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Polarization degree differences for the $3p\,^2P_{3/2}$–$3s\,^2S_{1/2}$ transition of $N^{4+}(3p\,^2P_{3/2})$ produced in N^{5+}-He and N^{5+}-H_2 collisions

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The magnetic substate-selective single-electron-capture cross sections in collisions of N^{5+} with He and H_2 are calculated using the two-center atomic orbital close-coupling method, and the polarization of emitted radiation from the excited state of N^{4+} is investigated for projectile energies between 1.2 and 7 keV/u. The polarization degrees for the $3p\,^2P_{3/2}$–$3s\,^2S_{1/2}$ transition of $N^{4+}(3p\,^2P_{3/2})$ produced in N^{5+} + He and $N^{5+} + H_2$ electron-capture collisions are in general agreement with the experimental measurements. It is found both experimentally and theoretically that there exists a large difference between the polarization degrees of this radiation resulting from the N^{5+} + He and $N^{5+} + H_2$ electron-capture collisions, namely, ∼0.25 and ∼0, respectively. By studying the time evolution of electron-capture dynamics in the two systems we have found that this difference is caused mainly by the difference in the interactions in the two systems at relatively small internuclear distances, consistent with the molecular picture of the collision dynamics.

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In electron-capture processes of highly charged ions with atoms and molecules, the captured electron usually populates the projectile excited states that decay by line radiation. Due to the fixed projectile velocity direction, the emitted lines can be polarized, and the polarization degree can give information about the magnetic substate populations. This allows one to probe the collision dynamics on a more fundamental level. Numerous experiments have been devoted in the past [1–8] to the determination of photon polarization degree in collisions of multicharged ions with atomic and molecular targets. In the keV energy region, most of the experiments have measured the polarization of emitted radiation following the single-electron capture in collisions of highly charged ions with alkali-metal atoms [4–6], and polarization degrees of 15∼40% have been observed. However, in the experimental studies involving the charge transfer processes of He-like C^{4+}, N^{5+}, and O^{6+} ions with He and H_2 targets [5,7,8], it was found that the polarization degree for the $3p\,^2P_{3/2}$–$3s\,^2S_{1/2}$ transition lies in two regions: 0∼7% for the O^{6+} + He and $N^{5+} + H_2$ collisions, and 17–21% and 16–24% for the N^{5+} + He and $C^{4+} + H_2$ collision systems, respectively. Particularly intriguing is the large difference in the polarization degrees for the N^{5+} + He and $N^{5+} + H_2$ systems in which the projectile is the same ion and both targets are two-electron systems.

On the theoretical side, the classical trajectory Monte Carlo (CTMC) method has been widely used to calculate the polarization degree of emitted radiation from excited states created in charge transfer processes of fully and partially stripped ions with alkali-metal atoms [2,9]. Although the agreement between experimental measurements and theoretical calculations in most cases was found good, obtaining the magnetic state-selective cross sections from the fully classical distributions is an art. Two decades ago, Salin [10] calculated the magnetic (m)-state-selective capture cross sections in hydrogen-atom fully stripped multicharged ion systems by using the semiclassical molecular orbital close-coupling (MOCC) method and discussed the role of the Stark effect in the angular momentum (l) distribution of captured electrons in the field of residual target ions (see also [11]). While the m-state distribution of captured electrons is determined mainly by the interactions at small internuclear distances (rotational coupling of molecular states), the radial couplings (at avoided molecular energy curve crossings) at intermediate internuclear distances and the Stark state mixing at large internuclear distances determine the distribution of its angular momentum states.

The l-state-selective electron-capture cross sections for the N^{5+} + He and $N^{5+} + H_2$ collision systems have been calculated by the MOCC method [12,13] and measured by the photon emission spectroscopy method [7]. But to the best of our knowledge, no polarization degree calculations have been reported for these collision processes and the question of the observed large difference in their polarization degree remains open.

