Calculations of Q values in single- and double-charge-transfer collisions of highly charged ions with atoms

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Close-coupling calculations are carried out for the Q values for electron capture processes in collisions of multiply charged ions with atoms over a broad range of energies. For single-capture processes the results for N^{7+} + He and O^{8+} + He collisions are in good agreement with the experimental data of Wu *et al.* [preceding paper, Phys. Rev. A **50**, 502 (1994)]. To compare with the experimental Q values for the transfer ionization (TI) and the true double-capture (TDC) processes, an independent-electron model was used to calculate double-electron-capture cross sections. By combining with the calculated average fluorescence yields, the theoretical Q values for TI and TDC processes are also found to be in fair agreement with the experimental data. We also compared the Q values calculated by the close-coupling method and by the classical-trajectory Monte Carlo method.

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I. INTRODUCTION

When a multiply charged ion collides with a neutral target atom or molecule in the keV/amu energy region, the dominant electron removal process is electron capture. The change in electronic energy in such reactions, called the Q value, for such a process is a direct measure of the distribution of final states populated on the projectile. Direct measurement of the energy gain (or loss) of the projectile has been used to determine this final state distribution [1,2]. However, as limited by the energy resolution, the energy gain spectroscopy is restricted to low energy projectiles and low-charge ions. For higher energies, measurement of Q value by the direct method becomes rather difficult. In recent years, it has been shown that recoil ion longitudinal momentum can yield direct information on the Q value. In a series of experiments, this technique has been used to determine the Q value of a number of ion-atom collision systems over a broad energy range [3,4].

In a typical collision between a multiply charged ion and atom at low energies, the electron-capture process is relatively selective in that only a small number of final states are populated at the end of the collision. The experimentally determined Q value is then a direct measure of the energy gain of the dominant final states populated. As the collision energy increases, the number of final states populated in the collision increases and the experimental Q value is the weighted average of the energy gain of this final-state distribution. The Q-value measurement, despite of failing to determine individual final states, does provide an alternative check on the theoretical prediction on the final-state distribution.

Among the theoretical models for describing the collisions between multiply charged ions and simple atoms, the close-coupling expansion method has been shown to provide the most detailed and reliable predictions [5,6]. In favorable situations where the number of final states populated is limited, the close-coupling calculations based on the two-center atomic orbitals have been shown to explain the very details of the final-state distributions measured by energy gain spectroscopy or by the photon spectroscopy. However, these measurements provide little information about the contribution of weaker final states which have smaller cross sections. The Q-value measurement, by the nature that it is an energy moment, provides additional check on the theoretical prediction of the final-state distribution. In this paper, we calculate the Q value for a number of collision systems between multiply charged ions and atoms using the two-center atomic orbital expansion method. We first calculate the Q values for the collisions of a number of bare multiply charged ions with atoms with atomic hydrogen using the close-coupling expansion method and compare the results with those obtained by the classical trajectory Monte Carlo method to establish the validity of the latter simpler calculations. We then examine in more details the Q values for the collisions between bare oxygen and bare nitrogen ions with helium atoms where the Q values have been determined for the single- and double-electron-capture processes experimentally by Wu *et al.* [3] in the preceding paper.

Section II provides a brief description of the parameters used in the close-coupling calculation. The results are presented in Sec. III where comparison with experimental data is also given. A brief summary is given in Sec. IV.

II. THEORETICAL METHODS

A. Single-electron-capture processes

The close-coupling methods where the wave functions are expanded in terms of atomic orbitals on the two collision centers has been well documented [5]. In the present calculations, the number of atomic orbitals used in the calculations has been substantially increased to account for the population of the weaker final states.

