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Production of $(2s^2)^1S$, $(2p^2)^1D$, and $(2s2p)^1P$ states by double electron capture in 150-500-keV ³He²⁺ + He collisions

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The production of $(2s^2)^1S$, $(2p^2)^1D$, and $(2s^2p)^1P$ states formed by double capture into projectile autoionizing states was measured with high resolution using 0° Auger spectroscopy in 150-500-keV ${}^{3}\text{He}^{2+}$ + He collisions. Absolute cross sections for these states were determined to lie approximately between 10^{-18} and 10^{-21} cm², the production of the $(2s^2)^1S$ state being dominant. An extension of a continuous distorted-wave independent-particle model to the calculation of double-electron-capture cross sections was found to overestimate our results by a large factor, ranging between 6 and 75.

In collisions of highly charged projectiles with atoms, single- and multiple-electron-capture processes proceed not only to the ground state but also to the formation of excited projectile states that later decay via photon or electron emission. Recently, the study of such state-selective electron-capture (SSEC) processes has become a very active field of interest, due to important practical implications of these processes for controlled thermonuclear fusion, astrophysics, and the creation of vuv and x-ray lasers among other things (for a recent review, see Ref. 1). In particular, high-resolution studies of *multiple* capture into specific excited states in contrast to single-capture studies, can provide information on the role played by electron-electron interactions.²

To date, however, very little information on stateselective multiple electron capture is available. Measurements of state-selective double-electron-capture (SSDEC) cross sections are tedious, measured yields usually being compromised by the high-energy resolution required. Furthermore, only a few, if any, calculations are applicable. A few experimental attempts have been reported: Energy gain^{3,4} or electron^{2,5} spectroscopy were used to study SSDEC in collisions of C^{4+} , N^{7+} , O^{6+} , and O^{7+} with H₂, He, or Ne, at low impact energies ($\leq 4.5 \text{ keV/u}$). Absolute SSDEC cross sections were not reported. At high energies, high-resolution Auger-electron data⁶ were reported for bare F^{9+} projectiles at $\frac{2}{3}$ MeV/u incident on noble gases. Limitations due to kinematic line broadening effects,⁷ allowed only for the determination of doubleelectron-capture (DEC) n distributions. These broadening effects, however, can be substantially reduced by observing⁸ ejected electrons at 0° with respect to the beam.

In the present work, high-resolution 0° Auger spectros-

copy is used to study SSDEC for the system:

³He²⁺+⁴He(1s²)
$$\rightarrow$$
 He^{**}(2lnl')+He²⁺
 \downarrow He⁺(1s)+e⁻

One or two electrons can be captured by the projectile. In particular, only doubly excited He^{**}(2lnl') ($n \ge 2$) projectile states formed by *double* electron capture decay via Auger electron emission after the collision. Thus, by detecting these electrons with high-resolution information about DEC into these intermediate states is gained. Here we report absolute cross sections for SSDEC only into the three lowest He autoionization states $(2s^2)^1S$, $(2p^2)^1D$, and $(2s2p)^1P$ which were well resolved in the overall electron spectra. Several projectile energies between 50-166 keV/u were utilized.

This is the first systematic study of SSDEC for the fundamental symmetric He^{2+} + He system and its dependence on the collision energy. Our reported SSDEC cross sections at these intermediate collision energies are expected to provide stringent tests of newly developed formalisms^{9,10} for few-electron systems which will bridge the gap between low- and high-energy collisions. In particular, such state-selective two-electron transfer data should provide basic information on the effects of electron-electron interactions.¹¹

The experimental apparatus has been previously described¹² and therefore will not be discussed in detail. A ³He²⁺ beam resulted from stripping ³He⁺ ions which were accelerated through the 500-kV AN Van de Graaff at the Hahn-Meitner-Institut für Kernforschung, Berlin. The beam was magnetically selected, tightly collimated, 1964

and focused ($\leq \sim 1 \text{ mm}^2$) into a 5-cm-long differentially pumped cell filled with 99.996% pure ⁴He gas.

