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13. ABSTRACT *(Maximum 200 words)*

The report describes research on non-perturbative solutions to problems in resonant nonlinear optics and on photorefractive nonlinear optics and its applications. Non-perturbative solutions have been found for optical double resonance and applied to optical switching and three-wave mixing in quantum-well materials and to degenerate four-wave mixing studies on the $^2\Sigma^+ - {}^2\Pi$ (0-0) band of OH. A non-perturbative analysis has also been carried out for simultaneous one-, two-, and three-photon resonances in coherent anti-Stokes Raman spectroscopy (CARS). This allows optimization of CARS signals in diagnostic systems that operate at very low particle densities. Research has also been carried on periodicall!!y driven two-level systems and two-level systems with permanent dipole moments. In other work, the beam propagation method has been developed for use in photorefractive nonlinear optics. This method has been applied to the problem of nonlinear beam cleanup and to studies of the statistical nature of amplified scattering. Development was carried out on phase conjugate interferometry and ellipsometry measurement techniques for thin films. For holographic storage systems that use different wavelengths for reading and writing, a birefringent phase matching technique has also been developed.

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A. Exact Solution for Optical Double Resonance

The optical double resonance problem is a familiar one in optics. For this process, a near-resonant field at frequency ω_a connects level 1 with level 2 while a near resonant field at frequency ω_b connects level 2 with level 3. When level 2 is below level 3, the excitation of level 3 involves two-photon absorption; if level 2 is above level 3, the excitation of level 3 is a Raman process. We have solved this problem for the case when both fields are strong.

Previously, we described a nonperturbative technique based upon iterative replacement in the density matrix equations so that couplings between the density matrix are bilinear in the field. When either the states have parity or the resonant couplings dominate in such a way as to produce an equivalent separation, we find two sets of density matrix elements where the elements of one set have been eliminated from the equations for the other set. Thus, the replacement process involves a multifold Gauss elimination. One set (the dipole set) involves those matrix elements for dipole transitions while the other set (the diagonal set) consists of the remaining elements including the diagonal elements and, for double-resonance, the two-photon coherence terms. To solve the double resonance case, we have extended our approach by further iteration to obtain coupled equations for diagonal elements only. This results in a pair of coupled equations for population differences which contain both one- (bilinear in the field) and two-photon (quadrilinear in the field) renormalization terms. One- and two-photon coherences can be readily obtained from the population differences.

B. Low Loss Optical Switching and 3-Wave Mixing in a Semiconductor Quantum Well

Using the results described in the previous section, we have made an analysis, which predicts significant changes in band-edge absorption, index of refraction, and 3-wave-mixing susceptibility in a quantum well when two conduction subbands are strongly coupled by an electromagnetic field.

Recently, the idea of using strong optical fields to improve nonlinear optical conversion efficiencies in atomic systems has received considerable attention. Harris *et al.* proposed that a field which strongly couples two upper states of a multi-state atomic system could, under appropriate conditions, resonantly enhance the nonlinear susceptibility while at the same time induce transparency. In this scheme, transparency is achieved at the center of a Rabi-split absorption line. Following this proposal, it was demonstrated that applying a strong dc-electric field to atomic hydrogen leads to resonantly enhanced second-order susceptibility with reduced absorption at the second harmonic wavelength. These results have motivated more recent suggestions that analogous effects may be possible in semiconductor quantum wells. Manipulating absorption in a quantum well is potentially important for a wide range of optics applications including sum-frequency generation and optical switching.

In fact, manipulation of quantum well optical properties through strong field effects has already been demonstrated. Strong field excitation below the absorption edge has been shown to lead to an instantaneous blueshift of the exciton absorption resonance, an effect, which is known as the ac, or optical Stark effect. Optically Stark-shifted exciton resonances have also been demonstrated when a strong field couples two quantum well conduction subbands. In the above configurations, and in the ones to be considered in this paper, the strong optical fields do not excite real populations; this is in contrast to recently discussed quantum well nonlinearities which depend on strong field excitation of real populations.

In atomic systems, Rabi splitting and optical Stark phenomena are closely related: an atomic resonance line which is Rabi split by a strong, near-resonant field becomes simply Stark shifted when the strong field is tuned sufficiently off of resonance. In our analysis, we determine the influence of a strong, near-resonant optical field on the optical properties of a semiconductor quantum well and thereby establish the relationship between atomic and semiconductor Rabi splitting and the optical Stark effect. We study the band-edge absorption, index of refraction, and wave-mixing susceptibility in a quantum well in which two unoccupied levels are strongly coupled by the optical field. The change in absorption and refractive index can be used for the switching of a weak field by a strong field. Because the strong field connects unoccupied levels the process can be optically lossless and without severe thermal imitations.

