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PAPER

Generation and detection of squeezed phonons in lattice dynamics by ultrafast optical excitations

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We propose a fully quantum treatment for pump and probe experiments applied to the study of phonon excitations in solids. To describe the interaction between photons and phonons, a single effective hamiltonian is used that is able to model both the excitation induced by pump laser pulses and the subsequent measuring process through probe pulses. As the photoexcited phonons interact with their surroundings, mainly electrons and impurities in the target material, they cannot be considered isolated: their dynamics needs to be described by a master equation that takes into account the dissipative and noisy effects due to the presence of the environment. In this formalism, the quantum dynamics of pump excited phonons can be analyzed through suitable probe photon observables; in particular, a clear signature of squeezed phonons can be obtained by looking simultaneously at the behavior of the scattered probe mean photon number and its variance.

1. Introduction

An effective optical experimental technique suitable for the study of the dynamics of phonon excitations in solids is pump and probe spectroscopy [8, 9, 21, 26]. It consists in exciting a crystal with a high intensity laser pulse, usually referred to as the pump; the resulting phonon excitations are studied by means of a second, less intense laser pulse, called the probe, that hits the material after a certain time delay. By analyzing either the reflected or transmitted probe photons (depending on whether the material is opaque or transparent, respectively), one may access the properties of the excited phonons; indeed, typically the time scale of the latter is of the order of picoseconds, much longer than the duration of the pump and probe pulses (on the order of femtoseconds). The whole process is repeatedly performed for different time intervals between pump and probe, thus obtaining information on the phonon dynamics, as a function of the pump and probe time delay, through the analysis of the behavior of suitable probe photon observables.

Typically only the intensity of the reflected or transmitted probe light had been considered in order to investigate the properties of photoexcited phonons. Probe intensity analysis is indeed sufficient for studying the most simple phonon excitations, the ones that, in analogy with quantum optics, are called ‘coherent phonons’ [8, 21, 26]. Most of the theoretical work that has been directed at the analysis of coherent phonons is based on semiclassical approaches [24], using either the so-called ‘impulsive stimulated Raman scattering’, mainly applied in the production of coherent phonons in transparent materials [33, 38], or ‘displacive excitation’ mechanism proposed as an explanation for observations made in certain opaque materials [13, 40].

However, also of interest, both theoretically and experimentally, is the possibility of the ultrafast optical excitation of lattice vibrational modes with certain specific quantum features [5, 30], for instance the creation of ‘squeezed phonons’, so named again in analogy with quantum optics (e.g. see [3, 14, 18, 29] and references

therein). Squeezing is a typical non-classical property, as squeezed states can achieve lower quantum noise than the zero-point fluctuations of vacuum or coherent states [36, 39].

Theoretical studies on squeezed phonons typically use analogies with quantum light [36] in order to explain their generation and dynamics: phonons are treated either as elements of complex compound objects, like polaritons [2, 19], or independently, with squeezing resulting from the interaction with either photons, through Raman scattering, or among themselves [3, 16, 17]. At the same time the detection process is still treated in a semiclassical way, while the quantum features of lattice vibrations ought to be accessed via the analysis of the full quantum statistics of the scattered probe light. More recent studies have been focused on alternative generation mechanisms for squeezing, in particular, optical excitations of phonons mediated by quantum dots [7]; however, reduction of the fluctuations below the vacuum level were obtained only in the case of excitations induced by multiple pump pulses [32, 37].

Nonetheless, the production and detection of squeezed phonons using optical techniques have attracted a lot of attention also from the experimental point of view. Most of these efforts have focused on identifying signatures for the presence of squeezing through the analysis of the intensity of the collected scattered probe light as a function of the time delay between pump and probe pulses, tacitly assuming that the measured photon properties exactly reproduce those of the excited phonons.

In this respect, squeezed phonons were first reported to be observed in a transparent material at low temperature, by relating semiclassically the measured probe transmittance to the periodic oscillations of the lattice displacement in the target material [14]. This result has drawn a lot of controversy because of the small magnitude of the detected modulation, which is compatible with thermal fluctuations [18].

Further information on the dynamics of the excited material can be obtained by looking at additional data characterizing the scattered probe light beyond its mean intensity. This idea was first explored and discussed in experiments studying phonon dynamics in opaque materials [27, 28]. In such experiments, probe data were repeatedly collected for each time delay between pump and probe pulses, so that not only the mean intensity of the scattered probe light, but also fluctuations around the mean could be obtained. Assuming a perfect correspondence between the features of the detected scattered photon state and those of the excited phonons in the material, the presence of oscillations in the fluctuations with twice the frequency of the excited phonon mode was seen as a sign of phonon squeezing. Still, a shadow of doubt was cast also on these results, since the observed oscillations in the fluctuation data could be attributed to an intrinsically imprecise phase determination in the used apparatus, giving rise to the so-called jittering effects [20]. More recent experiments have been performed using ultrafast x-ray diffraction, granting a reduced pulse duration, hence reduced jittering errors [23]; despite this experimental advantage, the results concerning the presence of squeezed phonons seem to need further analysis.

In essence, a clear signal for the presence of squeezed phonons in pump excited materials remains elusive. Recently, new experiments have been performed, focusing on time-resolved optical experiments. In particular, the displacement of photoexcited atoms in the lattice has been addressed by means of non-equilibrium optical experiments in the shot-noise limited regime [10]. It appears that the unveiling of photon squeezing in α -quartz can be achieved through the acquisition of the full statistics of quantum fluctuations of the scattered probe photon number. Many factors need to be taken into account and understood in interpreting experimental data, in particular those related to the mechanism of generation of squeezing, the interaction of the excited phonons with their surrounding environment, the process of detection, and how to combine them all. The aim of the present investigation is to address some of these issues with a fully quantum, phenomenological description based on, first, a photon–phonon interaction hamiltonian containing the first relevant gauge-invariant contributions in terms of phononic and photonic annihilation and creation operators and, second, a master equation of Lindblad type taking care of the weak-coupling of the phonons with their environment.

More specifically, the interaction between the photons in the laser pulses and the phonons in the target material will be described by an effective photon–phonon quantum hamiltonian, capable of describing both pump and probe processes: linear and bilinear in the photon and phonon operators, this hamiltonian can account for the generation of both coherent and squeezed phonons in the pump process, at the same time describing the probe reading.

Furthermore, phonons in the material cannot be considered an isolated system: they interact weakly with their environment, made of the constituents of the material other than the phonons themselves. As a consequence, their dynamics is no longer unitary and needs to be studied within the open quantum system paradigm, through the use of an appropriate master equation, accounting for dissipative and noisy effects.

In order to correctly relate collected photon data with phonon properties, a consistent quantum description of the probing process is necessary⁵; two physically relevant scenarios will be described. In this way, higher order photon correlations become accessible, supporting a precise analysis of the phonon quantum state, that is not

⁵ In the frequency domain, the relations between the quantum statistics of phonons and scattered light has been discussed in [34].

possible within the semiclassical approaches used so far. More specifically, we shall focus on the study of two related probe photon observables, the mean number and its corresponding variance. Through the analysis of their joint behavior as a function of the pump and probe time delay, unique signals associated with the presence of squeezing in the pump excited phonons will be identified.

In detail, section 2 is devoted to the systematic description of a typical pump and probe experiment. The quantum hamiltonian describing the photon–phonon interaction will be introduced and discussed: it will be used for studying both pump and probe processes. Two different types of dissipative environments will be studied, thermal and dephasing baths; they give rise to two different master equations describing the dynamics of excited phonons, whose solutions will be explicitly obtained and analyzed in section 3. In section 4, the quantum formalism so far developed will be applied to the description of phonon excitations, as reported in experiments. This is done by studying in detail the evolution of the mean probe photon number and the corresponding variance: their joint analysis reveals unique signs of the presence of squeezed phonons. The final section 5 comprises an outline of our results and a discussion of recent experimental studies, that are compatible with the theoretical predictions of the approach outlined above. Finally, the Appendices contain technical material inappropriate for the main text.

2. Effective quantum description

Most of the various proposed mechanisms for the generation of phonons in pump and probe experiments (like Raman scattering, multiphonon scattering, and polaritons) rely on semiclassical treatments, using either ad hoc classical equations of motion for the atom displacement in the crystal or mean-field approximations, whereas the optical fields are mostly described classically. Although these models may account for some average features of the phonon dynamics, they are unsuited to capture their quantum properties.

2.1. Quantum approach to pump and probe experiments

The characterization of a typical pump and probe experiment involves several steps:

(1) *Initial state.* Before being hit by the pump laser pulse, the material under study is in thermal equilibrium: the initial phonon state, described by the density matrix ρ_β , is then a thermal one, characterized by the inverse temperature β , which in experiments frequently corresponds to room temperature. On the other hand, the laser pump pulse can be described by a coherent photon state $|\nu\rangle$ [11, 35], of high intensity $|\nu| \gg 1$. The initial total (photon and phonon) state is then given by the density matrix

$$|\nu\rangle\langle\nu| \otimes \rho_\beta. \quad (1)$$

(2) *Pumping process.* In view of the difference between the laser pulse's duration (≈ 80 – 100 femtoseconds) and the typical phonon evolution time scale (a few picoseconds), the pumping process can be considered instantaneous at time $t = 0$. Consequently, it is described by a unitary operator \mathcal{U} (discussed in detail in section 2.5, see also section 2.7), that transforms the initial state of the composite system into a new state

$$\mathcal{U}(|\nu\rangle\langle\nu| \otimes \rho_\beta)\mathcal{U}^\dagger. \quad (2)$$

This new state contains information about both photons and phonons. Since the scattered pump pulse is discarded and we are interested only in the phonon behavior, the photon degrees of freedom can be traced out:

$$\rho = \text{Tr}_{\text{photon}}[\mathcal{U}(|\nu\rangle\langle\nu| \otimes \rho_\beta)\mathcal{U}^\dagger]. \quad (3)$$

The density matrix ρ thus describes the state of the phonons immediately after the excitation by the pump.

(3) *Phonon dynamics.* The excited phonons cannot be considered isolated from their environment, since they inevitably interact with it, albeit weakly. In this case, standard techniques describe their time-evolution by means of a master equation [1, 4, 12]

$$\partial_t \rho(t) = \mathbb{L}[\rho(t)], \quad (4)$$

where the generator \mathbb{L} takes into account the dissipative and noisy effects induced by the environment.

(4) *Probing process.* After a delay time t , the probing pulse acts again impulsively; in analogy with the pump process, probing can be described by a unitary operator \mathcal{U} (treated in detail in section 2.7, see also section 2.5). It acts on a total photon–phonon state of the form $|\alpha\rangle\langle\alpha| \otimes \rho(t)$, where $|\alpha\rangle$ is the corresponding coherent state of the probe laser light, with intensity $|\alpha|$ considerably smaller than the pump intensity $|\nu|$. Since we extract information about the phonons by analyzing the scattered probe light, we are only interested in the state of the photons, obtained by tracing away the phonon degrees of freedom:

$$\sigma(t) = \text{Tr}_{\text{phonon}}[\mathcal{U}(|\alpha\rangle\langle\alpha| \otimes \rho(t))\mathcal{U}^\dagger]. \quad (5)$$

The photon state $\sigma(t)$ is completely characterized by its correlation functions

$$\langle (\hat{a}^\dagger)^m \hat{a}^n \rangle(t) = \text{Tr}_{\text{photon}} [(\hat{a}^\dagger)^m \hat{a}^n \sigma(t)], \quad (6)$$

where \hat{a}^\dagger , \hat{a} are the photon creation and annihilation operators and m, n positive integers. While previous investigations have considered essentially the case $m = n = 1$, in the present approach higher order photon correlation functions can be obtained. As we shall show, they allow a precise characterization of the quantum features of the phonon states.

