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Tuning photoacoustics with nanotransducers via Thermal Boundary Resistance and Laser Pulse Duration

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The photoacoustic effect in liquids, generated by metal nanoparticles excited with short laser pulses, offers high contrast imaging and promising medical treatment techniques. Understanding the role of the thermal boundary resistance (TBR) and the laser pulse duration in the generation mechanism of acoustic waves is essential to implement efficient photoacoustic nanotransducers. This work theoretically investigates, for the paradigmatic case of water-immersed gold nanocylinders, the role of the TBR and of laser pulse duration in the competition between the launching mechanisms: the thermophone and the mechanophone. In the thermophone, the nanoparticle acts as a nanoheater and the wave is launched by water thermal expansion. In the mechanophone, the nanoparticle directly acts as a nanopiston. Specifically, for a gold-water interface, the thermophone prevails under ns light pulse irradiation, while the mechanophone dominates shortening the pulse to the 10 ps regime. For a graphene-functionalized gold-water interface, instead, the mechanophone dominates over the entire range of explored laser pulse durations. Results point to high-TBR, liquid-immersed nanoparticles as potentially efficient photoacoustic nanogenerators, with the advantage of keeping the liquid environment temperature unaltered.

Nanoscale photoacoustics generation in liquids, owing to its potential in nanoimaging and therapeutic applications, is a flourishing topic at the cross-road of condensed matter physics, nanomedicine and material science¹⁻⁵. In this context, liquid-immersed metal nanoparticles have proven to be efficient photoacoustic generators due to their tunable optical absorption properties⁶⁻⁹, high contrast imaging features^{10,11} and biocompatibility^{12,13}. Great efforts have been devoted to optimise the parameters allowing a more efficient photoacoustic conversion, such as size, geometry^{9,14-16} and transducer materials¹⁷. Yet, despite its relevance for applications, the combined effects of the pulse temporal width, τ_L ,¹⁸⁻²⁰ and the thermal boundary resistance²¹⁻²³ (TBR) tunabilities remain relatively unexplored and lack a thorough rationalization.

In brief, the photoacoustic effect of an individual liquid-immersed metal nanoparticle consists of three steps: (i) absorption of the laser pulse by the nanoparticle and its temperature rise, (ii) thermal interaction between the nanoparticle and the liquid environment and (iii) generation of the acoustic wave in liquid. The acoustic wave in liquid is triggered by two launching mechanisms: the mechanophone and the thermophone effects. The former is due to the thermal expansion of the metal nanoparticle, the latter mechanism to that of the liquid environment with the nanoparticle acting as a nanoheater. The photoacoustic generation in these systems is typically investigated under ns laser pulses irradiation,

in which case the mechanophone contribution is, in most instances, disregarded^{18,24-27}. Nevertheless, the mechanophone effect cannot be neglected in general, as recently demonstrated for the case of carbon nanotubes immersed in water²⁰. Once the size and composition of the nanoparticles and of the surrounding liquid are fixed by the specific application constraints, the relative contributions of the thermophone vs

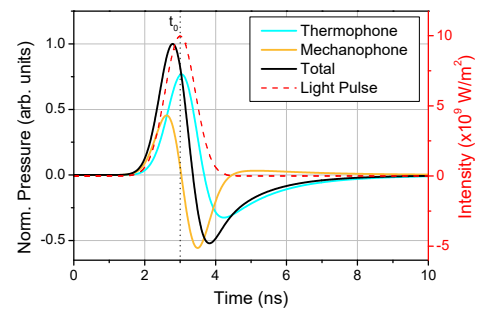


FIG. 1. Left axis: pressure time evolution in water at $r = R_{gnc} + 5$ nm for $\tau_L = 1$ ns, $\mathcal{R} = 1 \times 10^{-7}$ m²K/W. The curves are normalized to the total pressure maximum. Right axis: $I(t)$ (red dashed curve).

