Excitation of helium by protons and alpha particles

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Cross sections for the $1\,^1S \rightarrow 4\,^1S$ excitation of He under proton and α-particle impact are calculated with incident energy ranging from 10 to 5000 keV using the two-state distortion approximation and a corresponding second-Born approximation. Two sets of wave functions are used. The results are compared with the existing first-Born calculations and the experimental data. The generalized oscillator strengths for the transition are also presented. The nonorthogonality of the wave functions has a very marked effect on the cross sections for almost the entire energy range considered. For proton impact, use of a properly orthogonal set of wave functions reduces the first-Born cross-section values obtained from an improperly orthogonalized set of wave functions by a factor of almost 2 at high energies, in agreement with the experimental values. The second-Born and distortion approximations give better agreement at lower energies also and the latter correctly predicts the functional dependence of cross sections on energy down to an incident energy of 20 keV. The necessity of using properly orthogonal sets of wave functions in the calculation of scattering processes involving high atomic excitations is thereby stressed.

I. INTRODUCTION

Direct collisional formation of highly excited states of He by ionic impact is a topic on which considerable experimental interest has been concentrated so far, but relatively few theoretical studies are available. One reason behind this situation appears to be the nonavailability of accurate as well as simple wave functions for these states, so as to be useful in scattering calculations. Inaccuracies like nonorthogonality in the set of wave functions chosen, affect considerably the absolute values of the theoretical scattering cross sections, especially for the high excitations. However, a properly orthogonal set of one-parameter wave functions for some of the higher states of He has become available recently,[1] and comparison between theory and experiment may now be of interest.

The two-state distortion approximation of Bates[2] in the impact parameter formulation has been demonstrated to give good results for ion-impact excitation of He in the intermediate energy region, where the first-Born approximation fails to be sufficiently accurate. The results of Bell[3] and Davison[4] for ^1P and ^1D excitation by proton impact have shown good agreement with experiment at intermediate energies. Distortion is found to be more important for the higher states than the lower. However, calculations of Bates[2,5] on atomic hydrogen reveal that the effect of distortion is much more important for s-s transitions than for s-p ones. For proton-impact $3\,^1S$ excitation of He, the distortion calculation of Roy and Mukherjee[6] gives results nearly the same as experiment at low energies. In view of the above success, application of the distortion approximation to study higher ^1S-state excitations appears worthwhile.

Another method, which is expected to give results better than the first-Born approximation at intermediate energies, is the second-Born approximation. However, here the expression for amplitude involves an infinite summation over all the states of the target atom. Only under suitable approximations retaining a few intermediate states does this method become tractable. The distortion effects in the second-Born calculation can be taken into consideration by retaining couplings only to the initial and the final states in the infinite summation. Kingston *et al.*[7] and Chaudhuri and Bhattacharya[8] have applied the second-Born approximation in this form to study transitions in atomic hydrogen by the impact of protons and α particles, respectively. They obtained good agreement with the results of the distortion approximation. However, further inclusion of polarization effects in the calculation of Kingston *et al.*,[7] by retaining the intermediate $2p$ state, gave cross-section values close to the first-Born results. Hence, it may be useful to extend the second-Born approximation for the case of two-electron atoms, retaining couplings only to the initial and the final states, and to compare the cross-section results with those given by the two-state distortion approximation.

In the present investigation, we propose to apply the two-state distortion and the corresponding second-Born approximation to calculate the $4\,^1S$ excitation cross section of He under proton and α-particle impact. For proton-impact excitation of this state, there exist two theoretical calculations due to Van den Bos[9] and Oldham,[10] both in the first-Born approximation. Van den Bos[9] has used a single-parameter wave function[11] for the excited state of He that is not properly orthogonal to the ground state used. His result overestimates the observed cross-section values by a factor of almost 2 at high energies. Oldham,[10] on the other hand, has employed highly accurate many-parameter wave functions including electron correlations. His values are in much better absolute agreement with the experimental data in the intermediate-energy region than those of Van den Bos.[9] But again, there is some appreciable overestimation in the high-energy region. An estimation due to Gaillard,[12] is also available at high energies for this excitation cross section. He has used a scaling formula due to Bates and Griffing[13] to convert electron excitation data to the proton case. The scaled results agree with experiment, but exhibit an incorrect asymptotic high-energy behavior.