In the following we will discuss in detail each one of these steps.

2.2. Initial phonon state

Let us consider pump and probe experiments at room temperature. Before the pump pulse hits the target, the phonons can then be assumed to be in thermal equilibrium at a given inverse temperature β ; their state is thus described by the thermal density matrix

$$\rho_\beta = \frac{e^{-\beta H}}{\text{Tr}[e^{-\beta H}]}. \quad (7)$$

In general, the pump laser beam can excite different vibrational modes; however, in many situations, only one mode is seen to be physically relevant due to the specific experimental setups [3, 14, 18, 29]. In such cases, the phonon hamiltonian can be taken to be $H = \Omega \hat{b}^\dagger \hat{b}$, where only one mode of energy Ω contributes, and \hat{b}^\dagger and \hat{b} are the corresponding phonon bosonic creation and annihilation operators satisfying the canonical commutation relation $[\hat{b}, \hat{b}^\dagger] = 1$. In this equilibrium state, the mean phonon number is given by

$$n_{\text{eq}} = \text{Tr}[\rho_\beta \hat{b}^\dagger \hat{b}] = (e^{\beta\Omega} - 1)^{-1}. \quad (8)$$

We remark that the assumption of a single active phonon mode is frequently realistic. For example, crystal symmetries and laser polarization may be exploited to selectively excite a single mode [10], or the temperature dependence of some mode can be used to suppress it, even at room temperature [22]. Furthermore, the formalism we are describing is rather general and can easily be extended to account for the presence of more than one phonon excitation mode.

2.3. Pump photon state

In pump and probe experiments, a mode-locked pulsed laser is used. Although laser pulses contains many frequencies, mode-locking allows one to consistently adopt a single-mode description, which collectively accounts for the coherent properties of the laser beam as discussed in detail in [11]. Such single-mode treatment of laser pulses interacting with matter is of course tenable only when the dynamics does not distinguish the various pulse components.

The pump laser photon state will then be described by a single mode coherent state $|\nu\rangle$, satisfying $\hat{a}|\nu\rangle = \nu|\nu\rangle$, with \hat{a} , \hat{a}^\dagger the (collective) annihilation and creation operators, satisfying the canonical commutation relation $[\hat{a}, \hat{a}^\dagger] = 1$. The parameter ν is related to the laser intensity via the mean photon number $\langle \nu | \hat{a}^\dagger \hat{a} | \nu \rangle = |\nu|^2$. Since the pump laser pulse is of high intensity, we have $|\nu| \gg 1$.

2.4. Photon–phonon effective interaction

The effects of the laser pulses on the crystal vibrations can generically be described in terms of first and second order processes. Since the pulse duration is much shorter than any phonon characteristic time, the laser action on the target material can be considered instantaneous, and consequently may effectively be described by a time dependent interaction hamiltonian of the form

$$\mathcal{H}(t) = (\gamma \hat{a}^\dagger \hat{b} + \gamma^* \hat{a} \hat{b}^\dagger + \eta (\hat{a}^\dagger)^2 \hat{b}^2 + \eta^* \hat{a}^2 (\hat{b}^\dagger)^2) \delta(t), \quad (9)$$

where $\delta(t)$ embodies the impulsive character of the interaction, while γ and η are suitable complex coupling constants. To apply the scheme to a specific experiment, they have to be determined, either from first principles or phenomenologically; a study of typically possible behavior can be found in section 4.

In (9), a generic photon–phonon interaction hamiltonian has been expanded up to the second order in photon and phonon operators: higher orders can be safely neglected since their coupling constants are smaller and smaller, taking into account that the scattering processes they describe are less likely to occur. Notice that the hamiltonian (9) has been chosen to preserve the number of bosons, i.e. the annihilation of a certain number of photons results in the creation of the same number of phonons and vice versa. This assumption corresponds to the request that the interaction be gauge-invariant in all bosonic operators. The linear terms in the hamiltonian involve only phonons in a single mode at null momentum \mathbf{k} , while the quartic ones are not limited to $\mathbf{k} = 0$ and one should thus integrate over the entire optical phonon dispersion region including processes where the

momentum conservation is guaranteed by the creation of optical phonons with opposite momenta. However, the effective hamiltonian comprises only a single phonon mode at $\mathbf{k} = 0$ since, in essentially the great majority of experiments so far performed, the probing process is limited to the linear regime whereby phonons at $\mathbf{k} \neq 0$ do not practically affect the observed photon number fluctuations.

Moreover, since the hamiltonian is linear and bilinear in the phonon operators, the described interaction can generate not only coherent or squeezed phonons, but also both effects at the same time—a situation that is far more likely.

The above hamiltonian is time dependent and thus the evolution operator $\mathcal{U}(t, t_0)$ involves a time-ordered exponential. In this case, however, the integral can be calculated so that the $\mathcal{U}(t, t_0)$ is the same for all $t_0 < 0$ and $t > 0$. Consequently, we can describe the photon–phonon interaction by a single application of the operator

$$U = \exp[-i(\gamma \hat{a}^\dagger \hat{b} + \gamma^* \hat{a} \hat{b}^\dagger + \eta (\hat{a}^\dagger)^2 \hat{b}^2 + \eta^* \hat{a}^2 (\hat{b}^\dagger)^2)]; \quad (10)$$

it represents the action of the laser pulses on any photon–phonon state.

2.5. Pump process

As mentioned before, the pump process transforms the initial total state $|\nu\rangle\langle\nu| \otimes \rho_\beta$ into $\mathcal{U}(|\nu\rangle\langle\nu| \otimes \rho_\beta)\mathcal{U}^\dagger$. Since $|\nu\rangle$ represents a high intensity coherent photon state, the use of a mean-field approximation for the photon degrees of freedom is appropriate (see appendix A for a more detailed discussion); then, one may replace \hat{a} with ν and \hat{a}^\dagger with ν^* , so that \mathcal{U} in (10) reduces to

$$U_\nu^{\text{MF}} = \exp[-i(\gamma \nu^* \hat{b} + \gamma^* \nu \hat{b}^\dagger + \eta (\nu^*)^2 \hat{b}^2 + \eta^* \nu^2 (\hat{b}^\dagger)^2)], \quad (11)$$

containing only phonon operators. Therefore, immediately after the action of the pump, the state of the composed photon–phonon system is given by

$$|\nu\rangle\langle\nu| \otimes (U_\nu^{\text{MF}} \rho_\beta (U_\nu^{\text{MF}})^\dagger). \quad (12)$$

It turns out that U_ν^{MF} can be rewritten as a combination of displacement (D) and squeezing (S) operators⁶

$$U_\nu^{\text{MF}} = S(\xi_\nu)D(z_\nu), \quad (13)$$

with

$$D(z_\nu) = \exp(z_\nu \hat{b}^\dagger - z_\nu^* \hat{b}), \quad (14)$$

$$S(\xi_\nu) = \exp\left(\frac{1}{2}\xi_\nu^* \hat{b}^2 - \frac{1}{2}\xi_\nu (\hat{b}^\dagger)^2\right), \quad (15)$$

where the displacement z_ν and squeezing $\xi_\nu \equiv r_\nu e^{i\varphi_\nu}$ parameters are explicitly given by

$$z_\nu = \frac{|\gamma|}{2|\eta||\nu|} [(1 - \cosh(2|\eta||\nu|^2))e^{i(\theta_\nu + \theta_\gamma - \theta_\eta)} - \sinh(2|\eta||\nu|^2)e^{i(\theta_\nu - \theta_\gamma + \pi/2)}], \quad (16)$$

$$r_\nu = 2|\eta||\nu|^2, \quad \varphi_\nu = 2\theta_\nu - \theta_\eta + \frac{\pi}{2}, \quad (17)$$

with $\gamma = |\gamma| e^{i\theta_\gamma}$, $\eta = |\eta| e^{i\theta_\eta}$ and $\nu = |\nu| e^{i\theta_\nu}$.

The phonon state after the pump process

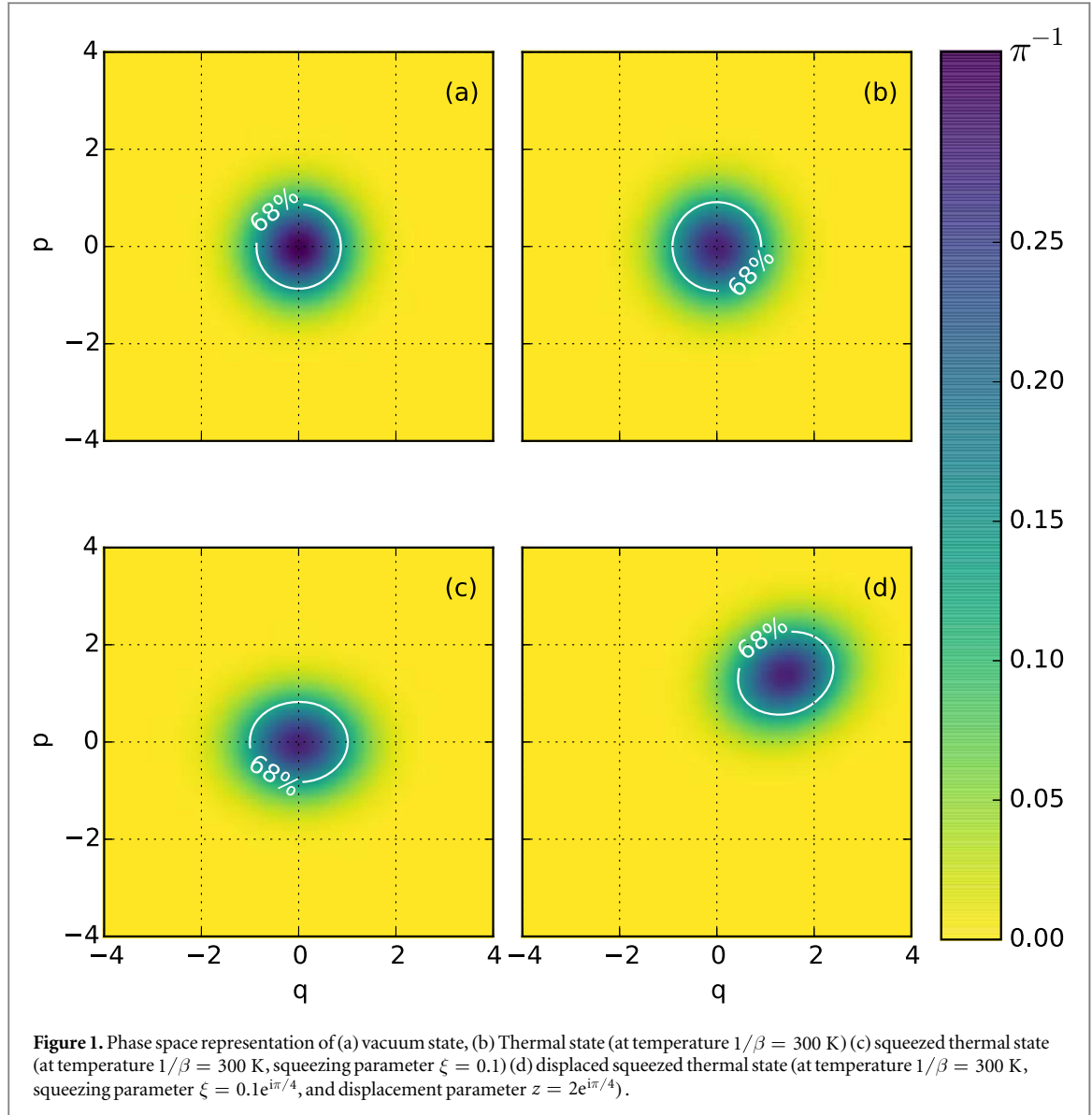
$$\rho = U_\nu^{\text{MF}} \rho_\beta (U_\nu^{\text{MF}})^\dagger, \quad (18)$$

