Anharmonic decay of subterahertz coherent acoustic phonons in GaN

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This letter examines the anharmonic decay in GaN of subthermal ($\hbar\omega \ll k_B T$) coherent longitudinal acoustic (LA) phonons with a frequency in the subterahertz range. In a collisionless regime ($\omega\tau > 1$), the anharmonic decay rate of subterahertz coherent LA phonons in GaN shows square dependence on the phonon frequency and cubic dependence on the crystal temperature. As confirmed by the authors experiments, this behavior agrees with a model based on Herring scattering, indicating its importance for the estimation of acoustic attenuation in the subterahertz range. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2433755]

In recent years, terahertz coherent acoustic phonons (CAPs) propagating in crystalline solids have drawn attention in a wide variety of scientific studies focusing on topics such as the spontaneous decay of longitudinal acoustic (LA) phonons,[1] picosecond ultrasonics,[2] nanoultrasonics,[3] and the development of acoustic phonon lasers.[4] However, the terahertz CAPs propagating in the crystals suffer strong attenuation caused by collisions with thermal phonons which degrades and limits the performances of related applications. In crystalline dielectrics, this loss can arise from both extrinsic and intrinsic mechanisms.[5] The extrinsic mechanisms are commonly dominated by phonon-scattering processes at lattice imperfections. When these extrinsic mechanisms are absent, the acoustic loss will ultimately be determined by intrinsic anharmonic decay. For most applications, CAPs are subthermal ($\hbar\omega \ll k_B T$) and the angular frequency ω is usually much smaller than the collision rate $1/\tau_{th}$ of thermal phonons. Therefore, acoustic attenuation due to crystal anharmonicity can be adequately described by macroscopic relaxation damping.[6] However, in studies of picosecond ultrasonics[2] and nanoultrasonics,[3] the frequency of optically initiated acoustic waves is so high that the product of ω and τ_{th} could be much greater than 1.[7] Under such conditions, the attenuation caused by relaxation damping will be independent of phonon frequency.[6] Quantum mechanical, three-phonon processes will emerge to play an important role in the anharmonic decay. This situation has been considered theoretically in Herring's early paper.[8] Damen *et al.* tried to verify this theory with TeO_2,[9] but Tamura *et al.* interpreted it as a case dominated by relaxation damping.[10] As suggested by Tamura *et al.*, the presence of subterahertz acoustic phonons is required to observe the occurrence and behavior of Herring's processes.[7] In our work, we employed InGaN/GaN multiple-quantum well (MQW) samples and a pulse-echo scheme[11] to study the anharmonic decay of acoustic phonons with a frequency up to the subterahertz range. The results not only validate Herring's theory but also reveal the importance of Herring scattering for the estimation of acoustic attenuation in the fields of picosecond ultrasonics and nanoultrasonics.

The sample we used in this study was grown on a 2-μm-thick wurtzite GaN buffer layer on a sapphire substrate by using metal organic chemical vapor deposition (MOCVD). It has ten periods of $In_{0.2}Ga_{0.8}N/GaN$ (3 nm/14 nm) MQW for the generation and narrow-band detection of subterahertz CAPs. On top of the MQW, the sample was capped with a 140-nm layer of unintentionally doped GaN. The femtosecond pulses for the pump-probe measurement came from a mode-locked Ti:sapphire laser. After frequency doubling by a beta-barium borate crystal, it provided 160 fs optical pulses at an above-band-gap wavelength of 390 nm. These 390 nm pulses were split into pump and probe pulses, whose temporal delay was controlled by a stepping stage. Then 84 mW pump pulses and 8 mW probe pulses were focused onto the MQW region by a lens with a 5 cm effective focal distance. To measure the anharmonic decay time at various temperatures, the sample was placed in a cryostat with fused-silica windows on opposite sides for the entrance and exit of laser pulses. The transmitted probe pulses were then collected and detected by a photodetector with lock-in detection.