Experimental measurements of the proton (or deuteron) impact $4\,^1S$ excitation cross section of atomic helium have been performed by a number of groups.[14-19]

The wave function we use for the excited $4\,^1S$ state of He belongs to a properly orthogonal set given recently by Winter and Lin[1] (WL). In addition to this, we use also the improperly orthogonalized wave function due to Van den Bos[11] (V). On comparing the two sets of results, we can make an estimate of the effect of nonorthogonality of the wave function on the present cross-section calculations.

Furthermore, as observed earlier,[20,21] important information regarding the accuracy of any Born-approximation result, as well as its sensitivity on the target wave functions used, may be obtained from the generalized oscillator strength (GOS) of the transition concerned. In the present paper, we calculate also the GOS of the $1\,^1S \rightarrow 4\,^1S$ transition in He using the WL wave functions, and compare the results with those obtained from other wave functions. Atomic units will be used throughout the present work.

II. THEORY

The incident structureless ion of charge Z_i is assumed to follow a classical straight-line trajectory with a velocity $\vec{v}$ relative to the target nucleus. The time-dependent Schrödinger equation for the bound system is

$$H\Psi(\vec{r}_1, \vec{r}_2, t) = i\frac{\partial}{\partial t}\Psi(\vec{r}_1, \vec{r}_2, t)\,, \tag{1}$$

where Ψ is the total atomic wave function and $\vec{r}_j$ is the position vector of the jth bound electron with respect to the target nucleus. The total Hamiltonian H is given by

$$H(\vec{R}, \vec{r}_1, \vec{r}_2) = H_0(\vec{r}_1, \vec{r}_2) + V(\vec{R}, \vec{r}_1, \vec{r}_2)\,,$$

H_0 being the unperturbed Hamiltonian of atomic helium and V the interaction potential, given by

$$V(\vec{R}, \vec{r}_1, \vec{r}_2) = \frac{2Z_i}{R} - 2\sum_{j=1}^{2}\frac{1}{|\vec{R}+\vec{r}_j|}\,, \tag{2}$$

where $\vec{R}$ is the vector from the projectile to the helium nucleus.

Expanding Ψ in terms of the bound eigenfunctions $\phi_n(\vec{r}_1, \vec{r}_2, t)$ of atomic helium as

$$\Psi(\vec{r}_1, \vec{r}_2, t) = \sum_n A_n(t)\phi_n(\vec{r}_1, \vec{r}_2, t)$$

and proceeding in the usual manner, we obtain the following set of coupled differential equations:

$$\frac{dA_m(s)}{ds} = \frac{i}{v}\sum_n A_n(s)F_{mn}(s)\,, \tag{3}$$

where

$$s = vt\,,$$

$$F_{mn} = -\langle\phi_m|V|\phi_n\rangle = V_{mn}\exp[i\epsilon_{mn}s/v]\,,$$

$$\epsilon_{mn} = \epsilon_m - \epsilon_n\,,$$

ϵ_m and ϵ_n being the eigenenergies corresponding to ϕ_m and ϕ_n.

The set of equations (3) is solved for $A_m(s)$ with the initial conditions $A_m(-\infty) = \delta_{m1}$, whence the probability of excitation from the ground state 1 to state m is given by

$$P_m = |A_m(\infty)|^2\,.$$

Formal integration of (3) yields

$$A_m(s) = \delta_{m1} + \frac{i}{v}\sum_n \int_{-\infty}^{s} A_n(s')F_{mn}(s')ds'\,. \tag{4}$$

The first-Born approximation corresponds to the substitution $A_m(s) = A_m(-\infty) = \delta_{m1}$ on the right-hand side of Eq. (4) yielding the first-Born amplitude

$$A_m^B(\infty) = \delta_{m1} + \frac{i}{v}\int_{-\infty}^{\infty} F_{m1}(s)ds\,. \tag{5}$$