The Q value is defined as the weighted average of energy gain of each populated state,

$$Q = \frac{\sum_i \sigma_i (E_i - E_0)}{\sum_i \sigma_i}$$

where σ_i is the cross section of populating a specific final state, E_i is the total electronic energy of the final state and E_0 is the initial electronic energy. The sum is over all the final states. As will be shown later, the Q value is sensitive to the distribution of the higher Rydberg states populated for collisions at higher energies. In reality, these higher Rydberg states cannot be included explicitly in the close-coupling calculation. For each collision system at a given collision energy, there is a particular final state with principal quantum number n where the electron capture cross section is maximum. In the close-coupling calculation, we make sure that atomic orbitals with principal quantum numbers $(n+1)$ and $(n+2)$ are included in the basis set. The electron capture cross sections for higher n's are then obtained by assuming the "standard" $1/n^3$ rule, from which the Q value is calculated by summing over all the final Rydberg states.

The close-coupling calculations cannot be extended to collisions involving highly charged ions since the number of basis states that need to be included in the calculation becomes rather large. There are few quantum mechanical methods to account for the final-state n distributions, especially at higher collision energies. An alternative model is the straightforward classical-trajectory Monte Carlo method (CTMC). The CTMC has been shown to be able to give a rough description ion-atom collisions at higher energies, but it is not clear whether it can give a reasonable description of the Q values. To this end, we performed some calculations for the simple one-electron collision systems involving bare ions with atomic hydrogen. By comparing the results from the CTMC and those from the close-coupling method, we explore the range of the validity of the CTMC method for the Q value predictions. We used only the standard CTMC where the initial state is sampled by the microcannonical ensemble and the final electron-capture state is not quantized artificially. We note that the final CTMC result can depend somewhat on how the sampling is done, but there is no reason to favor one over the other.

B. Double-electron-capture processes

We also examine the Q values for the double-electron-capture processes. Experimentally the Q values were determined for the so-called transfer ionization (TI) process and the true double-capture (TDC) process. Both processes, in the collision energy range considered here, are actually due to the double-electron-capture process to doubly excited states. They are distinguished by the decay channel of the final states. If the doubly excited states decay by autoionization, they are registered as TI, and if these states decay by radiative stablization, they are registered as TDC.

To obtain the Q values for such two electron processes from *ab initio* calculations is obviously a formidable task. First one needs to calculate the cross sections for double capture to individual doubly excited states where the density of states in a given manifold (n, n') is quite large, here n and n' refer to the principal quantum numbers of each of the electrons. Second, the fluorescence yield of each doubly excited state has to be calculated. Only after that one can calculate the Q values for the TI and the TDC processes and compare the results with experiment.

To simplify the calculations, we obtain double-electron-capture cross sections using the independent electron model. Instead of calculating double-electron-capture cross sections to individual doubly excited states which had been done for a few collision systems previously [8–10], we calculate the double-electron-capture cross section to each (n, n') manifold directly. For example, in the collision between O^{8+} on He, we treat the first electron in He to be as a hydrogenlike system with an effective charge $Z = 1.43$. The probabilities $P_n(b)$ for populating the different n levels are calculated for each impact parameter b. In the independent-electron model, we treat the second electron in He as a hydrogenlike system with charge $Z = 2$. The electron-capture probability $P_{n'}(b)$ to each n' level from this second capture are also calculated using the close-coupling method. The double-electron-capture cross section to each (n, n') manifold is then obtained by integrating the products $P_n(b)P_{n'}(b)$ over the impact parameter plane.

To obtain the fraction of the doubly excited states in each manifold that will decay by radiative or by electron emission processes, we carried out the fluorescence yield calculations for all the states within each manifold. Such calculations are tedious but are straightforward. We then calculate an average fluorescence yield for each manifold (n, n') by assuming that the doubly excited states are statistically populated in each manifold. This latter assumption is known to be incorrect, but this procedure still allows us to obtain the major trend of the fluorescence yield with respect to the values of n and n'. In terms of the average fluorescence yield we can then calculate the "cross sections" for TI and TDC, respectively, for each (n, n') manifold, and the cross sections can be extrapolated to higher n' using the $1/n'^3$ rule for a given n.

