Spin conservation in electron-capture collisions

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The coupling between electron spin and collisional angular momentum has been studied for He^{++}-He, Ne, H_2, and O_2 collisions (4–600 eV) by observing radiation resulting from double electron capture into excited states of He I and comparing the triplet and singlet emissions. The two-electron targets provide a rigid test of the spin-conservation rule, but of these only H_2 yields data in accord with the rule. The apparently anomalous He^{++}-He results are attributed to the domination of short-range effects. These short-range effects are not restricted to double-electron capture, but can apply in any process which involves spin flip.

Recently there has been considerable interest in atomic and molecular collisions involving multiply charged ions. The study of these processes has been stimulated by their importance in high-temperature plasmas,[1] interstellar space,[2] and the potential of multiply charged ions for soft-x-ray lasers.[3] Experimental studies have provided cross-section measurements for single and double charge transfer[4-8] and some data on the energetics of single charge transfer.[9,10] Very few identifications of specific final states have been made; for double charge transfer no observations are known to us. In this paper we present data on production of excited He atoms formed by two-electron capture by He^{++} in collisions with H_2, He, Ne, and O_2. These double-electron-capture processes provide an ideal opportunity to study the degree of coupling between electron spin and collisional angular momentum in atomic and molecular interactions, and the conditions under which spin nonconserving processes occur. Thus, although the data presented here are for double electron transfer, the conclusions concerning this coupling should be considerably more general and applicable to any inelastic process.

Because the data are in the form of collision-produced emission spectra, specific information on excited-state production is obtained. Thus, the couplings between the various angular momenta (spin, electronic orbital, and collisional) can be investigated. If the spin angular momentum couples only weakly to the collisional angular momentum, the Wigner spin-conservation rule[11] should be obeyed; no change in the total spin should occur in the collision. By comparing emission intensities from singlet and triplet excited states of He I it is possible to determine the extent to which the rule is obeyed. Table I lists the targets used in these studies, the energetics for specific product states, and whether these states are permitted by Wigner's rule. For He and H_2 targets, formation of triplet He I states is strictly forbidden since the He I carries all the final-state spin; the relative triplet: singlet radiation then

TABLE I. Energetics and applicability of the spin-conservation rule for He^{++} collisions with various target species.

Target[a]	Products	Q (eV)[b]	Wigner spin-conservation rule obeyed?
$H_2(X^1\Sigma_g^+)$	$He(3d\,^3D) + 2H^+$	24.2	No
	$He(3d\,^1D) + 2H^+$	24.2	Yes
$He(1s\,^1S)$	$He(3d\,^3D) + He^{++}$	−23.1	No
	$He(3d\,^1D) + He^{++}$	−23.1	Yes
$Ne(2p^6\,^1S)$	$He(3d\,^3D) + Ne^{++}\,(2p^4\,^3P)$	−6.7	Yes
	$He(3d\,^1D) + Ne^{++}\,(2p^4\,^3P)$	−6.7	No
	$He(3d\,^3D) + Ne^{++}\,(2p^4\,^1D)$	−9.9	No
	$He(3d\,^1D) + Ne^{++}\,(2p^4\,^1D)$	−9.9	Yes
$O_2(X^3\Sigma_g^-)$	$He(3d\,^3D) + 2O^+\,(2p^3\,^4S^0)$	23.6	Yes
	$He(3d\,^1D) + 2O^+\,(2p^3\,^4S^0)$	23.6	Yes

[a] The indicated state is the ground state.
[b] $Q > 0$ means exothermic process.

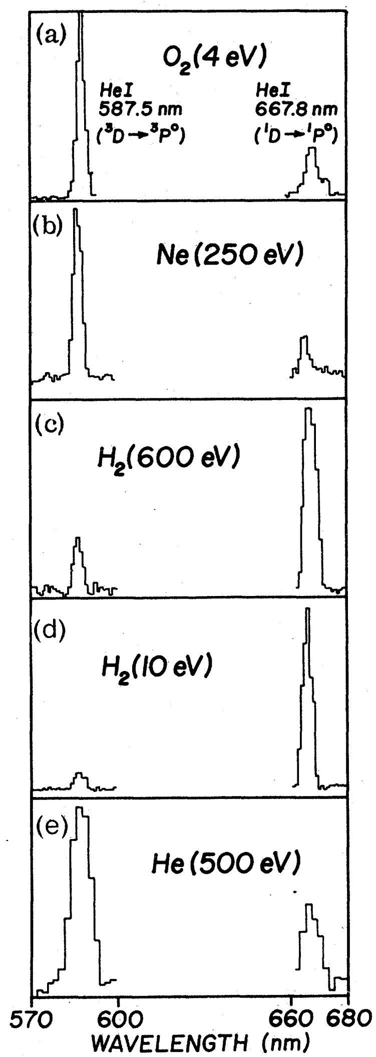

FIG. 1. Representative spectra (count rate vs wavelength) for He^{++} collisions with various neutral species at the indicated laboratory kinetic energies. The resolution for spectra (a)–(d) is 2.5 nm half width at half maximum (HWHM). Because of low photon yields with target He, the resolution in (e) is 5 nm (HWHM) and background counts have been subtracted. All data are corrected for spectral efficiency of the optical system. The vertical scales are adjusted to make the largest peak full scale and do not, therefore, represent absolute count rates. The wavelength region between 600 and 660 nm is omitted here although in some cases there were emissions from species other than HeI in that region; for example, H_α is produced in He^{++}-H_2 collisions.

provides unequivocal evidence of the extent of spin conservation. For the other targets both singlet and triplet helium states are allowed, either because of the initial state of the target (O_2) or because of the variety of possible final states of the twice-ionized product. Therefore, while inferences may be drawn from the Ne and O_2 data, these targets do not provide definitive information on the extent of spin conservation as do He and H_2. In the work reported here the observed relative intensities of the 667.8 nm ($3d\,^1D \to 2p\,^1P^0$) and 587.5 nm ($3d\,^3D \to 2p\,^3P^0$) HeI lines are used to study the applicability of the spin-conservation rule. Radiation from other charge-transfer–excitation processes, both single and double, was also observed but is not the subject of the present report.

