Evaluation of antiproton-impact ionization of He atoms below 40 keV

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We have performed extensive close-coupling calculations to obtain total impact ionization cross sections of He atoms by antiprotons at energies from 1 keV to 300 keV by expanding the time-dependent two-electron wave functions in terms of helium eigenstates. The stability of the calculated ionization probabilities and total ionization cross sections with respect to the choice of basis functions has been examined. Our total ionization cross sections are only slightly higher than the results from the forced-impulse approximation of Reading *et al.* [J. Phys. B **30**, L189 (1997)] but they disagree with the existing experimental data below 40 keV [Hvelplund *et al.*, J. Phys. B **27**, 925 (1994)]. We conclude that new measurements for the ionization of He by antiprotons are needed in the low-energy region.

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I. INTRODUCTION

Since antiproton beams became available at CERN for atomic collision experiments in 1986, a number of measurements have been carried out at high energies (above a few hundreds keV), for example, to study the ratio of cross sections for double ionization to single ionization of atomic helium targets [1,2]. The ratios have been found to be different for proton and antiproton projectiles. Since single-ionization cross sections are expected and found to be identical for proton and antiproton impact at high energies, the measured different ratios prompt many theoretical papers to study the role of the higher-order perturbation terms and the importance of electron correlation in the double-ionization processes. At lower energies, the cross sections for single ionization by protons and by antiprotons are expected to be different. The difference is understood to be more dramatic as the collision energies are lowered [3]. For proton collisions at low energies, electron capture process is dominant and ionization is a rare event. Without the electron capture channels, ionization by antiprotons is expected to have larger cross sections, but antiproton collision experiments are much more difficult to perform because of the degrading of the beams at low energies. Nevertheless, such a pioneering experiment has been carried out by Hvelplund *et al.* [4] for antiproton energies down to about 12 keV. Their results, together with the data taken earlier at higher energies [5], are displayed in Fig. 1. Note that the two experiments agree reasonably well at energies above 50 keV.

Since the report of the experimental results, various theoretical approaches have been used to obtain the ionization cross sections for this system. Except for the so-called CDW-EIS theory [6], which is not known to be valid in the low-energy region, all the other calculations fail to agree with experimental results below 40 keV. In the theoretical calculations, however, various approximations have to be employed and one wonders if there is still something missing in the theory that is responsible for the discrepancy. In performing such calculations one may classify the approximations into three categories. First, the scattering model: is the model expected to work in the low-energy region? Second, the He wave function: are the two electrons treated on equal footing with antisymmetrized wave functions and the electron-electron interaction properly accounted for? It is not advisable to employ the independent particle approximation without checking its validity. Third, how are the ionization channels represented? Most of the existing nonperturbative methods can treat ionization in some approximations only, namely, by representing them using pseudostates. How much is the error expected in such approximations?

In this paper we have used the semiclassical impact parameter close-coupling approximation to treat the collisions of antiprotons with helium atoms by expanding the time-dependent two-electron wave function in terms of the eigenstates of helium atoms. The computer program is the general

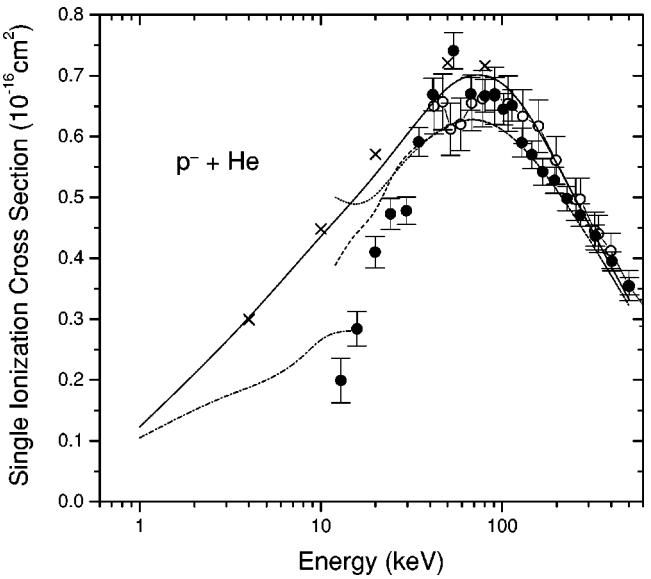

FIG. 1. Single-ionization cross sections for antiproton collisions with He. Experiments: solid circles, Hvelplund *et al.* [4]; open circles, Andersen *et al.* [5]. Theoretical results: solid line, present calculation with basis set 1; crosses: present results with basis set 2; dashed lines, multicut FIM theory [10]; dotted lines, one-cut FIM theory [10]; dash-dotted lines, multielectron hidden-crossing theory [13].