III. RESULTS AND DISCUSSION

A. Q-value calculations for charge transfer in He^{2+}, B^{5+}, and O^{8+} on H

We have calculated the Q values for the collision of bare ions with atomic hydrogen atom. Figure 1 shows

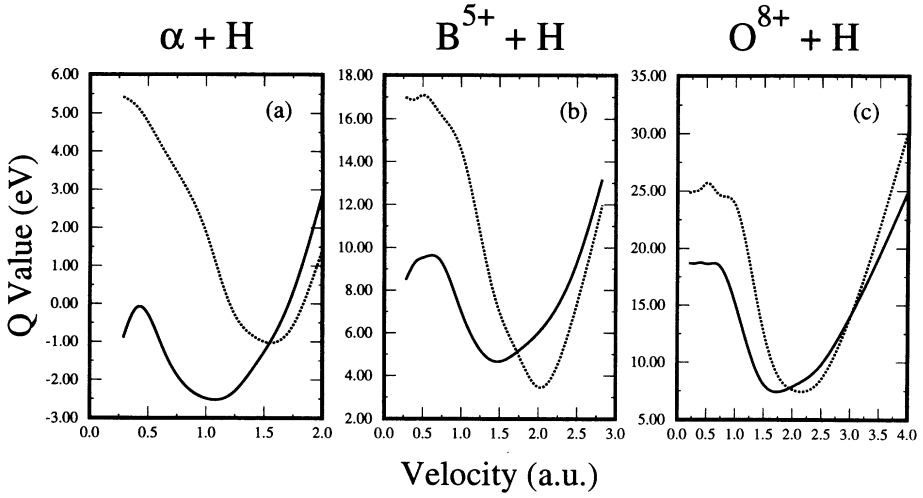

FIG. 1. The Q values calculated for electron-capture processes for three different collision systems: (a) α, (b) B^{5+}, and (c) O^{8+} on H. Solid lines: close-coupling calculation; dotted lines: classical-trajectory Monte Carlo (CTMC) calculation.

the Q values calculated for electron-capture processes in α + H, B^{5+} + H, and O^{8+} + H collisions. The solid lines are results from the close-coupling calculations and the dotted lines are from the classical-trajectory Monte Carlo (CTMC) method. The CTMC calculations were carried out to check its region of validity for estimating the Q values for a given collision. Both calculations give the same general shape for the Q values vs collision velocity though the absolute values are somewhat different. The relative difference is smaller at higher v and for higher incident charge. The difference is quite large at small v, especially for small Z incident ions. For low-charge projectiles, say α particles, the dominant capture channels are to the low n excited states. These quantized low n states are not well described by classical mechanics and that the density of states is also smaller. When the charge of the projectile is increased, electron is captured to high n states where the density of states is larger and that the classical description of the electronic motion is more valid such that deviation of the CTMC from quantal results is smaller.

The behavior of the Q values vs collision energy can be understood qualitatively. We note that in general the Q values decrease first with increasing collision velocity. At low collision energies, charge transfer is very selective and only a few n states are populated. As the collision energy increases, more states are populated and cross sections for populating other higher n states also increase. Since higher n states have larger density of states and correspond to lower Q value, this results in a decrease in the Q value. As the collision energy is increased further, the population of smaller n begins to dominate and the Q value becomes increasing with collision energies. One also notices that the Q value weights the low n states more than the high n states. Thus, the measurement of Q values provides a sensitive test of the n distribution of the final states populated in the charge-transfer collisions.

In carrying out the close-coupling calculations, the atomic orbitals included on the projectile are hydrogenlike states with principal quantum numbers $n = 1 \sim 6$ and the target atomic orbitals included are $n = 1, 2$ hydrogenic orbitals. The large basis set is needed so that the total electron-capture cross sections to each n manifold can be evaluated. We make sure that the cross sections for the highest two n's decrease with n so that cross sections for higher n's can be obtained by extrapolation using the $1/n^3$ rule. As will be shown in the next section, the contribution to the Q value from the higher n's is not negligible for collisions at higher energies even though the cross sections to individual n is not large. A more quantitative illustration of this procedure will be given in the next section.

In carrying out the CTMC calculations for the Q values, we used microcanonical distributions for the initial state, and the energy of the electron after capture is not quantized artificially.

