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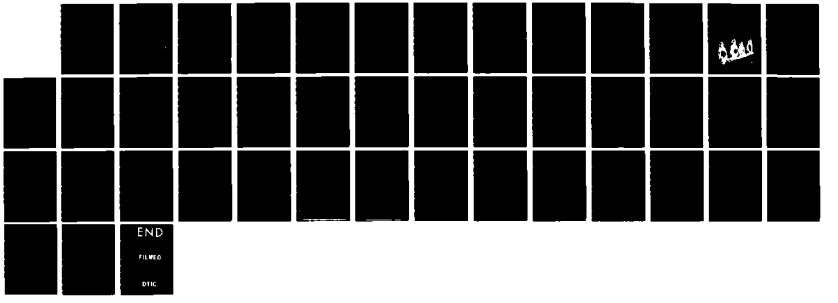

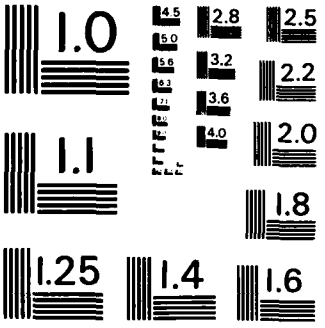

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Final Technical Report

Office of Naval Research Contract N00014-81-K-0513

APPLICATION OF FOUR-WAVE MIXING SPECTROSCOPY IN THE EXCITONIC REGION OF SEMICONDUCTORS

July 1, 1981 - December 31, 1983

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Contents

Foreword

During the funding period, two different areas of research were conducted in our laboratory. They are:

I. Development of a photoacoustic detection scheme using nanosecond laser pulses, and its application to semiconductor research, (in particular, the study of two-photon absorption in ZnS)

II. Study of nonlinear optical effects in the excitonic region of CdS. In this work, bound-exciton laser action was investigated in detail, employing a new cavity configuration.

List of Publications:

The list of publications which resulted from this research are as follows:

1) "Photoacoustic Detection of Polarization-Dependent Nonlinear Optical Effects in Crystals," Y. Bae, J.J. Song, and Y.B. Kim, Proceedings of the Second International Topical Meeting on Photoacoustic Spectroscopy (Opt. Soc. Am. 1981).

2) "Photoacoustic Detection of Nanosecond-Pulse-Induced Optical Absorption in Solids;" Y. Bae, J.J. Song, and Y.B. Kim, Appl. Opt. 21, 35 (1982).

3) "Photoacoustic Study of Two-Photon Absorption in Hexagonal ZnS;" Y. Bae, J.J. Song, and Y.B. Kim, J. Appl. Phys. 53, 615 (1982).

4) "Visits to Laser Research Laboratories in Japan;" J.J. Song, Scientific Bulletin (ONR-Far East) 7, 45 (1982).

5) "Laser Oscillations in the Bound-Excitonic Region of CdS," W.C. Wang and J.J. Song, Fourth International Conference on Dynamical

Processes in the Excited States of Solids, July 1983; J. Opt. Soc. Am. 73, 1387 (1983).

6) "Bound-Exciton Laser Action in CdS;" W.C. Wang and J.J. Song, Annual Meeting of Optical Society of America, October 1983; J. Opt. Soc. Am. 73, 1928 (1983).

7) "Laser Oscillations in the Bound-Excitonic Region of CdS," J.J. Song and W.C. Wang, J. Appl. Phys. 55, 660 (1984).

8) "Resonant Pumping of CdS Laser; study of formation and decay of I_1 bound excitons," J.J. Song and W.C. Wang (in preparation).

Next, the outcome of our research efforts will be presented, and its significance, discussed.

I. Photoacoustic Effect

1) Detection of nanosecond-laser-induced optical absorption.

Photoacoustic detection is a sensitive mehtod for observing weak optical absorption in a variety of materials. In most cases, however, chopped cw laser radiation or relatively long (microsecond) laser pulses were employed as the input laser sources. This was because the optical absorption depends on the energy (i.e. number of photons) of the input lasers, and consequently, longer pulses resulted in higher photoacoustic signals.

For nonlinear optical studies, what is important is the laser peak power, not the energy of each pulse. Unfortunately, there was no report of using short (nanosecond) laser pulses for photoacoustic detection. Furthermore, there was great confusion in deciding proper

(in terms of speed) transducers for short pulses.

We took a systematic approach to determining suitable (right speed) detectors and found that the detectors exhibit resonant behavior which depends on the length and material of transducers. (See Table 1, Bae et al, Appl. Opt. 1982.) This seemingly obvious and simple aspect of photoacoustic detection was overlooked or misunderstood by other research groups. We were thus the first group who successfully observed nanosecond-laser-induced absorption in solids. The sensitivity of our detection scheme was 10^{-6} cm/MW in case of two photon absorption. The application of the photoacoustic detection to the study of two-photon absorption in ZnS is reported next.

2) Photoacoustic study of two-photon absorption in Hexagonal ZnS.

In the study of two-photon absorption (TPA), one of the fundamental questions is whether the observed TPA is a two-step one-photon process or a one-step two-photon process. One way of finding this out is to change the polarization direction of the laser beam with respect to the crystalline axes of the ZnS sample. (See Bae et al, J. Appl. Phys. 1982.) By comparing the data with the group-theoretical analysis, it was concluded that the TPA we observed belonged to the latter case (i.e. one-step process).

By observing the dispersion of TPA, it was also found that TPA in ZnS occurs via allowed-forbidden type processes. This was further supported by the measurement of linear-circular dichroism, and by comparing our data with the theoretical prediction of Beregulin et. al (see Bae et al, J. Appl. Phys. 1982).

II. Optical Properties of CdS in the Excitonic Region

The changes in optical properties of CdS which occur under pulsed laser excitation are very complex, controversial and not well understood in spite of the fact that these optical properties of CdS have been widely investigated since the advent of pulsed laser sources (specifically the N_2 lasers). In the course of executing wave-mixing experiments, we also encountered complications. It was found that the experimental data were extremely sensitive to the quality of the input laser beams. For definitive interpretation of the data, the spectral and spatial purity of two incident laser beams was required. Since our home-made pulsed dye lasers were unable to deliver the desired high quality beams, we were forced to modify our goals, and investigated laser-action in CdS, which required only one laser. Also, the requirement of the excitation beam quality was not so stringent in this case. We made, however, significant contributions to the physics-aspect of CdS laser action as well as to the engineering-aspect of semiconductor lasers in general. To summarize:

i) We have utilized an external cavity configuration which was never applied to semiconductor lasers before.

ii) The lasing could be achieved in the themperature range of 5-250°K without heat sinking. This implies that room temperature laser operation may be feasible with proper heat sinking.

iii) At liquid helium temperature, the lasing was attributed to

acoustic-phonon-coupled I_1 bound exciton transitions.

iv) The resonant behavior of lasing processes was observed, from which the formation and decay routes of bound excitons can be revealed.

v) The lasing frequencies higher than I_1 bound exciton energy was observed.

In light of the fact that optically pumped semiconductor lasers are of great interest to the scientific community, it is worthwhile to elaborate on the significance of our cavity design. Also discussed next will be the physical significance of our findings on the bound exciton laser action in CdS.

1) Why external cavity? Why longitudinal pumping?

Typical semiconductor laser cavities are formed by two parallel surfaces of the semiconducting sample itself or by two parallel mirrors separated by short distance (~mm). In this case, there is no space to insert a frequency selecting element. Recently, a long (~20 cm) cavity was introduced by an MIT group. The lasing photons, however, were collected by a micorscopic objective lens which was located right next to the sample inside the cavity. This lens intorduced additional optical loss of the laser cavity resulting in the higher laser threshold and additonal difficulty in the cavity alignment. Our design of the hemispherical cavity condiguration is

unique in the sense that the cavity length was long enough (~5cm) for the insertion of a frequency selecting element, and that the microscopic objective lens was eliminated by a concave mirror. This design, of course, can be applied to any semiconductor lasers including heterostructure compound semiconducting superlattice lasers.

The longitudinal-pumping method we employed has advantages over traditional transverse-pumping. In the case of the latter, at least three sample surfaces need to be good (polished or cleaved), whereas in the case of the former, only two good surfaces are required. Furthermore, when combined with an external cavity-configuration, as-grown samples can be used, thereby greatly simplifying sample preparation procedures. These features are particularly attractive for selecting samples that are likely to lase prior to the detailed investigation of lasing characteristics. Particularly large applications will be found in heterstructure semiconducting superlattice lasers. There have been great efforts by researchers to lase the superlattices along the axis normal to the layers. (This is an important research topic at the Electrotechnical Laboratories in Japan, and PI had a lengthy discussion with Dr. Takafumi Yao on this subject during her visit to ETL, December 1983). Our laser configuration seems particularly suited to this purpose.

In summary, the pumping and lasing goemetry we employed has wide applications in tunable solid-state (semiconductors and color-centers, for example) lasers. It remains to be seen, however, whether superlattices such as GaAs/AlGaAs or GaSb/AlSb can be lased perpendicular to the layers.

2) Bound-Exciton Laser Action in CdS.

There are several optical transitons which can lead to laser oscillations in CdS at LHe temperature. The lasing at the P-bnad at 4905Å or at A-LO at 4927Å has previously been observed by many researchers. The P-band which appears only under a high excitation condition was first identified in 1970, as arising from exciton-exciton interactions. The A-LO band originating from the LO-phonon-assisted A exciton transitions, usually exhibits higher lasing threshold than the P-band.

The bound excitons have also been speculated to be associated with possible laser oscillations in the CdS. Nearly all as-grown CdS platelet samples contain impurity centers, to which excitons can be bound at low temperatures. Two prominent bound excitons at 4876Å and 4889Å are called I_2 and I_1, respectively. We have clearly observed I_1-related laser oscillations, but not the laser action involving I_2 contrary to previous predictions. The large gain (~300 cm^{-1}) observed in the I_1 region also indicates that there is a need to reexamine the previously reported electron-hole liquid picture in CdS. The claim was based on the gain-spectra which overlaps with the large gain we observed near I_1 under relatively low (~10 kw/cm^2) pumping density, which is too low even for electron-hole plasma formation. The relatively broad (~10 cm^{-1}) gain profile is believed to be associated with acoustic phonon coupling. What is noteworthy is that the lasing was observed at higher frequencies than I_1. That is, the anti-Stokes side wing of I_1 exhibited laser oscillations. To our knowledge, this is the first report on such lasing in any semiconductors.

We also observed that the threshold of laser action depended on the pumping frequencies (resonant-phenomenon). When the pumping energy coincided with the energy of the I_1-neutral-acceptor, bound to B-exciton instead of A-exciton, the laser output showed resonant enhancement. This observation indicates that one route of creating I_1 bound excitons is to form a B-bound exciton at the site of the participating neutral-acceptors, and then the hole is scattered from B- to A- band to form A-bound I_1 excitons. A wealth of information on bound exciton dynamics can be obtained when picosecond lasers are utilized for above-mentioned resonant pumping experiments.

Finally, it should be mentioned that CdS crystals are currently drawing a lot of attention from the optics (and physics) community. Optical Bistability was observed from CdS at 2K near I_2 bound exciton transitions. The exact mechanism for the observed OB, however, has not yet been identified. The OB due to the renormalization of the band gap of CdS was also recently reported. Since OB originates from the intensity-dependent change of optical properties, to which laser action is closely related, we believe that OB can also be observed near I_1 without much difficulty. We also believe that our findings will be of great help to understand complicated intensity-dependent optical changes that take place near excitons.

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SCIENTIFIC BULLETIN

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VISITS TO LASER RESEARCH LABORATORIES IN JAPAN

Jin J. Song

The high level of Japanese quantum electronics technology and the laser-related research is well-known. They keep abreast of the most up-to-date U.S. research activities in this area by sending their research personnel to U.S. laboratories, and also through the biannual topical meetings such as the U.S.-Japan light scattering symposium or the U.S.-Japan laser workshops.

During May 1981, I visited some of the leading Japanese laser laboratories which were geographically scattered from Kyoto to Sendai. Overall, the common interests of Japanese researchers were very much in line with the U.S. (or rather international) research trend. This again reflects their efficiency in information gathering. Not surprisingly, some of them acquired their early experience with lasers at U.S. laboratories. Their current interests include short duration subpicosecond pulse generation, high power UV excimer laser technology, short wavelength coherent light pulse generation, and their applications.

The research laboratories I visitied are in chronological order as follows:

- University of Electrocommunications (Chofu)
- Institute of Solid State Physics (Tokyo)
- Tsukuba University and Electrotechnical Laboratory (Tsukuba)
- Institute for Molecular Science (Okazaki)
- Kyoto University (Kyoto)
- Research Institute of Electrical Communication (Tohoku)

A brief summary of my observations and impressions at each laboratory is presented in the order shown above.

UNIVERSITY OF ELECTROCOMMUNICATIONS

Several laser-related research laboratories in Japan were visited during 11-20 May 1981. My first day on this trip was spent at the Institute for Laser Science (ILS), University of Electrocommunications in Chofu near Tokyo. The institute was established for the purpose of developing new lasers and laser materials. The director, and my host, Professor H. Takuma, is an internationally recognized laser scientist. Under his strong leadership, the several young researchers I met seemed very enthusiastic about their projects. One of the major programs was to construct electron-beam-pumped high power eximer lasers for future applications in fusion research. This project was to be more vigorously pursued with the completion of the new building now under construction. Some of the ongoing projects included the study of laser interaction with atomic vapors (sodium and rubidium) and nonlinear wave mixing laser spectroscopy in molecular beams (in particular, coherent anti-Stokes Raman spectroscopy). A project similar to the latter has been carried out by Professor R. Buyer's group at Stanford University. The research at ILS also included the investigation of lasing action in rare-earth ion-doped glass material. As an efficient pumping source for Tm^{3+}: fluorophosphate glass, the XeF laser was proposed.

