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Double Excitation of Helium by Fast Particles of Charge Z

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Double excitation of helium by fast particles of charge Z

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Results of calculations of double excitation of helium to $n=2$ states for fast projectiles of charge Z are presented. Nonzero Z^3 contributions for single and double excitation occur only when time-ordering contributions from the second-order amplitude are nonzero. For double excitation, electron correlation must also be nonzero to obtain Z^3 terms. The time-ordering effects arise from virtual off-energy-shell intermediate states. As with second-order amplitudes for Thomas singularities in electron capture, the energy-nonconserving amplitude is connected to the second-order energy-conserving amplitude by a dispersion relation. Comparison is made with experiment.

Higher-order terms in perturbation expansions contain information absent in simpler first-order terms. While first-order Born contributions are useful[1] in evaluating basic cross sections for excitation of a single atomic electron by a high-velocity projectile of charge Z, higher-order terms in Z are required for either: (i) multiple excitation in the absence of correlation, or (ii) understanding the nature of intermediate states of the collision. Leading higher-order contributions to observable excitation scattering probabilities and cross sections vary as Z^3. These higher-order Z^3 terms contain information about how a scattering event proceeds, information not included in the simpler first-order Z^2 terms. For example, both time-ordering[2] and intermediate energy-nonconserving contributions[3] are present only in higher-order amplitudes. This information is useful in characterizing few and many-body processes in scattering of atoms by fast charged particles.

Observations by Barkus *et al.*[4] of Z^3 effects, related to single ionization and excitation in atomic collision with positive and negative pions, were made over 30 years ago. In the last decade, observations of double ionization in helium by protons and electrons[5,6] have led to various calculations and interpretations[7–12] of observed Z^3 effects. Experiments in the last few years have been performed to measure differences in high-velocity scattering by protons and antiprotons and by electrons and positrons.[12,13] These differences may be attributed to higher-order effects since the leading-order Z^2 effects give no difference. A non-Z^2 dependence of the double excitation of helium at velocities of 1.5 MeV/amu (or 7.7 atomic units) have been recently reported by two groups.[14,15] In this paper we present an interpretation of Z^3 effects and compare detailed calculations with experiment. We demonstrate that a nonzero Z^3 contribution in single- and double-excitation cross sections requires the presence of time-ordering effects. For double excitation, electron correlation is also necessary[7,16] for the Z^3 term to be nonzero.

The probability amplitude for a transition from $|i\rangle$ to $|f\rangle$ in scattering a particle of charge Z from an atom may be expressed[16] in the interaction representation[17] as

$$a=\left\langle f\left|T\exp\left[-i\int V(t)dt\right]\right|i\right\rangle . \tag{1}$$

Here T is the time-ordering operator[2] and V contains a sum of Coulomb interactions between the projectile and each of the electrons in the atom, namely

$$V(t)=\sum_j e^{-iH_0t}\frac{Z}{|\mathbf{R}(t)-\mathbf{r}_j|}e^{iH_0t} .$$

The perturbation (or Born) expansion in Z is given[16] by

$$a=\langle f|i\rangle-i\int_{-\infty}^{\infty}dt\langle f|TV(t)|i\rangle + \frac{(-i)^2}{2}\int_{-\infty}^{\infty}dt\int_{-\infty}^{\infty}dt'\langle f|TV(t)V(t')|i\rangle+\cdots , \tag{2}$$

where V is linearly proportional to Z.

The zeroth-order term vanishes since the states are orthonormal. In the first-order term the time-ordering operator plays no role since only one time is involved and T may be replaced by unity. For single or double excitation the integral over t is purely real.[17] However, this first-order term is zero for double excitation if there is no correlation. Then V is a sum of single electron operators[16] and

$$\langle f|V|i\rangle=\sum_j(\phi_k^f\phi_j^f|V(r_j)|\phi_k^i\phi_j^i)\sim(\phi_k^f|\phi_k^i)=0$$

for orthogonal states.[18] Hence we may represent the first-order term in Eq. (2) by $-ic_1Z$ where c_1 is a real coefficient which is nonzero in double excitation only if electron correlation is nonzero.