B. Single capture for N^{7+} and O^{8+} on He

We have calculated the Q values for single-electron-capture processes for N^{7+} and O^{8+} collisions with He using the close-coupling method. We approximate the helium atom as a one-electron atom with an effective charge Z=1.43 so it gives the correct single ionization energy. Atomic orbitals including up to $n = 6$ are included in the close-coupling calculations and the cross sections for the higher n's are obtained by extrapolation using the procedure discussed in the previous section. The calculated Q values are compared to the experimental results of Wu *et al.* In Fig. 2 we compared the calculated Q values with the experimental data for O^{8+} and it is clear that the agreement is rather satisfactory. To understand the calculated results, we show in Fig. 2(a) the energy dependence of the electron-capture cross sections to different n's. In the energy range considered in this work, $n = 4$ is the dominant channel at low energies, and the the $n = 5$ and $n = 6$ states become relatively more important as the collision energy increases. This energy dependence of the n distributions is responsible for the decrease of Q values with increasing collision energies because more high n states correspond to lower Q values.

We have also calculated the Q values for the single-electron-capture processes in N^{7+}-He collisions. The cal-

culated results are compared with the experiment of Wu *et al.* in Fig. 3(b). In this figure two theoretical curves are shown to illustrate the importance of the contribution from the higher n's. The dashed curve is the calculated Q value if the contribution from states greater than $n = 6$ is neglected. Only when these higher states are included in the calculation of the Q value can one obtain results in agreement with experiment. We comment that at higher energies the cross sections for higher n, while still assumed to have the $1/n^3$ dependence, does decrease slower at higher energies. To illustrate the relative importance of the different n levels populated as the collision energy is varied, we show in Fig. 4 the calculated single-electron-capture cross sections to $n = 2 \sim 6$ at two energies, (a) 25 keV/amu and (b) 75 keV/amu. Figure 4(b) clearly indicates that it is not advisable to include up to $n = 6$ only in calculating the Q values.

In comparing the results for O^{8+} with those for N^{7+}, we note that the Q value for the latter system increases with energy initially (around 10 keV) before it decreases with increasing energies. This is simply understood from the behavior of the cross section for capture to the $n = 3$ states, Fig. 3(a). We note that this cross section increases with energies in the 5–20 keV region as the cross section for capture to the dominant $n = 4$ states decreases. Thus the Q value measurement reflects the relative importance of the cross sections to the different n levels.

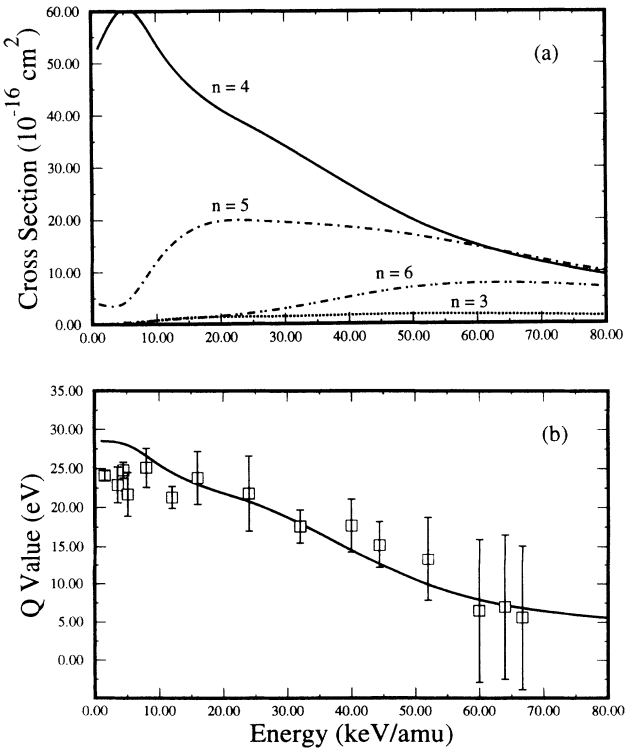

FIG. 2. (a) Single-electron-capture cross sections to individual final n states of O^{7+} for O^{8+} + He collisions. (b) The Q values calculated (solid line) for the same collision system. Symbols are experimental measurements [3].

