Double Ionization of Helium by Protons and Antiprotons in the Energy Range 0.30 to 40 MeV

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(Received 22 October 1986)

We present an *ab initio* calculation of the double ionization of helium by protons and antiprotons, using the *forced impulse* method. Cross sections are larger for antiprotons than for protons, in accord with recently reported experimental results.

PACS numbers: 34.50.Fa

The production of high-energy beams of bare ions of increasing charge has given atomic physicists the unique opportunity to change the coupling constant in nontrivial reactions and much has been learned. The report[1] of the acquisition of a beam of MeV/u antiprotons at the CERN Low Energy Antiproton Ring has been eagerly anticipated by theorists anxious to demonstrate the validity of their calculational methods in this negative coupling-constant regime.[2] Electron beams have been available for many years, and interesting differences between cross sections for the double ionization of helium produced by high-energy electrons and by protons traveling at the same speed have been observed.[3,4] But as these differences had no convincing immediate interpretation there was always the possibility that perhaps the relevant parameter being varied was the mass of the projectile and not the charge. The recently reported antiproton results indicate that to a large extent a high-energy electron is equivalent to an antiproton moving with the same speed. Though there are anticipated differences at lower energies between cross sections produced by antiprotons and electrons, the first point of interest is that at high energies antiprotons and protons have now been shown to give different cross sections. Furthermore, the cross section for the double ionization of helium is bigger for antiprotons, eliminating the obvious first guess that charge transfer might play a role in the cross section for protons. Not only would this give the opposite result to that found, but both measured and calculated charge transfer cross sections[5] indicate very little flux in this channel above 1 MeV/u.

There is a second and perhaps even more important challenge. At extremely high energies, greater than 20 MeV/u, the double ionization cross section, calculated as the sequential single removal of two independent electrons, is an order of magnitude less than the data. Another mechanism has therefore been invoked[4] called *shakeoff*. Here only one electron is supposed to be suddenly ejected by a primary collision with the projectile. The other is left in a single-particle orbital appropriate to neutral helium when in fact the system is singly ionized helium. The subsequent collapse of the wave packet to the appropriate linear combination of stationary states of the singly ionized helium system produces ionization. This mechanism is important in the double ionization of helium by energetic photons and in that context is well understood. However, the ejected electron in a high-energy ion-atom collision moves rather slowly away from the atom. Thus the shakeoff limit is not applicable and gives a cross section predicted to be an order of magnitude higher than that observed.[4]

This Letter reports on the results of a method[6] devised to accurately solve numerically the collision of a bare ion with a correlated many-electron atom. It is called the *forced impulse* method (FIM). Using it we can only reproduce the absolute value of the experiments to within 35%, but that is probably because the calculation only has s and p waves included for each electron. We do, however, obtain higher cross sections for antiprotons than protons, though the difference is not so great as has been observed.

At the high energies of interest here the projectile and atomic nuclei can accurately be assumed to travel past each other on straight-line paths and provide a time-dependent perturbation of the electrons in the system. The infinite spectrum of electronic eigenstates can be approximated by a finite set of pseudostates[7]; they are eigenfunctions of the unperturbed Hamiltonian projected onto a truncated Hilbert basis space. Transitions between these pseudostates are calculated by the integrating out of time-dependent coupled equations. Typically, for a single-electron system, a total of sixty s, p, and d target-centered pseudostates are needed to obtain accurate transition amplitudes. Projectile states are not needed if charge transfer is unimportant.[8] If, as has been almost universal practice, the electron-electron interaction is replaced by some average effective single-particle potential, then many-electron systems can be solved in the same way by formation of a determinant of single-particle orbitals each with a different initial starting condition.[9] This method yields answers at least an order of magnitude too small for the double ionization of helium at high energies.[4] If the electron-electron force is retained one can expect that the square of the number of pseudostates, approximately three thousand, is needed for convergence in a connected two-electron calculation. It is a formidable task to integrate directly such a large system of coupled equations. This is the problem that is

addressed by the FIM.[6]

If the projectile is moving very fast then the target electrons have no time to sense any binding forces before the collision is over. The collision can be accurately described with the impulse approximation.[10] Correlated systems are easily treated as the electron-electron force enters only through the initial or final state. The FIM provides a systematic method of extending the validity of this method to lower energies. It chops the time development of the system into segmented pieces chosen short enough that the electron-electron force may be neglected during the time evolution within each time segment. Initially, at time t_1, the beginning of a segment, the system is in one of a complete set of unperturbed fully correlated states. During each time segment it is allowed to develop as an independent-particle system. Before this approximation has time to become poor, at time t_2, the system is allowed to collapse back into a linear superposition of unperturbed correlated states.

