Technical Progress Report
on Contract No. AT(11-1)-1674
for the Period October 1, 1971 to September 30, 1972.

October 1, 1972

This year's activity has been marked by exploratory developments in new directions, while earlier work — chiefly applications of the frame transformation method — was being completed or continued at reduced strength. The number of students in the group is now at a long-time low and should be built up again.

1. Scope of the Report. This report covers the scientific activity of the following personnel:

- a) Principal Investigator: U. Fano, 100% of research activity.
 - b) Y.-K. Kim, Senior Research Associate, a guest from Radiological Physics Division of Argonne National Laboratory from October 1, 1971 to May 31, 1972.
 - c) Graduate Students:
 - T. Roth supported for 9 months.
 - C.M. Lee supported for 12 months.
 - C.D. Lin supported for 9 months by this project and in part (3 months) by NSF Contract NSF GH-33636-MRL-Light.
 - Dan Dill supported for 9 months by ARPA Contract DAHC-15-67-C-0220 Research and now by this project as Research Associate.
 - d) A.F. Starace and J.L. Dehmer, former students, continued work begun here at their present locations, at no cost to this project.
- K.T. Lu, also a former student, is now visiting and serving as a temporary consultant.

2. Two-electron Problem. The study of the joint motion of two excited electrons in a Coulomb field has not been pressed this year even though we still regard it as a main target. However a new approach to it has been discerned and we plan to follow it next year.

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

3. Channel Interaction and Spectroscopy. Applications of the frame transformation principle, and of the related Quantum Defect Theory, have continued this year, but with a view to termination of this program or to the development of selected offshoots. An invited survey of the whole program will be presented by Fano at the Annual Meeting of the APS-DEAP (COO-1674-68).

The work by Chang and Fano on the implications of Frame Transformation Theory for electron-molecule scattering has been completed and published (COO-1674-58). Detailed applications have not been carried out nor are planned at this time by our group.

Extensions of Lu's analysis of perturbed spectra have been continued at a slower pace. Starace's work on Ne, presented in abstract form last Fall, has been completed by him at his new location and accepted for publication (COO-1674-69). C.M. Lee's work on Ar and Kr has developed unexpected evidence which has delayed its completion; the matter is now being resolved under the impulse of a visit by K.T. Lu and we are looking to its early conclusion.

Rapid variations of cross sections for photoionization (and related phenomena) near higher thresholds had been surmised in earlier work as likely effects of channel interaction. They had been referred to as effects of "spectral repulsion" but may be also regarded as analogs of the Wigner "threshold cusps" which occur in collisions with neutral systems. C.D. Lin has demonstrated the existence of the phenomenon and its possible alternative manifestations through numerical calculation of typical examples. This work is being concluded and should be reported soon.

The next two paragraphs deal with offshoots of this program which have developed into new subprojects.

4. Theory of Angular Distributions of Reaction Products.

This theory has been developed long ago in a very general form for application to nuclear physics and involves elaborate procedures of Racah algebra. These procedures solve the geometrical, as contrasted to the dynamical, aspects of each problem and take different forms under different dynamical circumstances. Their purpose is to extract more accurate dynamical information from data on the distribution of reaction products. Their application to atomic problems is just beginning to expand in response to or anticipation of new experimental capabilities and presents new aspects related to the different dynamical characteristics of atomic and nuclear processes. In addition, the procedures of Racah algebra still prove clumsy in most cases and can profit by additional insight and streamlining.

This class of problems has been met in the course of completing Dan Dill's work on the angular distribution of photoelectrons from H_2 under conditions of rotational autoionization. This work itself has been published (COO-1674-61). It has given rise to three other papers by Dill and Fano (COO-1674-57, 66, and 70) which emphasize aspects of general significance. Dill has extended his H_2 work to calculate the angular distribution of photoelectrons from Xe in the Beutler autoionization range of the spectrum and in the near continuum. Future verification of his numerical predictions should provide a critical test on the internal consistency of our theoretical analyses. Publication of the results is waiting for an analysis of the sensitivity of the results to input parameters. Dill is also engaged in the analysis of the dependence of photoelectron distribution on various dynamical aspects of the process with a view to preparation of comprehensive reports.

