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Phonon-polaritons in quasiperiodic piezoelectric superlattices

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Phonon-polaritons are studied both theoretically and experimentally in a one-dimensional two-component generalized quasiperiodic piezoelectric superlattice. The experimental observation of phonon-polaritons through dielectric abnormality is carried out at the microwave region. Some potential applications are discussed.
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As an elementary excitation in solid-state physics, the polariton is due to the coupling between the photon and the polar elementary excitation. Owing to the unusual properties, the polaritons are of great interest from both a fundamental and an applied perspective. Recently, a polariton laser based on exciton-polaritons has been demonstrated in a semiconductor microcavity.[1] As for the phonon-polariton,[2,3] the rapidly varying refractive index is made use of in constructing prisms for infrared spectroscopy.[4] Ensued from the study of artificial microstructure materials, much effort has been devoted to the research on the phonon-polariton. In the periodic superlattice, a periodic potential with a giant period, in contrast with the atomic period, results in the formation of the miniature Brillouin zone. By virtue of this, the far-infrared Raman laser and Reststrahlen filter made of AlAs/GaAs superlattices become realizable.[5] Another property of the phonon-polariton, significantly reduced group velocity, can be utilized in solid-state traveling wave devices.[6] Moreover, the photonic band gap (PBG), in which the propagation of electromagnetic (EM) waves is forbidden, will be affected by the presence of the phonon-polariton in photonic crystals composed of polar materials.[7–9] It is shown that the phonon-polariton coupling flattens the photonic bands and is favorable for opening up the PBG.

On the other hand, the discovery of quasicrystals has fired up a new field of condensed-matter physics and given rise to many practical applications since l984.[10] For example, the multiwavelength second-harmonic generation and the direct third-harmonic generation have been realized in a Fibonacci superlattice.[11,12] In the field of photonic crystals, the complete PBG in 12-fold symmetric quasicrystals has been reported.[13] Furthermore, compared with a one-dimensional (1D) two-component Fibonacci superlattice, a 1D two-component generalized quasiperiodic superlattice (GQPSL) possesses more freedom for applications.[14]

In this Letter, we investigate the influence of structural variation of the piezoelectric superlattice upon the phonon-polaritons on the basis of a 1D two-component GQPSL. We measured the dielectric function of the GQPSL at the microwave region, thereby obtaining the physical information required to deduce the properties of polaritons.[15] The possible applications are discussed.

In the absence of translational symmetry, the Bloch theory is no longer adequate for quasiperiodic structures. But according to the projection method,[16] the 1D quasiperiodic structure may be considered as the projection of a two-dimensional (2D) periodic structure. As an example, we consider a GQPSL with a $3m$ point group which consists of two building blocks, A and B, with each block made up of one positive and one negative ferroelectric domain (a ferroelectric material is piezoelectric). The widths of blocks A and B are l_A and l_B, respectively. We assume that the negative domain of blocks A and B has the same width l, shown in Fig. 1(a). Different from the Fibonacci sequence, the projection angle becomes an adjustable structure parameter in the GQPSL, and its tangent is not fixed as a golden ratio, i.e., $(1+\sqrt{5})/2$. Figures 1(b) and 1(c) show two schematic diagrams of the GQPSL structures: one is the so-called side-by-side configuration, the other the head-to-head configuration.[17] In both cases, the piezoelectric coefficient, as an odd-rank tensor, will change signs for domains with different spontaneous polarization directions, which arouses quasiperiodically modulated piezoelectric coefficients in the GQPSL.

