In this letter, the recently predicted Side-band Inequivalence in quantum optomechanics is analyzed to the full order. It is shown that not only frequency shifts due to blue- and red-scattered (Stokes and anti-Stokes) mechanical side-bands are counter-intuitively inequal, but also there exists a maximum attainable side-band inequivalence which may be expected at an optimal operation point. The mathematical method employed here is a combination of operator algebra equipped with harmonic balance, which allows a clear understanding of the associated nonlinear process. This reveals the existence of three distinct operation regimes in terms of pump power or intracavity photon number, two of which have immeasurably small side-band inequivalence. Compelling evidence from Raman scattering data of various materials unambiguously confirms existence of a previously unnoticed side-band inequivalence.

PACS numbers: 42.50.-p, 42.50.Lc, 42.65.Dr

The nonlinearity of optomechanical interaction [1–9] causes scattering of incident photons with the annihilator $\hat{a}$ from the cavity unto either red- or blue-shifted photons through annihilation or generation of a cavity phonon with the annihilator $\hat{b}$, giving rise to the first-order mechanical side-bands. Taking the optical frequency $\omega$ to be at a detuning $\Delta = \omega_c - \omega$ from cavity resonance $\omega_c$, the $\nu$-th order sidebands are naturally expected to occur at the detunings $\Delta_{\pm \nu} = \Delta \mp \nu \Omega$, where $\Omega$ represents the mechanical frequency. As a results, the first-order mechanical side-bands of scattered red $\Delta_{+1}$ and blue $\Delta_{-1}$ processes must average out back to the original pump detuning $\Delta$.

Defining the side-band inequivalence as the deviation of this average from $\Delta$, as $\delta = \frac{1}{2}(\Delta_{+1} + \Delta_{-1}) - \Delta$, then one may conclude $\delta = 0$. A non-zero $\delta$ would have otherwise implied the so-called Side-band Inequivalence. This type of asymmetry appears to have a classical nonlinear nature, and is illustrated in Fig. 1.

There is, however, another well-known type of side-band asymmetry between the red and blue side-bands in the context of optomechanics, which has a quantum nature and may be used for instance to accurately determine the absolute temperature through a reference-free optomechanical measurement [10, 11]. This is based on the ratio of Stokes to anti-Stokes Raman transition rates, which is equal to $\exp(\hbar \Omega/k_B T)$ where $k_B$ is the Boltzmann’s constant and $T$ is the absolute temperature [12, 13]. Also illustrated in Fig. 1 clearly, side-band inequivalence is quite different from this type of side-band asymmetry.

While both time-reversal symmetry and energy conservation are fundamentally preserved in this scattering process, a nonlinear analysis of quantum optomechanics using the recently developed method of higher-order operators [14–18] necessitates a slight difference among detunings of blue and red-scattered photons, the amount of which was initially found to increase roughly proportional to the intracavity photon number $\bar{n}$. Here, $\bar{n}$ is defined as the steady-state mean-value of the number operator $\hat{n} = \hat{a}^\dagger \hat{a}$.

Surprisingly enough, this disagreement satisfying $\delta \neq 0$ does not violate the energy conservation law, actually allowed by the finite cavity linewidth as well as the single-photon/single-phonon nature of the process involved. Moreover, the time-reversal symmetry is also preserved.

Among the pool of available experimental data, only a handful of side-band resolved cavities reveal this disagreement [14]. Some initial trial experiments recently done at extremely high intracavity photon numbers $\bar{n}$, and/or extremely large single-photon optomechanical interaction rates $g_0$, though, failed to demonstrate its existence. This may raise the speculation that whether side-band inequivalence would have been merely a mathematical artifact, or something has been missing due to not doing the operator analysis to the highest-order.

