Relativistic effects on giant resonances in electron-impact double ionization

M. S. Pindzola
Department of Physics, Auburn University, Auburn, Alabama 36849
(Received 22 December 1986)

The electron-impact double-ionization cross section for Fr^+ is calculated in the distorted-wave Born approximation. A giant resonance in the $5d$ subshell ionization-autoionization contribution to the cross section is found to be quite sensitive to changes in the double-well potential caused by relativistic effects on bound-state wave functions.

Double ionization is an important electron-impact ionization mechanism for heavy atomic ions. Recent studies[1-5] of electron ionization in the xenon isonuclear sequence have found that the ratio of double to single ionization can vary from 0.2 to 0.7 as a function of electron energy. The increased likelihood of double-ionization processes in not only xenon ions but also in many other heavy atomic ions has important consequences for the modeling of the time evolution of ion state populations in a plasma.[6]

Contributions to the electron-impact double-ionization cross section can be made by the following collision processes:

$$e^- + A^{q+} \rightarrow A^{(q+2)+} + e^- + e^- + e^- \quad (1)$$

and

$$e^- + A^{q+} \rightarrow (A^{(q+1)+})^* + e^- + e^- \rightarrow A^{(q+2)+} + e^- \, . \quad (2)$$

The first process is called direct double ionization and the second is called ionization-autoionization. The further processes of excitation—double-autoionization and resonant-recombination—triple-autoionization can also contribute to the double-ionization cross section. For most heavy atomic ions the largest contribution to the electron-impact double-ionization cross section is made by ionization-autoionization. If we assume the branching ratio for autoionization of low-charge state atomic ions is close to unity, predicting the double-ionization cross section reduces to a calculation of the relevant innershell ionization cross sections. Thus insight into the validity of single-ionization theory can be made by comparison with the growing number of crossed-beam double-ionization experiments.

For Fr^+ the electron-impact double-ionization cross section is dominated near threshold by contributions from the $5d$ innershell ionization-autoionization process. The $5d$ ionization cross section for Fr^+ is found to be strongly affected by a shape resonance in the $l=3$ scattered electron continuum. Previous calculations[7,8] of the $4d$ and $4f$ ionization cross sections for various heavy ions have found similar giant resonances. In the following paragraphs we will show that relativistic effects play a strong role in determining the strength of giant resonances for a very heavy atomic ion like Fr^+.

The electron-impact ionization cross section may be calculated in a distorted-wave Born approximation.[9] The average-configuration cross section is described by a triple partial-wave expansion of the first-order Born scattering matrix element. An integration is performed over the possible distribution of final-state energy between the scattered and ejected electrons. The maximum interference approximation of Peterkop[10] is used to describe exchange between the two final-state electrons. The bound-state orbitals needed to evaluate the Slater radial integrals found in the cross-section expression are calculated using a wave-function code developed by Cowan.[11] We used either the standard Hartree-Fock method (HF) or the Hartree-Fock method with relativistic modifications (HFR), which includes the mass-velocity and Darwin relativistic corrections within modified differential equations.[12] The continuum-state orbitals needed to evaluate the Slater integrals are obtained by solving the radial Schrödinger equation in the distorted-wave approximation. A local semiclassical potential[13] is used for the exchange interaction in the distorted-wave equations. Although the major relativistic effect on the continuum orbitals is the relativistic modification of the target distorting potentials, a mass-velocity potential is also included in the relativistic formulation of the distorted-wave equations.

The results of distorted-wave Born calculations for the electron-impact double-ionization cross section of Fr^+ are shown in Fig. 1. The dotted curve in Fig. 1 is an estimate of the direct double-ionization process only, obtained using a binary-encounter approximation calculation.[14] The threshold energy for double ionization of Fr^+ is calculated to be 54 eV. The solid curve labeled HF in Fig. 1 is a nonrelativistic calculation for the total cross section including the contribution from $5d$ ionization-autoionization. The HF threshold for $5d$ ionization is 78.2 eV. The solid curve labeled HFR in Fig. 1 is a relativistic calculation for the total cross section including the contribution from $5d$ ionization-autoionization. The HFR threshold energy for $5d$ ionization is 71.6 eV, almost 7 eV lower than the HF value. An average-configuration Dirac-Fock calculation, using the wave-function code developed by Grant,[15] yields a $5d$ ionization energy of 71.4 eV, in good agreement with the HFR results.