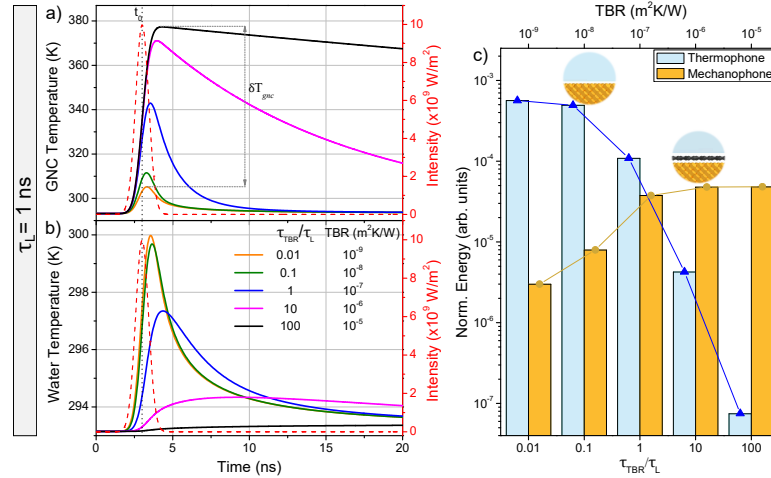


FIG. 2. Laser pulse: $\tau_L = 1$ ns. Panels (a,b): temperature time evolution, for increasing values of τ_{TBR}/τ_L and the corresponding TBR values, in the GNC ($r = 0$ nm): panel (a); in proximal water ($r = R_{gnc} + 5$ nm): panel (b). Right axis: $I(t)$ (red dashed line) maximized at the time t_0 . δT_{gnc} : difference between the GNC peak temperatures between the cases of $\tau_{TBR}/\tau_L \sim 100$ and 0.01. Panel (c): normalized mechanical energy generated in water by the thermophone and the mechanophone effects for different τ_{TBR}/τ_L (bottom axis) and the corresponding TBRs (top axis). The ratios τ_{TBR}/τ_L are rounded to the first significant figure. Values for Au/water and graphene-functionalized Au/water interface are identified by the two round sketches.

mechanophone effects may be tuned upon varying the TBR at the nanoparticle/liquid interface (interface engineering) and τ_L , their interplay making the focus of the present work.

We theoretically investigate the role of the thermophone and mechanophone effects in acoustic wave generation for the paradigmatic case of a water-immersed gold nanocylinder (GNC) of radius $R_{gnc} = 10$ nm and of high aspect ratio, because of their relevance in bio-medical applications^{28–34}. Formally, the GNC is assumed infinitely extended along its axis, the problem thus being radially symmetric with r the radial coordinate. The model, detailed in SI, comprises three steps: optics, thermics and mechanics. As for the optics, the system, at equilibrium at $t = 0$ s, is excited with a laser pulse at 530 nm wavelength, i.e. within water transparency window. The light intensity (W/m^2) has a Gaussian intensity profile, $I(t) = 2\sqrt{\frac{\ln(2)}{\pi}} \frac{\Phi}{\tau_L} \exp[-4\ln(2)\frac{(t-t_0)^2}{\tau_L^2}]$, where $\Phi = 10 \text{ J/m}^2$ is the pulse fluence, which is kept constant for all τ_L values, with $\tau_L = 1$ ns, 100 ps, 10 ps its temporal full-width-half-maximum, while the pulse temporal peak occurs at $t_0 = 3\tau_L$. Via the GNC absorption cross section we then calculate the absorbed power density. The latter serves as the source term for the thermics. The temperature $T(r, t)$ throughout the system (both GNC and water) is then obtained solving the thermal diffusion equation while imposing the continuity of the heat flux at the GNC/water interface, $\hat{q} = \frac{1}{\mathcal{A}} [T(R_{gnc}^-, t) - T(R_{gnc}^+, t)] \hat{r}$, which is controlled by the TBR, \mathcal{R} , with $T(R_{gnc}^\pm, t)$ the temperature at the inner (-) and outer (+) side of the interface and \hat{r} its normal vector. $T(r, t)$ serves as the source terms for the

mechanics via the thermal expansion coefficients of both the GNC and water, ultimately yielding the pressure, $p(r, t)$, and the radial velocity field, $v_r(r, t)$ in water. With p and v_r at hand, the acoustic Poynting vector, \mathbf{P} (W/m^2), and from it the mechanical energy radiated in water, U , are retrieved. The thermophone and mechanophone contribution to the total $p(r, t)$ and U are calculated forcing to zero the GNC and water thermal expansion coefficients, respectively.

