Breakdown of the Independent Particle Approximation in High-Energy Photoionization

O. Hemmers[1], P. Glans[1], D. L. Hansen[1], H. Wang[1], S. B. Whitfield[1], D. W. Lindle[1], R. Wehlitz[2], J. C. Levin[2], I. A. Sellin[2], R. C. C. Perera[3], E. W. B. Dias[4], H. S. Chakraborty[4], P. C. Deshmukh[4], and Steven T. Manson[5]

[1]Department of Chemistry, University of Nevada, Las Vegas, NV 89154-4003
[2]Department of Physics and Astronomy, University of Tennessee, Knoxville, TN 37996-1200
[3]Advanced Light Source, Lawrence Berkeley Laboratory, Berkeley, CA 94720
[4]Department of Physics, Indian Institute of Technology - Madras, Madras 600036, INDIA
[5]Department of Physics and Astronomy, Georgia State University, Atlanta, Georgia 30303

INTRODUCTION

The response of physical systems to ionizing electromagnetic radiation, photoionization, is a basic process of nature. Because of the weak coupling between incident photons and target electrons, the electromagnetic radiation exerts only a small perturbation on the target, thereby allowing the unambiguous study of target electron properties, e.g., correlation and many-body aspects of electron dynamics. In addition, the photoionization process, along with associated spectroscopies including photoelectron spectroscopy, is of importance in a variety of applications [1] including structural determination in crystalline solids, astrophysical modeling, radiation physics, etc. Owing to its importance, the field has seen a recent upsurge of activity, particularly in the x-ray range, due to the development of third generation synchrotron radiation sources on the experimental side [2], along with the dramatic increase in computer power available, on the theoretical side.
In recent years, a wide variety of studies, both theoretical and experimental, have shown the importance of correlation in the form of interchannel coupling on the photoionization process in the region of the outer shell thresholds [3-10]; in some cases, the single particle viewpoint breaks down completely. An outstanding example is the threshold behavior of Xe *5s*, which is completely dominated by interchannel coupling with the *5p* and *4d* channels [5]. In addition, in the vicinity of inner shell thresholds, dramatic effects are seen in outer shell cross sections due to interchannel coupling. Examples of this phenomenon abound [7], e.g., effects on the outer shell cross sections of atomic Ba in the vicinity of the *4d* threshold [11].

It is generally thought, however, that in the x-ray range (far from the first ionization potential) away from inner shell ionization thresholds, the photoionization process can be well characterized in a single channel [3,7,12,13], or independent particle approximation, theory which omits correlation entirely. If this assertion is not true, then doubt is cast upon the interpretation of a number of studies of atoms, molecules and condensed matter involving x-ray photoabsorption.

Consider the photoionization of an *np* electron, inner or outer, from any atom, molecule or solid. Not far above the *np* ionization threshold will always be an *ns* threshold. Thus, a bit above the *np* threshold, there will always be an *ns* cross section degenerate with the *np* cross section. However, no matter what the relative values of these cross sections are near the thresholds, at energies far above threshold the *ns* cross section will *always* dominate the *np*. This is because, at high energy, the electric dipole photoionization cross section for an *np* subshell falls off with energy as $E^{-(7/2+\ell)}$ [3,7]. Thus,

$$M_{np\to kd(s)}(E) = D_{np\to kd(s)}(E) + \wp\int \frac{<\psi_{ns\to k'p} \mid H - H_o \mid \psi_{np\to kd(s)}>}{E-\varepsilon} D_{ns\to k'p}(\varepsilon)d\varepsilon \tag{1}$$

Because the energies of the photoelectrons from the *np* and *ns* channels are similar, the interaction matrix element falls off only very slowly and remains large with increasing energy, much like the Xe *5s* case. Thus, for both *np* $\rightarrow$ kd and *np* $\rightarrow$ ks, the second term in Eq.(1) becomes a larger and larger contribution to the matrix element, with increasing energy. This is in sharp contradistinction to the notion that the single-particle characteristics of the electric dipole photoionization process dominate at high energy.

As a prototypical example, we consider photoionization of atomic Ne in the 1 keV photon-energy range.

EXPERIMENT

The experiments were performed on undulator beamline 8.0, [17], which covers the 100-1500 eV photon-energy range. The monochromator entrance slit was set to 70 μm and the exit slit to 100 μm yielding very high flux, because high photon resolution was not needed. During the measurements the ALS operated at 1.9 GeV in two-bunch mode with a photon pulse every 328 ns. Four time-of-flight (TOF) electron analyzers, equipped with microchannel plates for electron detection, collect spectra simultaneously at different angles. The total electron flight paths are 460 mm, and the analyzers have a full cone acceptance angle of 5°. The interaction region is formed by an effusive gas jet intersecting the photon beam, which has a diameter of about 2 mm. Energy resolution of the TOF analyzers with a focus size of 2 mm is 3% of the electron kinetic energy. Each spectrum was collected for about 600 s.

