Stokes – anti Stokes pair and their physical analogue to Cooper pairs.

Sanker Timsina

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ABSTRACT

In correlated Raman effect, photons may interact with each other with or without matching the vibrational energy difference of the medium. The out of resonance process has been linked to Cooper pair mechanism in superconductor. I derive the effective Hamiltonian for the Stokes anti-Stokes correlated photon pairs and briefly introduce this subject under intensive research.

INTRODUCTION

Raman spectroscopy is studied widely in physics and chemistry to understand the structural and chemical properties of matter [1]. Raman spectrum is the result of inelastic scattering of light by matter [2]. It is described in terms of shifts in a photon energy compared with the incident photon energy. Let us suppose that the initial energy of laser pulse incident on matter is $E_L = \hbar\omega_L$, ω_L is the frequency of laser pulse. The incident light may lose energy by exciting a phonon (quanta of atomic vibration) of frequency ω_p. As a result, the scattered photon has energy $E_s = \hbar\omega_L - \hbar\omega_p$. This photon is red shifted and called stokes photon. Similarly, incident photon may gain the energy from the lattice vibration and generate a blue shifted anti-Stokes photon with energy $E_{aS} = \hbar\omega_L + \hbar\omega_p$.

In addition to these two independent processes, a correlated Raman scattering or Stokes anti- Stokes (SaS) process may also occur [3]. In this case, a pair of incoming photons are scattered into a Stokes and an anti-Stokes photon. An incident photon interacts a Raman active material, creates a Stokes-photon and excites a quantum vibrational mode in the medium. Thus excited phonon is then annihilated by the second incident photon giving rise to anti-Stokes photon [1]. Theses processes are depicted in figure below.

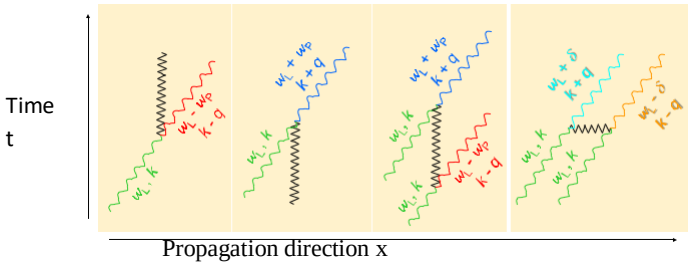

Figure1: Diagrammatic representation of Stokes, anti-Stokes, real and virtual Stokes anti-Stokes process [1].

The SaS process is quantified by the second order cross correlation defined as [4]

$$g^{(2)}_{S,aS}(\Delta t) = \frac{\langle n_S(t) n_{aS}(t')\rangle}{\langle n_S(t)\rangle\langle n_{aS}(t')\rangle}$$

Where, $\Delta t = t' - t$

In the classical limit, the cross-correlation is bound by the Cauchy-Schwartz inequality $[g^{(2)}_{S,aS}(0)]^2 \leq g^{(2)}_{S,S}(0) \times g^{(2)}_{aS,aS}(0)$ where, $g^{(2)}_{S,S}(0) and\ g^{(2)}_{aS,aS}(0)$ are autocorrelation of Stokes and anti-Stokes signal. It has been found that $g^{(2)}_{S,aS}(0)$ obtained in experiments strongly violate the classical bound [1]. The idea of SaS correlated photon pair production pioneered by Ado Jorio [5] are very interesting. They demonstrated that even water at room temperature can generate correlated photon pairs that violate the usual criteria for classical behavior by 5 orders of magnitude.

This establishes the fact that, the process is quantum mechanical leading to quantum entangled photon pairs. Non-classical correlations for photon pairs has been produced previously [6] and these entangled photons have been identified as one of the key resources in modern quantum information science. Various aspects of quantum information like quantum memory [7], quantum computation with linear optics [8], etc. are based on ability to entangle photons.
In this short report, I will briefly discuss the analogy between the formation of cooper pairs and correlated SaS pair by deriving the photon-photon interaction Hamiltonian. Description of experimental setup and experimental data is beyond the scope of this report. I will try to briefly summarize the result from the previous works.

