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1. Coherent transients
2. Laser spectroscopy
3. Pressure-induced resonances
4. Laser-assisted collisions
5. Quantum jumps
6. Interaction of atoms with broad band width radiation
7. Exchange collision kernel
8. Laser cooling of neutral atoms

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FINAL REPORT (MFR)

Title: "Theoretical Studies Relating to the Interaction of Radiation with Matter"
Supported by: The U.S. Office of Naval Research
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The final report for Contract No. N00014–87–K–0303 is divided into eight subject areas as follows:

1. Coherent transients
2. Laser spectroscopy
3. Pressure–induced resonances
4. Laser–assisted collisions
5. Quantum jumps
6. Interaction of atoms with broad bandwidth radiation
7. Exchange collision kernel
8. Laser cooling of neutral atoms

A detailed description of the work carried out under this Contract can be found in the articles listed at the end of this report as well as in Annual Reports MAR1–MAR3 associated with this Contract. In this Final Report, the research areas are summarized briefly, with references to the appropriate articles. Advances achieved during the last four months of the Contract (following Annual Report MAR3) in each subject area are indicated.

1. Coherent Transients

A calculation of the transient response of a two–level atom to a strong pump and a weak probe field has been carried out.[1*] It is shown that there can be gain for the probe field even in the absence of a population inversion of the levels. An interpretation of the results in terms of dressed states was given. This work has direct implications for "lasing without inversion," a subject area that has received a great deal of recent attention both experimentally and theoretically.

A detailed analysis of an experiment by Yodh and coworkers [Phys. Rev. Lett. 53, 659 (1984)] was undertaken.[14,15] Their experiment can be considered as an extended pulse photon echo, in which the second excitation pulse in the sequence can have a duration that is comparable with the inverse relaxations rates of atomic decay. The modification of the signal resulting from velocity–changing collisions was calculated and the results were compared with experiment. Good agreement with experiment was achieved, although the physical interpretation given for the results differed somewhat from that of Yodh and coworkers. Related calculations are in progress to characterize the "breakdown" of the optical Bloch equations in solids that has been observed in free–induction decay, rotary echoes and delayed saturated absorption.

A discussion of the quantum Zeno effect and quantum Zeno paradox has been given.[31] The quantum Zeno effect (inhibition of stimulated transitions by measurements) has been demonstrated recently by Itano et. al. [Phys. Rev. A41, 2295 (1990)] in a three–level atomic system. The quantum Zeno paradox (the fact that continuous measurement of a particle in a bubble chamber does not affect its lifetime) has not yet been observed. We analyze the experiment of Itano et. al. and show that the results follow simply from the atomic dynamics, with no need to invoke the idea of wave function collapse following each "measurement." We also introduce an atomic analogue of the bubble chamber experiment to help resolve the quantum Zeno paradox.

2. Laser Spectroscopy

We have shown that it is possible to model sodium – rare gas collisions using hard sphere collision kernels and transport coefficients obtained from theoretical intermolecular

*Superscripts refer to the numbers in the list of publications found at the end of this report.

potential curves. Using essentially only one free parameter, we could fit nonlinear spectroscopic data on *both* fine structure transitions in sodium.[2] These results, in turn, were used to model the optical piston.

In collaboration with the group of Duncan Steel at the University of Michigan, we carried out joint experimental–theoretical studies of very narrow resonances that can occur in four–wave mixing line shapes. The resonances under investigation are those characterized by some effective ground state width, rather than the spontaneous decay rate of the excited state. It was shown that the nonconservation of magnetic–state orientation or alignment can lead to narrow resonances, even in systems where the total state population is conserved and there are no collisions.[4,6,9] As is discussed below, this result has important implications for the sub–Doppler laser cooling of neutral atoms. We also showed that velocity–changing collisions can "open" a two–level quantum system and lead to the existence of narrow resonances.[7] Experimental verification of the theory was achieved [J. Liu and D. G. Steel, Phys. Rev. A**38**, 4639 (1988)]. In addition, our calculations relating to pump–probe line shapes for arbitrary ratios of atom–field detuning to Doppler width has appeared.[5]