**FIG. 1.** Illustration of two basic symmetry breakings in quantum optomechanical cavity at resonant pump, and in absence of cooling tone: (a) Side-band Inequivalence – The red detuning exceeds that of blue detuning, shown in blue dashed arrows; (b) Side-band Asymmetry – The red peak exceeds that of blue peak, shown in black dash-dotted arrows. Here, $S_{AA}(\Delta)$ represents the spectral noise density.
A careful analysis of this phenomenon, however, confirms the latter, thus classifying the quantum optomechanical interaction into three distinct regimes with different behaviors:

- **Fully Linear**: This regime can be investigated using the lowest-order analysis and first-order operators, which is conventionally done by linearizing the Hamiltonian around equilibrium points. This will require the four-dimensional basis of first-order ladder operators \( \{\hat{a}, \hat{a}^\dagger, \hat{b}, \hat{b}^\dagger\} \) and is indeed quite sufficient to understand many of the complex quantum optomechanical phenomena [19].

- **Weakly Nonlinear**: This regime requires higher-order operator analysis of at least second-order. This can be done using the three-dimensional reduced basis [14, 19] given as \( \{\hat{n}^2, \hat{a}b, \hat{ab}^\dagger\} \).

- **Strongly Nonlinear**: Full understanding of this interaction regime requires the highest-order analysis using third-order operators. Referred to as the minimal basis [13, 19], the convenient reduced choice is the two-dimensional basis \( \{\hat{n}^2, \hat{a}b\} \). While most of the quantum optomechanical experiments happen to fall in this regime, the striking behavior of governing equations is in such a way that a fully-linearized analysis of fluctuations mostly happens to work.

Side-band Inequivalence is essentially forbidden in the fully linear regime, and it also quickly fades away in the strongly nonlinear regime. But it may only happen in the weakly nonlinear regime. This is now also confirmed both by the higher-order operator method and extensive calculations. It typically does not exceed one part in a million to one part in ten thousand, and therefore, it is a very delicate phenomenon and elusive to observe.

This letter provides a direct route towards clear understanding of this complex nonlinear phenomenon. Using a combination of operator algebra and harmonic balance (used in analysis of laser diodes) [20], we obtain a closed form expression for side-band inequivalence \( \delta \) as a function of intracavity photon number \( \bar{n} \), which is expected to be valid through all three above operation regimes, and for any arbitrarily chosen set of optomechanical parameters. It can be shown that there is an optimal point at which the side-band inequivalence attains a maximum. Moving away from the optimal point, both at the much smaller and much larger pump rates, \( \delta \) attains much smaller values, tending to zero in the limit of very large \( \bar{n} \).

This will be greatly helpful to designate the investigation range of experimental parameters given any available optomechanical cavity. Furthermore, it marks a clear and definable border among the three above-mentioned operation regimes.

The analysis of side-band inequivalence proceeds with considering the behavior of optomechanical cavity under steady-state conditions. We will focus only on the first-order side-bands and discard all other contributions coming from or to the second- and higher-order side-bands. We consider a single-frequency pump with ideally zero linewidth at a given detuning \( \Delta \), which normally gives rise to two stable blue- and red- side-bands. Hence, the time-dependence of the photon annihilator will look like

\[
\hat{a}(t) = \hat{a}_0 e^{i\Delta t} + \hat{a}_b e^{i(\Delta + \frac{\delta}{2})t} + \hat{a}_r e^{i(\Delta - \frac{\delta}{2})t} + \cdots, \tag{1}
\]

where \( \hat{a}_0, \hat{a}_b, \) and \( \hat{a}_r \) respectively correspond to the central excitation resonance at pump frequency, and blue- and red-detuned side-bands. The steady-state time-average of central excitation satisfies \( \langle \hat{a}_0 \rangle = \sqrt{n} \), where \( n \) can be determined by solution of a third-order algebraic equation once the optical power pump rate \( P_{op} \), detuning \( \Delta \), external coupling \( \eta \) and all other optomechanical parameters are known. The standard set of basic optomechanical parameters needed here are mechanical frequency \( \Omega \), optical decay rate \( \kappa \), and mechanical decay rate \( \Gamma \). Therefore, the photon number operator up to the first side-bands will behave as

