Terahertz response of acoustically driven optical phonons

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(Received 8 April 2010; revised manuscript received 24 May 2010; published 17 June 2010)

The manipulation of transverse-optical (TO)-phonon polaritons and the associated terahertz (THz) light field by means of an ultrasound acoustic wave is proposed and illustrated by calculating the TO-phonon-mediated THz response of acoustically pumped CuCl and TlCl crystals. We show the high-contrast acoustically induced change in the THz reflectivity and multiple THz Bragg replicas, which are associated with the far-infrared-active TO-phonon resonance driven by the ultrasonic wave. The effect, which stems from phonon anharmonicity and deals with the resonantly enhanced acousto-optical susceptibilities, refers to an operating acoustic intensity \( I_{ac} \approx 1–100 \text{ kW/cm}^2 \) and frequency \( \nu_{ac} \approx 0.1–1 \text{ GHz} \). Due to the anomalously small interaction length between the acoustic and optical fields, possible applications of the effect are in THz spectroscopy and THz acousto-optic devices.

DOI: 10.1103/PhysRevB.81.245208

PACS number(s): 71.36.+c, 43.35.+d

I. INTRODUCTION

Since the pioneering work by Kun Huang,\(^1\) the physics of infrared polaritons associated with transverse-optical (TO) phonons has emerged as a well-established discipline. Recently, room-temperature polaritons was implemented for processing and coherent control of the terahertz (THz) light field.\(^2,3\) In addition to conventional infrared spectroscopy and continuous-wave (cw) Raman-scattering experiments, broadband THz time-domain spectroscopy has been developed and successfully applied to characterize picosecond (ps) and sub-ps dynamics of infrared-active TO phonons\(^4\) and to visualize the phonon dispersion associated with these vibrational modes.\(^5\) Furthermore, the THz polariton spectra allow the study of unusual lattice dynamics, e.g., in ferroelectrics\(^6\) and negative thermal-expansion compounds.\(^7,8\) However, the spectrally resolved control of the electromagnetic field associated with far-infrared polaritons still remains a very challenging task of THz optics.

In this paper, we propose an acoustic modulation of TO-phonon polaritons in order to drastically change their optical response in the THz band. Phonon anharmonicity, which can be large in some dielectric and semiconductor materials and particularly strong for soft TO-phonon modes, leads to the coupling between a coherent (pumping) acoustic wave (AW) and infrared-active TO phonons. In this case one deals with an acoustically induced Autler-Townes effect, which gives rise to spectral gaps \( \Delta_{\nu} \) in the THz polariton spectrum of AW-driven TO phonons, and is akin to the acoustical Stark effect for optically allowed excitons.\(^9–14\) The AW-induced gaps \( \Delta_{\nu} \), which open up in the polariton spectrum and develop with increasing acoustic intensity \( I_{ac} \), are due to the \( N \)th-order resonant acoustic-phonon transitions within the polariton dispersion branches. These forbidden-energy gaps strongly modify the optical response of TO-phonon polaritons and make possible the effective AW-controlled manipulation of the THz field.

The spectrally resolved AW control of the THz field propagation can also be interpreted in terms of Bragg diffraction of far-infrared polaritons by the pumping AW: we analyze the use of phonon anharmonicity to create an acoustically induced Bragg grating. In this case the contrast of the AW grating is dictated by the efficiency of the scattering channel “TO phonon \( \leftrightarrow \) acoustic phonon (two acoustic phonons) \( \leftrightarrow \) TO phonon” for cubic (quartic) phonon anharmonicity. Thus the scattering of THz light is mediated and strongly enhanced by the TO-phonon resonance. This results in large acousto-optical nonlinearities and therefore in an anomalously short interaction length \( \ell_{int} \) between the acoustic and optical fields needed for the formation of the Bragg replicas.

