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W. T. Htwe

Tibor Vajnai

M. Barnhart

A. D. Gaus

et. al. For a complete list of authors, see https://scholarsmine.mst.edu/phys_facwork/1320

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Differential Double Excitation Cross Sections in Proton-Helium Collisions Studied by Energy-Loss Spectroscopy

W. T. Htwe, T. Vajnai, M. Barnhart, A. D. Gaus, and M. Schulz
Department of Physics and the Laboratory for Atomic and Molecular Research, University of Missouri-Rolla, Rolla, Missouri 65401
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We have measured ion energy-loss spectra for 150 keV proton-helium collisions as a function of the projectile scattering angle. From the data we obtained double excitation cross sections differential in the proton scattering angle as well as the ratios of both double excitation and single ionization to single excitation. In these ratios pronounced peak structures are observed at about 0.7 mrad. Two alternative interpretations of these peak structures are offered: They may be due to binary collisions between the projectile and the target electrons, or they could be a manifestation of an interference between different transition amplitudes leading to the same final state of the collision.

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One important goal of studying ion-atom collisions is to increase our understanding about the dynamics of the fundamental forces acting in atomic systems. The forces that can lead to inelastic processes in ion-atom collisions are the force between the nucleus of one collision partner and the electron of the other collision partner and the force between any two electrons in the collision system. Even though the underlying fundamental interaction for both forces is the same (electromagnetic), the dynamics of the nuclear-electron force is significantly different from the one of the electron-electron force: In the nuclear-electron force a well localized particle (the de Broglie wavelength of a proton at energies studied here is negligible) interacts with a poorly localized, diffuse electron cloud. In the electron-electron force, in contrast, two diffuse electron clouds interact with each other. Our understanding of the dynamics of the electron-electron force is much less complete than the nuclear-electron force.

The electron-electron force is usually insignificant in one-electron processes such as single excitation; however, it can be very important in two-electron processes like double excitation [1]. Double excitation of the target atom can proceed through two different mechanisms. In one mechanism, referred to as uncorrelated double excitation, the projectile interacts sequentially with both target electrons independently through the nuclear-electron force. In the second process, correlated double excitation, the projectile interacts with only one target electron and this target electron then interacts with the second electron. The second process has gained some interest in recent years as it involves the electron-electron force explicitly. Furthermore, since both processes lead to the exact same final state of the collision, they are indistinguishable which can result in quantum mechanical interference between them.

A number of experiments have been performed studying double excitation of He by looking at the Auger electron spectra [2–5]. The projectile Z dependence of the total cross sections was measured in these experiments and a strong deviation from a Z^4 dependence of these cross sections was found [5]. Such a deviation indicates a strong electron-electron correlation. Interference effects between the correlated and uncorrelated processes, however, have not been identified yet. On the other hand, interference effects would provide a very sensitive tool to test the theoretical treatment of the dynamical aspects of the forces involved in double excitation. The phase difference between the transition amplitudes for the correlated and uncorrelated processes depends on the scattering angle. In a total cross section measurement, where one integrates over all scattering angles, this phase difference may be partially or completely averaged out. Therefore, if there are any significant interference effects present in double excitation, they should be much more pronounced in the differential cross sections as a function of scattering angle than in total cross sections. Another interference which could affect the double excitation cross sections is known as the Fano interference [6]. The doubly excited states of He decay to the ground state almost exclusively through autoionization. These autoionized electrons are indistinguishable from directly ionized electrons of equal energy which leads to the Fano interference.

In this Letter we present the first measured differential double excitation cross sections as a function of scattering angle for the proton-helium collision system. The data were obtained using ion energy-loss spectroscopy. Energy-loss spectra for proton-helium collisions were recently studied by Schiwietz *et al.* [7]. In their experiment, the energy resolution was not sufficient to discern double excitation peaks from the continuous ionization spectrum. They were thus focusing on single ionization whereas our primary interest lies in double excitation.