\[
\hat{n}(t) = \hat{a}_0^\dagger \hat{a}_0 + \hat{a}_b^\dagger \hat{a}_b + \hat{a}_r^\dagger \hat{a}_r \tag{2}
\]

\[
+ \hat{a}_b^\dagger \hat{a}_b e^{-i\left(\Omega + \frac{\delta}{2}\right)t} + \hat{a}_0^\dagger \hat{a}_0 e^{-i\left(\Omega - \frac{\delta}{2}\right)t} + \hat{a}_r^\dagger \hat{a}_r e^{-i\left(\Omega - \frac{\delta}{2}\right)t} + \cdots,
\]

while the mechanical annihilator will exhibit a closely spaced doublet around the mechanical frequency spaced within \( \delta \) as

\[
\hat{b}(t) = \hat{b}_0 + \hat{b}_b e^{-i\left(\Omega - \frac{\delta}{2}\right)t} + \hat{b}_r e^{-i\left(\Omega + \frac{\delta}{2}\right)t} + \cdots. \tag{3}
\]

Here, the average mechanical displacement satisfies

\[
\hat{b}_0 = \langle \hat{b}_0 \rangle = \frac{i\gamma_0 \bar{n}}{\Omega + \frac{\delta}{2}}. \tag{4}
\]

Now, let us get back to the Langevin equation for mechanical motions, which simply is

\[
\frac{d}{dt} \hat{b}(t) = -(i\Omega - \frac{\delta}{2})\hat{b}(t) + i\gamma_0 \hat{n}(t) + \sqrt{\gamma_0} \hat{b}_m(t), \tag{5}
\]

where \( \hat{b}_m(t) \) is the operator for mechanical fluctuations. For the purpose of our analysis here, all fluctuations can be discarded since they are irrelevant to the formation of side-band frequencies and average out to zero. Using (2) and (3) we get

\[
- \frac{i}{\Omega + \frac{\delta}{2}} \hat{b}_r e^{-i(\Omega + \frac{\delta}{2})t} - \frac{i}{\Omega - \frac{\delta}{2}} \hat{b}_b e^{-i(\Omega - \frac{\delta}{2})t} \tag{6}
\]

\[
\approx -(i\Omega + \frac{\Gamma}{2})\hat{b}_r e^{-i(\Omega + \frac{\delta}{2})t} - (i\Omega + \frac{\Gamma}{2})\hat{b}_b e^{-i(\Omega - \frac{\delta}{2})t} + i\gamma_0 \hat{a}_0^\dagger \hat{a}_0 e^{-i(\Omega - \frac{\delta}{2})t} + i\gamma_0 \hat{a}_r^\dagger \hat{a}_r e^{-i(\Omega + \frac{\delta}{2})t} + \cdots.
\]

From the above, we obtain two key operator equations

\[
\hat{b}_r = \frac{i2\gamma_0}{-i\delta + \Gamma} \hat{a}_b^\dagger \hat{a}_0, \tag{7}
\]

\[
\hat{b}_b = \frac{i2\gamma_0}{i\delta + \Gamma} \hat{a}_0^\dagger \hat{a}_b.
\]
In a similar manner, the Langevin equation for the photon annihilation is
\[
\frac{d}{dt} \hat{a}(t) = (i\Delta - \frac{1}{2} \kappa) \hat{a}(t) + ig_0 \hat{a}(t)[\hat{b}(t) + \hat{b}^\dagger(t)] + \sqrt{n} \hat{a}_m. \tag{8}
\]