In the present work, the polarization degrees for the $3p\,^2P_{3/2}$–$3s\,^2S_{1/2}$ transition of $N^{4+}(3p\,^2P_{3/2})$ produced in N^{5+} + He and $N^{5+} + H_2$ single-electron capture processes have been calculated and the dynamical mechanism of their differences in two processes has been established. The collision dynamics was described within the two-center atomic orbital close-coupling (TC-AOCC) method with plane-wave translational factors [14]. In the energy range 1.21–10 keV/u considered in our study, the TC-AOCC method should provide an adequate description of collision dynamics of the aforementioned processes provided the TC-AO basis is adequately large. Atomic units will be used throughout, unless explicitly indicated otherwise.

The atomic orbitals on the ion cores N^{5+}, He^+, and H_2^+ have been obtained by solving the Schrödinger equation with the model potentials [12], [15], and [16].

The eigenvalue problem with the aforementioned potentials has been solved variationally [17]. The calculated energy levels are in good agreement with the available data [18].

The present AOCC calculations included all the states centered on N^{5+} with $n \leqslant 6$, all the states centered on He$^+$ with $n \leqslant 3$, and the $1s$–$6s$ states centered on H_2^+. They include all magnetic m states within a given nl subshell. If the m distribution of (l, m) states is not statistical, then the emitted photon spectrum is polarized.

Due to the cylindrical symmetry around the beam axis, the magnetic substate-selective cross section $\sigma_{lm} = \sigma_{l-m}$. In this work, we consider only the polarization degree for the $3p\,^2P_{3/2}$–$3s\,^2S_{1/2}$ transition of N^{4+}($3p\,^2P_{3/2}$) and, consequently, only the population of the $3p_0$ and $3p_1$ electron-capture states is required. Polarization degree for the $3p\,^2P_{3/2}$–$3s\,^2S_{1/2}$ transition has the form

$$P = \frac{6\sigma(3p_0) - 3\sigma(3p_1)}{10\sigma(3p_0) + 7\sigma(3p_1)}, \tag{1}$$

and $\sigma(3p_1) = \sigma(3p_{+1}) + \sigma(3p_{-1})$. The populations of excited states are, thus, directly proportional to the electron-capture cross sections.

In Figs. 1(a) and 1(b) we present the state-selective cross sections for capture to $3l$ states of the N^{4+} ion for the N^{5+}-He [Fig. 1(a)] and N^{5+}-H$_2$ [Fig. 1(b)] collision systems, respectively, which are the predominantly populated capture states in the considered energy range. The corresponding experimental data [20,21] and the MOCC results [22] are also given in this figure for comparison. All the results are mutually consistent regarding the energy behavior of the cross section but there exist discrepancies regarding its magnitude. The disagreement of our results with experimental data may be ascribed to the inadequate account of the electron correlations by the adopted one-particle model potentials. We note that the MOCC results of Ref. [22] are also outside the experimental error bars for most of the energies, which demonstrates the difficulties in the calculations of state-selective electron cross sections in many-electron systems. It is noteworthy that in the N^{5+}-He collision system, the dominant channel in the low-energy region is the capture to the $3s$ state, while for the N^{5+}-H$_2$ reaction, it is the capture to the $3p$ and $3d$ states. This difference is mainly due to the different electronic structures of the He atom and the H$_2$ molecule.

Figure 2 shows the calculated polarization degree, P, of the line corresponding to the $3p\,^2P_{3/2}$–$3s\,^2S_{1/2}$ transition of the N^{4+} ion as a function of the projectile energy for N^{5+}-He [panel (a)] and N^{5+}-H$_2$ collisions [panel (b)], respectively. In