Our analysis is of a Type I quantum well in which the two lowest conduction subbands are strongly coupled by an optical field polarized perpendicular to the well while a weak optical field probes the interband response. It is noted that this is the same general configuration used to observe the optical quantum confined Stark effect. This problem differs from that of the optical Stark effect treated theoretically by Schmitt-Rink because it requires a three-band model instead of a two-band model and because the strong optical field considered here does not couple occupied valence subbands to either of the empty conduction subbands. Thus, the scheme does not involve the generation of a high density of either virtual or real electron-hole pairs and nonequilibrium many-body techniques are not necessary to describe the effect of the strong coupling field.

We have found dramatic changes in band-edge absorption, index of refraction, and wave-mixing susceptibility in a quantum well when two conduction subbands are strongly coupled by an electromagnetic field. Both the absorption-refractive index and wave mixing susceptibilities were calculated for a quantum well in which two conduction subbands are resonantly coupled by a strong optical field. The quantum well absorption spectra is modified dramatically by the strong coupling optical field, exhibiting plateau splitting which is the quantum well analog of atomic Rabi splitting. The corresponding peak change in quantum well index of refraction is calculated to be 0.01. These changes are predicted to be very fast, suggesting potential applications to high speed optical switching devices. It has also been shown that these changes evolve into the quantum well optical Stark effect as the strong coupling field is tuned off of resonance. A strong coupling field also significantly modifies the interband quantum well wave mixing susceptibility, although these changes do not lead to enhanced nonlinear conversion efficiencies as in analogous atomic systems. However, the radiative renormalization approach may be directly extended to semiconductor quantum wire and quantum dot systems, which, by virtue of their more atomic-like density of states, deserve further study for strong field nonlinear optics applications.

C. Four-wave Mixing in Optical Double Resonance

We have developed a model of a three-level system in interaction with three cross-polarized and co-propagating fields having the same frequency. Two pump waves have arbitrary intensities and a third one (the probe wave) is assumed to be non-saturating. Using the radiative renormalization method, the density matrix equations have been solved analytically.

In collaboration with a group at ONERA in France, we have applied this model to forward degenerate four-wave mixing (FDFWM) in OH. The signal has been calculated under thermodynamic conditions where the collision and Doppler broadening are comparable. The evolution of the FDFWM intensity is computed with respect to incident laser power densities and to pressure. The signal intensity is optimal and the pressure dependence is minimized when the laser intensity is approximately equal to the saturation threshold. FDFWM experimental spectra of the $^2\Sigma^+$ - $^2\Pi$ (0-0) band of OH obtained in a welding torch flame have been compared to the theoretical profiles. For incident laser intensities in the range of a few MW/cm^2, good agreement has been obtained between calculated and experimental profiles. Discrepancies arise under stronger conditions of saturation, which limits the range of application of our model.

D. Non-perturbative Analysis of the Four-Level System with Three Strong Fields

Radiate renormalization has been used to obtain an exact steady state solution of a four level system coupled by three arbitrarily strong, near resonant, monochromatic fields. The four wave mixing and absorption terms are given for a weak fourth field. The results demonstrate the effects of optically induced energy level mixing and shifting and population redistribution. An exact treatment of the strong field case in steady state is desirable for the optimization of signals in applications such as CARS spectroscopy phase conjugation, and electromagnetically induced transparency. Perturbation theory is not capable of such an analysis when effects such as Autler-Townes splitting, Stark shifting, power broadening, and population redistribution become important.

Using these results, a study of strong field effects on resonant CARS has been performed. We have calculated the exact four wave mixing component for various resonance detunings and field strengths. The effects of vibrational and electronic saturation have also been investigated. For the case of a four level system, we have obtained steady state expressions that allow one to analytically solve for the four wave mixing component of the polarization, which is independent of the anti-Stokes field, as well as to obtain simplified numerical solutions for the absorption, which is linear in the anti-Stokes field. We have considered the four wave mixing component in three limits. The first is the weak transfer limit in which the rate out of the ground state is so slow that no significant population transfer occurs but the dynamical Stark effect is important. The second limit considered is pure Raman saturation, where population is transferred to the Raman exited state and the dynamical Stark effect is relatively weak. The final limit considered is when full electronic and Raman saturation as well as the dynamical Stark effect occur. In this case, population may be transferred to any level. For all cases considered, we assume OH "like" molecules as the molecular species under test.