The experiment was performed at the University of Missouri-Rolla ion energy-loss spectrometer (UMRIELS). The details of the ion energy-loss spectroscopy method are described elsewhere [8] and only a brief summary is given here. A beam of protons with narrow energy spread (<1 eV) is created in a hot cathode

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ion source and is accelerated to an energy of 150 keV. The beam is then collimated and steered into a gas cell with a length of 1 cm containing He at a pressure of 70 mTorr. After passing through the target region, the beam is cleaned from charge changed components by a switching magnet. The protons then pass through a solid angle defining collimator before they are decelerated to an energy of 2 keV and energy analyzed by a parallel plate analyzer. An energy-loss spectrum is obtained by scanning an offset voltage on the accelerator relative to the decelerator potential. A scattering angle range of 0 to 1 mrad was covered by pivoting the accelerator around the center of the collision chamber. The overall energy resolution was 1.5 eV and the angular resolution was 0.15 mrad FWHM.

In Fig. 1 we show energy-loss spectra taken for scattering angles of 0.3 mrad (top), 0.55 mrad (center), and 0.8 mrad (bottom). Data were taken in the energy-loss region of 19 to 25 eV and from 45 to 70 eV. Pronounced structures can be seen in these spectra. The peak at 21 eV is due to single K and L excitation of He. The peak at 23 eV contains excitation from the K shell to all states with $n > 2$ which are unresolved. The ionization continuum starts at 24.6 eV. The region between 45 and 70 eV is dominated by single ionization. Double excitation is expected for energy losses around 60 eV. In the energy-loss spectrum for $\theta = 0.3$ mrad a very small peak is observed at 60 eV. However, at $\theta = 0.55$ mrad the magnitude of this peak is significantly increased relative to the single ionization background and the single excitation peaks. At $\theta = 0.8$ mrad two peaks can be resolved near 60 eV. A detailed analysis of the energy losses where these peaks occur reveals that the first peak (58 eV) is due to double excitation to the $(2s^2)^1S$ state and the second peak (60 eV) contains double excitation to the unresolved states $(2p^2)^1D$ and $(2s2p)^1P$ (for simplicity, we refer to this peak in the following as ^1D state). This examination of the energy-loss spectra shows two features of the data which are worth noting: (1) Both double excitation and single ionization relative to single excitation are more important at large scattering angles than at small angles and (2) double excitation to the $(2s^2)^1S$ state is only significant at large scattering angles.

