Excitation of Coherent and Incoherent Terahertz Phonon Pulses in Quartz Using Infrared Laser Radiation*

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We report piezoelectric surface excitation of coherent acoustic 0.891- and 2.53-THz phonon pulses in quartz by means of chopped infrared laser radiation and the detection of these highly collimated phonons 10 mm from the excitation area using a superconducting tin bolometer. At 2.53 THz the effect of dispersion is clearly visible. Absorption of infrared photons in the bulk material generates incoherent phonons which also can be detected by the bolometers.

We report in this paper first experiments showing the direct transformation, without change of frequency, of infrared laser radiation into phonon pulses by piezoelectric surface excitation[1] and, at higher frequencies, by one-phonon infrared absorption in quartz. These experiments are performed by focusing a chopped infrared-laser beam upon the X-cut face of a $10\times10\times10$-mm^3 quartz cube kept in contact with liquid helium. In the case of piezoelectric phonon excitation the phonons are produced only at the surfaces; in the case of strong infrared absorption the phonon generation takes place in a thin layer near the surfaces. In order to get a time separation between laser radiation and phonon signal at the superconducting bolometer at the rear face, the laser-pulse duration should be 1 μsec or less. Since pulsed molecular lasers usually show an output radiation far too long for our purpose, an external beam chopper has been constructed[2] and inserted.

Changing only gas fuel and some electrical parameters, laser emission can be observed in our setup at distinct frequencies between 337 (HCN) and 10.6 μm (CO_2). We report here only phonon-excitation experiments using single emission lines at 0.891 THz (337-μm line[3] of HCN), at 2.53 THz (118-μm line[4] of H_2O), and at 10.7 THz (28-μm line[5] of H_2O). These lines were controlled by a compact external Michelson interferometer. Usually peak powers of about 10 W at 2.53 THz and of about 50 W at 10.7 THz are attainable. The insertion loss of the mechanical chopper is nearly 60% at 0.891 THz and less at higher frequencies.

Our tin bolometers cover an area of 1 mm in diameter and have the shape of a meander. The stripe width as well as the free-space between the stripes is 0.1 mm. Vacuum deposition of the 600–800-Å films is done with the substrate cooled to liquid-nitrogen temperature. Whereas these films are highly sensitive detectors for phonons,[6] only a very small fraction of incident infrared photons are absorbed as a consequence of the high electronic conductivity.

Before discussing the experiments on coherent excitation, we give a rough estimate of the attainable power conversion from the incident electromagnetic wave into coherent phonons ("sound waves") at low frequencies where continuum acoustics can be applied. The method of piezoelectric surface excitation was introduced by Bömmel and Dransfeld[7] for the generation of coherent phonon pulses in the gigahertz range. As Jacobsen[8] pointed out, the generation of sound in a piezoelectric crystal is due to the discontinuity of the piezoelectric stress at the surface of the crystal. Since a free crystal face must be stress-free, a mechanical wave of the same frequency is generated at the surface and just compensates these driving piezoelectric stresses caused by the alternating electric field. Excitation and coherent detection by the inverse mechanism was reported at frequencies up to 114 GHz.[9] A consequent extension to higher frequencies is the use of far-infrared lasers[10] and of incoherent phonon detectors such as superconducting tunnel junctions or superconducting bolometers. For an estimate of the expected power conversion in such an experiment, we start with the following material equation of a piezoelectric crystal,[11] written in scalar form, i.e., we neglect all tensor properties in our estimate:

$$\varepsilon = s\sigma + dE. \tag{1}$$

ε is the effective strain, σ is the effective stress, E is the effective electric field, s is a typical compliance constant, and d the largest component of the piezoelectric tensor. At the crystal surface the stress σ vanishes and (1) gives directly

the strain $\varepsilon = dE$ of the generated sound wave. The particle velocity v is connected with the strain in a plane wave in the form $\varepsilon = v/c_q$, where c_q is the phase velocity of the sound wave. Eliminating ε, we get for the effective particle velocity of the generated sound wave the following estimate:

$$v = c_q dE. \tag{2}$$

On the other hand, the power carried by a plane wave per area A is given for an electromagnetic wave by $P_k = Ac_k\epsilon_0 E_0^2$ and for a sound wave by $P_q = Ac_q\rho v^2 = Ac_q^3\rho d^2E^2$. Here, c_k is the vacuum velocity of light for the exciting electromagnetic wave, E_0 is the corresponding effective electric field amplitude, and ρ is the mass density of the crystal. The electric field E in the crystal near the surface is given by $E = 2E_0/(\sqrt{\epsilon}+1)$, if normal incidence of the exciting electromagnetic wave is assumed and the crystal has dielectric constant ϵ. The power conversion in piezoelectric surface excitation is defined by the ratio P_q/P_k. Within our estimation, the above relations yield, for the assumed normal incidence, the following expression:

$$P_q/P_k = 4\rho d^2 c_q^3/[\epsilon_0 c_k(\sqrt{\epsilon}+1)^2]. \tag{3}$$

With use of $\rho = 2650$ kg/m^3, $d = 2.3\times10^{-12}$ C/N, $c_q = 5.75$ km/sec, and $\epsilon = 4.4$ for quartz, Eq. (3) yields a power conversion $P_q/P_k = 4.2\times10^{-7}$, i.e., -64 dB. This value is in good agreement with a detailed analysis of piezoelectric surface excitation by plane electromagnetic waves,[12] which shows in detail the influence of crystal face, angle of incidence, and electromagnetic polarization on the generation of phonons of special polarization. Because of the large difference in phase velocity of electromagnetic waves and sound waves, only phonons with a wave vector nearly perpendicular to the crystal face can be generated. Our experiments were performed with an X-cut quartz crystal and with nearly normal incidence. In this case, continuum acoustics allows the generation of transverse phonons only, propagating along the X direction. Longitudinal phonons should not be observed.

Surface excitation of coherent 0.891-THz phonons was first investigated by use of HCN-laser radiation. One result is shown in Fig. 1(a). Photons falling upon the tin bolometer at the rear face produce a time-reference pulse. The following broader pulse is due to phonons which are spatially coherent, and hence highly collimated. This spatial coherence can easily be verified experimentally by moving the laser spot over the front face of the quartz crystal and observing the bolometer signal at the same time. In principle, an enhancement of directional phonon intensity can also occur with incoherent phonons because of phonon focusing.[13] But a detailed numerical analysis within continuum acoustics reveals[14] that this is not the case in the X direction of quartz. This result is in accordance with our experimental observations using thermal phonon radiators. For these incoherent radiators a bolometer signal results as given in Fig. 1(b). The group velocities of phonons propagating in the X direction are at ultrasonic frequencies[15]: $c_L = 5.75$ km/sec for longitudinal phonons, $c_{T1} = 5.1$ km/sec for fast transverse phonons, and $c_{T2} = 3.3$ km/sec for slow transverse phonons. The expected pulse positions due to these velocities are given in Fig. 1. In the case of coherent excita-

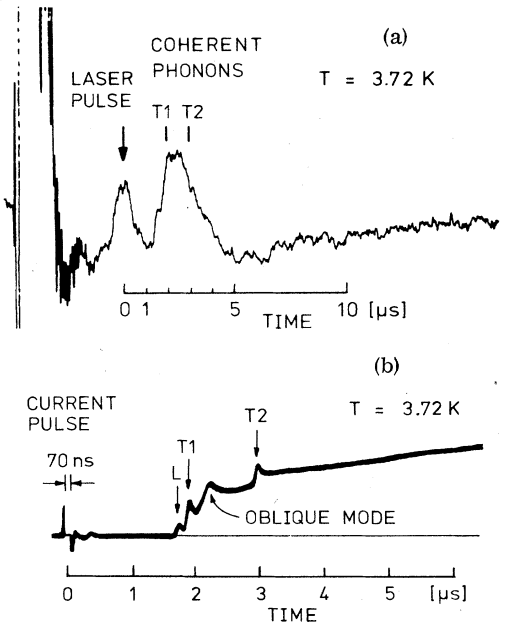

FIG. 1. Bolometer signals of phonon pulses in X-cut quartz. (a) Excitation of coherent phonons by piezoelectric surface excitation using focused 0.891-THz laser pulses. The focal spot at the front face of the crystal is about 2 mm in diameter. The bolometer signals are taken at the rear side of the crystal and are recorded by means of a boxcar integrator. The incident laser beam deviates 17° from the normal to the crystal face. (b) Excitation of incoherent phonons using a thermal Constantan radiator. Radiation temperature is 20 K, corresponding to a peak in the thermal Planck distribution at about 1.2 THz. Radiator and detector area are 1 mm^2. Crystal thickness: 10 mm. Oscilloscope trace.

tion [Fig. 1(a)] the time of flight of the different phonon groups cannot be determined with the present experimental setup with a satisfying time resolution since the exciting laser pulse is still too long and the strong laser-power fluctuations are much too disturbing. The absence of the large ramp which occurs in the case of the incoherent and broad-band excitation of Fig. 1(b) is significant. Only under very favorable conditions could a weak coherent echo signal be observed. This can be understood by the smallness of the expected echo signal due to the small reflection factor at the quartz-tin interface and the strong phonon absorption in tin.