With a GQPSL aligning along the x axis in Fig. 1(b), a vertically incident y-polarized EM wave propagates into it on the left-hand side surface. The properties of the phonon-polariton can be obtained from the following piezoelectric and motion equations:[18]

$$T_1(x,t) = C_{11}^E S_1(x,t) + e_{22}(x) E_2(x,t),$$

$$P_2(x,t) = -e_{22}(x) S_1(x,t) + \varepsilon_0(\varepsilon_{11}^S - 1) E_2(x,t),$$

$$\rho \frac{\partial^2 S_1(x,t)}{\partial t^2} = \frac{\partial T_1(x,t)}{\partial x^2}, \tag{1}$$

where T_1, S_1, E_2, and P_2 are the stress, strain, electric field, and polarization, respectively. C_{11}^E, $e_{22}(x)$, ε_{11}^S, and ρ are the elastic coefficient, piezoelectric coefficient, dielectric coefficient, and mass density, respectively. The damping of materials has been omitted here. The second equation of Eq. (1) implicates that a longitudinal wave S_1 introduces a transverse electric polarization P_2, which can interfere with the EM wave. For an infinite GQPSL structure, the quasiperiodically modulated piezoelectric coefficient $e_{22}(x)$ can be written, using Fourier transformation,[19] as $e_{22}(x) = e_{22} f(x) = e_{22} \sum_{m,n} f_{m,n} \exp(iG_{m,n} x)$ with

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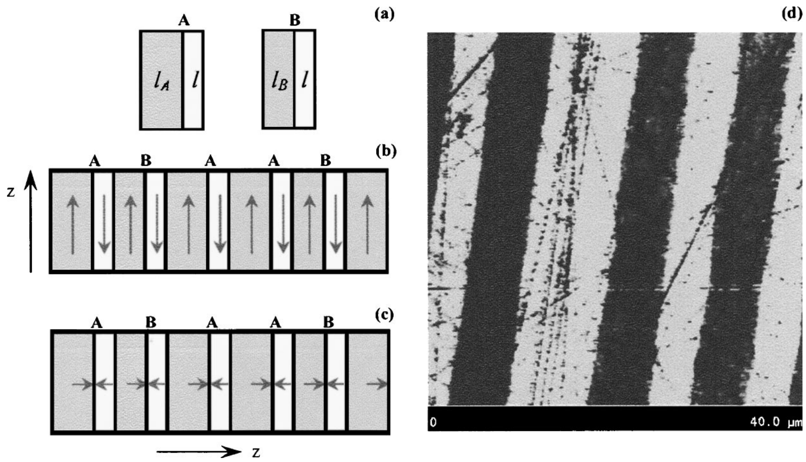

FIG. 1. (Color online) Schematic of a 1D two-component GQPSL structure (a) composed of two building blocks *A* and *B*, (b) with side-by-side 180° reversed ferroelectric domains, or c) with head-to-head 180° reversed ferroelectric domains. The arrows indicate the directions of spontaneous polarization. (d) Piezoresponse scanning force microscopy of a GQPSL based on the $LiTaO_3$ single crystal. It is the image of phase response.

$$f_{m,n} = \frac{2(1+\tau)l}{D}\frac{\sin(G_{m,n}l/2)}{G_{m,n}l/2}\frac{\sin X_{m,n}}{X_{m,n}}, \tag{2}$$

where $G_{m,n}=2\pi(m\tau+\mathrm{n})/D$ is the reciprocal vector (m,n are two integers), $D=\tau l_A+l_B$ is the average structure parameter of the GQPSL, $X_{m,n}=\pi(l+\tau)(nl_A+ml_B)/D$, $\tau=\tan\,\theta$, and θ is the adjustable projection angle. Using Eq. (1), the average dielectric function $\varepsilon_2(k,\omega)$ of the GQPSL is found to be

$$\varepsilon_2(k,\omega) = \varepsilon_{11}^S - \frac{e_{22}^2}{\varepsilon_0 L}\sum_{m,n,m',n'} f_{m,n}f_{m',n'} \times \frac{G_{m,n}^2+2G_{m,n}k+k^2}{\rho\omega^2 - C_{11}^E(G_{m,n}+k)^2}\int_0^L e^{i(G_{m,n}+G_{m',n'})x}dx, \tag{3}$$

where k and ω are the wave vector and angular frequency of EM waves. L is the whole length of the GQPSL, and m',n' are also two integers. From Eq. (3) we can see that the resonance, which engenders phonon-polaritons, will occur around $\omega^2=G_{m,n}^2 v^2$ [$v=(C_{11}^E/\rho)^{1/2}$ is the phase velocity of the longitudinal superlattice vibration]. Hence, the positional distribution of resonance peaks reflects the quasiperiodicity of the GQPSL structure.