RESULTS

New measurements have been made for the ratio of the Ne *2s* to the *2p* cross section which take into account the non-dipole contribution to the photoelectron angular distribution [16], and they are shown in Fig. 1, along with our theoretical results. New calculations also were performed within the framework of the relativistic-random-phase approximation (RRPA) [14,15] for the cross section, σ, and photoelectron angular distribution asymmetry parameter, β, of the *2p* subshell. Four levels of approximation were considered: (i) coupling of all of the relativistic single excitation channels arising from *2p*, *2s* and *1s*; (ii) from *2p* and *2s* only; (iii) from *2p* and *1s* only; (iv) from *2p* alone and *2s* alone. The measurements confirm the accuracy of the calculation by the excellence of the agreement.The most important result demonstrated by Fig. 1 is the divergence between the fully coupled and the uncoupled calculations at the highest energies, and the fact that it is the coupling with *2s* that is important as evidenced by the agreement between the full (*2p* + *2s* + *1s*) calculation and the *2p* + *2s* calculation. In addition, a central-field calculation [3,12,13] was performed using a Hartree-Slater potential [18] and the results (not shown) are virtually identical to the uncoupled *2p* RRPA result of Fig. 1, as expected. Thus, it is clear that the single-particle result does not agree with experiment at higher energies,while the coupled result does, in contrast to the conventional wisdom [3,7,12,13].

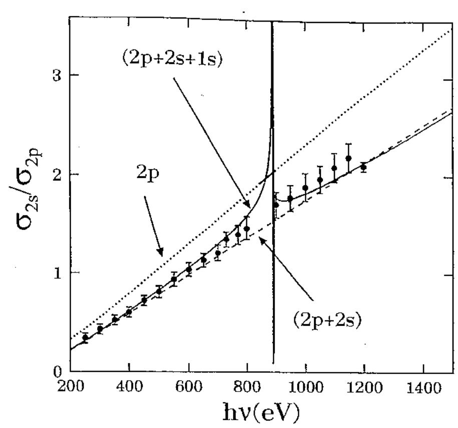

Figure 1. Ratio of the *2s* to *2p* cross section for Ne. The calculations employed the RRPA formalism with the single excitation channels arising from *2p*, *2s* and *1s* coupled (solid curve); *2p* and *2s* coupled (dash curve); and *2p* and *2s* *uncoupled* to each other (dot curve). The experimental points were measured in the manner discussed in Ref. 16.

Turning to the photoelectron angular distribution parameter, β, the experimental results [16], along with the various levels of calculated results, are shown in Fig. 2; all levels of calculation agree reasonably well at

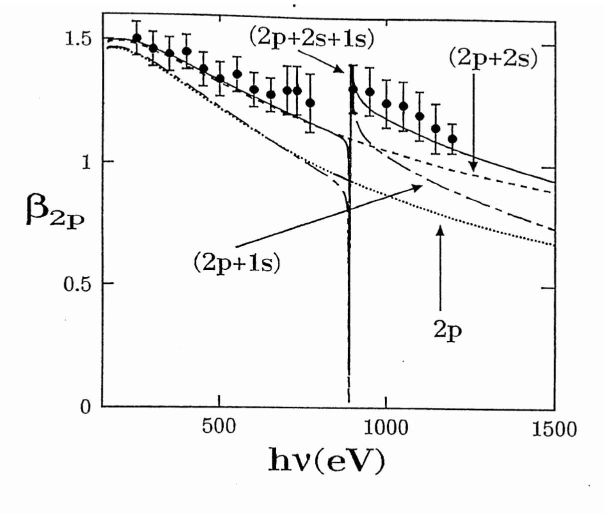

Figure 2. Photoelectron angular distribution asymmetry parameter, β, for Ne *2p* calculated using the RRPA formalism with the single excitation channels arising from *2p*, *2s* and *1s* coupled (solid curve); *2p* and *2s* (dash curve); *2p* and *1s* (dash-dot curve); and *2p* alone (dot curve). The experimental points are from Ref. 16 augmented by some new points reported here using the methodology of Ref. 16.

the lowest energies, but the separation into the same two groups occurs with increasing energy. Agreement of the experimental results with the full RRPA calculation is clear. Our single particle result for β (not shown) also is virtually indistinguishable from the *2p* alone calculation. At the highest energies considered, we see about a 30% shift in β from the single particle calculation, reiterating the point that even out at 1.5 keV, approximately 100 times the threshold energy, interchannel coupling does matter.