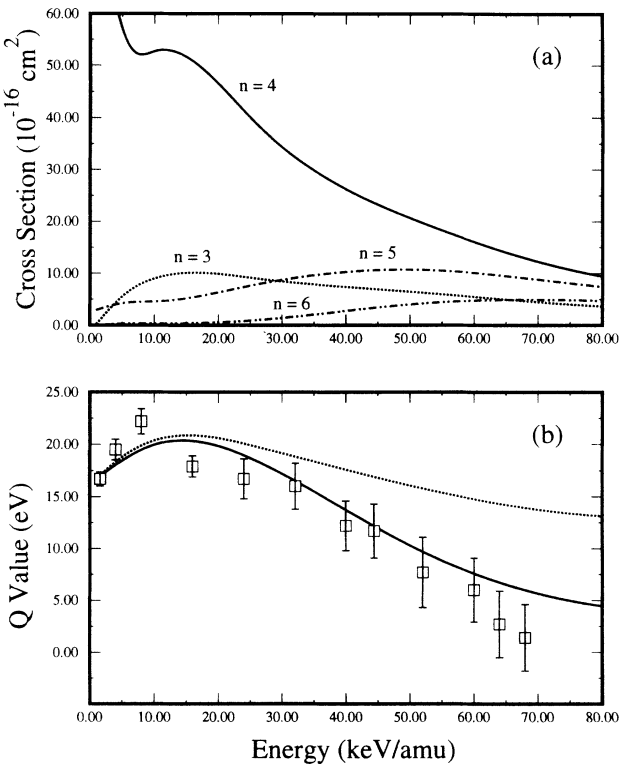

FIG. 3. Same as Fig. 2 but for N^{7+} + He collision. The dotted line in (b) is the Q value calculated by including single capture only up to $n = 6$ states.

C. Q-value calculation for TI and TDC processes

We next consider processes involving the capture of two electrons from helium by O^{8+} and N^{7+} ions. In the energy range up to 80 keV/amu, double-capture process populates doubly excited states which can stablize by either electron emission or by radiative process and the results are measured as TI and TDC processes, respectively.

Using the independent-electron approximation, the calculated double-electron-capture cross sections to each

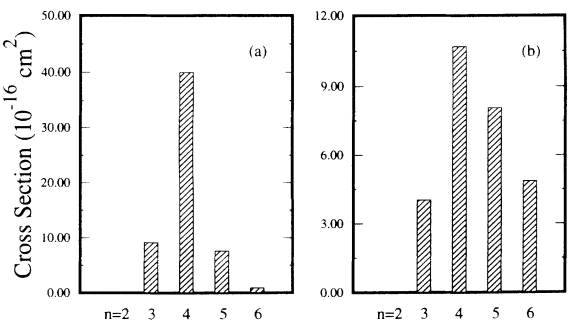

FIG. 4. The n dependence of single-electron-capture cross sections for N^{7+} + He collisions at 25 and 75 keV/amu collision energies.

of the dominant (n,n') manifolds are shown in Fig. 5 for N^{7+} on He. The dominant manifolds populated by double capture at 25 keV/amu are the $(3,3)$ and $(3,4)$ manifolds, with the latter being the larger one. At the higher 75 keV/amu, the $(2,n)$, $(3,n)$, and $(4,n)$ manifolds are also populated and the other higher manifolds are found to be still negligible.

To separate the Q values for TI and TDC processes, we need to calculate the fluorescence yield which is rather state dependent. Fortunately, we have existing programs which can be easily used to calculate the fluorescence yields of two-electron doubly excited states [7]. There is no need to tabulate the results for each individual states calculated. In Fig. 6 we show in graphical form the fluorescence yields for states in the $(2,n)$ and $(3,n)$ manifolds for N^{5+}. The energy range of states within each manifold is indicated by a horizontal bar. Note that the typical fluorescence yields increases rapidly with increasing n for the both the $(2,n)$ sequence and the $(3,n)$ sequence. For the $(2,2)$ and the $(3,3)$ manifolds the contribution to the TDC is negligible. For the other manifolds, we calculate an average fluorescence yield for each manifold by assuming that each state has equal population. This is by far from correct since it is known in general that the double-capture cross sections are rather state selective [8–14]. In view of the lack of such information from actual calculations for each manifold or from experiment, we calculate the average fluorescence yield for each manifold and the results are shown in Table I.