TABLE I. Comparison of the two sets of eigenenergies (a.u.) of He singlet states. The two sets differ primarily in the distributions of the pseudostates (for energies greater than −2.00 a.u.). Both sets of basis functions have been used in the close-coupling calculations. The experimental binding energies of He for the first few excited states are also shown.

$L=0$			$L=1$			$L=2$			$L=3$		
Set 1	Set 2	Experiment	Set 1	Set 2	Experiment	Set 1	Set 2	Experiment	Set 1	Set 2	Experiment
−2.8791	−2.8803	−2.9033	−2.1226	−2.1225	−2.1236	−2.0556	−2.0556	−2.0554	−2.0221	−2.0260	−2.0310
−2.1445	−2.1446	−2.1458	−2.0548	−2.0548	−2.0549	−2.0309	−2.0310		−2.0122	−2.0086	
−2.0608	−2.0608	−2.0160	−2.0299	−2.0303		−2.0047	−2.0114		−1.9890	−1.9984	
−2.0333	−2.0334		−2.0171	−2.0176		−1.8821	−2.0009		−1.8796	−1.9904	
−2.0209	−2.0212		−1.9861	−1.9955		−1.6749	−1.9674		−1.7803	−1.9808	
−1.9940	−2.0072		−1.9651	−1.9905		−1.3483	−1.9343		−1.2914	−1.9620	
−1.9218	−2.0029		−1.8283	−1.9879		−1.2153	−1.8751		−0.6591	−1.9192	
−1.7884	−1.9820		−1.5241	−1.9861		−0.6735	−1.8189		0.9311	−1.8801	
−1.5645	−1.9817		−1.0516	−1.9798		−0.2254	−1.6225		1.9266	1.2916	
−1.2751	−1.9291		−0.6405	−1.9714		0.5197	−0.7833				
−0.8984	−1.9273		−0.4641	−1.9162		1.0864	−0.6729				
−0.7585	−1.8956		−0.4445	−1.7475		2.8328	0.8915				
−0.5773	−1.4578		0.2480	−1.2090							
−0.0093	−0.7669		0.3348	−0.8324							
0.3044	−0.7159		0.7825	−0.4490							
0.5436	−0.5129		1.2465	0.2022							
0.6908	0.4831		1.5875	0.7121							
1.3421	0.9432		3.7802	0.9663							
			4.6755	1.9982							

two-electron two-center atomic orbital expansion code, which has been used extensively with great success in ion-atom collisions [7,8]. In applying this code to the present system, we only include the eigenstates on the helium center since there are no bound electronic states associated with the antiproton center. We used pseudostates to represent the ionization channels, and these pseudostates are centered also only on the target atom. It may be argued that it would be desirable to include some pseudostates around the antiproton to account for ''antielectron capture to the continuum.'' However, we will show that such a two-center calculation has been found not essential for the ionization of atomic hydrogen by antiprotons. Thus we expect that a similar equivalent one-center calculation be adequate for antiproton collisions with helium atoms.

In Sec. II we show calculations performed using two different basis sets and compare the total ionization cross sections obtained. The results are then compared to experimental data and with other calculations in Sec. III. By analyzing the theoretical results we believe that the discrepancy with experimental data is most likely coming from the experimental side and thus new measurements are called for when the new low-energy antiproton beams become available in the near future. Section IV gives a short summary to end this paper.

II. THEORETICAL CALCULATIONS AND BASIS SETS

We employed the semiclassical impact parameter model where the antiproton is assumed to travel on a rectilinear trajectory. Following the standard close-coupling approximation the time-dependent two-electron wave function is expanded in terms of the eigenstates of the helium atoms, the scattering amplitude to each final state is obtained by solving the resulting coupled first-order differential equations.