3. Pressure–Induced Resonances

In collaboration with G. Grynberg, we have published a series of papers in which the theory and interpretation of pressure–induced extra resonances (PIER) is developed. The collision–induced resonances under consideration are those that can arise in a number of nonlinear spectroscopic experiments such as pump–probe spectroscopy and four–wave mixing. Our first paper in this area[12] analyzed three–level atoms interacting with four incident laser fields. An atomic state population is monitored as a function of the difference in frequency of two of the applied fields. Even though a population is monitored, all the features encountered in studies of PIER using the collective emission of four–wave mixing are reproduced. The results could be interpreted simply using a dressed atom picture in which the dressed states contributing to the extra resonances are unpopulated in the absence of collisions owing to conservation of energy considerations.

In a second paper,[13] we used a semiclassical dressed atom approach to interpret the PIER that occur in pump–probe spectroscopy and four–wave mixing. Moreover, we derived a signal for pressure–induced resonances in fluorescence beats. In this approach, the dressing field is taken as the as the average of two incident fields with a time–dependent amplitude. The approach represents a relatively simple calculational tool in which the PIER can be traced to the existence of collision–induced modulated dressed–state populations. The calculation also clarifies the role played by atomic–state gratings produced by the incident fields; an interpretation in terms of atomic–state gratings may also prove useful in the analysis of sub–Doppler laser cooling.

In a third paper,[22] the semiclassical approach was extended to calculate PIER in (1) four–level atoms interacting with four incident fields, (2) three–level atoms (plus continuum) which are photoionized by four incident radiation fields and (3) three–level atoms in which the PIER is monitored by fluorescence. All of these examples could be interpreted in terms of the interference of different quantum pathways contributing to the same final state.

In a fourth paper,[16,20] a quantized–field dressed atom approach was used to analyze pressure–induced resonances in four–wave mixing. Three fields having frequencies Ω, $\Omega-\delta$, and Ω are incident on an ensemble of two–level atoms. This is by far the most difficult case to analyze since the atoms remain in the two–state subsystem after spontaneous emission. By defining dressed states in terms of operators, we were able to associate the pressure–induced resonances with collision–induced, modulated dressed state operators. Our approach reveals that the calculation is much more easily carried out in the Heisenberg representation than in the Schrodinger representation. This result generally applies to situations in which one measures operators that already involve an average over some of the system parameters (in this case the pure atomic operators imply an average over field variables). We are still exploring the differences between the Heisenberg and Schrodinger approaches as applied to a number of basic systems in quantum optics.

The interest in using the Schrodinger picture has been discussed further in a fifth paper,[29,30] in which pressure–induced resonances in four–wave mixing have been discussed using quantized radiation fields. Although the Heisenberg picture calculation is much simpler, the Schrodinger picture calculation reveals interesting correlations between the atomic and field variables that are hidden in the Heisenberg approach. The pressure–induced terms are seen to arise from interference effects between different atomic sites involving both collisions and spontaneous emission at those sites.

In all the above cases, we have shown how the pressure–induced resonances are always intimately linked to conservation of energy considerations. As such, we have achieved a consistent, physical picture of the pressure–induced resonances.

4. Laser Assisted Collisions

Earlier work on a three–state model of laser–assisted collisions was expanded and summarized.[3,8,10] In this model, the population of an intermediate state *during* a collision was not neglected as is commonly done in theories of laser–assisted collisions. This modified model accounted for a long–standing discrepancy between theory and experiment in the line wings of laser–assisted collisions. A calculation of light–induced collisional energy transfer (LICET) in the line wings, taking into account the effects of magnetic–state degeneracy, was also completed.[24] The final state magnetic polarization in the LICET reaction was calculated for a number of different level schemes and the results were compared with analogous results for the so–called optical collisions.

