Microscopic nanomechanical dissipation in gallium arsenide resonators

M. Hamouni,1 P. E. Allain,1 W. Hease,1 L. Morgenroth,2 B. Gérard,3 A. Lemaître,4 G. Leo,1 and I. Favero1

1Matériaux et Phénomènes Quantiques, Université Paris Diderot, CNRS UMR 7162, Sorbonne Paris Cité, 75013 Paris, France
2Institut d’Electronique, de Microélectronique et de Nanotechnologie, UMR CNRS 8520, Avenue Poincaré, 59652 Villeneuve d’Ascq, France
3III-V Lab, 1 Avenue Augustin Fresnel, 91677 Palaiseau, France
4Centre de Nanosciences et de Nanotechnologies, CNRS, Université Paris Sud, Université Paris-Saclay, C2N-Marcoussis, Route de Nozay, 91460 Marcoussis, France

(Dated: January 31, 2018)

We report on a systematic study of nanomechanical dissipation in high-frequency (∼300 MHz) gallium arsenide optomechanical disk resonators, in conditions where clamping and fluidic losses are negligible. Phonon-phonon interactions are shown to contribute with a loss background fading away at cryogenic temperatures (3 K). Atomic layer deposition of alumina at the surface modifies the quality factor of resonators, pointing towards the importance of surface dissipation. The temperature evolution is accurately fitted by two-level systems models, showing that nanomechanical dissipation in gallium arsenide resonators directly connects to their microscopic properties. Two-level systems, notably at surfaces, appear to rule the damping and fluctuations of such high-quality crystalline nanomechanical devices, at all temperatures from 3 to 300 K.

The physical origin of nanomechanical dissipation is a topic of curiosity and debate, motivated by a vast number of applications. Ultra-low dissipation nanomechanical resonators represent a key ingredient for optomechanics, which investigates the interaction of light and mechanical motion [1,2]. They are becoming crucial in weak-force resolution [3,4], mass sensing [5–8], or mesoscopic quantum operations such as ground-state cooling of mechanical motion [9,10] and entanglement between mechanical systems [11]. For example Gallium Arsenide (GaAs) nano-optomechanical disk resonators, whose high-frequency radial breathing modes (RBMs) strongly couple to optical whispering gallery modes (WGMs) [12,13], are expected to display low mechanical dissipation thanks to their constitutive crystalline epitaxial material, and they indeed achieved large Q-frequency products. However, despite the achieved control of clamping losses [14,15], their ultimate mechanical performances are still affected by residual damping processes. The investigation of these processes is the focus of the present work.

In this Letter, specific dissipation channels are made negligible by experimental conditions (vacuum operation that suppresses fluidic damping) or by design (pedestal engineering that suppresses anchoring losses [14,15]), enabling a direct analysis of intrinsic loss mechanisms. These are investigated by comparative measurement of identical resonators made out of two distinct epitaxial wafers, and accurately compared to models of phonon-phonon damping. Surface nanomechanical dissipation is investigated by observing the influence of an atomic layer deposition (ALD) of alumina Al2O3 onto the resonators. The temperature dependence between 3 and 300 K is systematically measured and fitted by two-level systems (TLS) models, allowing the emergence of a microscopic picture of damping processes in GaAs resonators. Our results indicate that TLS dissipation dominate at all temperatures, despite the crystalline nature of the material. By comparing distinct wafers, as well as pristine and surface-treated resonators, we provide evidences about the nature and localization of TLS. Our study finally provides a consistent picture of noise mechanisms affecting high-Q crystalline nanomechanical systems, which are generally regarded as best candidates for quantum applications.