To distinguish the imperfection-related attenuation from the intrinsic anharmonic decay, we exploited their different responses to changes in the sample's temperature. For imperfection-related loss, the attenuation constant is temperature independent,[12] while the impact of anharmonic decay on attenuation is temperature sensitive. The impact of

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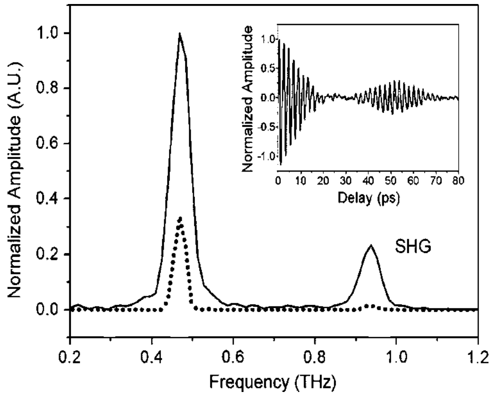

FIG. 1. Differential transient transmission trace of the studied sample with InGaN/GaN MQW at 80 K (inset) and its corresponding power spectra during the generation (solid curve) and echo (dotted curve) phases.

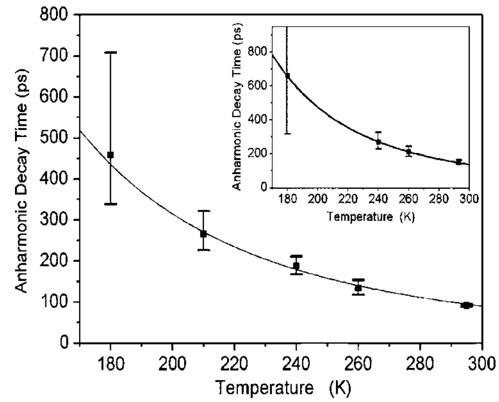

FIG. 2. Anharmonic decay time (solid squares) of 470 and 380 GHz (inset) longitudinal coherent acoustic phonons propagating in GaN at different temperatures. Error bars represent the standard deviation of measured values. Described by Eq. (2) (solid curve), the decay time shows an inverse cubic dependence on temperature.

anharmonic decay becomes negligible when the temperature is so low that the amplitude of thermal lattice vibrations becomes too small to produce significant nonlinear effects through the anharmonicity of the interatomic potential. In that situation, the thermal expansion coefficient, which arises from the anharmonicity of lattice force, is close to zero. For GaN, this critical temperature is around 100 K.[13] Therefore, we took a measurement at 80 K to isolate and measure the imperfection-related decay. The inset of Fig. 1 shows the measured differential transmission pump-probe trace at 80 K. When the pump pulse excited carriers in the MQW, it generated coherent LA phonons propagating along the (0,0,0,1) axis.[11] The wavelength of the CAPs was 17 nm in the MQW region, which is determined by the oscillation period of the MQW.[14] The observed oscillation period was 2.13 ps, which corresponds to an acoustic frequency of 470 GHz and a longitudinal sound velocity $V=8000$ m/s in the MQW. After a round trip in the GaN cap layer, the reflected CAPs from the cap-air interface returned to the MQW starting point. There was a 51.5 ps delay and the CAPs returned with smaller oscillation amplitudes. In order to measure the decay ratio at a specific frequency, we performed a fast Fourier transformation on the generation signal (before 30 ps, in the inset of Fig. 1) and the echo signal (after 30 ps, in the inset of Fig. 1) of the measured CAPs. The normalized power spectrum of the generation phase showed a 470 GHz fundamental peak and a 940 GHz second-harmonic peak (the solid curve in Fig. 1), which result from the asymmetric thickness between the barriers and quantum wells.[14] The power spectrum of the echo phase (the dotted curve in Fig. 1) decreased to 0.337 of the generation phase at 470 GHz, while the echoed second-harmonic peak was suppressed below the noise level. With a 51.5 ps traveling time, this imperfection-related decay corresponds to a 48 ps lifetime.

