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## Differential double capture cross sections in $p+\text{He}$ collisions

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We have measured differential double capture cross sections for 15 to 150 keV  $p+\text{He}$  collisions. We also analyzed differential double to single capture ratios, where we find pronounced peak structures. An explanation of these structures probably requires a quantum-mechanical description of elastic scattering between the projectile and the target nucleus. Strong final-state correlations have a large effect on the magnitude of the double capture cross sections.

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### I. INTRODUCTION

Inelastic processes in atomic collisions in which two or more electrons undergo a transition are particularly suitable to study the role of the electron-electron interaction. The reason is that the scattering dynamics tends to be dominated by first-order contributions in the projectile-target atom interaction. On the other hand, in such a first-order mechanism the projectile interacts with only one electron directly. Consequently, a two-electron process can then only occur in the presence of the electron-electron interaction. The theoretical treatment of the collision dynamics under the influence of the electron-electron interaction is particularly challenging because it leads to a dynamic screening of the nuclei. Treating two-electron processes in terms of an effective two-center potential, generated by nuclei with effective (partly screened) charges, is often inadequate. In contrast, for one-electron processes, where the electron-electron interaction tends to be much less important, such an approach is often quite successful.

To extract the maximum amount of information about the role of the electron-electron interaction, multiply differential, ideally fully differential, measurements on two-electron processes are highly desirable. There is a rich literature on experimental studies of such processes, such as, e.g., double ionization, double excitation, or double capture (for reviews see e.g. [1–3]). However, for most reactions the investigator faces serious problems in performing and/or interpreting fully differential experiments. In double ionization the final state is rather complex involving four unbound particles. As a result, experiments typically integrate over several kinematical parameters. Fully differential cross sections are only available for electron impact [4,5] and only one nearly fully differential measurement was performed for ion impact [6]. Theoretically, double ionization is very difficult to describe because the final state involves a two-electron continuum of a two-center potential.

Double excitation is kinematically simpler than double ionization. Considering that most doubly excited states decay predominantly by autoionization, the final state involves three unbound particles and a single electron continuum. However, a fully differential experiment requires measuring the autoionized electrons in high energy resolution (to sep-

rate, to the extent possible, autoionization from direct ionization) in coincidence with the momentum-analyzed scattered projectiles or recoil ions. Such an experiment is extremely difficult and has not been performed yet. Only cross sections differential in the electron (e.g., [7,8]) or projectile solid angle [1,9,10] are currently available. Furthermore, autoionization following double excitation is indistinguishable from direct ionization leading to pronounced interference structures in the ejected electron-energy spectra [11]. This Fano interference makes the interpretation of the data quite complicated.

Recently, we have pointed out that these problems encountered in studies of double ionization and excitation are not present for capture of one target electron with simultaneous excitation of a second target electron (transfer excitation) [12]. Kinematically, it is even simpler than double excitation since the final state involves only two unbound particles and no continuum electron. As a result, singly differential cross sections readily constitute fully differential cross sections. Furthermore, the theoretical problems related to many-electron continua or Fano interferences do not arise. Finally, for the collision system studied in [12], i.e.,  $p+\text{He}$ , the final electronic state is rather simple and can be represented by a product of hydrogenic wave functions. Therefore, in transfer excitation the electron-electron interaction is unimportant in the final state, in sharp contrast to e.g. double ionization [6,13].

Studies of double capture share some of the advantages of transfer excitation. Here too, the final state only involves two unbound particles (except for double capture to a doubly excited state decaying by autoionization), does not contain continuum electrons and it is not affected by Fano interference. In the case of  $p+\text{He}$  collisions an additional advantage is that the double capture process is intrinsically state selective since there is only one bound state for the  $\text{H}^-$  ion. An interesting aspect of double capture for this collision system is that the final state is highly correlated, in sharp contrast to transfer excitation. This unusually strong correlation manifests itself for example in the large asymmetry between the first and second ionization potential of  $\text{H}^-$  (0.7 and 13.6 eV compared to 24.6 and 54.4 eV for helium). Therefore double capture and transfer excitation measurements are complementary as they probe different aspects of electron-electron correlation effects.