Possible applications of governing far-infrared polaritons by using an ultrasonic acoustic field include frequency-tunable THz detectors and filters, Bragg switches, and frequency converters. The size of these devices would be much smaller than conventional acousto-optic equivalents, due to the rather small operating length scale, \( \ell_{int} \approx 100 \mu\text{m} \).

In Sec. II, we discuss a model for far-infrared polaritons parametrically driven by the ultrasonic acoustic field, derive the macroscopic equations for resonantly coupled THz light field and polarization associated with TO phonons, and finally calculate the total Bragg reflectivity of TO-phonon polaritons. In Sec. III, the AW-induced response of THz polaritons in bulk CuCl (cubic phonon anharmonicity) and TlCl (quartic phonon anharmonicity) is modeled to illustrate the high efficiency of the effect and its potential for device applications. In Sec. IV, we discuss the results. A short summary of the work is given in Sec. V.

II. MODEL

The Hamiltonian of far-infrared polaritons coherently driven by a cw bulk acoustic wave \( \{ \mathbf{k}, \omega_{ac}(k) \} \) is given by

\[
H = H_0 + \hbar \sum_p [2 \tilde{m}_\nu \tilde{J}_p \tilde{b}_p^\dagger \tilde{b}_p + (m_\nu^4 \tilde{J}_p \tilde{J}_\nu^4 e^{2i\omega_{ac} t} \tilde{b}_p^\dagger \tilde{b}_{-p} + \text{H.c.})] \\
+ (m_\nu^4 \tilde{J}_\nu^4 \tilde{b}_p^\dagger e^{-2i\omega_{ac} t} \tilde{b}_{-p} + \text{H.c.})
\]

(1)

with \( H_0 \) the conventional polariton Hamiltonian of infrared-active TO phonons,\(^15\) \( \tilde{b}_p \) the TO-phonon operator, \( \omega_{ac} = 2\pi \nu_{ac} = \nu_k, \nu_\nu \) the sound velocity, and \( m_\nu \) (\( \tilde{m}_\nu \)) the matrix element associated with cubic (quartic) anharmonic-
ity. The macroscopic equations, which describe the control of THz polaritons by applying the acoustic field of an arbitrary profile, \( I_{ac} = I_{ac}(r, t) \), are

\[
\frac{\partial^2}{\partial t^2} E(r, t) = -\frac{4\pi}{c^2} \frac{\partial^2}{\partial t^2} P(r, t),
\]

\[
\begin{align*}
\left[ \frac{\partial^2}{\partial t^2} + 2 \gamma_{TO} \frac{\partial}{\partial t} + \Omega_4^2 + 4 \Omega_{TO} \tilde{m}_4^*(r, t) \\
+ 4 \Omega_{TO} m_4 I_{ac}(r, t) \cos(\omega_{ac} t - kr) \\
+ 4 \Omega_{TO} m_4 I_{ac}(r, t) \cos(2\omega_{ac} t - 2kr) \right] P(r, t) = \frac{\varepsilon_b}{4\pi} \Omega_k^2 E(r, t),
\end{align*}
\]

where \( E \) and \( P \) stand for the field light and TO-phonon polarization, respectively, \( \Omega_{TO} \) is the TO-phonon frequency, \( \Omega_k \) is the polaron Rabi frequency, \( \varepsilon_b \) is the background dielectric constant in the infrared, and \( \gamma_{TO} \) is the damping rate of TO phonons, mainly due to their decay into short-wavelength acoustic phonons. Equations (2) and (3) refer to simple cubic lattices with spatially isotropic long-wavelength anharmonicity and optical response. For cw acoustic excitations, when \( I_{ac}(r, t) = I_{ac} = \text{const} \), Eqs. (2) and (3) yield the same quasienergy spectrum as that of the quadratic Hamiltonian (1). If \( I_{ac} = 0 \), Eqs. (2) and (3) reduce to the standard TO-phonon polaron equations.1 The fourth term on the left-hand side (lhs) of the polarization Eq. (3) yields a Stark shift of the TO-phonon frequency, which is \( \propto I_{ac} \) and is associated with the quartic phonon nonlinearity. The last two terms on the lhs of Eq. (3) give rise to the Bragg spectrum of TO-phonon polaritons driven by the AW.