This expression is substituted on the right-hand side of (4) and all couplings, except those involving the initial and the final state ($n=1$ and m) in the infinite summation, are neglected. The resulting second-Born amplitude is given by

$$A_m^{B2}(\infty) = \frac{i}{v}\int_{-\infty}^{\infty} F_{m1}(s)ds - \frac{1}{v^2}\int_{-\infty}^{\infty}\left(F_{m1}(s)\int_{-\infty}^{s}F_{11}ds' + F_{mm}(s)\int_{-\infty}^{s}F_{m1}ds'\right)ds\,. \tag{6}$$

Alternatively, retaining coupling only to initial and final states in the original set of coupled equations (3), one obtains

$$\frac{dA_1}{ds}=\frac{i}{v}[A_1F_{11}+A_mF_{1m}] \tag{7a}$$

and

$$\frac{dA_m}{ds}=\frac{i}{v}[A_1F_{m1}+A_mF_{mm}]\,. \tag{7b}$$

Neglecting the back coupling term involving F_{1m} on the right-hand side of (7a) and solving, we obtain the two-state distortion amplitude

$$A_m^D(\infty)=\exp\left(\frac{i}{v}\int_{-\infty}^{\infty}F_{mm}(s)\,ds\right)\times\frac{i}{v}\int_{-\infty}^{\infty}A_1F_{m1}(s)\exp\left(-\frac{i}{v}\int_{-\infty}^{s}F_{mm}ds'\right)ds\,. \tag{8}$$

For ^1S excitations, all the $V_{mn}(s)$ involved are even in s, and on simplification one obtains the first-Born probability

$$P_m^B=(A_m^B)^2\,,$$

where

$$A_m^B=\frac{2}{v}\int_0^{\infty}V_{m1}\cos(\epsilon_{m1}s/v)\,ds\,; \tag{9}$$

the second-Born probability

$$P_m^{B2}=P_m^B-\frac{2}{v}A_m^BA_m'^{B2}\,,$$

where

$$A_m'^{B2}=\frac{2}{v}\int_0^{\infty}V_{m1}(s)\sin(\epsilon_{m1}s/v)\times\left(\int_0^{s}(F_{11}-F_{mm})\,ds'\right)ds \tag{10}$$

(neglecting terms of the fourth order in interaction energy); one also obtains the distortion probability

$$P_m^D=(A_m^D)^2\,,$$

where

$$A_m^D=\frac{2}{v}\int_0^{\infty}V_{m1}(s)\cos\left[\frac{1}{v}\left(\int_0^{s}(F_{11}-F_{mm})ds'+\epsilon_{m1}s\right)\right]ds\,. \tag{11}$$

The double integrals are evaluated numerically; hence the total cross section for excitation from the ground state to the mth state is given by

$$\sigma_m=2\pi\int_0^{\infty}P_m p\,dp\,, \tag{12}$$

p being the impact parameter.

III. WAVE FUNCTION

For both V and WL wave functions, the ground states are of the product form:

$$\phi_1\equiv\phi(1\,^1S\,|r_1,r_2)=u(r_1)u(r_2)\,,$$

where

$$u(r)=N_0(e^{-\alpha r}+\eta e^{-\beta r})\,,$$

while the final-state wave functions are of the form

$$\phi_m\equiv\phi(4\,^1S\,|r_1,r_2)=\frac{N}{\sqrt{2}}[\psi(4s\,|r_1)\psi_0(z\,|r_2)+\psi(4s\,|r_2)\psi_0(z\,|r_1)]\,,$$

where $\psi_0(z\,|r)$ is the ground-state hydrogenlike orbital of a single electron in the field of a nuclear charge $z=2$ and $\psi(4s\,|r)$ is the excited-state orbital. N_0 and N are the normalization constants.

For V wave functions, the ground state is of Byron and Joachain,[22] while for WL wave functions, it is of Green *et al.*[23]

The function ϕ_m in case of V wave function is orthogonal only to the ground $1\,^1S$ state and has nonzero overlaps with the $2\,^1S$ and $3\,^1S$ states. For WL case, ϕ_m is orthogonal to all the lower states.

Generalized oscillator strength

The generalized oscillator strength $f_n(K)$ for the transition of a helium atom from its ground state (1) to the nth excited state with a momentum transfer $\vec{K}$ is defined as

$$f_n(K)=\frac{8\epsilon_{n1}}{K^2}\,|\langle\phi_n|e^{i\vec{K}\cdot\vec{r}}\,|\phi_1\rangle|^2\,, \tag{13}$$

where ϵ_{n1} $(=\epsilon_n-\epsilon_1)$ is the excitation energy from the ground state and $\vec{r}$ is the position vector of any bound electron.

