AD-A055 076 IBM THOMAS J WATSON RESEARCH CENTER YORKTOWN HEIGHTS N Y F/G 7/4
NONLINEAR SPECTROSCOPY.(U)
APR 78 J J WYNNE DAAG29-75-C-0016

UNCLASSIFIED ARO-13013.16-P NL

1 OF 1
AD A055076

END
DATE FILMED
7-78
DDC

ARO 13013.16-P



# NONLINEAR SPECTROSCOPY

Final Report for the Period March 1, 1975 - February 28, 1978

James J. Wynne

April 28, 1978

U. S. Army Research Office

Contract DAAG29-75-C-0016

IBM Thomas J. Watson Research Center

P. O. Box 218

Yorktown Heights, New York 10598

Approved for Public Release; Distribution Unlimited

**FINDINGS IN THIS REPORT ARE NOT TO BE CONSTRUED AS AN OFFICIAL DEPARTMENT OF THE ARMY POSITION UNLESS SO DESIGNATED BY OTHER AUTHORIZED DOCUMENTS.**

Unclassified

SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

## REPORT DOCUMENTATION PAGE

READ INSTRUCTIONS BEFORE COMPLETING FORM

| | |
|---|---|
| 1. REPORT NUMBER (18) ARO (19) 13013.16-P | 2. GOVT ACCESSION NO. |
| 3. RECIPIENT'S CATALOG NUMBER | |
| 4. TITLE (and Subtitle) (4) Nonlinear Spectroscopy. | 5. TYPE OF REPORT & PERIOD COVERED: Final 3/1/75-2/28/78 |
| | 6. PERFORMING ORG. REPORT NUMBER |
| 7. AUTHOR(s) (10) James J. Wynne | 8. CONTRACT OR GRANT NUMBER(s) (15) DAAG29-75-C-0016 |
| (9) Final rept. 1 Mar 75-28 Feb 78. | |
| 9. PERFORMING ORGANIZATION NAME AND ADDRESS: IBM Thomas J. Watson Research Center, P. O. Box 218, Yorktown Heights, New York 10598 | 10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS (11) 28 Apr 78 |
| 11. CONTROLLING OFFICE NAME AND ADDRESS: U. S. Army Research Office, P. O. Box 12211, Research Triangle Park, NC 27709 (12) 8p. | 12. REPORT DATE: April 28, 1978 |
| | 13. NUMBER OF PAGES: 6 pages |
| 14. MONITORING AGENCY NAME & ADDRESS(if different from Controlling Office) | 15. SECURITY CLASS. (of this report): Unclassified |
| | 15a. DECLASSIFICATION/DOWNGRADING SCHEDULE |

16. DISTRIBUTION STATEMENT (of this Report)

Approved for public release;
Distribution unlimited.

17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)

18. SUPPLEMENTARY NOTES

Findings in this report are not to be construed as an official Department of the Army position unless so designated by other authorized documents.

19. KEY WORDS (Continue on reverse side if necessary and identify by block number)

Spectroscopy
Nonlinear Optics
Laser
Infrared Generation
Ultraviolet Generation
Atomic Spectroscopy

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

Using the technique of multiphoton ionization spectroscopy, we have studied Rydberg series of states in the alkaline earth atoms Ca, Sr and Ba. ~~We observe~~ long series of states not previously identified. ~~We~~ analyze ~~this new data~~ using multichannel quantum defect theory. were observed and

DD FORM 1 JAN 73 1473

349250

Unclassified

SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

CL

## Background and Summary of Results

This contract began on March 1, 1975 under the title, "Generation and Detection of Infrared and Ultraviolet Radiation by Nonlinear Optical Techniques". At that time we had shown that nonlinear optical mixing of dye laser beams in atomic vapors was a way to generate tunable, coherent, infrared and ultraviolet radiation. The efficiency of these processes was strongly influenced by resonant enhancement when the lasers were tuned near various atomic energy levels. Under this contract, we began to investigate ways to improve the efficiency of these processes. We saw resonant enhancement from known levels and we also saw resonant enhancement where no levels were previously known. Thus we had found a new way of doing atomic spectroscopy. Subsequently, the discovery and analysis of new energy levels in alkaline earth atoms became our primary interest and the contract title was changed to "Nonlinear Spectroscopy".

During the time we were supported by this contract, the primary problem studied was multiphoton ionization spectroscopy of the alkaline earth atoms Ca, Sr and Ba.

The study of the bound states of atoms by optical spectroscopy has been fundamental in the development of the understanding of atomic structure. However, despite a long and continuing history of achievements, many aspects of the electronic structure of many-electron atoms are still not well understood. The states of many-electron atoms do not, in general, fall into simple Rydberg sequences. The combined effects of Coulomb repulsion and angular momentum recoupling have made it difficult to identify simple Rydberg series of states in the optical spectra of many-electron atoms. Absorption spectroscopy has been extremely successful in observing and identifying Rydberg series because of the well-defined nature of the initial state. In contrast, picking out Rydberg series from emission spectra is a formidable task when the series are complicated by configuration interactions and neither the initial nor final state of the transitions are known. Consequently, emission spectroscopy has had limited success in identifying Rydberg series in many-electron atoms.