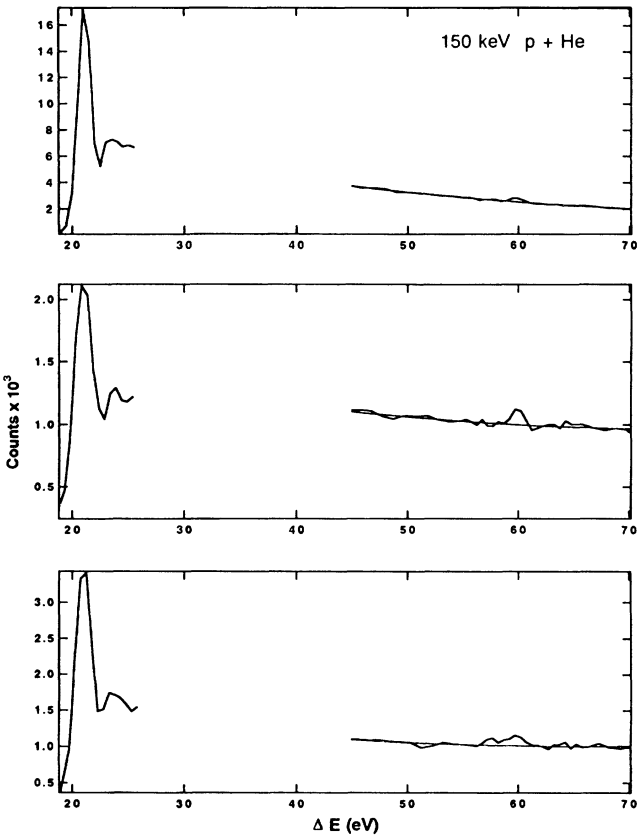

FIG. 1. Projectile energy-loss spectra for 150 keV p + He for scattering angles of 0.3 mrad (top), 0.55 mrad (center), and 0.8 mrad (bottom). The peaks at 21 and 24 eV are due to single K to L excitation and single excitation to $n > 2$, respectively. The region for energy losses larger than 45 eV is due to single ionization. The peaks near 60 eV are due to double excitation.

Since a discernible peak for the $(2s^2)^1S$ state was only observed for scattering angles equal or larger than 0.8 mrad, in the following we will analyze double excitation only for the ^1D state. In order to extract double excitation cross sections from the data we have fitted the ionization background between 45 and 70 eV excluding the region where the double excitation peaks occur by a polynomial. These fits, which are shown in Fig. 1, were then subtracted from the spectra and the remaining ^1D double excitation peak was integrated. This method of analyzing the data is not strictly legitimate because direct single ionization leading to energy loss equal to the double excitation energy has to be treated coherently with double excitation followed by autoionization, as mentioned above. Therefore, we have also determined the double differential single ionization cross sections $d^2\sigma/d\Omega dE_{\mathrm{SI}}$ as a function of scattering angle for an energy loss of 60 eV in a 2 eV bin (which includes the ^1D double excitation peak). This is the same bin size over which the ^1D double excitation line was integrated. These latter cross sections contain direct single ionization and double excitation, which are thus treated coherently. Double ionization does not contribute in this energy-loss region because the threshold energy for double ionization is 79 eV. Single K to L excitation cross sections were obtained by fitting the peak at 21 eV with a Gaussian function.

The fact that in our experiment single and double excitation and single ionization are measured simultaneously allows us to determine the ratios of the cross sections for double excitation $(d\sigma/d\Omega)_{\mathrm{DE}}$ to single excitation $(d\sigma/d\Omega)_{\mathrm{SE}}$ and for single ionization $(d^2\sigma/d\Omega dE)_{\mathrm{SI}}$ to single excitation almost free of systematic errors. Typical sources of systematic errors, such as detector efficiencies, target thickness, or beam fluctuations, cancel in this ratio if both cross sections are measured simultaneously with the same detector. The

ratios $R_{DE} = (d\sigma/d\Omega)_{DE}/(d\sigma/d\Omega)_{SE}$ are plotted in Fig. 2 as a function of scattering angles as full circles. The open circles in Fig. 2 show the corresponding ratios R_{SI} for the doubly differential single ionization cross section for an energy loss of 60 eV relative to single excitation. The contribution of double excitation followed by autoionization to total single ionization at this energy loss is only 5% to 15% depending on the scattering angle, as can be seen from the energy-loss spectra in Fig. 1. The ratios R_{SI} are thus dominated by direct ionization.

The scattering angle dependence of these ratios confirms the general trend that can be seen in the energy-loss spectra already: With increasing scattering angle double excitation and single ionization become increasingly more important relative to single excitation. However, apart from this trend, there is a pronounced and relatively narrow (~0.2 mrad FWHM) peak at about 0.7 mrad in R_{DE}. In R_{SI} there is a peak at almost the same angle (0.65 mrad) which is significantly wider (~0.4 mrad FWHM) than the peak in R_{DE}.