The employed GaAs optomechanical disks have a radius of 5.5 μm, are 200 nm thick, and sit on a 1.8 μm high aluminium gallium arsenide (AlGaAs) pedestal of radius 500 nm. They are measured by optical means. Light is brought to a disk WGM via evanescent coupling to an on-chip suspended waveguide [17], whose endings are shaped into inverted tapers to optimize coupling to micro-lensed fibers. The samples are fabricated from two distinct wafers (1 and 2) grown by molecular beam epitaxy (MBE) under distinct conditions but with the same nominal structure: 200 nm(GaAs)/1.8 μm(Al0.8Ga0.2As)/500 μm(semi-insulating GaAs). The disk and waveguides are first patterned in a negative resist using electron beam lithography. The resist is developed and serves as a mask during the Inductively Coupled Plasma Reactive Ion Etching (ICP-RIE) with a SiCl4/Ar chemistry. The pedestal is under-etched with a hydrofluoric acid solution, and the tips of the inverted tapers are freed using a BCK solution [18]. Fig. 1a shows an electron micrograph of a fabricated device.

In the following, we invariably measure the quality factor $Q_m$ of a mechanical mode or its energy dissipation rate $\Gamma_m = \omega_m/Q_m$, obtained from the full line width of the corresponding resonance. The mechanical
spectrum is measured optomechanically by tuning the laser on the flank of an optical resonance and analyzing the radio-frequency noise of the output light with a fast photo-detector connected to an electronic spectrum analyser [12]. To avoid dynamical optomechanical back-action modifying $\Gamma_m$, [1, 2], the measurements are taken as function of optical power and the linear evolution extrapolated at zero power. We focus here on the first order RBM of the above-discussed disks, which has a frequency of $f_m = \omega_m/2\pi = 260$ MHz and is in our case only subject to intrinsic dissipation channels. Indeed, in this work GaAs disk resonators are operated in a cryostat (accessible range 2.6 to 300 K) and under vacuum ($\leq 10^{-5}$ mbar). At such pressure, the gas damping of the breathing motion is negligible [3, 19]. The dimensions of the disk and pedestal are also chosen to render clamping losses negligible. The latter are simulated numerically by Finite Element Method (FEM), as shown in Fig. 1b, and our previous work in the clamping-limited regime showed good agreement with experiments [14, 15]. We adopt here a disk for which our tolerance on pedestal dimensions bounds clamping losses $\Gamma_m$ to below 3.26 kHz, corresponding to a $Q_m > 5 \cdot 10^5$. In what follows, this channel of dissipation can be neglected, whatever the temperature.

The measurements of $\Gamma_m$ between 3 and 300K are shown in Fig. 2a, for nominally identical resonators fabricated with the exact same process, but out of the two distinct epitaxial wafers (1 and 2). These results reveal two obvious features. Firstly, the intrinsic dissipation tends to increase with temperature, in a similar manner for the two wafers; secondly, the dissipation is larger in wafer 1 than in wafer 2. The temperature evolution of $\Gamma_m$ distinguishes three regimes: (1) Slow increase between 3 K and 150 K (2) Peak around 180 K (3) Quasi-plateau from 200 K to 300 K. The similar behavior of wafers 1 and 2 points towards some universality, whose origin remains to be elucidated. Fluidic and clamping losses being negligible, the dissipation processes must take place in the bulk or at the surface of resonators.

We first analyze the mechanical dissipation induced by interaction of the 260 MHz (mechanical) phonon with high-frequency ($\approx h/k_BT$) thermal phonons. This phonon-phonon damping was discussed in the bulk, using the Landau-Rumer approach (valid when $\omega_m \tau_{ph} \gg 1$) or a Boltzmann equation approach like employed by Akhiezer (valid when $\omega_m \tau_{ph} \ll 1$), where $\tau_{ph}$ is a relaxation time for thermal phonons. The relation between these approaches was discussed by Maris [20]. The Boltzmann equation description assumes thermal phonons to be localized with respect to the mechanical wavelength, a condition implying that $k_BT \gg hf_m$, which is satisfied here where $T > 3 K$. Upon incidence of a mechanical wave (even spatially uniform), the population of thermal phonons is perturbed as consequence of the lattice anharmonicity and dissipates energy via collisions to return to equilibrium. A collision time approximation can be adopted, provided $\omega_m \tau_{ph} < 1$, leading to an expression of this Akhiezer damping [20, 22]:

$$\Gamma_m = \omega_m \cdot \frac{C_p T (\Delta \gamma)^2}{\rho \bar{c}^2} \cdot \frac{\omega_m \tau_{ph}}{1 + (\omega_m \tau_{ph})^2}$$

