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Invited paper to be presented to the International
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Nonlinear Optical Processes in Solids, Aussois,
France, March 18-20, 1981

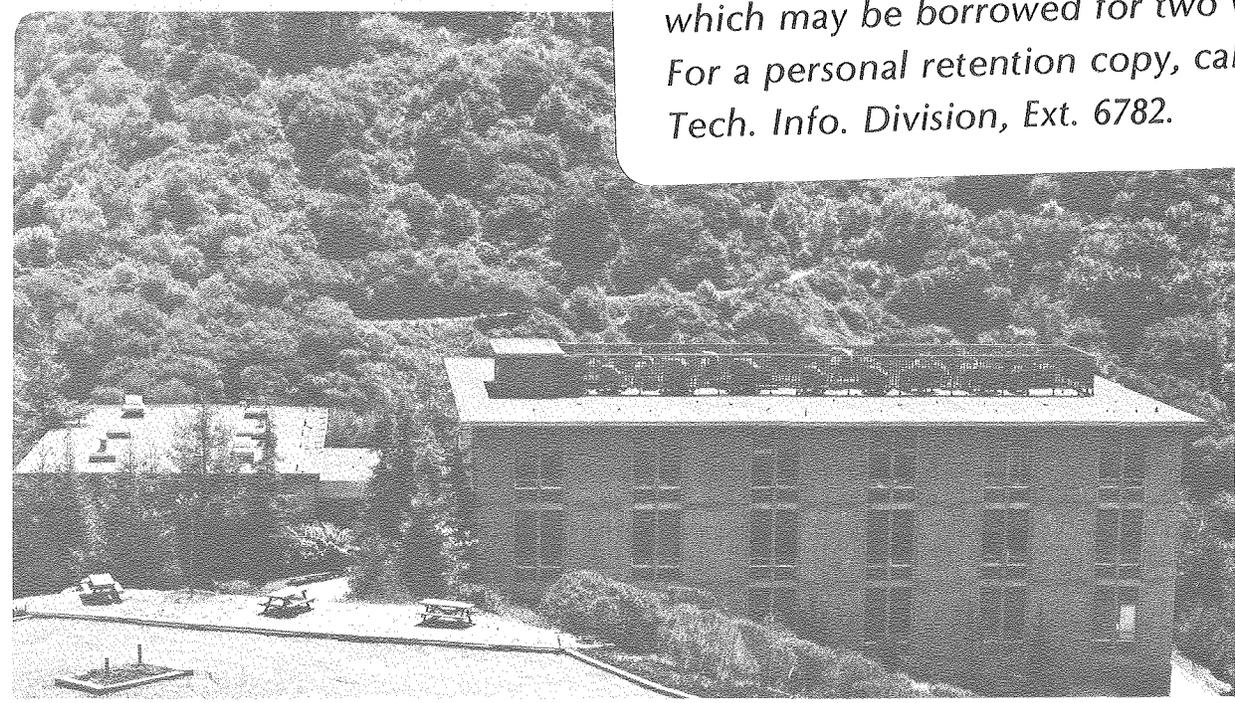
THEORETICAL ASPECTS OF FOUR-WAVE MIXING SPECTROSCOPY
WITH MULTIPLE RESONANCES

J.L. Oudar and Y.R. Shen

January 1981

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THEORETICAL ASPECTS OF FOUR-WAVE MIXING
SPECTROSCOPY WITH MULTIPLE RESONANCES

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THEORETICAL ASPECTS OF FOUR-WAVE MIXING SPECTROSCOPY WITH
MULTIPLE RESONANCES