The first conclusion that can be drawn from simulations results is that the mechanophone effect needs to be accounted for. Fig.1 shows $p(t)$ in water, 5 nm from the GNC/water interface, together with the thermophone and mechanophone contributions. Results are for the case of $\tau_L = 1$ ns and $\mathcal{R} = 1 \times 10^{-7} \text{ m}^2\text{K/W}$. The latter is representative of the general cases that might be encountered: its order of magnitude falls between that of the Au/water, $1 \times 10^{-8} \text{ m}^2\text{K/W}$ ^{18,35–37}, and that of the graphene-functionalized Au/water interface³⁸, $1 \times 10^{-6} \text{ m}^2\text{K/W}$. The thermophone and the mechanophone contributions have similar amplitudes, thus both contributing to the total pressure signal.

We now address the role played by the TBR and τ_L in the relative contribution between these two launching mechanisms. On the thermal side, upon absorption of the laser pulse, the GNC raises its temperature on a time scale τ_L . It cools down on a time scale τ_h transferring heat to the proximal water and raising the temperature of the latter. Finally, the GNC and the proximal water diffuse heat to distant water, relaxing to the initial temperature. The timescale τ_h has contributions from the TBR and heat diffusion effects arising from the GNC

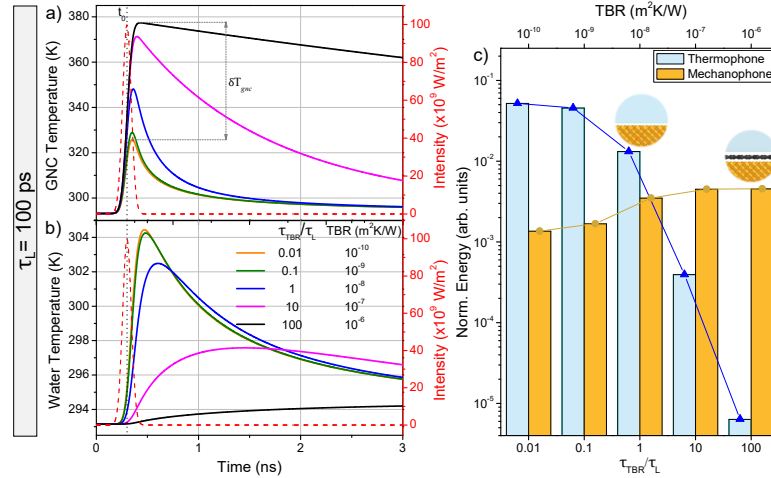


FIG. 3. Laser pulse: $\tau_L = 100$ ps. Same caption as for Fig.2. The TBR values are varied so as to span the same values of τ_{TBR}/τ_L as for the 1 ns pulse case.

and proximal water thermal impedances³⁹. We now discuss what might be intuitively foreseen in the two extreme-case scenarios.

For $\tau_{th} \gg \tau_L$, energy from the laser pulse is delivered to the GNC on a time scale τ_L , and, only after a time $\sim \tau_{th}$, the GNC temperature decreases substantially, while delivering heat to the proximal water. That is, on a time scale τ_{th} we should expect a high-temperature GNC, thermally isolated from the surrounding water still at its ambient temperature. On the mechanics side, the thermal expansion of the GNC should be at its maximum. On the contrary, the contribution of water thermal expansion should be at a minimum and set in on a timescale exceeding τ_{th} . The relevance of the mechanophone effect with respect to the thermophone should thus be highest for cases in which $\tau_{th} \gg \tau_L$.

For $\tau_{th} \ll \tau_L$ the situation is the opposite. The laser feeds energy to the GNC on a time scale τ_L , whereas the GNC delivers energy to the proximal water on a much faster time scale, τ_{th} : the GNC absorbs energy from the laser pulse and concomitantly delivers it to the proximal water. In this scenario the peak GNC temperature should be at its minimum, whereas the proximal water temperature should reach its greatest value. Accordingly, on the mechanics side, the peak thermal expansion of the GNC should be at its minimum, and that of proximal water at its maximum. The relevance of the mechanophone effect with respect to the thermophone should thus be lowest for $\tau_{th} \ll \tau_L$.

