUNNETT diodes [16, 17], uni-traveling-carrier photodiodes [18, 19, 20], resonant tunneling diodes [21, 22, 23], and plasmonic THz emitters [24, 25]. However, the interaction of optical phonons with photons frequently restricts the THz waves available, because it results in forbidden bands [26], where the THz waves are unable to propagate over a long distance in bulk media. In THz-wave generation by stimulated polariton scattering in LiNbO₃ (LN) [1, 2, 3, 4, 5, 6], the tuning range is limited to 0.7–3.0 THz due to the substantial absorption loss of the THz waves interacting with the lowest A_1 phonon mode (7.4 THz).

A possible way to generate THz waves at forbidden frequencies in bulk is to couple out the excited surface phonons [27]. In fact, thermally excited surface phonons on SiC gratings have been shown to exhibit infrared emission [28], and the spectra of this emission cover a wide

frequency range (more than several THz) in the forbidden band. Since surface waves have long-range coherence [29], coherent emissions occur even with incoherent excitations.

In this paper, we explore THz-wave generation from surface phonons, focusing on the A_1 modes in LN crystals. We discover surface phonon modes arising from the lowest and highest A_1 modes. Finite-difference time-domain simulations reveal the THz-wave radiation from a grating surface. The emission spectra as well as the dispersion relations are found using Fourier analyses.

2. Theoretical background

In the bulk of a polar crystal, the interaction between transverse optical (TO) phonons and transverse photons results in coupled modes, called polaritons [26]. With respect to a simple single-oscillator (two-atom) model in lossless media, the polariton wavevector (i.e., dispersion relation) $k(\omega) = (\omega/c)\sqrt{\varepsilon(\omega)}$ is determined using the following dielectric function:

$$\varepsilon(\omega) = \varepsilon_\infty + \frac{S\omega_T^2}{\omega_T^2 - \omega^2} = \frac{\varepsilon_\infty(\omega_L^2 - \omega^2)}{\omega_T^2 - \omega^2} \quad (1)$$

where ε_∞ is the permittivity for high frequencies, S is the oscillator strength, ω_T is the frequency of the TO phonon, and ω_L is the frequency of the longitudinal optical phonon. As $\varepsilon(\omega) < 0$ at $\omega_T < \omega < \omega_L$, the wavevector is purely imaginary ($k = i|k|$); thus, the amplitude of the electromagnetic wave decays as $\exp(-|k|x)$ according to its distance from the surface (forbidden band). The zero point of Eq. (1) [$\varepsilon(0) = \varepsilon_0$] provides the well-known Lyddane–Sachs–Teller (LST) relation as follows:

$$\frac{\omega_L}{\omega_T} = \sqrt{\frac{\varepsilon_0}{\varepsilon_\infty}} \quad (2)$$

Although the electromagnetic waves at the forbidden frequencies cannot propagate for long in bulk, surface waves (known as surface phonons [27]) can freely propagate along the surface with transverse magnetic (TM) polarization. On a flat surface between the crystal and air, the electromagnetic boundary conditions yield the following surface phonon wavevector:

$$k_{SP}(\omega) = \frac{\omega}{c} \sqrt{\frac{\varepsilon(\omega)}{\varepsilon(\omega) + 1}} \quad (3)$$

The dispersion curve of the surface phonon emerges from ω_T [$\varepsilon(\omega) = -\infty$], whereas the upper limit ω_s is specified by

¹ College of Engineering, Nihon University, Koriyama 963-8642, Japan

² Graduate School of Science, Tohoku University, Aoba-ku, Sendai 980-8578, Japan

³ RIKEN Center for Advanced Photonics (RAP), RIKEN, Aoba-ku, Sendai 980-0845, Japan

^{a)} shikata.junnichi@nihon-u.ac.jp

$$\omega_s = \omega_T \sqrt{\frac{\varepsilon_0 + 1}{\varepsilon_\infty + 1}} \quad (4)$$

from the condition $k_{SP} = \infty$ [$\varepsilon(\omega) = -1$] in Eq. (3).

