State-Selective Single-Electron Capture in Low-Energy N^+-He Collisions

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Abstract

Employing a reaction microscope spectrometer with a double-coincidence method, we experimentally measured the three-dimensional momentum of recoil He+ ions from single electron capture in collisions of 0.64 keV/u N+ with He, obtaining state-selective cross sections and angular distributions for electron capture into different quantum states of the projectile ion. The experimental results indicate that the ground-state projectile ion N+(1s22s22p2 3P) captures a 1s electron from the He target, populating predominantly the 2p shell with visible contributions from higher shells; the metastable projectile ion N+(1s22s2p3 5S) captures a 1s electron from the He target, populating mainly the 2s shell with almost no observable contribution from higher shells. The potential energy curves of the NHe+ quasimolecular ion provide a qualitative explanation for the experimental results, but the reaction window predictions of the molecular Coulombic over-the-barrier model show significant discrepancies with the experimental data. In the process of electron capture into the 2s orbital by the metastable projectile ion N+(1s22s2p3 5S), the angular differential cross section displays a pronounced oscillatory structure, which likely arises from Demkov-type transitions.

Full Text

State-selective single electron capture in slow N^+-He collisions

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Abstract

Using a reaction microscope with a two-fold coincidence method, we experimentally measured the three-dimensional momentum of recoil He^{+} ions from single-electron capture in 0.64 keV/u N^{+} collisions with He, obtaining state-selective cross-sections and angular distributions for electron capture into different quantum states of the projectile ions. The experimental results show that ground-state projectile ions N^{+}(1s\$$^{\{2\}2s}$\{2\}2$p$^\{2\}\$ ^{3}P) capture a 1s electron from the He target primarily into the 2p shell, with contributions to higher shells also observable; metastable projectile ions N^{+}(1s\$$^{\{2\}2s2p}$\{3\}\$ ^{5}S) capture a 1s electron from the He target mainly into the 2s shell, with almost no observable contribution to higher shells. The experimental results are qualitatively interpreted using potential energy curves of the NHe^{+} quasimolecular ion, but the reaction window predictions of the molecular Coulomb over-barrier model differ significantly from the experiments. During the process of metastable projectile ions N^{+}(1s\$$^{\{2\}2s2p}$\{3\}\$ ^{5}S) capturing target electrons into the 2s orbital, a clear oscillatory structure appears in the angular differential cross-section, which likely originates from Demkov-type transitions.

Keywords: single-electron capture; state-selective cross-section; angular differential cross-section; metastable states; ion-atom collisions

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Introduction

Studies of electron capture processes in ion-atom collisions are not only crucial for the development of fundamental theories of atomic structure and collision dynamics, but also have broad applications in astrophysics and plasma physics. For example, from an application perspective, de-excitation spectral lines of N atoms have been observed in many nebulae, attracting widespread theoretical attention [1-3]. These excited N atoms may originate from collisional excitation, radiative recombination, charge exchange, and other processes in low-energy ion-atom collisions. Experimental investigations of electron capture (charge exchange) for low-energy N^+ ions can provide important foundations for theoretical refinement [4].

As early as 1977, the Vujovic group [5] separated the ground-state and metastable incident ions (hereinafter uniformly referred to as projectiles) participating in the N^+-He capture reaction using beam attenuation techniques [6], obtaining ground-state projectiles and three types of metastable projectiles with high purity (within 10% error), and measured the single-electron capture cross-sections in collisions with He atomic targets. Based on quasimolecular theory analysis of their results, they found that the $N^+(5S)$ state dominated the single-capture reaction for metastable projectiles with He targets. To investigate the scattering cross-sections of metastable projectiles with different targets, Moran et al. [7] conducted studies on total electron capture cross-sections for 1-3 keV $N^+(3P)$ and $N^+(1D)$ collisions with various neutral molecular gas targets. They found that the influence of metastable projectiles on capture cross-sections for different targets was universal, meaning that the probability of target electrons being captured by projectiles depends on the internal energy of the projectiles. Subsequently, the Nutt group [8] measured single-electron capture cross-sections for ground-state C^+, N^+, and O^+ ions colliding with atomic hydrogen and hydrogen molecules in the 0.1-13 keV energy range. The McAfee group [9] studied single-electron capture processes of $N^+(1S)$ and $N^+(5S)$ ions with nitrogen gas targets. Toshio Kusakabe’ s group [10] obtained ground-state projectiles with high purity by controlling electron collision energy and reported systematic studies on single-electron capture in collisions of C^+, N^+, and O^+ projectiles with high ground-state purity with He atoms at 1-3 keV, investigating the effects of different projectile types on single-capture cross-sections. Although the dependence of capture cross-sections on projectile internal energy has been commonly observed experimentally [7], the underlying mechanisms remain poorly understood. To provide theoretical explanations, the Kimura group [11] employed a more accurate semiclassical close-coupling model to calculate the energy dependence of state-selective single-electron capture cross-sections. The cross-sections for $N^+(5S)$ and $N^+(3P)$ projectiles capturing target electrons to the ground state agreed well with the total single-electron capture cross-sections obtained by the Vujovic group. For N^+ ion collisions with He, they found that as collision energy increases, the capture cross-section for metastable projectiles exceeds that for ground-state projectiles at approximately 0.4 keV/u. The aforementioned work primarily focused on total single-electron capture cross-sections, and to date, no state-resolved single-electron capture experiments for low-energy N^+ ions colliding with He have been reported. In this work, using 0.64 keV/u N^+ projectiles (which may be in ground or metastable states) provided by the EBIS ultra-low-energy heavy ion experimental platform at the Institute of Modern Physics [12-13] and based on a reaction microscope [12-13], we have investigated state-selective electron capture processes in N^+ and He collisions, obtaining state-selective cross-sections and angular differential cross-sections for captured electrons populating both the projectile ground-state level and higher excited-state levels, and explored the relevant charge transfer mechanisms in the collision process.