Figure 6 also indicates that the range of energies for states within each manifold extends over a large interval, especially for states within the (n,n') manifolds where n and n' are small. We do not use the average energy for states within each manifold. Instead, we use the energy levels of states which are known to be populated significantly in the collision. Based on previous studies and experimental data [11–14], for example, we use the $^1F^o$ and $^1G^e$ energy levels for the $(3,3)$ and $(3,4)$ manifolds where these states are known to be predominantly populated. For the higher n's in the $(2,n)$ and the $(3,n)$ manifolds the states are all in a narrower energy range and there is no need to make additional assumptions. The $(2,2)$ manifold is hardly populated and thus there is no need to be concerned with the energies of the states there.

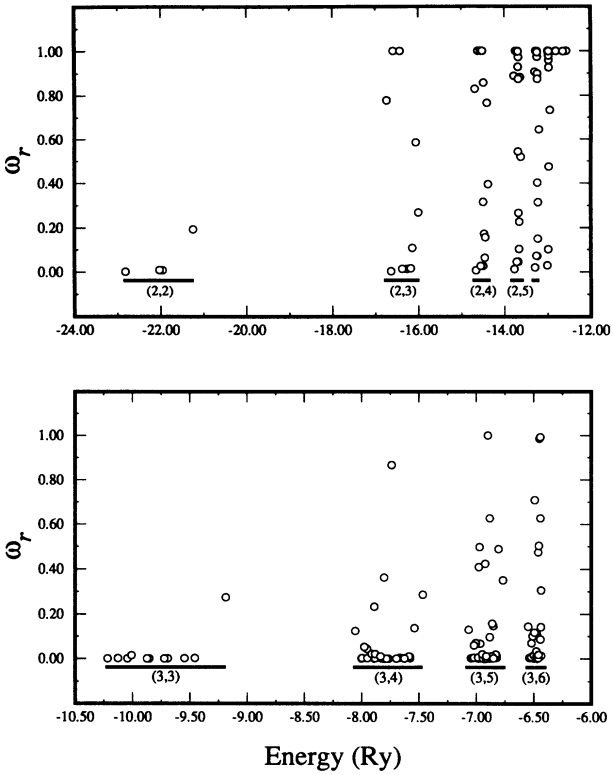

FIG. 6. Fluorescence yields for the $(2,n)$ and $(3,n)$ doubly excited states of N^{5+}. Each circle represents the fluorescence yield of one doubly excited state and is arranged according to its binding energy. States within each manifold are grouped by a horizontal bar.

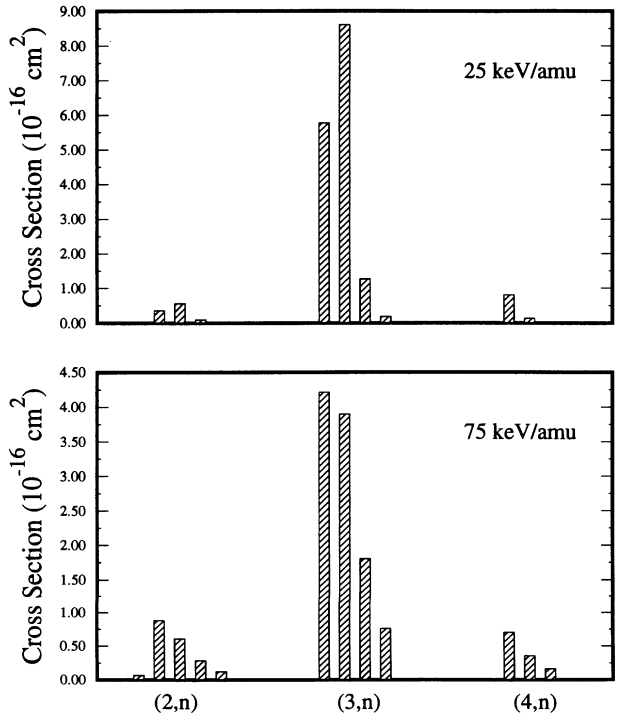

FIG. 5. Double-electron-capture cross section for N^{7+} + He at 25 and 75 keV/amu collision energies.