A similar peak structure has been observed in the ratios of the differential cross sections for double ionization relative to single ionization R_{DI} as a function of scattering angle [9]. However, in R_{DI} the peak occurs at an angle of 1 mrad and the peak is much wider (~0.5 mrad FWHM) than in R_{DE}. Several authors interpreted the peak in R_{DI} within an independent electron model as due to binary collisions of the projectile with both target electrons even though the magnitude of the peak was not well reproduced

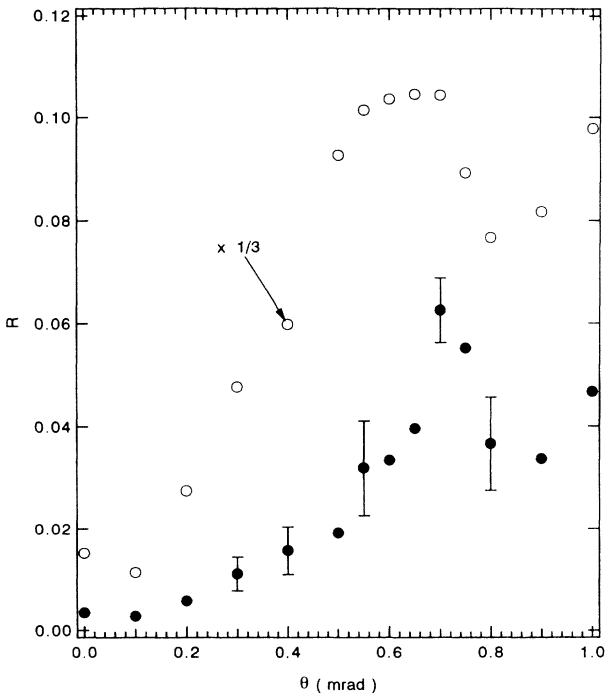

FIG. 2. Ratios of the differential cross sections for double excitation to the ^1D state to single excitation (full circles) and doubly differential single ionization cross section at 60 eV to single excitation (open circles) as a function of scattering angle.

[10–13]. Other interpretations have been given [14,15]. Some experimental results on single ionization were reported which support the importance of projectile-target electron scattering [16,17]. A similar model could be used to attempt to explain the peak structure in the present data for R_{SI} and R_{DE}. In a binary collision of the projectile with the target electrons, the deflection from the target nucleus is neglected and the scattering angle is unambiguously determined by the energy transfer to the target electron due to momentum and energy conservation. In the binary collision model, direct single ionization (which is the dominating contribution in R_{SI}) proceeds through a single collision of the projectile with one target electron, which is assumed to be initially at rest. The energy transfer of 60 eV in this collision corresponds to a scattering angle of 0.42 mrad. The fact that the peak in R_{SI} appears at a larger scattering angle could be explained by a non-negligible deflection of the projectile from the target nucleus. These nuclear contributions to the total deflection along with the momentum distribution of the electron in the initial state can also explain the width of the peak. The gross features of our data for R_{SI} are therefore not inconsistent with the binary collision model.

In double excitation to the ^1D state the total energy transfer to both electrons is 60 eV. In the correlated double excitation mechanism, the entire energy transfer occurs in a single collision of the projectile with one target electron. Therefore, in the binary collision model correlated double excitation should be completely equivalent to direct single ionization leading to an energy loss of 60 eV since the collision kinematics are completely determined by the energy transfer. In the uncorrelated mechanism this energy transfer occurs in two collisions of the projectile with both target electrons. The energy transfer in the first collision is 21 eV [single excitation to the $(1s2p)^1P$ state] which corresponds to a scattering angle of 0.27 mrad. The energy transfer in the second collision is 39 eV [excitation from the $(1s2p)^1P$ state to the $(2p^2)^1D$ state] corresponding to a scattering angle of 0.35 mrad. Therefore, the total scattering angle in such a double collision can be anywhere between 0.8 and 0.62 mrad, depending on the deflections from the two collisions relative to each other.

The peak position in R_{DE} is not inconsistent with the binary collision model if deflection of the projectile from the target nucleus is considered since it nearly coincides with the peak position in R_{SI}. However, it is not clear why the peak in R_{DE} is so much narrower than in R_{SI}. A difference in width could be due to some contributions from the uncorrelated double excitation process. However, we would expect that such contributions would make the peak wider rather than narrower since the total scattering angle is no longer unambiguously determined for two binary collisions.