In first instance, the ratio τ_{th}/τ_L therefore appears as a meaningful metric to inspect the thermophone to mechanophone transition. The TBR is though the *only* material parameter that can be tuned⁴⁰⁻⁵⁶, the thermal properties of the GNC and water being fixed. Under a practical stand-point it is therefore desirable to parameterize the prob-

lem in terms of a thermal decay time linked to the TBR only, rather than to τ_{th} , which comprises also the effect of proximal water and GNC thermal impedances. To this end, we link the TBR to the thermal decay time through the expression $\tau_{TBR} = \mathcal{R} R_{gnc} c_p \rho / 2$, with c_p and ρ the Au specific heat and mass density, respectively. This relation is somewhat approximate⁵⁷, nevertheless, it has the merit of providing a rule-of-thumb estimate. In the following, we therefore parameterize simulations results in terms of τ_{TBR}/τ_L rather than τ_{th}/τ_L . We now inspect what happens varying the TBR for a fixed laser pulse duration.

Nanosecond regime. Fig.2 shows the GNC (panel (a)) and the proximal water (panel (b)) temperature dynamics. The curves are calculated for a fixed value of $\tau_L = 1$ ns while varying the TBR, from 10^{-5} to 10^{-9} m^2K/W , so as to cover the range of τ_{TBR}/τ_L from 10^{-2} to 10^2 . For increasing τ_{TBR}/τ_L the GNC maximum temperature, $\max\{T_{gnc}(t)\}$, increases from 305 K to 377 K, whereas that of proximal water, $\max\{T_w(t)\}$, decreases from 300 K to 293 K.

The implications of the thermal problem on the competition between the thermophone and mechanophone contributions are shown in panel (c). The histogram shows the mechanical energy radiated in water by the sole thermophone (azure) and sole mechanophone (mustard) effects as a function of τ_{TBR}/τ_L (bottom axis) and TBR (top axis). Energies are normalized to the maximum mechanical energy observed in water in our simulations (i.e. the thermophone contribution of the $\tau_L = 10$ ps, $\tau_{TBR}/\tau_L = 10^{-11}$ case that will be described later). For increasing values of the TBR, acoustic wave generation in water switches from thermophone-dominated for $\tau_{TBR}/\tau_L \lesssim 1$, to mechanophone-dominated for $\tau_{TBR}/\tau_L \gg 1$, $\tau_{TBR}/\tau_L \approx 1$ being a cross-over value between the two regimes.

In real case scenarios, the TBR can be tuned engineering the

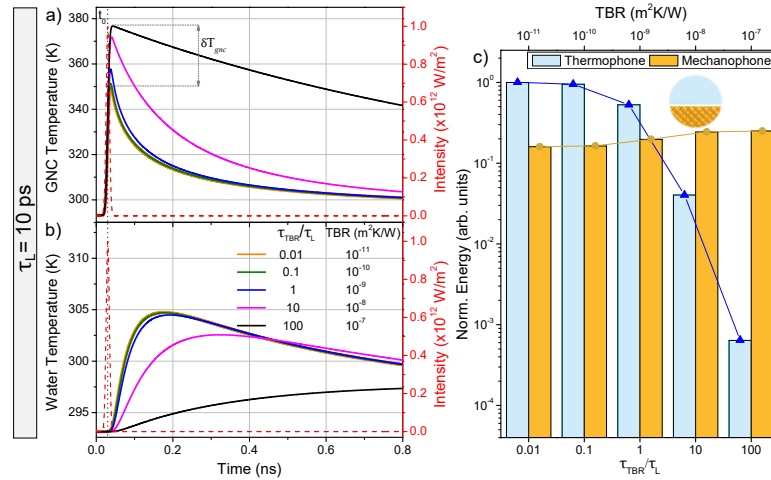


FIG. 4. Laser pulse: $\tau_L = 10$ ps. Same caption as for Fig.2. The TBR values are varied so as to span the same values of τ_{TBR}/τ_L as for the 1 ns pulse case. In panel (c) the case of the graphene-functionalized Au/water interface is here dominated by the mechanophone effect and is hence not reported.

GNC/water interface, and the acoustic wave launching mechanism accordingly, switching for instance from the thermophone for the bare Au/water interface, to the mechanophone for the graphene-functionalized Au/water interface, which cases are indicated by the two inset sketches. So far we spanned τ_{TBR}/τ_L for a fixed value of τ_L while varying the TBR. The question then arises as to whether a similar physics holds true also for shorter laser pulses, thus eventually allowing for an additional knob, τ_L , to select the launching mechanism.

