PHYSICAL REVIEW A VOLUME 32, NUMBER 2 AUGUST 1985

Alignment of Ne^{8+} n^1P states produced by collisions of Ne^{9+} with H_2 at 4 keV/amu

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(Received 26 December 1984)

Relative intensities and polarization rates of $n^1P \rightarrow 1^1S$ heliumlike Lyman x rays emitted in collisions of 4-keV/amu Ne^{9+} ions with H_2 have been measured. Our experimental results are discussed in connection with molecular calculations for the near-system $F^{9+}+H$. High polarization rates ($\sim 60\%$) are measured for direct x rays, indicating a strong predominance of $m=0$ substates in final n^1P states, in agreement with the theory. This shows the existence of a strong rotational coupling in the molecular frame. The difference of polarization observed for the $2P \rightarrow 1S$ and $3P \rightarrow 1S$ x rays may originate from a higher anisotropy of the low-angular-momentum states with respect to the high ones, as predicted by the theory.

I. INTRODUCTION

As electron capture by multicharged ions from atoms produces excited states, recent experimental effort has been devoted to the determination of the n and l distributions of these excited states, either by the energy gain technique or by an optical method.[1] In the low velocity range considered here, a molecular description of the collision is adequate and transitions to the most populated states, characterized by a restricted range of final n values, occur in the vicinity of molecular-level pseudocrossings. However, it has been shown[2] that the l distribution is not determined by this primary process of electron capture. Indeed, the Stark effect caused by the residual charge of the target after electron capture is strong enough to mix the various l substates for a given hydrogenic n state. Then only the m distribution is representative of the initial charge-exchange process; in the rotating molecular frame, $m=0$ and $m=1$ values are, respectively, generated by radial and rotational couplings. This shows the interest of determining this m population. Moreover, the m distribution governs the l one, which is clearly illustrated by the fact that this last one can be obtained with a good approximation just by sharing equally the m values among consistent l values (the so-called "complete l mixing" model).

In this paper we present experimental results about n,m populations in 4-keV/amu $Ne^{9+}+H_2$ collision and an argued comparison with theoretical predictions for the one-electron case $F^{9+}+H$. One-electron projectiles and singlet final states were selected to take advantage of the fact that they do not suffer from dealignment by spin-orbit coupling. Lyman x rays $1snp \rightarrow 1s^2$ have been observed and it is found that, except for the case of $n=2$, they sign exclusively the deexcitation of singlet states; indeed, $1snp\,^3P$ states decay to the $1s2s\,^3S$ state; however, for $n=2$ the intercombination line $^3P_1 \rightarrow {}^1S_0$ is observed with the normal $^1P_1 \rightarrow {}^1S_0$ line. Thus the polarization of $nP \rightarrow 1S$ Lyman x rays reflects (except in the case of $n=2$) the alignment of initial $n\,^1P$ states.

II. EXPERIMENTAL METHOD

The experiment was performed with 50-nA Ne^{9+} beam produced with a "Minimafios" type multicharged ion source[3] set up at the Centre d'Etudes Nucléaires de Grenoble (Agrippa GIS CEA/CNRS).

The gaseous target, whose characteristics have been described elsewhere[4] was an effusive jet with a known density profile. A pressure of about 10^{-5} mbar was maintained in these experiments and the single collision condition was checked. X rays emitted at 90° to the ion beam were detected by a high-resolution Bragg crystal polarimeter. A rubidium acid phtalate (RbAP) crystal with high integrated reflectivity was used. The resolution of the system was fixed by a 5-mm-wide slit. Reflected x rays were detected by a thin window (1.5-μm aluminized Mylar) position-sensitive flow proportional counter filled with $ArCH_4$ gas at a pressure of 1 atm regulated to better than 1%. The transmission T of the window was measured for Na, Al, and Cl K_α x rays (1.04, 1.49, and 2.62 keV, respectively) produced by fluorescence of Al and NaCl samples, and for As Lyman x rays (1.28 keV) from a radioactive source. The transmission for Ne^{8+} heliumlike Lyman x rays varies from 0.19–0.41 for $2P \rightarrow 1S$ to $6P \rightarrow 1S$ lines. Correspondingly, the detector efficiency ranges from 0.98–0.92.