Using (1) and (3) we obtain
\[
i \Delta \hat{a}_0 e^{i\Delta t} + i(\Delta - \Omega + \frac{\delta}{2}) \hat{a}_0 e^{i(\Delta + \Omega + \frac{\delta}{2}) t} \tag{9}
\]
\[
+ i(\Delta - \frac{\kappa}{2}) \left[ \hat{a}_0 e^{i\Delta t} + \hat{a}_0 e^{i(\Delta + \Omega + \frac{\delta}{2}) t} \right] \approx
\]
\[
+ i \left[ \frac{i(\Omega + \frac{\delta}{2}) + \frac{\kappa}{2}}{i \delta} \right] \hat{b}_0 e^{i(\Omega + \frac{\delta}{2}) t} + \hat{b}_0 e^{i(\Omega - \frac{\delta}{2}) t}
\]
\[
+ \hat{b}_0 e^{i(\Omega + \frac{\delta}{2}) t} + \hat{b}_0 e^{i(\Omega - \frac{\delta}{2}) t} \right].
\]

where \( \hat{x}_0 = \hat{b}_0 + \hat{b}_0^\dagger \). This will yield the further operator equations as
\[
\frac{\kappa}{2 i \delta} \hat{a}_0 = \hat{a}_0 \hat{x}_0 + \hat{a}_0 \hat{b}_0^\dagger + \hat{a}_0 \hat{b}_r, \tag{10}
\]
\[
\left[ \frac{i(\Omega + \frac{\delta}{2}) + \frac{\kappa}{2}}{i \delta} \right] \hat{a}_0 = \hat{a}_0 \hat{x}_0 + \hat{a}_0 \hat{b}_0^\dagger,
\]
\[
\left[ \frac{i(\Omega + \frac{\delta}{2}) + \frac{\kappa}{2}}{i \delta} \right] \hat{a}_r = \hat{a}_r \hat{x}_0 + \hat{a}_0 \hat{b}_r^\dagger.
\]

Now, substituting whatever we have in hand in the second equation of (10), and taking expectation values at the end, we obtain a key algebraic equation in terms of \( \delta \) as
\[
i \left(-\Omega + \frac{\delta}{2}\right) + \frac{\kappa}{2} = ig_0 \Delta x_0 + ig_0 \sqrt{2 i \delta / \Omega} \tag{11}
\]
\[
\text{with } x_0 = b_0 + b_0^\dagger. \text{ Rearrangement of the above gives rise to the equation}
\]
\[
\delta^2 - [2\Omega + i\gamma + 2g_0 x_0] \delta
\]
\[
+ \left[ (2\Omega - \kappa)\Gamma - 4g_0^2 \hat{n} + 2i\Gamma g_0 x_0 \right] = 0,
\]
\[
in which \( x_0 = b_0 + b_0^\dagger \) and \( \gamma = \kappa + \Gamma \) is the total optomechanical decay rate \( \Gamma \).
\]

This approximate nature of this equation will yield complex values for \( \delta \) the imaginary value of which has to be discarded. Furthermore, it leaves room to ignore the the square terms \( \delta^2 \), to admit the solution
\[
\delta = \Re \left[ \frac{(2\Omega - \kappa)\Gamma - 4g_0^2 \hat{n} + 2i\Gamma g_0 x_0}{2\Omega + i\gamma + 2g_0 x_0} \right]. \tag{13}
\]

This solution can be put into the more convenient form using (11) and further simplification as
\[
\delta(\hat{n}) = \Re \left[ \frac{A + B\hat{n}}{C - iD\hat{n}} \right]. \tag{14}
\]
\[
= \frac{\Re[AC^*] + (B\Re[C] - 3|A|D)\hat{n}}{|C|^2 - 2\Re[C]D\hat{n} + D^2\hat{n}^2}
\]
\[
= \frac{2i\Omega^2 + 2\Omega(B - i\Gamma D)\hat{n}}{|C|^2 - 4\Omega D\hat{n} + D^2\hat{n}^2},
\]
\[
\text{where}
\]
\[
A = \Gamma(2\Omega - \kappa),
\]
\[
B = 4g_0^2(\Omega - \frac{1}{2}\Gamma)^2 / (\Omega^2 + \frac{1}{4}\Gamma^2) = B^*,
\]
\[
C = i\gamma + 2\Omega,
\]
\[
D = \frac{4g_0^2\Omega}{\Omega^2 + \frac{1}{4}\Gamma^2} = D^*.
\]