It is worth noting that the dispersion curve is below the light line $k = \omega/c$ because $\varepsilon(\omega)/(\varepsilon(\omega) + 1) > 1$ in Eq. (3) at $-\infty < \varepsilon(\omega) < -1$. Since the energy and momentum conservations do not hold simultaneously, THz-wave emissions do not occur from surface phonons on a flat surface. However, if a Bragg grating (Fig. 1) is structured on the surface, the radiative coupling becomes possible in accordance with the condition of momentum conservation:

$$k_{SP}(\omega) = \frac{\omega}{c} \sin \theta + m \frac{2\pi}{\Lambda} \quad (5)$$

where θ is the radiation angle, m is the diffraction order (an integer), and Λ is the grating period.

Fig. 1 Schematic diagram of THz-wave generation from surface phonons on a grating surface of LN.

3. Dispersion relations of A_1 modes in LN

3.1 Bulk polariton modes

Since an LN crystal has four A_1 modes, the n th mode can be designated as $A_1^{(n)}$ in Table I. We used the multi-oscillator model of the dielectric function, including the non-negligible damping terms, described as

$$\varepsilon(\omega) = \varepsilon_\infty + \sum_n \frac{S_n \omega_{Tn}^2}{\omega_{Tn}^2 - \omega^2 - i \Gamma_n \omega} \quad (6)$$

where Γ_n is the damping coefficient.

Table I Optical parameters of A_1 modes in LN [30].

mode	ω_T (cm ⁻¹)	S	Γ (cm ⁻¹)	ε_∞	ω_L (cm ⁻¹)
$A_1^{(1)}$	248	16.0	21	4.6	273
$A_1^{(2)}$	274	1.0	14		306
$A_1^{(3)}$	307	0.16	25		423
$A_1^{(4)}$	628	2.55	34		869

Fig. 2 shows the dielectric properties calculated from Eq. (6). The strong resonances ($A_1^{(1)}$, $A_1^{(4)}$) almost determine the dispersive and loss characteristics of the polaritons in Figs. 2(a) and 2(b), whereas the weak resonances ($A_1^{(2)}$, $A_1^{(3)}$) contribute less. The resulting negative-permittivity regions emerge at 7.4–12.7 THz and 18.8–25.6 THz [the upper part of Fig. 2(a)]. Accordingly, we focused on the lowest and highest A_1 modes to study the surface phonon bands and their emissions, as shown below.

3.2 Surface phonon modes on a flat surface

To solve Eq. (3) for the lossy case [Eq. (6)], we employed the complex frequency scheme [31], where the frequency

Fig. 2 Dielectric properties associated with A_1 modes in bulk LN. (a) Complex dielectric functions. (b) Dispersion relations of the polaritons. The frequency is assumed to be real. The dashed lines denote the TO phonon frequencies (ω_T). The absorption coefficient is defined by $\alpha = 2 \text{Im } k$.

was assumed to be complex ($\omega = \omega' - i\omega''$), such that the amplitude decay was expressed as $\exp(-\omega''t)$. We modified Eq. (3) to the following system of nonlinear equations:

$$k_{SP} = \text{Re} \left[\frac{\omega' - i\omega''}{c} \sqrt{\frac{\varepsilon(\omega' - i\omega'')}{\varepsilon(\omega' - i\omega'') + 1}} \right] \quad (7)$$

$$0 = \text{Im} \left[\frac{\omega' - i\omega''}{c} \sqrt{\frac{\varepsilon(\omega' - i\omega'')}{\varepsilon(\omega' - i\omega'') + 1}} \right] \quad (8)$$

Eq. (7) as well as Eqs. (6) and (8) were numerically solved for each fixed real wavevector k_{SP} .