THEORY

The correlated Stokes anti-Stokes (SaS) process that takes place can be further classified as real and virtual processes. In the out of resonance process, correlation between Stokes and anti-Stokes photons takes place by the exchange of virtual phonons which is called virtual SaS process [9]. Unlike real process, the virtual SaS is expected to happen near instantaneously [9]. Interestingly, this virtual process has been linked to the formation of Cooper pairs in superconductors through the phonon mediated interaction [1].
The Hamiltonian for the inelastic Raman scattering can be described by the [4],

$H=H_0+H_1$ (1),
Where, $H_0 = \sum_k \hbar\,\omega_k {b_k}^\dagger b_k + \sum_q \hbar\,\vartheta_q {c_q}^\dagger c_q$ (2)
is the Hamiltonian describing the free electromagnetic vibration composed of photons of energy $\hbar\omega_k$ and material vibration or phonons of energy $\hbar\vartheta_q$. q is the vibrational mode index. Here, b (c), $b^\dagger(c^\dagger)$ is the annihilation and creation operator of photons (phonons), respectively.

And the interaction Hamiltonian is
$H_1 = \sum_{k,q} M_q\,(c_q^\dagger + c_{-q})\,b_k b_{k-q}^\dagger$ (3)

Equation (3) describes the interaction of photons with the atomic vibrations. The term H_1 describes two processes. In one of them, the photon loses energy and creates a phonon during the scattering and in the other, a photon annihilates the photon and gains energy [4]. The quantity M_q is the strength of the photon- phonon coupling. Now we use the unitary transformation to remove the phonon operators.

Let us consider the transformation

$$H' = e^{-s}He^{s} = (1 - s + \cdots)H(1 + s + \ldots)$$
$$= H + [H,s] + 1/2[[H,s],s] + \ldots$$
$$= H_0 + ((H1 + [H0\,, s]) + \tfrac{1}{2}[(H1 + [H0\,, s]), s] + \tfrac{1}{2}\,[H1\,, s] + \ldots$$

Now, we choose s to satisfy [10],
$H_1 + [H0\,, s] = 0$ (4)
With this condition satisfied, we write, the Hamiltonian to be
$H' = H_0 + \frac{1}{2}[H_1, s]$ (5)
The idea is to write the interaction term differently, preserving the form of H_0, which is exactly solvable [10],
Let $s = \sum_{k,q} (a_- c_q^\dagger + a_+ c_{-q}) b_k b_{k-q}^\dagger$

From equation (4), we calculate $[H_0,S]$,

$$[H_0,\ c_q^\dagger b_k b_{k-q}^\dagger] = \left[H_0, c_q^\dagger\right] b_k b_{k-q}^\dagger + c_q^\dagger [H_0, b_k] b_{k-q}^\dagger + c_q^\dagger\, b_k \left[H_0, b_{k-q}^\dagger\right]$$
$$= \hbar(\vartheta_q - \omega_k + \omega_{k-q})\, c_q^\dagger\, b_k b_{k-q}^\dagger$$

Similarly,

$$[H_0,\ c_{-q}^\dagger b_k b_{k-q}^\dagger] = \hbar(-\vartheta_{-q} - \omega_k + \omega_{k-q})\, c_{-q}^\dagger\, b_k b_{k-q}^\dagger$$

From equation (3), we get

$$\sum_{k,q} M_q\,(c_q^\dagger b_k b_{k-q}^\dagger + c_{-q} b_k b_{k-q}^\dagger) = \sum_{k,q} -\hbar\,\{a_-(\vartheta_q - \omega_k + \omega_{k-q})\, c_q^\dagger\, b_k b_{k-q}^\dagger + a_+(-\vartheta_{-q} - \omega_k + \omega_{k-q})\, c_{-q}^\dagger\, b_k b_{k-q}^\dagger\}$$