5. Quantum Jumps

We have shown that two identical "two–level" atoms separated by a distance R constitutes a system that is capable of exhibiting quantum jumps.[18,21] When the atoms are irradiated by an optical field having wavelength λ which is nearly resonant with the atomic transition, quantum jumps can occur if $R \ll \lambda$. The eigenstates of the two atoms consist of symmetrical and antisymmetrical components. If $R \ll \lambda$, the symmetrical component decays at rate 2Γ (Γ = decay rate of the excited state of an isolated atom), while the antisymmetrical state is very nearly metastable. Consequently, the antisymmetrical state can be used as a "shelving" level to produce quantum jumps. We have worked out the probability distributions for the dark and bright periods assuming both coherent and incoherent pumping of the atoms. Moreover, we calculated the photon statistics and frequency–resolved photon statistics of the spontaneously emitted light, using a method involving "frequency–resolved delay functions." It may be possible to observe such effects by implanting impurity atoms in a host crystal.

6. Interaction of Atoms with Broad Bandwidth Radiation

Broadband radiation can be used as a source of sub–picosecond time resolution. Rather than the pulse duration, it is the correlation time of the radiation source that determines the ultimate temporal resolution achievable in certain limits. Photon echo or stimulated photon echo experiments are used to exploit the inherent high temporal resolution of broadband optical noise. The theoretical analysis of this problem brings into play many profound problems, owing to the fact that the *same* light source interacts with the active medium at least twice, since one of the incident beams is split and sent into the medium as it time–delayed replica. Consequently, there is a memory of the initial pulse and the problem is highly non–Markovian. We have devoted a great deal of effort in trying to obtain solutions to this problem in the limit of intense fields. An effective–field method was developed[23] in which two of the time–delayed pulses are replaced by totally overlapping (but modified) pulses. This model has enabled us to gain additional physical insight into this very difficult problem.

The optical coherent transients that arise when a sample of two–level atoms is irradiated by a sequence of two or three broad–bandwidth pulses were studied theoretically.[17,23,26,31] The first two pulses are correlated with one another and can be strong enough to saturate the two–level atomic transition. Taking into account the effects of inhomogeneous and homogeneous broadening, we calculate the intensity of the transient signals, emitted in different directions, as a function of delay time. In particular, it is shown for strong excitation pulses, that the strongest signals exhibit a peak having a width given by the cross–correlation time of the pulses, τ_c^{12}. The preliminary experimental results obtained at Laboratoire Aime Cotton (France) by the group of J.–L. LeGouet confirm our theoretical conclusions. In the case of two–pulse transient theory, the peak is found to disappear when the Doppler width of the atomic ensemble becomes sufficiently large, in qualitative agreement with experiment [R. Beach, D. deBeer, and S. Hartmann, Phys. Rev. A**32**, 3467 (1985)]

The optical coherent transients induced in a sample of three–level atoms by time–delayed fluctuating correlated pulses are also being considered.[33] We have seen that the three–level dynamics leads to a signal which, as a function of the delay time, depends dramatically on the intensities of the excitation pulses. For strong pulses, the signal may vary significantly on a time scale much smaller than τ_c^{12}. This effect may permit one to obtain time resolution better than the cross–correlation time of the pulses. This work was carried out in collaboration with P. Tchenio of Laboratoire Aime Cotton. Experimental confirmation of the theoretical predictions has recently been reported by LeGouet's group.

7. Exchange Collision Kernel

A detailed description of the exchange collision kernel and its relation to quantities that can be measured in laser spectroscopic experiments has been given.[27] The exchange collision kernel is the probability density per unit time that an atom undergoing a collision with a "perturber" having velocity $\mathbf{v}_p$ will have a velocity $\mathbf{v}$ following a collision. In other words, the exchange kernel can be used to determine the way in which a velocity–selected ensemble of atoms can transfer memory of this velocity selectivity to an initially thermal ensemble of "target" atoms. The exchange kernel is complementary to the direct kernel which is the probability density per unit time that an atom has its velocity changed from $\mathbf{v}'$ to $\mathbf{v}$ as a result of collisions with (thermal) perturbers. The exchange kernel is calculated for a hard–sphere interaction and a phenomenological exchange kernel, similar to the Keilson–Storer direct kernel, is proposed. Two independent relationships between the exchange kernel and the collision integrals of transport theory are obtained. The theory is used to analyze an experiment in laser spectroscopy where, in effect, the exchange kernel was measured. The relationship between the exchange and direct kernels is discussed, incorporating our previous work on the direct kernel.[11]