Fig. 3 illustrates the resulting dispersion relations of the A_1 surface phonon modes. In Fig. 3(a), the lower branch emerges from $\omega_T = 7.4$ THz, asymptotically approaching the upper limit $\omega_s = 14.6$ THz. Moreover, in Fig. 3(b), the lower branch extends from $\omega_T = 18.8$ THz to $\omega_s = 22.7$ THz. Both the upper limits (ω_s) agree well with those resulting from Eq. (4). In this way, we found that both the lower branches in Figs. 3(a) and 3(b) were of surface phonons that were relevant to the $A_1^{(1)}$ and $A_1^{(4)}$ modes, respectively.

Fig. 3 Dispersion relations of surface phonon modes on a flat surface. (a) $A_1^{(1)}$ mode, $\omega_s = 14.6$ THz. (b) $A_1^{(4)}$ mode, $\omega_s = 22.7$ THz. The surface wavevector k_{SP} is assumed to be real.

Note that the $A_1^{(4)}$ surface phonon band (18.8–22.7 THz) is within the negative-permittivity region (18.8–25.6 THz) [Fig. 2(a)]; however, $\omega_s = 14.6$ THz of the $A_1^{(1)}$ mode exceeds the upper boundary of the forbidden band (12.7 THz). This anomaly results from the large oscillator strength S (i.e., ε_0) of the $A_1^{(1)}$ mode, which results in the breakdown of the LST relation [32]. Indeed, for the $A_1^{(1)}$ mode, the LST relation (2) is broken ($\omega_L/\omega_T = 1.1$, $\sqrt{\varepsilon_0/\varepsilon_\infty} = 2.1$), whereas the relation holds well for the $A_1^{(4)}$ mode ($\omega_L/\omega_T = 1.4$, $\sqrt{\varepsilon_0/\varepsilon_\infty} = 1.2$).

3.3 Surface phonon modes on a grating

Fig. 4 presents reduced band diagrams of the $A_1^{(1)}$ and $A_1^{(4)}$ surface phonons on a grating, which is based on a simple gapless model. To shift most of the dispersion curves (Fig. 3) within the light cone according to Eq. (5), we chose a grating period Λ such that the zone edge π/Λ was located close to $k_{\min}$ (the surface phonon wavevector at ω_T). In Fig. 4, the folded dispersion curves extend over the frequency ranges of 7.7–14.6 THz ($\Lambda = 19.4 \mu\text{m}$) and 19.1–22.7 THz ($\Lambda = 7.8 \mu\text{m}$) relevant to the $A_1^{(1)}$ and $A_1^{(4)}$ modes, respectively. Their properties of surface phonon emissions were discussed based on electromagnetic simulations, as shown below.

Fig. 4 Dispersion relations of surface phonon modes on a grating. (a) $A_1^{(1)}$ mode, grating period $\Lambda = 19.4 \mu\text{m}$. (b) $A_1^{(4)}$ mode, $\Lambda = 7.8 \mu\text{m}$.

4. THz-wave emissions from surface phonons

We now analyze the THz-wave emissions from surface phonons on a grating. The electromagnetic simulations were conducted using a finite-difference time-domain method (Fujitsu, Poynting for Optics). An LN grating on the x -plane (Fig. 1) was assumed because the polarization of the A_1 mode is along the c -axis (i.e., the z -axis). Also supposing an infinite length along the y -axis, we performed two-dimensional simulations in the x - z plane. To excite surface phonons, we used a point source [33] on the flat surface [orange filled circle in Fig. 5(a)]. The propagating surface waves were diffracted at a structured grating (period $\Lambda = 19.4 \mu\text{m}$, groove depth $t = 3 \mu\text{m}$), which might yield surface phonon emissions.

The resulting electric field distributions are shown in Figs. 5(b)–5(d). To illustrate both the surface waves and their emissions with TM polarizations, we extracted the amplitude of the E_z -fields, although the radiative fields had both the E_x - and E_z -components. As expected, the emissions were largely frequency dependent. Below the TO phonon frequency (7.4 THz), the emission as well as the surface wave were suppressed [Fig. 5(b)]; however, the emission was evident at 9.5 THz [Fig. 5(c)]. Surprisingly, THz-wave emission was also found at 19 THz [Fig. 5(d)], despite the grating period having been designed for the $A_1^{(1)}$ mode. These emission frequencies were both in the negative-permittivity regions relevant to the $A_1^{(1)}$ and $A_1^{(4)}$ modes. Note that the radiation angles were different (i.e., dispersive), similar to those in Ref. [28].