IV. RESULTS AND DISCUSSION

In Fig. 1, we have plotted the present GOS values using the WL wave function against K^2. The results are compared with those of Oldham[10] and of Van den Bos.[9] Our WL results are closer to the results of Oldham[10] in comparison with those of Van den Bos.[9] This clearly demonstrates the superiority of the WL wave functions to the V wave functions.

Our results for the proton-impact $4\,^1S$ excitation cross section of He with the V and WL wave functions are displayed in Fig. 2. The theoretical results of Oldham[10] and Van den Bos[9] as well as the experimental data[14-19] are also included in Fig. 2. In Fig. 3, we give the present results for α-particle impact excitation cross section of He.

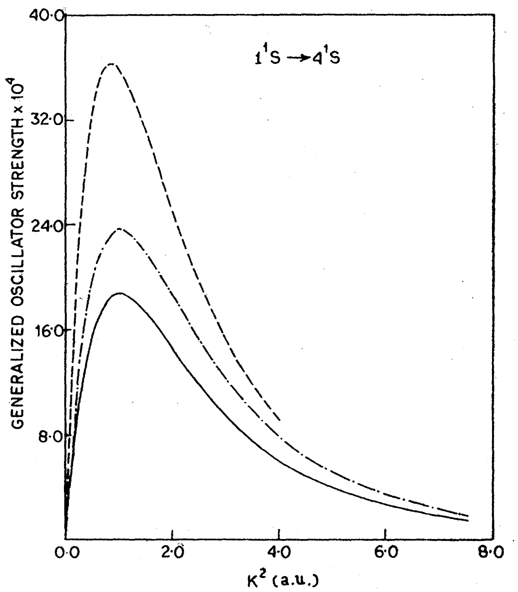

FIG. 1. Generalized oscillator strengths for the 4^1S excitation of helium: (——) present calculation; (–•–•–•) calculation of Oldham (Ref. 10); (– – –) calculation of Van den Bos (Ref. 9).

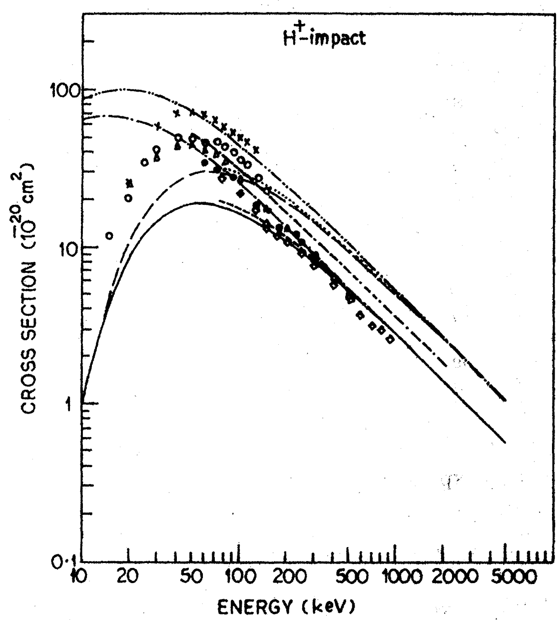

FIG. 2. Cross section for the 4^1S excitation of He atom by proton impact. Theory: (——) distortion approximation using WL wave function; (– – –) second-Born approximation using WL wave function; (–•–•–•) first-Born approximation using WL wave function; (— —) distortion approximation using V wave function; (• • • • •) second-Born approximation using V wave function; (—• • —• •) first-Born approximation using V wave function by Van den Bos (Ref. 9); (— —•— —•) first-Born approximation by Oldham (Ref. 10). Experiment: (X) Dodd and Hughes (Ref. 14); (●) Robinson and Gilbody (Ref. 15); (□) Thomas and Bent (Ref. 16); (Δ) Denis *et al.* (Ref. 17); (O) Van den Bos *et al.* (Ref. 18).