We have overcome these shortcomings of emission spectroscopy by using tunable lasers to excite levels in a well defined manner from the ground state. By multiple photon excitation, levels which are not connected to the ground state by one-photon transitions may be selectively excited. For example, with an even-parity ground state, even-parity excited states may be reached by two-photon transitions, but not by one-photon transitions. We have studied the excited states of alkaline earths by observing strong ionization signals when an atomic vapor is irradiated by lasers tuned to multiple photon resonances. This ionization is easily detected by inserting a simple ionization probe into a pipe containing the atomic vapor. If the probe is negatively biased relative to the walls of the pipe, thermionic emission from the probe will lead to a space-charge limited current. Ions produced by the laser excitation partially neutralize the space charge, thereby allowing an increased electron current to flow. Using pulsed lasers, one detects ionization by observing the pulsed current flowing through the external load resistor.

Using nitrogen-laser-pumped dye lasers, we have studied Rydberg series of states in Ca, Sr and Ba. Our experiments allowed us to observe long series of both odd and even parity states which had not been previously identified. Our important experimental findings are now summarized. Using two-photon excitation we have observed the msns $^1S_0$ and msnd $^1D_2$ and $^3D_2$ series in Ca (m=4) and Sr (m=5). Using three-photon excitation we have observed the msnp $^3P^o$ series in Ca (m=4), Sr (m=5) and Ba (m=6). We have also seen autoionizing levels of the type 3dns and 3dnd in Ca using two-photon excitation. Finally the Zeeman effect of the 5snd levels of Sr has been studied in the region where $^1D_2$ and $^3D_2$ states are strongly mixed.

The analysis of all of this data has been done using multichannel quantum defect theory (MQDT). Within its range of applicability, MQDT is essentially an exact parametrization of the energy levels and wavefunctions of interacting Rydberg series. It treats each whole series as a unit, known as a channel, and is often superior to conventional analysis on a level by level basis. MQDT has enabled us to interpret our data in terms of strong configuration interactions

with perturbing levels. For example, the 4snd $^1D_2$ series in Ca is strongly affected by the $4p^2$, $3d^2$ and 3d5s configurations. This interaction has prevented previous workers from identifying the levels belonging to $^1D_2$ configurations from emission spectroscopy. A triumph of our MQDT analysis was the prediction of the Landé g-factor for Sr 5snd states.

Details of our accomplishments may be found in the list of publications which follows.

## List of Publications

1. Tunable Generation by Optical Mixing in Atomic Vapors, J. J. Wynne and P. P. Sorokin, book chapter in Topics in Applied Physics, Vol. 16, Nonlinear Infrared Generation, ed. by Y. R. Shen (Springer-Verlag, Berlin, 1977) p. 159.

2. Multiphoton Ionization Spectroscopy of High-Lying, Even Parity States in Calcium, P. Esherick, J. A. Armstrong, R. W. Dreyfus and J. J. Wynne, Physical Review Letters 36, 1296 (1976).

3. Bound, Even-Parity J=0 and 2 Spectra of Ca: A Multichannel Quantum Defect Theory Analysis, J. A. Armstrong, P. Esherick and J. J. Wynne, Physical Review A 15, 180 (1977).

4. Bound, Even-Parity J=0 and J=2 Spectra of Sr, P. Esherick, Physical Review A 15, 1920 (1977).

5. Spectroscopy of $^3P^o$ States of Alkaline Earths, P. Esherick, J. J. Wynne and J. A. Armstrong, Optics Letters 1, 19 (1977).

6. High Rydberg States in Alkaline Earth Atoms - I: Multiphoton Spectroscopy, P. Esherick and J. J. Wynne, Comments on Atomic and Molecular Physics VII, 43 (1977).

7. Comments on the Spectra of Ca, Sr and Ba Near their Ionization Limits, J. A. Armstrong, P. Esherick and J. J. Wynne, in Proceedings of C.N.R.S. International Colloquium Highly Excited States of Atoms and Molecules Coupled to the Continuum, Aussois, France, 1977, p. 15.

8. Multiphoton Ionization Spectroscopy of the Alkaline Earths, P. Esherick, J. J. Wynne and J. A. Armstrong in Laser Spectroscopy, ed. by J. L. Hall and J. L. Carlsten,

Springer Series in Optical Sciences, Vol. 7, (Springer-Verlag, New York, Heidelberg, 1977), p. 170.

9. Multiphoton Ionization Spectroscopy of Alkaline Earth Atoms, P. Esherick, J. J. Wynne and J. A. Armstrong, Proceedings of SPIE 21st Annual Technical Symposium, Laser Spectroscopy I, Vol. 113, 121 (1977).

10. Zeeman Effect of J=2 States of Sr: g-Factor Variation for Interacting Rydberg Series, J. J. Wynne, J. A. Armstrong and P. Esherick, Physical Review Letters 39, 1520 (1977).

11. Coherent VUV Generation by Nonlinear Processes, J. J. Wynne, J. de Physique, Proceedings of VUV5, Montpellier, France, 1978 (to be published).

12. Multiphoton Spectroscopy of the Alkaline Earths, J. J. Wynne, J. A. Armstrong and P. Esherick in Multiphoton Processes, ed. by J. H. Eberly and P. Lambropoulos (John Wiley and Sons, New York, to be published).

13. VUV Spectroscopy, J. J. Wynne in Quantum Electronics, ed. by C. L. Tang (Academic Press, to be published).

## Participating Scientific Personnel

Dr. James J. Wynne

Dr. Peter Esherick

Dr. John A. Armstrong

Dr. Jean-Paul Hermann

Mr. Louis H. Manganaro