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ABSTRACTS

NON PERTURBATIVE THEORY OF ELECTRONIC RESONANT COHERENT RAMAN  
SCATTERING (CARS, CSRS)

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Preresonance enhanced coherent Raman scattering occurs when one of the input fields is close to resonance with an allowed electronic transition. Under these conditions, large deviations from what expected in usual CARS and CSRS occur (1,2), namely line-broadening and large non-linearities in the coherent generated intensity.

The expression for the third-order susceptibility in the fully perturbative approach does not account for the above observed effects. Actually, fully-perturbative approaches in quasi-resonant conditions loose their validity, when the Rabi frequencies of the radiation-matter interaction are of the same order of magnitude as the molecular dampings (3).

The creation of coherent excitations in the material system are treated here in a semi-perturbative way. Resonant strong fields are dealt in a "exact" way, while non-resonant and weak fields are considered as perturbations (3).

The emission of Stokes and anti-Stokes radiation rules the evolution of the coherent excitations, derived as usual by summation over all the intermediate states.

Saturation effects, line broadening and dispersive line-shapes are accounted, leading profiles similar to those observed in several experimental works (2).

REFERENCES

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