(1)

where $C_p$ and $\rho$ are the volume specific heat and density, $(\Delta \gamma)^2$ is the variance of the Gr"uneisen parameter over thermal phonons involved in the process, $3/\bar{c}^3 = 1/c_l^3 + 2/c_t^3$ is the mean Debye sound velocity with $c_l$ ($c_t$) the longitudinal (transverse) velocity. The relaxation of thermal phonons occurs both in the bulk and at the resonator’s surface $\tau_{ph} = \tau_{bulk}^{-1} + \tau_{surf}^{-1}$, where $\tau_{bulk} = 3\kappa/C_p \bar{c}^2$ is a temperature dependent relaxation time [22] with $\kappa$ the bulk thermal conductivity, and $\tau_{surf}$ a surface relaxation time governed by the resonator geometry. In the spirit of prior works on micro and nanoscale resonators [24, 25], we adopt the relation $\tau_{surf} = \sqrt{\nu_R/\bar{c}}$.
The Akhiezer prediction of Eq. (1) is reported in Fig. 2b, and accounts for a first part of the phonon-phonon damping. The strain field of the RBM being non-uniform, the anharmonicity of the lattice ($\gamma \neq 0$) additionally induces temperature gradients within the vibrating resonator, leading to irreversible heat flows and dissipation. This thermoelastic damping (TED) \cite{22, 25, 29, 31} can be simulated by FEM, resulting in the extra contribution reported in Fig. 2b, when the temperature dependence of thermal expansion is accounted for \cite{32}. The total phonon-phonon damping is finally plotted in Fig. 2b. It shows an overall increase with temperature, yet with no peak nor plateau. Whatever the temperature, its amplitude is also smaller than in measurements, being negligible for $T < 50$ K and representing a small contribution at higher temperatures. Our models hence indicate that phonon-phonon mechanisms do not govern the dissipation of our nanomechanical resonators. This conclusion is supported by the clear difference in dissipation amplitude between the two wafers shown in Fig. 2a, which points towards material-related effects that need to be elucidated.

In order to investigate the contribution of surfaces, we deposit a 6.5 nm layer of alumina by ALD onto resonators made out of wafer 2, and compare in Fig. 3a the temperature dependence of dissipation before and after ALD treatment. The ALD treatment increases the dissipation at all temperatures: the peak around 180 K vanishes and the plateau-like behavior is replaced by a monotonous increase. The outcome of this trial is that surfaces play an important role in the mechanical dissipation of GaAs nano-resonators. This will be further illustrated in the analysis below.

Indeed the exact temperature dependence of $\Gamma_m$, together with its variation with the employed surface treatment or wafer, can help identifying the microscopic origin of dissipation. With this mindset, we systematically analyse our results with TLS models initially developed for amorphous materials \cite{33, 34}. These models depict microscopic defects and configurations as potential energy double-wells (Fig. 3b) with the following parameters: the asymmetry $\Delta$, the barrier height $V_0$, or the well-to-well tunneling amplitude $\Delta_0 \approx 2E_0/\pi \cdot \exp\left[-d (mV_0/2\hbar^2)^{1/2}\right]$, with $d$ the separation between wells and $E_0$ the ground state energy of a single well \cite{33}. For a TLS distribution $P(\Delta, \Delta_0)$, the dissipation rate is in general given by \cite{33}:

$$
\Gamma_m = \omega_m \cdot \frac{2\eta^2 N_{TLS}}{\rho c_T^2 k_B T} \int \int_{\mathbb{R}^+} d\Delta d\Delta_0 \frac{\Delta^2}{E^2} \text{sech}^2 \left( \frac{E}{2k_B T} \right) \frac{\omega_m \tau}{1 + (\omega_m \tau)^2} P(\Delta, \Delta_0)
$$  \hspace{1cm} (2)