In Eqs. (1) and (3), the matrix elements \( m_{3,4} \) and \( \tilde{m}_4 \) are given by

\[
m_3 = 6 \left( \frac{v_0}{\hbar^2 \mu k} \right)^{1/2} V_3(k, p - k, -p),
\]

\[
m_4 = 12 \left( \frac{v_0}{\hbar^2 \mu k} \right) \left[ V_4(k, k, p - 2k, -p) + V_4(k, k, -p, p - 2k) \right],
\]

\[
\tilde{m}_4 = 24 \left( \frac{v_0}{\hbar^2 \mu k} \right) V_4(k, -k, p, -p),
\]

where \( v_0 \) is a volume of the primitive cell and

\[
V_3(k, p - k, -p) = \frac{1}{6} \left( \frac{\hbar^3}{8v_0 k \Omega_{TO}} \right)^{1/2} \Phi^{(3)}(k, p - k, -p),
\]

\[
V_4(k, -k, p, -p) = \frac{1}{24} \left( \frac{\hbar^2}{4v_0 k \Omega_{TO}} \right) \Phi^{(4)}(k, -k, p, -p).
\]

Here, \( \Phi^{(3)} \) (\( \Phi^{(4)} \)) is proportional to the Fourier transform of the third-order (fourth-order) spatial derivative of the interatomic potential. The explicit formulas for \( \Phi^{(3,4)} \) are given, e.g., in Refs. 16 and 17.
Acoustic phonons in CuCl driven by the bulk TA wave of $\nu_{ac}=50$ MHz, black lines, and the bare polariton spectrum, red (dark gray) lines. The wave vector $p$ is normalized to the acoustic wave vector $k$. The arrows highlight the N-TA-phonon resonant coupling between the polariton states ($N=1,2$). [(b)-(e)] The calculated total Bragg reflectivity (black lines), $R=\Sigma_{j}|r_{j}|^{2}$, against the light frequency $\nu=\omega/(2\pi)$ for $\nu_{ac}=50$ MHz, 100 MHz, 200 MHz, and 1 GHz, respectively. The bare polariton reflectivity is shown by the red (dark gray) lines. The Bragg signals labeled in (c) as $n=-1$ and $n=-2$ are mainly due to $|r_{-1}|^{2}$ and $|r_{-2}|^{2}$, respectively. The acoustic intensity $I_{ac}=25\text{ kW}/\text{cm}^{2}$.

and (3) with $m_{4}=0$, is plotted in Fig. 2(a) for CuCl parametrically driven by the TA wave of frequency $\nu_{ac}=50$ MHz and constant intensity $I_{ac}=25\text{ kW}/\text{cm}^{2}$. The spectrum, which arises from spatial and temporal modulation of the crystal lattice, can be interpreted in terms of the Brillouin-zone picture, with acoustically induced energy gaps $\Delta_{N}=I_{ac}^{1/2}$ due to the $N$-phonon resonant transitions within the polariton dispersion branches. The spectral positions of the transitions are indicated in Fig. 2(a) by the vertical arrows. For the frequency scale used in Fig. 2(a), only the gaps $\Delta_{N=1}$ and $\Delta_{N=2}$ in the upper polariton branch are clearly seen. From the air side, $z<0$ (see Fig. 1), the light field is given by

\[ E(z<0,t) = e^{i\omega t}e^{-i\omega t} + \sum_{n} r_{n}e^{-i\omega t}e^{-i(\omega+n\omega_{ac})t}, \]

where $q_{a}=(\omega+n\omega_{ac})/c$ with $-n_{max} \leq n \leq n_{max}$ (we proceed up to $n_{max}=60$) and $r_{n}$ stands for the amplitude of the outgoing Bragg replica $n$ normalized to the unity amplitude of the incoming light wave. The electric field propagating in the crystal ($z>0$) is

\[ E(z>0,t) = \sum_{n,j} A_{j}E_{nj}e^{i(p_{j}+nk)z-(\omega+n\omega_{ac})t}. \]