Picosecond regime. Fig.3 and Fig.4 are analogous to Fig.2 but for the cases of $\tau_L=100$ ps and 10 ps, respectively. For sake of comparison, the TBR is now varied from 10^{-6} to 10^{-10} m 2 K/W and from 10^{-7} to 10^{-11} m 2 K/W for the case $\tau_L=100$ ps and 10 ps, respectively. These changes allow covering the same range of τ_{TBR}/τ_L as for the case of the 1 ns laser pulse. On the thermal side, Fig.3(a,b) and Fig.4(a,b) encompass the general trend observed in Fig.2(a,b): for increasing τ_{TBR}/τ_L , $\max\{T_{gnc}(t)\}$ increases whereas $\max\{T_w(t)\}$ decreases. While reducing τ_L , a relevant parameter for the following discussion is δT_{gnc} , defined as the difference between the GNC peak temperatures between the cases of $\tau_{TBR}/\tau_L=10^2$ and 10^{-2} . δT_{gnc} ranges from 72 K for $\tau_L=1$ ns, to 27 K for $\tau_L=10$ ps, because of the increase of the peak temperature for the case of $\tau_{TBR}/\tau_L=10^{-2}$ while transitioning from Fig.2(a), across Fig.3(a) to Fig.4(a). Among the physical reasons behind this trend, is that reducing τ_L for a fixed value of $\tau_{TBR}/\tau_L=10^{-2}$ implies reducing the TBR, eventually to a point where the interfacial heat transfer is no more the limiting process, the GNC and the proximal water thermal impedances⁵⁸ as the only factors controlling the thermal dynamics⁵⁸.

The implications of the thermal problem on the competition between the thermophone and mechanophone contributions are shown in panel (c) of Fig.3 and Fig.4, for $\tau_L=100$ ps and 10 ps, respectively. The striking difference, comparing Fig.2(c), 3(c) and 4(c), is that the mechanophone contribution dependence on τ_h/τ_L weakens substantially upon reducing the pulse temporal width; not so for the thermophone contribution. Indeed, for the $\tau_L=1$ ns case, the mechanophone contribution changes by more than an order of magnitude when increasing τ_{TBR}/τ_L from 10^{-2} to 10^2 , whereas it changes by a factor of ~ 2 for the $\tau_L=10$ ps case. The physical reason stands in the thermal problem. The mechanophone effect is triggered by the GNC thermal expansion. The shorter the laser pulse, the smaller is δT_{gnc} , implying that the peak GNC temperature becomes rather uniform regardless of τ_{TBR}/τ_L , and the GNC thermal expansion accordingly.

When exciting with a 100 ps laser pulse, the mechanophone effect contribution to the radiated acoustic energy in water raises to 23% for the Au GNC/water interface, and dominates the graphene-functionalized GNC/water interface, see Fig.3(c). Further reducing the pulse duration to 10 ps, the mechanophone effect becomes the prevailing mechanism also for the Au GNC/water interface, see Fig.4(c).⁵⁹

In conclusion, we showed, for the case of a water-immersed Au nanocylinder, that the TBR and the laser pulse duration are two valuable control knobs, allowing to switch the acoustic wave launching mechanism in water from the thermophone to the mechanophone. The Au/water and graphene-functionalized Au/water interfaces were discussed as realistic show-case scenarios. Importantly, when the mechanophone is the dominant launching effect, the surrounding water temperature increase is minimized. These findings thus bare particu-

lar importance in situations requiring high frequency acoustic wave generation in water (i.e. short τ_L) while avoiding heating effects of the latter, as is the case for in-vivo bioimaging and theranostics applications at the nanoscale. Noteworthy, these findings may be expanded to include other nanosystems^{60,61}.