The connection between the distributions of electrons ejected by light and by collision of fast charged particles has been worked out by Dr. Kim (COO-1674-62) as a part of his study of secondary electrons (see Sec. 8).

5. Theory of Close Coupling Eigenchannels. The quantum defect and frame transformation procedures developed by our group solve, in effect, the motion of an excited or ionized electron in the field of the residual ionic core. The input parameters for the solution consist of eigenphases and eigenvectors of the close coupling problem, defined on the outer surface of the core. These parameters have been chosen by us in the past semiempirically, to account for experimental data. We have now turned to the task of determining them by ab initio calculation.

This task consists of adapting the self consistent field methods to the treatment of an excited neutral atom bounded by a surface that encloses its ionic core tightly. Boundary conditions on this surface are designed to single out the close coupling eigenchannels rather than energy levels; eigenvalues of this problem are eigenphases at fixed energy. The conceptual and computational methods for this new approach have been under development by C.M. Lee and Fano throughout this year. Much has been done but it is still unclear how well or in what form the approach is going to work. Further implications of this approach are indicated below.

6. Barrier Effects in Molecules and Crystals. The 1971 Technical Report described the point of view developed by Dehmer, that inner shell photoabsorption in molecules of the SF_6 class reveals the existence of high (up to 20 eV) barriers to the escape of photoelectrons. Verification of this empirical point of view by calculation of excited orbitals was considered but not planned in the 1972 Proposal. In fact the opportunity arose to transfer the computational problem outlined by us to a group of quantum chemists in Modena, Italy. They solved it for the BF_3 molecule which is simpler than SF_6 and displays less spectacular but more varied photoabsorption spectra. Their results confirm substantially, in some detail, the predictions of our empirical analysis. However we have been unable thus far to identify whether the effective barrier derives from static or exchange potentials or from orthogonality of orbitals. A preliminary joint paper (COO-1674-63) was regarded as not suited to the Physical Review Letters and was then submitted to Chemical Physics Letters.

This success added emphasis to extension of our analysis to photoabsorption in crystals, whose spectra seem akin to those of SF_6 -class molecules. Fano has given considerable attention to this problem, beyond the plans of the 1972 Proposal, and has discussed it at conferences (COO-1674-59 and 60). The vacancy created in a crystal by inner-shell photoabsorption behaves as a fixed "deep impurity" center. The interpretation of the observed spectra implies then a successful solution of the still open problem of deep impurity theory, extended to include very high energies of excitation. Fano's approach to this problem regards the crystal atom with an inner shell vacancy as akin to the ionic core of free-atom excitations and regards the crystal lattice surrounding the impurity as akin to the free space around the core, where the Coulomb field prevails. The problem is then seen in both cases as that of matching the photoelectron's motion within and without the ionic core. The motion within the ionic core is to be treated as in Sec. 5 above, which gives added importance to the task outlined there. The motion in the surrounding lattice is to be described by a superposition of Bloch states, perturbed by the Coulomb field and adjusted to match boundary conditions characteristic of the impurity eigenchannels. The adaptation of Bloch state concepts to the requirements of matching at an impurity boundary constitutes a new task which Fano has been exploring.

7. Effective Interactions and RPA Method. The calculation of rare earth photoabsorption spectra (reported in 1971) brought out the necessity of scaling down the strength of d-f subshell interaction by a semiempirical factor of $\sim 2/3$ to achieve agreement with experiments. This factor represents presumably the collective influence of high order perturbations; an order of magnitude check by Starace indicated that it might be reproduced by an RPA calculation of the type developed by Amusia in Leningrad. This fact raised the priority for finding a consistent procedure by which the RPA methods can be embedded in the ordinary configuration-interaction procedures of theoretical spectroscopy.

