Generation and Detection of Squeezed Phonon

P. Munkhbaatar*
Department of Physics, School of Science, NUM, Ulaanbaatar 210646, Mongolia

The variance of the time dependent atomic positions and momenta is directly mapped into the quantum fluctuations of the photon number of the scattered probing light. A fully quantum description of the non-linear interactions between photonic and phononic fields unveils evidences of squeezing of thermal phonons. In here a pump-probe spectroscopy was proposed to observe phonon squeezing in solids. Numerical simulation showed pump-probe response due to coherent phonon and squeezed phonon in α-quartz.

PACS numbers: 78.47.J-, 42.70.Ce, 78.47.jd, 78.47.D-

I. INTRODUCTION

Development in femtosecond lasers and ultrafast spectroscopy has enabled us to generate and observe the coherent and squeezed lattice oscillations in which atomic motions are excited with light through a nonlinear process and the resulting dynamics is directly observed in the time domain.

In the non-linear spectroscopy formalism, impulsive stimulated Raman scattering (ISRS) is a non resonant excitation mechanism of lattice vibrations in transparent materials by ultrashort laser pulses. It is a four-wave mixing process due to third order polarization effects. The Cartesian components of the third order non-linear polarization are given by

$$P_i^{(3)}(\omega) = \sum_{ijkl} \chi_{ijkl}^{(3)} E_1(\omega_1)_j E_2(\omega_1)_k E_3(\omega_1)_l, \quad (1)$$

where $\chi_{ijkl}^{(3)}$ is the susceptibility tensor, E_1 is the probe field and E_2 and E_3 are the pump fields. The susceptibility tensor determines the polarization selection of vibrational modes that can be excited via ISRS[1]. In particular, quartz Raman active vibrational modes are 4 totally symmetric modes of symmetry A_1 and 8 doubly degenerate modes of symmetry E (transverse and longitudinal)[2].

Both pump and probe come from the same laser source. The excited phonon state is detected via the scattering of the probe pulse which arrives on the sample with time delay τ with respect to the pump. The transmitted light undergoes a polarization selection trough a polarizer positioned after the sample. The pump direction is almost collinear with the probe one. Assuming that the involved optical fields propagate along the z direction, we can limit our analysis to the xy plane. In this case the quartz Raman tensors assume the form[3]

$$A = \begin{pmatrix} a & 0 \\ 0 & a \end{pmatrix}, E^T = \begin{pmatrix} c & 0 \\ 0 & -c \end{pmatrix}, E^L = \begin{pmatrix} 0 & -c \\ -c & 0 \end{pmatrix}. \quad (2)$$

Following the notation in[4], the susceptibility tensor can be expressed as:

$$\chi_{ijkl}^{(3)} = A_{ij}A_{kl} + E_{ij}^T E_{kl}^T + E_{ij}^L E_{kl}^L. \quad (3)$$

Where each index can assume the values 1, 2 associated to the direction x and y respectively. The first two indexes describe the polarization components of the emitted field (i index) and of the probe field (j index), while the last two indexes (k and l) describe the polarization components of the two pump fields.

In particular, we are interested in selecting the excitation of an E symmetry Raman mode. For this purpose we use a probe linearly polarized along x and we perform a polarization selection after the sample in order to detect the emitted field component orthogonal to the probe (along y). This polarization configuration allows the selection of the susceptibility matrix elements $\chi_{21kl}^{(3)}$ associated with the involved E phonon mode. Notice that such elements vanish when $k = l$ that is when the two pump fields are both polarized along x or along y. Thus, in order to activate the process, we need the two pump fields (two frequency components of the same laser pulse) to have orthogonal polarizations. This is possible when the pump pulse is linearly polarized along a direction in between x and y. In particular, the efficiency of the ISRS is maximal when the pump polarization is at 45 with respect to the x axis. This is indeed the configuration we chose and consequently the matrix elements involved in our experiment are $\chi_{21kl}(k \neq l) = c^2$, getting an emitted field almost collinear with the unscattered probe photons and with polarization orthogonal to the probe one. We configure a polarizer after the sample in order to transmit the emitted field polarization only. The residual probe acts as a local oscillator amplifying the emitted field within the total signal[5].