The expression (14) obtained for the side-band inequivalence has interesting properties at the limits of zero and infinite intracavity photon number. We may obtain here after some simplification easily the asymptotic expressions
\[
\lim_{\hat{n} \to \infty} \delta(\hat{n}) \sim \frac{\Omega}{\beta\hat{n}}, \quad \beta = 2g_0^2 / \Omega^2,
\]
\[
\lim_{\hat{n} \to 0} \delta(\hat{n}) \sim \frac{2\Gamma^2\Omega}{4\Omega^2 + \gamma^2} = 0,
\]
\[
\text{where } \beta = 2g_0^2 / \Omega^2, \text{ while noting that } \Gamma < \Omega \text{ and also for a side-band resolved cavity } \kappa < \Omega, \text{ together which we have } \kappa < \gamma < \Omega.
\]

One should take into account the fact that for Doppler cavities, side-bands normally resolve well enough for a decisive measurement \( |\bar{\hat{n}}| \), and the concept of side-band inequivalence is only practically meaningful for side-band resolved cavities. Therefore, the following approximations are valid
\[
A \approx 2\Gamma\Omega,
\]
\[
B \approx 4g_0^2 / \Omega^2,
\]
\[
C \approx 2\Omega, \quad D \approx \frac{4g_0^2}{\Omega^2},
\]
\[
\delta(\hat{n}) \approx \frac{2\Gamma^2\Omega + 8g_0^2\Omega\bar{\hat{n}}}{\gamma^2 + 4\Omega^2 \left[ 1 - 2(g_0 / \Omega)^2 \right] \bar{\hat{n}}^2}.
\]

As long as satisfies \( \bar{\hat{n}} < \frac{2|\Re[C]|}{D} = \frac{\Omega^2 / g_0^2}{\Omega} \), then second order term \( \bar{\hat{n}}^2 \) in the denominator of (15) is negligible and can be ignored. Under this regime, the side-band inequivalence varies almost linearly with \( \bar{\hat{n}} \) as
\[
\delta(\bar{\hat{n}}) \approx \frac{2\Gamma^2\Omega}{4\Omega^2 + \gamma^2 + \frac{8g_0^2\Omega(4\Omega^2 + \gamma^2 + 2\Gamma^2)}{\gamma^2 + 4\Omega^2} \bar{\hat{n}}}
\]
\[
\approx \frac{2\Gamma^2\Omega}{\Omega} \bar{\hat{n}}. \tag{18}
\]

This result is also well in complete agreement with the expression obtained earlier for the side-band inequivalence \( |\bar{\hat{n}}| \) in the limit of \( g_0 < \Omega \) given as \( g_0^2 / \Omega^2 \), considering
that a factor of $\frac{1}{2}$ must be added as a result of different definition of $\delta$.

The first immediate conclusion which can be obtained from (18) is that the side-band inequivalence $\delta$ is always positive, meaning that the detuning frequency of red-sideband should always be a bit larger in magnitude than the blue-sideband. This also agrees with the previous findings of higher-order operator algebra [14].

The unique mathematical form of (14) which is composed of a first- and second-order polynomials in terms of $\bar{n}$ respectively, offers a clear maximum at a certain optimum intracavity photon number $\bar{n}_{\text{max}}$. To do this, let us first define the dimensionless constants $\alpha = 4g_0^2/\Gamma^2$ and $\beta$ already defined under (16), $\vartheta = \gamma/2\Omega$, and $\psi = \Gamma^2/2\Omega^2$. Then, the side-band inequivalence (17) can be rewritten as

$$\delta(\bar{n}) = \Omega\vartheta\bar{n} \left[ 1 + \frac{\alpha\bar{n}}{\vartheta^2 + (1 - \beta\bar{n})^2} \right].$$

