AD-A111 462 ARGONNE NATIONAL LAB IL F/G 7/4
HIGH-RESOLUTION SPECTROSCOPY AND DYNAMICS OF MULTIPHOTON PROCES--ETC(U)
FEB 82 P M DEHMER, J L DEHMER N00014-81-F-0051
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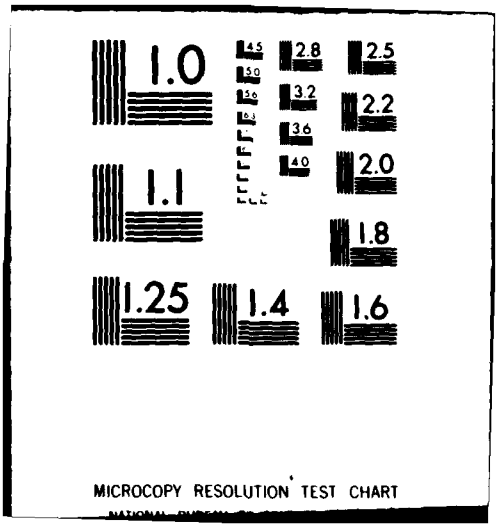
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MICROCOPY RESOLUTION TEST CHART

Unclassified

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Multiphoton ionization, multiphoton dynamics, multiphoton absorption, high-resolution spectroscopy, atoms and molecules, laser-induced fluorescence, optical-optical double resonance spectroscopy, electron spectroscopy, mass spectrometry, fluorescence spectroscopy, dye lasers, Nd:YAG laser, nonlinear frequency conversion.

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

This report summarizes research to establish the high-resolution spectroscopy and detailed mechanisms of sequences of resonant transitions induced in atoms and molecules by multicolor, multiphoton excitation. These processes are studied as a function of the independently tunable frequencies and polarization states of multiple dye laser beams in order to establish both the underlying physics and the high degree of selectivity of this advanced form of laser excitation. Detection methods include photoelectron analysis, mass

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20. (Continued)

spectrometry, and laser-induced fluorescence. The initial phase of work covered in this report included design, construction, and testing of all major components of the experimental apparatus needed to perform these studies. In addition, measurements of photoelectrons ejected from rare gas atoms by nonresonant, multiphoton excitation are described.

ANNUAL SUMMARY REPORT

High-Resolution Spectroscopy and Dynamics of Multiphoton Processes in Atoms and Molecules (Contract No. N00014-81-F-0051)

Submitted to

Office of Naval Research
Physical Sciences Division
Physics Program Office
Department of the Navy
Arlington, VA 22217

ATTN: Dr. Bobby R. Junker

Submitted by

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2. Contract Description

This contract covers research to establish the high-resolution spectroscopy and detailed mechanisms of sequences of resonant transitions induced in atoms and molecules by multicolor, multiphoton excitation. These processes are studied as a function of the independently tunable frequencies and polarization states of multiple dye laser beams in order to establish both the underlying physics and the high degree of selectivity of this advanced form of laser excitation.

3. Scientific Problem

This study is aimed at both determining the underlying physics and developing the high degree of selectivity of multicolor, multiphoton processes in atoms and molecules. This is a frontier research area which involves roughly three major challenges: First, it is necessary to establish the spectroscopy of excited states of molecules at vastly improved resolution. This results from the narrow line width of tunable dye lasers, typically 0.02-0.10 cm^{-1} for pulsed sources. This means that, in molecules, intermediate states of resonant excitation sequences will correspond to particular excited rotational-vibrational-electronic states of the molecule. Therefore, to specify a workable sequence of resonant transitions, it is necessary to know the excited state spectroscopy at this level. Second, it is necessary to establish the mechanisms and dynamical parameters governing multiphoton processes. For example, in order to design the most selective and sensitive excitation scheme, one must learn the cross sections for successive steps, rates and modes of decay of intermediate states, and cross sections for all competing excitation sequences, including non-resonant ones. This requisite basis for selective excitation is largely unknown at this time and requires a concerted basic research effort for

3. Scientific Problem (Continued)

adequate development. Third, this work involves harnessing the rapid technological advances in lasers, optics, and electronics to produce a composite laser probe which can stimulate a preselected sequence of resonant steps within the ten nanosecond duration of the laser pulse.