The complete basis set in such a calculation should consist of all the eigenstates of the He atom. They include the ground state, the singly excited states, doubly excited states, single-ionization states, ionization plus excitation states, and double-ionization states. At low energies states that have higher excitation energies are populated with much smaller probabilities, thus higher doubly excited states, excitation plus ionization states, and double-ionization states are not included in our basis set. Since the main interest is the single-ionization cross section, in the basis set we thus include the ground state, the singly excited states, and the single-ionization states that are approximated by pseudostates. Starting with primitive Slater functions for each electron with adjustable parameters, the two-electron basis functions with appropriate symmetries are constructed and used to diagonalize the two-electron Hamiltonian of the helium atom.

In Table I we show the resulting eigenenergies obtained from two different basis sets that are used in the scattering calculations. Since the total spin is considered as a good quantum number, only singlet states need to be included. For the first few bound states we also list the experimental energies for comparison [9]. Since the $L=0$ and $L=1$ states are populated mostly in the collision, efforts were made to increase the number of states in these two partial waves. For $L=2$ and $L=3$ the basis set was obtained without optimization in order to keep the total number of basis functions in

TABLE II. Comparison of the calculated total ionization probabilities from the two different basis sets used. The collisional impact parameter is fixed at $b=0.5$ a.u. The ionization probability to each partial wave L is also presented.

E (keV)	Set	$L=0$	$L=1$	$L=2$	$L=3$	Sum
4	1	0.1356	0.1149	0.0811	0.0294	0.3610
	2	0.1358	0.1396	0.0668	0.0186	0.3609
10	1	0.1518	0.1811	0.0855	0.0055	0.4238
	2	0.1603	0.1914	0.0379	0.0012	0.3908
20	1	0.1239	0.2348	0.0645	0.0079	0.4311
	2	0.1502	0.2425	0.0283	0.0021	0.4231
50	1	0.1004	0.2560	0.0497	0.0014	0.4202
	2	0.1286	0.2756	0.0195	0.0019	0.4256

the scattering calculation small. Since the binding energy of $He^+(1s)$ is -2.0 a.u., all the eigenstates with energies greater than -2.0 a.u. are pseudostates. The threshold for single ionization plus He^+ core excitation to $n=2$ is -0.5 a.u., thus the pseudostates with energies between -2.0 and -0.5 a.u. represent single-ionization channels. (Some doubly excited states exist below $E=-0.5$ a.u. They are partially excluded by choosing the orbital of one of the electrons to be close to the $1s$ of He^+.) For pseudostates with energies higher than -0.5 a.u. each pseudostate is a certain combination of single ionization, ionization plus core excitation, and double ionization. The pseudostates used were chosen not to contain such components as much as possible by forcing the inner electron orbital to represent a $He^+(1s)$ electron. This is only an approximation and it does not allow us to distinguish these channels rigorously unless careful projections are carried out. For the purpose of this work, we will present ionization cross sections as the sum of the cross sections to all pseudostates with energies higher than -2.0 a.u. This may introduce some small ambiguity in the cross sections obtained when compared to the experimental single-ionization cross sections. On the other hand, contribution of cross sections from pseudostates with energies higher than -0.5 a.u. is less than 5–10 % in the energy range considered.

A key element in determining whether the calculated total ionization cross section is stable or not is to see if the results depend on the basis set used. For this purpose we used two basis sets that have different pseudostate distributions. Another criterion is to make sure that there are more pseudostates with energies near the ionization threshold since that is the region where the ionization probability is large. The other basis set has been generated and used in the calculation but we only present complete results from these two sets since they have the better pseudostate distributions.

III. RESULTS AND DISCUSSION

In Table II we show the total ionization probabilities calculated at the fixed impact parameter $b=0.5$ a.u. at four different energies and how they are distributed among the different total orbital angular momentum states. In general the two basis sets give nearly identical partial wave contributions despite that the pseudostate distributions in each partial wave are different. Furthermore it is noticed that the total probability is even less sensitive to the two different basis sets used. The near independence of the total ionization probability with respect to the pseudostates used is essential. Thus we have confidence that the calculated cross sections are accurate to within a few percent with respect to the pseudostates used.