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ABSTRACT

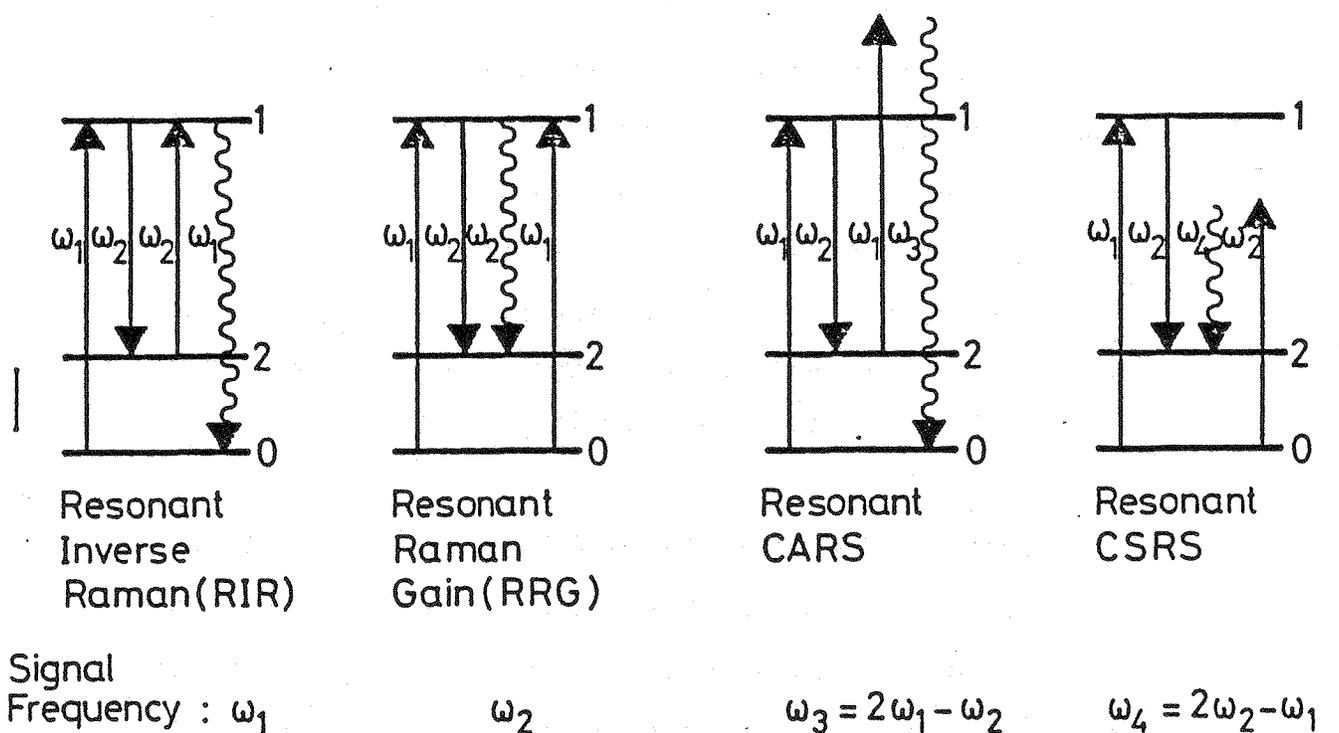
The use of multiresonant four-wave mixing processes in solid-state spectroscopy is analysed in detail. Specific information on the excited states of solids can be obtained from such techniques, including the reduction of inhomogeneous broadening, study of transitions between excited states, determination of ultrafast relaxation times through measurements in the frequency domain.

SUMMARY

Four-wave mixing can be used in several different ways for probing multiresonant optical nonlinearities in solids, and the efficiency of the interaction depends on a wide variety of different processes. A typical example is provided by the number of physical effects which may contribute to the generation of phase-conjugate waves in the degenerate case : optical Kerr effect, saturated absorption, gain saturation in laser media, non-linear refraction, two-photon resonances.

When the nonlinear optical process is multiresonant it is usually much more efficient, and in addition, it can provide more selective spectroscopic information, in the same way as resonant Raman scattering, compared to ordinary Raman scattering ⁽¹⁾. For instance it can be used for an accurate determination of matrix elements and damping parameters of transitions between excited states (transition dipole moments or excited state Raman cross-sections). However an increased complexity is the price to pay for such attractive features, and a careful analysis of the theoretical problem is needed in order to assess what new spectroscopic information can be obtained from such experiments. The density matrix formalism ⁽²⁾ with the help of time-ordered diagrams, ⁽³⁾ provides a general framework for discussing very different processes from a common viewpoint. It is especially well suited to comparing the optical nonlinearities associated with step-wise processes, involving successive real transitions, as opposed to direct processes.