*Electronic address: p_munhkbaatar@yahoo.com

II. GENERATION AND DETECTION OF COHERENT AND SQUEEZED PHONON

A. Generation

In order to address the fingerprint of the squeezed nature of the photo-excited vibrational state, I provide here a fully quantum model for time domain experiments. Standard theoretical descriptions of ISRS adopt semi classical approaches where the dipolar interactions in the crystal are treated quantum mechanically while the optical fields are described classically[6].

Before being hit by the pump laser beam, the relevant phonon mode at frequency ω can be appropriately described by a thermal state at inverse temperature

$$\rho = (1 - e^{\beta\omega})\, e^{\beta\omega b^\dagger b}. \tag{4}$$

The pump process is described by a photon-phonon interaction. In mean field approximation, the pump thus prepares the relevant phonon degree of freedom in a state ρ^* which is obtained by tracing over the photon degrees of freedom: $\rho_b = U\rho U^\dagger$. Where

$$U = \exp\{-i(c_1 b + c_1^*\, b^\dagger + c_2 b^2 + c_2^*\, b^{\dagger 2})\}. \tag{5}$$

c_1, c_2 are defined by pump modes in Ref[7]. The evolution operator generates coherent and squeezed phonon states, respectively, through the linear and quadratic terms in the phonon operators b and $b^\dagger$. The parameters β, ω, c_1 and c_2 used in the calculation are 5, 2, 5 and $-0.5i$, respectively.

B. Time evolution of coherent and squeezed phonon

The photoexcited phonon state ρ_b then undergoes a dissipative dynamics that effectively takes into account the interaction of the phonons with their environment until, after a delay time t , the target is hit by the probe laser beam. The phonon dynamics is considered to be that of an open quantum system in weak interaction with a large heat bath that will eventually drive the time-evolving phonon density matrix $\rho(t)$ to a thermal state $\rho_{\beta'}$ at temperature T' larger than that of the pre-pump phonon state: $\beta' < \beta$. Such a relaxation process is described by a master equation[8] for the phonon density matrix $b(t)$ of the form $\partial\rho_b(t)/\partial t = L[\rho_b(t)]$, where the generator of the time-evolution is given by

$$\begin{aligned} L(\rho_b(t)) \;=\; & -i[\omega b^\dagger b, \rho_b(t)] \\ & +\lambda(1+n')\left(b\rho_b(t)b^\dagger - \frac{1}{2}\left\{b^\dagger b, \rho_b(t)\right\}\right) \\ & +\lambda n'\left(b^\dagger\rho_b(t)b - \frac{1}{2}\left\{b\, b^\dagger, \rho_b(t)\right\}\right). \end{aligned}$$

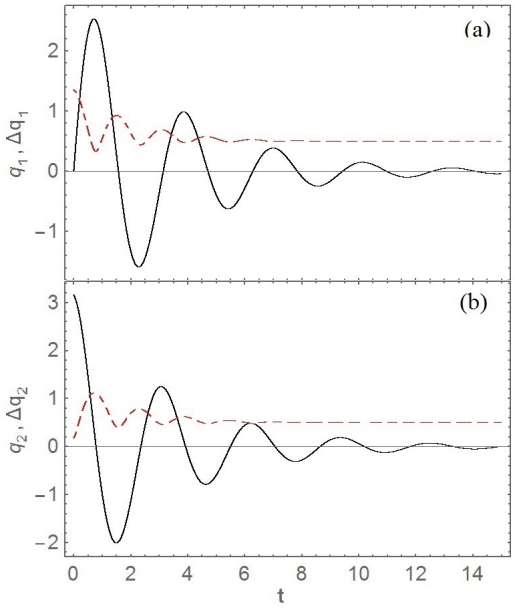

FIG. 1: Time dependence of two quadrature operators. Variances of the operators are shown by the dashed lines, respectively.

where $n' = \frac{1}{e^{\beta'\omega}-1}$, while λ is a coupling constant sufficiently small so that the non-negligible presence of the environment can nonetheless be accounted for, in the so-called weak-coupling limit regime[8, 9], by a master equation of the above type.

The parameters β' and λ used in the calculation are 4 and 0.6, respectively.

The first term of L generates the rotation in time of the phonon mode phase at its own eigen-frequency. The second two contributions consist of a so-called noise term $b\rho_b(t)b^\dagger$, respectively $b^\dagger\rho_b(t)b$ that has the property of transforming pure states into mixed states and of a dissipative term $-(1/2)\{b^\dagger\, b, \rho_b(t)\}$, respectively $-(1/2)\{b\, b^\dagger, \rho_b(t)\}$.