obtained from (12) by tracing out the photon degrees of freedom, is then a squeezed coherent thermal state; it represents the initial state of the phonon dynamics, whose description requires taking into account the weak interaction with the environment (see figure 1 for a phase–space representation of all the above introduced states).

2.6. Phonon dynamics

Phonons in the target material can hardly be considered as an isolated system, since they inevitably interact with the environment formed by the remaining crystal degrees of freedom, e.g. electrons or impurities. However, the interaction is in general weak and furthermore, initially, phonons and the environment can be considered uncorrelated. In such situations, the standard approach to open quantum systems allows to derive through the so-called ‘weak coupling limit’ techniques a Lindblad type master equation for the dynamics of the phonon state $\rho(t)$, that takes the general form [1, 4]

⁶ This can be easily proved by recalling the action of displacement and squeezing operators (14) and (15) on phonon creation and annihilation operators [15].



$$\begin{aligned} \partial_t \rho(t) = & -i[H, \rho(t)] \\ & + \frac{1}{2} \sum_k \lambda_k (2V_k \rho(t) V_k^\dagger - V_k^\dagger V_k \rho(t) - \rho(t) V_k^\dagger V_k). \end{aligned} \quad (19)$$

The first line alone would give rise to the usual unitary evolution generated by the phonon free hamiltonian $H = \Omega \hat{b}^\dagger \hat{b}$, while the additional contribution in the second line arises due to the presence of the environment; the quantities V_k represent suitable phonon operators, while the real and positive constants λ_k parametrize noisy and dissipative effects. Two different, physically relevant situations [12] will be studied explicitly:

Thermal environment. Experimental evidence indicates that, after being excited by the pump pulse, phonons relax in a few picoseconds to a thermal equilibrium state, at a final temperature possibly higher than the initial one. We have thus considered the typical master equation that describes this situation:

$$\begin{aligned} \partial_t \rho(t) = & -i[\Omega \hat{b}^\dagger \hat{b} \rho(t)] \\ & + \frac{\lambda_T}{2} [(n_f + 1)(2\hat{b} \rho(t) \hat{b}^\dagger - \hat{b}^\dagger \hat{b} \rho(t) - \rho(t) \hat{b}^\dagger \hat{b}) \\ & + n_f (2\hat{b}^\dagger \rho(t) \hat{b} - \hat{b} \hat{b}^\dagger \rho(t) - \rho(t) \hat{b} \hat{b}^\dagger)], \end{aligned} \quad (20)$$

where $n_f = (e^{\beta_f \Omega} - 1)^{-1}$ is the thermal mean phonon number at the final inverse temperature β_f , while $\lambda_T \geq 0$ is the phenomenological constant parametrizing the strength of noise and dissipation. The dynamics generated by this equation has a unique long-time asymptotic state, the Gibbs state at inverse temperature β_f .

Dephasing environment. A typical effect due to the presence of an external environment is loss of coherence, a phenomenon usually referred to as ‘dephasing’. This occurs when the number of phonons is constant in time,

since due to the Heisenberg uncertainty principle, the conjugate variable, the ‘phase’, must then be completely undetermined. A typical master equation describing dephasing effects is given by [12]

$$\partial_t \rho(t) = -i[\Omega \hat{b}^\dagger \hat{b}, \rho(t)] - \frac{\lambda_D}{2} [\hat{b}^\dagger \hat{b}, [\hat{b}^\dagger \hat{b}, \rho(t)]], \quad (21)$$

where $\lambda_D \geq 0$ is the phenomenological parameter describing the damping of the system. This master equation describes the situation in which there is no energy exchange between the phonons and their surroundings; indeed, the phonon number operator $\hat{b}^\dagger \hat{b}$ is left invariant by the time evolution generated by (21), which, as a consequence, does not possess a unique asymptotic state.

Besides their physical interpretation and applicability in the description of the phonon dynamics, these two different master equations can also be solved analytically.

2.7. Probing process

While the pump pulse is intense in order to excite the system out of equilibrium, the probe pulse is less intense and its use is to record the properties of the phonon state through the analysis of the properties of the scattered probe photons. By varying the time delay between pump and probe pulses, the relaxing phonon dynamics can thus be systematically examined.

As in the case of the pump, the probe laser pulses can be described using a coherent state $|\alpha\rangle$, so that the density matrix representing the state of the compound photon–phonon system just before probing can still be written in factorized form, $|\alpha\rangle\langle\alpha| \otimes \rho(t)$ (compare with (1)). However, one has $|\alpha| \ll |\nu|$, since in general the probe pulses are less intense than the pump ones. In such a case, the unitary operator $\mathcal{U} \equiv \exp(-iG)$ in (10), with $G = \gamma \hat{a}^\dagger \hat{b} + \gamma^* \hat{a} \hat{b}^\dagger + \eta (\hat{a}^\dagger)^2 \hat{b}^2 + \eta^* \hat{a}^2 (\hat{b}^\dagger)^2$, representing in general the action of the laser pulses on the target material, cannot be written as in (11), since the probe pulse can no longer be considered semiclassical, and therefore no mean-field approximation can be used. Nevertheless, when acting on $|\alpha\rangle\langle\alpha| \otimes \rho(t)$, \mathcal{U} can always be decomposed as

$$\mathcal{U} = U_\alpha + \Delta_\alpha, \quad (22)$$

in terms of the new unitary operator

$$U_\alpha \equiv \exp(-iG_\alpha), \quad G_\alpha = \gamma \hat{a}^\dagger \hat{b} + \gamma^* \hat{a} \hat{b}^\dagger + \eta (\alpha^*)^2 \hat{b}^2 + \eta^* \alpha^2 (\hat{b}^\dagger)^2 \quad (23)$$

and the difference Δ_α

$$\Delta_\alpha \equiv \mathcal{U} - U_\alpha = \int_0^1 ds \partial_s [U_\alpha^{1-s} \mathcal{U}^s] = i \int_0^1 ds U_\alpha^{1-s} (G - G_\alpha) \mathcal{U}^s.$$

Note that U_α is akin to a mean-field approximation in only the quadratic terms, thus introducing the parameter α . The full expression for \mathcal{U} in (22) is, however, still exact. Now we introduce an approximation that goes beyond the mean-field, while still guaranteeing the tractability of the calculation. The details of its application will be discussed in section 2.8. A first order approximation of Δ_α suffices, so that in the probing process \mathcal{U} can be approximated by the following expression:

$$\mathcal{U}_\alpha = U_\alpha \left\{ \mathbb{I} - i \int_0^1 ds (U_\alpha^\dagger)^s [\eta (\hat{a}^{\dagger 2} - \alpha^{*2}) \hat{b}^2 + \eta^* (\hat{a}^2 - \alpha^2) \hat{b}^{\dagger 2}] (U_\alpha)^s \right\}. \quad (24)$$

The state of the compound photon–phonon system after the probe pulse hits the material is then given by $\mathcal{U}_\alpha(|\alpha\rangle\langle\alpha| \otimes \rho(t)) \mathcal{U}_\alpha^\dagger$. In general, the bilinear terms in the exponential dominate over the quadratic ones, so that one can safely assume $\eta \approx 0$; however, there are situations in which, due to crystal symmetries or to specific experimental adjustments, the terms with the linear pieces in photon and phonon operators do not contribute [14], this situation is commonly considered for the sake of simplicity when the generation of a squeezed phonon state is the goal. In such cases $\gamma = 0$ and only the quadratic terms survive. Both situation will be discussed in detail in the following.

2.8. Photon correlations

Our aim is the study of the behavior of the photoexcited phonons as monitored by the probe pulses. This can be achieved by analyzing suitable scattered probe photon observables. Specifically, we shall focus on the behavior of the mean photon number $\langle \hat{a}^\dagger \hat{a} \rangle$ and on its variance $\Delta_{\hat{a}^\dagger \hat{a}}$. While $\langle \hat{a}^\dagger \hat{a} \rangle$ is connected to the reflectivity/transmittance of the probe light and thus to the semiclassical properties of the excited phonons, higher order photon correlations, as in $\Delta_{\hat{a}^\dagger \hat{a}}$, are needed in order to inspect its quantum aspects. The joint analysis of mean

number and its variance, two experimentally accessible photon observables, provides enough information for identifying some phonon quantum features, in particular the presence of squeezing.

The mean probe photon number at time t and its corresponding variance can be suitably written as⁷:

$$\langle \hat{a}^\dagger \hat{a} \rangle(t) = \text{Tr}[(\mathcal{U}_\alpha^\dagger \hat{a}^\dagger \hat{a} \mathcal{U}_\alpha) |\alpha\rangle \langle \alpha| \otimes \rho(t)], \quad (25)$$

$$\Delta_{\hat{a}^\dagger \hat{a}}(t) = \langle (\hat{a}^\dagger \hat{a})^2 \rangle(t) - (\langle \hat{a}^\dagger \hat{a} \rangle(t))^2, \quad (26)$$

with

$$\langle (\hat{a}^\dagger \hat{a})^2 \rangle(t) = \text{Tr}[(\mathcal{U}_\alpha^\dagger (\hat{a}^\dagger \hat{a})^2 \mathcal{U}_\alpha) |\alpha\rangle \langle \alpha| \otimes \rho(t)], \quad (27)$$

where the trace operation is now performed on both phonon and photon degrees of freedom. These quantities can be conveniently expressed in terms of suitable phonon correlations, $\langle (\hat{b}^\dagger)^m \hat{b}^n \rangle(t) = \text{Tr}_{\text{phonon}}[(\hat{b}^\dagger)^m \hat{b}^n \rho(t)]$, with $m, n = 0, 1, 2$; their explicit forms depend on which of the two probing processes mentioned above is relevant in the examined context (details and further discussion can be found in appendix B).