TABLE I. Average fluorescence yields for doubly excited states of the $(2,n)$ and $(3,n)$ manifolds for N^{5+}. The averaged fluorescence yield for each manifold is obtained from the calculated fluorescence yields of doubly excited states within the manifold by assuming that states are statistically populated in the double-capture process.

States	ω_r	States	ω_r
$(2,2)$	0.053		
$(2,3)$	0.378	$(3,3)$	0.033
$(2,4)$	0.472	$(3,4)$	0.079
$(2,5)$	0.606	$(3,5)$	0.133
$(2,6)$	0.720	$(3,6)$	0.204

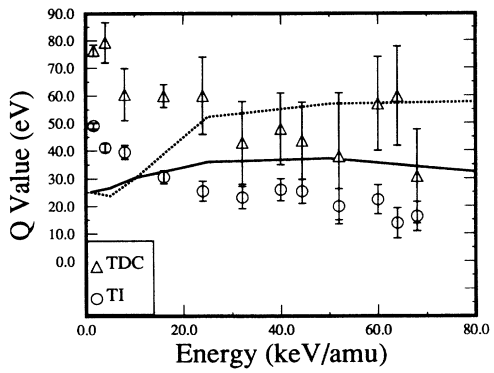

FIG. 7. Comparison of Q values for TI and TDC processes for the N^{7+} + He collision. Solid line: theoretical Q values for TI; dotted line: theoretical Q values for TDC.

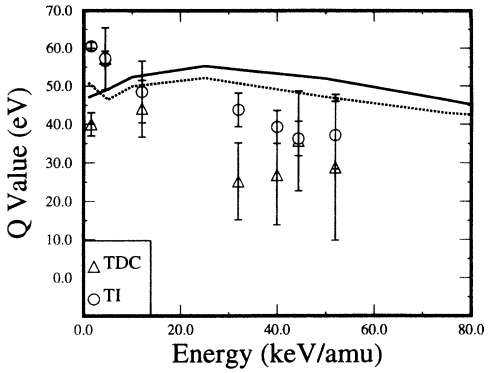

FIG. 9. Q values for double-charge transfer for O^{8+} + He collision. The theoretical calculations (curves) do not distinguish TI and TDC. Solid line: all captured states included; dotted line: the $(2,n)$ manifold is excluded in the calculation of Q values.

Under these three assumptions we have calculated the Q values for both the TI and the TDC processes. The calculated results are compared to the experimental data from Wu *et al.* in Fig. 7. The agreement is rather poor at low energies, but quite good at larger energies. The large discrepancy at low energies can be attributed mostly to the failure of the independent-electron model for calculating double-electron-capture cross sections. It is known experimentally that at the low energy range of Fig. 7 the dominant double-capture populates the $(3,3)$ manifold, but our calculation predicts that the $(3,4)$ manifold is populated predominantly. Thus our predicted Q values for both TI and TDC are too small.