Equating the left- and right-hand side of above equation, we get,

$$a_\pm = \frac{M_q}{\hbar(\omega_k - \omega_{k-q} \pm \vartheta_q)}$$

This gives our photon-photon interaction

$$H_{pp} = \frac{1}{2} \sum_{k,k',q} M_q (a_- - a_+) b_{k-q}^\dagger b_{k'+q}^\dagger\, b_k b_{k'}$$

Substituting the value of a_- *and* a_+, we further get,

$$H_{pp} = \sum_{k,k'q} \frac{M_q^2\, \vartheta_q}{\hbar[(\omega_k - \omega_{k-q})^2 - \vartheta_q^2]} b_{k-q}^\dagger b_{k'+q}^\dagger b_k b_{k'} \qquad (6)$$

This is analogous to the BCS interaction, except here the operators are for photons [1]. This equation describes the following process. Two incident laser pulse with wavevectors **k** and **k**' are scattered inelastically. One in scattered photon has wavevector less than the original pulse called the Stokes process and the other will have a wavevector greater than the original pulse called the anti-stokes process. Since we are interested in attractive interactions, we expect this interaction to be negative. This happens when $\omega_k - \omega_{k-q} \neq \vartheta_q$, that is out of resonance. This is same as in the formation of a cooper pair formation in superconductor where two electrons feel the attractive interaction mediated by phonons. The interaction potential is shown in figure 1 [4]. The potential diverges at resonance ($\omega_k - \omega_{k-q} = \vartheta_q$,) represented by the dashed line in the figure below and don't contribute to Stokes anti-Stokes pair formation. Inspired by the BCS model, the approximation potential, considered $-V_o$ in attractive photon-photon interaction [4].

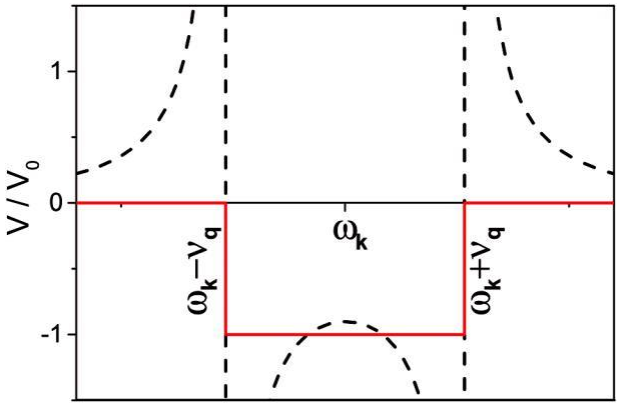

Figure 1. The dashed line shows the interaction potential. The red line is an approximation potential, as described in the text. The w_k gives the excitation laser frequency and v_q the vibrational frequency [4].

RESULTS

For the photonic counterparts of cooper pair, the coincident second cross correlation becomes,

$$g^{(2)}_{S,aS}(0) = \frac{\langle n_S(t) n_{aS}(t)\rangle}{\langle n_S(t)\rangle\langle n_{aS}(t)\rangle}$$

Experimental setup aims to measure this value. Figure 2 represents, the correlation function $g^{(2)}_{S,aS}(\Delta t = 0)$ (represented here with black circles joined by black dotted lines) as a function of Raman shift for a high purity 1.7 mm thick diamond slab [4]. The red line curve is the vibrational Stokes Raman spectrum.