This offers the optimum intracavity photon number and thus the maximum attainable side-band inequivalence as

$$\bar{n}_{\text{max}} = \frac{\sqrt{\alpha + \beta}^2 + \alpha^2\vartheta^2}{\alpha\beta} - 1 \approx \frac{1}{\alpha} \approx \frac{\Omega^2}{2g_0^2},$$

$$\delta_{\text{max}} = \delta(\bar{n}_{\text{max}}) \approx \frac{4\Omega^3}{\gamma^2}.$$

We should take note of the fact that the maximum practically measurable side-band inequivalence, which occurs at the optimum intracavity photon number $\bar{n}_{\text{max}} = \Omega^2/2g_0^2$, is actually at the onset of bistability, and under practical conditions, heating due to optical losses in dielectric.

In Fig. 2 variation of side-band inequivalence versus intracavity photon number and in terms of different settings for input parameters $\{\alpha, \beta, \vartheta\}$ is illustrated.

Another very important result which can be drawn from the above discussions, is marking the boundaries of linear, weakly nonlinear, and strongly nonlinear interaction regimes in quantum optomechanics. This follows by normalizing $\delta$ with respect to the mechanical frequency $\Omega$ first, as $\delta = \delta/\Omega$.

- Fully Linear: This regime is easily given by $\bar{n} << \bar{n}_{\text{max}}$, where intracavity photon number is essentially too low to cause any appreciable side-band inequivalence. Here, the behavior of normalized side-band inequivalence is proportional to $\bar{n}$.
- Weakly Nonlinear: This regime is next given by $\bar{n} \sim \bar{n}_{\text{max}}$ around the optimum operation point, where the side-band inequivalence rises to attain a maximum. The behavior of normalized side-band inequivalence is nearly Lorentzian centered at $\bar{n} = \bar{n}_{\text{max}}$, with an intracavity photon number linewidth of $\Delta\bar{n} = \vartheta\bar{n}_{\text{max}}$.
- Strongly Nonlinear: This regime at larger intracavity photon numbers satisfying $\bar{n} >> \bar{n}_{\text{max}}$ will push the system into strongly nonlinear regime where the side-band inequivalence quickly start to fade away. Here, the behavior of normalized side-band inequivalence is inversely proportional to $\bar{n}$.

These three behaviors in above operation regimes can be respectively displayed as

$$\tilde{\delta}(\bar{n}) \approx \left( \frac{\bar{n}}{\bar{n}_{\text{max}}} \right) \delta_{\text{max}}, \quad \bar{n} << \bar{n}_{\text{max}},$$

$$\tilde{\delta}(\bar{n}) \approx \left[ 1 + \vartheta^{-2} \left( \frac{\bar{n}}{\bar{n}_{\text{max}}} - 1 \right)^2 \right]^{-1} \delta_{\text{max}}, \quad \bar{n} \sim \bar{n}_{\text{max}},$$

$$\tilde{\delta}(\bar{n}) \approx \frac{\bar{n}_{\text{max}}}{\bar{n}} \delta_{\text{max}}, \quad \bar{n} >> \bar{n}_{\text{max}}.$$

It is easy to verify that the side-band inequivalence does not violate the two fundamental symmetries of the nature. Here, both the time-reversal symmetry as well as the conservation of energy are preserved. The energy of scattered red- and blue- photons $\hbar\omega \neq \hbar\Omega$ is normally expected to be within the energy of one phonon $\hbar\Omega$ where $\omega$ is the angular frequency of incident electromagnetic radiation. Per every annihilated photon, exactly one phonon is either annihilated, giving rise to a blue-shifted photon, or one phonon is created, giving rise to a red-shifted photon.

However, not all phonons are having exactly the same energies. This is permissible by the non-vanishing mechanical linewidth $\Gamma > 0$ of the cavity. One should expect that once this quantity vanishes, the side-band inequivalence is gone, since it is by [19] proportional to $\Gamma^2$. Hence, basically it should be not contradictory to have a possible non-zero side-band inequivalence.