The institute was very well-equipped and the experimental apparatus well-maintained. Among the lasers I saw at ILS were:

- two high power argon ion lasers (coherent radiation),

- one krypton ion laser (coherent radiation),
- two ring dye lasers (coherent radiation and spectra-physics),
- two Nd:YAG lasers and dye lasers (molectron), and
- small cw dye lasers and a homemade Nd:YAG laser.

In addition, there seemed to be no shortage in manpower as far as electronic technical support was concerned (may be due to its affiliation with the university). This was reflected in their computerized experimental apparatus, and a very sophisticated homemade high speed signal integrator which is far more efficient than a commercial boxcar integrator.

The general atmosphere of ILS was very much like that of any university laboratory in the U.S. It seemed that research topics could be chosen with a great deal of freedom and flexibility, and that their research interests were not confined to studying new laser systems. In this sense, the institute has great potential to attract imaginative scientists and to carry out frontier research in the very near future. The institute, however, was at a formative (developing) stage, and at the time of my visit they appeared to have more ideas and equipment than their research personnel could handle.

THE INSTITUTE FOR SOLID STATE PHYSICS

The Institute for Solid State Physics (ISSP) at the University of Tokyo was visited on May 12. It is widely known as one of the leading research institutes in the world with 25 years of history. Due to the limited time I had, my visit was restricted mainly to the laser physics and spectroscopy group. The group consists of three subgroups, each headed by Professors S. Shionoya, T. Yajima, and H. Kuroda, respectively. I had the opportunity to meet with all three distinguished scientists.

Professor Shionoya has been engaged in laser spectroscopy in solid state materials for more than a decade. In particular, he made significant contributions to the understanding of excited states in semiconductors, and those of ions in crystals and glasses by employing luminescence spectroscopy techniques. He was one of the first to extensively use short laser pulses to probe dynamics of optically excited states in semiconductors and insulators by observing the evolution of the luminescence spectra with nanosecond and picosecond time resolution. His work on high density electron hole plasma in direct gap semiconductors such as CdS, CdSe, and GaAs as well as on excitonic molecules in CuCl and CuBr has been the subject of an invited talk at numerous international laser spectroscopy conferences. One of the laser systems extensively used in his recent work is a broadly tunable Nd:YAG - parametric oscillator giving out 30 psec pulses. (Shorter pulses are desirable in some cases, but this system has the advantage that it can produce pulses from UV to infrared.)

Recently, Professor Shionoya's group has employed nonlinear laser spectroscopic techniques. Unlike spontaneous luminescence techniques, coherence effects of the excited states can be extracted from nonlinear spectroscopy. The wave mixing method they adopted has nothing new in itself, but very interesting material properties are being revealed. Their work is usually very thorough, meticulous, and reliable. This group is rather a newcomer in the nonlinear laser spectroscopy community. With their knowledge in materials and their background in basic spectroscopy, however, they will be as formidable and productive with their nonlinear optical work as they have been with their luminescence work.

Professor H. Kuroda's group carries out research on basic laser physics, as well as on

the interaction of high power laser pulses with matter. Their samples under investigation ranged from plasma to semiconductors under strong laser pulse irradiation. One of their goals is to produce high power, short (in duration as well as in wavelength) laser pulses. A high power (100 GW) KrF laser, ≈1 TW Nd:glass lasers and a hybrid system (Nd:YAG laser plus 5-stage excimer amplifiers) were being developed for fusion research. Picosecond VUV-pulse-generation and x-ray laser action were also under study, together with stimulated Raman/Brillouin scattering in plasma.

Professor T. Yajima is well-known for his pioneering work in quantum electronics and nonlinear optics. In 1975, he proposed to apply resonant Rayleigh-type wave mixing techniques to the study of ultrafast (usually less that psec) relaxation phenomena in condensed media. Nonlinear wave mixing in transparent region (laser wavelength is below the absorption gap of the material) was fairly well understood largely through Professor N. Bloembergen's work at Harvard University. (He received the 1981 Nobel prize in physics for his contribution in wave mixing phenomena, among other things.) When the laser community was moving toward resonant wave mixing in condensed media, Professor Yajimas's proposition and theoretical prediction was timely and prophetic. Ultrafast transient phenomena was usually investigated in time domain by use of (sub) picosecond lasers. With Rayleigh-type wave mixing, picosecond phenomena can be observed in the frequency domain by long (for example, nanosecond laser pulses which are easily accessible) pulses. Professor Yajimas's group experimentally demonstrated this in dye solutions and determined longitudinal and transverse relaxation rates of the excited states of the dye molecules. Our group at the University of Southern California adopted a somewhat different wave mixing technique to measure subpicosecond relaxation rates in liquids and solids. Our technique is sometimes termed as polarization spectroscopy of Kerr-type (as opposed to Rayleigh-type) mixing spectroscopy. In polarization spectroscopy, the polarization change of the probe beam induced by the strong pump beam is examined, whereas in Rayleigh-type mixing, signal frequency is different from the probe or pump frequency. Even though there are some differences in experimental requirements and observed spectra, fundamental principles can be considered almost the same for both cases.

Professor Yajima recently employed subpicosecond laser pulses for a comprehensive study of wave mixing processes. (Comprehensive in the sense that both transient and stationary nonlinear effects are studied.) One of the new schemes is a degenerate wave mixing method which is designed to yield some information on relaxation rates regardless of the relative magnitude of relaxation times and the pulse width.

One of Professor Yajima's projects, during the past couple of years, was to produce and analyze subpicosecond pulses from a dye laser pumped by an argon laser. In 1980, his group reported the generation of pulses of ~0.13 psec duration. (Their synchronously-pumped-passively-mode-locked laser scheme is similar to the one developed by J.C. Diels at North Texas State University.) In 1981, C.V. Shank and his coworkers at Bell Laboratories at Holmdel, New Jersey, reported successful generation of 90 femtosecond (0.09 psec) dye laser pulses by employing a novel colliding-pulse-mode-locking method. Yajima's group further introduced a method of amplifying subpicosecond pulses in which the dye laser medium acts as an amplifier pumped by an Nd:YAG laser. Now that they are equipped with short, strong (amplified) pulses for nonlinear subpicosecond laser spectroscopy, they can thoroughly investigate the relationship between the spectral and temporal responses of the nonlinear optical processes. From the theoretical as well as experimental point of view, Professor Yajima's group is ahead of any researchers in the world in the area of applying wave mixing methods to the study of ultrafast relaxation processes.

TSUKUBA UNIVERSITY

Tsukuba Science City was visited on May 13 and 14. The city looked very much like a large university campus in the U.S. It was designed to house government research laboratories. The whole research complex is called the AIST Tsukuba Research Center (Agency of Industrial Science and Technology). My host here was Professor H. Shirakawa of the University of Tsukuba. Professor Shirakawa's research was centered on the preparation and characterization of doped organic polymer systems such as iodine-doped polycetylene. Due to their high electric conductivity and large optical nonlinearity, these sort of polymers have been receiving much attention from both academia and industry. In the past, Professor Shirakawa and his coworkers employed optical spectroscopy to elucidate structural details of polycetylene.

The other laboratories I visited at Tsukuba University were those engaged in optics-oriented research. The laboratories were, in my opinion, not very well-equipped and their programs were oriented for student projects. Their Raman spectroscopy apparatus, however, was very up-to-date from the laser source to the computer-controlled detection scheme.

THE ELECTROTECHNICAL LABORATORY

The Electrotechnical Laboratory (ETL) which I visited is only one of the many national research laboratories in the Tsukuba Science City. It is the largest national research organization specializing in electricity and electronics in Japan. Although the research activities of ETL encompasses a very wide range of fundamental and applied research, my visit was mainly with the laser research group in the Radio and Optoelectronics Division. Among the many subjects of their research is the study of very fundamental processes in atomic vapor. Recently, they investigated theoretically as well as experimentally, the second harmonic generation (SHG) of strong ND:YAG laser pulses in sodium vapor without an externally applied electric field. Unlike usual SHG in vapors, the spontaneously generated electric field from the laser interaction caused SHG in their case.

Significant effort was being made to develop subpicosecond lasers and high power UV and VUV coherent light sources. In this sense, their program somewhat overlaps with that of the laser group at ISSP or of Professor Takuma's group at the Electrotechnical University. I was told during my visit that ETL was ahead of others in Japan in KrF eximer laser technology. They were also quite successful in operating a subpicosecond dye laser system. The design was similar to that of Yajima's group. Picosecond pulses were also generated from flashlamp-pumped dye lasers and semiconductor lasers.

It was interesting to find out that the major research personnel involved in the successful picosecond or KrF laser projects had recently been visiting scientists at the leading research laboratories abroad (namely the National Research Council of Canada and Professor D.J. Bradley's group at the Imperial College, London). I also noticed that they were very young scientists.

THE INSTITUTE FOR MOLECULAR SCIENCE

The Institute for Molecular Science (IMS) in Okazaki is truly an outstanding organization in terms of facility, equipment, and research personnel. The institute is involved with nearly all branches of molecular science. My host was Professor K. Yoshihara, leader of the division of electronic structure. Professor Yoshihara's team was studying electronic structure of excited states, photochemical reactions of organic

compounds, transient phenomena in biology, etc. One of their probing methods was to use picosecond lasers for time resolved spectroscopy. In their study, fluorescence decays from the excited states were monitored as a function of time. The technique in itself was similar to the one used in Professor Shionoya's laboratory for their semiconductor research.

The division of molecular structure employs a wide variety of spectroscopic techniques to identify transient species in chemical reactions and to understand details of reaction mechanisms. Their light sources cover a wide range in wavelength, from microwave to UV. Their projects included the development of sophisticated high resolution light scattering spectrometer employing a Fabrey-Perot interferometer and a multichannel analyzer. Various methods of two-beam spectroscopy were included in their probing techniques. To name a few, they were IR-visible double resonance spectroscopy and two-beam thermal lensing spectroscopy, which are somewhat related to nonlinear laser spectroscopy. A CARS setup (a nonlinear wave mixing method) was being developed using nanosecond pulses.

The Division of Molecular Assembly studies the electronic properties of the organic solids by means of UV and VUV photoelectron spectroscopy. Photoconduction of organic semiconductors, photoionization processes in diatomic molecules, light scattering from graphite intercalates, and electrical property of ferrocytochrome C were also among their research subjects. The more recent push of this group, however, seemed to lie in molecular beam spectroscopy. Many newly constructed molecular beam apparatus were noticed.

What I have observed at IMS amounts to only a very small portion of their research activities. The common denominator of the various research divisions was the study of photon-molecule interactions. The large laboratory space, abundant in equipment, including all kinds of U.S.-made lasers (among them were Spectra-physics Ring dye lasers and a color center laser manufactured by Burleigh Incorporated, New York) and the technical support from the Instrument Center or Computer Center, could easily be the subject of envy of any visitor.

From the viewpoint of laser spectroscopic techniques, they seemed more interested in modifying and using already established techniques for their purposes rather than developing or adopting any new (not so well established) methods. One area which I felt they were yet to explore was coherent transient phenomena (nonlinear optical effect). This area is being extensively studied by chemists at California Institute of Technology, the University of California at Berkeley, the University of Pennsylvania, and Stanford University.

KYOTO UNIVERSITY

Professor M. Matsuoka's laboratory at Kyoto University was briefly visited on May 18. Professor Matsuoka is one of the younger Japanese laser physicists who has received international recognition. He has worked with Professor Bloembergen at Harvard University, as well as with Professor S.R. Hartmann at Columbia University. Thus, he has had the opportunity to be associated with world authorities in nonlinear optics and the related phenomena.

Professor Matsuoka was studying coherent transient effects in vapors as well as in condensed phase samples. His group first demonstrated picosecond-backward photon echo effects in sodium vapor. The backward echo scheme is attractive for the application in picosecond spectroscopy since it alleviates the use of optical shutters with slow (nsec) responses. This scheme could be related to phase-conjugation effects or Professor Yajima's

recent time domain spectroscopy. Incidentally, Matsuoka carried out some of his work at IMS in Okazaki, and is a graduate of Yajima's laboratory.

A new picosecond laser system was under construction, based on the Kuizenga type Nd:YAG laser. Briefly, the Kuizenga model is a pseudo-cw-mode-locked, Q-switched laser. To my knowledge, the Kuizenga version of the Nd:YAG laser is not so simple to operate. In contrast to the modern, spacious IMS facilities, Professor Matsuoka's research was being carried out mostly in one large room in an old building. Nonetheless, they are an extremely productive group.

THE RESEARCH INSTITUTE OF ELECTRICAL COMMUNICATION

Professor H. Inaba's laboratory was located at the Research Institute of Electrical Communication, Tohoku University in Sendai. Professor Inaba had early access to laser research during his stay at Stanford University in the early 1960s. He has been a prominent figure in the international quantum electronics circle for a long period of time. His research reflects such diversity and flexibility that it did not seem possible to grasp all his activities in just one visit.

Both fundamental and applied research was being carried out. Some of their research topics were:

- study on laser radars for remote sensing of air pollutants,
- generation of coherent optical waves up to VUV,
- laser Raman and Mie scattering (application is extended to medical field),
- fabrication of semiconductor lasers-integrated nonlinear optics (picosecond, tunable, high repetition-rate), and
- development of new excimer lasers, etc.

Their recent efforts appeared to be more centered on integrated nonlinear optical devices.

As in Kyoto University, the laboratory building was old and there seemed to be a shortage of laboratory space. The research group consisted of twenty or so students and Ph.D.'s, and appeared to be well-organized and efficiently functioning. That could be one of the factors contributing to their continued success in the ever changing quantum electronics field.