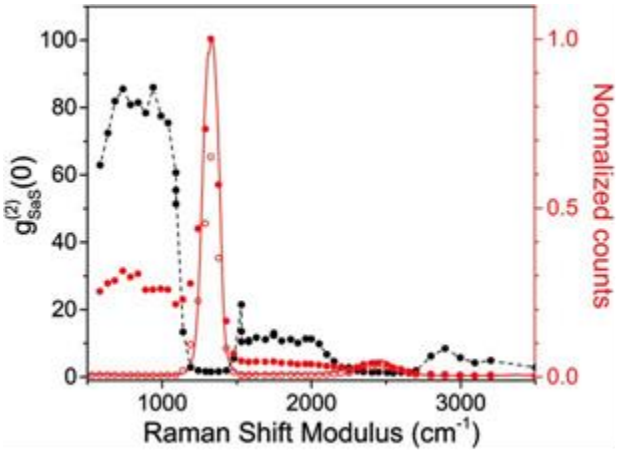

Figure 2: Observed Correlation function for diamond slab. Solid red circles represent the uncorrelated Raman process and the open red circle describe the correlated SaS process [4].

From the experimental result, it is inferred that out of resonance values of $g^{(2)}_{S,aS}(\Delta t = 0)$ are very high and decrease sharply as Raman shift approaches the real vibrational level (red line peak in figure 2). As can be seen from the figure, at resonance the number of uncorrelated stokes photons increases and obfuscates the correlated SaS process. Similar results were observed in water [1]. Various aspects of this experiments like momentum and energy dependence of correlation has also been studied recently [9].

CONCLUSIONS AND FUTURE WORK

The experimental results discussed above establish the fact that phonon mediated photon pairs dominate out of Raman resonance process and can be effectively utilized for the entangled photon generation. However, the efficiency of production of entangled SaS pair for bulk production is quite low [11]. The question I would explore in my research is whether in the presence of surface plasmons the production of SaS can be increased. If possible, the plasmonic- SaS pairs will be an important resource for photon-based quantum computers. Additionally, I briefly discussed how this virtual process has been linked to cooper pairs in superconductors and derived the effective Hamiltonian for the process. Understanding this analogy in greater detail and to what extend it can be extended is another of my research task.

. REFERENCES

1. A. Saraiva, F. S. de Aguiar Júnior, R. M. Souza, A. P. Pena, C. H. Monken, M. F. Santos, B. Koiller, A. Jorio, Phys. Rev. Lett. **119**, 193603(2017).
2. R. Acosta Diaz, C. H. Monken, A. Jorio, and Marcelo F. Santos, Phys. Rev B. **102**, 134304 (2020).
3. D. N. Klyshko, Sov. J. Quant. Electron. 7, **755** (1977).

4. F.S. de Aguiar Júnior, C. H. Monken, M. F. Santos, R. de Melo e Souza, A. Saraiva, B. Koiller, and A. Jorio Phys. Status Solidi B, 1900218 (2019).
5. M. Kasperczyk, F. S. de Aguiar Júnior, C. Rabelo, A. Saraiva, M.F. Santos, L. Novotny, and Ado Jorio Phys. Rev. Lett. **117**, 243603 (2016).
6. A. Kuzmich, W.P Bowen, A.D. Boozer, A. Boca, C.W. Chou, L.M. Duan and H.J. Kimble, Nature, 423, **731-734** (2003).
7. DS. Ding, W. Zhang, ZY. Zhou, S. Shi, B-Sen Shi & G-Can Guo, Nature Photonics 9, **332-338** (2015).
8. E. Knill, R. Laflamme & G.J. Milburn, Nature, 409, **46-52** (2001).
9. F.S. de Aguiar Júnior, A. Saraiva, M.F. Santos, B.Koiller, R. de Melo e Souza, A.P.Pena, R. A. Silva, C. H. Monken, and A. Jorio, Phys. Rev. B 99, 100503 (2019)
10. A. Saraiva, F. S. de Aguiar Júnior, R. M. Souza, A. P. Pena, C. H. Monken, M. F. Santos, B. Koiller, A. Jorio, Phys. Rev. Lett. **119**, 193603(2017), supplemental material.
11. A. Loot and V. Hizhnyakov, J. opt. **20,** 055502 (2018).