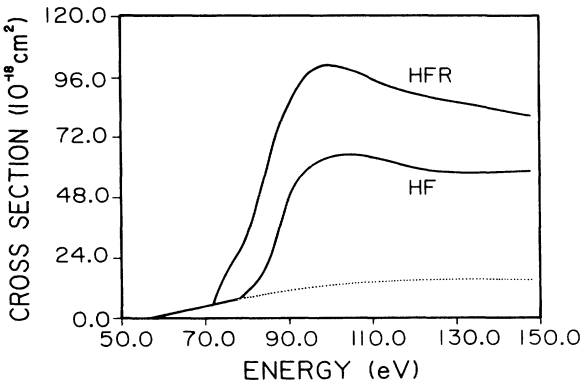

FIG. 1. Distorted-wave Born calculations for the electron-impact double-ionization cross section of Fr^+. The dotted curve is a binary-encounter approximation calculation for the direct double-ionization process only; the solid curve labeled HF is a nonrelativistic calculation for the $5d$ ionization-autoionization contribution plus direct double ionization; the solid curve labeled HFR is a relativistic calculation for the $5d$ ionization-autoionization contribution plus direct double ionization.

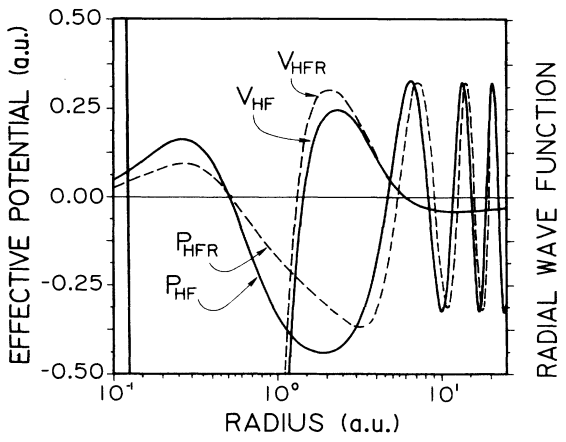

FIG. 2. Effective potentials and continuum wave functions for Fr^+. The solid curve labeled V_{HF} is the Hartree-Fock effective potential; the solid curve labeled P_{HF} is the Hartree-Fock k_sf continuum orbital; the solid curve labeled V_{HFR} is the Hartree-Fock relativistic effective potential; the solid curve labeled P_{HFR} is the Hartree-Fock relativistic k_sf continuum orbital. The energy $k_s^2/2=10$ eV.

At around 100-eV incident electron energy the distorted-wave Born results of Fig. 1 differ by about a factor of 1.5. The difference can mainly be attributed to relativistic changes in the bound-state orbitals used to construct the scattering potentials. Although all partial wave are affected to some degree, it is interesting to examine in detail the $l=3$ scattered partial wave because it contains a giant resonance. The radial Schrödinger equation for the $P_{k_sl}(r)$ distorted wave is given by (in atomic units)

$$\left[-\frac{1}{2}\frac{d^2}{dr^2}+V_{\text{eff}}(r)-\frac{k_s^2}{2}\right]P_{k_sl}(r)=0\ , \tag{3}$$

where k_s is the scattered electron momentum and $V_{\text{eff}}(r)$ is an effective local potential. In Fig. 2 the average-configuration effective potentials and k_sf continuum orbitals are plotted for the nonrelativistic and relativistic cases. The most obvious difference in the effective potential curves is that the positive energy barrier between the inner and outer potential wells is slightly larger in the relativistic case. The difference in barrier heights is caused by changes in the bound-state orbitals; the largest change in the outer orbitals is the relativistic contraction of the $6s$ orbital from a mean radius of $2.02a_0$ to $1.81a_0$. The energy of the scattered wave is 10.0 eV in Fig. 2. As the energy increases, the nodes of the scattered wave move further into the barrier region. Average-configuration effective potentials for the $l=3$ ejected partial wave also exhibit a double-well structure. The potential barrier for the ejected wave is much smaller than that for the scattered wave; in fact, it is negative. Again, however, the barrier height in the relativistic case is slightly larger.