Laser oscillations in the bound-excitonic region of CdS[a)]

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By employing a simple hemispherical resonator configuration, laser oscillations were observed from vapor-grown undoped CdS platelets pumped by 4-nsec dye laser pulses. The lasing shifted toward longer wavelengths from 4885 to 5180 Å by varying the temperature of the samples from 5 to 250 °K. The laser emission observed at low temperatures is attributed to the acoustic-phonon-coupled I_1 bound-exciton transitions. The resonant behavior of this laser process is also reported.

PACS numbers: 42.55.Px, 78.45. + h, 42.60.By, 42.60.Da

I. INTRODUCTION

Recently, an optically-pumped CdS laser in an external cavity was reported by Wünstel *et al.* and Roxlo *et al.*[1,2] In both cases, the resonator cavity was formed by two plane-parallel mirrors. As the pumping source, a pulsed Ruby laser (6943 Å) was used in the case of the former and a cw (or mode-locked) argon laser (4765 Å) was used in the case of the latter. The highest temperature at which the lasing could be accomplished in the above-mentioned work was 140 °K.[2]

By employing a hemispherical cavity configuration, we have observed laser oscillations from undoped CdS platelets in the temperature range between 5 and 250 °K. The samples were pumped by 4-nsec dye laser pulses at ≈4830 Å. At liquid-helium (LHe) temperature, the lasing was observed in the region of 4880 ~ 4900 Å with the polarization vector of the emission beam perpendicular to the *C* axis of the sample. The dominant mechanism for this laser process is attributed to the acoustic-phonon coupled I_1 bound-exciton transitions. With the increase of the sample temperature, the lasing wavelength increased so that at 250 °K the lasing wavelength was 5180 Å in the samples we studied.

There are several optical transitions which can lead to laser oscillations in CdS at LHe temperature. The lasing at the *P* band at 4905 Å or at *A*-LO at 4927 Å has previously been observed by many researchers.[3] The *P* band which appears only under a high excitation condition was first identified by Madge *et al.* in 1970, as arising from exciton-exciton interactions.[4] The *A*-LO band originating from the LO-phonon-assisted *A* exciton transitions, usually exhibits higher lasing threshold than the *P* band.[3,5]

The bound excitons have also been speculated to be associated with possible laser oscillations in CdS.[6,7] Nearly all as-grown CdS platelet samples contain impurity centers, to which excitons can be bound at low temperatures. Two prominent bound excitons at ≈ 4867 Å and ≈ 4889 Å are called I_2 (neutral donor-bound) and I_1 (neutral acceptor-bound), respectively.[8]

When highly excited by laser pulses or electron-beam (*e*-beam) bombardment the low energy sides of the I_2 and the I_1 show the stimulated emission-like behavior.[9,10] The lower energy wing of the I_2 in the emission spectra of CdS is sometimes referred as the *M* band and that of I_1, as the P_M band.[9] Although somewhat controversial, the *M* band is generally accepted as due to biexciton decay whereas the P_M band is regarded as an acoustic-phonon wing of the I_1[10].

Huber *et al.* observed indications of lasing from an *e*-beam-pumped compound semiconductor $CdS_{0.77}Se_{0.23}$ at 5382 Å and attributed it to I_2 transitions.[11] Liebing *et al.* observed laser-like sharp features in the P_M band of a CdS excited by the second harmonic of the Ruby laser radiation at the power density higher than 2.2 MW/cm^2.[11] In this case, however, other features such as the I_2, the *M* and the *P* band were still present in the spectra together with the laser-like spikes in the P_M region. In other words, the lasing effect was not clearly demonstrated. Based on the above-mentioned observation, Liebing *et al.* proposed a laser system involving I_2 or I_1 bound excitons and acoustic phonons.[12] Here we report on the clear observation of such laser processes in the I_1 region. We could not, however, observe the laser action involving I_2 bound excitons.

II. EXPERIMENTAL DETAILS

The CdS samples used in this work were undoped vapor-grown platelets with thickness ranging from 10 to 50 μ. The optical resonator cavity was formed by a flat back reflector (mirror A) and a concave output coupling mirror (mirror B) as shown in Fig. 1. The CdS platelets were mounted on the mirror A (sapphire substrates with ≈ 100% reflectivity dielectric coating) using silicon oil of high viscosity. The sapphire mirror was then mounted on the copper cold finger of a cryostat (Air Products Helitran LT-3-110 and a temperature controller model APD-E) using silicon grease. The cryogenic dewar was attached to an *xyz* translation stage. It also had two degrees of freedom of rotation, one rotation with respect to the *x* axis and the other, to the *z* axis. In this way, different spots of a sample could be examined more conveniently. The mirror B with 93% dielectric coating on the concave side, was 1 cm in diam and had 5 cm focal length. This output coupler was also mounted on an *xyz* translation stage. The distance between the two mirrors was 5 cm. It is to be noted that the only element between the sample and the mirror B was a quartz dewar window.

The sample was excited by a loosely focussed beam

[a)] A portion of this work was presented at the 4th International Conference on the Dynamical Processes in the Excited States of Solids, Stanford, California, July 1983.

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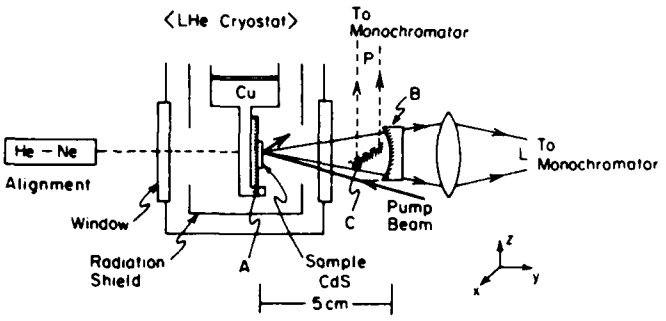

FIG. 1. Experimental setup. The optical cavity is formed by the mirrors A and B. The mirror C is translated out of the cavity when observing laser oscillations. P and L indicate the optical path through which the photoluminescence and the laser emission signals were collected.

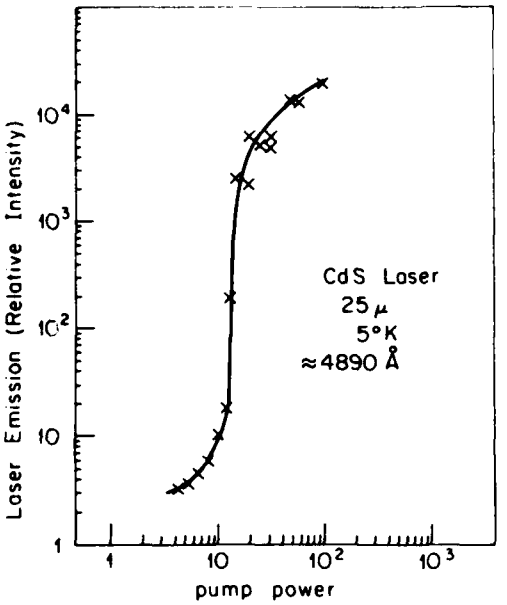

FIG. 3. Peak power of the laser emission is plotted against the pump power level. The peak power of 10 W in front of the sample corresponds to approximately 14 in the horizontal scale.

from a pulsed dye laser (10PPS) pumped by the third harmonic of 1.06 μ Nd:YAG laser radiation. In this experiment, a prism-type dye laser was used and the lasing medum was Coumarin 480 mixed with methanol. The dye laser beam was spatially filtered to ensure a relatively homogeneous beam profile on the sample. The peak power of the laser beam incident on the sample was less than 100 W. If necessary, the dye laser power was reduced by use of calibrated Kodak gelatin ND filters.

The CdS platelets used in this work were visually selected so as to have relatively flat and smooth surfaces on both sides. The surface quality was examined and the sample thickness measured under a microscope. Microscopic details of the sample such as impurity sites or concentrations are not known.

The initial alignment of the hemispherical laser cavity was carried out using a He-Ne laser. The 6328 Å laser beam was sent through the sample normal to its surface and to the center of the output mirror. Then the pulsed laser beam was overlapped with the He-Ne beam on the surface of the sample. First, the mirror B was coarsely aligned so that the He-Ne reflection from the mirror B coincided with the sample spot excited by the pulsed laser beam. The fine adjustment was then made by observing the laser emission signal detected through a double monochromator (SPEX 1403) in conjunction with a photomultiplier and a boxcar integrator (PAR 165). The monochromator was calibrated by using a Hg lamp. When we needed to observe the photoluminescence spectra without disturbing the cavity alignment, a small mirror (C) was inserted inside the cavity as shown in Fig. 1.

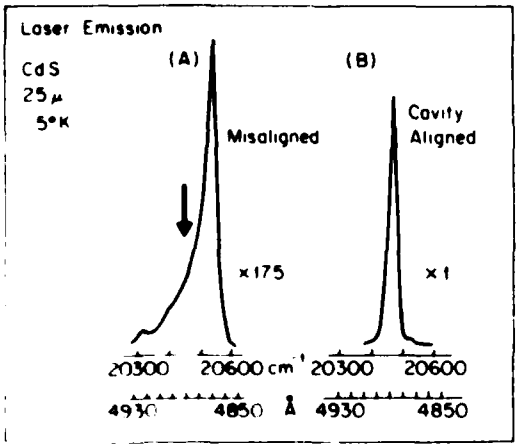

FIG. 2. Two spectra taken with the optical cavity misaligned (A), and aligned (B). The lasing frequency position is indicated by an arrow.

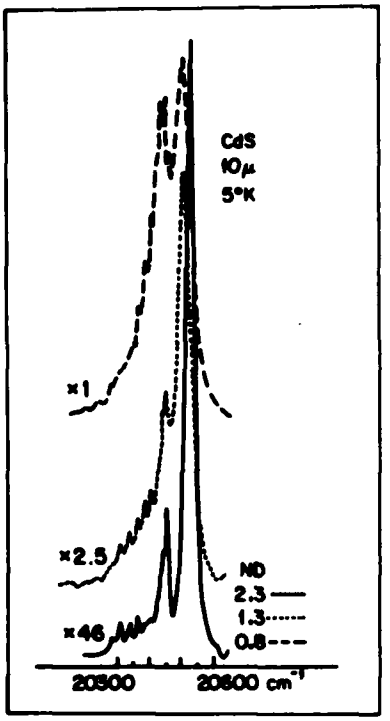

FIG. 4. Luminescence spectra taken from the 10-μ sample through the channel P (see Fig. 1). Relative pump power levels as well as the magnification factors are shown next to the spectra. See text for details.

III. RESULTS AND DISCUSSION

Laser oscillations were observed from CdS platelets by carefully aligning the mirror B while watching the intensity of the emission spectra. Over 600-fold increase in the signal level in the region of $\approx$4890 Å was detected with the alignment of the cavity. As an example, two spectra taken from a 25-μ-thick sample, before and after the cavity alignment, are shown in Fig. 2. Not only is the signal level but also the qualitative change in the spectral shape noticed. The lasing position is indicated by an arrow in Fig. 2(A). The halfwidth (HWHM) of the laser oscillation is about 3 Å in Fig. 2(B) but may vary with the pump power. The laser emission is plane-polarized perpendicular to the C axis of the CdS platelets.

In Fig. 3, the emission signal from the same sample as in the case of Fig. 2 is plotted as a function of the pump power in a log-log scale. The crosses are experimental points and the solid line is drawn to aid the eye. The lasing threshold is at $\approx$12-W peak power of the pump beam, which corresponds to the power density of approximately 20 KW/cm^2 at the sample surface. The lasing threshold may be lowered from the present value by increasing the dimension or the reflectivity of the mirror B.

The conversion efficiency is estimated to be 10% in this sample. It is not quite meaningful, however, to put too much emphasis on the absolute value of the lasing threshold or the conversion efficiency in this work, since the lasing is impurity related and thus varies from sample to sample or from spot to spot even in one sample. The surface quality was also found to greatly affect the lasing effect.

It is interesting to note that Hurwitz observed the lasing from an e-beam pumped CdS platelet at LHe temperature in the wavelength region of 4910 Å with the estimated conversion efficiency of less than 1%.[13] The CdS sample was supposed to be of high purity, and this fact was supported by the lasing in the P-band region. From the comparison between the conversion efficiency of this work and that of Hurwitz, it appears that the CdS sample with I_1 impurity center can be a more efficient lasing medium than the pure CdS.

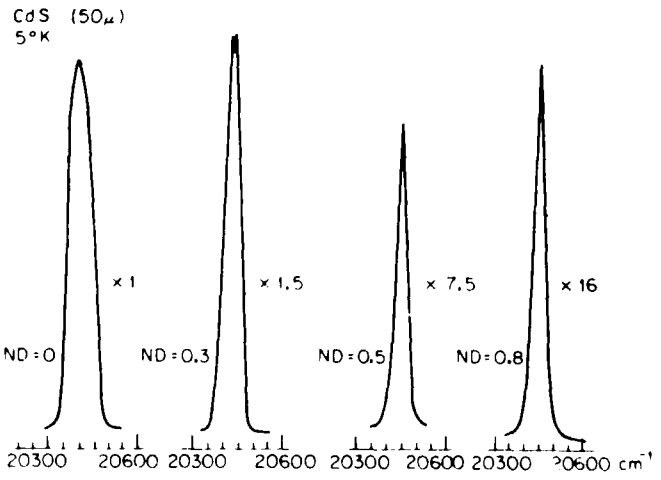

FIG. 5. Laser emission spectra from the 50-μ sample taken at different pump power levels. Changes in the peak position and the spectral halfwidth are noticed.

