# Mean-Field Approximation to $p$ + He Scattering

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(Received 30 March 1981)

The scattering of protons by helium atoms is studied in a mean-field approximation to the many-body $S$ matrix. The one-body equations governing the electron dynamics for elastic scattering are solved iteratively beginning with the interaction-picture time-dependent Hartree-Fock (TDHF) solution. At a representative proton laboratory energy of 20 keV, the converged $S$-matrix elements are within 10% of the TDHF value. Inclusive single-charge transfer probabilities calculated in the interaction-picture TDHF approximation agree reasonably well with the experimental data.

PACS numbers: 34.10.+x, 34.70.+e

The study of colliding many-body systems is a problem common to both nuclear and atomic physics. Recently, the time-dependent Hartree-Fock (TDHF) method [1-3] has been shown to provide a good description of the inclusive aspects of nuclear collisions. It is therefore of interest to consider the extent to which these methods can be applied to atomic systems. The first steps in this direction were taken in Ref. 4, where the finite-difference numerical techniques developed for the TDHF theory were successfully applied to the single-electron problem of $H^+$-H collisions. However, for many-electron systems, the deficiencies[5] of TDHF as a theory for exclusive properties make it difficult to obtain cross sections to particular final channels. As an improvement to the TDHF theory in this connection, Alhassid and Koonin[6] have recently proposed a time-dependent mean-field approximation to the $S$ matrix for the excitations of a many-body system induced by an external time-dependent potential, $V(t)$. Our aim in this work is to study some of the features of this approximation by considering a two-electron system, $p$ + He. The motion of proton is approximated by a classical Coulomb trajectory and only the intrinsic excitations of the electrons are treated in the mean-field approximation.

When dealing with many-body collision processes, a heirarchy of one-body approximations can be formulated. In the simplest of these, the many-body Hamiltonian is replaced by the time-independent HF Hamiltonian.[7] Here, dynamical changes of the self-consistent field due to the external time-dependent potential are neglected completely. A more detailed treatment including these changes can be formulated within the TDHF approximation. For heavier colliding systems and for processes involving outer-shell electrons, the dynamical changes in the mean field can be important, although they may not greatly affect processes involving deeply bound inner-shell electrons. At the most sophisticated level, the mean-field approximation replaces the full many-body Hamiltonian by a self-consistent one-body time-dependent potential in a way so as to avoid the difficulties in obtaining individual transition amplitudes within the TDHF framework.

A detailed derivation of the mean-field approximation to the $S$ matrix is given in Ref. 6; here we only quote the relevent equations. The Hamiltonian is taken to be

$$H(t) = H_0 + V(t),$$

where $H_0$ is the unperturbed many-body Hamiltonian consisting of the kinetic energy (and possibly a one-body potential, as the case may be) and a local two-body potential, $v(x - x')$. Apart from a phase factor, the $S$ matrix in the mean-field approximation for a transition from an initial state $|\beta\rangle$ to a final state $|\beta'\rangle$ is given by

$$\langle\beta'|S|\beta\rangle = \lim_{t\to\infty} \langle\beta'|U_{\sigma_f}(0,t)U_\sigma(t,-t)U_{\sigma_i}(-t,0)|\beta\rangle, \quad (1)$$

where the one-body evolution operator $U_\sigma$ is

$$U_\sigma = T\exp[-i\int h_\sigma(\tau)d\tau]$$

corresponding to the one-body Hamiltonian

$$h_\sigma = -\frac{\hbar^2}{2m}\nabla^2 + V(x,\tau) + \int v(x-x')\sigma(x',\tau)dx'.$$

Here, $x$ stands for both space and spin coordinates, $T$ denotes time-ordered product, and $U_{\sigma_i}$, $U_{\sigma_f}$ are similar to $U_\sigma$, but with $V(\tau)$ absent. Denoting $\sigma_i$, $\sigma$, or $\sigma_f$ by the general symbol $\sigma_l$, the mean fields are defined by

$$\sigma_l(x,\tau)=\mathrm{Re}\frac{\langle\beta_l'(\tau)|\sum_{i=1}^{2}\delta(x-x_i)|\beta_l(\tau)\rangle}{\langle\beta_l'(\tau)\beta_l(\tau)\rangle}. \quad (2)$$