2 Experimental Setup

[Figure 1: see original paper] shows a schematic diagram of the experimental apparatus, which has been described in detail in previous publications [12-13]; we provide a brief introduction here. Ions are produced in an electron beam ion source (EBIS-A) and extracted, then filtered by a Wien velocity selector to obtain N^{+} ions. The ions are accelerated to 0.64 keV/u by a high-voltage platform, focused by electrostatic lenses, calibrated by deflection plates, and collimated through a slit before entering the collision chamber. In the collision chamber, the N^{+} ions intersect with a supersonic helium atomic jet directed opposite to the Y direction. Recoil ions from electron capture reactions are extracted from the collision region by a transverse electric field (3.7 V/cm) perpendicular to the beam direction, pass through a time-of-flight (TOF) spectrometer system, and are finally detected by a two-dimensional position-sensitive detector with a delay-line anode. The spectrometer’ s acceleration and drift region lengths are 107.5 mm and 215 mm, respectively, satisfying the 1:2 one-dimensional time-focusing condition. Neutral nitrogen atoms produced by capture reactions are collected by a two-dimensional position-sensitive microchannel plate detector located downstream of the beam. Projectile ions that did not participate in capture reactions are deflected by a charge analyzer downstream of the beam and collected by a Faraday cup (FC). Finally, the three-dimensional momentum of recoil ions is reconstructed from their two-dimensional positions and flight times. In the experiment, the beam direction is defined as the Z direction. The recoil ion momentum component along Z is called longitudinal momentum, which is related to the reaction energy loss Q. The Q-value is defined as the change in binding energy of the participating electrons before and after the reaction; thus, the experimentally obtained Q-value spectrum reflects the state of the projectile after electron capture. For example, the Q-value corresponding to metastable projectile N^{+}(1s\$$^{\{2\}2s2p}$\{3\}\$ ^{5}S) capturing a 1s electron from He to the 2s shell is (14.53-24.59+5.8) eV, where 14.53 eV is the first ionization energy of N, 24.59 eV is the first ionization energy of He, and 5.8 eV is the energy difference between the metastable N^{+}(1s\$$^{\{2\}2s2p}$\{3\}\$ ^{5}S) and the ground-state N^{+} level. The recoil ion momentum component perpendicular to the beam direction is called transverse momentum, which is related to the scattering angle of the scattered ion.

As described in the experimental setup section, the data collected by the reaction microscope for pure electron capture processes can be analyzed to obtain primarily two types of information: energy loss spectra and angular distributions. The energy loss spectrum reveals the population states of electrons captured by the projectile, while the angular distribution reflects the dynamics of electron capture. Under the small-angle scattering approximation and based on momentum and energy conservation before and after the collision, the energy loss Q-value for single-electron capture processes relates to the recoil ion longitudinal momentum P_z as follows (unless otherwise specified, all subsequent discussions are in atomic units):

$$Q = -v_p \cdot P_z -$$

where v_p is the projectile velocity and n is the number of captured electrons. Under the small-angle approximation, the relationship between recoil ion transverse momentum P_t and projectile scattering angle θ is:

$$\theta = P_t / P_0$$

where P_0 is the initial projectile momentum.