E. Periodically Driven Two-Level Systems

We have generalized the steady-state, non-perturbative results obtained earlier for resonant, sinusoidal modulation of the two-level system to the case of arbitrary periodic modulation. The results can be greatly simplified if the damping constants are small compared to the frequency of periodic modulation. We have derived the reduced Bloch equations for arbitrary amplitude modulation on exact resonance. We have studied in detail the simple case where the longitudinal damping constant is equal to the transverse damping constant. We give the general steady state result, the approximate solution when the damping is weak, and the exact solution when the damping is zero. We have extended the analysis to unequal damping and give weak damping and zero damping results. We have also extended the analysis to include perturbation in detuning. Finally, we have considered the extent of the spectra and the nature of the variability of the components.

F. Two-Level Systems with Permanent Dipole Moments

The radiative renormalization technique has been used to analyze two-level systems with permanent dipole moments under the influence of a single-frequency applied field. For this problem, the advantage of radiative renormalization over the usual perturbative or harmonic balance techniques lies in the computational ease with which the density matrix operators can be determined to any order in applied field. We have characterized the absorption and dispersion of a typical two-level system within this framework and find that although the absorption saturates as expected with increasing incident field strength, additional oscillatory behavior is evident at extremely high field strengths. The structure and period of these oscillations is dependent upon the magnitude of the permanent dipole moment. The formalism can easily be applied to the problem of higher harmonic generation.

G. Diode Laser Pumped 3-4 μm Semiconductor Lasers

A student (D. Coppeta) supported under the program has collaborated with MIT Lincoln Laboratory researchers in the development of high-power semiconductor lasers emitting in the 3-4 mm region. The diode-array-pumped GaInAsSb/GaSb and InAsSb/GaSb double heterostructure lasers were operated at 85 K and yielded 95 mW average and 1.5 W peak power per facet at 3 μm, and 50 mW average and 0.8 W peak power at 4 μm. The highest operation temperature was 210 K for the 3-μm quaternary and 150 K for the 4-μm ternary.

H. Beam Propagation Method for Photorefractive Nonlinear Optics

We have extended our two beam coupling code developed earlier to four-wave mixing and are executing it on the CM-2 supercomputer at the Pittsburgh Supercomputer Center. We have made comparisons of its performance to results of previous models including one dimensional plane wave theory and the quasi-plane wave two dimensional theory developed by us previously. The comparisons hold up very well: when uniform plane waves are propagated through the crystal, the phase conjugate reflectivities predicted by the beam propagation method (BPM) and the plane wave theory are very close. Also, the beam profiles for in the quasi-plane wave model match the BPM profiles very closely.

We have also developed a method to extend our BPM to a full vector model in uniaxial crystals, in which ordinary and extraordinary polarized beams are propagated separately, and the tensor nature of the electrooptic tensor is taken into consideration.

To properly include photorefractive grating effects in the vector BPM, and to make our two-transverse-dimension models more accurate, we have had to develop a method for solving the photorefractive rate equations in two and three dimensions. These equations are very stiff, and defied solution for a long time, but we have recently been able to use the simultaneous over-relaxation method to solve the problem in 2D. Convergence is still very slow, and we are seeking techniques to speed it up.

An important consideration in photorefractive devices is the effect of amplified photorefractive scattering (commonly known as fanning). In image processing devices such as two beam coupling amplifiers, fanning is often reduces the signal to noise ratio substantially. Fanning can, however, be advantageous. For example, it is the basis of the fanning optical limiter, and is the source of seeding for many self-pumped phase conjugate mirrors. In preparation for developing a model for the scattering source, to be used in conjunction with the beam propagation method, we measured the first order statistics of unamplified and amplified scattering, and found that in the case of unamplified scattering, the statistics corresponded closely to predictions of a theoretical model for random walk scatterers. Detailed measurements of the angular distribution of scattered light have also been made. We have extended our BPM code to include the effects of such scatterers distributed throughout the crystal.

We have completed our adaptation of the Filon quadrature method to accurately evaluate the oscillatory integrals appearing in the beam propagation code. This should enhance the accuracy of our fanning model.

In the light of our new expertise in analysis of spatial interactions in nonlinear optics, we are making a careful reevaluation of nonlinear optical beam cleanup. It can be shown that the autocorrelation of the noisy pump beam appears on the amplified clean beam. This may be used to advantage in designing joint transform correlators, but will degrade the effectiveness of beam cleaners.

