REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 0704-0188

...n is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, ...ting and reviewing the collection of information. Send comments regarding this burden estimate or any other aspect of this ...ing this burden to Washington Headquarters Services, Directorate for Information Operations and Reports, 1215 Jefferson ...nd to the Office of Management and Budget, Paperwork Reduction Project (0704-0188), Washington, DC 20503

1. AGENCY USE ONLY (Leave blank)	2. REPORT DATE	3. REPORT TYPE AND DATES COVERED
	20 May 1991	Final Report/1 Jan 88-31 Dec 90

4. TITLE AND SUBTITLE
Laser-Atom Interaction at High Intensities

5. FUNDING NUMBERS
61102F/2301/A4

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8. PERFORMING ORGANIZATION REPORT NUMBER
AFOSR-TR- 91 0593

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)
AFOSR/NP
Bolling AFB DC 20332-6448

10. SPONSORING/MONITORING AGENCY REPORT NUMBER

11. SUPPLEMENTARY NOTES

12a. DISTRIBUTION/AVAILABILITY STATEMENT
Approved for public release; distribution is unlimited.

12b. DISTRIBUTION CODE

13. ABSTRACT *(Maximum 200 words)*

A general program has been developed to calculate multiphoton processes in atomic hydrogen to arbitrarily high order in perturbation theory. This program can be used to calculate quantities in intense field multiphoton processes, such as the generalized cross section of multiphoton absorption, the a. c. Stark effect (shift and the broadening of the level due to the field interaction), and the parameters in nonlinear optics such as the nonlinear index of refraction and the nonlinear susceptibility. Effective parametrization of high-order susceptibilities has been achieved over a wide range of frequencies, utilizing concepts from quantum defect theory to express the results of lengthy calculations in a compact form that also permits extrapolation across thresholds and resonance poles. Significant progress has been made in direct numerical solution of the time-dependent Schroedinger equation. In addition, contributions have been made in: understanding the role of spatial dimensionality in the solution of model systems (e.g., the one-dimensional delta-function potential); the development of a general R-matrix code for perturbative computations of multiphoton processes in many-electron atoms; and multiphoton ionization with two commensurate laser frequencies.

14. SUBJECT TERMS

15. NUMBER OF PAGES
05

16. PRICE CODE

17. SECURITY CLASSIFICATION OF REPORT	18. SECURITY CLASSIFICATION OF THIS PAGE	19. SECURITY CLASSIFICATION OF ABSTRACT	20. LIMITATION OF ABSTRACT
UNCLASSIFIED	UNCLASSIFIED	UNCLASSIFIED	UL SAR

Final Report to the

Air Force Office of Scientific Research

for Grant No. AFOSR-880098

LASER-ATOM INTERACTION AT HIGH INTENSITIES

1 Jan 88 - 31 Dec 90 Period of Performance

Principal Investigators:

T. J. McIlrath
C. W. Clark

During this period our program for evaluating high-order perturbation theory was essentially completed, and significant progress was made in direct numerical solution of the time-dependent Schrödinger equation. In addition, contributions were made in: understanding the role of spatial dimensionality in the solution of model systems (e.g. the one-dimensional delta-function potential); the development of a general R-matrix code for perturbative computations of multiphoton processes in many-electron atoms; and multiphoton ionization with two commensurate laser frequencies.