This interchannel coupling effect should also be in evidence for *nd* and *nf* subshells as well. In addition, although the detailed example was for an atom, the arguments are exactly the same for molecular and condensed matter targets. One *caveat* should be mentioned, however. At extremely high energies (tens of keV or higher), where relativistic interactions take over [19-21], the photoionization cross sections no longer behave as $E^{-(7/2+\ell)}$ and these arguments no longer apply. But for a very significant energy region below that, they do.

In conclusion, we have shown that the high energy photoionization of all $n\ell$ ($\ell > 0$) subshells will exhibit a breakdown of the independent particle approximation owing to the effect of interchannel coupling with the nearby *ns* channels, and this effect has been demonstrated for Ne *2p* employing both theory and experiment. It is predicted that the same effect applies equally to molecules and condensed matter, as well as atoms.

ACKNOWLEDGMENTS

This work was supported by the National Science Foundation, NASA, the Department of Energy, the Research Corporation and The Petroleum Research Fund. The authors thank the staff of the ALS for their support and the IBM, LBNL, LLNL, University of Tennessee and Tulane University collaboration for beam time at beamline 8.0. R.W. and O.H. thank for financial support by the AvH and DFG, respectively.

REFERENCES

1. H. S. W. Massey, E. W. McDaniel and B. Bederson, eds., *Applied Atomic Collision Physics* (Academic Press, NY, 1983), 5 volumes.
2. A. S. Schlachter and F. J. Wuilleumier, eds., *New Directions in Research with Third Generation Soft X-Ray Sources* (Kluwer, Dordrect, Netherlands, 1992), NATO ASI Series E, Vol. 254.
3. A. F. Starace, in *Handbuch der Physik*, Vol. 31, ed. by W. Mehlhorn (Springer-Verlag, Berlin, 1982), pp. 1-121.
4. J. A. R. Samson, in *Handbuch der Physik*, Vol. 31, ed. by W. Mehlhorn (Springer-Verlag, Berlin, 1982), pp. 123-213.
5. V. Schmidt, Rep. Prog. Phys. **55**, 1483 (1992).

6. B. Sonntag and P. Zimmermann, Rep. Prog. Phys. **55**, 911 (1992).
7. M. Ya. Amusia, *Atomic Photoeffect* (Plenum Press, NY, 1990).
8. T.-N. Chang, ed., *Many-Body Theory of Atomic Structure and Photoionization* (World Scientific, Singapore, 1993).
9. J. Berkowitz, *Photoabsorption, Photoionization, and Photoelectron Spectroscopy* (Academic Press, NY, 1979).
10. H. P. Kelly, in *X-Ray and Inner-Shell Processes*, ed. by T. A. Carlson, M. O. Krause and S. T. Manson (American Institute of Physics, NY, 1990), pp. 292-314.
11. J. M. Bizau, D. Cubaynes, P. Gerard and F. J. Wuilleumier, Phys. Rev. A **40**, 3002 (1989).
12. S. T. Manson and D. Dill, in *Electron Spectroscopy*, ed. by C. R. Brundle and A. D. Baker (Academic Press, NY, 1978), Vol. 2, pp. 157-195.
13. S. T. Manson, Adv. Electronics Electron Phys. **41**, 73 (1976).
14. W. R. Johnson and C. D. Lin, Phys. Rev. A **20**, 964 (1979).
15. W. R. Johnson, C. D. Lin, K. T. Cheng and C. M. Lee, Phys. Scripta **21**, 409 (1980).
16. O. Hemmers, G. Fisher, P. Glans, D. L. Hansen, H. Wang, S. B. Whitfield, D. W. Lindle, R. Wehlitz, J. C. Levin, I. A. Sellin, R. C. C. Perera, E. W. B. Dias, H. S. Chakraborty, P. C. Deshmukh and S. T. Manson, J. Phys. B **30**, L727 (1997).
17. R. C. C. Perera, Nucl. Instrum. Methods **A319**, 277 (1992).
18. F. Herman and S. Skillman, *Atomic Structure Calculations*, (Prentice-Hall, Englewood Cliffs, N.J., 1963).
19. R. H. Pratt, A. Ron and H. K. Tseng, Rev. Mod. Phys. **45**, 273 (1973).
20. I. P. Grant, J. Phys. B **7**, 1458 (1974).
21. A. Ron, I. B. Goldberg, J. Stein, S. T. Manson, R. H. Pratt and R. Y. Yin, Phys. Rev. A **50**, 1312 (1994).

Principal Investigator: Dennis W. Lindle, University of Nevada, Las Vegas. Email: lindle@nevada.edu.
Telephone: 702-895-4426.