Here, $p_{j}=p_{j}(\omega)$ is the wave vector associated with the quasienergy dispersion branch $j$ ($-n_{max} \leq j \leq n_{max}$) of the acoustically driven polariton, $E_{nj}$ are the corresponding normalized eigenvectors, and $A_{j}$ are the eigenmode amplitudes. Both $p_{j}=p_{j}(\omega)$ and $E_{nj}$ are the forced-harmonic solutions of Eqs. (2) and (3) for real-valued frequency $\omega$. The exponential on the right-hand side of Eq. (8) as well as the basic relationships $p_{j+s}(\omega)=p_{j}(\omega-s\omega_{ac})+sk$ and $E_{n+j}(\omega)=E_{n}(\omega-s\omega_{ac})$, with integer $s$, reflect the acoustic wave vector and frequency translational invariance of the quasiparticle spectrum. The Maxwellian boundary conditions at $z=0$ together with Eqs. (7) and (8) yield

\[ \delta_{n,0} + r_{n} = \sum_{j} A_{j}E_{nj}, \quad \delta_{n,0} - r_{n} = \sum_{j} A_{j}E_{nj} p_{j} + nk. \]

The above set of $2(n_{max}+1)$ linear equations determines $r_{n}$ and $A_{j}$.

Table I summarizes values of the parameters used in numerical simulations.

| TABLE I. Parameters of bulk CuCl and TICl used in the numerical calculations. |
| CuCl | TICl |
|---|---|
| $\hbar\Omega_{TO}$ (meV) | 20.28 | 7.81 |
| $\hbar\Omega_{LO}$ (meV) | 14.53 | 17.97 |
| $\hbar\gamma_{TO}$ (meV) | 0.20 | 0.92 |
| $v_{s}$ ($10^{4}$ cm/s) | 2.02 | 2.19 |
| $h\nu_{1}$ ($10^{-12}$ meV$^{1/2}$ cm s$^{1/2}$) | 0.10 | 0 |
| $h\nu_{4}$ ($10^{-27}$ cm$^{2}$ s) | 0 | 0.64 |

III. NUMERICAL MODELING

In this section, by solving numerically Eqs. (2), (3), and (9), we model the terahertz response of acoustically pumped copper chloride and thallium chloride crystals. For the first (second) compound, we set $m_{3}=m_{4}=0$ ($m_{3}=0$) in Eq. (3), according to the relative strength of the cubic and quartic anharmonic coefficients in CuCl (TICl).

A. Acoustically driven THz polaritons in bulk CuCl

In Figs. 2(b)–2(e), we compare the calculated total Bragg reflectivity, $R=R(\omega)=\Sigma_{n}r_{j}^{2}$ (black solid lines), with the reflectivity of the acoustically unperturbed THz polariton, $R^{(0)}=R^{(0)}(\omega)$ [red (dark gray) solid lines]. The sharp spikes in the Bragg spectrum of the acoustically driven CuCl crystal
are clearly seen for the one-TA-phonon and two-TA-phonon transitions, both for the upper-polariton (UP) and lower-polariton (LP) branches [see Fig. 2(a) against Fig. 2(b)]. For $\nu_{\text{ac}} \approx 30-300 \text{ MHz}$, the backward scattered Bragg replica $|r_{-1}|^{2}$ peaks at the energy of the one-phonon transition and is highly efficient, with $|r_{-1}|^{2}/R \approx 50-70 \%$. The peak position and its strength are effectively tunable by changing the frequency and intensity of the AW [see Figs. 2(b)–2(d)]. This can be used for the frequency down conversion by $\omega_{\text{ac}} = 2\pi v_{\text{ac}}$ of the optically induced THz polariton and thus of the incident light field. Generally, the backward Bragg scattering signals $|r_{0}|^{2} \propto I_{\text{ac}}^{0}(n < 0)$ peak at the spectral position of the gap $\Delta_{\text{ac}}/|n|$, i.e., for the light frequency $\omega = \omega_{\text{ac}}$ which satisfies the resonant Bragg condition $p_{\text{int}}(\omega_{N}) = Nk/2$ with $N=1,2,\ldots,n$.