Unlike photoionization calculations, the triple-partial-wave nature of electron ionization complicates the interpretation of what effect any particular node penetration delay due to a larger barrier height has on the total cross section for Fr^+. Substantial changes are found in the dominant $l_i=4$, $l_e=3$, $l_s=3$ partial-wave cross section. The HFR value for the direct-dipole Slater integral, $R^1(5dk_ig,k_efk_sf)$, is about 40% larger than the HF value at small ejected energies for an incident energy 20 eV above threshold. This results in an HFR ejected-energy differential cross section which is about 2.6 times the HF value at small ejected energies. Integrating over all ejected energies, the HFR value for the $l_i=4$, $l_e=3$, $l_s=3$ partial-wave cross section is found to be 1.4 times the HF value. Summing over all partial waves, the relativistic $5d$ ionization cross section for Fr^+ is about 1.5 times the nonrelativistic cross section in the threshold energy region.

Electron-ionization calculations[2,7] that go beyond the average-configuration method, outlined above, to include either LS term-dependent or ground-state correlation effects are reasonably successful for moderately heavy ions. For heavy ions, like Fr^+, attempts to go beyond the average-configuration method are complicated by the presence of strong spin-orbit splittings. A Dirac-Fock calculation for Fr^+ yields a $5d\,(J=\frac{5}{2})$ ionization energy of 68.4 eV, while the $5d\,(J=\frac{3}{2})$ ionization energy is 73.4 eV. Atomic structure calculations for high Rydberg states indicate that a substantial breakdown of LS coupling will occur for ejected electron configurations such as $Fr^{2+}5d^96s^26p^6k_ef$. Thus it appears that an intermediate-coupled formulation of the ionization cross section is needed to go beyond an average-configuration calculation.

In conclusion, the distorted-wave Born calculations for

the $5d$ ionization cross section of Fr^+ are found to be quite sensitive to relativistic effects on bound-state wave functions in the vicinity of a giant resonance. Such sensitivity, combined with the long-standing problem of the choice of phase between direct and exchange contributions, calls into question the potential accuracy of such first-order Born calculations for any giant resonance in very heavy atomic ions. No doubt future experimental measurements of the double-ionization cross section in heavy ions will shed light on some of the problems facing ionization theory.

The author wishes to thank S. M. Younger and D. C. Griffin for a number of helpful discussions, and to thank R. D. Cowan for making available his atomic structure program. This research was supported by the Office of Fusion Energy, U.S. Department of Energy, under Contract No. DE-FG05-86ER53217 with Auburn University.

[1]C. Achenbach, A. Muller, E. Salzborn, and R. Becker, Phys. Rev. Lett. **50**, 2070 (1983).

[2]M. S. Pindzola, D. C. Griffin, C. Bottcher, D. H. Crandall, R. A. Phaneuf, and D. C. Gregory, Phys. Rev. A **29**, 1749 (1984).

[3]A. Müller, C. Achenbach, E. Salzborn, and R. Becker, J. Phys. B **17**, 1427 (1984).

[4]D. C. Griffin, C. Bottcher, M. S. Pindzola, S. M. Younger, D. C. Gregory, and D. H. Crandall, Phys. Rev. A **29**, 1729 (1984).

[5]C. Achenbach, A. Müller, E. Salzborn, and R. Becker, J. Phys. B **17**, 1405 (1984).

[6]A. Müller, Phys. Lett. **113A**, 415 (1986).

[7]S. M. Younger, Phys. Rev. Lett. **56**, 2618 (1986).

[8]S. M. Younger, Phys. Rev. A **35**, 2841 (1987).

[9]M. S. Pindzola, D. C. Griffin, and C. Bottcher, in *Atomic Processes in Electron-Ion and Ion-Ion Collisions,* Vol. 145 of *NATO Advanced Study Institute, Series B: Physics,* edited by F. Brouillard (Plenum, New York, 1986), p. 75.

[10]R. K. Peterkop, Zh. Eksp. Teor. Fiz. **41**, 1938 (1961) [Sov. Phys.—JETP **14**, 1377 (1962)].

[11]R. D. Cowan, *The Theory of Atomic Structure and Spectra* (University of California Press, Berkeley, 1981).

[12]R. D. Cowan and D. C. Griffin, J. Opt. Soc. Am. **66**, 1010 (1976).

[13]M. E. Riley and D. G. Truhlar, J. Chem. Phys. **63**, 2182 (1975).

[14]M. Gryzinski, Phys. Rev. **138**, A336 (1965).

[15]I. P. Grant, B. J. McKenzie, P. H. Norrington, D. F. Mayers, and N. C. Pyper, Comput. Phys. Commun. **21**, 207 (1980).