SUPPLEMENTARY MATERIAL

See the supplementary material for the details on the simulations design.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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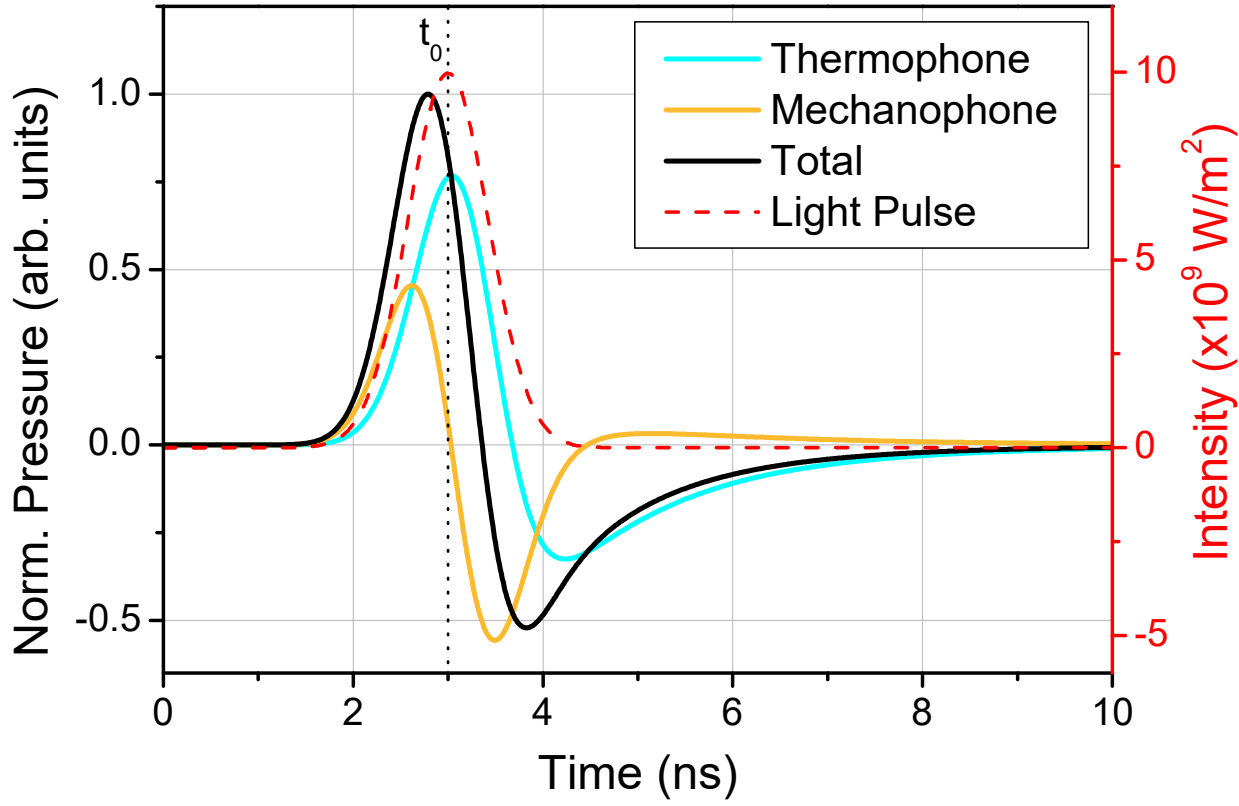