Several papers on these topics are presently in preparation, and should be submitted in the next six months:

Difference between the effects of volume and surface scattering.
Application of the Filon method to Hankel transforms
Application of the Filon method to the beam propagation method
Vector beam propagation
Two and three dimensional photorefractive rate equation model
Cross-talk in photorefractive beam cleanup.
Operation of the double phase conjugate mirror.

We have also completed a study of the statistical nature of amplified scattering in photorefractive crystals. Using a random walk model for scattering in the beam propagation code, we simulated amplified scattering (fanning). The characteristics of the scatterers were verified by comparison of the first order statistics of the scattered intensity with a theoretical prediction of the probability density function based on the random walk scattering model. Both the size and separation of the scatterers could be varied enabling us to tailor our numerical simulations to experimental measurements of non-amplified and amplified scattering. We showed that a one-dimensional model does not accurately simulate amplified scattering for crystals with a non-diagonal electro-optic tensor, and that surface scattering alone does not fully account for the angular dependence of the amplified scattering distributions.

I. Phase conjugate interferometry and ellipsometry

We have extended our research on phase conjugate interferometric measurement of thin films to the development of a phase conjugate ellipsometer suitable for the measurement of films on opaque substrates and for highly transparent films. The end mirror after the sample is replaced by a polarization preserving phase conjugate mirror. Using a phase conjugate mirror in place of a conventional mirror has many advantages. One is that the sampling arm is self-aligning because the phase conjugate beam retraces the path of incidence exactly so that no careful alignment is needed even when changing the sample. This makes possible real time evaluation of films during deposition. Also, most of the distortions caused by substrate irregularities and other imperfections in the optical system including viewports in growth chambers are canceled by phase conjugation.

A technical point to be considered is that photorefractive phase conjugate mirrors generally work only for one polarization direction. A polarization preserving photorefractive phase conjugate mirror can however be made by using the Basov scheme. Another point is that self-pumped phase conjugate mirrors frequently have temporal instabilities both in phase and amplitude of the reflection. Fortunately, phase instability does not affect measurement results because only the phase difference between the two detector outputs is used. Meanwhile, drift in the reflectivity can be controlled to within 0.1% during a period of one minute which is plenty of time for completion of a measurement by the ellipsometer.

J. Interferometric Polarimeter

In the course of investigating the interferometric ellipsometer, we developed a new type polarimeter for measuring the polarization state of quasimonochromatic light. This phase shifting elliptic interferometer (PEI) can measure the polarization state of light quickly with high accuracy. We expect that it will be fast, compact and relatively inexpensive.

The conventional technique for measuring polarization state is to align a rotatable wave-plate and linear polarizer followed by a photodetector in the optical path of the beam. Measurements are made of wave-plate and polarizer angle at nulls of the intensity at the photodetector. This technique, although well developed in automated instruments, is time consuming and somewhat expensive.

The layout of the PEI is basically a Michelson interferometer in which both polarization components travel common paths. The wave-plate in one arm switches the s and p polarizations in that arm so that the interferences in the interferometer outputs give the relative amplitudes of both input components as well as the phase difference between them. The two detectors measure the same quantities but in different quadratures at the same time. The detector combination thus never needs biasing to obtain maximum sensitivity.

The device has several positive features:

i) The measurement is independent of vibration in the system and the linearity of the piezoelectric phase shifting element. This is accomplished by plotting the outputs of the detectors against each other, forming an ellipse. The polarization state can be found by fitting ellipse parameters. The robustness comes about because the interferometer is the common path for the s and p components and the detector outputs are measured simultaneously. To show this, we made an experimental demonstration on an optical table without vibration isolation.

ii) The effects of angular misalignments of the input beam cancel each other out if the path lengths in both arms are equalized.

iii) The polarization ellipse can be displayed on a monitor during data acquisition. This is useful for checking the reliability of the device.

The common path and dual quadrature characteristics of the polarimeter described above can be useful for many other types of interferometers such as position sensors. We expect that such devices will have 0.1 nm sensitivity, 1 nm accuracy, and range equal to half the coherence length of the laser used.

L. Waveguide Theory

Wieslaw Krolikowski and Udo Trutschel designed a novel multichannel soliton switch consisting of an array of fibers embedded in an infinite cladding. Both the fibers and cladding are taken to have a weak self-focussing nonlinearity. By exciting five adjacent fibers with signals of suitable amplitude and phase, a spatial soliton like excitation is launched along the fiber array which delivers the bulk of its power to a specified output fiber.