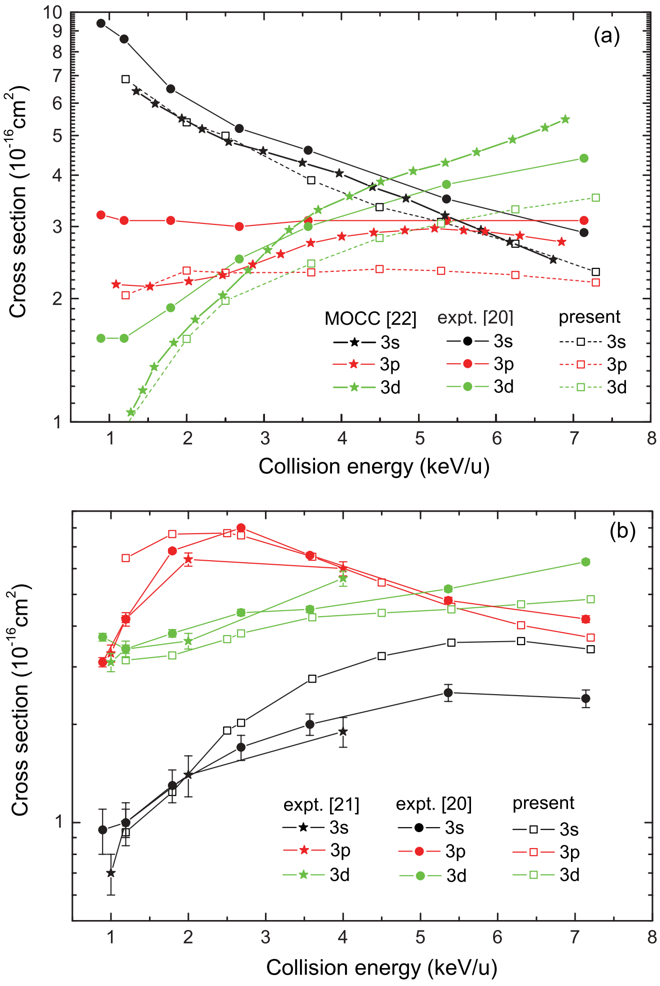

FIG. 1. (Color online) Cross sections for electron capture to the $3l$ states of the N^{4+} ion for the N^{5+}-He (a) and N^{5+}-H$_2$ (b) collision systems. The solid symbols are the experimental data of Refs. [20,21] and the MOCC calculation of Ref. [22] and the open symbols are the present AOCC results.

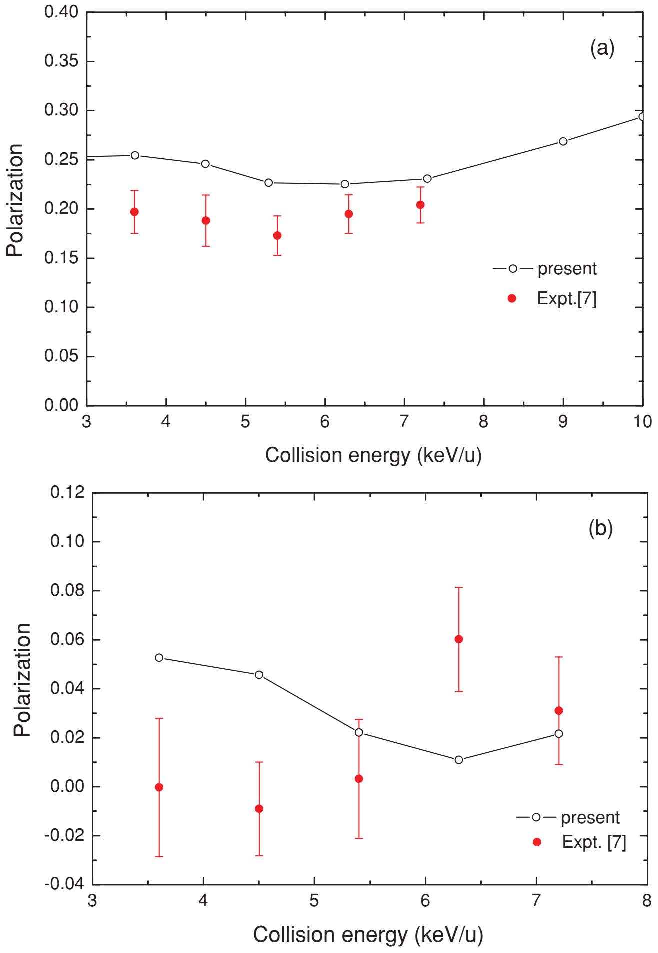

FIG. 2. (Color online) The degree of polarization for the line corresponding to the $3p\,^2P_{3/2}$–$3s\,^2S_{1/2}$ transition as a function of the projectile energy for N^{5+}-He (a) and N^{5+}-H$_2$ (b) collisions. Open circles: present result; solid circles: experimental data of Ref. [7].