Using density matrix given by equation we can study the dynamical properties of the system under consideration. Firstly we calculate the expectation value of the two quadrature operators given by

$$q_1 = (a^\dagger + a)/2, \quad q_2 = i(a^\dagger - a)/2.$$

Figure 2(a) and (b) shows q_1 and q_2, i.e. time dependence of two quadrature operators, respectively.

Now we turn our attention to the dynamics of the $Q(\alpha, \alpha^*)$ function. Quasiprobability distributions arise naturally in the study of quantum mechanics when treated in phase space formulation, commonly used in quantum optics, time-frequency analysis. One of such function is the Q Husimi function. Q always remains positive and bounded:

$$Q(\alpha, \alpha^*) = \frac{1}{\pi}\langle\alpha|\hat{\rho}|\alpha\rangle.$$

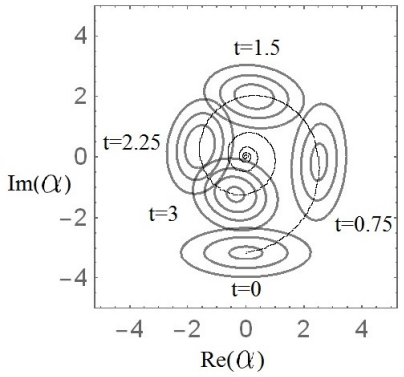

FIG. 2: Contour lines of $Q(\alpha, \alpha^*, t)$ in the complex α plane for coherent and squeezed phonon, at the times $t_0 = 0$, $t_1 = 0.75$, $t_3 = 1.5$, $t_4 = 2.25$, $t_5 = 3.0$. The thin line indicates the time evolution of center on quasidistribution function.

In figure 2, the contour plots of the $Q(\alpha, \alpha^*)$ function are shown for five different values of the times.

C. Detection

Finally, the probe process is again described by interaction phonon-photon. However, the corresponding impulsive unitary operator U now acts on a photon-phonon state of the form $\rho_{\overline{\alpha}} \otimes \rho_b(t)$. Here, $\rho_{\overline{\alpha}}$ is the multi-mode coherent state associated with the probe laser beam which contains x and y polarized components and is much smaller intense than the pump one, while $\rho_b(t)$ is the phonon state dissipatively evolved up to the delay time τ between pump and probe. Differently from the pump process, the lower probe intensity allows one to neglect in H the terms responsible for the squeezing effects. Moreover, we can apply the mean field approximation only to the field operators with x polarization, since the x-polarized probe components are much more intense than those polarized along y. Then, by replacing a_{xj} and $a^\dagger_{xj}$ by α_{xj} and α^*_{xj} the probe process is described by

$$U_{\alpha'} = \exp\{-i||\alpha'||(Ab^\dagger + A^\dagger b)\}, \qquad (6)$$

where A is the collective photon annihilation operator[7].

We can thus consistently let the probe process affect an initial state $|\alpha'\rangle\langle\alpha'| \otimes \rho_b(t)$, where $|\alpha'\rangle$ is the coherent state involving only the y polarization components. Notice that, unlike in, $U_{\alpha'}$ acts on the photon-phonon state as a whole and transforms it into

$$U_{\alpha'}|\alpha'\rangle\langle\alpha'| \otimes \rho_b(t) U^\dagger_{\alpha'}.$$

Unlike in the semi-classical theoretical approaches to pump and probe experiments attempted so far, one can here confront the experimental data with the scattered probe beam intensity, namely with the mean photon number $\langle N_y\rangle_\tau$, where $N_y = A^\dagger A$ and and with its variance $\Delta_\tau N_y^2 = \langle N_y^2\rangle_\tau - \langle N_y\rangle^2_\tau$. Then one uses that

$$\begin{aligned} U^\dagger_{\alpha'} N_y U_{\alpha'} &= A^\dagger A \cos^2(|\alpha|^2) + b^\dagger b \sin^2(|\alpha'|) \\ &+ \frac{i}{2}\sin(2|\alpha'|)(A\, b^\dagger + A^\dagger\, b). \end{aligned}$$

Thus, we can explicitly compute:

$$\begin{aligned} \langle N_y\rangle_\tau &= I_y \cos^2(|\alpha'|) + \langle b^\dagger b\rangle_\tau \sin^2(|\alpha'|) \\ &+ \frac{i}{2}\sqrt{I_y}\sin(2|\alpha'|)\langle b^\dagger + b\rangle_\tau. \end{aligned}$$

where I_y is the intensity of probe light.