The Bragg signal $n=0$ appears as the AW-induced change in the reflectivity at incident frequency $\omega$, $|r_{0}(\omega)|^{2} - R^{(0)}(\omega)$. The strength of the $n=0$ signal sharply increases with decreasing detuning $|\omega_{N}=1-\omega_{\text{TO}}|$ from the TO-phonon resonance so that $|r_{0}|^{2} - R^{(0)}$ becomes dominant over $|r_{-1}|^{2}$. For $\nu_{\text{ac}} \approx 1 \text{ GHz}$, when the one-phonon-acoustic transition within the LP branches occurs very close to $\omega_{\text{TO}}$, the AW-induced change in the reflectivity [see Fig. 2(e)] is completely determined by the $n=0$ replica and has no $\nu_{\text{ac}}$-down-converted frequency components. Thus, this operating mode can be used for TO-phonon polariton deflectors and acoustically controlled THz filters.

The interaction length $\ell_{\text{int}} = \ell_{\text{int}}(\omega_{0})$ required for the formation of the Bragg signals and thus for AW control of the THz light field is plotted in Fig. 3(a) for various $\nu_{\text{ac}}$. The sharp troughs at $\omega = \omega_{N=1}$ and $\omega_{N=2}$ in the $\ell_{\text{int}} = \ell_{\text{int}}(\omega)$ profile are due to the $n=1$ and $-2$ Bragg replicas [see Fig. 3(a)]. Figure 3(b) shows the light field distribution associated with the optically induced THz polariton in the acoustically driven CuCl crystal. Apart from the broad black band [see Fig. 3(b)], which corresponds to the Reststrahlen band with rather weak penetration of the light field into the crystal, the narrow stripes of alternating color illustrate the formation of the Bragg replicas. The interaction length for the $n=1$ replica at its resonant frequency $\omega = \omega_{N=1}$ is given by

$$ \ell_{\text{int}} = \frac{\hbar k c^{2}}{4 m_{3} v_{\text{ac}}^{2} \omega_{N=1}^{2} - \omega_{\text{TO}}^{2} + \Gamma_{R}^{2}}. $$

Equation (10), which is valid for $|\omega_{\text{TO}} - \omega_{N=1}| > \Gamma_{\text{TO}}$ and $\ell_{\text{int}}^{N>1}$, shows the resonant decrease in $\ell_{\text{int}}^{N=1}$ with decreasing frequency detuning from the TO-phonon resonance. In this case the interaction between the light field and pumping AW is mediated by the TO-phonon resonance, giving rise to $\ell_{\text{int}}^{N=1}$ of only a few tens of acoustic wavelength $\lambda_{\text{ac}}$ (see Fig. 3).

**B. Acoustically driven THz polaritons in bulk TlCl**

The calculated room-temperature reflectivity $R = R(\omega)$ of a TlCl crystal driven by the TA wave of frequency $\nu_{\text{ac}} \approx 25 \text{ MHz}$ and $125 \text{ MHz}$ is plotted in Figs. 4(a) and 4(b), respectively. In this case, in Eqs. (1) and (3) we put $m_{3}=0$, and therefore only even-order TA-phonon-assisted transitions occur. The Bragg signals $n=-2$, due to $|r_{-2}|^{2} \propto I_{\text{ac}}^{2}$, are indicated in Fig. 4(a) by arrows, for the transitions within the LP and UP branches, respectively. Similarly to the previous case (CuCl), for the resonant frequency $\omega_{N=2}$ close to $\omega_{\text{TO}}$ the AW-induced change in $R$, $\Delta R = R - R^{(0)} - I_{2}^{2}$ [see Fig. 4(b)], is mainly due to the $n=0$ Bragg replica. The quartic nonlinearity leads to the Stark blueshift by $2m_{3} I_{\text{ac}}^{2}$ of the TO-phonon frequency, according to Eqs. (1) and (3), as is clearly seen in Figs. 4(a) and 4(b). The Stark shift $\sim 0.1-0.2 \text{ THz}$ has a rather sharp contrast on the blue side of the THz reflectivity.