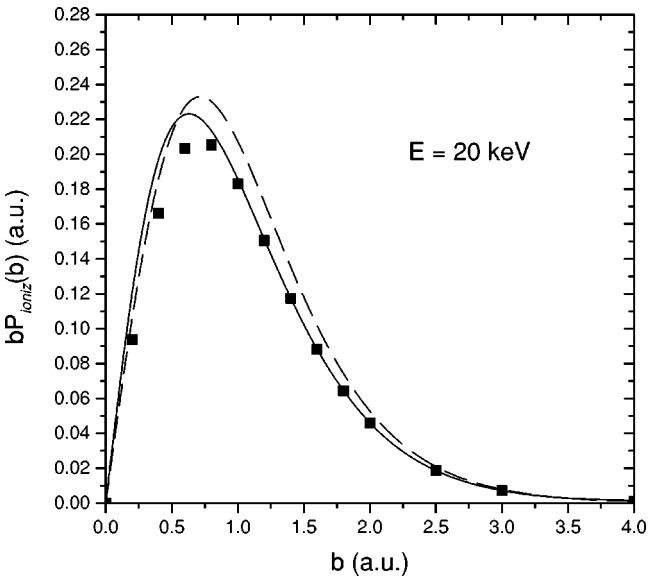

FIG. 2. Impact-parameter-weighted total ionization probability vs impact parameters for antiproton-He collisions at 20 keV. The solid line is calculated from basis set 1. Results from basis set 2 are shown as dashed lines. The squares indicated results from summing over pseudostates up to $E=0.0$ a.u. only, from basis set 1 (see text for explanation).

To assess the reliability of the calculated ionization cross sections we further show the impact-parameter-weighted total ionization probability as a function of impact parameters calculated from the two basis sets. In Fig. 2 we show the results for $E=20$ keV. Note that the difference appears to be larger at the larger impact parameters. This is traced to the fact that the higher L's are making more contributions in this energy region at larger impact parameters. In Fig. 2 we also show the weighted probabilities obtained by including pseudostates with energies from -2.0 to 0.0 a.u. only. The result would give a lower limit to the single-ionization cross section. In this approximation it is assumed that all the pseudostates with energies greater than 0.0 a.u. are double-ionization channels, which, of course, is not correct. Note that the difference is significant only at small impact parameters. This is easily understood since large energy transfer to the electron(s) occurs only in close collisions. Under this approximation, the total ionization cross section calculated is 0.538×10^{-16} cm^2, to be compared to the value 0.571×10^{-16} cm^2 obtained by summing over all the pseudostates. The difference can be used as an upper bound of the double-ionization cross sections. Since the pseudostate distributions for these higher energy states are very sparse, this estimate is probably not very accurate.

We have used the same two basis sets to calculate the

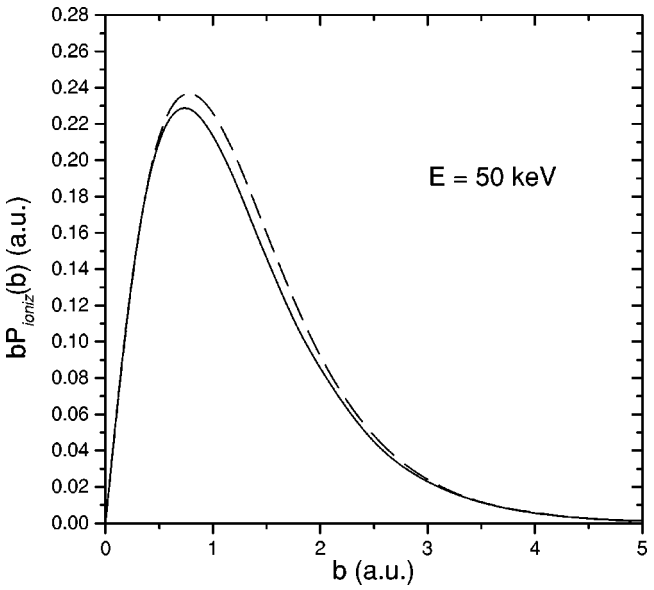

FIG. 3. Impact-parameter-weighted total ionization probability vs impact parameters for antiproton-He collisions at 50 keV. The solid line is calculated from basis set 1. Results from basis set 2 are shown as dashed lines.

ionization probabilities at $E=4$ keV and $E=50$ keV, and the resulting weighted ionization probabilities are shown in Fig. 3 and Fig. 4, respectively. Clearly we can claim that the results are relatively insensitive to the basis sets used.

We can now return to Fig. 1 to discuss the results of the present calculation in comparison with other existing calculations and experiments. For collision energies above 70 keV the experimental results of Hvelplund *et al.* [4] and of Andersen *et al.* [5] agree quite well with the results from the present calculation, which tend to agree better with the latter and the results from Reading *et al.* [10] tend to agree better with the former. However, we mention that our results in principle include some small contributions of double-ionization cross sections.

