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

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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\cite{1, 2, 3, 4, 5, 6, 7, 8, 9}, photoconductive antennas\cite{10, 11, 12}, THz quantum cascade lasers\cite{13, 14, 15}, TUNNETT diodes\cite{16, 17}, untraveling-carrier photodiodes\cite{18, 19, 20}, resonant tunneling diodes\cite{21, 22, 23}, and plasmonic THz emitters\cite{24, 25}. However, the interaction of optical phonons with photons in the forbidden bands of polar crystals, focusing on the $A_1$ phonon 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\cite{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:

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

where $\varepsilon_\infty$ is the permittivity for high frequencies, $S$ is the oscillator strength, $\omega_T$ is the frequency of the TO phonon, and $\omega_L$ is the frequency of the longitudinal optical phonon. As $\varepsilon(\omega) < 0$ at $\omega_L < \omega < \omega_T$, 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:

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

Although the electromagnetic waves at the forbidden frequencies cannot propagate for long in bulk, surface waves (known as surface phonons\cite{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:

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

The dispersion curve of the surface phonon emerges from $\omega_T [\varepsilon(\omega) = -\infty]$, whereas the upper limit $\omega_L$ is specified by
To solve Eq. (3) for the lossy case [Eq. (6)], we employed

$$\omega_s = \frac{\omega}{c} \sqrt{\frac{\varepsilon_0 + 1}{\varepsilon_\infty + 1}}$$  \hspace{1cm} (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:

$$k_{SP}(\omega) = \frac{\omega}{c} \sin \theta + \frac{2\pi}{\Lambda}$$ \hspace{1cm} (5)

where \(\theta\) is the radiation angle, \(m\) is the diffraction order (an integer), and \(\Lambda\) is the grating period.

![Fig. 1 Schematic diagram of THz-wave generation from surface phonons on a grating surface of LN.](image)

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

$$\varepsilon(\omega) = \varepsilon_\infty + \sum_n \frac{\varepsilon_n \omega_n^2}{\omega_n^2 - \omega^2 - i \Gamma_n \omega}$$ \hspace{1cm} (6)

where \(\Gamma_n\) is the damping coefficient.

![Fig. 2 Dielectric properties associated with \(A_1\) modes in bulk LN. (a) Complex dielectric functions. (b) Dispersion relations of the polaritons.](image)

Fig. 2 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 (\(\omega_s\)) 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.](image)

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 of the LST relation \[32\]. Indeed, for the \(A_1^{(1)}\) mode, the LST relation (2) is broken \((\omega_T/\omega_T = 1.1, \sqrt{\varepsilon_0}/\varepsilon_\infty = 2.1, \sqrt{\varepsilon_0}/\varepsilon_\infty = 1.2)\), whereas the relation holds well for the \(A_1^{(4)}\) mode \((\omega_T/\omega_T = 1.4, \sqrt{\varepsilon_0}/\varepsilon_\infty = 1.2)\).
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 $\Lambda$ such that the zone edge $\pi/\Lambda$ was located close to $k_{\text{min}}$ (the surface phonon wavevector at $\omega \gamma$). In Fig. 4, the folded dispersion curves extend over the frequency ranges of 7.7–14.6 THz ($\Lambda = 19.4 \, \mu m$) and 19.1–22.7 THz ($\Lambda = 7.8 \, \mu m$) 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 $\Lambda = 19.4 \, \mu m$. (b) $A_1^{(4)}$ mode, $\Lambda = 7.8 \, \mu m$.](image)

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 $\Lambda = 19.4 \, \mu m$, groove depth $t = 3 \, \mu m$), 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, 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. 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 $\Lambda = 19.4 \, \mu m$.](image)

![Fig. 6 THz-wave spectrum emitted from the $A_1$ surface phonons on a grating ($\Lambda = 19.4 \, \mu m$).](image)

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 ($k_1P/2\pi \sim 120 \, \text{cm}^{-1}$) 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...
by the \( \alpha_1 \) 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 \( \alpha_1 \) surface phonons on an LN grating of \( \Lambda = 7.8 \) \( \mu \)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 \( \alpha_1 \) 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.

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