with $\eta$ the deformation potential coupling of a TLS to the mechanical phonon (in eV), $N_{TLS}$ the density of TLS, $\rho$...
and $c_s$ the density and sound velocity of the material, $E = \sqrt{E_0^2 + \Delta^2}$ and $\tau$ the TLS relaxation time. For $T \geq 10$ K, several energy levels of the wells are typically populated. In this so-called thermally activated regime, the relaxation time is given by a quasi-Arrhenius law $\tau = \tau_0 e^{\frac{\Delta E}{k_B T}} \text{sech} \left( \frac{\Delta}{2k_B T} \right)$ where $\tau_0^{-1}$ is of the order of the Debye frequency of the material [33], and $\Delta \approx E$, leading a simplified expression of the dissipation [36], where a distribution $P(\Delta, V_0)$ is used equivalently.

In order to fit the measured temperature-dependent dissipation, we consider two distinct distributions introduced in the literature [36, 38] and sum up their contribution using the linearity of Eq. (2). The single defect distribution (1) $P(\Delta', \Delta_0') = \delta(\Delta - \Delta') \cdot \delta(\Delta_0' - \Delta_0)$ considers both $\Delta$ and $\Delta_0$, and hence $E$, as fixed, with $\delta$ the Dirac function. This distribution assumes a single type of TLS, and makes use of an alternative deformation potential $D = \frac{2}{3} \eta$ for conciseness. The amorphous distribution (2) $P(\Delta, V_0) = f(\Delta) \cdot g(V_0)$ where $f$ and $g$ are Gaussian functions in $\Delta$ and $V_0$, with mean value 0 and width $\Delta_1$ and $V_1$ [36]. This model is typically used for amorphous materials where a broad distribution of TLS couple to acoustic waves. In Fig. 3c, these two contributions sum-up with the prior phonon-phonon contribution to reproduce the mechanical dissipation measured on resonators of wafer 2 in the range $T \geq 10$ K, with TLS parameters shown in Table I. We note that the dissipation below 10 K is difficult to model using the thermally activated regime, such that the two lowest temperature points are fitted with a coherent version of the relaxation time [33, 37] $\tau^{-1} = \frac{\pi^2 \Delta^2 E}{2k_B c_s}$ \text{coth} \left( \frac{E}{2k_B T} \right)$. The obtained level of agreement sheds light on the microscopic nature of dissipation. The mechanical damping up to 100 K is well explained by an amorphous TLS model, which suggests a role of the surface reconstruction layer, whose amorphous nature was observed by transmission electron microscopy [39]. In order to model the dissipation at higher $T$, the single defect model must however be used on top. The related defect has an activation energy of $\approx 0.1$ eV, consistent for example with GaAs $DX$ centers [40]. For a deformation potential $\approx 10$ eV, the inferred density of TLS is of $\approx 10^{19}$ cm$^{-3}$, well above the unintentional doping of our epitaxial GaAs ($10^{14}$ cm$^{-3}$ range), suggesting the formation of TLS at the surfaces in a density superior to the bulk. The localization of TLS can further be investigated by looking at ALD-treated resonators and resonators fabricated out of wafer 1 (see Table I). The ALD surface treatment appears to modify both the amorphous and single defect distribution, indicating again that both types of TLS can be at surfaces. It enhances the density of amorphous TLS, which is consistent with the amorphous nature of deposited alumina; but decreases the density of single defect TLS, which is reminiscent of the passivation action of ALD [16]. The superior mechanical dissipation in wafer 1 compared to 2 seems to originate from a different density of amorphous TLS, which is again consistent with their localization at surfaces, since surface absorption of photonic resonators fabricated out of wafer 1 was also superior to that of wafer 2 [16].