4. Scientific and Technical Approach

This program utilizes a complementary set of experimental techniques to address the problems described above. The total experimental facility can be summarized in terms of its six major components: (1) A Nd:YAG oscillator/amplifier is used as the pump laser for all the dye lasers used to produce the composite laser beam. (2) We currently have three dye laser oscillator/amplifiers to produce three laser beams with independently tunable frequencies and polarization states. One laser is a commercial laser outfitted with nonlinear conversion (frequency doubling and summing) capability for producing ultraviolet frequencies. Two other dye lasers were manufactured to produce narrow line width visible beams for probing transitions among upper states. (3) A time-of-flight mass spectrometer is used to measure the ionic products of multiphoton ionization. (4) A high-resolution hemispherical electron spectrometer is used to analyze the kinetic energies of the electrons produced by multiphoton ionization. (5) A fluorescence spectrometer is used to monitor fluorescence from laser excited states throughout the excitation chain. (6) The multiple control and detection channels are centrally controlled by a microprocessor.

Summarizing, we are able to probe atoms and molecules with a fully flexible composite laser probe and measure most major excitation products - ions, electrons, and fluorescence. The remaining major product, namely neutral fragments, could also be monitored by further ionization or by laser-induced fluorescence, but this is not in our initial plans. Spectroscopic and dynamical information is then obtained by monitoring these detection channels as a function of the frequencies and polarization states of the input composite laser probe.

5. Progress

During this initial contract period, all of the major components described in the last section were put into operation: The commercial Nd:YAG pump laser and dye laser, including frequency doubling capability, were put into full operation. The two custom made dye lasers were designed, constructed, and tested. The three detection channels were adapted from previous uses for operation in conjunction with the laser probes. Also, roughly half of the computer automation is complete. In addition the Nd:YAG second harmonic beam (532nm) was used in a preliminary experiment (see next section) to study the angular distribution of electrons ejected from rare gas atoms by high-order nonresonant multiphoton excitation. Finally, we have specified and acquired the necessary optics to perform the

5. Progress (Continued)

initial two-color (optical-optical double resonance) resonance ionization experiment on a molecular system. We have chosen CO as the initial system to study and will excite the $X^1\Sigma \rightarrow A^1\Pi$ transition with a UV beam, followed by transitions to higher excited states (such as the $B\ ^1\Sigma^+$ or $E\ ^1\Pi$ states) using visible beams. Total ion production, photoelectrons, and fluorescence will be monitored using the general strategy described in item 4, above.

6. Publications

J. L. Dehmer, E. D. Poliakoff, and P. M. Dehmer
PHOTOELECTRON ANGULAR DISTRIBUTIONS FROM MULTIPHOTON IONIZATION. SEVEN PHOTON IONIZATION OF Kr AT 532nm.
Bull. Am. Phys. Soc. 26, 1322 (1981).

7. Extenuating Circumstances

Appointment of Dr. Stephen T. Pratt (Ph.D., Yale, 1982) as a postdoctoral appointee on this project occured on February 1, 1982, so that full staffing was not achieved until that time.

8. Unspent Funds

Due to the delay in making a postdoctoral appointment, $10K will remain unspent at the end of the current contract period. It is requested that this amount be carried over to the next contract period.

9. Graduate Students Receiving Degrees

None

10. Other Federal Contract Support

J. L. Dehmer is a co-principal investigator on Office of Naval Research Contract N00014-82-F-0011, "Triply Differential Studies of Atomic and Molecular Photoionization Using Synchrotron Radiation," 10/1/81 - 9/30/82.