Optical Nonlinear and Amplification Effects in CdS Nano-Rods

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Abstract

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Anti-Stokes enhancement over the Stokes intensity is observed in nano-rods of CdS with diameter of less than 2 nm. In these extreme nano-scales constriction of the impinging laser fields induce nonlinear effects, amplification of the Stokes and anti-Stokes radiation fields and enhanced CARS and possible lasing without inversion in a double lambda scheme and SASER.

Many aspects of nanostructures are presently under intense study. These nanostructures provide unique quantum arena for nonlinear and other optical processes. Our nano-crystals have dimensions of about 2 nm in diameter and about 10 nm in length. These scales confine the electrons and localize excitations. The quantum confinement results among others, in a blue shift of the optical absorption and the photoluminescence with decreasing size. The ability of these nano-structures to constrict weak CW electromagnetic field modes of an impinging laser renders strong intensity radiation fields inside them, that enable nonlinear effects to play important role in determining the optical properties.
In the present work we explore these effects in CdS nano- rods. The Raman spectra of CdS nanorods exhibit the typical Longitudinal Optical modes at 300 cm^{-1} (1LO) and the first overtone at 600 cm^{-1} (2LO). The size of the nano-particles affects their absorption and photoluminescence spectra and the ratio of the overtone to the fundamental LO frequency related to the exciton phonon coupling, see figure 1.

Figure 2 displays the basic process of the Raman scattering and the Stokes and anti-Stokes transitions of the phonon levels

Osgood et. al. have predicted [1] enhancement and even amplification of the Stokes field as a result of the laser constriction in nano-rods. Such a field along with the incident coherent laser field can generate a four wave mixing process leading to coherent enhancement of the anti-Stokes component namely, enhanced CARS seeFig. 3.

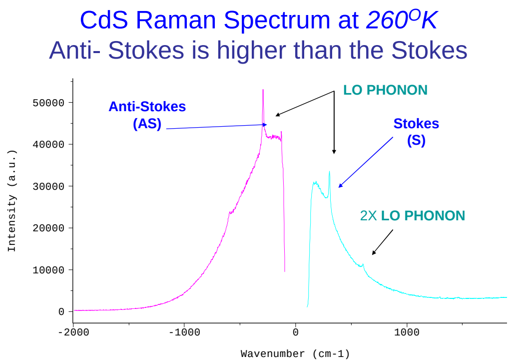

Figure 1. Raman spectra of CdS nano-rods of 1.7 nm and of diameter and 10 nm length, exhibiting unusual intensity ratio of anti-Stokes to Stokes, at low temperature. Also shown are the relevant second order phonon scattering. The background is the relevant photoluminescence emission. The exciting laser wavelength is 488 nm of the argon
lase

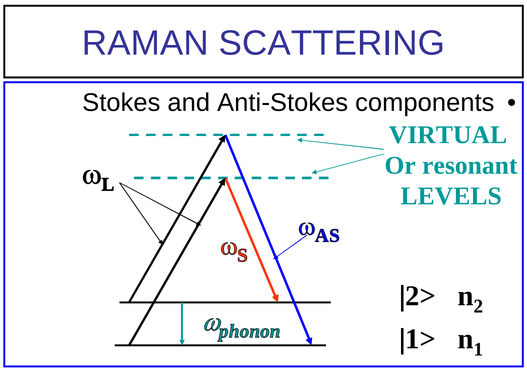

Figure 2. Raman scattering scheme showing the Stkes and anti-Stokes components and the phonon levels.

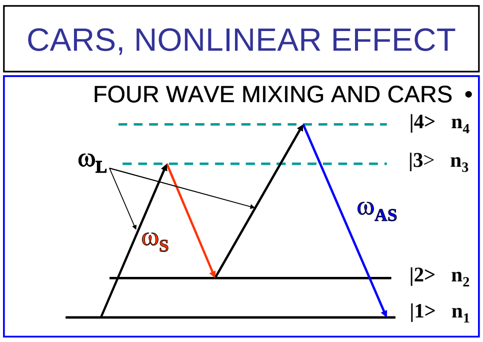

Fig. 3 CARS nonlinear model. In the present experiment two laser photons and one Stokes photon generates the fourth anti-Stokes photon in a four wave mixing process.