The wave function $|\beta_l(\tau)\rangle$ satisfies the TDHF-like equation

$$i\hbar\dot{\beta}_l=h_{\sigma_l}\beta_l, \quad (3)$$

with the initial condition $|\beta_i(\tau=0)\rangle=|\beta\rangle$ and is obtained by solving this equation around a time "loop": along $(0,-t)$ with $h_{\sigma_i}$, along $(-t,t)$ with $h_\sigma$ and along $(t,0)$ with $h_{\sigma_f}$. The state $|\beta_l'(\tau)\rangle$ is defined similarly, but with the boundary condition $|\beta_f'(\tau=0)\rangle=|\beta'\rangle$. The $S$ matrix (1) is then approximated by

$$\langle\beta'|S|\beta\rangle=\langle\beta_l'(\tau)|\beta_l(\tau)\rangle, \quad (4)$$

which is independent of $\tau$. Note that the mean field $\sigma(x,\tau)$ depends on both the initial and final channel wave functions, whereas in TDHF it depends on only the initial state $|\beta\rangle$.

To solve Eqs. (2)–(4) self-consistently, we use static HF solutions generated in coordinate space[8] for the channel wave functions $|\beta\rangle$ and $|\beta'\rangle$. A trial solution $|\beta_l\rangle$ with the boundary condition $|\beta_i(\tau=0)\rangle=|\beta\rangle$, is obtained in the TDHF approximation by replacing $|\beta_l'\rangle$ by $|\beta_l\rangle$ in Eq. (2) and solving Eq. (3) along the time loop discussed above. A similar procedure is followed to obtain a trial $|\beta_l'\rangle$. With these wave functions, new fields $\sigma_l(x,\tau)$ are obtained from Eq. (2) and new wave functions $|\beta_l\rangle$ and $|\beta_l'\rangle$ are calculated with use of Eq. (3). This iteration procedure is carried out until the value for $\langle\beta'|S|\beta\rangle$ given by Eq. (4) converges.

For $p$+He scattering, the unperturbed Hamiltonian $H_0$ and the external potential $V(t)$ are

$$H_0=-\frac{\hbar^2}{2m}\sum_{i=1}^{2}\left(\nabla_i^2-2\frac{e^2}{\tau_i}\right)+\frac{e^2}{|r_1-r_2|}+2\frac{e^2}{R(t)},$$

$$V(t)=-\sum_{i=1}^{2}\frac{e^2}{|r_i-R(t)|},$$

where $r_i$ are the space coordinates of the electrons and $R(t)$ is the position of the proton relative to the origin. We work in the laboratory frame and neglect the target recoil. Hence the origin is fixed at all times at the He nucleus. The calculations are done in a cylindrical coordinate system if one assumes the electron wave functions to be axially symmetric and spin degenerate. The computational frame therefore rotates so that the internucleus separation coincides with the $z$ axis at all times. This approximation,[1] which is reasonable for small impact parameters or low bombarding energies, reduces the one-body Eqs. (3) to two spatial dimensions. Details of the numerical procedure for the time integration in cylindrical coordinates are given in Ref. 1. The long-range nature of the Coulomb interaction requires spatial mesh dimensions much larger than those used to study nuclear collisions. In the present calculation a $40\times130$ mesh with a spacing of $0.227a_0$ was used. To smooth the singularities of the potential at the nuclear positions, the nuclei were given a small radius[4] of $0.227a_0$. With this choice, the HF energy of He ground state is 75.575 eV, which is within 4.5% of the experimental value and is in good agreement with the exact HF value of 77.993 eV (Ref. 9) when corrected for our finite nuclear size. The time evolution was done with a time step of 12 Å/$c$, and preserved the norm of the wave function to one part in $10^3$ over the entire collision time. The computational time taken for one iteration around the time loop at each impact parameter is of the order of 1 min on a Control Data Corp. CDC-7600 computer.