With use of the 2.53-THz radiation of an H_2O laser the bolometer signal of Fig. 2 was observed. In this case, two single-phonon pulses are clearly visible and both are spatially coherent. From the time of flight, group velocities of about 2 and 3.1 km/sec can be deduced. Such low group velocities for ballistically propagating phonons in quartz can not be explained by continuum acoustics and are due to the dispersion for lattice vibrations. Both group velocities are in agreement within experimental error with recent inelastic-neutron-scattering experiments on phonon dispersion in quartz.[16] The first phonon pulse can be identified with phonons of the TA_1 mode with $q = 0.4q_{max}$ ($\lambda_q = 1.8$ nm); the second pulse is due to TA_2 phonons with $q = 0.7q_{max}$ ($\lambda_q = 1.0$ nm). We have verified that the observed coherent phonons are produced by the 2.53-THz emission line by plotting the height of the phonon signal as a function of the laser-cavity length. From this experiment, from the observed spatial coherence, and from the group velocities in connection with the sharpness of the detected pulses we conclude that coherent phonons of 2.53 THz are produced by surface excitation and that they have free path lengths[17] exceeding 10 mm. However, the coherent phonon pulses can only be detected if the exciting laser power is quite low. This is due to the weak, but finite, bulk absorption of photons at this frequency. Each absorbed photon generates two acoustic phonons of half the frequency and opposite wave vectors[18] by a two-phonon process. Hence, photon absorption in the bulk produces (at low absorption rates where spontaneous decay is dominant) incoherent acoustic phonons along the laser beam. Coherent phonons, excited at the front face, meet these high-frequency phonons and can be scattered inelastically by phonon-phonon interaction. Since these interactions increase with the number of phonons produced, at higher laser intensities no coherent phonons are detectable. Under such conditions the large incoherent-phonon signal shows a peak, whose position depends, if the laser beam is moved through the crystal, on the lateral distance between bolometer and laser beam. This peak is due to the time of flight of those incoherent phonons which are generated nearest to the detector. In Fig. 2 the laser power was reduced and chosen for best detection of the coherent phonon pulses.

Figure 3 gives typical phonon signals observed with 10.7-THz radiation. Near this frequency many optical-phonon branches exist in quartz.[19] A direct optical-phonon excitation and direct as well as indirect excitations of two phonons at half

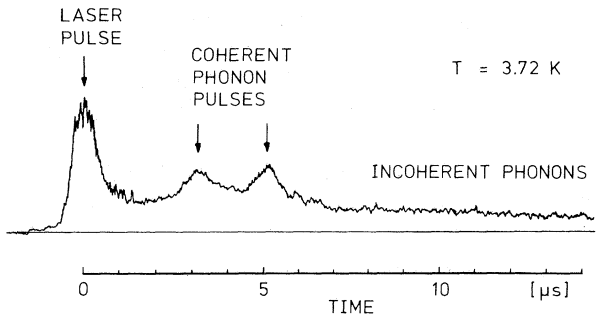

FIG. 2. Phonon generation in X-cut quartz using a focused 2.53-THz laser beam at normal incidence. Spot size at the front of the crystal is about 1 mm in diameter. The bolometer signal is taken at the rear crystal face at a position just opposite to the focal spot at the front face.

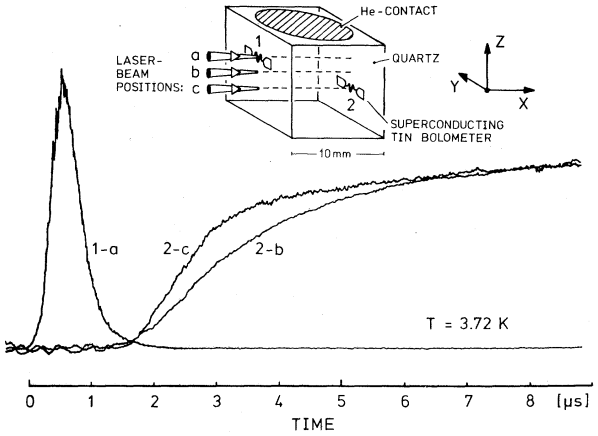

FIG. 3. Phonon generation in X-cut quartz using a focused 10.7-THz laser beam at normal incidence. The position of the laser beam and of the detecting bolometer is indicated in the inset for the three detector signals.

the frequency are possible. Even higher-order phonon processes may occur. A further decay into lower phonon branches follows. The photon absorption is so strong that practically all photons are absorbed within a very thin layer near the surface. In Fig. 3 three different signal traces are shown. If the laser beam meets bolometer 1 and the crystal face between the metal film too, the photon pulse appears at this bolometer and shows in addition a rapidly decaying phonon signal in the tail. The phonon signal of bolometer 2 at the rear of the crystal starts for laser-beam positions *a* and *b* after the time of flight of acoustic long-wavelength phonons ($\tau_L = 1.7$ μsec, $\tau_{T1} = 2$ μsec) and rises to a nearly constant value. Since for times between 2 and 6 μsec more phonons arrive at detector 2 with laser position *c* than with position *b*, there is a pronounced decay in the normal direction to the crystal face. On the other hand it is remarkable that the relatively weak asymptotic phonon signal at the rear face of the crystal lasts longer than 50 μsec, whereas at the front face a bolometer of nearly the same sensitivity shows no noticeable phonon signal at these times. It seems that no homogeneous diffuse phonon field is attainable in the crystal within this time interval. This phenomenon may be explained by a relatively long life time of the generated phonons in combination with an extremely small group velocity. On the other hand the shape of the detection signals is of course also strongly influenced by the expected frequency dependence of phonon reflection, by the change of polarization and phonon decay at the quartz-tin interface, and by the cutoff frequencies of the phonon states in the tin bolometer.

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[17]All experiments were performed with Brasilian natural quartz of the highest quality available.

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