The algorithm we have used is

$$U_{\beta\beta'}(t_2,t_1)=\delta_{\beta\beta'}+\sum_{\alpha\alpha'} a_\alpha^\beta \int_{t_1}^{t_2} dt\, e^{i\Delta t}\frac{d}{dt}U_{\alpha\alpha'}(t,t_1)a_{\alpha'}^{\beta'}.$$

Here the label β refers to the pseudostates obtained by diagonalization of the full unperturbed two-electron Hamiltonian. The label α refers to the independent-particle model pseudostates, i.e., the two-dimensional determinant produced by diagonalization of the separable two-electron Hamiltonian with the electron-electron force turned off. The a_α^β are the time-independent amplitudes for the expansion of the β states in the α representation. If we represent the energies of these pseudostates by ε, then

$$\Delta=\varepsilon_{\beta'}-\varepsilon_\beta-\varepsilon_{\alpha'}+\varepsilon_\alpha.$$

This is an *off-energy-shell* effect which simply reflects the fact that though there may be admixtures of α states in the β states they propagate with the energy ε_β.

The complete collision is described by a product of consecutive segment U matrices. The characteristic computer time needed to find the integrand for each segment is that of a single-particle problem, not a many-particle problem. The integration, however, is still time consuming. And, of course, if the length of each segment is as short as the time interval needed to integrate out the single-particle system nothing has been gained.

For the system at hand we tested the FIM at 7 MeV/u. Using a small basis set we first solved the coupled equations by direct integration. We then used the FIM correlated at three points: initially at the start of the collision, at the distance of closest approach, and finally at the end. Satisfactory agreement between these two methods was obtained. In the rest of this work this three-stage formulation was used from 0.30 to 40 MeV without further study of its convergence in the number of steps. More details of the method are given in earlier work.[6]

In the single-electron system a *positive*-energy pseudostate can be identified unambiguously as representing a group of ionized states with energy centered at the pseudostate energy. With this *interpretation* ionization is easily calculated by summation over positive-energy pseudostate transition probabilities. In a two-electron system no such clear identification is possible for a pseudostate with energy above the two-electron ionization threshold because of the energy degeneracy of the single- and double-ionization spectra. The FIM may have solved the problem of determining the collisional amplitudes for transitions between pseudostates but these amplitudes are useless without a reliable interpretation of what the states represent.[11]

A way to proceed is first to construct, numerically, specific single- or double-ionization *channel* wave functions over some appropriate energy range and then to project these onto the calculated FIM wave function. Unfortunately, the single-ionization cross section at high energies is 2 to 3 orders of magnitude greater than the double-ionization cross section. A small numerical error in the double-ionization wave function will inevitably introduce a random amount of single-ionization wave function into it. When one uses this to project out the hoped for double-ionization amplitude a small amount of single-ionization amplitude is picked up. This leads to numerical instabilities which we could not eliminate and still keep the underlying single-particle basis small enough for a numerically practical calculation.

The method we finally applied was to construct a complete set of single- and double-ionization channel wave functions at each two-particle pseudostate energy. Each pseudostate was then reexpressed as a sum of partitioned single- and double-ionization channel states, by use of a method especially designed for projected channel wave functions which will be described more fully in a lengthier publication. This partition was inserted into the U matrix algorithm described above. Thus with each β state now written as a sum of single and double partitions, β^s and β^d, the U matrix for double ionization is given by

$$U_{\beta^d\beta}(\infty,-\infty)=\textstyle\sum_{\beta'}U_{\beta^d\beta'}(\infty,0)U_{\beta'\beta}(0,-\infty).$$

The motivation for this approach is that it is numerically more accurate to calculate a small $U_{\beta^d\beta'}$ directly by integration than to try to extract it from a knowledge of the (possibly large) $U_{\beta\beta'}$ by projection.

In the three-stage formation described here we had to integrate only the positive time segment; we exploited time-reversal symmetry to infer the negative-time-segment U matrix.