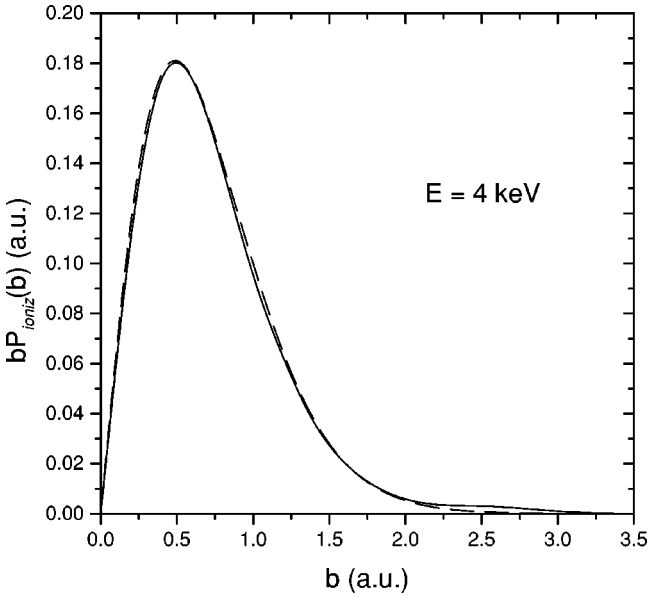

FIG. 4. Same as Fig. 3 but for $E=4$ keV.

The issue that we would like to address is the discrepancy between experiment of Hvelplund *et al.* [4] and the existing theoretical calculations at energies below 40 keV, but more notably below 20 keV. As mentioned in the Introduction, there have been many previous theoretical calculations attempting to interpret the low-energy experimental result. The only model that gives ''perfect'' agreement with the data is the CDW-EIS calculation of Fainstein *et al.* [6] (not shown). This agreement is considered to be fortuitous since the CDW-EIS model is a high-energy theory and is not known to work in the low-energy region considered here. There are other calculations [11,12] carried out within the context of the independent electron model and at higher energies; they do not address the discrepancy discussed here. Two other calculations have been made to address the ionization cross sections in this low-energy regime. One is the so-called forced-impulse approximation (FIM) of Reading *et al.* [10]. In the FIM method the two-electron wave function is propagated in time in a set of two-electron basis functions, but the electron-electron interaction is turned on at discrete time steps. In the single-cut FIM this interaction was turned on only once. In the multicut FIM the interactions were turned on up to seven times. The results from such a single-cut and multicut FIM are shown in Fig. 1. These authors showed that a single-cut FIM is not adequate, especially at lower energies, but the multicut FIM results appear to approach toward the experimental data of Hvelplund *et al.* [4], except that the experimental data show a much steeper drop with decreasing energies. Another calculation in the low-energy region is the hidden-crossing theory result of Bent *et al.* [13]. This is based on treating atomic collisions in the molecular basis but the molecular potential energy surfaces are calculated in the complex plane of the internuclear axis. Using the analytical property of the potential surface, the transition probabilities can be calculated in terms of simple contour integrals around the branch points. This method has been used extensively [14–16] to study one-electron collision systems to obtain ionization cross sections and the work of Bent *et al.* [13] is the first attempt for a two-electron system. Since the potential surface was calculated with an approximation where full configuration interaction (CI) was not included, it is not clear how to evaluate the accuracy of the reported results. Nevertheless, they are shown for comparison in Fig. 1. It must be mentioned that their results above 4 keV had been obtained by extrapolating to the region where the validity of the hidden-crossing theory is a concern.

Let us now discuss the present calculated total ionization cross sections. The results from basis set 1 are shown as a solid line, by connecting smoothly the calculated points. The actual results obtained from basis set 2 are indicated by crosses that are quite close to those from basis set 1. In comparing to the multicut FIM results of Reading *et al.*, there is a quite reasonable agreement within the energy range of 12–50 keV where the two results differ by about 10–15 %. Most importantly the energy dependence from the two calculations is essentially identical. The drop with decreasing collision energy is not as steep as indicated in the experi-

ment. We have extended the calculation down to 1 keV. For calculations at even lower energies one may need to use nonrectilinear trajectories in the semiclassical calculation. We comment that the result from the multielectron hidden-crossing theory is quite close to ours at 1 to 2 keV, though it has a different energy dependence.