For  $\text{He}^{2+} + \text{He}$  collisions several measurements of total [14–16] and differential [16–18] double capture cross sections, covering a broad range of projectile energies, have been reported. For  $p + \text{He}$  total cross sections have been extensively studied as well [19–22]. However, to the best of our knowledge no differential data for this collision system are available. Only for an Argon target has double capture to protons been measured differentially at very small energy [23]. Various approaches were employed in theoretical studies of double capture, ranging from close coupling methods at small and intermediate projectile energies (e.g., [24–27]) to perturbative techniques, such as e.g. the continuum distorted wave (CDW) approach, at large energies [28–31]. Often, one-electron transition amplitudes are calculated and the double capture cross sections are obtained with an independent electron model, i.e., electron-electron correlation effects are then not accounted for. Correlations in the initial and final states have been accounted for by Deco and Grun [32] and Belkic and Mancev developed a four-body (two nuclei and two electrons) CDW approach, which, at least conceptually, fully accounts for the electron-electron interaction [29,30].

Pronounced structures in the differential double capture probabilities have been experimentally observed at small projectile energies [17] and theoretically predicted at large energies [29,31]. At small energies, these structures can be explained in terms of an interference between transitions occurring at spatially separated regions in the incoming and outgoing paths of the collision. The predicted structures at large energies can be associated with a multiple scattering process, which was originally described in terms of a classical model by Thomas for the case of single capture [33]. This Thomas peak was experimentally observed for single capture [34,35], but not yet for double capture.

In this paper we report first measured differential double capture cross sections for  $p + \text{He}$  collisions for small and intermediate projectile energies ranging from 15 to 150 keV. Furthermore, we present differential double to single capture cross section ratios as a function of the scattering angle. In contrast to the absolute cross sections, pronounced peak structures are observed in these ratios.

## II. EXPERIMENT

The experiment was performed at the ion accelerator of the University of Missouri-Rolla. A proton beam was produced in a hot cathode ion source and extracted at an energy of 2 keV. The beam was then accelerated to energies ranging from 15 to 150 keV and steered through a differentially pumped target gas cell. A set of slits right in front of the gas cell was used to collimate the beam to a size of  $0.1 \text{ mm} \times 0.1 \text{ mm}$ . The projectile charge states after the collision were separated with a switching magnet. The  $\text{H}^-$  ions produced in the collision were then passed through a solid angle defining collimator located after the switching magnet and detected by a channel-plate detector. The projectile scattering angle was scanned by pivoting the accelerator about the center of the target chamber.

The gas cell was about 1.2 cm in length and had entrance and exit holes for the projectile beam with a diameter of

1.25 mm. The helium target gas pressure was kept at 70 mTorr. A pressure dependence of the detected  $\text{H}^-$ -ion rate was recorded for a projectile energy of 50 keV, which is close to the maximum of the total double capture cross sections, and found to be linear up to at least 150 mTorr. Therefore, single collision conditions were realized in the experiment. The base vacuum was about  $1 \times 10^{-7}$  Torr throughout the entire beam line. Because of the earth's magnetic field and the beam-steering elements  $\text{H}^-$  ions and neutralized beam components generated in collisions with the residual gas were spatially separated from the protons and could thus be collimated out of the projectile beam by the very narrow beam-defining slits before the target chamber. As a result, background from the residual gas contributed to the detected  $\text{H}^-$  signal only between the beam-defining collimator and the switching magnet, corresponding to a path of about 1.0 m.

Before the actual measurement of the angular  $\text{H}^-$  distribution an angular scan of the incident beam with no target gas was taken for each projectile energy. For this part of the experiment the field of the switching magnet was reversed so that the protons were recorded with the same detector as used for the  $\text{H}^-$  detection. From the angular distribution of the incident beam, we found an overall angular resolution of  $\pm 75 \mu\text{rad}$ . In the data analysis the incident beam profile was deconvoluted from the measured angular distributions of the  $\text{H}^-$  ions resulting from double capture following the procedure of Park *et al.* [36]. Finally, an angular distribution of  $\text{H}^-$  ions without target gas was taken to determine the background due to capture from the residual gas. Although this background was found to be very small, even at large scattering angles, it was later subtracted from the angular scans taken with the target gas. The deconvoluted and background-subtracted differential cross sections were normalized to a Chebyshev fit to recommended data for total cross sections [37].