In Fig. 4, we show the luminescence spectra taken from a 10-μ sample at different power densities. These spectra were collected in the following way; when lasing was observed, the mirror C was inserted into the cavity (see Fig. 1) in order to detect the luminescence from the very spot where the lasing originated. The pump power was reduced (by using ND filters) to $\approx$0.4 W to obtain the spontaneous emission spectrum shown at the bottom of Fig. 4. The strongest peak at 4867 Å is I_2, the small peak at 4888.5 Å is I_1 and A-LO shows up as a shoulder at 4927 Å. With the increase of the pumping power, qualitatively different spectra were observed. The top spectrum in Fig. 4 was taken at the power density near the lasing threshold. The emergence of the so-called M band at 4871 Å makes it difficult to quantitatively discuss the spectral changes. It is obvious though, the luminescence intensity in the I_1 region increases much faster than that in the rest of the spectrum. It is to be noted that the P band at 4905 Å still did not clearly show up at the power level near the threshold.

As the pumping power is increased further, the lasing shifts toward longer wavelengths accompanied by the increase in its spectral halfwidth as shown in Fig. 5. The increase in the halfwidth is partly due to the emergence of the P band at $\approx$4905 Å. At a high excitation level, with the increase of the number of electrons excited to the conduction band, the exciton density increases, and some of the excitons decay through exciton-exciton scattering (P band).[4] It is not surprising that in the presence of bound excitons the P band appears at a higher excitation level than the bound exciton luminescence, since a bound exciton is a complex of *one* exciton and an impurity center, whereas the P band originates from the interaction between *two* excitons.

Now let us discuss the mechanisms for laser oscillations in the I_1 bound exciton region. At LHe temperature the I_1 transition is at 4888.5 Å with the halfwidth of $<$0.5 Å.[8] Nevertheless, we have observed the lasing effect over a wide wavelength range between $\approx$4880 Å and $\approx$4900 Å. In other words, the gain profile was quite broad compared to the halfwidth of the I_1 transition itself. It is most likely that the broad gain profile is due to the acoustic-phonon-assisted I_1 transitions. I_1 bound excitons are known to couple strongly (compared to I_2, for example) to acoustic phonons.[4,14] Also the stimulated emission has been observed in the acoustic phonon wing of the I_1 (sometimes referred to as the P_M band).[12] This observation led several researchers to propose a laser scheme involving I_1 or I_2 bound excitons and acoustic phonons.[12,15] To our knowledge, however, a clear demonstration of the lasing in this region has not been reported until now.

What is noteworthy in our work is that the lasing is observed not only at the lower energy side (Stokes wing) but also at the higher energy side (anti-Stokes wing) of the I_1. For instance, the lasing at frequencies higher than that of I_1 by $>$10 cm^{-1} has been observed in all three samples (10, 25, 50 μ) reported in this work. It is conceivable that the temperature rise induced by the pulsed-laser irradiation onto the sample causes the laser oscillations at the higher energy side of the I_1. The lasing at the anti-Stokes wing, however, is more noticeable at low pumping levels. Thus, the temperature ef-

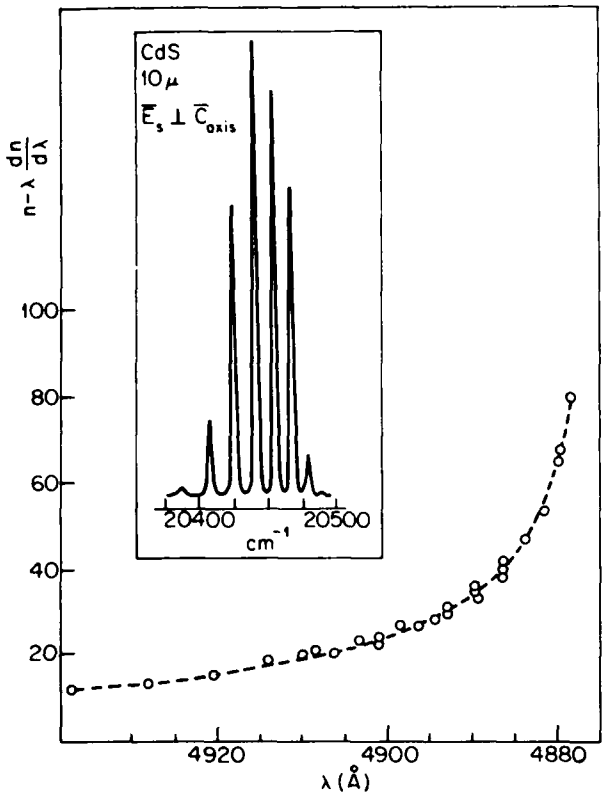

FIG. 6. The dispersion of $[n - \lambda (dn/d\lambda)]$ values deduced from fringe spacings. The broken line is drawn only as a visual aid. The inset shows a laser-emission spectrum exhibiting interference fringes with the resolution limited by the instruments. The true FWHM of each spike is about 1 Å. See text for details.

fect does not seem to be the sole cause of laser oscillations about the I_1 frequency. Further study is needed to clearly understand the origin of this phenomenon.

As the temperature of the sample is raised, the lasing shifts to longer wavelengths. The laser oscillations could be observed up to 250 °K. To our knowledge this is the highest temperature at which an optically-induced laser action has been observed in CdS. Up to 40 °K, the lasing frequency does not change much. At higher temperatures, the lasing frequency decreases at the rate of -6.45×10^{-4} eV/°K, which is faster than the rate of the reported band-gap energy change, -5.8×10^{-4} eV/°K.[16] Since the I_1 bound excitons are supposed to dissociate between 30 and 50 °K,[8,17] processes other than I_1 transitions are involved with the lasing at higher temperatures. According to the theoretical calculations in Ref. 5, exciton-electron scattering is the dominant mechanism for lasing in the high temperature region. Fischer *et al.* also regarded the laser action they observed in an *e*-beam pumped CdS as due to exciton-electron scattering.[18] The lasing wavelength of ≈5180 Å we observed at 250 °K agrees well with the result of Fisher *et al.*[18] Thus it seems reasonable to assume that the laser action were observed at temperatures beyond the dissociation point of the I_1 is associated with exciton-electron scattering.

When the two surfaces of the sample are parallel to each other as in the case of the 10-μ sample used in this work, Fabry–Perot type interference fringes were observed as shown in the inset of Fig. 6. The fringe spacings are > 1 cm^{-1}, too large to be related to the external cavity length of 5 cm. Besides, the fringe spacing varies with the emission wavelength. The spacing gets larger toward the longer wavelengths, reflecting the dispersion of the refractive index, n, of the sample. By using the relationship between n and the fringe spacing $\Delta\lambda$ such as

$$\Delta\lambda = \frac{\lambda^2}{2d}\frac{1}{\left(n - \lambda\frac{dn}{d\lambda}\right)} = \frac{\lambda^2}{2d}\frac{1}{\bar{n}},$$

where d is the sample thickness, the dispersion of $\bar{n}$ can be derived. The results are shown in Fig. 6. Our data agree well within experimental uncertainty with the value of $\bar{n}$ obtained at 4.2 °K by Hurwitz near 4910 Å from an *e*-beam pumped (parallel) CdS laser with no external cavity.[19,20] At 100 °K, the fringe spacing we observed at the lasing wavelength of ≈4975 Å is > 50 cm^{-1}, which implies that a single mode operation is possible with a CdS platelet of thickness $< 5\mu$ at this temperature.[2]

We finally report that the lasing in CdS could be achieved by resonant pumping at LHe temperature without the front mirror B. In this case, the back reflector (mirror A) and the front surface of the sample form a resonator cavity. The polarization direction of the pumping laser beam was made parallel to the C axis of the CdS sample and its wavelength was set in the vicinity of the I_{1B} bound exciton at 4862 Å.[21,22] This resonant behavior of the laser action we observed again confirms that the laser emission reported in this work is indeed associated with I_1 bound excitons. Experiments involving resonant pumping are still in progress, and will be published elsewhere.

IV. SUMMARY AND CONCLUDING REMARKS

We have demonstrated lasing effects from the CdS semiconductor platelets in an external cavity by optically pumping the samples with 4-nsec dye laser pulses. A very simple experimental geometry was used employing a hemispherical resonator cavity, thereby simplifying alignment procedures. Furthermore, in our experimental configuration, the pump beam characteristics such as the power density, the wavelength and the polarization direction can be easily changed without disturbing the cavity alignment. The only medium (other than the sample) inside the cavity is the cryostat dewar window so that the cavity loss is kept minimal. The low pump threshold value of ≈12 W found in this work suggests that the sample can be pumped by light sources other than lasers. In fact, the lasing effect has been observed in our laboratory by exciting the sample with the superflourescence from the dye mixture. We have also shown that the lasing can be achieved up to 250 °K, the highest temperature for the optically-pumped CdS laser operation reported so far. Green laser emission near 5180 Å is

observed at this temperature. As is well known of semiconductor lasers, a broader range of tunability from UV to IR can be achieved by using different semiconductors as lasing media, including compound semiconductors and heterostructure semiconducting superlattices.[23]

We have identified the observed lasing phenomena as associated with I_1 bound excitons in view of the lasing frequency position. A relatively broad gain profile is attributed to the participation of acoustic phonons in the lasing transition processes. The involvement of the I_1 in the observed lasing action is more firmly established in the resonant pumping experiments, where I_{1B} states are directly excited. Detailed mechanisms for the lasing processes, however, cannot be understood fully by the experiments reported here alone. The dynamics of the formation and the decay of the bound excitons, their interaction with acoustic phonons, and the competition between extrinsic (impurity related) and intrinsic processes will become more clearly understood if high-resolution time-domain experiments are carried out.[24] The origin of the lasing at frequencies higher than the I_1 may also be better elucidated through time-domain experiments. We hope that our work reported here will evoke the interests of the theorists in this area.

ACKNOWLEDGMENTS

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Photoacoustic study of two-photon absorption in hexagonal ZnS

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Photoacoustic detection technique is employed to investigate two-photon absorption (TPA) effects in ZnS over a wide frequency range with polarized 5 nsec dye laser pulses. By analyzing the dispersion and polarization dependence of TPA, we find that near the band gap, the TPA in ZnS occurs via allowed-forbidden type one-step, two-photon transitions.

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INTRODUCTION

The high sensitivity of photoacoustic spectroscopy (PAS) technique has been amply demonstrated in various one-photon absorption measurements.[1,2] As previously reported, the pulsed PAS technique is also suited for the study of nonlinear absorption in solids.[3–5] In this work, extensive two-photon absorption (TPA) study in hexagonal ZnS is reported. The TPA measurements were made over a wide frequency range, using 5 nsec polarized dye laser pulses and the sample-transducer configuration. The TPA effect in ZnS was previously investigated by two-beam transmission technique employing a flash lamp and a strong pump laser (Ruby or Nd:YAG).[6–8] The dispersion data were limited to near the band gap region below 4.2 eV or a few discrete points between 4 and 5 eV. No significant attempt was made to compare the dispersion with theory.

In this work, the dispersion of two-photon absorption was photoacoustically detected between 3.65 and 5.5 eV with a dye laser beam plane polarized along the optic (z axis) of the ZnS sample. The data were compared with the TPA coefficients measured with a circularly polarized beam. The dependence of TPA on the mutual orientation of the polarization direction of the light and the crystalline axes was also investigated at a few frequency points. By adopting the theoretical approach of Berequlin *et al.*,[9] our measurements could be used to determine the dominant TPA mechanism (allowed-allowed, or allowed-forbidden transition) as well as the two-photon transition processess (two-photon absorption vs two-stage, one-photon absorption) in the ZnS sample.

I. THEORY

The attenuation of a laser beam of intensity I, travelling along the x axis of a crystal sample, can be described by

$$dI/dx = -(\alpha + \beta I)I, \tag{1}$$

where α and β are the one-photon and two-photon absorption coefficients of the sample, respectively, and higher-order processes have been neglected.[10] By using third-order optical susceptibility tensor $\chi^{(3)}$, β can be expressed as

$$\beta = \frac{96\pi^2\omega}{c^2 n^2(\omega)} Im\chi^{(3)}_{ijkl}(-\omega,\omega,\omega,-\omega), \tag{2}$$

where n is the refractive index of the light at frequency ω, and the spatial indices i, j, k, l are determined by the sample symmetry and the incident laser field polarization, in such a way that for a laser beam plane polarized along the z axis of the sample, the relevant quantity is $Im\chi^{(3)}_{zzzz}$.[11,12] For crystalline samples with no center of inversion symmetry such as ZnS, additional contribution to β arises from non-zero second-order susceptibility terms $\chi^{(2)}$. In this case, $\chi^{(3)}$ in Eq. (2) is replaced by effective $\chi^{(3)}$ which can be written as[12,13]

$$\chi^{(3)\text{eff}}_{ijkl} = \chi^{(3)}_{ijkl} + \frac{2}{3}\frac{4\pi}{\epsilon(2\omega)-\epsilon(\omega)}\chi^{(2)}_{ijm}\chi^{(2)}_{mkl}, \tag{3}$$

where $\epsilon(\omega)$ denotes the dielectric coefficient at ω.

The calculation of the frequency dependence of β in semiconductors requires the knowledge of the parameters of the several bands that participate in the two-photon transition processes. Exact calculation of β, therefore, is in general very difficult, if not impossible.

There are several different approaches in calculating β.[9,14–17] Here we follow the formalism which Berequlin *et al.* developed specifically for II-IV compounds to apply for their CdS experiments. Briefly, the theory deals with a nonparabolic (Kane) two band model,[18] and the intermediate virtual states are taken to be the valence and conduction band themselves. The possible two-photon transitions were group-theoretically investigated using the irreducible representation of the energy band extrema and the dipole momentum operators. The result of their calculation, therefore, can be directly applied to the specific polarization conditions of the TPA measurements near the band gap.

