LBL-12154

CONF-810312--2

Lawrence Berkeley Laboratory

UNIVERSITY OF CALIFORNIA

Materials & Molecular Research Division

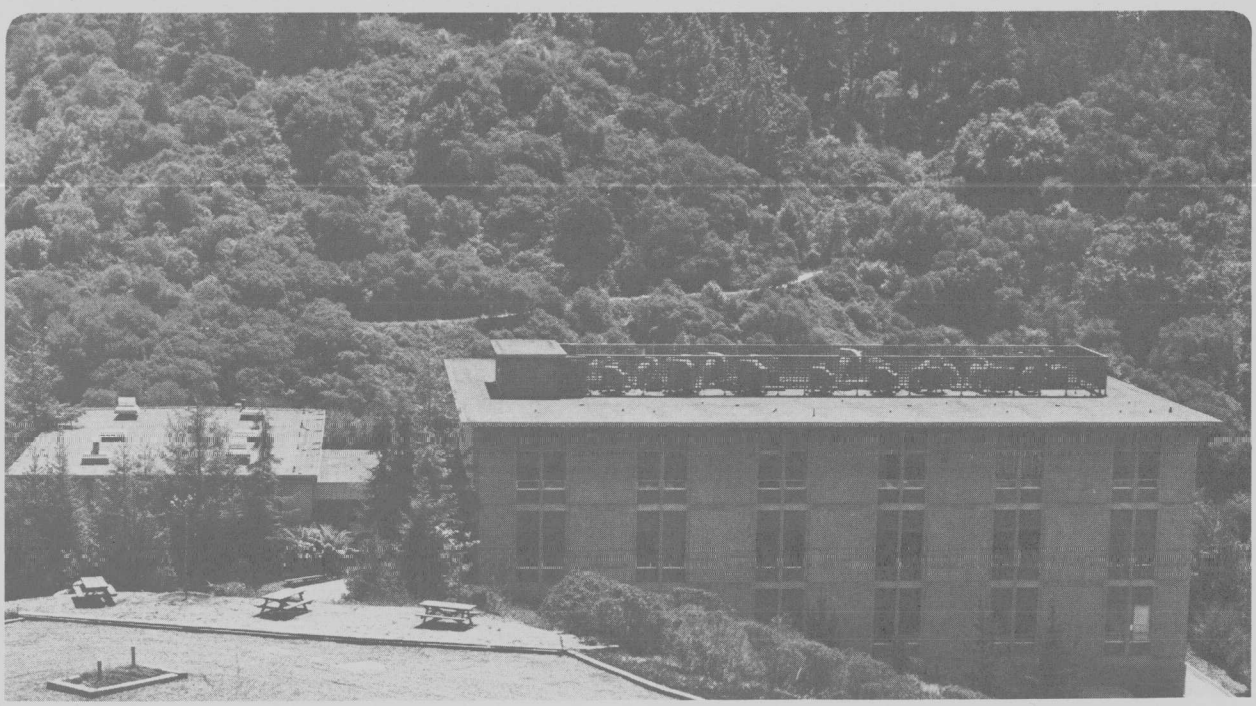
MASTER

Invited paper to be presented to the International Conference on Excited States and Multiresonant 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



DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

Prepared for the U.S. Department of Energy under Contract W-7405-ENG-48

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

LEGAL NOTICE

This book was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

THEORETICAL ASPECTS OF FOUR-WAVE MIXING SPECTROSCOPY WITH MULTIPLE RESONANCES

(invited)

J.L. OUDAR

Centre National d'Etudes des Télécommunications

196, rue de Paris - 92220 Bagneux (FRANCE)

and

Y.R. SHEN

Department of Physics, University of California, LBL,

Berkeley 94720 -(U.S.A.)-

DISCLAIMER

This book was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

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.

This work was partially supported by the Division of Materials Sciences, Office of Basic Energy Sciences of the U.S. Department of Energy under Contract W-7405-ENG-48 and the Centre National d'Etudes des Telecommunications, Bagneux, France.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