## III. RESULTS AND DISCUSSION

The differential double capture cross sections are shown in Fig. 1 for eight projectile energies ranging from 15 to 150 keV. The data exhibit the typical steeply decreasing dependence on the scattering angle, but neither oscillating structures characteristic of interference effects nor peaks reminiscent of the Thomas peak found for single capture at large energies [34,35] are observed. Of course, at the relatively small projectile energies studied here, structures due to Thomas scattering could not have reasonably been expected. On the other hand, interference oscillations due to quasimolecular transitions in spatially separated coupling regions have been observed for similar projectile energies in  $\text{He}^{2+} + \text{He}$  collisions [17]. The absence of such oscillations in our data is probably mostly due to two factors: first, double capture for  $\text{He}^{2+} + \text{He}$  is a perfectly resonant process, while for  $p + \text{He}$  the difference between the electronic final and initial energies is 64.7 eV (i.e., almost five times as large as the final energy in  $\text{H}^-$ ), which leads to a strong damping of the oscillations. Second, in the case of  $\text{He}^{2+} + \text{He}$  probabilities at fixed scattering angles, rather than differential cross sections were measured, i.e., the double capture rates were normal-

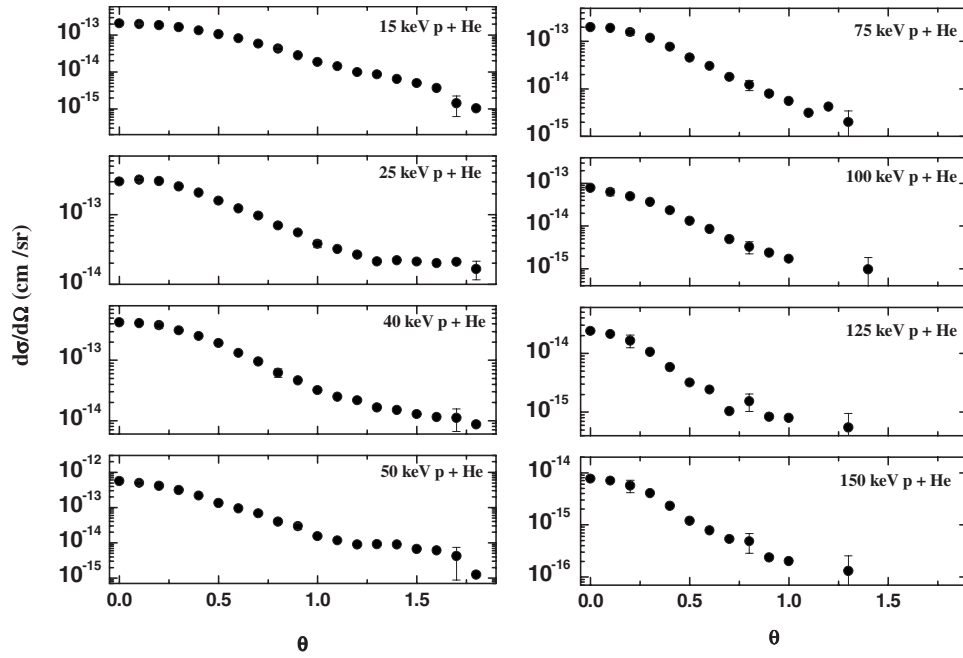


FIG. 1. Differential double capture cross sections in  $p$ +He collisions for projectile energies as indicated in the legends.

ized to the elastic scattering rates. This normalization largely removes the steep dependence on the scattering angle which is characteristic of differential cross sections and which can overshadow structures such as interference oscillations.

In this experiment we did not measure the elastic scattering rate so that we cannot analyze scattering-angle dependent double capture probabilities. However, another possibility to remove the steep angular dependence of the cross sections is to analyze the ratios of double to single capture differential cross sections. Similar ratios between the cross sections for a

two-electron process and the corresponding one-electron process were measured for transfer-ionization (TI) ( $R_{\text{TI}}$ ) [38], double ionization (DI) ( $R_{\text{DI}}$ ) [39], double excitation (DE) ( $R_{\text{DE}}$ ) [9,10], and transfer-excitation (TE) ( $R_{\text{TE}}$ ) [12]. In all cases except for transfer-excitation (where the studied projectile energies were relatively small) peak structures were observed which were not visible, or not as pronounced, in the absolute cross sections.