The main development in the work on high-order perturbation theory involved the extension of our Sturmian function approach to treat final states in the photoionization continuum. This was done via a complex-coordinate-rotation technique, which gives us a perturbative expansion for the complex energies of a non-Hermitean Hamiltonian in powers of the external field strength. In this picture, all atomic states become resonances in the presence of the field; the imaginary part of the complex energy of a resonance is inversely proportional to its lifetime. The coefficients in the expansion are independent of the complex-coordinate rotation angle, which is a significant advantage over previous methods. Using this approach, we are able to compute the energy shifts associated with above-threshold ionization (ATI) channels, in which photons are absorbed by a continuum electron. We have found that the onset of ATI can either *increase* or *decrease* the total ionization rate, in a manner which depends in a complicated fashion upon the radiation frequency. This is differs from the results of the model of Deng and Eberly, in which ATI always acts to decrease the total ionization rate. Comparisons made with independent, non-perturbative calculations by Chu and Cooper showed that corrections to lowest-order perturbation theory can be significant in intensity regimes where perturbation theory is effectively convergent, so the utility of this approach is not limited to cases in which lowest-order perturbation theory is dominant. However, the range of validity of perturbation theory, as determined by the convergence properties inferred from numerical calculations, is much lower than has been expected: for hydrogen in a Nd:YAG field the highest intensity at which perturbation theory converges is of the order 3×10^{11} W cm^{-2}. The nonlinear susceptibilities that describe high-harmonic generation were also calculated by this method. The qualitative behavior of the high-order susceptibilities changes when the highest virtual state lies in the continuum vs. the discrete spectrum: the critical intensity I_c, at which perturbation theory breaks down, decreases with increasing order when ionization is energetically forbidden, but exhibits an increase as the ionization threshold is crossed. These results have been published in the publications numbered 4-6 below, and have been presented in invited (1,2,4) and contributed (4) talks at conferences.

Perturbation theory was also applied to the excitation of hydrogen by radiation at two frequencies, ω and 2ω, with a fixed phase relationship. This is a problem that has recently become of experimental interest. Our results show that the ionization rate depends upon the phase, in a manner that is determined by the continuum phase shifts of the electronic wavefunction. This is a completely different mechanism than the enhanced tunneling that is predicted in classical treatments of the problem. This work was reported in a contributed talk (2), but has not yet been submitted for publication.

Work continued on a collaborative project with K. T. Taylor and M. Smith at the University of London, in which a general R-matrix code is being developed for the computation of multiphoton processes in many-electron atoms within the framework of lowest-order perturbation theory. This code is nearly complete, and the work done in this reporting period consisted of making extensive comparisons of its results for hydrogen (e.g. seven photon ionization) with those obtained by the Sturmian function method. Relatively good agreement (at the 10% level) has been obtained, except near intermediate resonances, whose energies are slightly different in the two methods. This work was reported in a contributed talk (3), and is being written up by M. Smith as his Ph.D. thesis.

The frequency-dependent polarizability of an electron bound by a short range potential was investigated in a general framework, which allowed the spatial dimensionality of the system to be treated as a continuously variable parameter. This was motivated by the frequent use of delta-function potentials for modelling the response of atoms to strong radiation fields. It was found that a delta-function potential taken to have the same binding energy as the hydrogen atom has a polarizability much lower than that of the atom, even in cases far from resonance. The polarizability exhibits a monotonic decrease with increasing spatial dimensionality. Thus short-range potentials will tend to underestimate the response of a real atom on the rising edge of a laser pulse. This work was published (1) and presented in a contributed talk (1).

Numerical integration of the time-dependent Schrödinger equation for hydrogen in a radiation field began in earnest, building upon preliminary work done late in 1989. The main advance involved the adoption of massively parallel processing techniques on a Connection Machine computer. This approach divides configuration space into many (up to several thousand) distinct regions, each of which is handled by a dedicated central processing unit (CPU) with a small amount of local memory. Each CPU integrates the equations of motion within its own region, using the contents of its own local memory and that of its nearest neighbors. Thus the time taken to solve the equations is roughly independent of the number of regions, if a sufficient number of processors are available. We have integrated the equations of motion for hydrogen in radiation fields of various frequencies and intensities (up to 10^{15} W cm^{-2}), over about ten optical cycles. Machine performance of several gigaflops has been attained, which is still somewhat below the theoretical limit for the configuration we are using (the CM-2 facility at the Northeast Center for Parallel Architectures, Syracuse University). We find the plateau phenomenon in the harmonic radiation spectrum, and get field-induced shifts of above-threshold ionization peaks that are in good agreement with recent experimental results from the University of Bielefeld. Work has been started on the dynamics of two-electron atoms in strong radiation fields. Our presentation of this work, which is still in a developmental phase, is limited to a single invited talk (7). A significant amount of analytical work has been done in support of the numerical effort. This has led to a comprehensive understanding of the propagation of a free wavepacket on a discrete mesh, and to a closed-form solution of the bound s states of hydrogen in the discretized representation. This has been presented in an invited talk (3), and is being written up for publication.