Figure 1 shows the convergence of $|\langle\beta'|S|\beta\rangle|^2$ for elastic scattering ($\beta'=\beta$) at impact parameters 0 and 0.1 Å and at a laboratory proton energy $E=20$ keV. Convergence is achieved in about 7 iterations to a value which differs from that of interaction picture TDHF[6] (the trial value) by only 6%. This is to be expected since the electron-electron interaction is weak relative to the electron-nucleus interaction.

At energies above 1 keV, one of the dominant processes is charge transfer. Experimental measurements of the total single-charge transfer probability, $P_{t_1}$ for the $p$+He system have been made[10] for $E=1.6$–$180$ keV and laboratory scattering angles 0.5°–4°. We have calculated $P_{t_1}$ in the TDHF approximation for $E=5$–$80$ keV for a fixed impact parameter of 0.1 Å (scattering angle 3.5°–0.2°). This corresponds to the value of $|\langle\beta'|S|\beta\rangle|^2$ after the first iteration. The exit

channel wave function in this case is

$$|\beta_{\lambda,\mu}'\rangle = \psi_\lambda(\vec{r}_1)\varphi_\mu[\vec{r}_2 - \vec{R}(t)]\exp\left[-i\vec{k}\cdot\vec{r}_2 - i\left(\epsilon_\lambda + \eta_\mu + \frac{\hbar^2k^2}{2m}\right)t\right].$$

Here, $\psi_\lambda$ and $\varphi_\mu$ are the hydrogenic single-electron wave functions for $He^+$ and H, respectively, with binding energies $\epsilon_\lambda$ and $\eta_\mu$, and $\vec{k}$ is the wave vector of an electron moving with the proton velocity. The total single-charge transfer probability is $\sum_{\lambda,\mu}|\langle\beta_{\lambda\mu}'|S|\beta\rangle|^2$. That this probability can be meaningfully compared with the experiment follows because of two reasons. First, as mentioned above, the rapid convergence of the $S$ matrix implies that the converged result would not be very different. Second, the TDHF value for $P_{t_1}$ shows very little time variation at long times when the evolution is carried out in the interaction picture; i.e., along the loop described above. In fact, it has been shown by model calculations[6,11] that the TDHF value for inclusive averages of the $S$ matrix can be an excellent approximation to the exact results.

In Fig. 2, we compare the calculated TDHF inclusive single-charge transfer probabilities with the experimental data.[10] (By inclusive probability we mean the probability for detecting a neutral hydrogen time, ground state or excited, irrespective of the state of the rest of the system. Hence this does not include two-electron capture processes.) The results of the atomic-state expansion method[12,13] are also plotted for comparison. The better agreement of the present theoretical results with the experiment is evident. The lack of distortion effects (important at low energies) and intermediate continuum states (important at high energies) in the atomic state expansion[14,15] leads to errors at both low and high energies. However, these effects are automatically included in our coordinate-space implementation of TDHF. The deviation of our calculated probabilities from the experimental data at high energies is probably due to our imposition of axial symmetry and neglect of recoil, approximations which are expected to become worse with increasing energy.

To summarize, we have applied the mean-field approximation to the many-body $S$ matrix to the case of $p$ + He scattering. The convergence of the $S$ matrix is achieved in 6 to 7 iterations when the trial solutions are chosen to be the interaction-picture TDHF solutions. The converged values of the $S$-matrix elements differ from the TDHF values by only ≈6%. Applications of this method to exclusive charge transfer processes and to heavier systems seem feasible and are in progress. As a first step in this direction, we have calculated the total single-charge transfer probability in the TDHF approximation over a wide energy range for extreme forward scattering. These results are in significantly better agreement with experiment than are those of the atom-