In Fig. 2 we show the double to single capture differential cross section ratios  $R_{\text{DC}}$  for 25, 50, 75, and 100 keV projec-

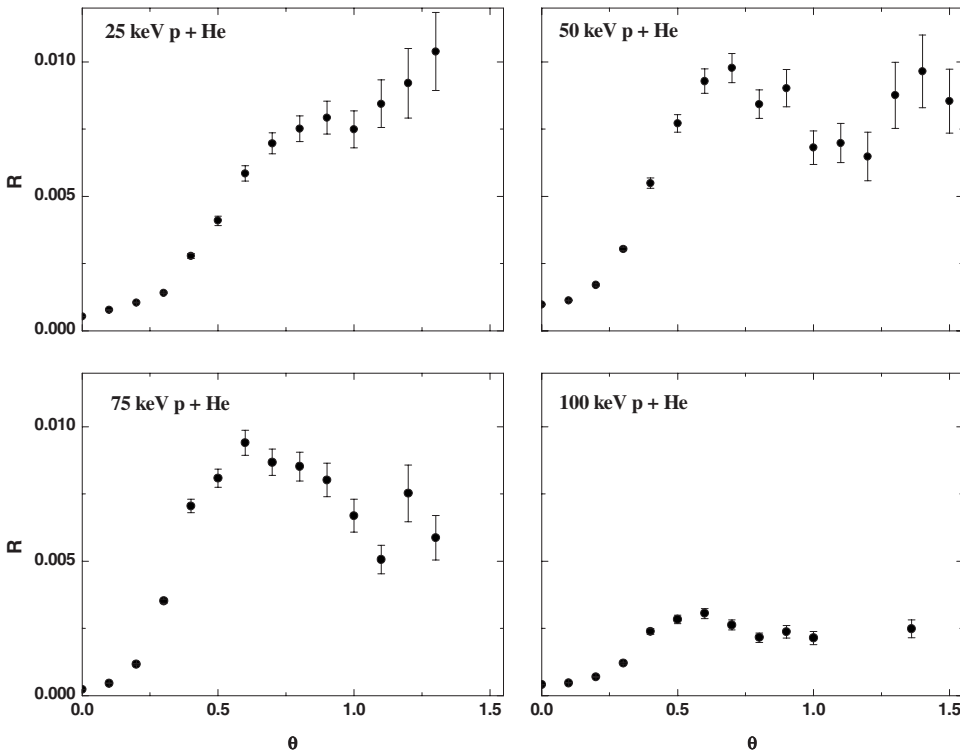


FIG. 2. Differential double to single capture ratios. The single capture cross sections were taken from Hasan *et al.* [12] for 25, 50, and 75 keV (see Fig. 3) and from Martin *et al.* [40] for 100 keV.

tile energies. The single capture cross sections were not measured in this experiment. We therefore used data which we recently obtained from a cold target recoil-ion momentum spectroscopy (COLTRIMS) study [12] for the three smaller projectile energies, shown in Fig. 3, and data of Martin *et al.* for 100 keV [40]. By not measuring the double and single capture cross sections simultaneously using the same detector systems, we are giving up one advantage of analyzing the cross section ratios: the uncertainties due to e.g. the initial beam intensity, the target thickness, or detector efficiencies do not divide out in the ratios, which are thus afflicted with systematic errors just as the absolute cross sections are. On the other hand, for projectile energies of 25 and 50 keV we have two data sets for single capture at our disposal [12,40]. Using both data sets in the ratios yields identical results within the statistical errors.

At all projectile energies, except for 25 keV clear peak structures are observed in  $R_{DC}$ . Even at 25 keV, a pronounced shoulder can be seen at around 0.8 mrad. With increasing projectile energy the peak position slowly moves down in scattering angle to about 0.6 mrad at 100 keV. This insensitivity of the peak position to the projectile energy is a strong indication that the structures in the ratio do not reflect interference oscillations. The phase factor is essentially determined by a time integral of the difference between the energies of the electronic states between which the transition takes place. The involved time scale, in turn, is proportional to the inverse square root of the projectile energy. Therefore, going from the smallest to the largest projectile energy (i.e., from 25 to 100 keV) should change the phase factor roughly by a factor of two. Of course, the effect on the interference pattern would nevertheless be small if the phase factor was small to begin with. However, if the peak structures were indeed due to an interference, the phase factor could not be small because the ratios drop fairly rapidly within a small angular range.

