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THE STUDY OF THE INTERACTION OF INTENSE PICOSECOND LIGHT PULSES WITH MATERIALS

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13. ABSTRACT

This report contains the summary of the works performed in the entire period of this grant. We have studied the two-photon and three-photon photoconductivity effects in various semiconductors. The effect was then applied to measure the ultrashort optical pulses. Two-photon pump GaAs bulk laser action was demonstrated. A theoretical calculation on the optical third harmonic generation with broad band picosecond pulses was made. Finally various aspects of the nonlinear optical interaction at the boundary of nonlinear media were investigated. The data agreed well with Bloembergen and Pershan's theory. The observation of the nonlinear Brewster's angle was reported for the first time in a transparent medium.

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I. Introduction

The research effort in this work has been devoted to the understanding of the interaction between intense picosecond pulses with matter. Since the power associated with such laser pulses is so high, the interactions are usually nonlinear in nature. We have studied, during this grant period, the two-photon and three-photon effects the semiconductors. We have also applied these effects to the measurements of the ultra-short laser pulses. Optical harmonic generation is one of the most important nonlinear optical effects. In the past, there are limited data on the behavior of such nonlinear interaction near the boundary of a nonlinear medium. In this work we have been able to obtain good experimental data which verifies many important aspects of Bloembergen and Pershan's theory in nonlinear optics. For example, we have now established a one to one corresponding between the laws of linear and nonlinear optics. In particular, we have shown convincingly for the first time, in a KDP crystal, the experimental observations of nonlinear Snell's law, nonlinear Brewster's angle, nonlinear critical angle for total internal reflection.

In this final report, we shall summarize some of the important findings for the work mentioned above, toghether with other works in this period, such as the laser induced damage studies, theoretical calculation of third harmonic generations by picosecond pulses and two-photon pumped semiconductor lasers.

For detail technical information, we referred the readers to the appropriate technical reports and/or the published papers.

II. Summary of the research effort.

(1) Multi-photon conductivity in semiconductors and its application for the measurement of picosecond pulses (Jayaraman and Lee, period: Dec. 1969 to March 1973).

(a) Two-photon conductivity in GaAs with nanosecond and picosecond pulses (Jayaraman and Lee[1], Dec. 1967 to Dec. 1971).

The effect of photoconductivity in GaAs single crystals was investigated by using both nanosecond and picosecond light pulses for excitation so that we can study both the steady state and transient state response from the samples. The two-photon conductivity in GaAs was found to exhibit a square law dependence on laser intensity, however, only over a very narrow range of laser power. The two-photon absorption coefficient was measured to be 5.6 cm/MW. This agrees favorably with the experimental value of Basov and also with our theoretical calculation. Fast relaxation of the photo-induced carriers suggests that GaAs is a good material as fast photo-detection.

(b) Three-photon conductivity in CdS[2] (Jayaraman and Lee, Dec. 1971 to June, 1972).

The photon-conductivity in CdS single and polycrystals wan investigated by using mode-locked Nd:glass laser pulses for excitation and was found to exhibit a power law of $I^{2.8 \pm 0.2}$, where I is the peak excitation intensity The three photon absorption coefficient can be estimated from the photo-condictivity measurement. They are 0.04 cm^3/GW^2 for polycrystal and 0.013 cm^3/GW^2 for single crystal of CdS. These data agreed well within an order of magnidutde with the thoeoretical values.

(c) Measurement of ultrashort optical pulses by two-photon photoconductivity techniques [3] (Lee and Jayaraman, July 1972 to March 1973.)

Two-photon conductivity in $CdS_{0.5}Se_{0.5}$ was investigated and compared to that obtained from GaAs with a view to determine which one is more suitable for picosecond pulse width measurement. The contrast ratio using GaAs as two-photon conductor was 1.8 and it increased to 2.4 when $CdS_{0.5}Se_{0.5}$ was used. The improved contract ratio in the case of $CdS_{0.5}Se_{0.5}$ was partly due to the improved resolution of the crystal thickness (2 psec) and partly to the extended square-law region. This technique clearly shows that it should be especially useful to measure short pulses in the 2 to 10 μm region and should be very adequate to measure the infra-red pulse of 20 psec duration or longer. Furthermore, because the fabrication of such a dectection device is quite compatible with integrated electronic technology, it is conceivable that an opto-electronic device consisting of high density packed two-photon conductors with multichannel data-requisition capability can be built based on priciple demonstrated here.

(2) Investigations of laser action in GaAs with two-photon pump, (Park and Lee, Sept. 1971 to March 1972).

Laser action in GaAs single crystal by using a Nd:glass laser beam as a two-photon pump source was investigated[4]. External mirrors were employed to form the optical cavity. Laser action in GaAs was observed from the following experiments. Firstly, we observed a line narrowing of the fluorsecence spectrum of GaAs from 300 Å abruptly down to 16 Å when the threshold pump power was reached. Secondly, the output radiation at 8500 Å showed order of magnitude increase when the pump power is above 6 MW/cm^2.