To confirm the THz-wave emissions arising from the A_1 surface phonons, we analyzed the frequencies and wavevectors of the E_z -fields, monitored at a detection area located in the air [green line in Fig. 5(a)]. Fourier analyses of

Fig. 5 (a) Schematic of the studied structure. (b) E_z distribution at 7.0 THz, (c) 9.5 THz, (d) 19 THz. The grating period $\Lambda = 19.4 \mu\text{m}$.

the temporal E_z -fields, normalized by the incident fields at a flat portion, yielded the emission spectra. To eliminate contributions from the surface waves as well as the direct radiations from the point source [see Figs. 5(b)–5(d)], we subtracted the spectrum for the flat surface from that for the grating surface, as shown in Fig. 6. In the 7.4–12.0 THz range, radiation relevant to the $A_1^{(1)}$ mode was prominent. Furthermore, the second peak at approximately 19 THz, attributed to the $A_1^{(4)}$ mode, was also noteworthy. The negative portions ($< 7.3 \text{ THz}$, $> 20.6 \text{ THz}$) might have resulted from material absorption or reflection at the grating that was under investigation.

Fig. 6 THz-wave spectrum emitted from the A_1 surface phonons on a grating ($\Lambda = 19.4 \mu\text{m}$).

Furthermore, the spatial Fourier transform of the E_z -fields (k_z -components) at each frequency led to the dispersion relations, as shown in Fig. 7. The bright portion at approximately 9 THz deviated from the light line and was in excellent agreement with the $A_1^{(1)}$ surface phonon branch, shifted by the first-order diffraction according to Eq. (5). Despite the limited frequency range, the second bright portion at approximately 19 THz ($k_{SP}/2\pi \sim 120 \text{ cm}^{-1}$) also agreed well with the first-order diffracted surface phonon branch of the $A_1^{(4)}$ mode. These behaviors were similar to the plasmonic resonance on a metal grating [34, 35]. Therefore, we decided that the present THz-wave radiations were those supported

Fig. 7 Dispersion relations of the A_1 surface-phonon emissions from a structured grating ($\Lambda = 19.4 \mu\text{m}$), depicted in the vicinity of the first Brillouin zone.

by the A_1 surface phonons in LN. A single periodicity of an LN grating opens the emission channels at approximately 9 and 19 THz. We did not find significant emissions from the $A_1^{(4)}$ surface phonons on an LN grating of $\Lambda = 7.8 \mu\text{m}$. However, we consider that emissions might be possible if the structural parameters (especially, the groove depths) were optimized. Although we have herein studied the linear optical phenomena on THz electromagnetic waves, the results of our research might also hold for nonlinear-optic THz-wave generation [1, 2, 3, 4, 5, 6, 7, 8, 9]. Laser-induced nonlinear polarizations are equivalent to the coherent excitation sources exploited here. Therefore, surface phonon emission might present a potential path to novel THz-wave sources at the forbidden frequencies in solids, including nonlinear optical crystals and semiconductors.

5. Conclusion

We have studied THz-wave generation in the forbidden bands of LN crystals. It has been shown that the surface phonons can be excited in relation to the lowest and highest A_1 modes, which can radiate widely tunable THz waves with a grating of single periodicity. These findings provide a basis for realizing coherent THz-wave sources at unexplored forbidden frequencies.

Acknowledgments

This work is partly supported by JSPS KAKENHI Grant Numbers JP19K05286 and JP18K04967 as well as the cooperative research aid from RIKEN.