**FIG. 3.** (Color online) (a) The interaction length between the THz polariton and TA pumping wave in CuCl, $\ell_{\text{int}} = \ell_{\text{int}}(\omega_{0})$, calculated for $I_{\text{ac}} = 25 \text{ kW/cm}^{2}$ and $\nu_{\text{ac}} = 50, 100$, and $200 \text{ MHz}$. (b) The electric field profile $|E(z>0, \omega)|^{2}$ evaluated for $\nu_{\text{ac}} = 100 \text{ MHz}$ ($\lambda_{\text{ac}} = 20.2 \mu\text{m}$). The gray-black color scale is logarithmic with black color corresponding to $E=0$.

**FIG. 4.** (Color online) The total Bragg reflectivity $R = R(\omega)$ of a TlCl crystal driven by the TA wave of frequency (a) $\nu_{\text{ac}} = 25 \text{ MHz}$ and (b) $125 \text{ MHz}$, $I_{\text{ac}} = 100 \text{ kW/cm}^{2}$ (black dotted lines) and $200 \text{ kW/cm}^{2}$ (black solid lines). The acoustically unperturbed THz spectrum $R^{(0)} = R^{(0)}(\omega)$ is shown by the red (dark gray) lines. Inset: the acoustically induced Stark shift of the TO-phonon line.
[see inset of Fig. 4(b)]. This can be used in device applications, e.g., for frequency tunable THz filters and deflectors.

IV. DISCUSSION

The third- and fourth-order anharmonic nonlinearities tend to cancel each other\(^\text{17,23}\) so that the values of \(m_3\) (CuCl) and \(m_4\) (TICl) we have inferred from the experimental data are in the lowest limit of their actual values. The proposed acoustic modulation of THz polaritons generally allow us, by comparing \(n=-1\) and \(n=-2\) Bragg replicas evaluated with Eqs. (2), (3), and (9), to distinguish the two nonlinearities as well as to analyze interference between the cubic and quartic anharmonic coefficients. The latter process occurs, e.g., for soft TO phonons in ferroelectric LiTaO\(_3\) and LiNbO\(_3\).

Within the used nonperturbative approach, each Bragg replica \(n\) integrates all \(n+s−s\) TA-phonon transitions with \(s\leq n_{\text{max}}\); with increasing \(I_{\text{ac}}\) the bare \(n\)-phonon transitions become dressed by higher-order processes when \(n+s\) phonons are emitted and \(s\) phonons absorbed. For the \(n=-1\) replica shown in Fig. 2(b), e.g., the multiphonon transitions \(-1+1−1\), \(-1+2−2\), etc., account for about 90% of \(|\langle r_0\rangle|^2\). For \(\nu_{\text{ac}}\geq 1\) GHz [see Fig. 2(e)], the dominant contribution to \(|\langle r_0\rangle|^2−R(0)\) stems from \(-s+s\) multi-TA-phonon transitions with \(s\geq 1\).