Then we performed the same measurement at room temperature (295 K), and the decay ratio decreased to 0.193 due to the effect of anharmonic decay. After excluding the imperfection scattering, the anharmonic decay time τ_{anh} for 470 GHz longitudinal CAP was 92 ps. We also varied the crystal temperature T and measured the corresponding τ_{anh}. Figure 2 shows the mean value of the measured τ_{anh} (the solid squares) at different temperatures and the error bar represents the corresponding standard deviation. As T was reduced to 180 K, the average τ_{anh} measured at 470 GHz was found to be as long as 450 ps, with a large measurement standard deviation due to weak signal attenuation during the 51.5 ps traveling time. To further investigate the dependency for τ_{anh} versus angular frequency ω, we studied another sample of ten periods of $In_{0.2}Ga_{0.8}N/GaN$ (3 nm/19 nm) MQW with a 380 GHz longitudinal CAP frequency. We repeated the same measurements. The imperfection-related decay time of acoustic power measured at 85 K was 67.7 ps. Factoring this out, the measured τ_{anh} versus temperature is shown in the inset (the solid squares) of Fig. 2). The τ_{anh} was increased to 147 ps at room temperature.

The temperature and frequency dependences of these measured τ_{anh} can be explained either by relaxation damping or by Herring scattering. Regarding the relaxation damping model, according to the derivation of Tamura *et al.*, the amplitude absorption constant can be described as follows:[6]

$$\alpha(\omega,T)=\frac{C_\nu T}{2\rho v_D^3}[\langle\gamma_{kj}^2\rangle-\langle\gamma_{kj}\rangle^2]\frac{\omega^2\tau_{th}}{1+\omega^2\tau_{th}^2}, \tag{1}$$

where C_v is the constant-volume heat capacity, T is the temperature, ρ is the mass density, v_D is the average of the inverse cubic velocity for thermal phonons, γ_{kj} is the Grüneisen constant of thermal phonons with a wave vector k and a polarization j, and $\langle\cdots\rangle$ denotes an average over the thermal phonon spectrum. The parameter C_v is temperature dependent and can be calculated from the parameters of constant-pressure heat capacity $C_p(T)$, the thermal expansion coefficient $\alpha_v(T)$, the bulk modulus B, and the molar volume V_{mol}.[15] Calculated from related parameters,[15–17] C_v is 33.77 J/mol K at 295 K. The thermal phonon lifetime τ_{th} in Eq. (1) can be calculated from the kinetic formula of thermal conductivity $\kappa=C_v\langle v^2\rangle\tau_{th}/3$, where $\langle v^2\rangle$ is the average of the squared velocity for thermal phonons. Using $\kappa=1.3$ W/cm K and the sound velocity of LA and TA phonons along the z axis,[18,19] the obtained τ_{th} is 4.8 ps. Employing C_v and τ_{th} calculated above with related parameters,[15,19,20] the absorption α is estimated to be 0.047 μm^{-1}, corresponding to a 1.32 ns lifetime of acoustic power at subterahertz phonon frequency. This lifetime is much longer than the τ_{anh} we observed, and the effects of relaxation damping can thus be discounted. Besides, in the subterahertz frequency range, the τ_{th} is so long that $\omega\tau_{th} \gg 1$. Therefore, the anharmonic LA+TA−>TA scattering processes described by Herring[8] must be responsible for the decay. Because the temperatures in our experiments were

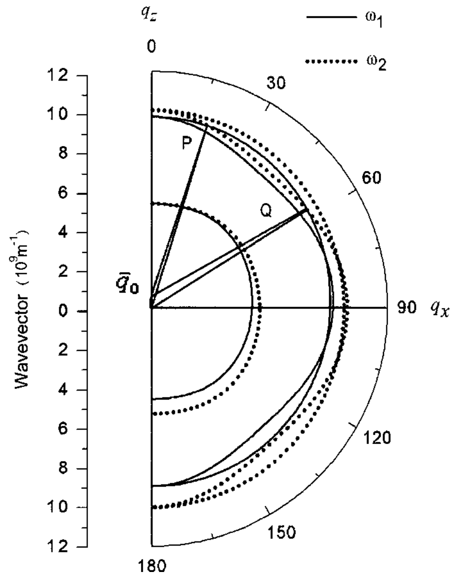

FIG. 3. Conservation surfaces of ω_1 (solid curve) and ω_2 (dotted curve) with the origin of the ω_1 surfaces moved by the wave vector of LA phonons. Point P is related to the occasional intersection close to the q_z direction and point Q is related to the nearly occasional intersection close to the q_x axis.