3 Results and Discussion

3.1 Energy Loss Spectrum

[Figure 2: see original paper] presents the Q-value spectrum for single-electron capture reactions in 0.64 keV/u N^+ collisions with He. The dotted-solid line represents the experimental results. As can be seen from the figure, the rightmost peak corresponds to a channel with a singlet final state (4S), and its full width at half maximum reflects the experimental energy resolution, approximately 1.4 eV. The solid line shows predictions from the molecular over-barrier model [15] considering two molecularized electrons, while the dashed line considers only one molecularized electron. Above Figure 2 are the reaction equations for the single-electron capture process, where *nl* corresponds to the labels above each peak; upright labels represent ground-state projectile capture processes, and italic labels represent metastable projectile capture processes. The figure reveals that the capture reaction primarily involves ground-state projectiles $\mathrm{N}^+(1\mathrm{s}\$^{\{2\}2s}\{2\}2p\hat{}\{2\}\$\ {}^3\mathrm{P})$ and metastable projectiles $\mathrm{N}^+(1\mathrm{s}\$^{\{2\}2s2p}\{3\}\$\ {}^5\mathrm{S})$ (the metastable lifetime is about 5.4 ms [16], and the ion flight time from source to collision region is approximately 6 s). For ground-state projectiles, the dominant contribution to single-electron capture is the process where projectile $\mathrm{N}^+(1\mathrm{s}\$^{\{2\}2s}\{2\}2p\hat{}\{2\}\$\ {}^3\mathrm{P})$ captures a 1s electron from He into the 2p shell, forming $\mathrm{N}(1\mathrm{s}\$^{\{2\}2s}\{2\}2p\hat{}\{3\}\$)$ scattered ions, with a secondary contribution from capture into higher *nl* shells. For metastable projectiles, the main contribution is the process where projectile $\mathrm{N}^+(1\mathrm{s}\$^{\{2\}2s2p}\{3\}\$\ {}^5\mathrm{S})$ captures a 1s electron from He into the 2s shell, forming $\mathrm{N}(1\mathrm{s}\$^{\{2\}2s}\{2\}2p\hat{}\{3\}\$\ {}^4\mathrm{S})$ states, with almost no contribution from capture into higher shells. Based on the total single-electron capture cross-sections for N^+(5S) and N^+(3P) projectiles with He targets given in reference [11], combined with the count ratio of captured ground-state and metastable projectiles measured in our experiment, we determined that the fraction of metastable 5S projectiles in the beam is about 5%. This method has been reported in previous work for determining the fraction of C^{3+} ions in a mixed beam with the same mass-to-charge ratio (C^{3+} and O^{4+} mixed beams, both with mass-to-charge ratio of 4) [17].

[Figure 3: see original paper] shows potential energy curves (partial) for the NHe^+ quasimolecular ion calculated in reference [14], with three line thicknesses labeled as type A, B, and C from thick to thin. In the quasimolecular picture, type A solid lines correspond to potential energy curves for quasimolecular ions formed by ground-state projectiles and He, type B solid lines correspond to excited-state projectile N^+(3D) and He, and type A dotted lines correspond to metastable projectile N^+(1s\$$^{\{2\}2s2p}$\{3\}\$ ^{5}S) and He. Due to transition selection rules, type C lines correspond to possible final-state channels for ground-state projectiles, and type B dotted lines correspond to possible final-state channels for metastable projectiles; see the legend in Figure 3 for details. For easier illustration, some guiding arrows are marked in the figure. Thick arrows represent possible potential energy change paths during capture processes for ground-state projectiles. The possible path is as follows: for the process where ground-state projectile N^+(1s\$$^{\{2\}2s}$\{2\}2$p$^\{2\}\$ ^{3}P) captures a 1s electron from He into the 2p shell to form N(1s\$$^{\{2\}2s}$\{2\}2$p$^\{3\}\$ ^{4}S), the ground-state projectile first undergoes a collisional excitation process with the target atom at an internuclear distance of about 2 a.u. (avoided crossing), exciting the ground-state projectile to the 3D state, then at about 3 a.u. there is a high probability of transitioning to the ground state 4S [11] (due to a real crossing between type B solid and type C dashed lines). Subsequent population of 2D and 2P states lacks avoided crossings and real crossings, which hinders radial coupling paths similar to capture to 4S [14]. This process may be caused by rotational coupling or by perturbative symmetric electron transitions occurring at large internuclear distances, also known as Demkov-type transitions [18-19], which often correspond to oscillations in angular differential cross-sections. Thin arrows represent possible potential energy change paths during capture processes for metastable projectile N^+(1s\$$^{\{2\}2s2p}$\{3\}\$ ^{5}S). The process where N^+(1s\$$^{\{2\}2s2p}$\{3\}\$ ^{5}S) projectile directly captures a 1s electron from He into the 2s shell to form N(1s\$$^{\{2\}2s}$\{2\}2$p$^\{3\}\$ ^{4}S) (type A dotted and type B dotted lines) also lacks avoided crossings and real crossings, and similar rotational coupling and Demkov-type transition processes may occur. Under the current experimental resolution, this process appears as an isolated peak; further explanation will be provided in the subsequent angular distribution discussion. In summary, single-electron capture to the 4S state by ground-state projectiles is a radial coupling mechanism, while capture to 2D and 2P by ground-state projectiles and capture to the 4S ground state by metastable projectiles are likely dominated by the Demkov-type mechanism. Further investigation of this mechanism requires more experimental and theoretical studies.