For a laser beam linearly polarized along the optic (z) axis ($\hat{e}||z$), the TPA dispersion can be expressed as follows:

(i) in case of allowed-allowed transition

$$\beta^{a}_{(\hat{e}\,||\,z)}{}^{a} \propto (\zeta-1)^{1/2}\zeta^{-5}. \tag{4}$$

(ii) in case of allowed-forbidden transition

$$\beta^{a}_{(\hat{e}\,||\,z)}{}^{f} \propto (\zeta-1)^{3/2}\zeta^{-5/2}[18(1.5\zeta-1)^{-2} + 2(\zeta+1)^{3/2}(1.5+\zeta^{2}+6\zeta^{-2})\zeta^{-7/2}], \tag{5}$$

where $\zeta = 2\hbar\omega/(Eg)$, Eg is the energy gap of the semiconductor, and we assumed that the index of refraction $n(\omega)$ does not vary in one-photon frequency range of our interest (far below the energy gap). By comparing the measurements of β with Eqs. (4) and (5) we can determine whether the two-photon transitions in the sample we study are of allowed-allowed or allowed-forbidden type.

Additional information on the sample material can be ascertained by investigating the anisotropy of β. The anisotropy can be revealed when the laser polarization direction is

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varied relative to the sample orientation. For a plane-polarized laser beam travelling normal to the optic axis of the hexagonal crystal (C_{6v}), the polarization angular dependence of β can be expressed as follows:

$$\beta(\theta)/[\beta(\theta=0)] = 1 + A\sin^2\theta + B\sin^4\theta, \tag{6}$$

where θ is the angle between the optic z axis and the incident laser field $\hat{e}$, and $\beta(\theta)$ was normalized to 1 when $\hat{e} \parallel z$. The two coefficients A and B in Eq. (6) can be expressed in terms of third-order nonlinear susceptibilities or vice versa,[19]

$$\frac{Im\chi^{(3)eff}_{yyyy}}{Im\chi^{(3)eff}_{zzzz}} = 1 + A + B,$$
$$\frac{Im\chi^{(3)eff}_{yyzz}}{Im\chi^{(3)eff}_{zzzz}} = \frac{A+2}{4}. \tag{7}$$

The above equation (7) shows that there are three independent tensor elements involved in our experimental geometry and once the absolute value of one element, for example $\beta(\hat{e}\parallel z)$, is known, the rest can be determined by measuring the angular dependence of β. It should be noted that due to the birefringence of the sample, the initially plane-polarized laser field becomes elliptical as it traverses through the sample along the x direction. Equation (6) was derived for a sufficiently thick (compared to the thickness of a quarter-wave plate) sample, incorporating the birefringence effect.[9] In the case of ZnS, the quarter-wave plate thickness at the laser wavelength 5000 Å is 31 μ.[20]

For the purpose of comparison, we show the angular dependence of the linear absorption for the same geometry as in the case of Eq. (6),

$$\alpha(\theta)/[\alpha(\theta=0)] = 1 + C\sin^2\theta, \tag{8}$$

where α was again normalized to 1 for $\hat{e}\parallel z$. For Eqs. (6) and (8), we see that there is a distinct difference between the angular dependence of the linear and the two-photon absorption effects.

II. EXPERIMENTAL DETAILS

The photoacoustic two-photon absorption experimental setup used for this work is essentially the same as that reported in Ref. (4). The 5-nsec, 15-pps nitrogen laser-pumped dye laser was the excitation source. The hexagonal ZnS sample (C_{6v}) was oriented and cut along the major axes to the dimension of $10\times10\times2$ mm. The light travelled normal to the optic z axis lying on the 10×10 mm surface. A 1/2 in.-long PZT transducer was glued to the 2×10 mm side of the sample. Both the large front and back surfaces were highly polished to minimize the direct scattering of the light to the transducer.

The photoacoustic signal and the laser power in front of the sample were simultaneously monitored. At a fixed frequency, the laser power dependence of the photoacoustic signal was investigated by using ND filters to ensure that our signal is indeed of the two-photon absorption in origin. An example of such measurement is shown in Fig. 1. Up to a certain power level the square-law power dependence was observed as expected from two-photon absorption processes. At higher laser power levels, however, a saturation effect

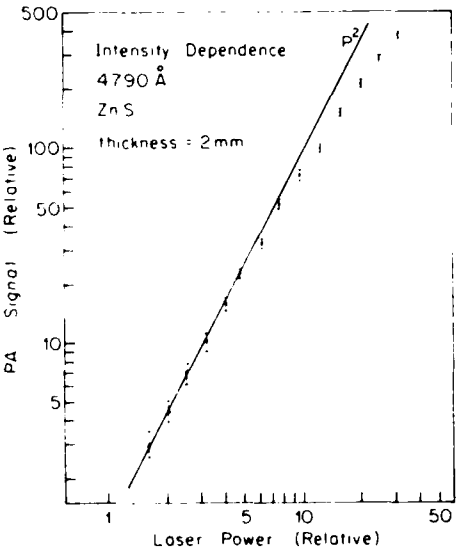

FIG. 1. Laser power dependence of photoacoustic signals from the ZnS crystal. Experimental data are represented by dots with the error bracket affixed to them. The solid line indicates the quadratic dependence of the signal on the laser power.

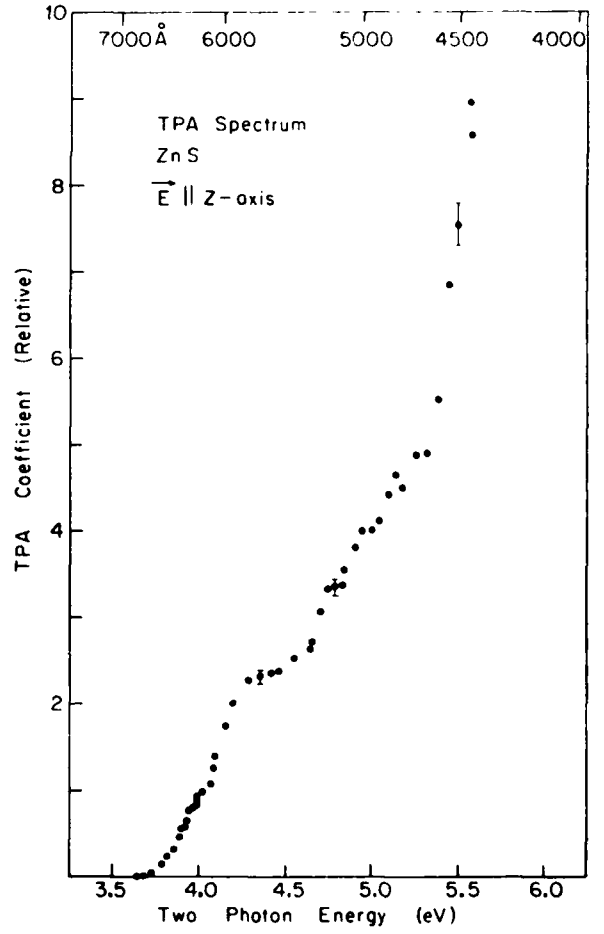

FIG. 2. Photoacoustic TPA spectrum of ZnS taken with the laser field plane polarized along the optic axis of the crystal. The top horizontal scale represents the incident laser wavelengths.

was noticed. This phenomenon can also be explained within the framework of TPA theory associated with thick bulk samples.[21]

When the laser frequency was varied (while maintaining the same power level), the signal level increased or decreased exhibiting the dispersion of the TPA signal.[19] At each frequency, the laser power dependence measurements such as that in Fig. 1 was repeated and the quadratic dependence was confirmed. The wide range of the laser wavelength, 4400–6800 A, was investigated in this work by using different laser dyes. In order to increase the accuracy of our measurements, we tried to use as many different dyes as possible so that the lasing range of one dye overlaps that of another dye. Altogether nine different dyes were used for the measurements shown in Fig. 2.

The polarization state of the laser beam was made linear or circular by using a Glan laser polarizer and/or a Fresnel Rhomb, respectively. For the anisotropy measurements shown in Fig. 3 the laser beam was made circular first and then the linear polarizer was rotated to maintain the same power level. It should be noted that each data point in Figs. 2 and 3 was deduced from several measurements made at different power levels to ensure the p^2 dependence.

In estimating the laser power inside the sample, the reflection from the sample surfaces was considered independent of the polarization state or the wavelength of the laser beam. The anisotropy of the hexagonal ZnS is rather small, for example, at 5000 Å, $n_e - n_0 = 0.004$. The dispersion of the index of refraction is also weak in our wavelength region; n changes by less than 6% between 4400 and 6800 Å.

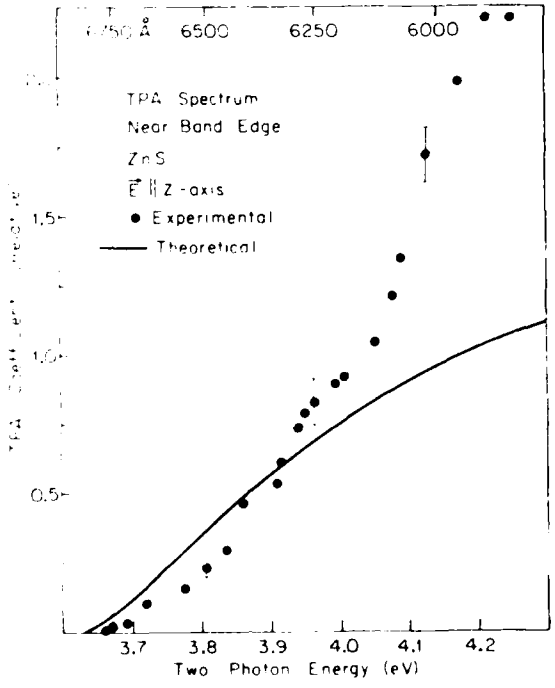

FIG. 3. The photoacoustic TPA spectrum of ZnS near the band edge is shown with the theoretically calculated curve using Eq. (5).

III. RESULTS AND DISCUSSION

The TPA spectrum in the range of 3.65–5.6 eV is shown in Fig. 2. The TPA coefficients increase smoothly up to 5 eV, then increases sharply beyond that point. Overall, the β values vary by about four orders of magnitude in the energy range we examined.

The previously reported TPA data in ZnS were obtained by two-beam transmission technique. The absorption of the weak probe light (flash lamp) was induced by the strong pump laser beam (Ruby or Nd:YAG) and the probe beam transmission through the sample was monitored. The TPA dispersion data were limited to near the band gap region below 4.2 eV[6,7] or a few discrete points between 4 and 5 eV.[8] No attempt was made to compare the dispersion data with theory. The TPA coefficients measured at 4 eV varied between 2×10^{-3} and 4×10^{-3} cm/MW, which correspond to $Im\chi^{(3)}$ values of 3.5×10^{-13} esu and 7×10^{-13} esu, respectively.[6–8]

Since our experiments were carried out using a single dye laser source and the previous measurements were made with two beams at different frequencies, ω_1 and ω_2, our data $\beta(2\omega)$ cannot be directly compared with previous data, $\beta(\omega_1+\omega_2=2\omega)$. Also, depending on relaxation mechanisms of the excited electronic states in the conduction band, the photoacoustically-detected TPA dispersion data can differ from those measured by optical techniques. However, general variation of the spectrum in the energy range previously investigated appears to agree reasonably well with our data within the experimental uncertainties.

When comparing with the previous data, it should be noted that for the TPA measurements made at two beams at different frequencies ($\omega_1\neq\omega_2$), the coefficient 96 in Eq. (2) increases by a factor of 2 due to the change in the frequency arguments of $\chi^{(3)}(-\omega_1,\omega_1,\omega_2,-\omega_2)$. Thus the β measured with one-beam at ω, $\beta(\omega+\omega=2\omega)$ can be approximated by one-half of $\beta(\omega_1+\omega_2=2\omega)$ for the same two-photon energy.[13] With this approximation and by taking $\beta(\omega_1+\omega_2=4$ eV$)=3\times10^{-3}$ cm/MW, the smallest β value we measured is deduced to be $\simeq3\times10^{-6}$ cm/MW. This value, however, should not be taken as the sensitivity limit of the present technique since only a few KW laser peak power was used and the signal to noise ratio was still about 5 in this measurement. Therefore the present technique appears to have a potential as a sensitive tool for the detailed study of the absorption edge.

The TPA spectrum near the band edge was compared with the theory. The experimental data agreed well with the dispersion calculation based on allowed-forbidden type transitions β^{af} (Eq. 5), but showed clear discrepancy with the calculation for allowed-allowed transitions, β^{aa} (Eq. 4). The dominant term in the two expressions β^{aa} and β^{af} is $(\xi-1)^{1/2}$ and $(\xi-1)^{3/2}$, respectively, and the increase of experimental β values near the edge is too fast to be fitted by the square-root dependence, $(\xi-1)^{1/2}$. In Fig. 3 we show the TPA data near the band edge together with the calculated curve β^{af} using the energy gap $E=3.63$ eV.[22] The experimental point at 3.9 eV was normalized to the theoretical value at the same energy. The deviation of the experimental

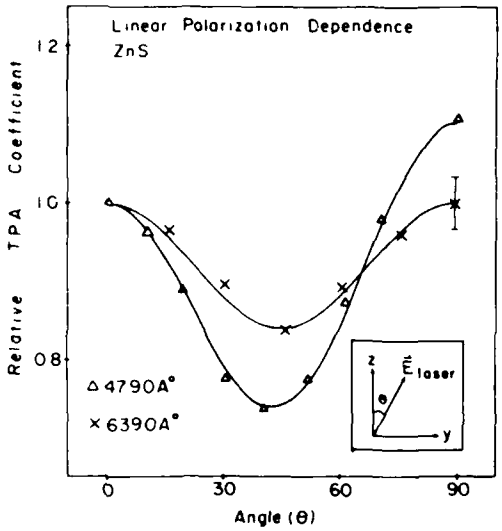

FIG. 4. Linear polarization dependence of TPA in ZnS. The experimental data are fitted with the theoretical formula in Eq. (6). θ represents the angle between the polarization direction of the incident laser and the optic axis of the crystal.

points from the theory toward the high energy values is not surprising in view of the simplified band model used in the calculation.