In summary, we have reported a systematic study of intrinsic nanomechanical dissipation in GaAs resonators. Microscopic models indicate that two-level systems dominate damping at any temperature between 3 and 300 K. While in conflict with the common sense that crystalline devices are less affected by TLS than their amorphous counterparts [11, 12], this conclusion is consistent with the presence of an amorphous reconstruction layer at their surface. Such a layer already rules the optical dissipation of high-$Q$ GaAs resonators with large surface to volume ratio [39], and we bring here a series of evidences that TLS impacting their nanomechanical dissipation mainly localize at surfaces as well. Our models anticipate that the freezing of these fluctuating TLS would be beneficial, predicting a mechanical quality factor $Q_m$.

FIG. 3. (a) Intrinsic mechanical dissipation before and after ALD of 6.5 nm of Al$_2$O$_3$. (b) Double-well model for TLS. (c) Modeling of the intrinsic mechanical dissipation in wafer 2 using phonon-phonon interactions and TLS models.
beyond $10^9$ at 10 mK. The related Q-frequency product $Q_m \times f_m$ would reach the $10^{17} - 10^{18}$ range for GaAs resonators, equaling the performances of other crystalline devices in quartz [43] and silicon [44]. Ultra-low temperature experiments, possibly below the milliKelvin, may ultimately reveal how far the performances of nanomechanics can be pushed, for metrological and quantum applications.

This work was supported by the European Research Council (ERC) through the Ganoms project (No.306664). The authors thank Bernard Perrin and Eddy Collin for fruitful comments.

