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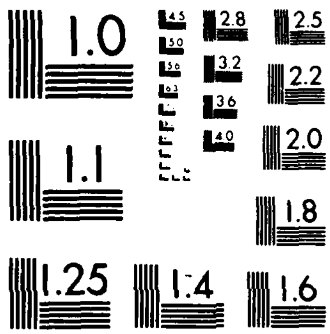

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Department of Physics

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Chemical Physics Institute

Relativistic Calculations and Measurements of Energies, Auger Rates, and Lifetimes

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This project is devoted to the study of the structure and dynamics of deep hole states in atoms and of few-electron systems. During the reporting period, work has been completed on the gauge dependence of atomic inner-shell transition rates, M x-ray emission rates in Dirac-Fock approximation, Auger and radiative deexcitation in P^{4+} ions, inelastic x-ray scattering cross sections of neon, on two book-length monographs on inner-shell processes, and on two reviews of atomic physics with synchrotron radiation. Threshold excitation phenomena in deep inner shells are being explored through gas-phase electron spectrometry with synchrotron radiation, and investigations of the resonant Auger Raman effect and post-collision interaction are being pursued.

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1. Introduction

This project is devoted to the study of the structure and dynamics of deep hole states in atoms and of few-electron ions. In such systems, relativistic and quantum-electrodynamic effects are strong, and the distinction between excitation and deexcitation becomes blurred, leading to threshold resonance phenomena and post-collision interaction effects. These phenomena have only recently become amenable to study, due to the advent of large computers and of tunable hard synchrotron radiation. The present project is focussed on deep inner-shell excitation experiments conducted with gas-phase electron spectrometry in the Stanford Synchrotron Radiation Laboratory, complemented by large-scale computational efforts utilizing facilities in the NASA Ames Research Center as well as modest computer resources on the University of Oregon campus. Current efforts are directed toward (a) continued investigation of the Auger resonant Raman effect that occurs in inner-shell threshold excitation, (b) post-collision interaction following photoionization of extremely short-lived hole states, (c) double photexcitation in which one incident x-ray quantum promotes two electrons, and (d) effects of atomic relaxation and configuration interaction on the lifetimes of deep hole states.

A-1

2. Status of the Research

a) Gauge dependence of atomic inner-shell transition rates

In relativistic calculations of radiative atomic transition probabilities on the basis of self-consistent-field (SCF) Dirac-Fock wave functions, the results depend on the gauge because of the nonlocal potential introduced in the Dirac-Fock effective Hamiltonian. All existing relativistic SCF calculations of x-ray emission probabilities have, however, been carried out in the Coulomb or length gauge. Similarly, all relativistic calculations of radiationless (Auger or Coster-Kronig) transition probabilities have heretofore been based on the Møller two-electron operator, which is in the Lorentz gauge. The question of gauge dependence of relativistic radiationless transitions computed with SCF wave functions has never been explored heretofore.

Because the calculated lifetimes of inner-shell vacancies depend directly on the radiative and (mostly) radiationless transition probabilities that fill these vacancies, and x-ray emission probabilities are given by the former, it is of interest to examine whether the choice of gauge makes a significant difference in the theoretical rates and hence introduces a perhaps unsuspected uncertainty in the calculated results. We have therefore calculated relativistic radiative and radiationless atomic inner-shell transition rates with Dirac-Fock wave functions, using matrix elements that correspond to different gauges. Radiative rates computed in the length gauge were found to be larger than Coulomb-gauge results; for K-shell radiative widths, the difference is approximately 20% at Z=10 and falls to 4% at Z=30; the difference for L_2 radiative widths is a factor of 2 at Z=18 and approximately 7% at Z=48. Especially large discrepancies between length- and Coulomb gauge-results were found for radiative transitions between levels with the same principal quantum number. Auger transition rates calculated in the Lorentz and Coulomb gauges agree to better than 1% in all cases that we tested; it is inferred that in first-order perturbation theory the Auger rate is practically gauge-invariant. [See Sec. 3, Items 1, 7, 9].

b) M x-ray emission rates in Dirac-Fock approximation

Atomic inner-shell vacancies can be filled through either of two major types of decay processes---radiative or radiationless. Measurements of the spectra of resultant x rays or Auger electrons can provide much information on atomic structure and dynamics, given the requisite theory for their interpretation. Numerous calculations of atomic radiative transiton rates have been performed, including recent computations that include the effects of relativity and relaxation. Most of the existing calculations, however, deal with x-ray emission rates associated with the filling of K- and L-shell vacancies. Only one calculation had heretofore been performed on M-shell x-ray emission rates, and this was based on Hartree-Slater wave functions and included only 6 elements. We have consequently undertaken a calculation of radiative transition rates to M-shell vacancies, in both the Coulomb and length gauges, for 10 elements with atomic numbers between Z=48 and Z=92. The computations are relativistic, based on Dirac-Fock wave functions. As previously found for K and L shells, length-gauge results exceed rates computed in the Coulomb gauge, by approximately 34% at Z=48, with the discrepancy decreasing to 4% at Z=92 for the M_1 shell, and by 44% at Z=48 to 3.5% at Z=92 for the M_3 shell. Dirac-Fock rates calculated in the