The allowed-forbidden transition processes are further evidenced by our two-photon linear-circular dichroism (LCD) measurements.[9] The TPA coefficients for linearly polarized beam, $\beta_{\rm lin}$, were measured to be larger by 10%–20% than those for the circularly polarized beam, $\beta_{\rm cir}$. According to the theory based on $a-f$ transition, the ratio $\beta_{\rm lin}/\beta_{\rm cir}$ is larger than 1, but does not exceed 1.2 up to 5.5 eV.[23] Our measurements show reasonable agreement with the theoretical values. Thus, from the measurements of the dispersion as well as those of LCD, we can conclude that the TPA in ZnS takes place via allowed-forbidden transition processes.

Finally, we discuss our data of the linear polarization dependence of TPA in ZnS (See Fig. 4). We show our anisotropy measurements of β at two different wavelengths, 4790 and 6390 Å. The anisotropy is more pronounced at the shorter wavelength. In the mixed crystal $Zn_{0.6}Cd_{0.4}S$, Brodin *et al.* also observed that $\beta(\theta=90°)>\beta(\theta=0)$, as in the case of our measurements at 4790 Å.[24] The experimental data were compared with theoretical angle dependence shown in Eq. (6) using A and B as adjusting parameters. The best fits are shown as solid curves. From this fit, the relative $\gamma^{(3)}$ values could be deduced using the relationship in Eq. (7). The results are listed in Table I.

We should point out that the polarization dependence data in Fig. 3 cannot be fitted with Eq. (8) describing linear absorption effect. When inspecting Eq. (8), we notice that, contrary to our observation, the minimum of α occurs either at $\theta=0$ or at $\theta=90°$ depending on the sign of the coefficient C. Since the two-stage transition (two successive one-photon absorption) should also reflect the linear polarization dependence, our data imply that the linear absorption as well as

TABLE I. Relative $\gamma^{(3)}$ values[a] deduced from the linear polarization dependence of TPA measured in ZnS.

Wavelength (Å)	$\frac{\gamma^{(3)}_{yyyy}}{\gamma^{(3)}_{zzzz}}$	$\frac{\gamma^{(3)}_{yyzz}}{\gamma^{(3)}_{zzzz}}$
4790	1.10	0.22
6390	1.00	0.34

[a] $\gamma^{(3)}$ denotes $Im\gamma^{(3)}_{\rm eff}$.

two-stage absorption processes did not make noticeable effects on our measurement.

IV. CONCLUDING REMARKS

The photoacoustic spectroscopy technique is demonstrated to be a sensitive method of studying nonlinear absorption effects in a solid sample. From the anslysis of the photoacoustically-detected dispersion as well as the linear-circular dichroism (LCD) of β, the allowed-forbidden type transition was found to be the dominant two-photon absorption mechanism in ZnS. The linear-polarization dependence was also observed to be consistent with what is expected from TPA theory. This polarization dependence together with LCD indicates that the contribution of any linear absorption or of two-stage absorption to our experimental data is not significant.

It should be mentioned that the present TPA theories do not offer full description of our experimental data. The dispersion was interpreted toward the band edge leaving the high-energy region unexplained. The interpretation of the linear polarization dependence relies only on the group-theoretical formalism. Extension of the present theory including the effect of several bands would not only provide better understanding of the nonlinear optical absorption processes, but also reveal more accurate information on the band structure itself.

ACKNOWLEDGMENTS

The authors are grateful to Dr. Y. S. Park and Dr. J. Owen for providing the ZnS samples used in this work. Y. Bae thanks P. S. Lee for his technical assistance. This work was supported in part by the Office of Naval Research.

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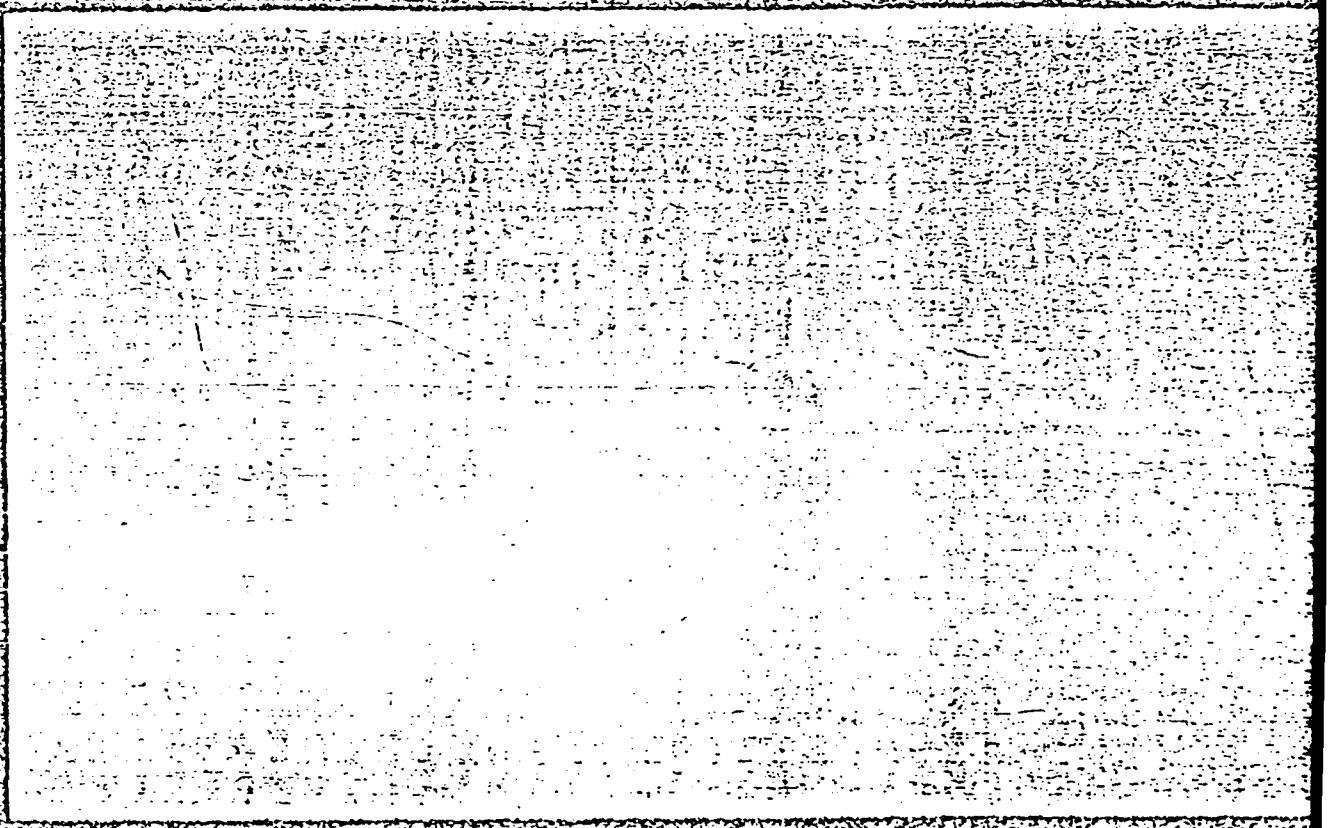

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(Title of paper. Capitalize only the first letter of each principal word.)

W. C. Wang and J. J. Song
(Author)

University of Southern California
(Institution)

Laser Oscillations were observed from optically-pumped CdS platelets from 5°K to 250°K. Bound-excitons are involved in the lasing transitions at low temperatures.

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BOUND-EXCITON LASER ACTION IN CdS.

(Title of paper. Capitalize only the first letter of each principal word.)

W. C. Wang and J. J. Song

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Department of Physics, University of Southern California

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Los Angeles, CA 90089-0484

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The stimulated emission and the laser action were investigated in CdS platelets at low temperatures with, and without external cavities. The undoped, vapor-grown samples 10μ to 50μ thick were optically pumped by 4 nsec dye laser pulses. Typical lasing in CdS at low temperatures is known to occur at wavelengths longer than 4900 Å and involves the P-band or A-LO transitions. We have observed the lasing from CdS at shorter wavelengths in the region of I_1 bound-excitons at the pumping power density far below the level sufficient for the observation of the P-band. The lasing in this region is attributed to the acoustic-phonon-coupled I_1 bound exciton transitions. With the increase of the sample temperature, the lasing shifted toward longer wavelengths at the rate of -6.45×10^{-4} eV/°K so that at 250°K the laser emission was observed at 5180 Å. The lasing could also be observed at low temperatures without the front coupling mirror of the resonator cavity when pumping directly into the I_{1B} states of CdS. The simple hemispherical resonator configuration as well as the resonant pumping method used in this work can be applied to the study of a variety of nonlinear optical effects in semiconductors.

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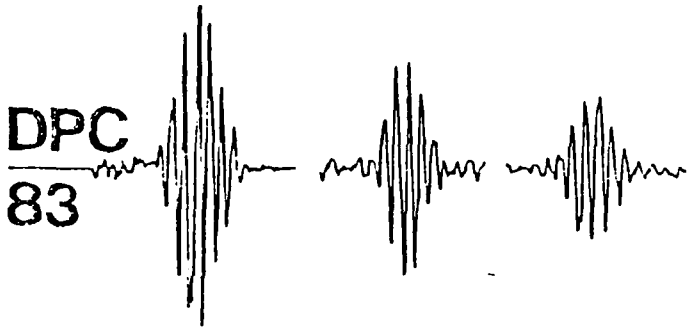

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Laser Oscillations in the Bound-Excitonic Region of CdS
(Title of paper. Capitalize only the first letter of each principal word.)

W. C. Wang and J. J. Song
(Author)

Dept. of Physics (Department) University of Southern California (Institution)

University Park, Los Angeles, CA 90089-0484
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Laser action was observed from optically pumped undoped CdS platelets inside a hemispherical resonator cavity from 5°K to 250°K. At low temperatures, the lasing was observed in the region of excitons bound to neutral acceptors (I_1 bound exciton). The conversion efficiency is estimated to be ~10% in contrast to less than 1% reported for intrinsic laser action involving exciton-exciton interactions (P-band). The laser oscillations were observed in the Stokes as well as anti-Stokes side I_1. Acoustic phonons seem to participate in the lasing transitions. With increasing pumping power the lasing shifts to longer wavelengths accompanied by an increase in halfwidth and onset of saturation. A significant drop in the lasing threshold level was noticed when the frequency of the pumping beam was approximately at resonance with the I_{1B} bound exciton states. Implications of this resonant behavior will be discussed.

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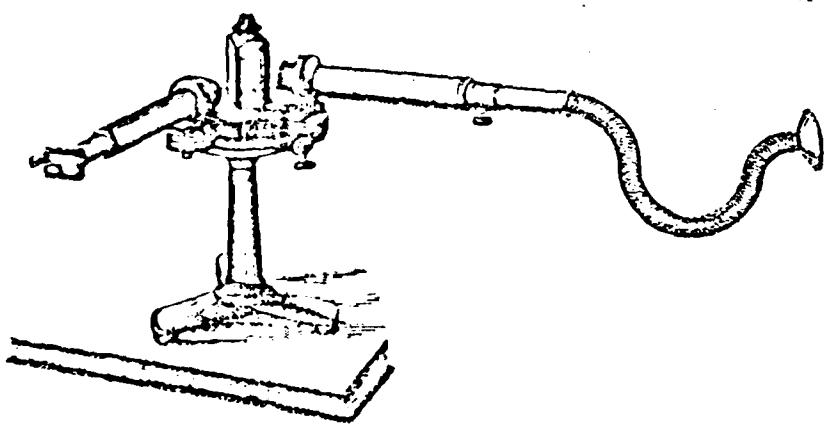

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PHOTOACOUSTIC DETECTION OF POLARIZATION-DEPENDENT NONLINEAR OPTICAL EFFECTS IN CRYSTALS

By

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Using photoacoustic detection technique, we have investigated two-photon absorption (TPA) effects in oriented single crystals, ZnS (hexagonal), ZnO and $SrTiO_3$, with particular attention to the dependence of TPA on the frequency and polarization of the incident laser beam.

A nitrogen-laser-pumped dye laser (5ns, 15pps) was the excitation source. The photoacoustic signal was detected using a PZT transducer and a gated boxcar integrator. Significant efforts were made to determine the optimum design of the transducer, theoretically as well as experimentally. TPA coefficients, β, less than 10^{-6} cm/MW could be detected using laser powers of a few KW.

Since one-photon absorption and TPA obey different polarization selection rules, the measurements of the

polarization-dependent TPA effects in crystals are valuable in understanding their energy band structures. The measurement of the linear-circular dichroism of β is also useful in studying the nature of the TPA processes in semiconductors.[1]

In II-VI semiconductors, β for a linearly polarized beam was measured to be larger than β for a circulary polarized beam. It was also found that β is dependent on the relative orientations of the crystals with respect to the polarization direction of the laser field. Such variation was detected even in a cubic crystal $SrTiO_3$, in contrast to the case of linear absorption. In II-VI compounds, however, β was found to be independent of the crystal orientation if the laser beam traveled along the sixfold z-axis of the samples. These observations are consistent with what is expected from a group-theoretical analysis.

In Fig. 1, we show, an example of our experimental data obtained in ZnS at $4800A^{o}$ as a function of the angle Θ between the z-axis of the crystal and the polarization direction of the electric field. β was normalized to unity at Θ = 0. For this geometry, the angle dependence of β should behave according to the following equation:

$$\frac{\beta(\Theta)}{\beta(\Theta=0)} = 1 + A \sin^2\Theta + B \sin^4\Theta \qquad \text{Eq. 1}$$

where A and B depend on the excitation energy and the sample

properties. Our data were fitted with Eq. 1 using A and B as fitting parameters, and the best fit is shown as a solid curve in Fig. 1.

We will present our experimental results and discuss their implications, based on the theoretical work in Ref. 1.