both collision systems the $3l$ capture states are the dominantly populated, so in the considered energy range the contribution to the $3l$ populations from the radiative cascades from the higher nl states can be neglected. (For the H_2 target the $4d$ state is only about two times less populated than the $3p$ state, but its radiative decay to the $2p$ states is about an order of magnitude larger than that to the $3p$ state.) The polarization degree shown in Fig. 2 has been, therefore, calculated directly by Eq. (1). The calculated polarization degrees are compared with the experimental values measured in Ref. [7]. For the N^{5+}-He system, the calculated polarization degree is around 0.25 for collision energies of 3–8 keV/u. This corresponds to the ratios of m-partial cross sections: $\sigma(3p_0) : \sigma(3p_{-1}) : \sigma(3p_{+1}) = 2.71 : 1 : 1$, indicating a strong departure from the statistical distribution of magnetic sublevel population in $N^{4+}(3p)$. The polarization varies smoothly in the considered energy range, and this behavior, including the minimum around 5.3 keV/u, is similar to that observed experimentally [7]. It is, however, about 15–25% higher than the measured value of about 0.20. In view of the use of model potentials to represent the interactions of the active electron with the ion cores in its initial and final states, this level of agreement can be considered quite satisfactory.

For the N^{5+}-H_2 collision system, the calculated polarization degree is less than 0.06 in the energy range 3–8 keV/u. This corresponds to a near-statistical distribution of magnetic sublevel populations, with calculated m-partial cross section ratios $\sigma(3p_0) : \sigma(3p_{-1}) : \sigma(3p_{+1}) = 1.27 : 1 : 1$ for $P = 0.06$ and $1.08 : 1 : 1$ for $P = 0.02$. In the experiment in [7] a polarization degree of less than 0.06 has also been observed in this energy range, but the error bars are too large, so that the present result can be considered consistent with the experiment.

Figures 2(a) and 2(b) show a striking difference in the polarization degree P of the $3p\,^2P_{3/2}$–$3s\,^2S_{1/2}$ radiation in N^{5+}-He ($P \sim 0.25$) and N^{5+}-H_2 ($P \sim 0$) collision systems. In order to reveal the origin of this difference, we plot in Fig. 3 the time evolution of weighted m-selective electron-capture probabilities as a function of the impact parameter for N^{5+}-He [panel (a)] and N^{5+}-H_2 [panel (b)] collision systems at the energy of 3.61 keV/u and distances along the direction of the projectile velocity vector $z = vt = -4, 0, 4, 20$, where $z = 0$ is the distance of closest approach. The interpretation of these figures can be most easily done if we invoke the molecular picture of the collision dynamics, in particular the fact that the initial molecular s_σ state in both systems has diabatic potential energy curve crossings with the N4$^{4+}(3l) + He^+, H_2^+$ states at about $R_{3s} = 6.4$, $R_{3p} = 7.7$, and $R_{3d} = 8.2$ for the $N^{5+} + He$ system and at about $R_{3s} = 4.2$, $R_{3p} = 4.7$, and $R_{3d} = 4.5$ for the $N^{5+} + H_2$ system. When passing these energy crossing regions in the incoming stage of the collision, the molecular states correlating to the asymptotic $3s$ and $m = 0$ substates of the $3p$ and $3d$ states of $N^{4+} + He^+, H_2^+$ are populated by radial coupling. At very small internuclear distances (the region of united atom), the rotational coupling populates also the $m = 1$ substates of molecular states that correlate to the $3p_1$ and $3d_1$ asymptotic states. In the outgoing stage of the collision, both the $3p_0$ and $3p_1$ substates enter the radial coupling region around R_{3p} but the interaction with the initial molecular s_σ state affects (reduces) only the $m = 0$ substate population. On the other hand, in the outgoing stage of the collision the returning flux along the s_σ state also enters the R_{3p} strong coupling region and populates the $3p_0$ capture channel by radial coupling. At large internuclear distances (well outside the "molecular region") the $3s$, $3p_0$, and $3d_0$ substates, as well as the $3p_1$ and $3d_1$ substates, are mixed by the electric field of the residual ion (Stark mixing).