The ejected electrons were detected at 0° with respect to the beam direction by a tandem spectrometer composed¹² of an entrance and exit 45° parallel-plate electrostatic analyzer, subtending an effective solid angle of $\sim 2.2 \times 10^{-4}$ sr. The measured yields were due to Auger electrons ejected at an angle of 0° in the emitter frame from the decay of projectile autoionizing states (21nl'), corresponding to an electron energy range of 32-42 eV in the emitter rest frame. A final resolution of about 120 meV (in the projectile rest frame) was attained by decelerating all detected electrons to 7 eV between the two



FIG. 1. Electron spectra (projectile rest frame) produced in ${}^{3}\text{He}^{2+}$ + He collisions at different projectile energies.

analyzers. The kinematic broadening caused by the finite acceptance (half)-angle of the spectrometer^{7,8} ($\Delta\theta \sim 1^{\circ}$) was only about 30 meV (projectile frame) for a projectile energy $E_p = 500$ keV (worst case) and was neglected. Beam currents ranged between 0.030-1.5 μ A for E_p between 150-500 keV, respectively.

In Fig. 1, high-resolution electron spectra are shown resulting from He²⁺ + He collisions. The measured yields are shown after subtraction of background continuum electrons and transformation to the projectile rest frame. Only spectral lines resulting from the decay of autoionizing states with n=2 are shown, i.e. $(2s^2)^{1}S$, $(2s2p)^{3}P$, $(2p^2)^{1}D$, and $(2s2p)^{1}P$, these being well resolved in the overall electron spectra. The ¹S, ¹P, and ¹D states can be formed by DEC. However, the ³P state cannot be formed by DEC, since this would require one of the two captured electrons to spin flip in order for a spin triplet to be produced. Such spin-flipping processes should be weak in low-Z systems where the spin-orbit interaction is small.

The ${}^{3}P$ state is assumed to be formed by successive oneelectron capture events taking place in two collisions. Evidence for this is demonstrated in Fig. 2, where the target pressure (P_t) dependence for the different states is shown in a typical case of 300 keV He^{2+} + He collisions. In this figure, a linear dependence of the yields on pressure, implying single collision conditions, is depicted as a straight line parallel to the pressure axis. Deviations from such behavior indicate the existence of multiple collision contributions. The three singlet states, ${}^{1}S$, ${}^{1}D$, and ${}^{1}P$, are seen to be well fitted by straight lines parallel to the pressure axis over most of the lower-pressure region, clearly demonstrating their single-collision production origin. However, this is clearly not so for the ${}^{3}P$ state which shows a strong quadratic behavior. For the ${}^{3}P$ state, it appears (see Fig. 2) that when $P_t \ge 1$ mTorr both collisions take place in the target gas, while for $P_t < 1$ mTorr, the first collision takes place predominantly within the beam line before the scattering chamber. This was further verified by making a small leak in the beam line (normally at 5×10^{-7} Torr) at the lowest P_t studied (0.15 mTorr). Only the ³P yield relative to the other three states was increased, thus ruling



FIG. 2. Target pressure dependence of the ${}^{1}S$, ${}^{3}P$, ${}^{1}D$, and ${}^{1}P$ states. Statistical errors are indicated.

out the possible observation of spin flip. The quadratic and linear pressure dependence of the triplet and singlet states, respectively, was checked and confirmed for all projectile energies studied.

Absolute differential cross sections for DEC were extracted directly from the measured electron yields in the single collision region ($P_t \approx 0.68-2 \text{ mTorr}$) using methods described elsewhere.¹² It can be readily shown that for the states studied here total SSDEC cross sections $\sigma_{20}(^1L)$ can be obtained by multiplying the differential cross sections measured at 0° (Fig. 1) by 4π , $4\pi/3$, and $4\pi/5$ for the 1S , 1P , and 1D states, respectively, to account for the angular dependence of the M=0 substrates which are predominantly selected⁷ by observing the electrons at 0°. These DEC cross sections are shown in Fig. 3. Only statistical errors are indicated. Absolute errors are estimated to be smaller than about 30%. The 1S state is seen to be dominantly populated, $\sigma_{20}(^1D)$ and $\sigma_{20}(^1P)$ being about 6-20 times smaller than $\sigma_{20}(^1S)$. DEC to the 1P state is slightly larger than to the 1D . The projectile energy dependence of the three states is seen to be quite similar at the higher energies.