References

- [1] K. Kawase, *et al.*: “Coherent tunable THz-wave generation from LiNbO₃ with monolithic grating coupler,” *Appl. Phys. Lett.* **68** (1996) 2483 (DOI: 10.1063/1.115828).
- [2] J. Shikata, *et al.*: “Tunable terahertz-wave parametric oscillators using LiNbO₃ and MgO: LiNbO₃ crystals,” *IEEE Trans. Microw. Theory Techn.* **48** (2000) 653 (DOI: 10.1109/22.841956).
- [3] K. Kawase, *et al.*: “Terahertz wave parametric source,” *J. Phys. D* **35** (2002) R1 (DOI: 10.1088/0022-3727/35/3/201).
- [4] S. Hayashi, *et al.*: “Ultrabright continuously tunable terahertz-wave generation at room temperature,” *Sci. Rep.* **4** (2014) 5045 (DOI: 10.1038/srep05045).
- [5] Y. Takida, *et al.*: “Terahertz-wave parametric gain of stimulated polariton scattering,” *Phys. Rev. A* **93** (2016) 043836 (DOI: 10.1103/physrev.93.043836).
- [6] K. Nawata, *et al.*: “Tunable backward terahertz-wave parametric oscillation,” *Sci. Rep.* **9** (2019) 726 (DOI: 10.1038/s41598-018-37068-7).
- [7] T. Tanabe, *et al.*: “Frequency-tunable terahertz wave generation via excitation of phonon-polaritons in GaP,” *J. Phys. D* **36** (2003) 953 (DOI: 10.1088/0022-3727/36/8/302).
- [8] T. Taniuchi, *et al.*: “Continuously tunable THz and far-infrared wave generation from DAST crystal,” *Electron. Lett.* **40** (2004) 60 (DOI: 10.1049/el:20040380).
- [9] K. Miyamoto, *et al.*: “Widely tunable terahertz-wave generation using an N-benzyl-2-methyl-4-nitroaniline crystal,” *Opt. Lett.* **33** (2008) 252 (DOI: 10.1364/ol.33.000252).
- [10] E.B. Brown, *et al.*: “Photomixing up to 3.8 THz in low temperature-grown GaAs,” *Appl. Phys. Lett.* **66** (1995) 285 (DOI: 10.1063/1.113519).
- [11] K.A. McIntosh, *et al.*: “Terahertz measurements of resonant planar antennas coupled to low-temperature-grown GaAs photomixers,” *Appl. Phys. Lett.* **69** (1996) 3632 (DOI: 10.1063/1.117006).
- [12] I.S. Gregory, *et al.*: “Continuous-wave terahertz system with a 60 dB dynamic range,” *Appl. Phys. Lett.* **86** (2005) 204104 (DOI: 10.1063/1.1935032).
- [13] R. Köhler, *et al.*: “Terahertz semiconductor-heterostructure Laser,” *Nature* **417** (2002) 156 (DOI: 10.1038/417156a).
- [14] S. Kumar: “Recent progress in terahertz quantum cascade Lasers,” *IEEE J. Sel. Topics Quantum Electron.* **17** (2011) 38 (DOI: 10.1109/jstqe.2010.2049735).
- [15] T.-T. Lin and H. Hirayama: “Improvement of operation temperature in GaAs/AlGaAs THz-QCLs by utilizing high Al composition barrier,” *Phys. Status Solidi C* **10** (2013) 1430 (DOI: 10.1002/pssc.201300216).
- [16] J. Nishizawa, *et al.*: “GaAs TUNNETT diodes oscillating at 430-655 GHz in CW fundamental mode,” *IEEE Microw. Wireless Compon. Lett.* **15** (2005) 597 (DOI: 10.1109/lmwc.2005.855381).
- [17] J. Nishizawa, *et al.*: “706-GHz GaAs CW fundamental-mode TUNNETT diodes fabricated with molecular layer epitaxy,” *Phys. Status Solidi C* **5** (2008) 2802 (DOI: 10.1002/pssc.200779256).
- [18] T. Ishibashi, *et al.*: “High-speed response of uni-traveling-carrier photodiodes,” *Jpn. J. Appl. Phys.* **36** (1997) 6263 (DOI: 10.1143/jjap.36.6263).
- [19] T. Ishibashi, *et al.*: “Unitraveling-carrier photodiodes for terahertz applications,” *IEEE J. Sel. Topics Quantum Electron.* **20** (2014) 3804210 (DOI: 10.1109/jstqe.2014.2336537).
- [20] T. Kurokawa, *et al.*: “Over 300 GHz bandwidth UTC-PD module with 600 GHz band rectangular waveguide output,” *Electron. Lett.* **54** (2018) 705 (DOI: 10.1049/el.2018.0932).
- [21] M. Asada, *et al.*: “Resonant tunneling diodes for sub-terahertz and terahertz oscillators,” *Jpn. J. Appl. Phys.* **47** (2008) 4375 (DOI: 10.1143/jjap.47.4375).
- [22] S. Suzuki, *et al.*: “Fundamental oscillation of resonant tunneling diodes above 1 THz at room temperature,” *Appl. Phys. Lett.* **97** (2010) 242102 (DOI: 10.1063/1.3525834).
- [23] R. Izumi, *et al.*: “Resonant-tunneling-diode terahertz oscillator with a cylindrical cavity for high-frequency oscillation,” *AIP Adv.* **9** (2019) 085020 (DOI: 10.1063/1.5114963).
- [24] Y.M. Mezziani, *et al.*: “Room temperature generation of terahertz radiation from a grating-bicoupled plasmon-resonant emitter: Size effect,” *Appl. Phys. Lett.* **90** (2007) 061105 (DOI: 10.1063/1.2459879).
- [25] A.A. Dubinov, *et al.*: “Terahertz plasmon-emitting graphene-channel transistor,” *Opto-Electron. Rev.* **27** (2019) 345 (DOI: 10.1016/j.opelre.2019.11.003).
- [26] C. Kittel: *Introduction to Solid State Physics* (Wiley, New York, 2005) 8th ed. 410.
- [27] Y.R. Shen: *The Principles of Nonlinear Optics* (Wiley, New York, 1984) 481.
- [28] J.-J. Greffet, *et al.*: “Coherent emission of light by thermal sources,” *Nature* **416** (2002) 61 (DOI: 10.1038/416061a).
- [29] R. Carminati and J.-J. Greffet: “Near-field effects in spatial coherence of thermal sources,” *Phys. Rev. Lett.* **82** (1999) 1660 (DOI: 10.1103/PhysRevLett.82.1660).