The peak structures observed in the transfer-ionization to single capture ratios  $R_{TI}$  have been interpreted in terms of Thomas scattering of the second kind (T2) [38]. Clear signatures of this process were later found in the recoil-ion momentum spectra [41]. In this two-step mechanism, the projectile first interacts with one target electron which subsequently interacts with a second target electron. The requirement that one electron needs to move at the same speed and in the same direction as the projectile in order to get captured leads to a critical scattering angle of 0.55 mrad for this process, which is consistent with the peak position in our data at least for the two larger projectile energies. However, it is well known that the relative importance of the T2 mechanism increases with increasing projectile energy. In our data, in contrast, the peak appears to be significantly weaker at 100 keV than at 50 and 75 keV, where the relative importance of the structures seems to maximize. This is illustrated in Fig. 4, where the peak height of the structure in  $R_{DC}$  around 0.6 to 0.8 mrad (see Fig. 2) is plotted as a function of projectile energy. For 25 keV a maximum can barely be identified, i.e., the structure is not nearly as pronounced as suggested by Fig. 4. Therefore, the peak structures appear to be most important at projectile energies between 50 and 75 keV. Based on this energy dependence, we discard Thomas scattering as a possible explanation.

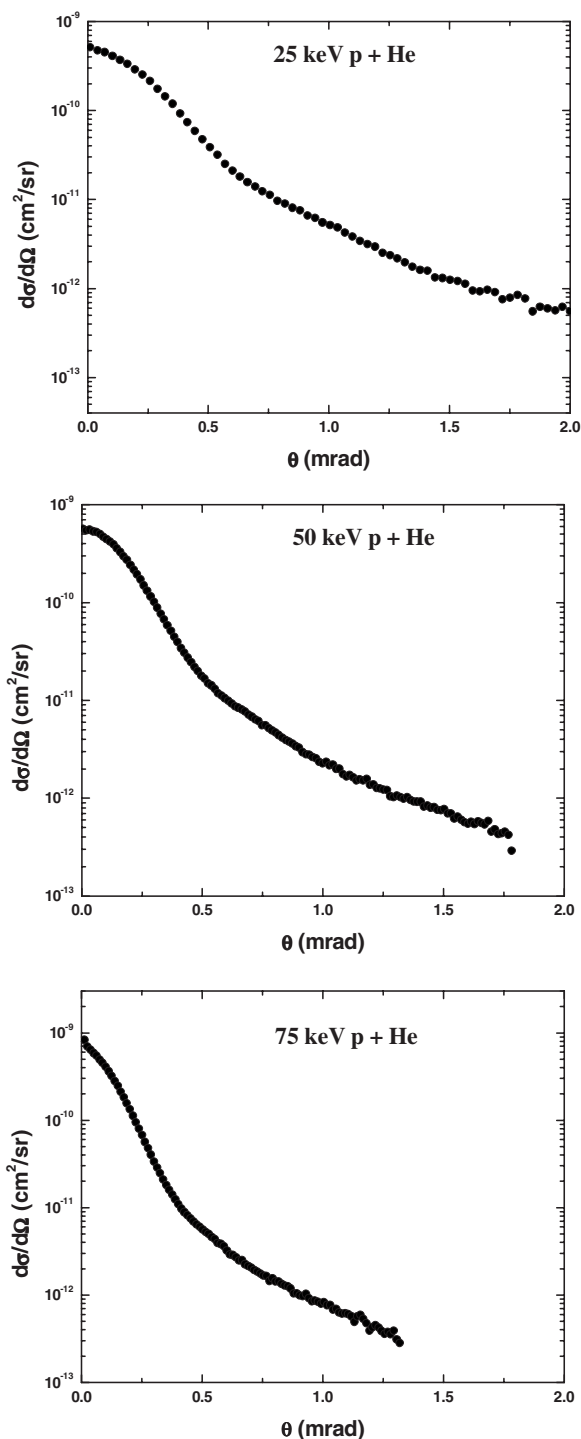


FIG. 3. Differential single capture cross sections in  $p$ +He collisions for projectile energies as indicated in the legends. The data were obtained in the experiment reported in Ref. [12].