M. Self Pumped Phase Conjugators for Fiber Multiplexing

In collaboration with Baruch Fischer of the Technion, we have investigated the use of the double phase conjugate mirror (DPCM) for efficient multiplexed coupling into single mode optical fibers. Several independent laser sources can be coupled through a DPCM into a single mode fiber.

N. Four-wave mixing and the double phase conjugate mirror

Using our new model of amplified scattering in photorefractive crystals, we were able to calculate the spatial behavior of the double phase conjugate mirror (DPCM) and have linked time series images into an animation.

Various one-and two-dimensional analyses of the double phase conjugate mirror (DPCM) have classified it as either an amplifier or an oscillator. We have attempted to bridge the gap in this controversy by using two-dimensional scalar numerical simulations of the device for two different cases which have previously been considered to exhibit either amplifier or oscillator behavior: the overlap region of interacting beams lies entirely inside the crystal (amplifier), and the overlap region partially overlaps the input boundaries (oscillator). By defining an oscillator as a device whose output remains high if it is above threshold when its input seeding is cut off, we demonstrate the behavior of the DPCM for both types of interaction regions. The results follow the theoretical qualitative predictions quite well. We find that the fanning associated with wide beams tends to increase the interaction length and so make the DPCM more oscillator like than a simple geometrical argument would show. Up until the end of 1994, we had been using Connection Machines for these calculations. Now, however, we are running code just as quickly using Fortran-

90 on Tufts University's DEC Alpha farm.

The fanning beams usually split up into narrow filaments. In an attempt to investigate the origin of this effect, we generalized the photorefractive model for the beam propagation method to include the effects of secondary trap levels in the photorefractive band conduction model. This model was first introduced to explain the sublinear intensity dependence of the photorefractive effect observed in some experimental set-ups. We performed numerical simulations of beam propagation in photorefractive crystals using this model for the space charge field solution. We found that the intensity distribution of beam fanning is shifted to higher angles compared to the intensity distributions of fanning obtained using the single trap model.

O. Ferroelectric and photorefractive waveguide devices

We have been collaborating with NZ Applied Technologies in the development of electro-optic thin film modulator and photorefractive devices. The company has expertise in chemical vapor deposition of perovskite like materials such as strontium barium niobate and barium titanate, and one of our graduate students has been evaluating the films for waveguide properties using prism coupler measurements. At the moment, the optical losses of the film are so high that they are unsuitable for waveguide applications. Current devices being investigated use the films in transmission as electrooptic modulators. The high electro-optic coefficients of some of the materials, PLZT in particular, have enabled high contrast thin film modulators. Our student is now also involved in building a newly designed CVD system to improve the film uniformity and to reduce scattering losses.

P. Photorefractive surface waves

We have shown that photorefractive gain can confine optical energy to the surface of a photorefractive crystal. This increases the average intensity of the beams in the interaction region and should lead to devices such as self-pumped phase conjugate mirrors with fast response times.

Q. Birefringent phase matching

In some image processing and data storage applications, it is advantageous to be able to read out holograms with light whose polarization and/or frequency is different from that of the light used to write the holograms. For example, this offers optical isolation of the laser source in phase conjugation. In another application, different write and read frequencies can be used for nonvolatile holographic data storage. The hologram is written at a wavelength where the crystal is optically sensitive, and read out at a longer wavelength so the hologram will not be erased by the readout process.

We have developed a birefringent phase matching technique that increases the useable image spatial bandwidth for holographic systems operated this way.

R. Synchrotron x-ray topography of photorefractive crystals

We have been continuing to interpret our x-ray topographs of barium titanate and strontium barium niobate. These show the structure of ferroelectric domains. They also allow direct tests of photorefractive grating theories, and have required the inclusion of surface strain effects to adequately model the experimental results.

S. Insertion loss of DPCM

The double phase conjugate mirror shows promise for applications such as power combining and distortion tolerant laser cavity design. The main drawback has been that its insertion loss has been high; typical insertion losses were of the order of 70%. Beam propagation calculations show that the losses should be much smaller than that. We have been studying ways to reduce the insertion loss. One thought is that minimizing the path taken by the beams in the crystal will reduce absorption losses. However, absorption losses turned out to be not a significant issue in the devices we were using, the main losses seem to be from fanning and surface reflections. One of our students, Dan Ball, has discovered a way to feed back reflection losses into the phase conjugate beams by using total internal reflection. The loss is down to 25% now, with the DPCM operating stably.

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