A. Proton-impact excitation

From Fig. 2, it can be seen that the present second-Born and distortion cross section using the V and WL wave functions approach the respective Born values in the high-energy limit, as is expected. For each wave function, the second-Born and distortion values are coincident above 500 keV. Marked difference between the cross sections using the V and WL wave functions is seen for almost the whole energy region covered. The second-Born and distortion WL results (i.e., our results with the WL wave function) agree with the experiments above 150 keV within the accuracy of measurements, while V results settle to values higher by a factor of 2 approximately. This is attributable to the lack of proper orthogonality of the V wave function.

The Born results of Oldham[10] show a good agreement with the experiments in the intermediate-energy region. But the agreement deteriorates at high energies. However, Oldham[10] has compared the slope of his theoretical curve with that obtained from the experimental data of Thomas and Bent[16] in a logarithmic plot of cross section against incident energy, as in Fig. 2. In the high-energy region, a least-squares straight line fitted to the data of Thomas and Bent[16] gives a value, −0.99, for the slope, while the result of Oldham[10] gives, −0.98. Our WL Born result gives the value −0.99 for the slope in Fig. 2. The result of Gaillard[12] for proton impact scaled from electron-impact data is not shown in Fig. 2. This agrees with experiment above 400 keV incident energy, but has a wrong slope.

In the intermediate energy range, discrepancies among different experimental data widely exceed the estimated error limits of the respective authors. No rigorous comparison of absolute values of the theoretical and the experimental cross sections is possible under such circumstances.

However, Oldham[10] observed that there is reasonable agreement in the functional dependence of the cross section on energy in different set of data, and indicated that the experimental curves could be normalized for convenient comparison with theory. Later, Thomas,[24] in a critical review of the experiments, normalized all the observed results at a suitable energy. We also compare the energy dependence of the experimental data with our distortion WL results by normalizing each to unity at 100 keV. This is shown in Fig. 4. Except for the measurements of Denis *et al.*,[17] which are not performed under properly single-

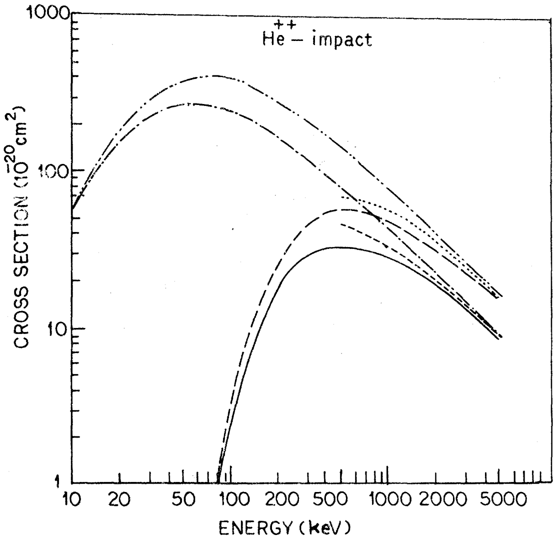

FIG. 3. Cross section for the $4\,^1S$ excitation of He atom by α-particle impact: (——) distortion approximation using WL wave function; (---) second-Born approximation using WL wave function; (—•—•) first-Born approximation using WL wave function; (— —) distortion approximation using V wave function; (•••••) second-Born approximation using V wave function; (—••—••) first-Born approximation using V wave function.

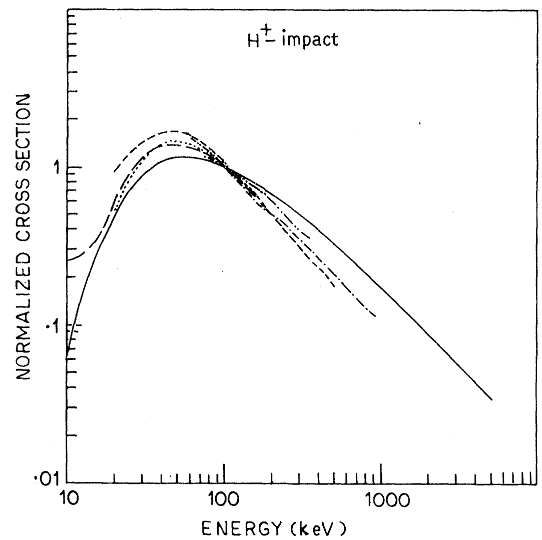

FIG. 4. Cross section for the $4\,^1S$ excitation of He atom by proton impact normalized to unity at 100 keV. Theory: (——) distortion approximation using WL wave function. Experiment: (•••••) Dodd and Hughes (Ref. 14); (—••—••) Robinson and Gilbody (Ref. 15); (—•—•) Thomas and Bent (Ref. 16); (– – – –) Denis *et al.* (Ref. 17); (—— ——) Van den Bos *et al.* (Ref. 18).