The polarimeter was oriented in such a way that the more-reflected polarization component was either parallel to the beam axis (vertical reflection plane) or perpendicular to the beam axis (horizontal reflection plane[5]) (see Fig. 1). Let $I_\parallel$ and $I_\perp$ be the intensities of light emitted at 90° from the beam with electric vectors, respectively, parallel and perpendicular to the beam and let R_π and R_σ be the crystal reflectivities for electric vectors, respectively, perpendicular and parallel to the reflection plane. Then light intensities detected for a vertical or a horizontal reflection plane are, respectively,

$$I_V = R_\pi I_\parallel + R_\sigma I_\perp \ , \quad I_H = R_\sigma I_\parallel + R_\pi I_\perp \ . \tag{1}$$

Lyman–x-ray polarizations and m populations in nP states were expressed as

$$P = \frac{I_\parallel - I_\perp}{I_\parallel + I_\perp} = \frac{I_V - I_H}{I_V + I_H}\frac{(1+R_\sigma/R_\pi)}{(1-R_\sigma/R_\pi)} \ , \tag{2}$$

$$\frac{p(m=+1)}{p(m=0)} = \frac{I_\perp}{I_\parallel} = \frac{I_H - I_V R_\sigma/R_\pi}{I_V - I_H R_\sigma/R_\pi} \ . \tag{3}$$

Let us note that integrated reflectivities calculated for ideal mosaic and ideally perfect crystals with absorption lie very near each other;[6] R_σ/R_π ratios are even closer to one another. For RbAP crystals, measured reflectivities lie between the two extreme values and differ by less than 10% from the mosaic one. We have thus adopted for R_σ/R_π ratios the $\cos^2 2\theta_B$ values predicted by the ideal mosaic case with a maximum error taken at 10%.

Observed Lyman–x-ray intensities were normalized with respect to the total number of ions and gas-jet density by monitoring the total x-ray yields with a Si(Li) detector placed in front of the crystal spectrometer and viewing the same interaction region. A geometrical normalization was also necessary since the two orientations of the polarimeter (horizontal and vertical reflection plane) need, respectively, a vertical and horizontal defining slit and the observed interaction region is not the same in both cases. The geometrical normalization was determined from the more intense ($2P \rightarrow 1S$) line by comparing, for each orientation of the polarimeter, the intensities obtained with the 5 mm vertical or horizontal slit in front of the source with the one obtained from a 5-mm×5-mm overlap. The normalization factor was measured with a 11% uncertainty.

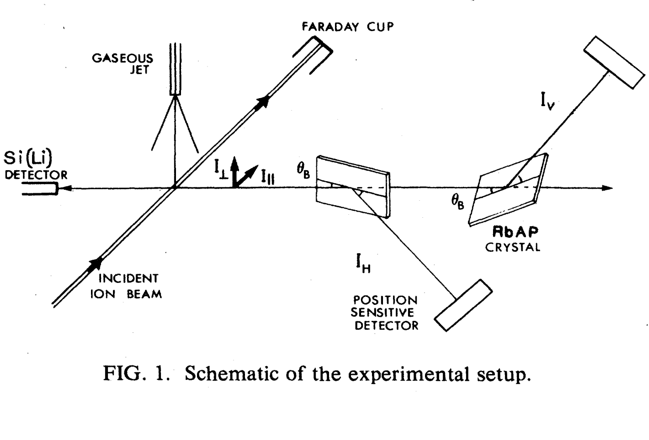

FIG. 1. Schematic of the experimental setup.

III. RESULTS AND DISCUSSION

Figure 2 shows Lyman spectra obtained with the RbAP crystal used in first order in one of the two geometries —the horizontal one. Total Lyman–x-ray intensities, calculated as $I_\parallel + 2I_\perp$, are given in Table I.

The lack of $nP \rightarrow 1S$ Lyman x rays for $n > 6$ indicates that the corresponding n states are not populated by capture. On the other hand, Lyman x rays for $n < 6$ may be partially or totally due to cascades events. This contamination has been estimated by assuming the l distributions in $n = 6$ and 5 levels (the most populated according to theory) to be the ones calculated for the F^{9+} +H case as explained in (2); calculations were based on transition probabilities in Li II (Ref. 7), which show that branching ratios for $1snl\,^1L$ states with $l \neq 1$ are nearly the hydrogenic ones[8] and that $1snp\,^1P$ states decay mainly to the $1s^2\,^1S$ level. In order to test the sensitivity of the result to the chosen initial l distributions, we have also used for comparison the rather different distributions calculated by Shipsey, Green, and Browne[9] and Fritsch and Lin[10] in the O^{8+} +H case (l distributions within the same n state appear to be not much different for neighboring systems). Calculations performed with these three l distributions give the same value for the $5P \rightarrow 1S$ Lyman–x-ray contamination—2%. For the $4P \rightarrow 1S$ and $3P \rightarrow 1S$ Lyman x ray, cascades appear to contribute for 35% and 23% with l distributions (Ref. 2), 43% and 35% with l distributions (Ref. 9), and 43% and 36% with l distributions (Ref. 10).