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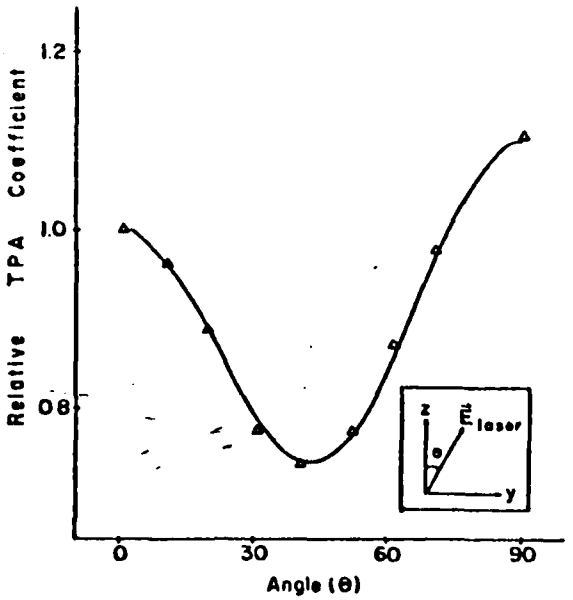

Fig. 1. Polarization-dependence of β in ZnS. Experimental geometry is shown in the inset.

Photoacoustic detection of nanosecond-pulse-induced optical absorption in solids

Y. Bae, J. J. Song, and Y. B. Kim

Photoacoustic detection of optical absorption in bulk solids is investigated with sample-transducer configuration using 5-nsec tunable dye laser pulses. The technique is shown to be sensitive for the measurements of nonlinear as well as linear absorption. The analysis of the resonant character of the piezoelectric transducer and the photoacoustic response of solids is also presented.

I. Introduction

Recently the photoacoustic (PA) detection technique has received much attention as a sensitive tool for the measurement of low absorption in various forms of materials.[1-11] In particular, pulsed photoacoustic spectroscopy became very attractive because strong short pulses can induce nonlinear optical effects, such as Raman gain and two-photon absorption (TPA), and they also enable the study of various phenomena that occur in a short time scale.[4-8]

In this paper we report our investigation of photoacoustic effects in bulk solid samples arising from linear or nonlinear absorption of 5-nsec laser pulses over a wide frequency range. Previous photoacoustic TPA investigations in solids were limited to the measurements at a fixed (1.06 μm) (Ref. 6) or a few discrete wavelengths using microsecond long laser pulses.[5]

Among the various experimental configurations to measure photoacoustic spectra of solids, we chose the solid-transducer (ST) technique over the solid-gas-microphone (SGM) or the solid-liquid-transducer (SLT) technique.[9] The piezoelectric detection method offers a distinct advantage in the time domain over a gas-microphone system. Also ST is expected to be more sensitive than SLT due to superior impedance matching between the solid sample and the transducer. Moreover direct contact between sample and detector will minimize the acoustic wave attenuation in the intermediate medium (liquid in SLT, for example).

We first theoretically analyzed the function of the piezoelectric transducer as an acoustic wave detector. Then we experimentally compared the PA signal response from the several transducers with various lengths and confirmed the resonance behavior of the transducer, which is expected from our theoretical analysis. With proper choice of the transducer, we could measure two-photon absorption coefficients β smaller than 10^{-6} cm/MW in $SrTiO_3$ and ZnS. We also found that around the 5000-Å region, the PA spectrum in Nd^{3+}: glass is consistent with the transmission data. To our knowledge, this work is the first report on PA detection in solids by the ST technique with nanosecond laser pulses.

II. Operation of Piezoelectric Transducer as an Acoustic Wave Detector

For application of the piezoelectric transducer to PA detection, we need to understand the electrical response of the transducers under acoustic excitation. As the first step toward understanding this effect, we consider the operation principle of the piezoelectric length-expander with end electrodes as shown in Fig. 1(a).[12,13] V and I denote the voltage and current driven to the transducer, respectively. The velocity of the end surface and the force exerted upon them are denoted by U and F, respectively. The transducer dimension is represented by l, w, and d. Figure 1(b) shows the general electrical equivalent circuit of this transducer, which was derived in Ref. 12 for the case of alternating temporal motion of the piezoelectric medium with angular frequency ω. The sound velocity of the medium is denoted as v. N, C_0, and Z_0 are the material-dependent constants here (for details, see Ref. 12).

Next we apply this idea of an electrical equivalent circuit to the piezoelectric transducer as a detector of sound waves [Fig. 2(a)]. The transducer produces the voltage V between the electrodes at both ends when

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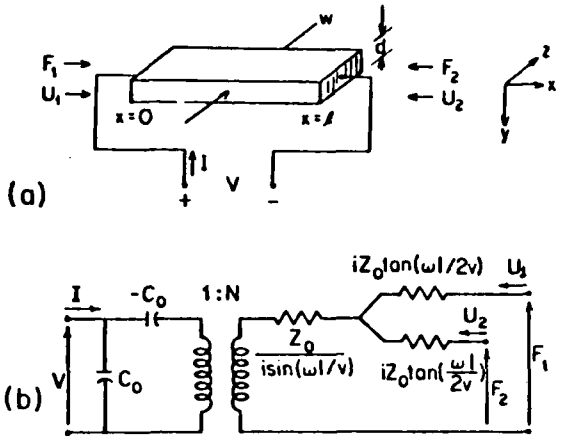

Fig. 1. (a) Piezoelectric transducer with end electrodes. The exciting electric field is parallel to its length; (b) complete equivalent circuit of the length-expander (see text for details).

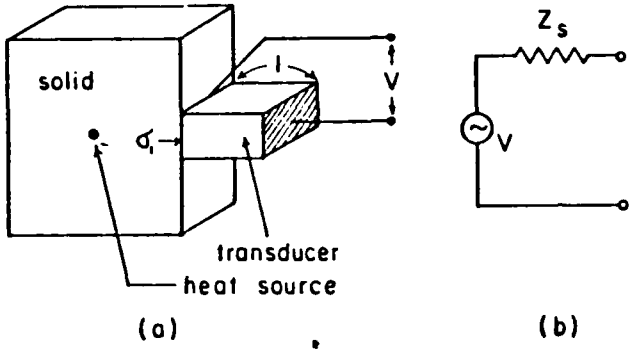

Fig. 2. (a) Sample-transducer configuration; (b) equivalent circuit of a piezoelectric detector shown in Fig. 2(a).

excited by the stress wave σ_1 originating from the heat source. The electrical response of the transducer can be deduced from Fig. 1(b). The F_2 terminal can be considered as shorted for the free end at $x = l$ and the F_1 terminal as coupled with the stress wave. By taking $F_1 = wd\sigma_1$ and $F_2 = 0$ we obtain, in a straightforward way, the Thevenin equivalent circuit. The source voltage V and the source impedance Z_s are given as follows:

$$V = \frac{wd\sigma_1 N}{Z_0 C_0 \omega} \tan\frac{\omega l}{2v}, \tag{1}$$

$$Z_s = \frac{1}{iC_0\omega}\left(1 - \frac{2N^2}{Z_0 C_0 \omega}\tan\frac{\omega l}{2v}\right). \tag{2}$$

From Eq. (1) we see that the source voltage becomes infinity (i.e., resonant) when the transducer length l is an odd multiple of $\lambda/2$:

$$l = \frac{\lambda}{2}, \frac{3\lambda}{2}, \frac{5\lambda}{2}, \ldots, \tag{3}$$

where λ is the wavelength of the acoustic wave. Another striking aspect of the function of the piezoelectric detector is that the source impedance Z_s also becomes infinity at the resonance. For actual measurements we should consider the signal voltage drop across the source impedance when using an instrument of finite input impedance.

Suppose that a finite impedance Z_L is loaded across the output terminal of the transducer. Then the load voltage V_L will be

$$V_L = V\frac{Z_L}{Z_L + Z_s}. \tag{4}$$

Near resonance such that

$$\tan\frac{\omega l}{2v} \gg \frac{Z_0}{2N^2}\sqrt{1 + \omega^2 C_0^2 Z_L^2}, \tag{5}$$

the load voltage becomes

$$V_{L,\mathrm{res}} \simeq -i\frac{wd\sigma_1\omega C_0 Z_L}{2N}. \tag{6}$$

We can also easily prove that the load voltage obtained near resonance conditions turns out to be the maximum for the finite load Z_L. Thus it is desirable to choose the transducers that satisfy the resonance conditions.

It will be interesting to investigate the response of the detector to an acoustic wave having overtone frequency components of the fundamental resonant frequency. According to Eq. (1), the source voltage tends to become smaller for higher overtones. This tendency, however, becomes reversed for the load voltage in Eq. (6).

III. Experimental Setup

Figure 3 shows our experimental setup. The dye laser was pumped by a (doubled or tripled) Nd:YAG laser (Quanta-ray) operating at 10 pps with 5-nsec duration. The dye laser beam passed through a number of optics and was incident on the sample through a converging lens. Both the photoacoustic signal from the PZT transducer glued to the sample and the incident laser power were simultaneously monitored through the signal and the reference channel, respectively. The piezoelectric signal was measured with a boxcar integrator (PAR 162) in conjunction with an amplifier with 1-MΩ input impedance and 1-MHz bandwidth. The laser power was detected by the combination of a silicon photodiode and a radiometric

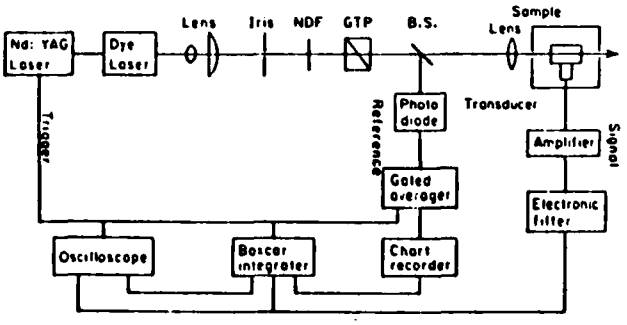

Fig. 3. Photoacoustic experimental setup. NDF, GTP, and BS represent the neutral density filter, the Glan-Thomson polarizer, and the beam splitter, respectively.

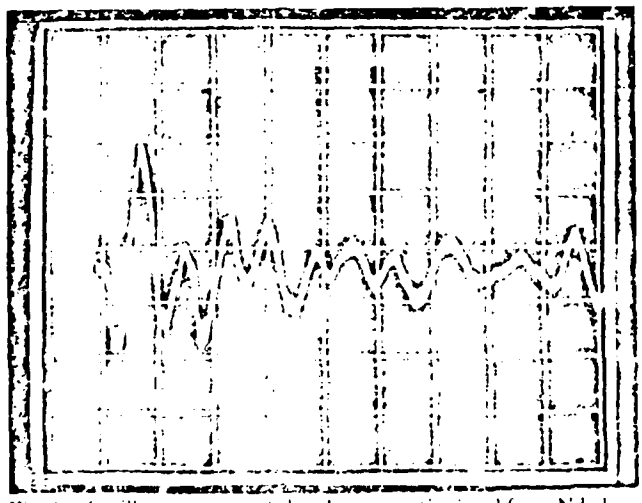

Fig. 4. Oscilloscope trace of the photoacoustic signal from Nd:glass excited by 5-nsec laser pulses (transducer length = 6.35 mm, 5 μsec/div, 0.2 V/div).

filter.[14] Together they produce a flat spectral response between 4500 and 9000 Å. The outputs from both channels were observed on the oscilloscope (Tektronix 465) and recorded by a dual-channel strip chart recorder. The dye laser was of Hänsch type and stepping-motor controlled. The laser halfwidth was 0.7-cm^{-1} HWHM. We checked carefully that the spatial characteristics of the beam did not change during the laser scanning.

The transducer tested ranged from 1.3 to 44 mm in length with the same cross-sectional area, 5 × 7 mm. Each was glued to the 5- × 50-mm side of the Nd:glass rod, which had dimensions of 5 × 5 × 50 mm, and its output voltage was measured in identical experimental conditions. The transducer material was PZT ceramic C5500.[15] The frequency constant of the ceramic is 1.95 kHz m when used as a thickness expander.[13,15]

The absorption of the Nd:glass was measured at ~5000 Å using a 12.7-mm long transducer which was found to show the highest responsivity from the previously mentioned transducer test.

The experimental arrangement for the two-photon absorption measurement is essentially the same as that for the linear absorption measurement of the Nd:glass.

In both the linear and two-photon absorption measurements, the input laser power dependence of the signal was investigated by varying the laser power with neutral density filters. In most cases, the laser peak power measured in front of the sample did not exceed ≈10 kW. Further details of the TPA experiments will be discussed elsewhere.[16,24]

IV. Behavior of the Photoacoustic Signal

An example of the time-dependent photoacoustic signal from the PZT transducer attached to Nd:glass is shown in Fig. 4. The Nd:glass sample was 5 mm in thickness, and the transducer used was 6.35 mm in length. The signal exhibits a damped wave train as well as beatings.

The largest peak in the wave train is the first positive one. The signal starts ~2 μsec after the triggering point, which closely corresponds to the incidence of the laser pulse on the sample. Considering the sound wave velocity, 5000 m/sec in glass[17] and 3980 m/sec in the PZT ceramic,[15] we estimate that the 2-μsec interval is the time taken for the acoustic wave to travel through the sample and the transducer. The main frequency appears to be ~250 kHz. This value is close to the resonant frequency, calculated using the frequency constant of the PZT material mentioned in Sec. III. The frequency of the acoustic wave in a sample is theoretically expected to be dependent on the laser beam diameter inside the sample.[18] We observed, however, that the main frequency of the wave train, i.e., 250 kHz, remained fixed with the change in the beam focusing geometry. We also noted that the frequency did not change with different samples or even in the case where the sample was severely damaged due to high laser power. These observations can be regarded as an indication that the transducer is sensitive only to the acoustic waves with the resonant frequency of the transducer itself. At the same time this behavior may also be regarded as indirect experimental evidence of the resonant characteristics expected from Eq. (1). The beating effect observed may be due to the interference between the primary waves and the waves reflected at the sample boundaries.