scattering conditions, agreement between normalized theoretical and experimental cross sections is, in general, within the accuracy of the measurements down to 20 keV in the low-energy side. Only in case of the results of Van den Bos *et al.*,[18] does discrepancy between theory and experiment exceed the error limits given by the authors. Thomas[24] has observed that the authors do not appear to have made the error estimates adequately.

A peak of our WL results in the distortion approximation occurs at 55 keV. Experimentally it occurs near about 50 keV.

The asymptotic high-energy behavior of Born cross sections was originally considered by Bethe[25] and subsequently by many authors.[26,27] The cross section σ_{nl} for optically forbidden transitions of a single electron from the ground state to (nl) states can be represented by[28]

$$\sigma_{nl} = C_{nl}\, Z^2 M/E\,, \tag{14}$$

where C_{nl} is related to the generalized oscillator strength for the transition concerned (precise interpretation of C_{nl} has been given by Kim and Inokuti[20]) and Z, M, and E are the charge, mass, and energy of the projectile, respectively. The asymptotic behavior of the cross sections allows us to write $C_{nl} = C'_{nl}/n^3$. Expressing σ_{nl} in cm^2 and E in keV and putting $Z = M = 1$ for protons, Thomas[29] gives from $n\,^1S$ excitation data, $C'_{nl} = 1.47 \times 10^{-15}$ for E above 450 keV, with an uncertainty of ±25%. Our first-Born WL result for proton impact gives $C'_{nl} = 1.77 \times 10^{-15}$ for E above 300 keV. The two values agree within the error limits specified by Thomas.[29] The result of Oldham,[10] however, corresponds to a value, $C'_{nl} = 2.14 \times 10^{-15}$.

B. α-particle impact excitation

The α-particle impact cross sections also show the same general behavior as described above for proton impact. Here also the V results at high energy exceed the WL ones by a similar factor of 2. A peak in distortion WL results occurs at 400 keV. The constant C'_{nl} is 1.67×10^{-15} in this case for E exceeding 500 keV.

V. CONCLUSION

Whereas the first-Born calculation in case of proton impact using the V wave function, gives cross section values almost a 100% higher than experiment at its region of validity (high energies), the present Born calculation with the WL wave function gives correct values at high energies. The second-Born and distortion results with the WL wave function gives agreement with experiment at lower energies also, and the latter correctly predicts the energy dependence of the cross section up to 20 keV on the low-energy side. This illustrates the necessity of using a properly orthogonalized set of wave functions in the scattering calculation for high atomic excitations.

In the low-energy region, the present results with the distortion approximation are much smaller than those given by the Born approximation. However, the Born approximation is not reliable in this energy region and as such, the corresponding results do not at all follow the experimental trend, which our distortion results can exhibit in much better ways. Further, for optically forbidden transitions, the Born approximation has been found to be unreliable, even at much higher energies.[21,30] This can be attributed to the neglect of the optically allowed virtual transitions via some strongly coupled intermediate target states.

There exists slight underestimation in our absolute theoretical cross sections at the intermediate-energy region in comparison with the experiments. It is to be noted in this respect that in the present two-state calculations, we neglect coupling to the intermediate target states. Van den Bos[30] has performed coupled-state calculations for the $2\,^1S$, $2\,^1P$, $3\,^1P$, and $3\,^1D$ excitation cross sections of He under proton impact. For the optically allowed transitions, his theoretical results are in fair agreement with the experiment. However, this is not true for the optically forbidden transitions, and no conclusive evidence follows regarding the effect of coupling to the intermediate states on such transitions. The full elucidation of this question with regard to the $4\,^1S$ excitation must hence await a close-coupling calculation retaining the intermediate states.

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