First-order probing process. When the quadratic terms in the hamiltonian are negligible, i.e. $\eta \approx 0$, a situation most likely occurring in opaque materials, the impulsive unitary operator describing the probe process (24) reduces to

$$\mathcal{U}_\alpha = \exp(-i[\gamma \hat{a}^\dagger \hat{b} + \gamma^* \hat{a} \hat{b}^\dagger]). \quad (28)$$

Note that this operator does not depend on α anymore, and that the subscript merely denotes the approximation used. In this case, the mean value of the photon number can be expressed as

$$\begin{aligned} \langle \hat{a}^\dagger \hat{a} \rangle(t) &= |\alpha|^2 \cos^2(|\gamma|) + \langle \hat{b}^\dagger \hat{b} \rangle(t) \sin^2(|\gamma|) \\ &+ i \frac{\sin(|\gamma|) \cos(|\gamma|)}{|\gamma|} (\gamma^* \alpha \langle \hat{b}^\dagger \rangle(t) - \gamma \alpha^* \langle \hat{b} \rangle(t)), \end{aligned} \quad (29)$$

while the explicit expression of the variance $\Delta_{\hat{a}^\dagger \hat{a}}(t)$ is rather more involved; it is collected in appendix B.

Second-order probing process. On the other hand, when there is no contribution from the linear terms in phonon creation and annihilation operators ($\gamma \approx 0$), no generation of coherent states out of the pump process can be achieved. This situation describes the process in which the excited phonon state resulting from the pump process is a squeezed thermal one, a typical situation encountered in transparent materials [14, 18]; in this case, \mathcal{U}_α is still given by (24), but with the simplified \mathcal{U}_α :

$$\mathcal{U}_\alpha = \exp(-i[\eta (\alpha^*)^2 \hat{b}^2 + \eta^* \alpha^2 (\hat{b}^\dagger)^2]). \quad (30)$$

The mean value of the photon number takes now the form

$$\langle \hat{a}^\dagger \hat{a} \rangle(t) = |\alpha|^2 - \sinh^2(r_\alpha) (1 + 2 \langle \hat{b}^\dagger \hat{b} \rangle(t)) + \sinh(2r_\alpha) \Re[\langle \hat{b}^2 \rangle(t) e^{-i\varphi_\alpha}], \quad (31)$$

where r_α and φ_α are as in (17), but with ν replaced by $\alpha = |\alpha| e^{i\theta_\alpha}$, while \Re signifies real part; similarly, for the variance one gets:

$$\begin{aligned} \Delta_{\hat{a}^\dagger \hat{a}}(t) &= |\alpha|^2 - 2(1 + 2 \langle \hat{b}^\dagger \hat{b} \rangle(t)) \sinh^2(r_\alpha) \\ &- (1 + 2 \langle \hat{b}^\dagger \hat{b} \rangle(t))^2 \sinh^4(r_\alpha) + 2 \sinh(2r_\alpha) \Re[\langle \hat{b}^2 \rangle(t) e^{-i\varphi_\alpha}] \\ &+ \sinh^2(r_\alpha) \sinh(2r_\alpha) (1 + 2 \langle \hat{b}^\dagger \hat{b} \rangle(t)) \Re[\langle \hat{b}^2 \rangle(t) e^{-i\varphi_\alpha}] \\ &- \sinh^2(2r_\alpha) (\Re[\langle \hat{b}^2 \rangle(t) e^{-i\varphi_\alpha}])^2. \end{aligned} \quad (32)$$

In order to obtain the explicit time dependence of these two observables, $\langle \hat{a}^\dagger \hat{a} \rangle(t)$ and $\Delta_{\hat{a}^\dagger \hat{a}}(t)$, one now has to analyze the phonon dynamics generated by the master equations (20) and (21) and compute the needed phonon correlation functions.

3. Dissipative phonon dynamics

As explained in section 2, after the action of the pump, the excited phonons will dynamically evolve as an open quantum system, since they interact weakly with the surrounding environment. Their evolution in time is described by a Lindblad master equation of the form (19); its explicit solution depends on the type of the environment, and the two cases presented in section 2.6 need to be treated separately.

Thermal environment. In the case of a thermal reservoir, the master equation takes the form (20). In this case, it is useful to express the phonon density matrix ρ in phase space using the displacement operator D ,

⁷ Notice that if \hat{a}, \hat{a}^\dagger were substituted by α, α^* in the expression of the operator implementing the probing process (see (10)), there would be no reading of the phonon properties by the probe photons, since in this case $\mathcal{U}_\alpha^\dagger \hat{a}^\dagger \hat{a} \mathcal{U}_\alpha = \hat{a}^\dagger \hat{a}$.

by writing [15]

$$\rho = \frac{1}{\pi} \int_{\mathbb{C}} d^2w \chi(w) D(-w), \quad (33)$$

with w a complex variable; the characteristic function $\chi(w)$ can be obtained through the equation

$$\chi(w) = \text{Tr}[\rho D(w)]. \quad (34)$$

For the squeezed coherent thermal state (18) resulting from the pump process, one finds [25]

$$\chi(w) = e^{-(A+\frac{1}{2})|w|^2 - \frac{1}{2}(B^* w^2 + B (w^*)^2) + C^* w - C w^*}, \quad (35)$$

with

$$A = n_{\text{eq}} + (2n_{\text{eq}} + 1) \sinh^2(r_\nu), \quad (36)$$

$$B = (2n_{\text{eq}} + 1) e^{i\varphi_\nu} \sinh(r_\nu) \cosh(r_\nu), \quad (37)$$

$$C = z_\nu \cosh(r_\nu) - z_\nu^* e^{i\varphi_\nu} \sinh(r_\nu), \quad (38)$$

making apparent its gaussian character. Since the master equation (20) contains at most quadratic terms in phonon creation and annihilation operators, this gaussian character will not be spoiled by the finite time evolution generated by it. As a result, the time dependent density operator, solution of (20), can also be written in a form similar to (33); specifically, one finds

$$\rho(t) = \frac{1}{\pi} \int_{\mathbb{C}} d^2w \chi_t(w) e^{i\phi_t(w)} D(-w), \quad (39)$$

where

$$\phi_t(w) = i \left(n_f + \frac{1}{2} \right) (1 - e^{-\lambda_T t}) |w|^2, \quad (40)$$

while $\chi_t(w)$ is as in (35) with the replacement of w with $e^{i\Omega t} e^{-\lambda_T t/2} w$. From this expression, any phonon correlation function $\langle (\hat{b}^\dagger)^m \hat{b}^n \rangle(t) \equiv \text{Tr}[(\hat{b}^\dagger)^m \hat{b}^n \rho(t)]$ can easily be obtained through the formula:

$$\langle (\hat{b}^\dagger)^m \hat{b}^n \rangle(t) = (-1)^n \partial_w^m \partial_{w^*}^n \left[\chi_t(w) e^{i\phi_t(w)} e^{\frac{|w|^2}{2}} \right]_{|w=0, w^*=0}. \quad (41)$$

For instance, one gets:

$$\langle \hat{b} \rangle(t) = C e^{-i\Omega t} e^{-\lambda_T t/2}, \quad (42)$$

$$\langle \hat{b}^2 \rangle(t) = (C^2 - B) e^{-2i\Omega t} e^{-\lambda_T t}, \quad (43)$$

$$\langle \hat{b}^\dagger \hat{b} \rangle(t) = (A + |C|^2) e^{-\lambda_T t} + n_f (1 - e^{-\lambda_T t}). \quad (44)$$

Dephasing environment. In the case of the reservoir that preserves the phonon number, the corresponding master equation (21) can be explicitly solved in the Fock representation. Indeed, in the standard Fock basis formed by eigenstates of the number operator, $\{|n\rangle, n = 1, 2, \dots\}$, with $\hat{b}^\dagger |n\rangle = (n+1)|n+1\rangle$, the master equation reduces to

$$\dot{\rho}_{m,n}(t) = \left[-i\Omega(m-n) - \frac{\lambda_D}{2}(m-n)^2 \right] \rho_{m,n}, \quad (45)$$

where $\rho_{m,n}(t) = \langle m | \rho(t) | n \rangle$ are the elements of the phonon density matrix, so that

$$\rho_{m,n}(t) = e^{-i\Omega(m-n)t} e^{-\lambda_D(m-n)^2 t/2} \rho_{m,n}. \quad (46)$$

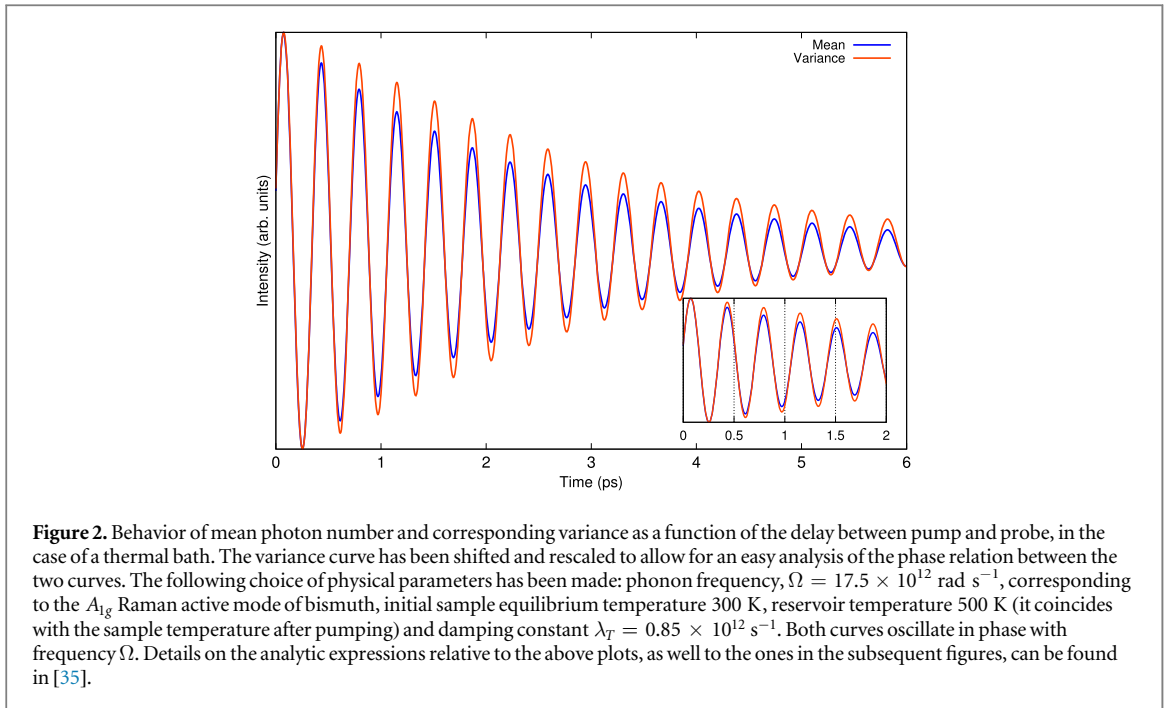
The phonon correlation functions at time t

$$\langle (\hat{b}^\dagger)^m \hat{b}^n \rangle(t) = e^{i(m-n)\Omega t} e^{-(m-n)^2 \lambda_D t/2} \langle (\hat{b}^\dagger)^m \hat{b}^n \rangle, \quad (47)$$

can then be expressed in terms of known, time independent correlations

$$\langle (\hat{b}^\dagger)^m \hat{b}^n \rangle = \text{Tr}[(\hat{b}^\dagger)^m \hat{b}^n \rho], \quad (48)$$

i.e. the correlations in the initial state ρ , the squeezed coherent thermal state (18) obtained from the pumping process.