Indeed, we observe an enhanced anti Stokes component of the LO phonon at 300 cm^{-1} in CdS nano-rods. In the following, we examine the Raman scattering properties of CdS nano rods. While there are well known particle size related optical properties such as phonon confinement and tunable photoluminescence, not much is discussed about their Stokes and anti-Stokes scattering characteristics. Our temperature dependent measurements of the Raman spectra of CdS nano-rods exhibit peculiar behavior of the anti-Stokes to Stokes intensity ratio. In the studied temperature range this ratio indicates higher intensity of the anti Stokes than that of the Stokes, figure 4.

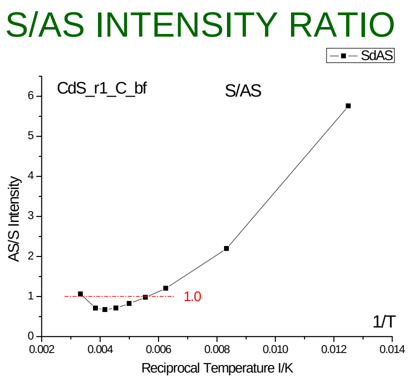

Figure 4. Results of measured Stokes to anti-Stokes intensity ratio in the temperature range of 100 °K to room temperature. Notice the unusual "inverted" ratio in most of the temperature range of the experiments.

The plausible consequence is that there should be an inverted phonon population and no thermal equilibrium under continuous laser exposure. A possible mechanism for such a scenario is the generation of LO phonons by a decay of excitons to a non-thermal population level [2]. This makes possible phonon super-radiance and sound amplification, namely SASER [3]. Moreover, enhancement of the coherent anti-Stokes component, namely CARS, in these elongated nano-rods is made possible by the high intensity that results from the constriction of the impinging laser field inside the nano-rods. However, there is competition among the two Raman sidebands for which the anti-Stokes component has a larger cross-section.
Finally, laser without inversion (LWI) [4] in two Lambda scheme in the presence of two em fields of the incident laser and the Stokes laser, Fig.5, is also a viable explanation yet to be investigated in further research of this interesting characteristics of the nano-particle behavior.

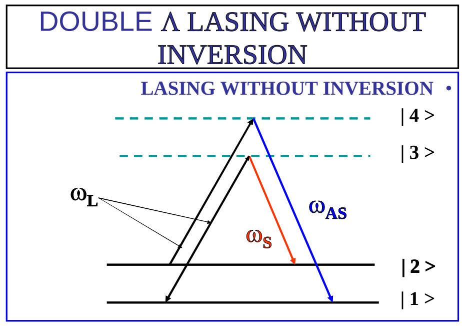

Figure 5. The coherent two laser photons and the Stokes photon generates a lasing without inversion in a double Lambda scheme, via a quantum interference process.

Results and summary

We have shown non-standard behavior of the intensity of the anti-Stokes scattering vs. that of the Stokes component in the temperature range of 100-300 °K. It is a unique to nano-size particles of CdS For he excitation laser used in the experiment, resonant Raman scattering on the incoming and the outgoing channels, the Stokes and the anti-Stokes is achieved.
The photoluminescence peak (near band edge) of nano-crystal CdS shifts with temperature roughly between 2.525e V to 2.465 eV [5]. The higher cross-section of the anti-Stokes provides a factor of 1.125. However, both of these causes are insufficient to explain the measured ratio.
It is plausible to incorporate nonlinear effects and Stokes amplification due to the laser light constriction in the nano-rods, followed by CARS four wave mixing to generate the anti-Stokes emission particularly, due to the enhancement factor of 1.125 due to the frequency difference. LWI process is also a good candidate process for the anti-Stokes enhancement seen in the experiment. This can provide basis for nano-lasers in a wide range of the spectrum particularly in wide band gap material. These can also provide nano-markers in biosensors studies.

References

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