Together with Lyman–x-ray intensities, we have reported in Table I partial n cross sections for single-charge transfer to $n = 6$ and 5 states. They were deduced from the experimental np cross sections and the calculated percentages of p substates in a given n state. We note that the ratio of these cross sections is compatible with the predictions of the classical decay model (preferential n value = 5 in this case).

As discussed before, $nP \rightarrow 1S$ Lyman x rays for $n \leq 4$ come, for a large part, from the cascades. They are also generated by double events, transfer ionization (TI) or autoionizing double capture (ADC), which can occur with a molecular hydrogen target. Indeed these processes are known to populate levels with n values smaller than the pre-

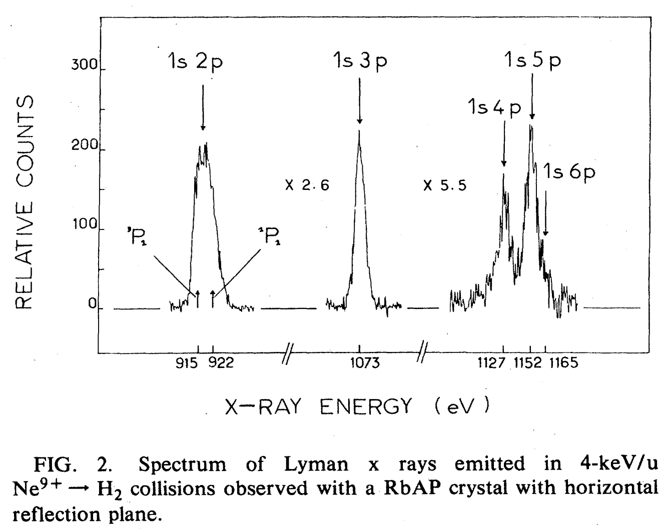

FIG. 2. Spectrum of Lyman x rays emitted in 4-keV/u $Ne^{9+} \rightarrow H_2$ collisions observed with a RbAP crystal with horizontal reflection plane.

ferential n one. Justiniano *et al.*[11] and Groh, Müller, Schlachter, and Salzborn[12] have found for the $Ne^{8+}+He$ and $Ne^{7+}+He$ cases that they amount to 20 or 25% of total capture, nearly independently of projectile energy. Furthermore, the "true" double electron capture is found to be very weak, which ensures a small satellite contamination of our heliumlike Lyman x rays.

Thus $6P \rightarrow 1S$ and $5P \rightarrow 1S$ Lyman x rays reflect the direct single charge transfer whereas $4P, 3P, 2P \rightarrow 1S$ x rays are generated partially ($4P$) or totally ($3P, 2P$) from the cascades and double events (TI and ADC).

Lyman-x-ray polarizations and the m populations in corresponding np states are given in Table I. Reported uncertainties include all previously discussed errors plus statistical errors on crystal and monitoring Si(Li) spectra.

Experimental results for the $Ne^{9+}+H_2$ collision have been compared with theoretical predictions for the $F^{9+}+H$ case. The following arguments show that such a comparison indeed makes sense. For stripped projectiles colliding with atomic hydrogen targets, two effects determine the l and m distributions:[2]

(i) One is the primary process of charge exchange in the vicinity of the pseudocrossings. It populates preferentially σ states with respect to the direction of the projectile velocity.[2]

(ii) Another is the long-range linear Stark effect caused by the residual ion on the final excited states of the projectile as the two ions separate.

When dealing with a H_2 molecule target, the relevant ionization energy is larger than that of H by 0.105 a.u. (since the H_2 molecule hardly rotates and vibrates during the collision, one has to use the "vertical" ionization energy[13]). This increases the position of the crossings by 15% and affects the n distribution, but to a lesser extent the l and m distributions, since the orientation effect (i) is unaffected and the Stark mixing shows the same pattern for various systems,[2] including $F^{9+}+H$, for which the crossing distance is appreciably different.

Furthermore, all these processes take place for Ne-H_2 distances much larger that the H_2 equilibrium distance.

The presence of a core electron in Ne^{9+} is more significant. It damps the linear Stark effect for distances R_C such that the Stark effect is of the same order as the level shift in Ne^{8+} with respect to the hydrogenic value. R_C is of the order of 17 a.u. in the present case. This effect excludes a fully quantitative agreement between Ne^{9+} and F^{9+} cases, but still R_C is large enough to justify a comparison between both collision systems.