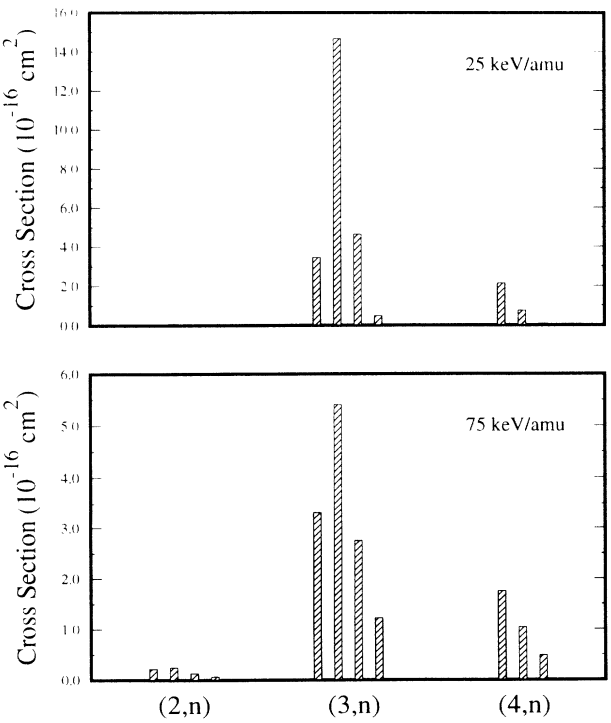

FIG. 8. Double-electron-capture cross section for O^{8+} + He at 25 and 75 keV/amu collision energies, showing the dependence of cross sections vs n for the $(2,n)$, $(3,n)$, and $(4,n)$ manifolds.

We have also calculated the Q values for the TI and the TDC processes in the collisions of O^{8+} with He. Figure 8 shows the calculated double-electron-capture cross sections to individual (n,n') manifolds. The dominant double-capture manifolds are the $(3,n')$, with $n' = 3,4$. By comparing with Fig. 5, we note that the cross sections for populating the $(2,n')$ $(n' > 2)$ are quite small.

To calculate the Q values, we assumed that the average fluorescence yield for each $(3,n')$ and $(4,n')$ manifold is the same for O^{6+} and for N^{5+} and thus the results calculated from Table I are used to calculate the Q values for the TI and TDC processes. The average energy of doubly excited states within each manifold is also assumed to be from the dominant channels. This assumption is in agreement with the experimental observation [11–13] for the $(3,3)$ manifold which has been measured using electron spectroscopy.

The calculated Q values for the TI and TDC processes are shown in Fig. 9. The solid line is the theoretical calculation for TI and the dotted line for TDC. Since the $(2,n)$ manifolds are not populated by the primary double-capture process, they are not included in the calculation. The Q values are mainly determined by the $(3,n')$ and $(4,n')$ manifolds and thus the Q values are smaller for O^{8+} projectiles than for N^{7+} projectiles. Furthermore, the fluorescence yields for the dominant $(3,n')$ and $(4,n')$ channels do not vary much and thus the Q values for the TI and TDC processes are not very different. Because TDC is process is much weaker than TI, the TDC is more sensitive to the $(2,n)$ population. At higher collision energies, the TDC Q values become larger when $(2,n)$ is included, But TI Q values remain the same.

IV. SUMMARY AND CONCLUSIONS

In this paper, we presented the Q values for charge-transfer processes for collisions between multiply charged ions with atoms. We first compared the Q values using

electron-capture cross sections calculated from the close-coupling method and compared the results with those where the electron-capture cross sections are calculated from the classical-trajectory Monte Carlo method. These calculations were carried out for the collisions between bare ions and atomic hydrogen. We showed that the Q values from the CTMC calculations do not agree in general with those obtained from the close-coupling calculations. But agreement improves with higher charge ions. For Q value calculations of very highly charged ions colliding with atoms where the close-coupling calculations are not desirable due to large number states, one may expect the CTMC method to give reasonable results. It is also established that electron capture to the higher excited states ($n > 6$) contributes significantly to the calculated Q values for collisions at higher energies.

We also have examined the Q values for single-electron capture and the transfer ionization and true double-capture processes for collisions between N^{7+} and O^{8+} on He. We used an independent-electron model to calculate single and double-electron-capture cross sections. The fluorescence yields for the doubly excited states are also calculated such that the Q values for the TI and TDC are extracted. The calculated results are compared with the experimental results of Wu *et al.* For single-electron-capture processes, the agreement is very good. For the TI and TDC processes, there is also general agreement except at low energies where we attributed that the discrepancy is due to the limitation of the independent-electron approximation. We also interpreted the large Q values for the TDC in N^{7+} on He collisions are due to the relatively larger population of the $(2, n')$ manifolds.

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