The second-order term in Eq. (2) may be analyzed by setting $T=1+(T-1)$. Here $T=1$ is the limit in which time-ordering effects vanish. Hence we define $T-1$ to be the operator which carries the effects of time ordering. Keeping this in mind and using the step function Θ, the intermediate state propagator from the $e^{\pm iH_0t}$ factors between $V(t)$ and $V(t')$ in Eq. (3) is[19]

$$\frac{T}{2}e^{-i\epsilon(t'-t)}=\Theta(t-t')e^{-i\epsilon(t'-t)}=\frac{i}{2\pi}\int_{-\infty}^{\infty}e^{-i\Omega(t'-t)}\frac{1}{\Omega-\epsilon+i\eta}d\Omega$$
$$=\frac{i}{2\pi}\int_{-\infty}^{\infty}-e^{-i\Omega(t'-t)}\left[-i\pi\delta(\Omega-\epsilon)+\mathrm{P}\frac{1}{\Omega-\epsilon}\right]d\Omega . \tag{3}$$

The principal-value term $P[1/(\Omega-\epsilon)]$ vanishes in the limit $T\rightarrow 1$. Thus the $-i\pi\delta(\Omega-\epsilon)$ term in Eq. (5) corresponds to $T=1$ and the $P[1/(\Omega-\epsilon)]$ term corresponds to $T-1$, which carries the effects of time ordering. We note that in the second-order term $T-1$ may be replaced[2] by $2\Theta(t-t')-1=2(\Theta-\bar{\Theta})$ where $\bar{\Theta}=1/2$ so that $T-1$ corresponds to a time variation of the integrand in Eq. (2) from its average values. In this sense $T-1$ may be regarded as a time correlation. Using this in Eq. (2) it may be shown that all matrix elements for both single and double excitation are real[17] so that the second-order term may be represented by $Z^2(ic_2-\bar{c}_2)$ where $\bar{c}_2$ arises from $T=1$ in Eq. (3) and c_2 arises from the operator $T-1$ which gives the effects of time ordering.

Collecting terms, Eq. (2) may be expressed as

$$a=ic_1Z-(\bar{c}_2-ic_2)Z^2=-i(c_1-c_2Z)Z-\bar{c}_2Z^2, \quad (4)$$

where all the c's are real in double excitation. We have seen that a nonzero c_1 carries spatial correlation and a nonzero c_2 carries time ordering. We note that $\bar{c}_2$, which has no time ordering, may carry some electron correlations and does include the lowest-order independent electron approximation, where a reduces to a simple product of first-order probability amplitudes for double excitation.

Cross sections σ and scattering probabilities $|a|^2$ may be expressed by

$$\sigma=\int|a|^2d\mathbf{B}\simeq\int(c_1^2Z^2-2c_1c_2Z^3+c_2^2Z^4+\bar{c}_2^2Z^4)d\mathbf{B}$$
$$\equiv C_1^2Z^2-2C_{12}Z^3+C_2^2Z^4+\bar{C}_2^2Z^4+O(Z^5), \quad (5)$$

where $\mathbf{B}$ is the impact parameter of the projectile. The difference in double excitation by particles of opposite charge is given by

$$\sigma(-)-\sigma(+)=4C_{12}|Z|^3. \quad (6)$$

This is nonzero only if effects due to both spatial correlation (in c_1) and time ordering (in c_2) are present.

The time-ordering term c_2 also represents effects of energy nonconservation in intermediate states during the collision. This may be seen in Eq. (3) where $T-1$ is associated with $P[1/(\Omega-\epsilon)]$ which restricts $\epsilon\neq\Omega$ so that intermediate energy is not conserved. The energy nonconserving contributions are purely quantum mechanical and may be ascribed to virtual intermediate states. Energy-conserving intermediate states, corresponding to the $-i\pi\delta(\Omega-\epsilon)$ terms in Eq. (3), also contribute to the double-excitation probabilities and cross sections for the $\bar{c}_2$ terms. The energy-conserving terms may be directly related to on-shell physically observable processes, and are also present in classical calculations. It may further be that c_2 and $\bar{c}_2$ obey a dispersion relation, namely

$$c_2(\epsilon)=-\frac{i}{\pi}P\int_{-\infty}^{\infty}\frac{\bar{c}_2(\Omega)}{\Omega-\epsilon}d\Omega, \quad (7a)$$

$$\bar{c}_2(\epsilon)=+\frac{i}{\pi}P\int_{-\infty}^{\infty}\frac{c_2(\Omega)}{\Omega-\epsilon}d\Omega. \quad (7b)$$

Such a dispersion relation has been found to hold in the vicinity of a Thomas singularity in electron capture in a calculation[20] using the Schrödinger representation.