The resonant behavior of the transducer is well illustrated in Fig. 5, which shows the PA signal vs the length of the transducers. Among the eight transducers, maximum responsivity was obtained at 12.7 mm.

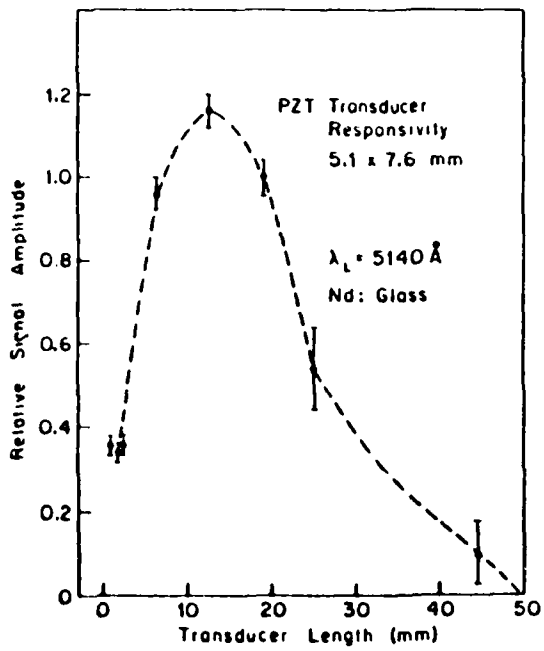

Fig. 5. Length-dependent responsivity of the transducers. Solid circles represent the experimental points with uncertainties indicated by the error bracket. Dashed line is drawn to aid the eyes (see text for details).

Table I. Calculated Fundamental Resonance Frequency and the Observed Signal Frequency Detected by the Various Length Transducers Directly Attached to Nd:Glass Under the 5-nsec Laser Pulse Excitation

Length (mm)	Observed signal frequency	Calculated fundamental resonance frequency
1.27	2.0 MHz	1.6 MHz
1.91	1.4 MHz	1.1 MHz
2.54	1.0 MHz	780 kHz
6.35	240 kHz	310 kHz
12.7	120 kHz	160 kHz
19.1	80 kHz	100 kHz
25.4	30 kHz	78 kHz
44.4	20 kHz	45 kHz

Not much change in the signal was observed below l = 2.5 mm. Table I lists the observed frequency of the PA wave train as well as the calculated fundamental resonance frequency for each transducer. The observed frequency appears to be close to the calculated values. The larger discrepancy for long ones may be due to the fact that the long transducers deviate from the assumed thickness-expander geometry. We also noticed that not only the signal level but also the SNR was poorer for longer transducers, possibly due to the slow environmental noise.

The acoustic signal from the solid is considered to consist of many waves with different frequencies (broadband spectrum). Each transducer senses an acoustic wave whose frequencies are close to its own resonance frequency. At this point, we would like to mention that we should not be surprised to observe the slow acoustic response of the solid in the 100-kHz range to the excitation by the short (~100 MHz) laser pulses. This can be understood by taking into account the elasticity of solids. In a simplified picture, we may consider the heat injection area [Fig. 2(a)] as a vibrating circle. During heat injection the circle will expand to the maximum size. After the heat injection is over, the vibrating circle will shrink to a size smaller than its equilibrium value due to the restoring force arising from the elasticity of the medium, while the heat diffuses to the surrounding medium. The analogous case may be found in a simple pendulum which is excited within 5 nsec and then released to oscillate freely. The free oscillation frequency of the pendulum will be governed by its own characteristic frequency.

V. Photoacoustic Detection of Linear and Nonlinear Absorption in Solids

When a laser pulse of power P is incident on a sample, the stress developed in a solid can be written in the form

$$\sigma = K_1 \alpha P + K_2 \beta P^2, \tag{7}$$

where α and β are the linear and two-photon absorption coefficients of the sample, respectively, K_1 and K_2 depend on material properties and the laser beam characteristics.

We measured the PA signals from Nd^{3+}:glass in the range from 4800 to 5400 Å (coumarin dye 500) using a 12.7-mm thick transducer. The photoacoustic spectrum shown in Fig. 6 is the plot of the amplitude of the first peak which appeared 7 μsec after the laser pulse heat injection. The distinct two peaks are identified as $^4G_{9/2}$ and $^2G_{9/2}$ manifolds. For the purpose of comparison, we measured the transmittance spectrum of the same sample using a Beckman spectrophotometer (ACTA MVI). The total absorption derived from the PA spectrum agreed well with that measured with the spectrophotometer in the wavelength range examined.

Farrow *et al.*[19] measured the absorption of Nd:glass using a photoacoustic technique with the light source modulated at 135 Hz. The resulting PA data were quite different from the absorption spectrum taken by the transmission technique in the same wavelength range. The existence of the peak $^4G_{9/2}$ was not clear in their PA spectrum. It is reported that the lifetime of the level ($^4F_{3/2}$) that dominantly contributes to the radiative emission processes of Nd^{3+} ions is of the order of 700 μsec.[20] In Ref. 19 the photoacoustic signal was monitored for a long (compared with 700 μsec) period of time, while the ions are continuously excited by the incident photons. In our experiments, however, PA signals are monitored shortly (a few-microsecond range) after the nanosecond laser excitation. The complicated relaxation dynamics of Nd^{3+} ions may have caused the discrepancy observed between the transmittance data and the PA results taken by the cw technique. The resemblance of our PA data taken with nanosecond pulses and the transmission data indicates that the two absorption peaks have similar mechanisms for generating acoustic waves in the time scale of our experiments. It will be interesting to compare the PA measurements with the absorption data over a much wider wavelength range using short laser pulses.

We also report our nonlinear absorption measurement in transparent bulk crystals ($SrTiO_3$, ZnS, and

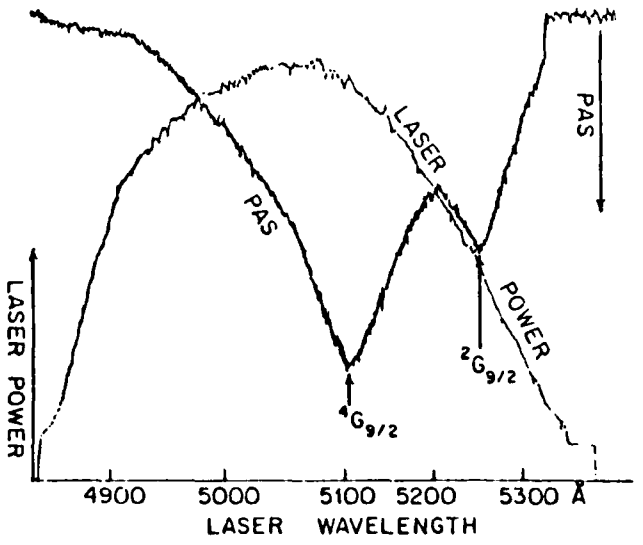

Fig. 6. Chart recorder trace of the laser power and the photoacoustic signal from Nd:glass.

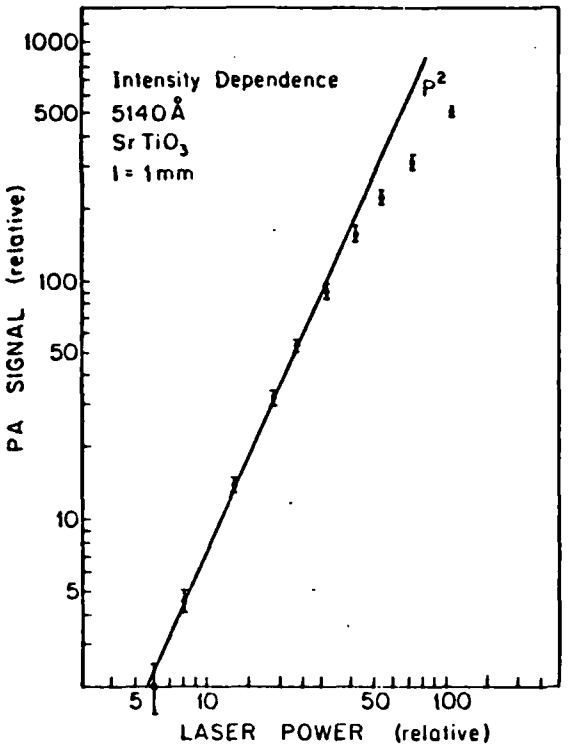

Fig. 7. Laser power dependence of a photoacoustic signal from $SrTiO_3$ crystal. Experimental points are represented by dots with error bracket. Solid line indicates the quadratic dependence of the signal on the laser power.

TiO_2) with energy gaps larger than 3 eV. As shown in Fig. 7, the PA signal in these samples exhibited quadratic dependence on the incident laser power as expected from the TPA processes. Other effects such as linear absorption or laser scattering from the transducer would not exhibit such a square dependence. The deviation of the signal from p^2 dependence with increasing laser power levels may be due to the optical limiting effect as in the case of two-photon transmission measurements.[21]

We measured the dispersion of two-photon absorption coefficient β in these crystals over a wide frequency range by using several different dyes. Figure 8 shows one example of our experimental results obtained from $SrTiO_3$. By comparing the PA dispersion data with the absolute value of β, measured using the transmission technique, we could deduce that the PA signal we measured near the band gap of $SrTiO_3$ and ZnS corresponds to $\beta < 10^{-6}$ cm/MW.[22-24] In general, β of TiO_2 was found to be much larger than that of the ZnS or $SrTiO_3$.

Finally, we would like to mention that the dispersion of β near the energy gap of ZnS and $SrTiO_3$ could be well fit by the theoretically calculated curves. Details of the experimental results and the corresponding theoretical analysis on the dispersion will be discussed elsewhere.[16,24]

VI. Summary and Concluding Remarks

We have successfully applied the photoacoustic detection technique to the linear as well as TPA measurements in bulk solids by using 5-nsec tunable dye laser pulses.

The electrical response of the piezoelectric transducer under acoustic excitation was theoretically analyzed and shown to exhibit resonant characteristics [see Eqs. (1)–(3)]. The resonant behavior was experimentally demonstrated by measuring the PA signal from several transducers with different lengths attached to an Nd:glass sample.

The PA spectrum from Nd:glass near 5000 Å was measured to be consistent with the transmission measurement. We demonstrated the high sensitivity of the PA technique by measuring very small nonlinear TPA effects near the band gap of transparent crystals. We further utilized the technique to study the various aspects, for example, dispersion and polarization dependence, involved in the TPA effects in crystals. Manuscripts are in preparation for publication on this (TPA) subject.[16,24]

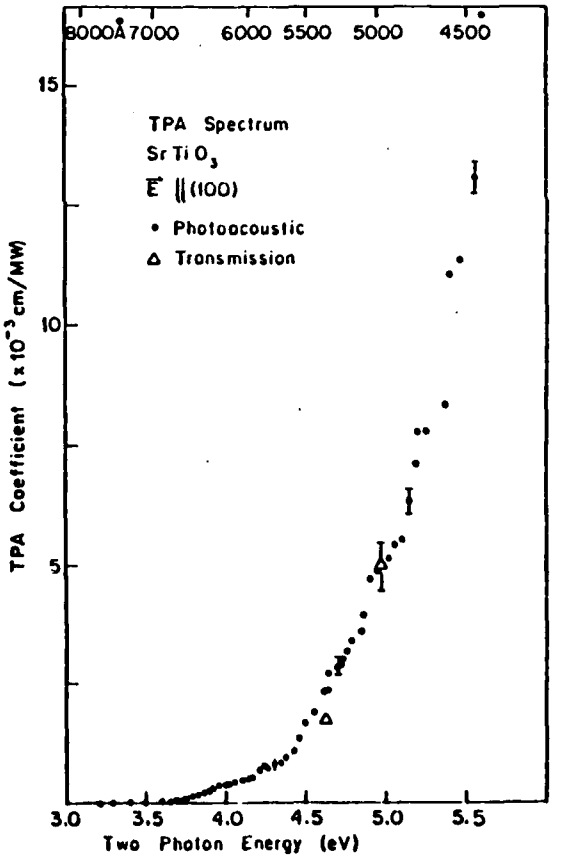

Fig. 8. TPA dispersion of $SrTiO_3$ obtained by the PA technique. Dots represent our PA measurements. Top horizontal scale represents the incident laser (one-photon) wavelength. The laser field was linearly polarized along the (100) axis of the sample. Triangles represent absolute value measurements of β by the transmission technique.[23] Data were normalized to absolute β at 5.0 eV.

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Meetings Schedule

OPTICAL SOCIETY OF AMERICA
1816 Jefferson Place N.W.
Washington, D.C. 20036

6–8 January 1982 INTEGRATED AND GUIDED-WAVE OPTICS, SIXTH TOPICAL MEETING, Asilomar Information: Meetings Department at OSA or CIRCLE NO. 48 ON READER SERVICE CARD

8–10 March 1982 LASER TECHNIQUES FOR EXTREME ULTRAVIOLET SPECTROSCOPY, TOPICAL MEETING, Boulder Information: Meetings Department at OSA or CIRCLE NO. 50 ON READER SERVICE CARD

13–15 April 1982 OPTICAL FIBER COMMUNICATION, FIFTH TOPICAL MEETING, Phoenix Information: Meetings Department at OSA or CIRCLE NO. 49 ON READER SERVICE CARD

14–16 April 1982 CLEO 82, CONFERENCE ON LASERS AND ELECTROOPTICS, Phoenix Information: Meetings Department at OSA or CIRCLE NO. 64 ON READER SERVICE CARD

17–21 May 1982 SPRING CONFERENCE ON APPLIED OPTICS, Rochester Information: Meetings Department at OSA or CIRCLE NO. 56 ON READER SERVICE CARD

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