To date, no calculations exist for SSDEC. Recently, Salin, Bachau, and Gayet¹³ calculated SSDEC cross sections into the ¹S, ¹D, and ¹P autoionizing states for this system by extending the independent electron model of Gayet, Rivarola, and Salin.¹⁴ These states were described through a configuration interaction treatment¹⁵ using antisymmetrized hydrogenic orbitals. This model assumes that the electron-electron interaction is switched off during the transition. Each electron is then captured independently to a final He⁺ (nlm) state with amplitude $a_{nlm}(b)$, computed in a continuous distorted wave (CDW) approximation¹⁶ and a total amplitude A(LMS) for DEC is obtained from the product of the single-electron amplitudes, i.e.,

$$A^{LMS} \propto \sum_{n_1 l_1 n_2 l_2} \sum_{m_1 m_2} \langle l_1 l_2 m_1 m_2 | LM \rangle a_{n_1 l_1 m_1}(b) a_{n_2 l_2 m_2}(b) ,$$

from which cross sections

$$\sigma_{20}(LMS) = 2\pi \int b \, db \left| A^{LMS}(b) \right|^2$$

are obtained. The most important impact parameters b range between 0-1 a.u. Calculations break down below ~ 250 keV, as the limit of validity of the CDW method is reached.

The results of these calculations for $\sigma_{20}(LMS)$, with M=0, are also included in Fig. 3. The CDW SSDEC cross sections are seen in general to be much larger than the measured ones by factors ranging from 6 for the ¹S state up to 75 for the ¹P state. A more sophisticated calculation in which the first electron is captured into a He²⁺ ion and the second into the remaining He⁺ ion, rather than both electrons being captured into a He²⁺ ion (as assumed above), should lead to smaller DEC cross sections, since σ_{10} can be about an order of magnitude smaller¹⁷ than σ_{21} . Furthermore, electron correlation effects¹¹ not treated by this independent particle calculation should partially account for some of this discrepancy.

In conclusion, state-selective double-electron capture



FIG. 3. Absolute SSDEC cross sections σ_{20} for the ¹S, ¹D, and ¹P states. Only statistical errors are indicated. Absolute errors $\leq 30\%$. Dashed lines: CDW calculation (Ref. 10) with M=0 already divided by numbers in parentheses.

(SSDEC) into autoionizing states of He^{**}(2lnl') was studied for He²⁺ + He collisions at different projectile energies between 150-500 keV. Using the simplicity and high-resolution capabilities of 0° Auger spectroscopy, the three lowest doubly excited projectile states of He, i.e., the $(2s^2)^1S$, $(2s^2p)^1P$, and $(2p^2)^1D$ states, were clearly resolved and SSDEC cross sections into these states were measured with excellent statistics. These were found to be of the same order of magnitude as the transfer-excitation cross sections measured for $He^+ + He$ collisions.¹² The only available SSDEC calculation,¹³ an independent particle CDW approximation, was found to overestimate SSDEC cross sections by large factors ranging from 6 to 75, pointing to the lack of adequate understanding of SSDEC even for the simplest existing two-electron system. These and other such measurements could be an important testing ground for newly developed more sophisticated formalisms that include electron-electron interactions,¹¹ such as time-dependent Hartree-Fock⁹ or atomic-orbital coupled-channel¹⁰ calculations which are expected to be quite accurate in the intermediate velocity regime and should soon be available for the study of SSDEC.

Note added in proof. Improved agreement (about a factor of 2 or better than the CDW calculation) with our SSDEC's has very recently been obtained by an independent particle coupled-channel approach (AO+) valid over the entire 100-500-keV energy range.

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