[1] I. Favero and K. Karrai, Nature Photonics 3, 201 (2009).
[2] M. Aspelmeyer, T. J. Kippenberg, and F. Marquardt, Reviews of Modern Physics 86, 1391 (2014).
[3] H. Miao, K. Srinivasan, and V. Aksyuk, New Journal of Physics 14, 075015 (2012).
[4] A. G. Krause, M. Winger, T. D. Blasius, Q. Lin, and O. Painter, Nature Photonics 6, 768 (2012).
[5] J. Tamayo, P. M. Kosaka, J. J. Ruz, A. San Paulo, and M. Calleja, Chemical Society Reviews 42, 1287 (2013).
[6] F. Liu, S. Aliae, Z. Leseman, and M. Hosseinzadeh, Optics Express 21, 19555 (2013).
[7] E. Gil-Santos, C. Baker, D. T. Nguyen, A. Lemaitre, C. Gomez, S.ucci, G. Leo, and I. Favero, Nature Nanotechnology 10, 810 (2015).
[8] E. Gil-Santos, C. Baker, D. T. Nguyen, W. Hease, C. Gomez, A. Lemaitre, S.ucci, G. Leo, and I. Favero, in Proceedings of the 29th International Conference on Micro Electro Mechanical Systems (IEEE MEMS) (IEEE, 2016) pp. 238–241.
[9] J. Teufel, T. Donner, D. Li, J. Harlow, M. Allman, K. Ci-cak, A. Sirois, J. D. Whittaker, K. Lehnert, and R. W. Simmonds, Nature 475, 359 (2011).
[10] R. Peterson, T. Purdy, N. Kampel, R. Andrews, P.-L. Yu, K. Lehnt, and C. Regal, Physical Review Letters 116, 063601 (2016).
[11] K. Berkje, A. Nunnakamp, and S. M. Girvin, Phys. Rev. Lett. 107, 123601 (2011).
[12] J. C. L. Ding, C. Baker, A. Andronico, D. Parrain, P. Senellart, A. Lemaitre, S.ucci, G. Leo, and I. Favero, “Gallium arsenide disk optomechanical resonators,” in Handbook of Optical Microcavities (Pan Stanford, 2014) p. 381.
[13] I. Favero, “Gallium arsenide disks as optomechanical resonators,” in Cavity Optomechanics (Springer, 2014) pp. 149 – 156.
[14] D. T. Nguyen, C. Baker, W. Hease, S. Sejil, P. Senellart, A. Lemaitre, S.ucci, G. Leo, and I. Favero, Applied Physics Letters 103, 241112 (2013).
[15] D. T. Nguyen, W. Hease, C. Baker, E. Gil-Santos, P. Senellart, A. Lemaitre, S.ucci, G. Leo, and I. Favero, New Journal of Physics 17, 023016 (2015).
[16] B. Guha, F. Marsault, F. Cadiz, L. Morgenroth, V. Ulin, V. Berkovitz, A. Lemaitre, C. Gomez, A. Mo, S. Com-brié, et al., Optica 4, 218 (2017).
[17] C. Baker, C. Belacel, A. Andronico, P. Senellart, A. Lemaitre, E. Galopin, S.ucci, G. Leo, and I. Favero, Applied Physics Letters 99, 151117 (2011).
[18] W. Hease, Gallium Arsenide disk optomechanical resonators approaching the quantum regime, Ph.D. thesis, Paris Diderot University (2016).
[19] S. S. Verbridge, H. G. Craighead, and J. M. Parpia, Applied Physics Letters 92, 013112 (2008).
[20] H. J. Maris, “Interaction of sound waves with thermal phonons in dielectric crystals,” in Physical Acoustics, Vol. 8 (Academic Press, 1971) pp. 279 – 345.
[21] H. Maris, Physical Review 175, 1077 (1968).
[22] B. Perrin, Physical Review B 24, 6104 (1981).
[23] A. Cleland, Foundations of Nanomechanics (Springer, 2003).
[24] W. Fon, K. Schwab, J. Worlock, and M. Roukes, Physical Review B 66, 045302 (2002).
[25] K. Kunal and N. Aluru, Physical Review B 84, 245450 (2011).
[26] J. Blakemore, Journal of Applied Physics 53, 123 (1982).
[27] R. Cottam and G. Saunders, J. Phys. C Solid State Phys. 6, 2105 (1973).
[28] R. Cottam and G. Saunders, J. Phys. C Solid State Phys. 7, 2447 (1974).
[29] G. Zener, Physical Review 53, 90 (1938).
[30] R. Lifsitz and M. Roukes, Physical Review B 61, 5600 (2000).
[31] A. Kiselev and G. Iafrate, Physical Review B 77, 115 (1980).
[32] W. A. Phillips, Reports on Progress in Physics 50, 1657 (1987).
[33] D. Tielburger, R. Merz, R. Ehrenfels, and S. Hunklinger, Physical Review B 45, 2750 (1992).
[34] S. Rau, C. Ens, S. Hunklinger, P. Neu, and A. Würger, Physical Review B 52, 7179 (1995).
[35] R. Vacher, B. Courtenes, M. Foret, Physical Review...
[37] W. A. Phillips, Physical Review Letters 61, 2632 (1988).
[38] S. Hunklinger and W. Arnold, “Ultrasonic properties of glasses at low temperatures,” in Physical Acoustics, Physical Acoustics, Vol. 12 (Academic Press, 1976) pp. 155 – 215.
[39] D. Parrain, C. Baker, G. Wang, B. Guha, E. Gil Santos, A. Lemaître, P. Senellart, G. Leo, S. Ducci, and I. Favero, Optics Express 23, 19656 (2015).
[40] K. Yamanaka, S. Naritsuka, K. Kanamoto, M. Mihara, and M. Ishii, Journal of Applied Physics 61, 5062 (1987).
[41] T. S. Faust, J. Rieger, M. J. Seitner, J. P. Kotthaus, and E. M. Weig, Phys. Rev. B 89, 100102 (2014).
[42] L. G. Villanueva and S. Schmid, Phys. Rev. Lett. 113, 227201 (2014).
[43] M. Goryachev, D. L. Creedon, E. N. Ivanov, S. Galliou, R. Bourquin, and M. Tobar, Applied Physics Letters 100, 243504 (2012).
[44] S. Meenehan, J. D. Cohen, G. S. MacCabe, F. Marsili, M. Shaw, and O. Painter, Phys. Rev. X 5, 041002 (2015).