- [10.1103/physrevlett.82.1660](https://doi.org/10.1103/physrevlett.82.1660)).
- [30] A.S. Baker, Jr. and R. Loudon: “Dielectric properties and optical phonons in LiNbO₃,” Phys. Rev. **158** (1967) 433 (DOI: [10.1103/physrev.158.433](https://doi.org/10.1103/physrev.158.433)).
 - [31] J. Le Gall, *et al.*: “Experimental and theoretical study of reflection and coherent thermal emission by a SiC grating supporting a surface-phonon polariton,” Phys. Rev. B **55** (1997) 10105 (DOI: [10.1103/physrevb.55.10105](https://doi.org/10.1103/physrevb.55.10105)).
 - [32] Y. Ichikawa, *et al.*: “Direct observation of the soft-mode dispersion in the incipient ferroelectric KTaO₃,” Phys. Rev. B **71** (2005) 092196 (DOI: [10.1103/physrevb.71.092196](https://doi.org/10.1103/physrevb.71.092196)).
 - [33] B. Hecht, *et al.*: “Local excitation, scattering, and interference of surface plasmons,” Phys. Rev. Lett. **77** (1996) 1889 (DOI: [10.1103/physrevlett.77.1889](https://doi.org/10.1103/physrevlett.77.1889)).
 - [34] S.A. Maier: “Plasmonics: metal nanostructures for sub-wavelength photonic devices,” IEEE J. Sel. Topics Quantum Electron. **12** (2006) 1214 (DOI: [10.1109/jstqe.2006.879582](https://doi.org/10.1109/jstqe.2006.879582)).
 - [35] S.A. Maier: *Plasmonics: Fundamentals and Applications* (Springer Verlag, 2007) 30.