4. Phonon quantum properties: squeezing

Having obtained explicit expressions for the time evolution of mean number and corresponding variance of the scattered probe light, we can now show how the joint analysis of these observables allows a much more complete description of the quantum properties of excited phonons in pump and probe experiments than so far achieved.

As mentioned in the introduction, the creation and detection of squeezed phonons in optically excited materials have been subject of debate: based on the experiments so far performed and the related theoretical studies, a definite sign, indicator for the presence of squeezed phonons in pump excited materials has not yet been identified. In order to study this question within our fully quantum approach, we have explored a broad set of values for pump intensity, coupling constants, probe intensity and damping parameters. We will present our results separately for thermal and dephasing bath, further distinguishing between opaque and transparent materials (see [35] for a more comprehensive treatment).

4.1. Thermal environment

The thermal reservoir has a unique equilibrium state, a Gibbs state determined by the temperature of the bath. This temperature is larger than the initial equilibrium temperature of the target material (here considered to be the usual room temperature of about 300 K), since the highly intense pump pulse is partially absorbed by the crystal. Typical temperatures reached by an absorbing opaque crystal after the pump are around 500 K, while for transparent materials, where the absorption effects can be neglected, the increase in temperature can be estimated to be at most few degrees higher than the initial one. As discussed earlier, the pump pulse excites the phonons, producing displacement and squeezing in the initially thermal state: their subsequent dissipative relaxation back to equilibrium as driven by the presence of the thermal environment, can be studied through the analysis of the typical oscillating behavior of probe photon mean number and variance (see figure 2).

At this stage, the difference between opaque and transparent materials needs to be taken into account; as mentioned, the final temperature is considerably higher in the case of opaque crystals, like bismuth, due to the absorption of energy from the pump pulse. Meanwhile, in transparent materials, like α -quartz, the final temperature can be expected to be only marginally higher than the initial one. In the model described here, this parameter needs to be either estimated from the crystal properties or fitted from the experimental results. The possibility of melting the target due to the absorption of energy from high intensity pump pulses establishes a natural threshold for opaque and transparent materials. For opaque materials, a limit in the intensity used in the pump pulse is naturally given by the properties of the crystal, while for non-absorbing crystal, i.e. transparent crystal targets, a broad range of intensities (typically referred to as fluence in experiments) can be used.

Concerning the probe process, there is no distinction between the two cases described above, since the probe pulse is always weak in order to avoid an additional excitation of the crystal target.

Linear probing process. The physical situation in which the probe interaction is dominated by mechanisms involving linear terms in photon and phonon operators is adequately described by the linear probing process.

This regime is usually achieved by means of a probe pulse that is much weaker than the pump pulse. Some particular situations are worth discussing in detail, listed below according to characteristics of the state ρ of the excited phonons after pumping.

(i) *Thermal state.* If the pump pulse creates neither any displacement nor squeezing in the initial thermal equilibrium state, no oscillating terms appear in the time evolution of the mean photon number or variance. Only a thermalization process is observed, driving the phonon state to a new equilibrium state, the one supported by the thermal bath; it is characterized by the damping constant λ_T and the bath temperature.

(ii) *Coherent thermal state.* Another physically likely and relevant situation is when the pump process displaces the thermal state, but does not squeeze it, i.e. $\rho = D(z_\nu)\rho_\beta D^\dagger(z_\nu)$, so that the excited phonons are described by a coherent thermal state. This particular case can be achieved by either a not so intense pump pulse or by a negligible coupling constant η . In this situation, the mean photon number and the corresponding variance both behave in the same way as a function of the time delay between pump and probe: both observables show typical damped oscillations with frequency given by the excited phonon mode Ω , as depicted in figure 2. This is also the typical behavior of the reflectivity/transmittance reported in experiments on optically excited coherent phonons.

(iii) *Squeezed thermal state.* Another possibility is given by the excitation of phonons in a squeezed thermal state, $\rho = S(\xi_\nu)\rho_\beta S^\dagger(\xi_\nu)$. It should be noted that this case is less likely to occur since it corresponds to a physical situation in which the bilinear contributions in the effective hamiltonian describing the pumping process can be neglected. In this case the mean photon number does not present oscillations, whereas the variance oscillates with twice the phonon mode frequency Ω ; furthermore, both observables contain damping factors characterized by the dissipative constant λ_T . The presence of the frequency 2Ω in the variance oscillations is a signal of squeezing in the excited phonon state; indeed, if the squeezing parameter ξ_ν is zero, then this contribution disappears.

(iv) *Squeezed coherent thermal state.* The most general situation occurs when the pumping process excites a phonon state which is both squeezed and coherent, as in (18), a likely situation in this kind of non-equilibrium optical experiments. In this case, the mean photon number oscillates with frequency Ω , with an amplitude that, besides depending on displacement, squeezing and probe intensity parameters, is damped in time by the factor $e^{-\lambda_T t/2}$. On the other hand, the associated variance shows oscillating terms with frequency Ω , which are damped by factors $e^{-\lambda_T t/2}$ or $e^{-3\lambda_T t/2}$, and terms with frequency 2Ω , damped by $e^{-\lambda_T t}$. This is due to the different damping behavior in (42)–(44). Furthermore, one finds that the oscillating piece with frequency 2Ω is present only if the squeezing parameter ξ_ν is non-vanishing, i.e. only in presence of squeezed phonons. However, it is in general difficult to isolate such a contribution or to have evidence of a 2Ω contribution in the corresponding Fourier spectrum; indeed, at least for high intensity pump and probe pulses, commonly used in pump and probe experiments where the crystal target is non-absorbing, the mean photon number and variance show a behavior in time similar to the one shown in figure 2.

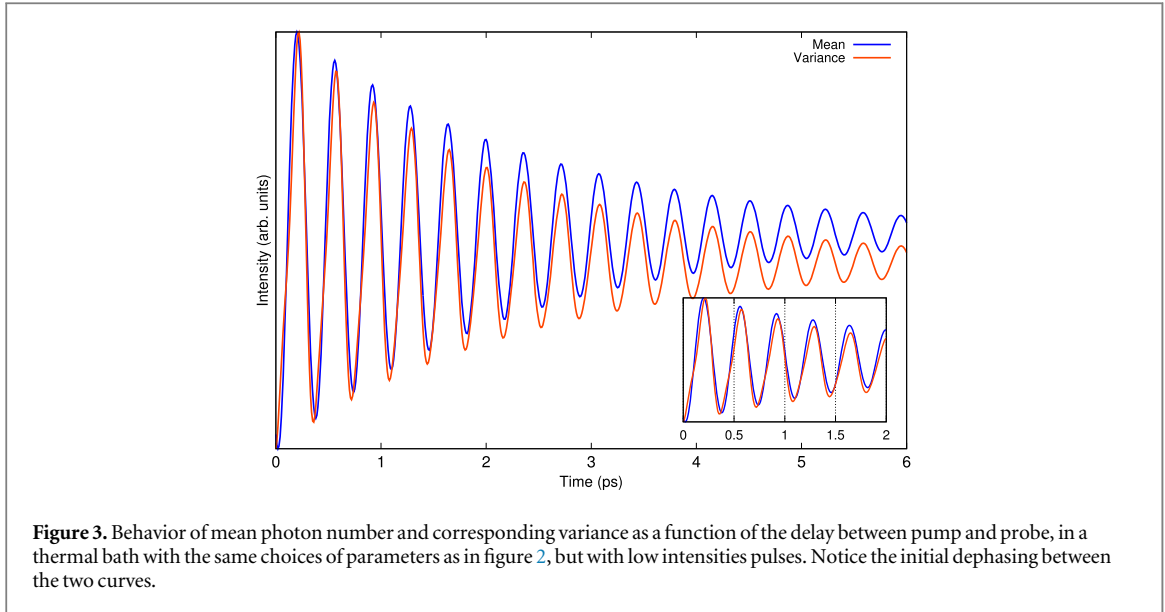
For low intensity probe pulses $|\alpha|^2 < 1$ and pump intensity $|\nu|^2$ two orders of magnitude larger, describing experiments with absorbing materials, one can notice the occurrence of a phase shift between the oscillations of mean number and variance, which is present only if the phonon state out of the pump process has non-vanishing squeezing (see figure 3). This shift is time dependent and in general vanishes after a few oscillations (see appendix C for details). The detection of such a shift can be taken as clear signal of the presence of squeezed phonons in pump and probe experiments involving opaque materials. The visibility of the shift in actual experiments depends on the specific features of the setups: each given experimental situation fixes the magnitude of the phenomenological parameters associated with both the pump and probe processes and the dissipative dynamics and, as a consequence, also the magnitude of the shift.

Quadratic probing process. As already mentioned before, due either to crystal symmetries or to specific experimental situations, optical excitation of phonons in some crystal targets do not involve coherent contributions: only the quartic pieces in the hamiltonian (9) are relevant [14]; in other words, $\gamma = 0$. We focus below on relevant physical situations, listed according to the characteristics of the excited phonon state ρ out of the pumping process; note that, in the present situation, the displacement parameter z_ν is always vanishing.

(i) *Thermal state.* When the pump is not able to excite the target material, i.e. the phonon state after pumping is still a thermal state, the behavior of the mean photon number and its variance is given by exponential damping terms, signaling thermalization of the phonon state to the bath temperature.

(ii) *Squeezed thermal state.* In this case, while the photon mean number oscillates with frequency 2Ω , the corresponding variance contains oscillatory contributions with frequency 2Ω and 4Ω . In addition, one finds that, in both cases, these oscillations are present only when the squeezing parameter ξ_ν is non-zero: their detection would then constitute a clear signal of the presence of squeezing in the pump excited phonons.

Notice that for small values of the squeezing parameter ξ_ν , the oscillating contribution with frequency 4Ω in the variance can be neglected; in this case, both the mean and variance oscillate with the same frequency and in



addition these oscillations are perfectly in phase (the behavior is similar to the one reported in figure 2, although the oscillations now occur at frequency 2Ω).

4.2. Dephasing environment

As discussed in section 3, the characteristic property of the dephasing bath is the conservation of the number of phonons in the material. Thus, if the pump pulse is too weak to produce excitations, leaving the phonons in thermal equilibrium, the phonon dynamics do not show any time dependence since the thermal state is a stationary state of the master equation (21). A non trivial time evolution occurs only when the pump pulse is able to generate phonon displacement and squeezing. As in the case of the thermal reservoir, also for a dephasing environment the behavior of the photon observables depends on the reading mechanism considered in the probe process.