An alternative explanation for the peak structure in R_{DE} is an interference effect either between the uncorrelated

and correlated double excitation mechanisms or between double excitation followed by autoionization and direct ionization as described above. Interference patterns tend to depend very sensitively on parameters determining the phase difference between the involved wave functions. It is therefore hard to predict the features of possible interference effects in double excitation even on a qualitative level without a full calculation. The impact parameter dependence of double excitation has been calculated for 1.5 MeV p and $\overline{p}$ + He by Moribayashi *et al.* [18]. They attributed differences between the probabilities for p and $\overline{p}$ impact to an interference between the correlated and uncorrelated double excitation mechanisms. However, direct ionization was not taken into account in this calculation. No calculation on differential double excitation cross sections for collision energies studied in this work is currently available.

In Fig. 3 we show the differential cross sections for double excitation to the ^1D state. A similar peak structure as in R_{DE} is observed at 0.7 mrad. This shows that the peak in R_{DE} is due to the angular dependence of double excitation rather than single excitation. Neither the angular dependence of the single ionization nor the one of the single excitation cross sections shows any structure, so that it is not clear whether the peak in R_{SI} is due to single ionization or single excitation. The peak in the double excitation cross section accounts for about 10% of the integrated cross section and is thus non-negligible in spite of the relatively large scattering angle of its occurrence. We also obtained the total cross section by integrating over the scattering angle. For the sum of the $(2p^2)^1D$ and the $(2s2p)^1P$ states we obtain a total cross section of

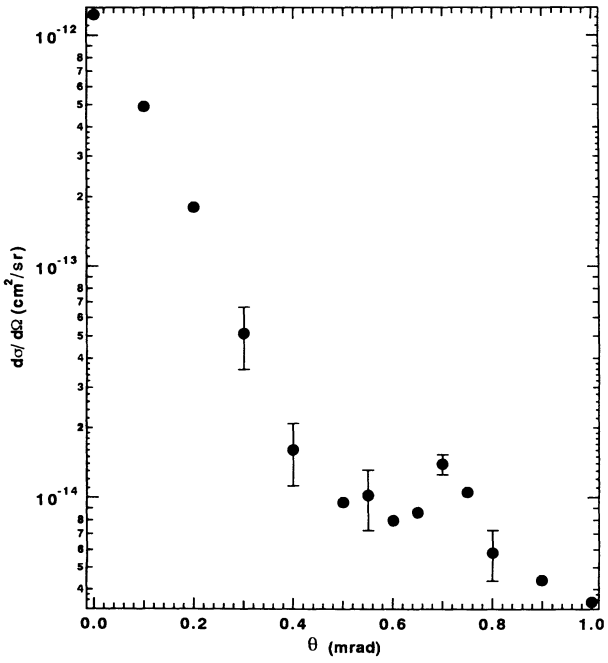

FIG. 3. Absolute differential cross sections for double excitation to the ^1D state as a function of scattering angle.

$(1.1 \pm 0.4) \times 10^{-19}$ cm^2. This value is consistent within experimental uncertainties with the cross sections reported by Bordenave-Montesquieu, Gleizes, and Benoit Cattin [2] of $(1.8 \pm 0.6) \times 10^{-19}$ cm^2 which was obtained for the same collision system using electron spectroscopy.

In summary, we have measured differential double excitation cross sections as a function of scattering angle for the first time. A pronounced peak structure was observed at 0.7 mrad. The peak position is not inconsistent with a binary collision model which was used to explain similar structures in double ionization. However, we cannot explain the width of the peak within the simple one or two binary collision model. Interference effects between the uncorrelated and correlated double excitation mechanisms or between double excitation followed by autoionization and direct ionization offer an alternative explanation for our data. Hopefully, this work will stimulate theoretical calculations which will shed further light on these important collision mechanisms.

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