Fano has devoted considerable effort to this task with the help of Starace himself, of C.D. Lin and of Professor L. Armstrong of Johns Hopkins. An approach has been devised which appears suitable but has not been brought to the stage of report writing.

8. Other Activities. In the Summer of 1970, Dehmer had applied the Phase-Amplitude-Method (PAM) of COO-1674-30 to investigate the empirical formula for the spin-orbit coupling strengths throughout the periodic system given by Fano and Martin in COO-1674-34. Dehmer has now completed his work providing a theoretical foundation not only for the formula itself but also for periodic deviations from it (COO-1674-64). Dr. Kim's work in our group centered on his ANL-based program of analyzing data on the spectra and direction of slow secondary electrons; lack of this information has been a major obstacle to the analysis of radiation effects on physical, chemical, and biological systems. A preliminary report on Kim's results is included in the ANL Annual Report and listed here as COO-1674-67. Remarks by Fano on the anisotropic aspects of oscillator strengths and derived quantities are being published as COO-1674-65.

- COO-1674-57 Angular Momentum in the Theory of Angular Distributions, U. Fano and Dan Dill, Phys. Rev. A 6, 185 (1972).
- COO-1674-58 Theory of Electron-Molecule Collisions by Frame Transformations, E. S. Chang and U. Fano, Phys. Rev. A 6, 173 (1972).
- COO-1674-59 Virtual Electronic Levels in Molecules and Crystals, U. Fano, Comments on Atomic and Molecular Physics, Vol. III, No. 3, 75 (1972).
- COO-1674-60 Two-Electron Processes, U. Fano, abstract for invited talk at International Conference on Inner Shell Ionization Phenomena, Atlanta, Ga., April 17-21, 1972. Interpreting Inner Shell Photoabsorption, U. Fano, abstract for Invited Talk at the "High Energy Excitations in Solids" Symposium (APS meeting) April 24-27, 1972. Bull. Am. Phys. Soc. 17, No. 4 436 (1972).
- COO-1674-61 Angular Distributions of Photoelectrons from H₂. Effects of Rotational Autoionization, Dan Dill, Phys. Rev. A 6, 160 (1972). (Ph.D. Thesis)
- COO-1674-62 Angular Distributions of Secondary Electrons in the Dipole Approximation, Y.-K. Kim, Phys. Rev. A 6, 666 (1972).
- COO-1674-63 Barrier to Electron Passage through Electronegative Atoms, B. Cadioli and U. Pincelli, Modena, Italy; E. Tosatti, Rome, Italy; and U. Fano and J. L. Dehmer, accepted for publication in Chem. Phys. Letters (1972).
- COO-1674-64 Phase-Amplitude Methods in Atomic Physics. II. Z dependence of Spin-Orbit Coupling, J. L. Dehmer, Phys. Rev., in press.

- COO-1674-65 Tensor Polarizability and Related Parameters, U. Fano and R. D. Deslattes, Comments on Atomic and Molecular Physics, Vol. No. (1972).
- COO-1674-66 Diagrammatic Theory of Angular Distribution, Dan Dill and U. Fano, Abstracts of papers of Third International Conference on Atomic Physics, August 7-11, 1972, Boulder, Colorado, p. 58.
- COO-1674-67 Report on the Energy and Angular Distributions of Secondary Electrons, Y.-K. Kim, Informal report for Argonne National Laboratory Annual Report (1972).
- COO-1674-68 Collisions and Spectra by the Quantum Defect Method, U. Fano, abstract for Invited Talk for the Fourth Annual Meeting of the Division of Electron and Atomic Physics, Stanford Research Institute, Menlo Park, Calif., November 29-December 1, 1972.
- COO-1674-69 (Replacing COO-1674-53)
Absolute Line Strengths by Analysis of Lu-Fano Plots with Application to Excited-State Transitions in Neon, A.F. Starace, J. Phys. B, in press.
- COO-1674-70 Parity Unfavoredness and the Distribution of Photo-fragments, Dan Dill and U. Fano, submitted to Phys. Rev.
- COO-1674-71 Technical Progress Report 1972.