Linear probing process. We shall first consider the situation in which the probing mechanism can be described using only the bilinear terms in the hamiltonian (9) describing photon–phonon interactions. Again we describe different cases, classified according to the phonon state ρ .

(i) *Coherent thermal state.* When the pump process excites coherent phonons only, the mean photon number oscillates with frequency Ω , damped by a factor $e^{-\lambda_D t/2}$. On the other hand, the variance shows oscillations both with frequency Ω and 2Ω ; however, the latter are more damped, the decaying factors being proportional to $e^{-\lambda_D t}$ and $e^{-2\lambda_D t}$, respectively. In practice, mean number and variance are seen to oscillate in time with the same frequency and in addition with the same phase. In the literature, the presence in the variance of oscillating terms with frequency 2Ω has been repeatedly considered to be a signal of the presence of phonon squeezing. Instead, here we see that, due to the effects of the dephasing bath, a 2Ω oscillating contribution can arise even in absence of squeezing, and therefore that the appearance of such oscillations in the variance cannot be taken as an unquestionable signal of squeezing.

(ii) *Squeezed coherent thermal state.* The most likely situation is, however, the excitation of squeezed coherent thermal phonons. In this more general case, for low intensity pulses, the dominant terms in the mean photon number and variance take the form [35]:

$$\begin{aligned} \langle \hat{a}^\dagger \hat{a} \rangle(t) &\sim -|\alpha||z_\nu| e^{-\lambda_D t/2} \sin(2|\gamma|) [\mathcal{A} \sin(\Omega t + \phi_A) - \mathcal{B} \sin(\Omega t + \phi_B)], \\ \Delta_{\hat{a}^\dagger \hat{a}}(t) &\sim -|\alpha||z_\nu| e^{-\lambda_D t/2} \sin(2|\gamma|) [\mathcal{C} \sin(\Omega t + \phi_A) - \mathcal{D} \sin(\Omega t + \phi_B)], \end{aligned} \quad (49)$$

where

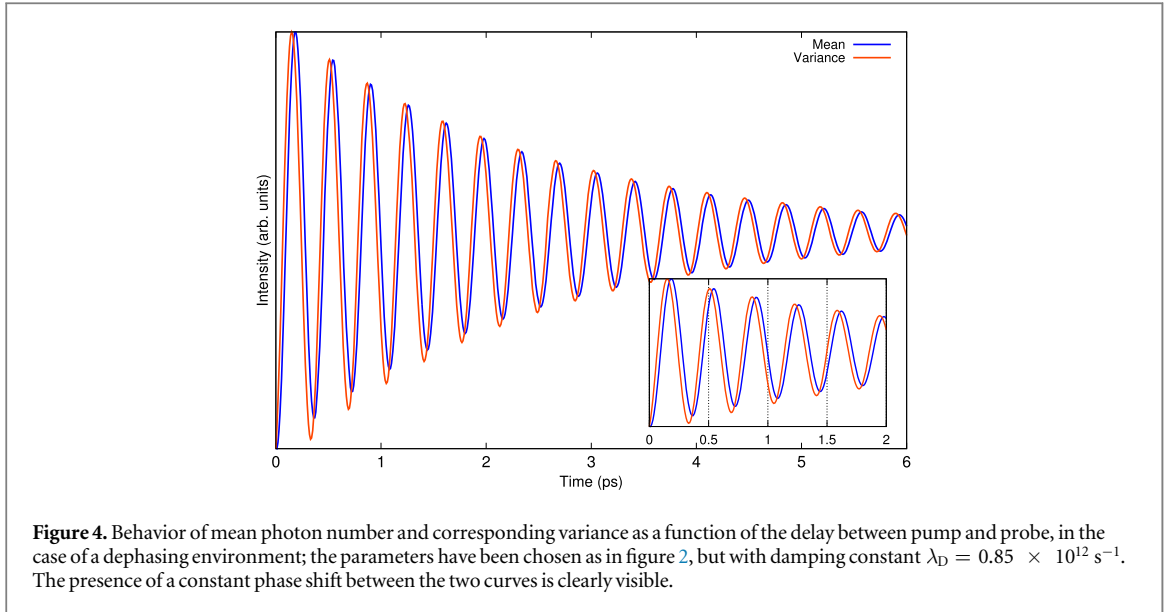
$$\mathcal{A} = \cosh(r_\nu), \quad \phi_A = \theta_\alpha - \theta_\gamma - \theta_z, \quad (50)$$

$$\mathcal{B} = \sinh(r_\nu), \quad \phi_B = \theta_\alpha - \theta_\gamma + \theta_z - \varphi_\nu, \quad (51)$$

$$\mathcal{C} = [\cos^2(|\gamma|) + (1 + 2n_{\text{eq}})(2 \cosh(2r_\nu) - 1) \sin^2(|\gamma|)] \cosh(r_\nu), \quad (52)$$

$$\mathcal{D} = [1 + 2(n_{\text{eq}} + (1 + 2n_{\text{eq}}) \cosh(2r_\nu)) \sin^2(|\gamma|)] \sinh(r_\nu). \quad (53)$$

Both observables oscillate with the frequency Ω ; however, there is a phase-shift $\delta\Omega$ between the two curves given by



$$\delta\Omega = \arctan \left[\frac{(\mathcal{A}\mathcal{D} + \mathcal{B}\mathcal{C}) \sin(\varphi_\nu - 2\theta_z)}{\mathcal{A}\mathcal{C} + \mathcal{B}\mathcal{D} - (\mathcal{B}\mathcal{C} + \mathcal{A}\mathcal{D}) \cos(\varphi_\nu - 2\theta_z)} \right]. \quad (54)$$

This is clearly visible in the plots of figure 4. Notice that this shift is constant in time; further, it vanishes as the squeezing parameter r_ν tends to zero. Therefore, in the specified physical situation, the detection of a phase shift between mean number and variance surely represents a clear signal of the presence of pump induced squeezed phonons.

Quadratic probing process. One can similarly analyze the case in which the most relevant mechanism accounting for the photon–phonon interaction is due to the quartic terms in the proposed hamiltonian; in this case, as discussed before for the thermal bath, no displacement can be generated in the phonon states in both pumping and probing processes. The most likely situation to occur due to the action of the pump is then the excitation of a

(i) *Squeezed thermal state.* In this situation, the mean photon number is found to oscillate with frequency 2Ω , while the corresponding variance has oscillating contributions with both frequencies 2Ω and 4Ω . Nevertheless, when the probe pulses are of low intensity, $|\alpha|^2 < 1$, both observables contain essentially only 2Ω oscillations, and in addition with exactly the same phase: the obtained behavior is qualitatively the same as the one shown in figure 2.

5. Outlook

Interactions of light with lattice vibrations have recently been the focus of many investigations. In particular, pump and probe techniques have been used to study coherent phonon dynamics, both in transparent and opaque materials. Most of these studies focused on detecting the presence of squeezed phonons using various experimental methods; however, all results so far obtained are based on the study of the mean reflectivity/transmittance of probe photons using semiclassical techniques. Squeezed states are characterized by quadrature fluctuations smaller than in vacuum, and this makes their experimental identification rather challenging. These facts, together with the debate that the various experiments stimulated, have kept the unquestionable identification of squeezed phonons an open problem.

As a contribution to the solution of this issue, in the present work a fully quantum description of both pumping and probing process has been developed. As a first step, a unique hamiltonian describing the effective coupling between phonons and photons has been introduced: it characterizes the impulsive interaction between the laser light and the lattice vibrations in the target material and can account for the excitations of both coherent and squeezed phonons.

Once excited by the pump, the phonons have been treated as an open quantum system, i.e. as a system in weak interaction with its external environment, here made by the electrons and impurities in the crystal. Two different physical situations have been investigated: first a thermal environment, where the phonon system reaches an asymptotic equilibrium thermal state at the bath temperature; second, a dephasing environment, preserving the number of phonons in the crystal target. In both settings, the corresponding master equation,

describing the dissipative phonon dynamics, admits an exact analytic solution allowing direct access to phonon correlations.

In the subsequent probing process, described by means of the same photon–phonon hamiltonian used for the pumping process, two different experimentally relevant frameworks have been analyzed: first, the case in which only bilinear contributions in photon and phonon operators are relevant, higher order terms being negligible due to the low intensity of the probe pulse; second, the case in which the only relevant contribution to the photon–phonon interaction comes from quartic contributions in photon and phonon operators. As previously discussed, these situations and the range of pump intensities can be related to the description of the photoexcited phonons in opaque and transparent materials.

In order to study the quantum properties of the pump excited phonons, it is not sufficient to focus on the behavior of average observables, like probe photon reflectivity/transmittance, as was done so far: higher order photon correlations need to be taken into account. We have thus considered two observables, the mean photon number and its variance, and analyzed in detail their evolution as functions of the delay time between pump and probe pulses. In this way, by studying jointly the dynamics of these two observables, clear signals for the presence of excited squeezed phonons have been identified.

More specifically, in the case of opaque materials, a distinctive signal of squeezing is given by the presence of a relative phase shift in the oscillating behavior of reflected probe photons mean and variance: it is non-vanishing only when the squeezing parameter r_r is nonzero. In presence of a thermal environment, the phase shift between the two curves is time dependent and vanishes after few oscillations; instead, for the dephasing environment, the phase shift is constant in time.

In the case of transparent materials, where higher intensities for pump and probe pulses are allowed, no phase shift between mean and variance can be observed. However, for the case in which the difference between pump and probe intensities is such that the probe process is dominated by linear mechanisms, i.e. linear probing process, the presence of oscillations in the transmittance with twice the frequency of the photoexcited vibrational mode is a signature of squeezing, reachable only in sophisticated experiments where thermal fluctuations can be reduced and only quantum fluctuations are observed [11]. More generally, when the most relevant contribution to the photon–phonon interaction is due to the quadratic terms in photon and phonon operators, it has not been possible to distinguish any feature clearly associated with the presence of squeezed phonon states by just looking at mean photon number and its variance; the study of higher order photon correlations is presumably needed.

As a further remark, note that for both opaque and transparent materials, in presence of a dephasing environment, oscillations with frequency 2Ω , i.e. twice the phonon mode frequency, can be observed in the behavior of the two observables even with vanishing squeezing parameter. Since any realistic environment is likely to contain a dephasing contribution, the presence of oscillations with such double frequency can not be in general considered a definite signal of squeezing. So, particular interest in the associated environment for the excited phonons is needed.

Finally, let us observe that the results and predictions of our investigation seem to be confirmed by recent, preliminary experimental findings. Indeed, under conditions similar to those analyzed here, experiments have been performed using both opaque [31] and transparent [10] target materials. In the case of bismuth, an opaque crystal, a damped oscillatory behavior has been reported for the mean reflectivity and its corresponding variance; in addition, a phase shift between these two observables is visible in the raw experimental curves. On the other hand, in order to excite coherent phonons in transparent materials, like the quartz crystals used in [10], more intense laser pulses are needed, making the excitation of squeezed phonons more likely. However, in this situation, no phase shift between mean photon number and variance is expected, but due to the particular setup and data treatment used in the experiments, purely quantum fluctuations in the modulation of the transmittance and its variance can be observed. The experimental results are matched and predicted by a phenomenological quantum description of the impulsive stimulated Raman scattering that fits within the purely quantum approach to pump and probe experiments developed in this manuscript.