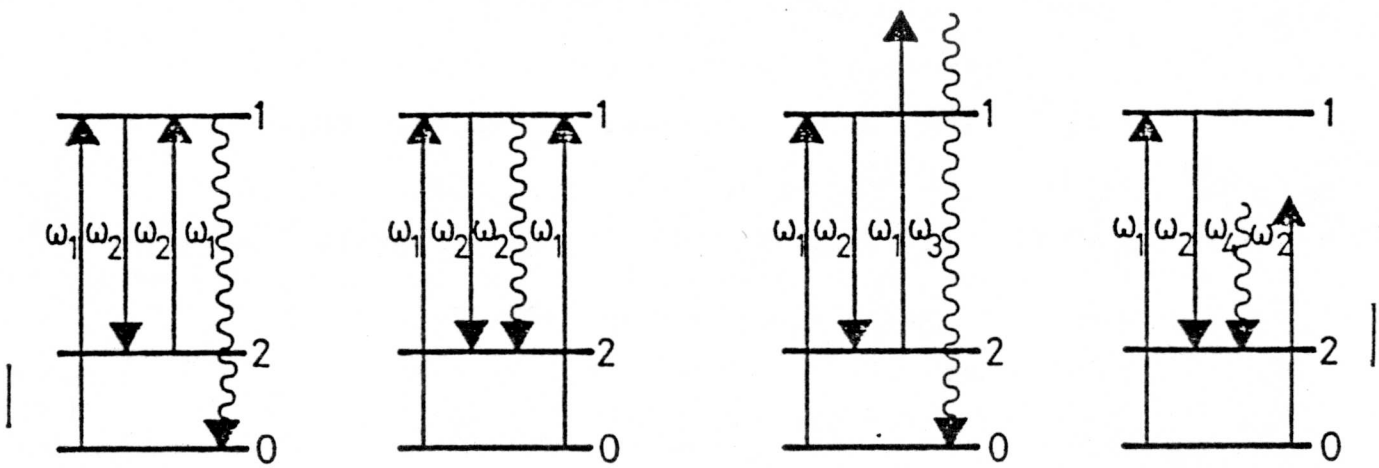
MGW

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 :



Resonant Inverse Raman(RIR) | Resonant Raman Gain(RRG) | Resonant CARS | Resonant CSRS

Signal Frequency : ω_1 ω_2 $\omega_3 = 2\omega_1 - \omega_2$ $\omega_4 = 2\omega_2 - \omega_1$

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 proportionnal 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.

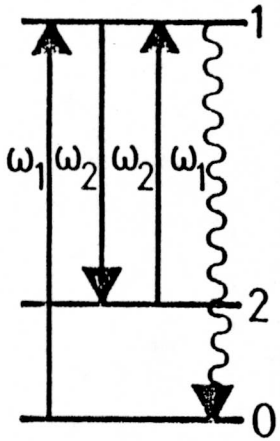
REFERENCES

(1) J.L. OUDAR and Y.R. Shen, Phys. Rev. A22, 1141 (1980)

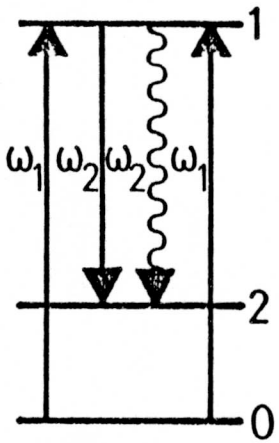
(2) N. BLOEMBERGEN, Nonlinear Optics (Benjamin, New-York, 1965) ; N. BLOEMBERGEN, H. LOTEM and R.T. LYNCH, Indian J. Pure and Appl. Phys. 16, 151 (1978)

(3) S.Y. YEE, T.K. GUSTAFSON, S.A.J. DRUET and J.P.E. TARAN, Optics Commun. 23, 1 (1977) ; S.Y. YEE and T.K. GUSTAFSON, Phys. Rev. A18, 1597 (1978)

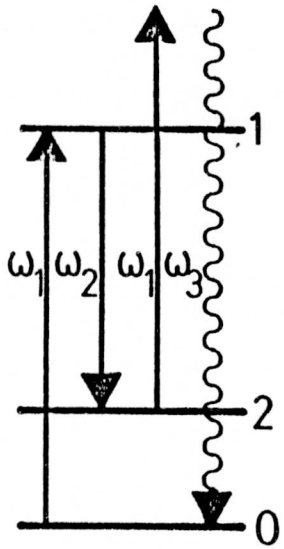
(4) A.E. SIEGMAN, Appl. Phys. Lett. 30, 21 (1977).



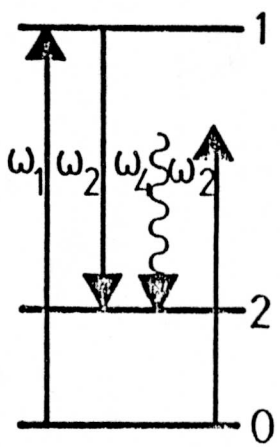
Resonant Inverse Raman (RIR)



Resonant Raman Gain (RRG)



Resonant CARS



Resonant CSRS

Signal Frequency : ω_1 ω_2 $\omega_3 = 2\omega_1 - \omega_2$ $\omega_4 = 2\omega_2 - \omega_1$

This report was done with support from the Department of Energy. Any conclusions or opinions expressed in this report represent solely those of the author(s) and not necessarily those of The Regents of the University of California, the Lawrence Berkeley Laboratory or the Department of Energy.

Reference to a company or product name does not imply approval or recommendation of the product by the University of California or the U.S. Department of Energy to the exclusion of others that may be suitable.

TECHNICAL INFORMATION DEPARTMENT
LAWRENCE BERKELEY LABORATORY
UNIVERSITY OF CALIFORNIA
BERKELEY, CALIFORNIA 94720