The large relative population of $m=0$ observed experimentally for the $6P$ and $5P$ states is clearly at variance with the statistical distribution between substates (all m values equally populated within one l value) predicted by some authors.[14] However it is consistent with the results of our molecular calculation. In this consideration, one has to notice that the predominance of $m=0$ substates observed with respect to the direction of the projectile velocity must be related[2] to the fact that the electron orbital is nearly frozen during the collision (superposition of $m \neq 0$ molecular orbitals in the rotating frame), and thus indicates a strong rotational coupling. Calculations[2] show that this coupling is very important around molecular crossing due to the near degeneracy of parabolic states.

The experiment shows also a high predominancy of $m=0$ substates for the $4P$ and $3P$ states. As seen before, these levels are partially ($4P$) or totally ($3P$) populated by the cascades and by transfer ionization or autoionizing double capture. Calculations for m values reached in these two last processes are not done at the moment. As to the fraction of $4P$ and $3P$ states populated by the cascades, calculations predict indeed that they are highly anisotropic:

(i) nl branching ratios show that these states proceed mainly from low-angular-momentum initial states for which a high predominancy of $m=0$ substates is predicted.

(ii) nlm branching ratios[15] show that the cascading process does not destroy in this case the anisotropy.

Finally, it appears experimentally that the $2P \rightarrow 1S$ transition is weakly polarized. This can be explained in the following way. This transition is the sum of the two components $-2\,^3P_1 \rightarrow 1\,^1S_0$ and $2\,^1P_1 \rightarrow 1\,^1S_0$-, which on statistical grounds are expected to have equal intensities.

(i) The $2\,^3P_1 \rightarrow 1\,^1S_0$ component is nearly totally depolarized by spin-orbit coupling.[16]

(ii) The $2\,^1P_1 \rightarrow 1\,^1S_0$ transition comes for a large part from the cascades from high-angular-momentum initial states for which calculations predict small anisotropy.

TABLE I. np states populated in 4-keV/u charge-exchange collisions of $Ne^{9+} \rightarrow H_2$: Lyman-x-ray intensities, partial transfer cross sections, polarizations, and m populations.

n	Lyman-x-ray intensity[a]	Partial transfer cross section[b]	Polarization Experiment	Polarization Theory[c]	$p(m=1)/p(m=0)$ Experiment	$p(m=1)/p(m=0)$ Theory[c]
2	29.2 ± 5.3		$(7 \pm 7)\%$	$\sim 15\%$[d]	0.88 ± 0.11	
3	3.8 ± 0.6		$(69 \pm 19)\%$	$\sim 45\%$[e]	0.18 ± 0.13	
4	0.78 ± 0.11		$(69 \pm 20)\%$		0.18 ± 0.14	
5	1	1	$(59 \pm 21)\%$	83%	0.26 ± 0.17	0.09
6	0.16 ± 0.05	0.35 ± 0.13	$> 58\%$	80%	< 0.21	0.11

[a]Corrected for the anisotropy and normalized to $n=5$.
[b] Semiempirical value (see text) normalized to $n=5$.
[c]$F^{9+} \rightarrow H$.
[d]Calculated for the sum of a $2\,^1P_1 \rightarrow 1\,^1S_0$ cascade transition from the $1s5g$ level and an unpolarized $2\,^3P_1 \rightarrow {}^1S_0$ transition (see text).
[e]Calculated for a $3\,^1P_1 \rightarrow {}^1S_0$ cascade transition from the $1s5d$ level (see text).

IV. CONCLUSION

In conclusion, this work provides original information about m distributions which give a direct insight into the mechanism of the charge-exchange reaction. It confirms the importance of symmetries for this process and explains the failure of oversimplified classical models. The observed predominance of $m=0$ substates with respect to the direction of the projectile velocity, well reproduced by molecular calculations, indicates a strong rotational coupling in the rotating molecular frame. Experimental results are also consistent with theoretical predictions of a higher anisotropy (strong predominance of $m=0$ substates) for the low-angular-momentum states than for the high ones.

Finally, one may conclude that an experimental and theoretical comparison between the l and m distributions in Ne^{9+} and F^{9+} collisions would give direct information on the long-range Stark mixing and its sensitivity to the presence of an additional electron in the projectile.

ACKNOWLEDGMENTS

We are very grateful to R. Geller and S. Bliman for their constant help and we thank J. J. Bonnet for his technical assistance. This work was partly supported by the Centre National de la Recherche Scientifique ATP No. 920027. This work was performed at AGRIPPA, Centre d'Etudes Nucléaires de Grenoble, Centre National de la Recherche Scientifique, F-38041 Grenoble Cédex, France. Equipe associée au Centre National de la Recherche Scientifique No. 071018.

[1]See, e.g., the contribution in the *Proceedings of the First International Conference on the Physics of Highly Ionized Atoms, Oxford, 1984,* edited by H. Rofendeel (North-Holland, Amsterdam, in press).

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