Based on the results obtained from our calculation and from those of Reading *et al.*, we believe that it is appropriate to challenge the experimental ionization cross sections reported in Hvelplund *et al.* [4] at lower energies. We believe that the cross section should not drop as rapidly as reported. In view of the difficulty of performing experiments with diminishing beams at these low energies, such error in the measurement is not inconceivable. One may argue that ionization cross section is the easiest measurement that can be performed for collisions with antiprotons in any energy regime, thus the existing discrepancy certainly calls for new measurements once the new antiproton beams become available in the next year or two [17,18] before any other collision experiments are carried out.

The above conclusion is based on our estimation that the final total ionization cross sections will not be changed significantly by the approximations employed in the present calculation. In this work the electron-electron interaction is included explicitly. The main approximation would be the representation of the ionization channels with a limited number of pseudostates. We believe such an approximation would not cause large errors in the total ionization cross sections based on experience from calculations in many ion-atom collision systems. We further checked that the reported total ionization cross section is not sensitive to the pseudostates chosen. Another possible concern is the single-center basis functions used in the present calculation and the truncation of the partial waves up to $L=3$ only. Such a truncated expansion is not expected to describe fully the final state interaction between the electron and the antiproton, but such an effect will not change the total ionization cross section significantly. This speculation is drawn from the study of antiproton collisions with atomic hydrogen where many more detailed calculations have been made by various methods. To illustrate this, we present the ionization cross sections for this system from 1 to 300 keV in Fig. 5. Calculations carried out using the present single-centered expansion method are shown as a solid line, from another single-centered calculation of Hall *et al.* [19] calculations are shown as dashed lines. They are to be compared with results obtained from integrating the time-dependent Schrödinger equation on space grid points of Wells *et al.* [20] where no basis functions were employed. In our close-coupling calculations we also used up to $L=3$, and the errors is clearly within 10% of the direct numerical solution results. There are other single-centered calculations up to $L=6$ [22], but the total ionization cross sections obtained differ little from the ones we have reported here. In Fig. 5 the experimental ionization cross sections [21] have been plotted also but the data is available only at the higher energies with large errors.

As a side product, we also report the excitation cross sections to $1s2s(^1S^e)$ and $1s2p(^1P^o)$ states that are likely to be measured in future experiments. These data can be checked by other theoretical calculations as well in the future. In Fig. 6 the cross sections are plotted. The excitation cross sections calculated with basis set 2 are also shown in crosses and asterisks, for the $1s2s(^1S^e)$ and $1s2p(^1P^o)$ states, respectively. They agree well with those obtained from basis set 1. Note that these cross sections are smaller than the total ionization cross sections by a factor of 5 to 10.

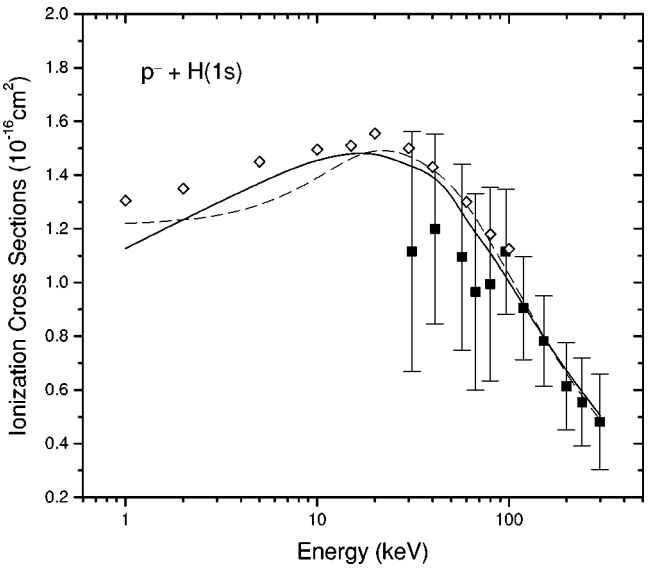

FIG. 5. Total ionization cross sections for antiprotons colliding with atomic hydrogen. Solid line: present single-center close-coupling calculation; dashed lines: single-center calculation of Hall *et al.* [19]; diamonds: direct integration of Schrödinger equation results of Wells *et al.* [20]; solid squares: the experimental results of Knudsen *et al.* [21].