We have evaluated (4) using correlated configuration-interaction (CI) wave functions for the initial and final states. The effective charge Z_{eff} of the helium atom was varied to minimize the ground-state energy. For $Z_{\text{eff}}=1.76$, the initial state wave function,

$$|i\rangle=0.9916|1s^2\rangle-0.1251|1s2s\rangle-0.0230|2s^2\rangle$$
$$+0.0251|2p^2\rangle,$$

gives an energy within 1.6% of the exact energy. The final states were $\langle 2s2p(^1P)|=1.0\langle 2s2p|$, and $\langle 2p^2(^1D)|=1.0\langle 2p^2|$.

In arriving at the second-order amplitudes in (4), we have inserted a complete set of two-electron, correlated states in (2), have performed the energy decomposition (3), and then have used an average-energy approximation to perform the sum by closure. We chose the degenerate energy $\bar{E}$ of the intermediate states to be 51.8 eV above the ground state, which is in line with the reasoning of Lodge,[21] Day,[22] and Harley and Walters[23] except that in this present case we have two ionization thresholds and have chosen $\bar{E}$ to be midway between them. An additional argument in support of choosing the intermediate-state energy to be in the range of the singly-ionized states is that these are precisely the states that play the major role of interfering with the bound states (at about 60 eV above the ground state) in the observation process.[14,15]

Cross sections for projectile-impact excitation into the $2s2p(^1P)$ state are shown in Fig. 1. The proton-to-antiproton ratio is a factor of $\frac{3}{2}$ in the projectile energy region of 0.1 to 0.3 MeV/amu but decreases to 9% at 1.5 MeV/amu where Giese *et al.*[15] made their observation (for which the error bars mask a definitive statement

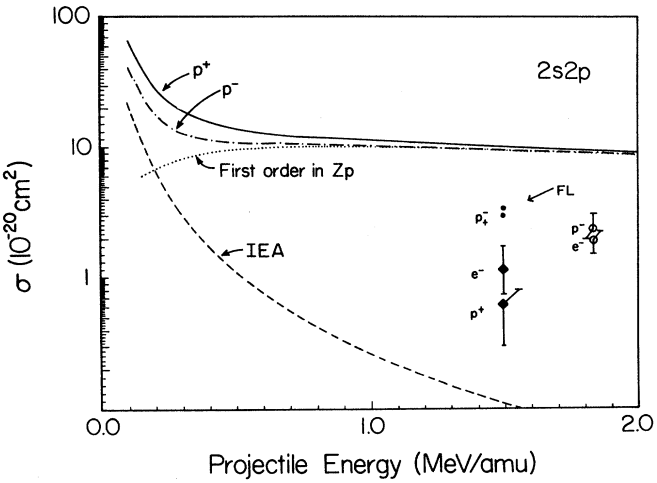

FIG. 1. Cross section for the excitation of helium into the $2s2p(^1P)$ state. The solid is the second-order result for protons and the dot-dashed curve is for antiprotons. The first-order (dotted curve) and (uncorrelated, energy-conserved second order) independent-electron approximation (IEA), (dashed curve) are also shown. The solid circles are the close-coupling result of Fritsch and Lin (FL, Ref. 24). The solid diamonds are experimental results of Giese *et al.* (Ref. 15) and the open circles are data from Pedersen and Hvelplund (Ref. 14) who summed the $2s2p(^1P)$ and $2p^2(^1D)$ cross sections.