The advantage in this method of excitation is the possibility of achieving uniform population inversion through out the bulk of the semiconductor instead of just near the surface. The use of external reflections for optical cavity also offers the possibility for obtaining simulataneous Q-switching and mode-locked pulse in the near infrared region. This pulse again can be wavelength tuned by changing the temperature of the GaAs sample.

(3) Laser-induced damage studies by X-ray diffraction techniques[5], (Wu, Armstrong and Lee, Dec. 1970 to June 1971).

The Berg-Barrett X-ray diffraction contrast technique has been utilized to study the process of laser-induced damage in a zinc crystal and, also to make observations on the annealing characteristics of the dislocation substructure before and after the laser treatment. The laser-induced damage required a threshold enery which cause melting, possible vaporization and appreciable plastic deformation of the matrix material by deformation twining, nonbasal slip, and microkinking. By comparision with the structural rearrangement observed in the preannealed specimen, the laser damage zone showed only very retarded changes.

(4) Third harmonic generation by picosecond optical pulses, (Coffey and Lee, June 1972 to December 1972).

A theory for the optical third harmonic generation of the broad-band picosecond pulses was developed. Using Maxwell's equations as the starting point[6], we have derived and solved the wave equation for the third harmonic field resulting from the propagation of a broad-band fundamental beam through an isotropic nonlinear medium. The material parameters considered were the nonlinear susceptibility giving rise to the third harmonic generation, the respective phase and group velocities, and the nonlinear interaction length. Three examples consisting of a sine wave, a Gaussian pulse and a linearly chirped pulse were then examined.

(5) Nonlinear optical interaction in KDP prisms with picosecond pulses. (Lee and Bhanthumnavin, June 1972 to June 1973).

KDP is one of the most studied nonlinear materials. However most of the works performed on KDP have been in transmission with normal incidence under the phase-matching condition. To extend the optical laws of reflection and refraction from the linear case to its nonlinear counterpart one must perform the experimeriments with oblique incidence over a wide angular range. The behavior of the light waves at the boundary of a nonlinear optical medium was analyzed by Bloembergen and Pershan[(7)] in 1962. Since then many experimental works have been devoted to the verification of those predictions. For example, the nonlinear equivalence of the Snell's law was verified by Ducuing and Bloembergen[(8)] in semiconductor, nonlinear Brewster's angle in absorbing semiconducting materials by Chang and Bloembergen[(9)], and nonlinear total reflection by Bloembergen, Lee and Simon[(10)] in $NaClO_3$.

In this work, we present new and improved experimental data on KDP, which include the observation, for the first time, the existence of a real nonlinear Brewster's angle in a transparent medium. The achievement on the improved data was attributed to the use of the mode-locked laser beam from a Nd:glass laser. This not only extends the dynamic range for non-linear study but also drastically reduces the intrinsic fluctuation of the data due to phase-locking.

The KDP prism was immersed in an optically dense fluid, 1-bromonapthalene which has larger indices of refraction than KDP at both ω and 2ω. This arrangement facilitates the observation of nonlinear Brewster's angle and the enhanced phase-matched relected second harmonic signal at total relection in the same experimental run. The KDP crystal had the face normal of its entrace surface aling the [111] direction and the fundamental laser beam was polarized along the $[1\bar{1}0]$ direction. The p^{NLS} was then in the [001] direction which is in the plane of incidence. The reflected harmonic intensity $I_R(2\omega)$ generated from the crystal was observed as a function of the angle of incidence, which varied from 20° to 75°. The experimental data showed a pronounced peak at the critical angle $\theta_i = 66.78^\circ$ and pronounced dip (zero signal) at $\theta_i = 42.83^\circ$ which corresponds to the theoretical predicted nonlinear Brewster's angle for this geometry. The reflected harmonic intensity varied between these two extreme cases by more than seven orders of magnitudes.

Data for the detailed variation of the intensity of both the transmitted and reflected second harmonic beams near the critical angles for total reflection in a KDP prism will also be presented for various crystal orientations and polarizations. In some case, the incident angle varied over a very wide range, the corresponding transmitted harmonic signal showed maximum corresponding to the usual phase-matching and minimum corresponding the case that the p^{NLS} is along the direction of radiation. The intensity variation was more than ten order of magnitude in this case. The data are compared and appear in excellent agreement with the theory of Bloembergen and Pershan. The deviation of the data points from the theoretical curve is less than 5 percent, mainly due to the use of the mode-locked laser.

The transmitted second harmonic intensity under noncollinear phase matching condition by means of two beam spatial mixing in KDP crystal was demonstrated as a function of the angle of incidence. Again the result was in good agreement with the theory of Bloembergen and Pershan. The application of this result for the measurement of the second order intensity auto-correlation function of the picosecond pulses will also be discussed.

III. Publications in this grant period.

A. Papers published in the journals.

1. Observation of two-photon conductivity in GaAs with nanosecond and picosecond light pulses, S. Jayaraman and C. H. Lee, Appl. Phys. Letters Vol. 20, 392 (1972).