With regard to the time-reversal symmetry, one must take notice of the fact that all optical frequencies are physically positive, since we first must move back out of the rotating reference frame. For instance, the blue- and red-scattered photons have frequencies given by $\omega_b = \omega_c + \Omega - \frac{1}{2}\delta$ and $\omega_r = \omega_c - \Omega - \frac{1}{2}\delta$. Therefore, blue and red processes are not time-reversed processes of each other.

![FIG. 2. Variation of side-band inequivalence around the maximum point in terms of various settings of parameters.](image-url)
as they both stay on the positive frequency axis. Negative frequency images corresponding to both processes do however exist and exactly satisfy the time-reversal.

Remarkably, the Stokes and anti-Stokes peaks of Raman spectra also exhibit the same phenomenon of side-band inequivalence. This has so far skipped the attention since normally the nonlinear interaction rate causing the formation of Raman scattering is too small for majority of bulk materials and liquids. Again, a full linear theory of Raman scattering \[21\] disallows side-band inequivalence. However, with the recent advent of low-dimensional materials, optical nonlinear interactions leading to Raman scattering \[22\] and Kerr effect \[30\] are available at much stronger rates.

Table 4 provides a summary of measurable side-band inequivalence based on the reported Raman scattering of different materials. Calculations of normalized side-band inequivalence and errors are done according to the best possible resolution of measurement graphs. It has to be noted that for every given material, the side-band inequivalence is a strong function of scattering order, excitation wavelength, polarization, angle of incidence, as well as intensity. Therefore, it is not possible to reconstruct a fit to varying function for \(\delta\) as \[19\]. However, the existence of a previously unnoticed side-band inequivalence, which is always leaning toward the red side-band seems to be conclusive. Therefore, a rigorous quantum theory of side-band inequivalence for Raman scattering from continuum optomechanics is yet to be developed, such as the one recently developed for materials \[21\].

In the end and following the above, let us now plug-in \[4\] and \[7\] into the first of \[10\]. Some simplifications, while ignoring the inequilibrium quantum thermal effects on the population of side-bands, gives the equation

\[
\bar{n}_r - \bar{n}_h \approx \left( \frac{\Omega \bar{n}}{\Omega^2 + (\frac{1}{4} \Omega^2) \delta} \right) \delta \quad \text{(22)}
\]

Here, \(\bar{n}_r = |\langle \hat{a}_r \rangle|^2\) and \(\bar{n}_h = |\langle \hat{a}_h \rangle|^2\) respectively refer to the number of scattered photons unto red and blue side-bands. Then, from \[19\], and assuming that \(\bar{N} = (\bar{n}_r - \bar{n}_h)/\bar{n}\) denotes the normalized asymmetry of side-bands, we get

\[
\bar{N}(\bar{n}) = \frac{\psi(1 + \alpha \bar{n})}{\bar{n}^2 + (1 - \beta \bar{n})^2}. \quad \text{(23)}
\]

Accordingly the asymmetry is increases up to a positive maximum, before decreasing back to zero at sufficiently high powers.

Finally, it is easy to see that the same nonlinear symmetry breaking can lead to asymmetry in the particle pair production or parametric down conversion, which can be considered as the dual of optomechanical process \[45\] \[46\]. In order to observe this fact, consider an optomechanical system with a mechanical frequency roughly double the optical frequency \(\Omega \approx 2\omega\). If the mechanics is driven strong enough at the frequency \(\Omega\), then the effective interaction Hamiltonian will be simply

\[
\mathcal{H}_{\text{eff}} = \hbar \Omega (\hat{a}^\dagger \hat{a}^\dagger \hat{b} - \hat{a} \hat{b} \hat{b}^\dagger),
\]

where a phonon with energy \(\hbar \Omega\) dissociates into two photons with energies \(\hbar (\omega \pm \delta)\) with