8. Laser Cooling of Neutral Atoms

We have begun a systematic study of the relationship between nonlinear spectroscopic line shapes and laser cooling. The theory of nonlinear spectroscopy and laser cooling can be described by essentially the same formalism. We develop this formalism as applied to transition between two sets of electronic levels, each set of levels consisting of a number of fine and hyperfine levels. The magnetic sublevels of each of the hyperfine levels are also considered. It is shown[25,28] that the very narrow resonances (on the order of some effective ground state width) that can occur in nonlinear spectroscopy have a direct connection with laser cooling of atoms below the so–called Doppler limit. The existence of resonances are related to the fact that the certain physical parameters characterizing the atoms (e.g. magnetic state alignment and orientation) are not conserved. Our explanations are complementary to earlier discussions of sub–Doppler cooling based on field polarization gradients and light–induced atomic level shifts. The formalism, as developed, can be applied to arbitrary level schemes to analyze laser cooling in one, two and three dimensions. As a first example of the formalism, we obtained analytical expressions for the friction force of laser cooling in several cases of current experimental interest. Our work in this area is continuing. Recently, we have obtained some results for cooling in two–dimensions in which the friction force of laser cooling does not depend linearly on the velocity for small velocities. Our work also indicates that "standard" methods for calculating the atomic populations, valid for small velocities in one–dimension, need no longer retain their validity in two or three dimensions.

Publications

1. N. Lu and P. R. Berman, "Transient Probe Spectra in a Driven Two–Level Atom," Phys. Rev. A36, 3845–3861 (1987).
2. J. E. M. Haverkort, J.P. Woerdman, and P. R. Berman, "Experimental Determination of Na–Noble Gas Velocity–Changing and Fine Structure Changing Collision Kernels," Phys. Rev. A36, 5251–5264 (1987).
3. P. R. Berman, "Light –Induced Collisional Energy Transfer–Revisited," in **Atomic and Molecular Processes with Short Intense Laser Pulses,** edited by P. Bandrauk (Plenum, New York, 1988) pp. 75–85.
4. J. Liu, G. Khitrova, D. Steel, and P. R. Berman, "Narrow Resonances in Four–Wave Mixing Due to Radiative Decay," in **Laser Spectroscopy VIII,** edited by W. Persson and S. Svanberg (Springer–Verlag, Berlin, 1987) pp. 291–293.
5. G. Khitrova, P. R. Berman, and M. Sargent III, "Theory of Pump–Probe Spectroscopy," J. Opt. Soc. Amer. B5, 160–170 (1988).
6. P. R. Berman, D.G. Steel, G. Khitrova and J. Liu, "Effects of Radiative Decay in Four–Wave Mixing Spectroscopy – Narrow Resonances Produced by Nonconservation of Population, Alignment, and Orientation," Phys. Rev. A38, 252–262 (1988).
7. M. Gorlicki P. R. Berman, and G. Khitrova, "Theory of Resonances in Four–Wave Mixing Resulting from Velocity–Changing Collisions," Phys. Rev. A38, 4340–4350 (1988).
8. A. Agresti, P. R. Berman, A. Bambini and R. Stefanel, "Analysis of the Far Wing Behavior in the Spectrum of the LICET Process," Phys. Rev. A38, 2259–2273 (1988).
9. J. Liu, D.G. Steel, P. R. Berman, M. Gorlicki and G. Khitrova, "High Resolution Four–Wave Mixing Spectroscopy of Narrow Resonances in Doppler–Broadened Systems," in **Proceedings of XVI International Quantum Electronics Conference.**
10. P. R. Berman, "Three–State Model for Laser Assisted Collisions," in **Spectral Line Shapes,** Vol. 4, ed. by J. Szudy (Ossolineum, Warsaw, 1989) pp. 713–732.
11. P. R. Berman, "On the Connection Between Collision Kernels and Transport Coefficients," in **Proceedings of Workshop on Light–Induced Kinetic Effects in Gases** edited by L.J.F.Hermans (Univ. of Leiden, 1988) pp.1–8.
12. P. R. Berman and G. Grynberg, "Theory and interpretation of pressure–induced resonances," Phys. Rev. A39, 570–585 (1989).