Coulomb gauge agree well with Dirac-Hartree-Slater results, to approximately 7% throughout. Contributions from higher multipoles are found to be small, the largest being 1.5% electric quadrupole for 3s and 3p and 0.07% electric quadrupole for 3d initial vacancies. [Sec. 3 Item 8].

c) Auger and radiative deexcitation of P^{4+} ions

The properties of highly stripped, excited atoms are very interesting from both theoretical and applied points of view. Intermediate coupling and configuration interaction must be incorporated in successful calculations, and measurements of transition energies and rates can constitute delicate tests of theory. Excited ions play important roles in certain laboratory plasma systems and in astrophysical processes, and metastable ionic states have been considered in connection with the design of short-wavelength lasers. A part of the Ph.D. thesis of Kh R. Karim in our group consists of calculations of x-ray wavelengths, Auger energies, and decay rates for various states of the P^{4+} ion, with configurations that have been studied through beam-foil spectroscopy. Intermediate coupling and configuratin interaction were taken into account. The energies and decay rates were found to be strongly affected by configuration interaction. The theoretical results have been compared with observations in ion-atom collision experiments. Good agreement with measured spectra is found, and the calculations characterize a number of lines that had not perviously been identified. [Sec. 3, Item 4].

d) Effects of exchange, electron correlation, and relaxation on the $L_1-L_{2,3}M_1$ Coster-Kronig spectrum of argon

A long-standing problem in atomic theory concerns the fact that the observed lifetimes of 2s (and 3s) hole states of atoms throughout the periodic table are substantially longer than predicted by the most elaborate of calculations; the discrepancy can reach a factor of 2 or 3. For several years now we have examined this puzzling problem, looking into the possibility that the discrepancies might be produced by relativistic effects, many-body interactions, or lack of orthogonality between initial- and final-state wave functions. An elaborate study of the problem has been completed by Kh R. Karim as part of his doctoral thesis. The effects of exchange, electron correlation, and relaxation on the L_1-$L_{2,3}M_1$ Coster-Kronig spectrum of argon were examined. We have extensively investigated the effects of final-state relaxation and of exchange between the continuum electron and bound electrons in the final state, and we have included some of the many-body interactions through initial- and final-state interaction. The conclusion drawn from these very detailed and exhaustive calculations is that substantial effects arise from relaxation and from the exchange between the Coster-Kronig electron and bound-state electrons. While these effects cannot be omitted from a successful calculation, they do not adequately account for the physical situation. Clearly, the independent-particle model of atomic structure is insufficient to treat the pronounced many-body aspects of radiationless transitions of the type we have considered, and more elaborate approaches are required to explain the observed rates. [Sec. 3, Item 5].

e) Inelastic x-ray scattering cross sections of neon

X-ray scattering constitutes an important method for the exploration of atomic and molecular form factors, electronic charge and momentum distributions. Scattering cross sections are particularly sensitive to correlation effects that generally lie beyond the reach of calculations with atomic wave functions based on independent-particle models. Experimental possibilities in the field of x-ray scattering from atoms and molecules have been vastly enhanced with the advent of intense, tunable synchrotron radiation sources. A study utilizing these new possibilities was conducted by F. Parente within the framework of his doctoral thesis research in our group; the work has now been published. Inelastic x-ray differential scattering cross sections of Ne were computed to compare the effect of three nonrelativistic approximations: Waller-Hartree, impulse, and excited-state. The same Hartree-Slater wave functions were employed throughout, and momentum transfer between $K/4\pi=0.1$ and 1.2 $Å^{-1}$ were included. For momentum transfer below 0.5 $Å^{-1}$ the Waller-Hartree and excited-state results were found to agree within 4%, whereas the impulse-approximation results are up to 300% higher. The Compton profiles of the individual atomic shells were computed in the excited-state approximation for three values of incident x-ray energy and are found to vary in height and shape with energy and scattering angle, in contrast with the constant profiles predicted by the impulse approximation. Results of theoretical ratios of x-ray inelastic and elastic scattering cross sections were compared with data from an experiment that we performed with synchrotron radiation at SSRL. Waller-Hartree and excited-state results agree fairly well with experiment. The work points toward the importance of more extensive and elaborate x-ray scattering experiments with tunable synchrotron radiation; we plan to explore these possibilities in the future. [Sec. 3, Item 3].

