Conservation law of angular momentum in Raman spectroscopy using circularly polarized light

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In Raman scattering of two-dimensional material, it is known that the helicity of circularly polarized light (left- or right-handed circularly polarized light) becomes opposite for incident and scattered light for some Raman active modes such as the in-plane optical IMC mode of transition metal dichalcogenides and the G band of graphene [Nano Lett. **15**, 2526 (2015) and Phys. Rev. B **95**, 165417 (2017)]. Since circularly polarized light has an angular momentum,$\pm\hbar$, the angular momentum of a photon is transferred to the material when the helicity of the scattered light is changed from that of incident light after the Raman scattering.

In the first-order Raman scattering, since a phonon is emitted in the Raman process, it may be natural to think that the angular momentum of the incident photon is transferred to the emitted phonon. L. Zhang *et al.* discuss the angular momentum of phonon and they show that the degenerated Γ-point phonon can have an angular momentum $\pm\hbar$ by making the superposition of two degenerate eigenvectors of the phonon [Phys. Rev. Lett. 115, 115502 (2015)]. However, if the angular momentum of a photon changes from $+\hbar$ to $-\hbar$, the change of angular momentum of a photon is $2\hbar$ which cannot transfer to a phonon with $\pm\hbar$.

In this presentation, we discuss the conservation law of angular momentum for first-order Raman scattering by considering the N-fold and $N\nu$-fold rotational symmetries of crystal and phonon, respectively [1]. H. J. Simon and many reseachers discussed the conservation law of angular momentum of crystal for 3-fold rotational symmetry. [Phys. Rev. 171, 1104 (1968)]. Here we give a general conservation law for $N = 1,2,3,4,\text{and } 6$ of the crystal for Raman and Rayleigh scattering. Further, we calculate the Raman and Rayleigh spectra for circularly polarized light by using the first principles calculations [2]. The calculated results explain all existing experiments and theories and predict some phonon modes in general N-fold rotational symmetry. We hope that the predicted, helicity exchanged Raman spectra that is not observed yet will be observed by experiment.

References:

1. Y. Tatsumi, T. Kaneko, R. Saito, unpublished,
2. Y. Tatsumi, R. Saito, Phys. Rev. B97, 115407 (2018).