The acoustically induced modulation of far-infrared polaritons has to be particularly strong for ferroelectric soft TO phonons (e.g., in LiTaO\(_3\) and LiNbO\(_3\))\(^6\) and bismuth titanate\(^5\). In this case, a multilayer potential local for the displacive ferroelectric mode has a considerable low-wavevector component and therefore yields large values of \(V_3\) and \(V_4\). Very far-infrared optical phonons (2–10 meV) in zirconium tungstate (ZrW\(_2\)O\(_8\)) indicate anomalously high anharmonicity\(^7,8\). The normal modes associated with soft TO phonons in this negative thermal-expansion compound are a mixture of librational and translational motion. The latter strongly couples with acoustic phonons giving rise, as we foresee, to manipulation of the THz polaritons by using ultrasonic waves of modest \(I_{\text{ac}}\)\(^,27\).

In accordance with Eq. (4), \(m_3\) and \(m_4\) scale to the sound velocity as \(v_s^{-3/2}\) and \(v_s^{-3}\), respectively. As a result, the use of a surface AW in order to control the propagation of THz polaritons considerably reduces the operating acoustic intensity \(I_{\text{ac}}\); \(v_s=v_{\text{SAW}}\) is usually by about 20% less than \(v_s=v_{\text{TA}}\).

The realistic values of the damping constant, \(\hbar\gamma\)\(\text{TO}=0.9\) meV for CuCl at \(T=5–10\) K (Refs. 21 and 23) and \(\hbar\gamma\)\(\text{TO}=0.9\) meV for TICl at room temperature\(^,25\) are used in our numerical simulations. Note that \(\gamma\) is rather weakly temperature dependent, e.g., roughly doubling its value for CuCl with increasing temperature from cryogenic values to the room one. In all the materials mentioned in the paper, the polariton effect associated with TO phonons persists at room temperature. This is because the Rabi frequency of THz polaritons, \(\hbar\Omega_{\text{g}}\sim 10\) meV, is much larger than the damping constant, which even for soft TO-phonon modes is usually \(\hbar\gamma\)\(\text{TO} \approx 0.5–1\) meV only.

V. SUMMARY

In this paper we have proposed, theoretically described, and numerically modeled the THz response of far-infrared polaritons parametrically driven by a coherent acoustic field. The cubic and quartic phonon anharmonicity processes give rise to an acoustically induced Bragg grating. In this case, the TO-phonon resonance mediates and strongly enhances the acousto-optical nonlinearity leading to the anomalously small interaction length \(r_{\text{int}}\sim 100\) \(\mu\)m between the optical and acoustic fields, which is required for the formation of the THz response (Bragg replicas). The main results of the work are: (i) the macroscopic Eqs. (2) and (3), which describe spatially temporal or wave-vector-frequency effective control of far-infrared polaritons and THz light associated with them by applying the acoustic field. (ii) The nonperturbative approach, based on Eqs. (7)–(9), to calculate the total Bragg reflectivity of TO-phonon polaritons modulated by the ultrasonic acoustic wave. (iii) The illustration of the high efficiency of the proposed effect by numerical modeling of the THz response of bulk CuCl and TICl pumped by the coherent acoustic field. (iv) The large potential of the effect for device applications: the small interaction length between the THz light and subgigahertz acoustic field would potentially lead to a new family of THz microdevices, such as frequency-tunable THz detectors and filters, Bragg switches, and frequency converters.

ACKNOWLEDGMENTS

We thank S. G. Tikhodeev, D. R. Khokhlov, P. Mauskopf, and R. Zimmermann for valuable discussions. This work was supported by RS (Grant No. JP0766306), EPSRC and WIMCS.

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The operating acoustic intensities used in the simulations of bulk CuCl and TiCl driven by a TA wave are similar to those applied to a GaAs-based microcavity (MC) in the experiments (Refs. 12 and 13). The latter study deals with the acoustical Stark effect for MC polaritons. Our recent evaluations yield a much smaller operating intensity, $I_{ac} = 1 - 10 \, \text{kW/cm}^2$, needed for the effective manipulation of TO-phonon polaritons in LiNbO$_3$ driven by a bulk TA wave. For ZrW$_2$O$_8$, we anticipate still further reduction in the operating acoustic intensities toward a subkilowatt per square centimeter scale.