3.2 Angular Differential Cross-Section

[Figure 4: see original paper] presents the angular differential cross-sections for single-electron capture reactions in 0.64 keV/u N^+ collisions with He. The black dotted-solid line shows the total angular differential cross-section, the red line shows the angular differential cross-section for the metastable projectile capture to the ground-state channel, and the blue and green lines show the angular differential cross-sections for ground-state projectile capture to the ground state and to higher shells, respectively. The red solid line exhibits clear oscillations.

One possible oscillation mechanism is the Demkov-type transition mechanism mentioned earlier. General characteristics of Demkov-type transitions include [19]: oscillations appear in angular differential cross-sections, transitions occur at large internuclear distances, transition probabilities generally increase with collision energy, and the adiabatic potential energy curves in the quasimolecular picture typically have no avoided crossings or real crossings. According to the results in reference [11], the energy dependence of state-selective cross-sections for this channel is consistent with the Demkov mechanism. As seen in [Figure 3: see original paper], the potential energy curves of the entrance and exit channels have no avoided crossings or real crossings, and our experimental results also show strong oscillations in the angular differential cross-section for this channel. Another possible mechanism is Fraunhofer diffraction of the projectile ion' s matter wave on the target atom [20-24], which mainly appears in s-s channel charge transfer. This mechanism was first proposed by M. van der Poel [20] through studies of single-electron capture in low-energy Li^+ ion collisions with Na targets, providing a good explanation for experimental results. Later, Agueny [23] calculated charge transfer angular differential cross-sections for ^3He ions with He and protons with He from a theoretical perspective, showing good agreement with existing experiments [25]. Wang et al. [22] successfully applied this mechanism to explain single-electron capture in He^{2+} and He collisions, and Guo et al. [24] used it to explain single-electron capture in C^{4+} and He collisions. Analogous to optical Fraunhofer diffraction, for s-s channel electron capture in ion-atom collisions with reduced mass and velocity v, the capture differential cross-section under the Eikonal approximation has the following approximate form:

$$\sigma(\theta) = k^2 \left| \int db\, b J_0(kb\theta) a(b) \right|^2$$

where $k = 2\pi/\lambda_{dB} = \mu v/\hbar$, λ_{dB} is the matter wavelength, $a(b)$ is the scattering amplitude for this channel, and J_0 is the Bessel function. For the s-s channel transfer in this system, using a two-state model [22] and considering polarization effects in the entrance and exit channels, the calculated maximum value of the interaction region is about 2 a.u. Based on this maximum interaction region, we calculated a set of alternating bright and dark fringes (black portions) as shown in [Figure 5: see original paper], where the black vertical line represents the calculation result for an "aperture" radius of 2 a.u. As can be seen, the first dark fringe corresponds to the experimental results, while other fringes do not match the experimental results, indicating that this mechanism may contribute to the experimental results but is not the main cause of the oscillations. In summary, the oscillations in the angular differential cross-section for metastable projectile capture to the ground-state channel are most likely caused by the Demkov mechanism.

Using a reaction microscope, we have measured state-selective cross-sections and angular differential cross-sections for single-electron capture in 0.64 keV/u N^+-He collisions. The energy loss spectrum shows that capture of He 1s electrons into the 2p shell by ground-state projectiles is the dominant reaction channel, with contributions from capture into higher *nl* shells also observed. For metastable projectiles N^{+}(1s\$$^{\{2\}2s2p}$\{3\}\$ ^{5}S), capture of He 1s electrons into the 2s shell is the main contribution, with almost no observable capture into higher *nl* shells. Analysis of the angular differential cross-sections for each channel reveals that capture of He 1s electrons into the 2p shell by ground-state projectiles contributes primarily at small angles, while capture into 3s and higher shells favors larger angles. For metastable incident ions N^{+}(1s\$$^{\{2\}2s2p}$\{3\}\$ ^{5}S), capture of He 1s electrons into the 2s shell favors small angles and exhibits oscillatory behavior. The study indicates that single-electron capture to the 4S state by ground-state projectiles is a radial coupling mechanism, while capture to 2D and 2P by ground-state projectiles and capture to the 4S ground state by metastable projectiles likely originate from Demkov transitions. Further investigation of this mechanism requires additional experimental and theoretical studies.

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