f) Inner-shell threshold excitation with synchrotron radiation

The production of deep-lying hole states in atoms and the deexcitation of such states under emission of x rays or Auger electrons has traditionally been considered a two-step process: the decay is separated from the excitation phase, and the photon and electron emission are treated as independent processes by means of first-order perturbation theory. An important advance in the fundamental understanding of atomic transitions will be brought about by a description that unifies the formation and decay of autoionizing and inner-shell vacancy states. Substantial inroads have already been made in the difficult problem of creating a unified theory of atomic excitation and deexcitation. Only quite limited experimental information exists, however, against which the unified theories can be tested---even to the extent that specific quantitative predictions can be derived from them. This is particularly true with regard to deep inner-shell transitions. Yet inner-shell hole states offer particular promise for the experimental exploration of the breakdown of the two-step model of atomic excitation/deexcitation: this breakdown becomes most pronounced when the states are excited near threshold, and the large widths of inner-shell vacancies allows one to probe the threshold phenomena in great detail, especially if excitation is accomplished with highly monochromatized synchrotron radiation that can be tuned precisely across the hole-state level energy. A further advantage of experiments with single inner-shell hole states is that the systems are usually well-described by independent-particle (self-consistent-field) atomic models, whence the interpretation of the results is greatly simplified.

We are engaged in a continuing investigation of the resonant Auger Raman effect and post-collision interaction. The Auger resonant Raman effect, which we were the first to observe, represents the epitome of the breakdown of the two-step model: excitation and deexcitation are represented by a single matrix element. The effect can be thought of as the radiationless, inelastic analog of resonance fluorescence. At resonance, the Auger line narrows below the natural lifetime width, and it exhibits linear dispersion with incident x-ray energy.

Above threshold, post-collision interaction (PCI) links the atomic excitation and deexcitation processes by feeding energy from the receding photoelectron to the Auger electron. As the incident-x-ray energy is lowered so as to approach the ionization threshold, the PCI blends smoothly into the Auger resonant Raman effect. In the other limit, if the incident-x-ray energy lies far above threshold, PCI vanishes and the two-step approximation becomes good.

As described in detail in our preceding Semi-Annual Technical Report, we have conducted measurements of the resonant Auger Raman effect and PCI of various Auger and Coster-Kronig lines in rare gases, following photoionization with hard synchrotron radiation. For narrow hole states, the PCI Auger-electron shifts are close to the predictions of semi-classical theory; the

"Niehaus" theory), but for wide hole states, such as Xe L_2, we observe a PCI shift that substantially exceeds the semiclassical prediction (by almost a factor of two). In the priod covered by this Report, we have performed calculations that remove some of the drastic approximations in the "Niehaus" theory, viz., the classical treatment of the receding photoelectron. Large-scale computations on the NASA ARC facilities have been initiated to carry out a completely quantum-mechanical calculation of PCI shifts. Unfortunately, AFOSR-provided computing funds expired at the end of the last fiscal year, in September 1983, and new funds have not yet been transferred as of this writing. We are extremely eager to complete these calculations to see if the more sophisticated theory reproduces the larger shifts which we observe. We also plan additional measurements of PCI shifts in Coster-Kronig lines arising from wide hole states, to gain additional evidence on the magnitude of the shifts. [For a general survey of synchrotron-radiation studies of atomic inner-shell physics, see Sec. 3, Items 6 and 10].

3. Publications

Published Papers

1. M. H. Chen and B. Crasemann: "Gauge Dependence of Atomic Inner-Shell Transition Rates from Dirac-Fock Wave Functions," Phys. Rev. A 28, 2829 (1983).

2. B. Crasemann: X-82 "International Conference on X-Ray and Atomic Inner-Shell Physics." Comments At. Mol. Phys. 13, 253 (1983).

3. Fernando Parente: "Inelastic X-Ray Scattering Cross Sections of Ne." J. Phys. B 16, 3487 (1983).

4. K. R. Karim, M. H. Chen, and B. Crasemann: "[illegible] Radiative Deexcitation of P^{4+} Ions." Phys. Rev. A [illegible], [illegible] (1983).

In Press and Submitted

5. K. R. Karim, M. H. Chen, and B. Crasemann: "Effects of Exchange, Electron Correlation, and Relaxation on the L_1-$L_{2,3}M_1$ Coster-Kronig Spectrum of Argon." Phys. Rev. A, in press.