Several multiresonant configurations can be used to probe a three-level system, depending on the frequencies of the incoming and outgoing waves with respect to the transition frequencies of the medium. For a three-level system of the resonant Raman type, four basic configurations are considered, with particular reference to the spectroscopy of impurities in a solid at low temperature. With two incident frequencies ω_1 and ω_2 , these configurations can be schematized by the following energy diagrams :



From one case to another, the multiresonant third-order susceptibility can vary by many orders of magnitude for

a given three-level system. Several factors contribute to these differences :

- two processes (RIR and RRG) are triply resonant, whereas the two others are doubly resonant ;
- for RRG only, one contribution at exact resonance is proportional to the longitudinal relaxation time T_1 of level 1, which can be much longer than the dephasing time T_2 of that level ;
- finally these processes exhibit a different behaviour with regards to the inhomogeneous broadening. In the same way as in fluorescence line-narrowing experiments, one can expect spectra with a reduced linewidth, because one of the incident frequencies (for instance ω_1) can selectively excite a restricted number of impurity sites within the broad absorption band. A lineshape analysis shows that this is not always the case, the linewidth reduction is predicted to occur in special cases only.

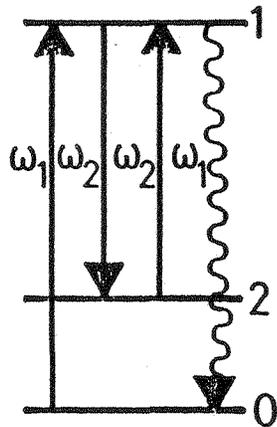
Among these possible configurations, the RIR looks fairly interesting because it is triply resonant, it involves only two different frequencies, and still it is intrinsically a coherent process. As a result, it is free of complications due to energy relaxations within level 1 or towards other levels lying close to it. On another hand, resonant CARS and CSRS involve the generation of a third frequency, and can be of great value for probing a four-level system such as realized by the vibrational structures of the electronic ground and excited states.

Finally, multiresonant four-wave mixing with three input frequencies can probe the ultrafast relaxations within a broadband continuum of levels ^(1,4). For that purpose, two of the incident frequencies are close together and tunable, thus forming a rapidly moving grating of excited states, which can be probed by the third incident beam.

In conclusion, four-wave mixing spectroscopy with multiple resonances is a powerful tool for studying the excited states of solids, their vibrational structure, dynamics, or their coupling with other excited states.

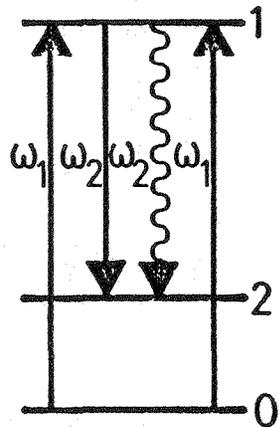
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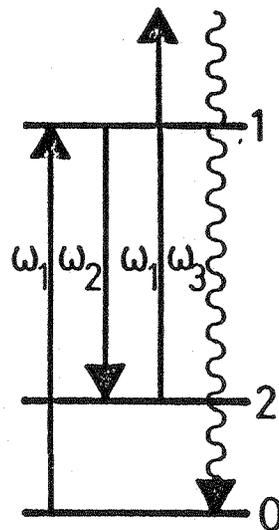
Resonant
Inverse
Raman (RIR)

Signal
Frequency : ω_1



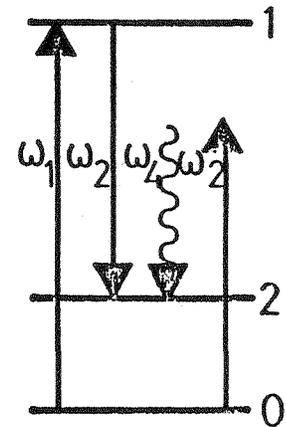
Resonant
Raman
Gain (RRG)

ω_2



Resonant
CARS

$\omega_3 = 2\omega_1 - \omega_2$



Resonant
CSRS

$\omega_4 = 2\omega_2 - \omega_1$