much lower than the Debye temperature of GaN ($\sim$900 K),[17] the decay rate of subterahertz LA phonons can be formulated as

$$\frac{1}{\tau(\omega,T)} = g\omega^d + c\omega^a T^b. \tag{2}$$

For temperatures below 100 K, only the imperfection-related part $g\omega^d$ remains in Eq. (2). From the measured imperfection-related decay times in both cases, we obtained $g=1.51\times10^{-9}$ and $d=1.64$. The $\omega^{1.64}$ dependence could be attributed to the combination of scattering processes from the boundary roughness, the mass difference, and the threading dislocations commonly observed in MOCVD-grown GaN samples.[12] Then we fit $1/\tau_{\text{anh}}$ by $c\omega^a T^b$ with the least sum of squared error separately for two frequencies (the solid curve in Fig. 2 and in the inset of Fig. 2). The parameter b is 3.2 ± 0.12 for the 470 GHz case and 3.06 ± 0.03 for the 380 GHz case (both are within an acceptable range of error), implying a cubic dependence of $1/\tau_{\text{anh}}$ on the crystal temperature. When the parameter b is set as 3.08, which is acceptable for both cases, the frequency dependence a is 1.98 ± 0.27 and the coefficient c is $(6\pm0.2)\times10^{-23}$. In brief, the $1/\tau_{\text{anh}}$ of LA phonons in wurtzite GaN show $\sim\omega^2T^3$ dependence. This trend satisfies Herring's derivation for the C_{6v} symmetry of wurtzite GaN at temperatures much lower than the Debye temperature.[8] But this happens only for TA phonons close to the occasional intersection points of the conservation surfaces. Just like with the case of TeO_2, the direction of the wave vector of the TA phonons should not be close to the c axis of wurtzite GaN. To make sure that there is another occasional intersection of conservation surfaces between TA branches, we employed related parameters[19,21,22] and plotted the conservation surfaces at the frequency $\omega_1 = k_BT/\hbar$ ($T=295$ K) and at the frequency $\omega_2=\omega_1+\omega_0$ ($\omega_0 = 2\pi\times470$ GHz). Then the required momenta of TA phonons in scattering processes could be determined by the intersection between the conservation surfaces of ω_1 (the solid curve in Fig. 3 and ω_2 (the dotted curve in Fig. 3), with the origin of the ω_1 surfaces moved to $\mathbf{q}_0$, which is the wave vector of 470 GHz LA phonons. As shown in Fig. 3, there are two intersection points P and Q between these two surfaces, close to the q_z axis and the q_x axis, respectively. The scattering processes related to point P do not contribute significantly to the analyzed anharmonic decay of LA phonons propagating along the c axis because, by symmetry principles, the nonlinear parameter in the considered class of crystals is equal to zero for the points exactly on the q_z axis.[7,8,10] We believe that the dominant contribution comes from points of the type Q, which in the limit $\hbar\omega \ll k_BT$ are forming a cone in the space of thermal phonon wave vectors, theoretically[23] contributing $\sim\omega^2T^3$ to acoustic absorption.

In conclusion, we have measured the anharmonic decay time of subterahertz LA phonons in a wurtzite GaN. By changing the temperature of samples and the angular frequency of LA phonons, we found that the anharmonic decay rate showed $\sim\omega^2T^3$ dependence. After factoring out the possible effects of relaxation damping and making sure of the occurrence of Herring scattering, we validated Herring's theory for the scattering processes of subthermal LA phonons. This verification reveals the importance of Herring scattering for the estimation of acoustic attenuation in the fields of picosecond ultrasonics[2] and nanoultrasonics.[3]

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