The results of our calculation of the ratio, R, of double to single ionization for protons and antiprotons are presented in Fig. 1. We generally limited the basis to nine states for each single-particle angular momentum

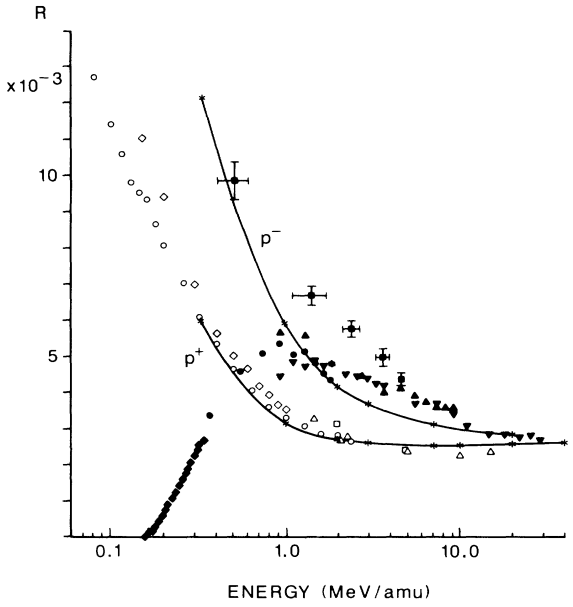

FIG. 1. The ratio R between double- and single-ionization cross sections for p^-, p^+, and e^- colliding with He. The solid curves labeled p^+ and p^- are our calculations for these projectiles, scaled by 1.35 as described in the text. The closed symbols are for p^- and e^-. The antiproton data are due to Andersen *et al.* (Ref. 1), squares with error bars. The electron data are due to Adamczyk *et al.* (Ref. 12), circles; Nagy, Skutlartz, and Schmidt (Ref. 13), triangles; Schram, Boerboom, and Kistemaker (Ref. 14), inverted triangles; and Stephen, Helm, and Märk (Ref. 15), lozenges. The open symbols are proton data due to Andersen *et al.* (Ref. 1), squares; Knudsen *et al.* (Ref. 16), triangles; Shah and Gilbody (Ref. 17), circles; and Puckett and Martin (Ref. 18), lozenges.

quantum number, l, and each azimuthal quantum number, m, We also limited ourselves to s and p states only, so that there are 36 single-electron orbitals. Angular momentum coupling leads to two-electron symmetries of ^1S, $^1P^e$, $^1P^o$, and ^1D that couple via the collision to the ^1S ground state. The single-particle orbitals used to construct the two-electron states were taken to be of definite time-reversal symmetry, so that this symmetry could be exploited. This leads to 423 two-electron β states. Convergence studies were then carried out at various energies with use of eleven states for each l and m, which yields 627 two-electron β states. Numerical stability against this change was established at the 2% level for R.

Experimentally R appears to be a constant at high energies and independent of the sign of the projectile. The FIM calculation we performed gave R with the same characteristics but it was some 35% lower than the observed value. We tentatively assign this discrepancy to the absence of d states in the single-particle basis. So in Fig. 1 we multiplied all our results for R, both for p^+ and for p^-, by the same factor of 1.35, to allow one more easily to compare our calculated p^+,p^- difference to that observed experimentally. We emphasize again that sudden-approximation calculations, appropriate to shakeoff in high-energy photon ionization, give a value of R that is a factor of 10 larger than the R we calculate and that is observed experimentally for these collisions.[3,4] Ours is the first *ab initio* calculation of R appropriate to ion-atom collisions, even in the high-energy limit.

As Fig. 1 shows, proton-impact results from several different groups are in rather good agreement with each other. Similarly, the e^--projectile results from various groups are in reasonably good agreement with each other. The downturn in R for e^- below the equivalent proton energy of 1 MeV/u is due to approach to the double-ionization threshold energy at 79 eV electron energy, which is 0.145 MeV/u. The very recent p^- data from Andersen *et al.*[1] are in fair agreement with the e^- data for energies well above the e^- threshold.

As the energy is lowered there is a marked difference in our calculated R for protons versus antiprotons that comes almost exclusively from differences in the double-ionization cross sections for the two projectiles. Though this difference is of the right sign it is not as large as experimentally measured. Once again this may be due to the absence of d states in the single-particle basis. If the effect is due to a low-energy electron-electron correlation then we might expect high angular momentum to be important as the single-particle basis attempts to mock up a two-body effect. Indeed we may eventually have to put functions of the displacement vector $\mathbf{r}_{12}$ between the two electrons into the basis set to reproduce the experiment.

Nevertheless, this first application of the FIM to this practical problem encourages the belief that significant progress can be made in the study of ion collisions with correlated atoms especially if guided by such stimulating experimental results.

This work was supported by U. S. National Science Foundation Grant No. PHY-8407263, a Los Alamos Laboratory subcontract No. 9X25Q3283, and a grant from the Texas A&M Center for Energy and Mineral Resources. We thank H. Knudsen for communication of the results of Ref. 1 prior to publication.

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