The peak structures observed in the double to single ionization ratios  $R_{DI}$  were explained in terms of binary collisions between the projectile and the target electrons [39]. For scattering of an ion by a free electron there is a maximum deflection angle, which is 0.55 mrad for protons. Therefore, in double ionization in  $p$ +He collisions caused by two independent binary interactions between the projectile and the electrons, the maximum angle is 1.1 mrad, which is roughly



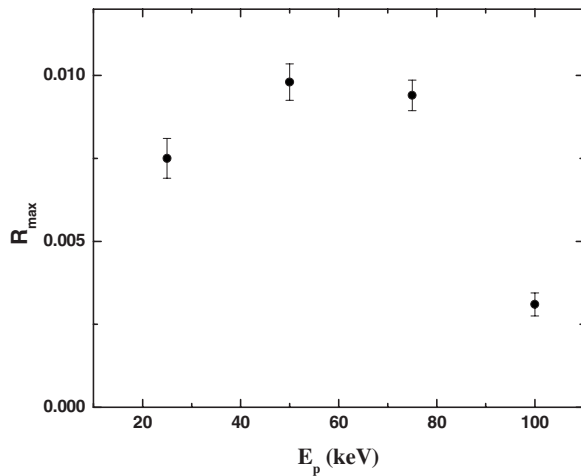


FIG. 4. Peak height of the structure in  $R_{DC}$  around 0.6 to 0.8 mrad in Fig. 2 as a function of projectile energy.

where the peak structures were observed. However, here again, the structures were found to become more pronounced with increasing projectile energy. At small energies the initial bond of the electrons to the target atom and the interaction of the projectile with the target nucleus become increasingly important and both factors have a tendency of diffusing the binary kinematics leading to the maximum scattering angle of 1.1 mrad. Therefore, for the present double capture results the interpretation of the peak structure based on binary projectile—electron collisions can also be discarded.

Both the explanation for the peak structures based on binary projectile-electron collisions in double ionization and the one based on the T2 process in transfer ionization have been criticized by Gayet and Salin [42]. They showed that the experimental ratios can qualitatively be reproduced by theory even when the electron-electron interaction, which is an essential component of the T2 process, is not accounted for. Furthermore, they argued that the collision cannot be analyzed in terms of classical scattering, as it is implied to some extent in the model based on binary projectile-electron interactions. They described both the projectile – electron interaction and elastic projectile-nucleus scattering quantum-mechanically using the eikonal approximation [43] and obtained good agreement with experimental data. Recently, structures observed in the double ratio  $R=R_{TE}/R_{DE}$  [12] were qualitatively reproduced using a similar approach, but the structure did not occur if the elastic scattering was treated classically [44]. The need for a quantum-mechanical description of the elastic scattering could be important for two- and one-electron processes. In our data both the double and the single capture cross sections fall off monotonically with increasing scattering angle. Therefore, the structures in  $R_{DC}$  cannot be unambiguously traced to either process.

As outlined above, both Thomas scattering and binary projectile-electron interactions can essentially be ruled out as explanations for the structures we observe in  $R_{DC}$ . At the same time it was demonstrated that structures observed in  $R_{TI}$  and in  $R_{TE}/R_{DE}$  can be traced to a quantum-mechanical description of the elastic projectile–target nucleus scattering. It therefore seems likely that the structures in  $R_{DC}$  also have their origin in the projectile–target nucleus interaction, rather than in the projectile–electron or electron-electron interaction. However, this does not necessarily imply that the electron-electron interaction is insignificant. As argued above, its influence is expected to be particularly important in the final state involving the highly correlated  $H^-$  ion. Since this final-state correlation does not directly involve the projectile, we expect it to mainly affect the magnitude of the double capture cross sections. We performed a crude qualitative estimate of what the total double capture cross section would be within an independent electron model assuming that both electrons are captured to the same projectile state of equal energy. A comparison to the actual measured cross sections suggests that the final state correlation leads to a reduction of about two orders of magnitude. However, for a complete analysis of final-state correlations calculations with fully correlated wave functions are needed.

#### IV. CONCLUSIONS

We have measured differential double capture cross sections for 15 to 150 keV  $p+He$  collisions and analyzed differential double to single capture ratios. While the cross sections only exhibit a monotonically decreasing angular dependence, clear peak structures around 0.6 to 0.8 mrad are found in the ratios. These structures are probably not due to an interference between transition amplitudes from spatially separated coupling regions, as observed in the angular dependence of double capture probabilities in  $He^{2+}+He$  collisions. The absence of this interference in our data is probably due to the large energy-asymmetry between the initial and final electronic state. We also rule out Thomas scattering as an explanation because the structure does not get more pronounced with increasing impact energy. Rather, the peak structures appear to have their origin in the elastic scattering between the projectile and the target nucleus, which needs to be described quantum-mechanically. Finally, final-state correlations strongly suppress the magnitude of the double capture cross sections.

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