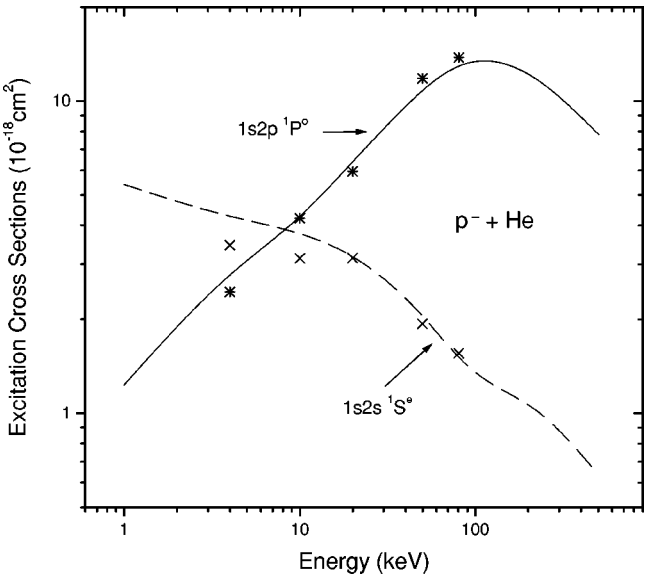

FIG. 6. Single-excitation cross sections to $1s2s$ $(^1S^e)$ and $1s2p$ $(^1P^o)$ states for antiprotons colliding with He from the present close-coupling calculations. Dashed line: $1s2s$ $(^1S^e)$ results with basis set 1; solid line: $1s2p$ $(^1P^o)$ results with basis set 1; crosses: $1s2s$ $(^1S^e)$ results with basis set 2; stars: $1s2p$ $(^1P^o)$ results with set 2.

IV. SUMMARY

In summary, we have performed a detailed calculation on the total ionization cross sections of He by antiprotons using two-electron basis functions in a close-coupling expansion method. Results from two different basis sets are shown to support the accuracy of the cross sections presented. While general agreement between theory and experiment is achieved for collision energies above 50 keV, we believe that deviations below 40 keV are likely due to the previous experimental difficulties. We have made the case to rule out that the discrepancy is due to approximations in the theoretical calculations and concluded that ionization cross sections of He by antiprotons below 40 keV should be remeasured again when the new antiproton beams become available in the next few years.

ACKNOWLEDGMENTS

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[1] L. H. Andersen *et al.*, Phys. Rev. Lett. **57**, 2147 (1986).
[2] H. Knudsen and J. F. Reading, Phys. Rep. **212**, 203 (1992).
[3] M. Kimura and M. Inokuti, Phys. Rev. A **38**, 3801 (1988).
[4] P. Hvelplund *et al.*, J. Phys. B **27**, 925 (1994).
[5] L. H. Andersen *et al.*, Phys. Rev. A **41**, 6536 (1990).
[6] P. D. Fainstein *et al.*, J. Phys. B **21**, 2989 (1988).
[7] W. Fritsch and C. D. Lin, J. Phys. B **19**, 2683 (1988).
[8] W. Fritsch and C. D. Lin, Phys. Rep. **202**, 1 (1991).
[9] C. Moore, *Atomic Energy Levels*, Natl. Bur. Stand. (U.S.) Circ. No. 467 (U.S. GPO, Washington, D.C., 1974), Vol. I.
[10] J. F. Reading *et al.*, J. Phys. B **30**, L189 (1997).
[11] L. A. Wehrman, A. L. Ford, and J. F. Reading, J. Phys. B **29**, 5831 (1996).
[12] G. Schiwietz *et al.*, J. Phys. B **29**, 307 (1996).
[13] G. Bent, P. S. Krstic, and D. R. Schultz, J. Chem. Phys. **108**, 1459 (1998).
[14] S. Y. Ovchinnikov and E. A. Solov'ev, Comments At. Mol. Phys. **22**, 69 (1988).
[15] R. K. Janev and P. S. Krstic, Phys. Rev. A **44**, R1435 (1991).
[16] M. Pieksma and S. Y. Ovchinnikov, J. Phys. B **27**, 4573 (1994).
[17] Y. Yamazaki (private communications).
[18] H. Knudsen (private communications).
[19] K. A. Hall *et al.*, J. Phys. B **29**, 6123 (1996).
[20] J. C. Wells *et al.*, Phys. Rev. A **54**, 593 (1996).
[21] H. Knudsen *et al.*, Phys. Rev. Lett. **74**, 4627 (1995).
[22] A. Igarashi (unpublished).