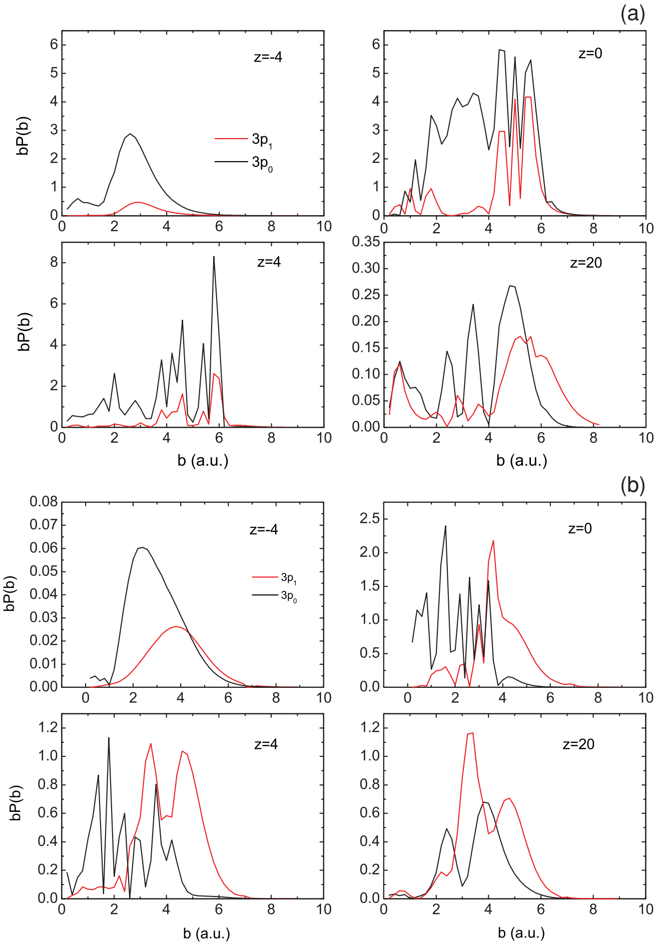

FIG. 3. (Color online) Weighted probabilities $b \times P(b)$ as a function of impact parameter for different collision times for N^{5+}-He (a) and N^{5+}-H_2 (b) collisions at the collision energy of 3.61 keV/u.

It is worth noting in Fig. 3 that the weighted probabilities for $z = -4$ in panel (a) and panel (b) of the figure are drastically (2 orders of magnitude) different. This indicates that in the $N^{5+} + He$ case the R_{3p} radial coupling region has been reached by the system, while in the $N^{5+} + H_2$ case it has not. In the time interval between $z = -4$ and $z = 0$, both collision systems have reached the internuclear distance region where rotational coupling is strong, and in the panels for $z = 0$ we see a sizeable population of the $3p_1$ state. At $z = 4$, both the $3p_0$ and $3p_1$ populations are reduced with respect to the $z = 0$ case due to radial coupling effects in the R_{3p} region. The significant reduction of the $3p_0$ and $3p_1$ populations observed at $z = 20$ in the $N^{5+} + He$ system can be attributed to the large extension of the radial coupling region around $R_{3p}^{He}(=7.7)$, to the strong nonadiabatic coupling, and to their Stark mixing with the $3d_0$ and $3d_1$ states, respectively. In the $N^{5+} + H_2$ case ($R_{3p}^{H_2} = 4.7$), it appears that all these effects are much weaker and the population of $3p_0$ and $3p_1$ states does not change considerably with respect to that at $z = 4$.

The aforementioned molecular dynamics interpretation of the difference between the polarization degrees of $3p\,^2P_{3/2}$–$3s\,^2S_{1/2}$ radiation in N^{5+}-He ($P \sim 0.25$) and N^{5+}-H_2 ($P \sim 0$) collision systems indicates that this difference is caused predominantly by the difference in the radial couplings between the states in the R_{3p} curve-crossing region which in the two systems lies at different internuclear distances. In the AOCC description of the collision dynamics employed in the present work, the molecular-state radial couplings are represented by the electron-exchange (nondiagonal) matrix elements of the Hamiltonian between the atomic states, the maxima of which are also distributed in the internuclear regions around the crossing of diabatic potential energies (the diagonal matrix elements of the Hamiltonian). Therefore, the conclusion regarding the difference in the polarization degrees of $3p\,^2P_{3/2}$–$3s\,^2S_{1/2}$ radiation in the two considered systems, derived on the basis of the molecular picture, remains valid in the AOCC dynamics picture as well.

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