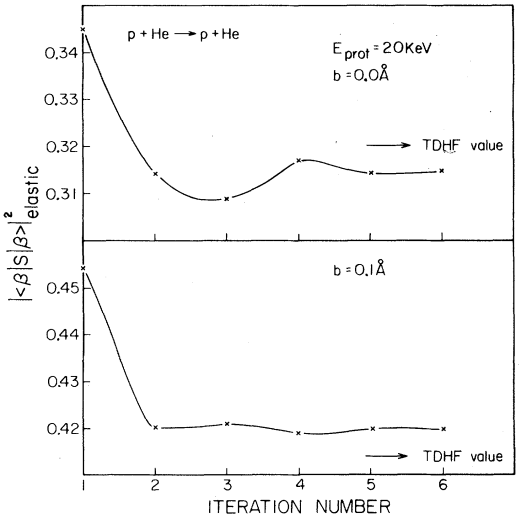


FIG. 1. Convergence of the absolute values of the $S$-matrix elements for elastic scattering at $E = 20$ keV and impact parameters $b = 0$ and 0.1 Å.

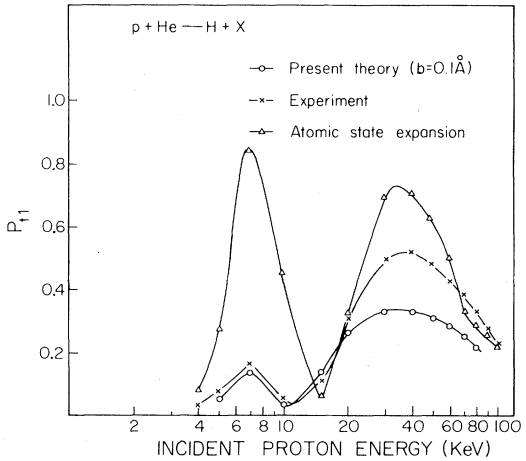


FIG. 2. Inclusive single-charge transfer probabilities, $P_{t1}$.

ic-state expansion method.

Useful comments and suggestions by Dr. Y. Alhassid at various stages of this work are gratefully acknowledged. We would also like to thank Dr. S. Levit, Dr. C. Bottcher, Dr. K. Kulander, and Dr. M. R. Strayer for fruitful discussions. This work was supported in part by the National Science Foundation under Grant No. PHY77-21602. One of us (S.E.K.) would like to thank the Alfred P. Sloan Foundation for providing a fellowship.

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# *Ab Initio* Calculation of the Third-Order Susceptibility of $H_2$

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(Received 17 March 1981)

The third-order susceptibility of $H_2$ has been calculated under coherent anti-Stokes Raman-scattering conditions, with use of wave functions specifically designed for accurate calculations of sum-rule properties. The theoretical values are in good agreement with experiment.

PACS numbers: 31.20.Di, 33.20.Fb, 33.80.Kn

The advent of tunable lasers has facilitated the probing of the nonlinear response of atoms and molecules to intense radiation fields. In an isotropic medium, the lowest-order nonlinear optical effect is due to the third-order susceptibility $\chi^{(3)}$. This property has been exploited to develop frequency converters, to provide high-resolution Raman spectra by means of coherent anti-Stokes Raman scattering (CARS), and to provide two-photon spectra.[1] In spite of its wide range of applications, first-principles calculation of $\chi^{(3)}$ for molecules with explicit frequency dependence is noticeably lacking.[1,2] We report here an *ab initio* calculation of $\chi^{(3)}$ for $H_2$ with wave functions designed to give accurate values of the sum-rule properties $S(0)$ and $S(-1)$. We have chosen to use CARS conditions because of the availability of experimental data,[1,3-5] but the method can be readily applied to other experimental conditions.

We use the expression of $\chi^{(3)}$ derived from perturbation theory.[6,7] We have chosen to do an explicit summation over states instead of using a variation-perturbation approach because application of the Dalgarno-Epstein (DE) condition[8] enables us to replace the infinite sum by a finite sum; and, once the wave functions are determined, $\chi^{(3)}$ can be calculated easily for a range of frequencies and experimental conditions with little additional effort. The good agreement between our calculation and experiment indicates that the replacement can be reliably carried out. Our method therefore resolves the outstanding