According to Maxwell's relation, the dispersion relation of the phonon-polariton in GQPSL can be gotten easily. That is,

$$c^2k^2/\omega^2 = \varepsilon_2(k,\omega), \tag{4}$$

where c is the phase velocity of the EM wave in free space.

In the very long wavelength region, the wave vector is very close to zero, and to a good approximation the dielectric function can be considered to depend only upon the frequency of waves. Taking the damping of the material into account, Eq. (3) can be changed to the form

$$\varepsilon_2(\omega) = \varepsilon_{11}^S\left[1 - \frac{K^2}{L}\sum_{m,n,m',n'}\frac{f_{m,n}f_{m',n'}\omega_{m,n}^2}{\omega^2-\omega_{m,n}^2+i\omega\gamma} \times \int_0^L e^{i(G_{m,n}+G_{m',n'})x}dx\right], \tag{5}$$

where $K^2=e_{22}^2/(C_{11}^E\varepsilon_0\varepsilon_{11}^S)$ is an electromechanical coupling coefficient, $\omega_{m,n}^2=G_{m,n}^2v^2$, $\omega_{m,n}$ is resonance frequencies of longitudinal superlattice vibrations in the GQPSL, i.e., the frequency positions of phonon-polaritons, and γ represents a damping constant with the dimension of frequency.

The GQPSL based on the congruent $LiTaO_3$ single crystal is fabricated by the method of electric-field poling.[20] The GQPSL used in our experiment is 640 blocks with a total length of about 8.2 mm along the x axis. The other structure parameters are $\tau=0.5645$, $l=5.75$ μm, $l_A=14.41$ μm, and $l_B=10.04$ μm respectively. The widths of domains after poling are determined with piezoresponse scanning force microscopy, in which the inverse piezoelectric effect is used. The z surface of the GQPSL was polished, and the phase response was recorded, as shown in Fig. 1(d). In this Letter, the material constants of the congruent $LiTaO_3$ crystal are selected from Ref. 21. The dispersion relation of the phonon-polariton in the GQPSL calculated from Eq. (4) is shown in Fig. 2(a). Figure 2(b) shows that for a phonon-polariton in the GQPSL, there are three branches forming two separate band gaps, while in the ionic crystal there are only two branches with one band gap. There are not only transverse phonon-polaritons, but also longitudinal phonon-polaritons in piezoelectric superlattices.[22,23] As shown above, the phonon-polaritons in this case are longitudinal phonon-polaritions. When a y-polarized EM wave propagates along the x axis of the GQPSL, it will be strongly reflected as long as its frequency lies in the band gap of the phonon-polariton. By use of this property, the reflector and polarizer based on the phonon-polariton can be made.

The frequency distribution of the phonon-polaritons exhibits a quasiperiodic property of the GQPSL, in accordance with the reciprocal vector. In the periodic structure. the frequency positions of phonon-polaritons are equidistant.[23] Be-

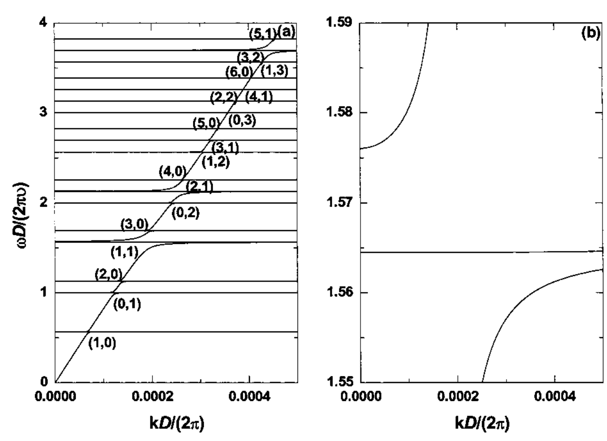

FIG. 2. Normalized dispersion curves of (a) the longitudinal phonon-polaritons of the GQPSL and (b) the enlarged one labeled as (1,1).

