Entangled Photon Pairs from Raman Scattering

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Raman scattering is a bona fide experimental setup to probe all sorts of systems on their chemical composition, inner structure, symmetries, layer orientation and so on. It is an inelastic light scattering process where incoming photons of a given frequency lose or gain energy due to the creation or annihilation of vibrations in the material in order to generate outgoing photons of different frequencies. Correlations between the two possible processes, of adding or subtracting a vibration, with consequential correlation between the outgoing light has been theoretically predicted in the 1970s and lies at the core of well established techniques such as Coherent Anti-Stokes Raman Spectroscopy (CARS). In most applications of Raman spectroscopy, all the quantized fields involved are intense and the quantum state of the vibrations can be taken semi-classically so that the inelastic scattering can be described as a standard four-wave mixing interaction.

In recent years, however, the quantized nature of the vibrations gained new focus due to their potential applications in quantum information theory. Early experiments tried to establish them as a potential quantum memory for information encoded in the light fields. In fact, typical pump-and-probe setups were used to demonstrate that a phonon created in diamond due to a spontaneous Raman scattering could save information of the pumping light and transmit it to an outgoing anti-Stokes photon generated by the probe. A similar setup was then used to entangle a phonon in two separated diamonds and then probe this entanglement.

In parallel, the effect of quantized vibrations in the properties of the scattered light gained momentum when it was shown to be a necessary element for the correction of the extraction of temperature through intense Stokes-anti-Stokes (SaS) spectroscopy [1]. More important, a theoretical and experimental result showed that the vacuum of the vibrational field played an essential role in the generation of time-correlated SaS photon pairs in a boson version of the creation of Cooper pairs in superconducting materials [2-4].

Entanglement in the frequency degrees of freedom was given but difficult to investigate, In polarization, however, it was unclear considering that the polarization of the formed pair mostly followed that of the pump [5]. In this work, we experimentally show that the SaS pairs are indeed entangled in polarization and we offer a theoretical explanation for this entanglement as a quantum combination (superposition) of two possible and independent origins for each pair: electronic or vibrational four wave mixing. We also show that the degree of entanglement depends on the frequencies of the generated pair and, in particular, whether the vibrational origin involves the exchange of real or virtual phonons in the used diamond sample. [6]

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