2. Berg-Barrett X-ray observation of annealing and laser-induced damage in zinc, C.C. Wu, R. Armstrong and C. H. Lee, J. Appl. Physics, Vol. 43, 821, (1972).

3. Observation of three-photon conducticity in CdS with mode-locked Nd:glass laser pulses, S. Jayaraman and C. H. Lee, J. Appl. Physics, Vol. 44, 5480, 1973.

4. Measurement of Ultrashort optical pulses by two-photon photoconductivity techniques, C. H. Lee and S. Jayaraman, Opto-electronics, Vol. 6, 115, 1974.

B. Paper presented in meetings.

1. Observations of two-photon conductivity in GaAs by a giant pulse Nd:glass laser, S. Jayaraman and C. H. Lee, Bull. Am. Phys. Soc., Vol. 16, p. 653 (1971).

2. Observation of three-photon conductivity in single and polycrystalline CdS, S. Jayaraman and C. H. Lee, Bull. APS, Vol. 18, p. 91, (1973).

3. Optical nonlinearity at total reflection in KDP, V. Bhantumnavin and C. H. Lee, Bull. of APS, Vol. 18, p. 657 (1973).

4. Multiphoton conductivity in semiconductors and its application to ultrashort pulse measurement, S. Jayaraman and C. H. Lee presented at the Conference on Laser Engineering and Applications, May 30 - June 1, 1973 at Washington, D.C.

C. Papers in submission.

1. Nonlinear optical interactions in KDP prisms, C. H. Lee and V. Bhanthumnavin, submitted to the 8th International Quantum Electronics Conference, June 1974.

2. Theory of third harmonic generation by picosecond optical pulses, D. W. Coffey and C. H. Lee, submitted in the American Physcial Society Spring Meeting at Washington, D.C., April 1974.

D. Other related papers published and presented during this period but not supported by this grant.

1. Measurement of ultrashort light pulses using optical third harmonic generator, R. C. Eckardt and C. H. Lee, Bull. of APS, Vol. 15, p. 88, (1970).

2. Investigation of the Nd:glass mode-locked laser with a ring cavity, R. C. Eckardt and C. H. Lee, Bull. of APS, Vol. 16, p. 592 (1971).

3. Temporal and spectral development of mode-locking in a ring-cavity Nd:glass laser, R. C. Eckardt, C. H. Lee and J. N. Bradford, Appl. Phys. Letters, Vol. 19, 420, (1971).

4. Evolution of high power picosecond pulse in a pulsed mode-locked laser, R. C. Eckardt, C. H. Lee and J. N. Bradford, paper R.8, presented at the 7th International Quantan Electronics Conferences at Montreal, Canada, May 1972.

5. Effect of self-phase modulation on the evolution of picosecond pulses in a Nd:glass laser, R. C. Eckardt, C. H. Lee and J. N. Bradford, Opto-Electronics, Vol. 6, 1974.

IV. Graduate students supported by this grant.

1. S. Jayaraman, obtaining his Ph.D. degree in May, 1973.
2. V. Bhanthumanavin, obtaining his Ph.D. degree in August, 1973.
3. Y. H. Park, obtaining his M.S. degree in Spring, 1972.
4. S. Mak, working toward his M.S. degree.

References

1. Observation of two-photon conductivity in GaAs with nanosecond and picosecond light pulses, S. Jayaraman and C. H. Lee, Appl. Phys. Letters, Vol. 20, 392, (1972).

2. Observation of three-photon conductivity i CdS with mode-locked Nd:glass laser pulses, S. Jayaraman and C. H. Lee, J. Appl. Phys., Vol. 44, 5480 (1973).

3. Measurement of ultrashort optical pulses by two-photon photoconductivity techniques, C. H. Lee and S. Jayaraman, Opto-Electronics, Vol. 6, p. 115 (1974).

4. Investigation of laser action in GaAs with two-photon pump, Y. H. Park, Master thesis, unpublished.

   N.G. Basov et al, JETP 23, 366 (1966).

5. Berg-Barrett X-ray observation of annealing and laser-induced damage in zinc, C. C. Wu, R. W. Armstrong and C. H. Lee, J. Appl. Physics, Vol. 43, 821, (1972).

6. Second harmonic generation by picosecond optical pulses, W. H Glenn, IEEE J. QE., Vol. QE-5, 2841 (1969).

7. N. Bloembergen and P.S. Persan, Phys. Rev. Vol. 128, p. 606 (1962).

8. J. Ducuing and N. Bloembergen, Phys, Letters, Vol. 6, p. 5 (1963).

9. R. K. Chand and N. Bloembergen, Phys. Rev., Vol. 144, p. 775 (1966).

10. N. Bloembergen and C. H. Lee, Phys. Rev. Letters, Vol. 19, p. 835 (1967); N. Bloembergen, H. J. Simon and C. H. Lee, Phys. Rev., Vol. 181, p. 1261 (1969).