about differences due to the sign of the projectile charge). Also shown are the close-coupling calculation of Fritsch and Lin[24] at 1.5 MeV/amu and the experimental results of Pedersen and Hvelplund[14] for excitation into either $2s2p(^1P)$ or $2p^2(^1D)$ at 1.84 MeV/amu. One may see that our result is almost all first order at 1.5 MeV/amu, so the difference in magnitude from experiment is not due to our use of a closure approximation. Figure 2 shows the cross sections for excitation into the $2p^2(^1D)$ state. Even though the first- and second-order contributions (not shown) are of the equivalent magnitude over the entire energy range in this case, projectile charge differences are only of order 10%. Our result is in near agreement with the data of Giese *et al.*[15] and lies above the result of Fritsch and Lin by a factor of 2 or 3 (p^+ or e^-).

Our analysis gives some insight into the nature of Z^3 terms in double excitation. However, this analysis does not explicitly distinguish between various specific physical mechanisms such as shakeoff, TS1 (two step with one interaction with the projectile), or polarization,[7-12,24,25] which have been proposed to explain observations.[4-6,12-15,25-29] In our opinion, this question of the specific nature of the mechanism for double excitation is still both interesting and open. In our analysis we have ignored interference effects between the Auger resonance and the continuum background, which is consistent with Fano's interpretation.[30] If the continuum background varies with the Auger electron energy and the Fano q parameter[27] is not larger than one, then interference with the continuum background could be non-negligible for this data. It is safer to compare the sum of the $2s2p$ and $2p^2$ results for experiment and theory. Then our calculations still lie well above experiment, however. Coupling to continuum intermediate states, ignored by both us and Fritsch and Lin,[24] could be significant. Our calculations use configuration-interaction bound-state wave functions with real coupling coefficients. It is not clear to us that these terms remain real for continuum states. Hence here we avoid analysis of important data for double ionization. Our analysis may be applied to single excitation with the understanding that c_1 is generally nonzero with or without electron correlation and that interference with the ionization continuum is ignored. Hence for a nonzero Z^3 term, time ordering is essential in a single-excitation cross section, but electron correlation is not. For higher-order terms in Z it is evident that odd-Z contributions to $|a|^2$ disappear when energy-nonconserving virtual intermediate states are eliminated since even and odd terms in Z contributing to the amplitude a then differ by a factor of i from the even terms.[31,32] However, further understanding of the pattern of time-ordering and energy nonconservation in higher order terms in Z remains open to further investigation.

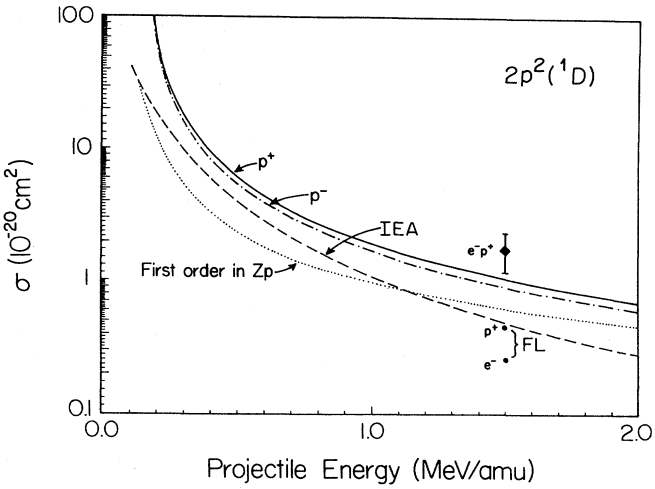

FIG. 2. Excitation into the $2P^2(^1D)$ state. The notation is the same as in Fig. 1.

In summary we have shown that Z^3 contributions to atomic excitation contain time-ordering contributions from the Z^2 amplitude. These time-ordering contributions may also be regarded as contributions from energy-nonconserving intermediate states of the collision. For double-excitation correlation must be present, in addition to time ordering, for a nonzero Z^3 term to occur. Proton versus antiproton cross sections for excitation into the $2s2p(^1P)$ state differ by a factor of $\frac{3}{2}$ in the energy region between 0.1 and 0.3 MeV/amu. At other energies, and all energies in this region for the $2p^2(^1D)$ final state, such differences are of order 10%.

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[32]This has also been noted by J. Macek (private communication).