The agreement between these experimental results and the predictions of our theoretical model is encouraging and we hope that it will stimulate additional experimental studies.

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Appendix A. Pump process: effective description

Here, we shall give a precise justification for the substitution of photon creation and annihilation operators by scalar quantities in the operator governing the pumping process as described in section 2.5.

We shall be quite general and take the effective hamiltonian describing the photon–phonon interaction of the form

$$H(\hat{a}, \hat{a}^\dagger) = \gamma \hat{a}^\dagger \hat{B} + \gamma^* \hat{a} \hat{B}^\dagger + \eta (\hat{a}^\dagger)^2 \hat{C} + \eta^* \hat{a}^2 \hat{C}^\dagger, \quad (55)$$

where \hat{a} and \hat{a}^\dagger are the photon annihilation and creation operators, while \hat{B} , \hat{B}^\dagger , \hat{C} and \hat{C}^\dagger are generic phonon operators. The dependence of the hamiltonian on \hat{a} , \hat{a}^\dagger has been made explicit because we want to justify here the substitution of $H(\hat{a}, \hat{a}^\dagger)$ with $H(\nu, \nu^*)$ where ν labels the coherent state $|\nu\rangle$ describing the photon state in the pumping laser beam. Denoting with $|\phi\rangle$ a generic pure phonon state, the initial state of the compound photon–phonon system is then in factorized form, $|\nu, \phi\rangle = |\nu\rangle \otimes |\phi\rangle$.

Proposition. Let \mathcal{O} be a self-adjoint operator depending on both phonon and photon variables and let

$$\mathcal{O}(t) = e^{itH(\hat{a}, \hat{a}^\dagger)} \mathcal{O} e^{-itH(\hat{a}, \hat{a}^\dagger)}$$

denote its time-evolution under the hamiltonian (55). Then

$$\begin{aligned} \langle \nu, \phi | \mathcal{O}(t) | \nu, \phi \rangle &= \langle \nu, \phi | e^{itH(\nu, \nu^*)} \mathcal{O} e^{-itH(\nu, \nu^*)} | \nu, \phi \rangle \\ &+ \int_0^t ds \Delta(\nu, s), \end{aligned} \quad (56)$$

where $\Delta(\nu, s)$ is a correction such that, for each fixed finite time $t \geq 0$, its integral from 0 to t can be made arbitrarily small by choosing $|\nu|$ large enough.

Proof. Let us consider the expectation value of the time-evolution equation for $\mathcal{O}(t)$ in the state $|\nu, \phi\rangle$:

$$\partial_t \langle \nu, \phi | \mathcal{O}(t) | \nu, \phi \rangle = i \langle \nu, \phi | [H(\hat{a}, \hat{a}^\dagger), \mathcal{O}(t)] | \nu, \phi \rangle. \quad (57)$$

Using $\hat{a} |\nu\rangle = \nu |\nu\rangle$ and the completeness relation for coherent states,

$$\frac{1}{\pi} \int d^2\beta |\beta\rangle \langle \beta| = \mathbb{I}, \quad d^2\beta = d\beta_1 d\beta_2, \quad \beta = \beta_1 + i\beta_2,$$

one finds

$$\begin{aligned} \partial_t \langle \nu, \phi | \mathcal{O}(t) | \nu, \phi \rangle &= i \langle \nu, \phi | (\gamma \nu^* \hat{B} + \eta (\nu^*)^2 \hat{C}) \mathcal{O}(t) \\ &- \mathcal{O}(t) (\gamma^* \nu \hat{B}^\dagger + \eta^* \nu^2 \hat{C}^\dagger) | \nu, \phi \rangle \\ &+ \frac{2}{\pi} \Re \left[\int d^2\beta \langle \nu, \phi | \mathcal{O} | \beta \rangle \langle \beta | (\gamma \beta^* \hat{B} + \eta (\beta^*)^2 \hat{C}) | \nu, \phi \rangle \right]. \end{aligned}$$

By passing to the new integration variable $\mu = \beta - \nu$, one reconstructs a hamiltonian operator with the photon operators being replaced by the complex scalars ν and ν^* :

$$\begin{aligned} \partial_t \langle \nu, \phi | \mathcal{O}(t) | \nu, \phi \rangle &= i \langle \nu, \phi | [H(\nu, \nu^*) \mathcal{O}(t)] | \nu, \phi \rangle \\ &+ \frac{2}{\pi} \Re \left(\int d^2\mu \langle \nu, \phi | \mathcal{O}(t) | \mu + \nu \rangle \langle \mu + \nu | \gamma \mu^* \hat{B} + \eta (\mu^*)^2 \hat{C} + 2\eta \mu^* \nu^* \hat{C} | \nu, \phi \rangle \right). \end{aligned} \quad (58)$$

In order to complete the proof, one needs to analyze the large $|\nu|$ behavior of the quantity

$$\begin{aligned} \Delta(\nu, t) &= \frac{2}{\pi} \Re \left[\int d^2\mu e^{-|\mu|^2/2} e^{-i\Im(\mu\nu^*)} \right. \\ &\times \left. \langle \phi | \langle \nu | \mathcal{O}(t) | \nu + \mu \rangle (\gamma \mu^* \hat{B} + \eta ((\mu^*)^2 + 2\mu^* \nu^*) \hat{C}) | \phi \rangle \right], \end{aligned} \quad (59)$$

where the scalar product of coherent states $\langle \mu + \nu | \nu \rangle = \exp[-|\mu|^2/2 - i\Im(\mu\nu^*)]$ has been used (\Im signifies imaginary part). We then consider separately the three terms that contribute to $\Delta(\nu, t)$; using Schwarz's inequality, one gets ($\mu = \mu_1 + i\mu_2$):

- First term:

$$\begin{aligned}
 & \left| \int d^2\mu e^{-|\mu|^2/2} e^{-i\Im(\mu\nu^*)} \mu^* \langle \nu, \phi | \mathcal{O}(t) \hat{B} | \mu + \nu, \phi \rangle \right| \\
 & \leq \int d\mu_1 d\mu_2 e^{-|\mu|^2/2} |\mu| \|\hat{B}^\dagger \mathcal{O}(t) | \nu, \phi \rangle\| \\
 & \leq \sqrt{2} \pi^{3/2} \|\hat{B}^\dagger \mathcal{O}(t) | \nu, \phi \rangle\|.
 \end{aligned} \tag{60}$$

- Second term:

$$\begin{aligned}
 & \left| \int d^2\mu e^{-|\mu|^2/2} e^{-i\Im(\mu\nu^*)} (\mu^*)^2 \langle \nu, \phi | \mathcal{O}(t) \hat{C} | \mu + \nu, \phi \rangle \right| \\
 & \leq \int d\mu_1 d\mu_2 e^{-|\mu|^2/2} |\mu|^2 \|\hat{C}^\dagger \mathcal{O}(t) | \nu, \phi \rangle\| \\
 & \leq 4\pi \|\hat{C}^\dagger \mathcal{O}(t) | \nu, \phi \rangle\|.
 \end{aligned} \tag{61}$$

- Third term:

$$\begin{aligned}
 & |\nu^*| \int d^2\mu e^{-|\mu|^2/2} e^{-i\Im(\mu\nu^*)} \mu^* \langle \nu, \phi | \mathcal{O}(t) \hat{C} | \mu + \nu, \phi \rangle \right| \\
 & \leq |\nu| \int d\mu_1 d\mu_2 e^{-|\mu|^2/2} |\mu| \|\hat{C}^\dagger \mathcal{O}(t) | \nu, \phi \rangle\| \\
 & \leq \sqrt{2} \pi^{3/2} |\nu| \|\hat{C}^\dagger \mathcal{O}(t) | \nu, \phi \rangle\|.
 \end{aligned} \tag{62}$$

Next, by using coherent states completeness, the following quantity

$$\begin{aligned}
 \int \frac{d^2\beta}{\pi} \|\hat{X}^\dagger \hat{Y} | \beta, \phi \rangle\|^2 &= \int \frac{d^2\beta}{\pi} \langle \beta, \phi | \hat{Y}^\dagger \hat{X} \hat{X}^\dagger \hat{Y} | \beta, \phi \rangle \\
 &= \langle \phi | \text{Tr}_{\text{photon}} (\hat{Y}^\dagger \hat{X} \hat{X}^\dagger \hat{Y}) | \phi \rangle,
 \end{aligned}$$

is finite for sufficiently well-behaved operators \hat{X} , \hat{Y} and phonon state $|\phi\rangle$. Then, by passing to polar coordinates, $d^2\beta = |\beta| d|\beta| d\theta$, the integrand of the integration over $|\beta|$, i.e. $|\beta| \|\hat{X}^\dagger \hat{Y} | \beta, \phi \rangle\|^2$, must vanish faster than $1/|\beta|$ when $|\beta| \rightarrow \infty$. As a result, by choosing $\hat{X} = \hat{B}$ or \hat{C} , and $\hat{Y} = \mathcal{O}(t)$, one sees that all three contributions listed above, and therefore $\Delta(\nu, t)$ itself, become arbitrarily small for large $|\nu|$. Finally, by integrating (58) over time, the statement of the Proposition follows. \square

Although a pure initial state has been used in the above proof, the results remains true also when the phonon state is described by a density matrix ρ instead of a pure state $|\phi\rangle$, since any density matrix can always be expressed as a convex combination of rank-one projectors. As the pumping laser beam is of high intensity, $|\nu|^2 \gg 1$, the substitution of \hat{a} with ν and \hat{a}^\dagger with ν^* in the operator \mathcal{U} representing the pumping process in section 2.5 appears now completely justified.

Appendix B. Probe photon correlations

As discussed in section 2.8, the explicit expressions of the correlation functions for the scattered/transmitted probe photons depend on the two mechanisms responsible for the probing process; the two situations need to be considered separately.