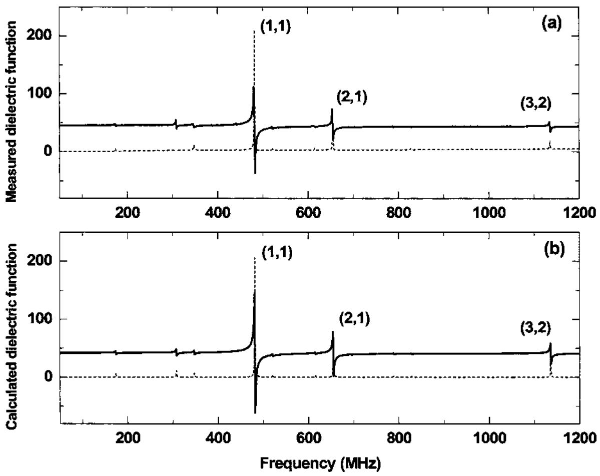

FIG. 3. The dielectric function curves for the GQPSL. (a) The measured results and (b) the calculated results choosing damping constant $\gamma = 0.003\ \omega_{1,1}$. The solid lines represent the real part, and the dashed lines the imaginary part.

sides the piezoelectric coefficient, Eq. (3) also tells us that the Fourier coefficients are responsible for the size of band gaps, correlated with the coupling intensity. As we have seen, many adjustable parameters allow one to conveniently tune the frequency positions of phonon-polaritons and the magnitude of the Fourier coefficients according to specific applications. For example, the arbitrarily adjustable projection angle enables us to achieve tunable channels with a narrow wavelength interval in wavelength division multiplexing (WDM) devices. The major property of the phonon-polariton is prohibiting simultaneously incident EM waves or sound waves within some special frequency ranges from penetrating through materials. This makes it possible that the phonon-polariton material can be used as the barrier for harmful EM waves or noises. It is also beneficial to miniaturization of devices as the wavelength of EM waves is much larger than the size of building blocks, whereas they should be comparable with each other in photonic materials.

The experimental dielectric function of the GQPSL measured with an HP4291B RF impedance/material analyzer is shown in Fig. 3(a). There is a one-to-one correspondence between the frequency position and magnitude of abnormal dielectric function peaks in Fig. 3(a), and the frequency position and the band-gap size of phonon-polaritons in Fig. 2, especially between the three most intense peaks and the three largest band gaps, labeled as (1,1), (2,1), and (3,2). In order to simulate the experimental dielectric function, the damping constant γ of the GQPSL must be given beforehand. By virtue of the damped harmonic-oscillator model, the damping constant γ approximates to either the full width at half maximum of a peak in the imaginary part of the dielectric function, or the difference between the two frequencies corresponding to the maximum and minimum around a point of abnormality in the real part of the dielectric function. From the measured dielectric function peaks the damping constant y is approximately equal to $0.003\ \omega_{1,1}$. Figure 3(b) shows the calculated dielectric function of the GQPSL. The profiles of theoretical curves are quite similar to those measured.

The frequency region for phonon-polaritons of the GQPSL falls into the microwave band, which is determined by its average structure parameter. It is noteworthy that the epitaxial technique, such as magnetron sputtering, can bring the frequency of phonon-polaritons up to the far-infrared region by adjusting the average structure parameter of the GQPSL. Then, the frequeny of phonon-polaritons can span the operational range of most optoelectronic devices.

In conclusion, the phonon-polariton in the 1D two-component GQPSL was studied in theory and experiment. The quasiperiodic modulation gives rise to the quasiperiodic frequency distribution of phonon-polaritons. If the piezoelectric coefficient is modulated aperiodically, it will introduce aperiodical frequency distribution of phonon-polaritons. This property can lead to the practical applications, such as for WDM devices in optical communications, and for EM wave (or sound wave)-proof materials in environmental protection.

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