The personnel employed on this project, and their sources of support, were as follows:

T. J. McIlrath, co-principal investigator	1 month (AFOSR)
C. W. Clark, co-principal investigator	3 months (NIST)
L. Pan, research associate	12 months (AFOSR)
J. Parker, research associate	6 months (NIST)
S. Blodgett-Ford, graduate student	6 months (NIST)

The following publications and conference presentations were attributed to AFOSR support during this period:

Publications

1. Clark, C.W., "Frequency-Dependent Polarizability of an Electron Bound by a Zero-Range Potential," J. Opt. Soc. Am. B **7**, 488 (1990).

2. Clark, C.W., Pan, L., and Taylor, K.T., "High-order Harmonic Generation by Hydrogenic Ions," in Advances in Laser Science-IV, ed. by J.L. Gole, D.F. Heller, M. Lapp, and W.C. Stwally (AIP, New York, 1990) p. 504.

3. Pan, L., "Treatment of Continuum-Continuum Coupling in the Theoretical Study of Above-Threshold Ionization," in Atoms in Strong Fields, ed. by C. Nicolaides, C.W. Clark and M. Nayfeh (Plenum Publishing, New York 1990) p. 447.

4. Pan, L., Taylor, K.T., and Clark, C.W., "Perturbation Theory Study of High Harmonic Generation," J. Opt. Soc. Am. **B 7**, 509 (1990).

5. Pan, L., Taylor, K., and Clark, C.W., "Perturbative Calculation of the AC Stark Effect by the Complex Coordinate Method," Phys. Rev. A (in press).

6. Pan, L., Taylor, K., and Clark, C.W., "Convergence of Rayleigh-Schrödinger Perturbation Theory in Calculations of Multiphoton Processes," Radiation Effects and Defects in Solids (in press).

Invited Talks

1. Clark, C.W., "Progress Towards the Generation of Coherent XUV Radiation," Winter College on High Resolution Spectroscopy, International Center for Theoretical Physics, Trieste, Italy, January 23, 1990.

2. Clark, C.W., "High-Order Perturbation Theory of Atoms in Strong Radiation Fields," International Conference on Coherent Radiation Processes in Strong Fields, Catholic University of America, Washington, DC, June 19, 1990.

3. Clark, C.W., "The Solution of the Schrödinger Equation on a Discrete Lattice," Surface Lunch Bunch Meeting, NIST, Gaithersburg, MD, November 5, 1990.

4. Pan, L., "Theoretical Study of Intense-Field Laser-Atom Interactions: Progress and Outstanding Issues," Institute for Physical Sciences and Technology, University of Maryland, College Park, MD, September 12, 1990.

5. Pan, L., "Theoretical Study of Intense-Field Laser-Atom Interactions: Progress of a Dressed-State Approach," Naval Research Laboratory, Washington, DC, September 26, 1990.

6. Parker, J.S., "ATI in Atomic Hydrogen - Role of Coherent Population Trapping," International Conference on Coherent Radiation Processes in Strong Fields, Catholic University of America, Washington, DC, June 19, 1990.

7. Parker, J.S., "Crunching Schrödinger's Equation on the Connection Machine," Laser Lunch Bunch Meeting, NIST, Gaithersburg, MD, November 21, 1990.

Contributed Talks

1. Clark, C.W., "The Frequency-Dependent Susceptibility of an Electron Bound by a Zero-Range Potential," Spring Meeting of the American Physical Society, Washington, DC, April 18, 1990.

2. Clark, C.W., "Interference of Multiphoton Ionization Final States in a Coherent Two-Color Radiation Field," Division of Atomic, Molecular and Optical Physics Meeting, American Physical Society, Monterey, CA, May 23, 1990.

3. Clark, C.W., "R-Matrix Method for Multiphoton Processes: Application to the H Atom," Division of Atomic, Molecular, and Optical Physics Meeting, American Physical Society, Monterey, CA, May 22, 1990.

4. Pan, L., "Convergence of Rayleigh-Schrödinger Perturbation Theory in the Calculation of Multiphoton Processes," 1990 Spring Meeting of the American Physical Society, Washington, DC, April 18, 1990.