13. G. Grynberg and P. R. Berman, "Pressure–induced effects in two–level atoms: New approach and simple physical interpretation," Phys. Rev. A39, 4016–4025 (1989).
14. R. Sung and P. R. Berman, "Collisional relaxation in an extended pulse photon echo: Weak field limit," Phys. Rev. A39, 6284–6297 (1989).
15. R. Sung and P. R. Berman, "Theory of optical coherent transients including collisional effects: Application to an extended–pulse photon echo," Phys. Rev. A39, 6298–6309 (1989).
16. P. R. Berman and G. Grynberg, "Dressed–atom approach to collision–induced resonances," in **Laser Spectoscopy IX**, edited by M. Feld, A. Mooradian and J. E. Thomas (Academic Press, New York, 1989) pp. 68–70.
17. V. Finkelstein and P. R. Berman, "Stimulated photon echo induced by broad bandwidth pulses," in **Coherence and Quantum Optics 6**, edited by J. H. Eberly, L. Mandel and E. Wolf.(Plenum, New York, 1989) pp. 305–308.
18. K. Yamada and P. R. Berman, "Macroscopic quantum jumps form a two–atom system," in **Coherence and Quantum Optics 6**, edited by J. H. Eberly, L. Mandel and E. Wolf (Plenum, New York, 1989) pp. 1243–1247.
19. V. Finkelstein, "Excitation into a quasicontinuum by a fluctuating laser field," in **Coherence and Quantum Optics 6**, edited by J. H. Eberly, L. Mandel and E. Wolf (Plenum, New York, 1989) pp. 301–304.
20. P. R. Berman and G. Grynberg, "Quantized field approach to pressure induced resonances," Phys. Rev. A40, 6921–6930 (1989).
21. K. Yamada and P. R. Berman, "Macroscopic quantum jumps from a two–atom system," Phys. Rev. A41, 453–462 (1990).
22. G. Grynberg and P. R. Berman, "Pressure induced extra resonances in nonlinear spectroscopy," Phys. Rev. A41, 2677–2686 (1990).
23. V. Finkelstein and P. R. Berman, "Optical coherent transients induced by time–delayed fluctuating pulses. I: three–pulse transients," Phys. Rev. A41, 6193–6224 (1990).
24. P. R. Berman, F. Schuller and G. Neinhuis "Generation of magnetic polarization in light induced collisional energy transfer," Phys. Rev. A42, 459–473 (1990).
25. P. R. Berman, "Nonlinear spectroscopy and laser cooling," Phys. Rev. A (to appear).
26. V. Finkelstein and P. R. Berman, "Optical transient signal induced by strong fluctuating pulses," Phys. Rev. Rapid Comm. A42, 3145–3148 (1990).
27. G. Rogers and P. R. Berman, "The exchange collision kernel," Phys. Rev. A (submitted).

28. P. R. Berman, "Narrow resonances in laser spectroscopy," in **Workshop on Light–Induced Kinetic Effects**, edited by E. Arimondo, C. Gabbanini, S. Gozzini, L. Moi, and F. Strumia, in press

29. P. R. Berman, "Pressure–induced resonances in multiwave mixing," in **Proceedings of Xth International Vavilov Conference on Nonlinear Optics**, in press.

30. G. Grynberg and P. R. Berman, "Quantized–field approach to parametric mixing and pressure–induced resonances: the Schrodinger picture," Phys. Rev. A, in press.

31. V. Finkelstein and P. R. Berman, "Optical coherent transients ind[illegible]ed by time–delayed fluctuating pulses," to be published in SPIE Proceedings (1990).

32. E. Block and P. R. Berman, "Quantum Zeno effect and quantum Zeno paradox in atomic physics," Phys. Rev. A, submitted.

33. V. Finkelstein, "Atomic response to optical fluctuating fields: temporal resolution better than the pulse correlation time," Phys. Rev. A, in press.