Despite its complicated expression, we report also the number variance predicted by the model as $\langle N_y\rangle_\tau$ and $\langle \Delta N_y^2\rangle_\tau$ are the quantities computed numerically:

$$\begin{aligned} \langle \Delta N_y^2\rangle_\tau &= I_y \cos^4(|\alpha'|) + \sin^4(|\alpha'|)(\langle (b^\dagger b)^2\rangle_\tau - \langle b^\dagger b\rangle^2_\tau) + \sin^2(|\alpha'|)\cos^2(|\alpha'|)\langle b^\dagger b\rangle_\tau \\ &- I_y \sin^2(|\alpha'|)\cos^2(|\alpha'|)[\langle b^{\dagger 2}\rangle_\tau - \langle b^\dagger\rangle^2_\tau + \langle b^2\rangle_\tau - \langle b\rangle^2_\tau] \\ &+ I_y \sin^2(|\alpha'|)\cos^2(|\alpha'|)[2\langle b^\dagger b\rangle_\tau + 1 - 2\langle b^\dagger\rangle_\tau\langle b\rangle_\tau] \\ &+ i\sqrt{I_y}\sin(|\alpha'|)\cos^3(|\alpha'|)[\langle b^\dagger\rangle_\tau - \langle b\rangle_\tau] \\ &+ i\sqrt{I_y}\sin^3(|\alpha'|)\cos(|\alpha'|)[2(\langle b^{\dagger 2} b\rangle_\tau - \langle b^\dagger b\rangle_\tau\langle b^\dagger\rangle_\tau + \frac{1}{2}\langle b^\dagger\rangle_\tau) \\ &- 2(\langle b^\dagger b^2\rangle_\tau - \langle b^\dagger b\rangle_\tau\langle b\rangle_\tau + \frac{1}{2}\langle b\rangle_\tau)]. \end{aligned} \qquad (7)$$

Figure 3 shows pump-probe signal or mean value and the variance of the number operator. The parame-

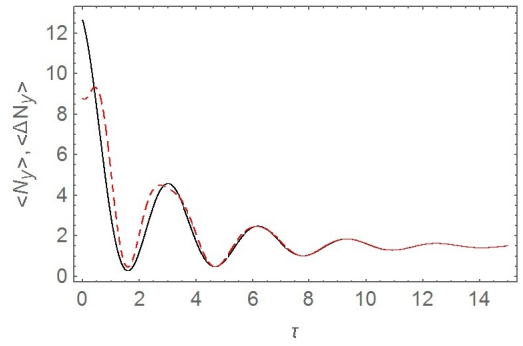

FIG. 3: Theoretical calculations of the mean value and the variance of the number of photons of the emitted field. Variance of the operator are shown by the dashed line.

ters α' and I_y used in the calculation are $\pi/4$ and 1, respectively. As the theoretical results, deference of mean value and the variance of the number of emitted photons is due to squeezed phonon. It is a evidence of squeezing.

For a given time delay τ a reference pulse, which has not interacted with the sample, is subtracted from each pulse transmitted through the sample by a differential detector capable of single pulse acquisition. The integral of the voltage produced by the differential pulse constitutes the measurement of the transmittance variation $\Delta T(\tau)$. The mean value $\Delta T(\tau)$ and the variance $\Delta T(\tau)$ can be measured for each time delay τ.

III. CONCLUSION

A pump-probe spectroscopy method was proposed to investigate of squeezed phonon. The fluctuations of the time dependent atomic positions and momenta are mapped directly into the quantum fluctuations of the probe photon number, thereby providing an absolute reference for the vibrational quantum noise.

Finally, the generality of our fully quantum approach, used here to reveal the squeezed nature of photo-excited phonon states in quartz, offers a novel framework to unveil distinctive quantum properties of vibrational states in matter. This paves the way for future studies addressing the thermodynamics of lattice and molecular vibrations of complex systems, possibly, in quantum regimes.

Acknowledgments

This research was supported by the National Research Foundation of Mongolia (Grants No. SSA_014/2016 and SSA_017/2016).

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