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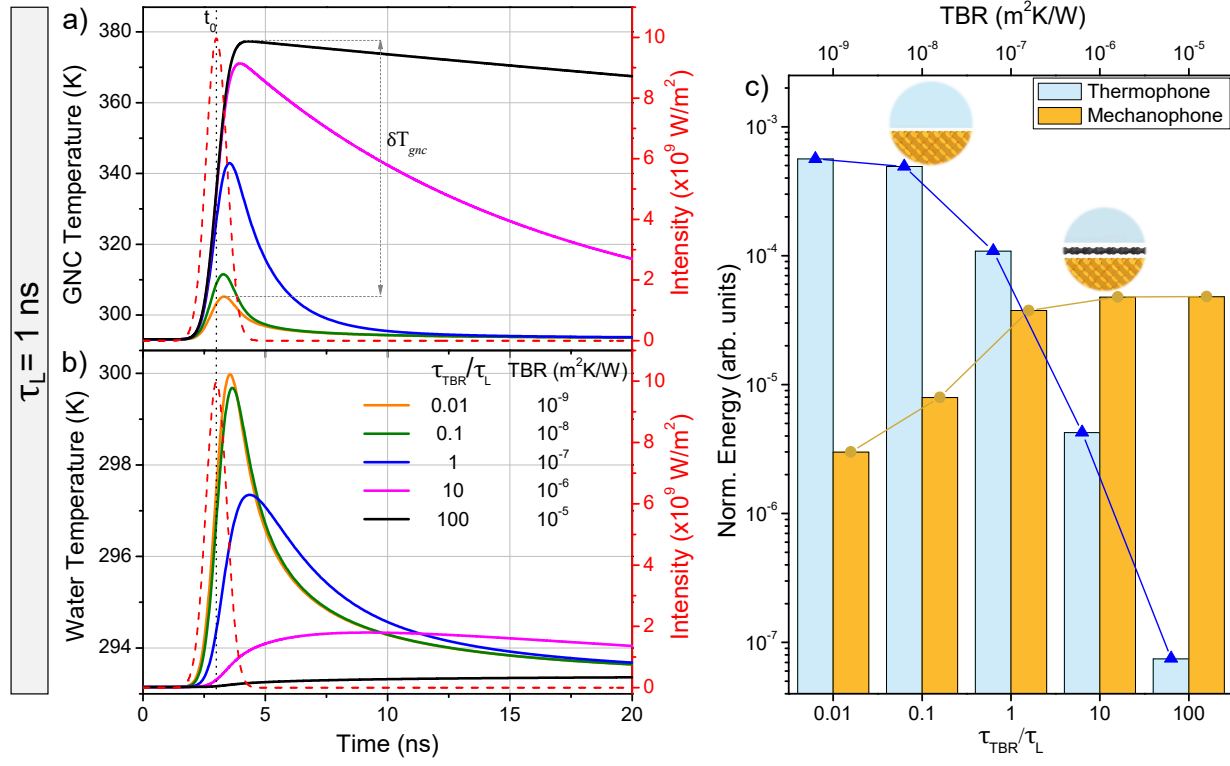
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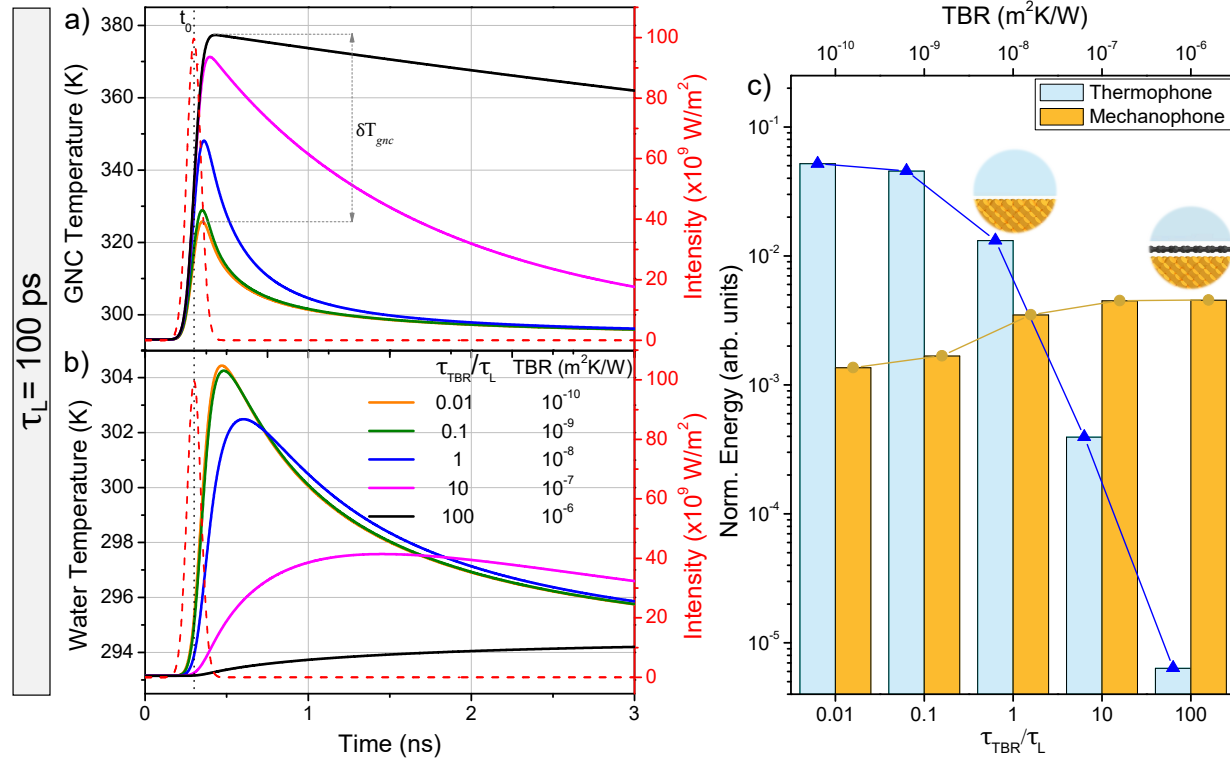
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