| Material Side-band Inequivalence | Remarks |
|---------------------------------|---------|
| MoTe₂ | 7% ± 0.8% | 6-layer |
| MoS₂ | 0.3% ± 0.1% | 6-layer |
| 0.78% ± 0.15% | flake |
| CNT | 0.6% ± 0.21% | (10, 5) SC |
| 0.2% ± 0.1% | 1st-order; SC |
| 1.22% ± 0.13% | 2nd-order; SC |
| 1.5% ± 0.18% | single-wall |
| 2.11% ± 0.35% | single-wall |
| Gr | 0.8% ± 0.16% | 4-layer |
| 0.5% ± 0.17% | 3-layer |
| 1.63% ± 0.48% | (1, 2)-twisted |
| 2.2% ± 1.00% | C₃₁ (1, 3)-twisted |
| 3.6% ± 1.8% | C₃₂ (1, 3)-twisted |
| C | 1.15% ± 0.57% | Bulk |
| S | 1.03% ± 0.25% | 1st-order; grains |
| 0.72% ± 0.18% | 2nd-order; grains |
| Ethanol | 0.18% ± 0.04% | 1st-order; EX |
| H₂ | 9.83% ± 0.214% | MOM² |
| Gr | 0.267% ± 0.0074% | \(P_{\text{op}} = 7.0\text{mW}^b\) |
| 0.167% ± 0.0165% | \(P_{\text{op}} = 3.8\text{mW}^b\) |
| 0.067% ± 0.0063% | \(P_{\text{op}} = 1.8\text{mW}^b\) |
| B₄ | 0.34% ± 0.018% | 1090nm line |
| SRA | 0.1424% ± 0.001% | Yb-doped fiber |

\(a\) CNT: Carbon Nano-tube
\(b\) SC: Semiconducting
\(c\) Gr: Graphene
\(d\) 1.96eV excitation
\(e\) S: Sulphur
\(f\) EX: Excitation at 532nm
\(g\) MOM: Molecular Optical Modulation; Measurement at 1015kPa done on ortho–H₂, which has a 17.6THz rotational motion frequency.
\(h\) 3ps Ultrafast Excitation: Experimental data received from the authors through private communication. Data has to be shifted to adjust for \(\delta(0) = 0\), and exhibits a linear rate of 0.03867%/mW versus input power.
\(i\) B4: Single Biphenyl-4-thiol molecule in monolayer confined to optical picocavity; Experimental data available online \[42\]
\(j\) 4-point moving average applied. Gaussian 10-point filtering results in the much larger value of 1.06%.
\(k\) SRA: Stimulated Raman Amplifier; Experimental data available online \[43\]
\(\delta\) representing the corresponding symmetry breaking in pair frequencies caused by side-band inequivalence. Parametric down conversion for phonons has recently been observed and reported, too [17].

Also, based on the duality of effective interaction Hamiltonian in linear electro-optic modulation (within the validity of rotating wave approximation), with the optomechanical Hamiltonian [18–50], one could predict that the same nonlinear inequivalence to appear in relevant experiments, too. Similar arguments should be valid for enhanced Raman scattering of single molecules by localized plasmonic resonances as well [51].

We presented a complete analysis of side-band inequivalence in quantum optomechanics, and showed it undergoes a maximum and obtained closed-form expressions for optimum intracavity photon number as well as maximum attainable side-band inequivalence. We classified the operation into the linear, weakly nonlinear, and strongly nonlinear regimes, in which the behavior of system is markedly different. The results of this investigation can provide the accuracy constraints as well as necessary experimental set up to resolve the elusive side-band inequivalence. Analysis of high resolution measurements of Raman scattering for different materials confirms the existence of side-band inequivalence. One could speculate that precise measurement of the variation of side-band inequivalence in terms of various system parameters could provide further insight into unexplored nonlinear properties of different material.

This article is dedicated to the celebrated artist, Anastasia Huppmann.

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