MASTER**THEORETICAL STUDIES OF HIGHLY IONIZED SPECIES****Progress Report**

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Abstract

The relativistic random phase approximation has been used to study discrete outer and inner shell transitions of the neon isoelectronic sequence, and to calculate photoionization cross sections of elements of the magnesium and zinc isoelectronic sequences. Model potentials have been constructed for the copper and zinc isoelectronic sequences. Molecular processes have been investigated and the possibility of an enhanced radiative recombination rate for molecular ions has been demonstrated. Detailed calculations of molecular resonance states have been completed for H_2 and the experimental data on ion velocity distributions produced in the photoionization of H_2 have been interpreted. Charge transfer processes involving multiply-charged ions at thermal energies have been examined and a preliminary selection has been made of those systems for which charge transfer is probably rapid. The mechanisms for the radiative destruction of the lithium excimer have been explored; it appears that destruction results preferentially by an accidental predissociation.

In order to interpret recent experiments on multiphoton absorption by molecular sodium and the photoionization of Na_2^+ , calculations were carried out of the oscillator strengths of transitions in Na_2 .

1. Introduction

We have continued with theoretical studies of a diverse range of atomic and molecular processes that we believe to be of interest to the mission of the Basic Energy Sciences Division of the Office of Energy Research of the Department of Energy. A list of papers published in 1978 or in press for which the research was supported by the Department of Energy is given in the appendix.

2. Applications of the Relativistic

Random Phase Approximation

We have been applying the Relativistic Random Phase Approximation systematically to the elucidation of relativistic effects on atomic structure and to the prediction of energy levels and transition probabilities. Of special interest has been the search for ionized systems having simple spectra for which the oscillator strength is concentrated in a small number of transitions with large excitation rates. We have studied most recently the neon sequence (Shorer 1978). Whereas for neutral neon the transitions to the ground state from the lowest lying excited levels have small oscillator strengths our calculations show that as the nuclear charge increases the distribution changes in important ways and strong individual transitions occur.

The calculations on the neon sequence also reveal the existence of anomalous behavior as a function of

nuclear charge which can be interpreted as a consequence of level crossings. However it demonstrates that extrapolation along an isoelectronic sequence, a widely used procedure, can be seriously misleading in specific cases.

We have extended the applications of the Relativistic Random Phase Approximation to photoionization phenomena. Preliminary studies have been made of the photoionization of elements of the magnesium and zinc isoelectronic sequences and values of the background cross sections have been obtained. Our treatment is general enough to include the inner shell resonances but the numerical procedures become slowly convergent when the energy under study falls within the resonance width. Some modifications will be necessary.

3. Model potential methods

We have obtained model potentials for several members of the copper isoelectronic sequence and the programs for the calculation of energy levels and oscillator strengths are at hand.

We have developed procedures for incorporating the effects of spin-orbit interactions so that the model potential method can be used to predict the probabilities of transitions in which a change of spin multiplicity occurs. We have used the model potential method to calculate transition probabilities for allowed and forbidden transitions along the beryllium sequence as far as NeVII⁺ (Laughlin, Constantinides and Victor 1978). The results agree well with those of the

Relativistic Random Phase Approximation (Lin and Johnson 1977). For higher nuclear charges, the values given by the Relativistic Random Phase Approximation can therefore be regarded as reliable.

Work was continued on a refined analysis of the experiments of Lucartoto and McIlrath (1976) and McIlrath and Lucartoto (1977) who obtained high ionization efficiencies by the application of long-pulse-length laser emission in sodium and lithium. It appears that sodium and lithium dimers may play a role in addition to two-photon ionization (Laughlin 1978) and laser-induced ionizing collisions (Geltman 1977). A manuscript is in preparation.

Studies were continued in collaboration with Dr. Shih-I Chu and Dr. W. Reinhardt of the possibility of combining model potential methods with a complex coordinate rotation transformation and an L^2 -discretization to investigate the effects of intense electric fields. Initial consideration was given to the related and important problem of atomic behavior in strong electric and magnetic fields (Chu 1978).

4. Resonating states

Our studies of resonating states in atomic systems were confined to an attempt to employ the Relativistic Random Phase Approximation. Numerical difficulties are severe but we have succeeded in locating the positions of

the most important states for a selected number of the elements of the magnesium and zinc sequences. The Relativistic Random Phase Approximation does in itself take proper account of the inner shell resonances and of the contribution to dielectronic recombination but different numerical procedures will have to be used.

5. Processes in low temperature plasmas

In collaboration with Professor A. Herzenberg of Yale University, we have explored the possibility that radiative recombination of molecular ions

may be enhanced by capture into resonating states produced by the excited rotational and vibrational states of the electronic Rydberg states. Such states are the source of the indirect mode of dissociative recombination described by Bardsley (1968).

We carried out model calculations that were then directed specifically to H_2^+ for which we could use experimental data on autoionization (Dehmer and Chupka 1977). We found that for H_2^+ the background radiative rate coefficient at low temperatures is doubled by the participation of the resonating states. The model suggests that for more complex species substantial enhancements can occur so that

polyatomic species may be very stable to ionizing radiation.

During the study, we calculated the properties of resonating states associated with the repulsive state of H_2^+ . Experimental data on the production of H^+ ions in the photoionization of H_2 show an effective threshold lying below that for direct absorption into the repulsive state of H_2^+ which must be attributed to repulsive Rydberg states (cf. Browning and Friar 1973, Strathdee and Browning 1978).

Our predictions are in close agreement with the experimental data of Browning and Friar (1973) and of Strathdee and Browning (1978) on both the resonance contribution to photoionization and the velocity distribution of the H^+ ions by postulating absorption into the lowest $(2p\sigma_u, 2s\sigma_g)^1\Sigma_u^+$ resonance state. There is an element of chance about the quantitative agreement because our treatment of the nuclear motion is not adequate. A more careful analysis is in progress.

6. Charge transfer

An elementary treatment of charge transfer in near resonance collisions was developed by Bottcher (1978). Most of our attention was given to thermal collisions between multiply-charged ionic systems and neutral hydrogen and helium. Analyses were presented (Dalgarno and Butler 1978, Dalgarno 1978) and a preliminary list was compiled of those

charge transfer reactions that might be rapid at thermal energies. Of particular importance to a number of astrophysical objects, including planetary nebulae, diffuse HII regions, quasars and Seyfert galaxies is the reaction

In collaboration with Dr. P. Bender we have calculated the quartet molecular potential energy curves and it appears the reaction may in fact be rapid, as indeed Pequignot et al. (1978) had concluded from an analysis of spectral emissions from the planetary NGC 7027.

A comprehensive study of similar processes that may be important in high temperature plasmas is in progress.

7. Laser processes

A study of the interaction of photons with the lithium and sodium excimers was continued. It was established that lithium is dissociated by a process of accidental predissociation and the dependence upon incident photon wavelength was investigated. There are interesting differences between different isotopes. A paper is in preparation.

Similar calculations were carried out for the sodium dimer. They acquired a special interest because of the laboratory experiments of Nitz, Hogan, Scheerer and Smith (1978) who studied the resonant two-photon ionization of

Na_2 and the subsequent third-photon dissociation of Na_2^+ using a high powered pulsed dye laser. Our theoretical study is not yet complete.

Percentage of time devoted to this project: A. Dalgarno,
Principal Investigator: 15%; G. A. Victor, Co-Investigator: 25%.

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Appendix

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