6. B. Crasemann and F. Wuilleumier: "Atomic Physics with Synchrotron Radiation." Physics Today, in press.

7. B. Crasemann, M. H. Chen, and H. Mark: "Atomic Inner-Shell Transitions." J.O.S.A. B: Optical Physics, in press.

8. M. H. Chen and B. Crasemann: "M X-Ray Emission Rates in Dirac-Fock Approximation." Submitted to Phys. Rev. A.

9. M. H. Chen: "Relativistic Calculations of Atomic Transition Probabilities." A chapter in the book Atomic Inner-Shell Physics, edited by B. Crasemann (Plenum Publishing Company, New York, to be published).

10. B. Crasemann and F. Wuilleumier: "Atomic Physics Research with Synchrotron Radiation." A chapter in the book Atomic Inner-Shell Physics, edited by B. Crasemann (Plenum Publishing Company, New York, to be published).

Abstracts

11. B. Crasemann: "Atomic Inner-Shell Transitions." Abstract of Invited Paper, in Abstracts and Program, SAS-83 Symposium on

Atomic Spectroscopy, edited by John Conway, Lawrence Berkeley Laboratory Report No. LBL-16509, 1983 (unpublished), p. 17.

12. B. Crasemann: "High-Resolution Spectroscopy in Few-Electron Systems." Abstract of Invited Paper, Bull. Am. Phys. Soc., in press.

In Preparation

13. B. Crasemann, Editor: Atomic Inner-Shell Physics. Treatise to be published by Plenum, New York. (Manuscript expected to be complete in March, 1984).

14. B. Crasemann and G. E. Ice: Synchrotron Radiation---a Tool for Research. Monograph to be published by Academic Press, New York. (Expected completion date: summer 1984).

4. Professional Personnel

Bernd Crasemann, Professor of Physics, Principal Investigator

Mau Hsiung Chen, Research Associate Professor (to 1-3-1984)*

Kh Rezaul Karim, Research Assistant to 8-31-1983, Research Associate from 9-1-1983

Wolfgang Tepperwien, Research Associate (1-15-1984 to 1-31-1984)+

S. N. Tiwary, Research Associate and Visiting Assistant Professor (from 1-1-1984)**

G. Bradley Armen, Research Assistant

Jon C. Levin, Research Assistant

Mahendadasa Yakabadda Gamage, Research Assistant (from 9-1-1983)

Off-Campus Collaborators:

George S. Brown, Stanford Synchrotron Radiation Laboratory

Gene E. Ice, Oak Ridge National Laboratory

Hans Mark, National Aeronautics and Space Administration

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*Dr. Chen accepted a position as Physicist, Lawrence Livermore National Laboratory, as of 3 January 1984. He will continue to collaborate with our group at an appropriate level.

+Dr. Tepperwien was forced to resign on account of illness, almost immediately upon his arrival from Germany.

**Dr. Tiwary holds an 0.5-time appointment in our research group, 1 January - 30 June 1984.

5. Interactions

a) Papers Presented at Meetings, Conferences, etc.

1. B. Crasemann: "Atomic Inner-Shell Physics with Synchrotron Radiation." Colloquium lecture, SRI International, Menlo Park, California, June 1983.

2. B. Crasemann: "Atomic Inner-Shell Transitions." Invited review lecture, SAS-83 Symposium on Atomic Spectroscopy, Berkeley, 12-16 Septembr 1983.

3. B. Crasemann: "Investigations of Atomic Inner Shells with Synchrotron Radiation." Colloquium lecture, Physics Department, University of Wisconsin, Madison, 30 September 1983.

b) Consultative and Advisory Functions (B. Crasemann)

1. Associate Editor, Atomic Data and Nuclear Data Tables (Academic Press).

2. Proposal Review Panel, Stanford Synchrotron Radiation Laboratory.

3. Divisional Councillor and Member, Executive Committee, American Physical Society. Member of APS Publications Committee, Committee on Committees, Fellowship Committee.

4. Visiting Scholar, Physics Department, Stanford University, and Stanford Synchrotron Radiation Laboratory, 1 January - 30 June 1983.

probabilities on the basis of self-consistent-field (SCF) Dirac-Fock wave functions, the results depend on the gauge because of the nonlocal potential introduced in the Dirac-Fock effective Hamiltonian. All existing relativistic SCF calculations of x-ray emission probabilities have, however, been carried out in the Coulomb or length gauge. Similarly, all relativistic calculations of radiationless (Auger or Coster-Kronig) transition probabilities have heretofore been based on the Møller two-electron operator, which is in the Lorentz gauge. The question of gauge dependence of relativistic radiationless transitions computed with SCF wave functions has never been explored heretofore.

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