B.1. Linear probing process

When the photon–phonon interaction is dominated by processes in which the interaction between the light and the lattice vibrations is such that one photon is transformed into one phonon, only linear terms in phonon creation and annihilation operators are relevant in the expression (24) of the operator \mathcal{U}_α describing the probe process. In this case, since the quartic coupling η vanishes, its expression reduces to the one given in (28)

$$\mathcal{U}_\alpha = \exp(-i[\gamma \hat{a}^\dagger \hat{b} + \gamma^* \hat{a} \hat{b}^\dagger]). \tag{63}$$

In order to obtain probe photon correlations

$$\langle (\hat{a}^\dagger)^m \hat{a}^n \rangle(t) = \text{Tr}[\mathcal{U}_\alpha^\dagger (\hat{a}^\dagger)^m \hat{a}^n \mathcal{U}_\alpha |\alpha\rangle \langle \alpha| \otimes \rho(t)], \quad (64)$$

one has to compute the action of this operator on monomials in photon creation and annihilation operators. This can be done conveniently by introducing the bilinear operators

$$\begin{aligned} J_1 &= \frac{\gamma^* \hat{a} \hat{b}^\dagger + \gamma \hat{a}^\dagger \hat{b}}{2 |\gamma|}, \\ J_2 &= i \frac{\gamma^* \hat{a} \hat{b}^\dagger - \gamma \hat{a}^\dagger \hat{b}}{2 |\gamma|}, \\ J_3 &= \frac{\hat{a}^\dagger \hat{a} - \hat{b}^\dagger \hat{b}}{2}, \end{aligned} \quad (65)$$

that obey the $su(2)$ algebraic commutation relations

$$[J_i, J_j] = i \epsilon_{ijk} J_k. \quad (66)$$

For instance, since $\hat{a}^\dagger \hat{a} = N/2 + J_3$, with $N = \hat{a}^\dagger \hat{a} + \hat{b}^\dagger \hat{b}$ the total boson number, commuting with all J_j , one easily finds:

$$\mathcal{U}_\alpha^\dagger \hat{a}^\dagger \hat{a} \mathcal{U}_\alpha = \frac{N}{2} + \cos(2|\gamma|) J_3 + \sin(2|\gamma|) J_2. \quad (67)$$

Recalling (64) above, from this result one immediately gets the expression of the mean photon number $\langle \hat{a}^\dagger \hat{a} \rangle(t)$ reported in (29). Using similar techniques, one can compute the corresponding variance [35]

$$\begin{aligned} \Delta_{\hat{a}^\dagger \hat{a}}(t) &= |\alpha|^2 \cos^2(|\gamma|) + (\langle (\hat{b}^\dagger \hat{b})^2 \rangle(t) - \langle \hat{b}^\dagger \hat{b} \rangle^2(t)) \sin^4(|\gamma|) \\ &\quad + \langle \hat{b}^\dagger \hat{b} \rangle(t) (2|\alpha|^2 + 1) \cos^2(|\gamma|) \sin^2(|\gamma|) \\ &\quad - \frac{\cos^2(|\gamma|) \sin^2(|\gamma|)}{|\gamma|^2} [(\gamma^*)^2 \alpha^2 (\langle (\hat{b}^\dagger)^2 \rangle(t) - \langle \hat{b}^\dagger \rangle^2(t)) \\ &\quad + \gamma^2 (\alpha^*)^2 (\langle \hat{b}^2 \rangle(t) - \langle \hat{b} \rangle^2(t)) + 2|\gamma|^2 |\alpha|^2 \langle \hat{b}^\dagger \rangle \langle \hat{b} \rangle(t)] \\ &\quad + i \frac{\cos(|\gamma|) \sin(|\gamma|)}{|\gamma|} (\gamma^* \alpha \langle \hat{b}^\dagger \rangle(t) - \gamma \alpha^* \langle \hat{b} \rangle(t)) \\ &\quad + 2i \frac{\cos(|\gamma|) \sin^3(|\gamma|)}{|\gamma|} [\gamma^* \alpha (\langle \hat{b}^\dagger \hat{b} \rangle(t) - \langle \hat{b}^\dagger \rangle \langle \hat{b} \rangle(t)) \\ &\quad - \gamma \alpha^* (\langle \hat{b}^\dagger \hat{b}^2 \rangle(t) - \langle \hat{b}^\dagger \hat{b} \rangle \langle \hat{b} \rangle(t))], \end{aligned} \quad (68)$$

where $\langle (\hat{b}^\dagger)^m \hat{b}^n \rangle(t) = \text{Tr}_{\text{phonon}}[(\hat{b}^\dagger)^m \hat{b}^n \rho(t)]$, with $m, n \in \{0, 1, 2\}$, represent phonon correlations.

B.2. Quadratic probing process

When processes in which one photon is absorbed and one phonon is produced, and vice versa, are not contributing to the photon–phonon interaction, i.e. the coupling constant γ is zero, the operator \mathcal{U} implementing the probing process in (24) can be approximated by:

$$\mathcal{U}_\alpha = U_\alpha \left[\mathbb{I} - i \int_0^1 ds (U_\alpha^\dagger)^s (\eta ((\hat{a}^\dagger)^2 - (\alpha^*)^2) \hat{b}^2 + \eta^* (\hat{a}^2 - \alpha^2) (\hat{b}^\dagger)^2) (U_\alpha)^s \right], \quad (69)$$

with now

$$U_\alpha = \exp[-i(\eta (\alpha^*)^2 \hat{b}^2 + \eta^* \alpha^2 (\hat{b}^\dagger)^2)]. \quad (70)$$

Since U_α involves no photon operators but only phonon operators, the photon correlations in (64) can be expressed in terms of phonon correlations only. For instance, in the case of the mean number, one finds:

$$\langle \hat{a}^\dagger \hat{a} \rangle(t) = |\alpha|^2 - 2 \Re \left\{ 2i\eta (\alpha^*)^2 \int_0^1 ds \text{Tr}[(U_\alpha^\dagger)^s \hat{b}^2 U_\alpha^s \rho(t)] \right\}. \quad (71)$$

Further, notice that U_α acts on phonon states as a squeezing operator (see (15)), with squeezing parameter $\xi_\alpha = r_\alpha e^{i\varphi_\alpha}$ explicitly given by:

$$r_\alpha = 2|\eta||\alpha|^2, \quad \varphi_\alpha = 2\theta_\alpha + \frac{\pi}{2} - \theta_\eta,$$

where $\alpha = |\alpha| e^{i\theta_\alpha}$ and $\eta = |\eta| e^{i\theta_\eta}$. Then, one sees that

$$(U_\alpha^\dagger)^s \hat{b}^2 U_\alpha^s = \hat{b}^2 \cosh^2(sr_\alpha) + (\hat{b}^\dagger)^2 e^{2i\varphi_\alpha} \sinh^2(sr_\alpha) - (1 + 2\hat{b}^\dagger \hat{b}) e^{i\varphi_\alpha} \cosh(sr_\alpha) \sinh(sr_\alpha). \quad (72)$$

Inserting this result in (71), one finally obtains for the mean photon number the expression reported in (31). The expression (32) for the corresponding variance $\Delta_{\hat{a}^\dagger \hat{a}}(t)$ can be similarly computed [35].

Appendix C. Phase shift

As mentioned in section 4.1, in presence of a thermal bath and for low intensity probe beam, $|\alpha|^2 < 1$, with $|\nu|^2$ two orders of magnitude larger, both photon mean number and variance are seen to oscillate with the same frequency Ω , but with a relative phase shift. Indeed, explicit evaluation shows that the dominant contributions to these two photon observables are as follows [35]:

$$\langle \hat{a}^\dagger \hat{a} \rangle(t) \simeq -|\alpha| |z_\nu| e^{-\lambda_T t/2} \sin(2|\gamma|) [\mathcal{A} \sin(\Omega t + \phi_A) - \mathcal{B} \sin(\Omega t + \phi_B)] \quad (73)$$

and

$$\begin{aligned} \Delta_{\hat{a}^\dagger \hat{a}}(t) \simeq & -e^{-\lambda_T t/2} |z_\nu| |\alpha| (1 + 2n_f \sin^2(|\gamma|)) \sin(2|\gamma|) \\ & \times [\mathcal{A} \sin(\Omega t + \phi_A) - \mathcal{B} \sin(\Omega t + \phi_B)] \\ & - 2e^{-3\lambda_T t/2} |z_\nu| |\alpha| \sin^2(|\gamma|) \sin(2|\gamma|) [\tilde{\mathcal{A}} \sin(\Omega t + \phi_A) - \tilde{\mathcal{B}} \sin(\Omega t + \phi_B)]. \end{aligned} \quad (74)$$

where

$$\begin{aligned} \mathcal{A} &= \cosh(r_\nu), & \phi_A &= \theta_\alpha - \theta_\gamma - \theta_z, \\ \mathcal{B} &= \sinh(r_\nu), & \phi_B &= \theta_\alpha - \theta_\gamma + \theta_z - \varphi_\nu, \\ \tilde{\mathcal{A}} &= \cosh(r_\nu) [(1 + 2n_{\text{eq}}) \cosh(2r_\nu) - 1 - n_{\text{eq}} - n_f] \\ \tilde{\mathcal{B}} &= \sinh(r_\nu) [(1 + 2n_{\text{eq}}) \cosh(2r_\nu) + n_{\text{eq}} - n_f]. \end{aligned}$$

In absence of squeezing, $r_\nu = 0$, the coefficients \mathcal{B} and $\tilde{\mathcal{B}}$ vanish, and the two observables $\langle \hat{a}^\dagger \hat{a} \rangle(t)$ and $\Delta_{\hat{a}^\dagger \hat{a}}(t)$ oscillate in phase. A non vanishing relative phase shift $\delta\Omega$ is therefore a signal of the presence of squeezed phonons; its magnitude can be evaluated focusing on the position of the maxima (or minima) of the two functions in (73) and (74). Explicitly, one finds:

$$\delta\Omega = \left[\arctan\left(\frac{N}{D}\right) - \arctan\left(\frac{\tilde{N}(t)}{\tilde{D}(t)}\right) \right], \quad (75)$$

where

$$\begin{aligned} N &= 2\Omega [\mathcal{A} \sin(\phi_A) - \mathcal{B} \sin(\phi_B)] + \lambda_T [\mathcal{A} \cos(\phi_A) - \mathcal{B} \cos(\phi_B)], \\ D &= 2\Omega [\mathcal{A} \cos(\phi_A) - \mathcal{B} \cos(\phi_B)] - \lambda_T [\mathcal{A} \sin(\phi_A) - \mathcal{B} \sin(\phi_B)], \\ \tilde{N}(t) &= (1 + 2n_f \sin^2(|\gamma|))N + 2e^{-\lambda_T t} \sin^2(|\gamma|) [2\Omega (\tilde{\mathcal{A}} \sin(\phi_A) - \tilde{\mathcal{B}} \sin(\phi_B)) \\ & \quad + 3\lambda_T (\tilde{\mathcal{A}} \cos(\phi_A) - \tilde{\mathcal{B}} \cos(\phi_B))], \\ \tilde{D}(t) &= (1 + 2n_f \sin^2(|\gamma|))D + 2e^{-\lambda_T t} \sin^2(|\gamma|) [2\Omega (\tilde{\mathcal{A}} \cos(\phi_A) - \tilde{\mathcal{B}} \cos(\phi_B)) \\ & \quad - 3\lambda_T (\tilde{\mathcal{A}} \sin(\phi_A) - \tilde{\mathcal{B}} \sin(\phi_B))]. \end{aligned}$$

Note that $\delta\Omega$ depends on time through the coefficients $\tilde{N}(t)$ and $\tilde{D}(t)$; as t become large, the ratio $\tilde{N}(t)/\tilde{D}(t)$ approaches N/D and the phase shift becomes vanishingly small. As shown in figure 3, for typical experimental conditions, $\delta\Omega$ becomes unobservable after a few oscillations.

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