The experiments were performed by directing a magnetically selected beam of He^{++} into a collision cell containing neutral target atoms or molecules at 1 mtorr pressure. Photons resulting from decay of excited species produced in the collisions were focused on the entrance slit of a $\frac{1}{4}$-m scannning monochromator with a double lens arrangement and detected by counting output pulses from a cooled photon-multiplier tube; dark current was ~1/sec. A PDP/8E computer controlled operation of the apparatus. Signal averaging to obtain quality spectra was necessary and was achieved by repetitively scanning the wavelength region of interest. For systems having high photon yields, such as He^{++}-O_2, count rates of ~50/sec were observed. Signal-to-noise ratios of ~0.5 were not uncommon for low-yield collisions such as He^{++}-He. Nevertheless, data of sufficient quality could be obtained with multiple scans.

Figure 1 shows representative spectra for each of the targets: differences in the relative triplet-singlet intensities for a given target specie as a function of kinetic energy were much smaller than intersystem differences. The 587.5-nm (triplet) intensity is greater than that of the 667.8 nm (singlet) with Ne and O_2. Both transitions are allowed with these targets according to the Wigner spin rule, but, as discussed above, do not yield unequivocal information about spin conservation. These data are included as illustrative of the results obtained when the *total* final-state spin cannot be determined. On the other hand, He^{++}-H_2 collisions, which provide a rigid test of Wigner's rule, produce only minor yields of the spin-forbidden 587.5-nm radiation. Thus, the data for H_2, Ne, and O_2 targets are consistent with the spin rule; for the H_2 case there is clear evidence that there is relatively little interaction between spin and collisional angular momentum. The results for the He^{++}-He system, which (like He^{++}-H_2) provides a rigid test, show a much greater degree of spin nonconservation; this may be seen by comparing the relative HeI line intensities in spectra (c) and (d) to those in (e).

The apparent discrepancy between the He and H_2

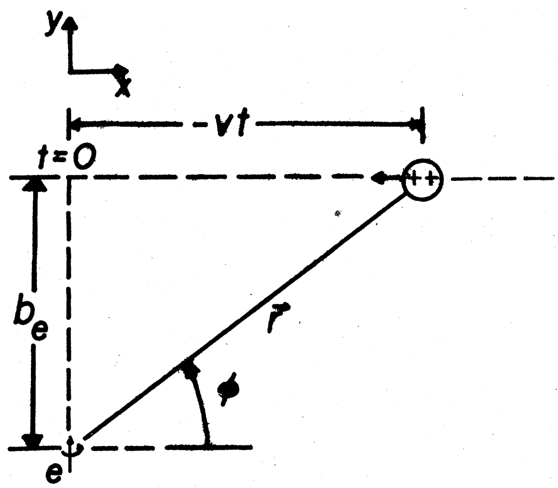

FIG. 2. Coordinates used in the derivation of the expression for the change in angular momentum ΔL resulting from coupling between electron spin and collisional angular momentum [Eq. (5)].

results may be resolved by classically computing the torque impulse on an electron magnetic moment produced by the magnetic field associated with an incident ion moving on a straight-line trajectory. Figure 2 shows the coordinates used in this calculation. The magnetic induction $\vec{B}$ is given (in Gaussian units) by[12]

$$\vec{B} = (\vec{v}/c) \times \vec{E}, \quad (1)$$

where $\vec{E}$ is the electric field of the ion,

$$\vec{E} = -(2e/r^3)\vec{r},$$

and

$$\vec{v} = -v\hat{i}$$

is the ion velocity. In terms of the angle ϕ, $\vec{B}$ may be written

$$B = (2ev/b_e^2 c)\sin^3\phi\hat{k}, \quad (2)$$

where $\hat{k}$ is the unit vector in the z direction, and b_e is the "impact parameter" for the He^{++}-e system. The torque on the electron magnetic moment, $\vec{\mu} = (e\hbar/2mc)\vec{\sigma}$, is given by

$$\vec{\tau} = \vec{\mu} \times \vec{B} \quad (3)$$

which, when integrated from $t = -\infty$ to $t = +\infty$, gives the torque impulse on the electron magnetic moment. The change in angular momemtum $\Delta\vec{L}$ is equal to this torque impulse

$$\Delta\vec{L} = \frac{2e^2}{b_e}\left(\frac{1}{mc^2}\right)\frac{\hbar}{2}(\vec{\sigma} \times \hat{k}) . \quad (4)$$

Since $\vec{\sigma} \times \hat{k}$ is of order unity and $e^2/a_0 = 27.2$ eV, where a_0 is the Bohr radius, Eq. (4) may be written

$$|\Delta L| \approx \frac{54.4 \text{ eV}}{0.5 \text{ MeV}}\frac{a_0}{b_e}\hbar$$

$$\cong 10^{-4}\frac{a_0}{b_e}\hbar . \quad (5)$$

TABLE II. Emission cross sections σ for HeI 587.5 nm ($3d\,^3D \to 2p\,^3P^o$) and 667.8 nm ($3d\,^1D \to 2p\,^1P^o$) radiation from He^{++} collisions with various targets.

Target	Kinetic energy (lab), eV	σ(Å^2) 587.5	σ(Å^2) 667.8
He	200	1×10^{-3}	0.5×10^{-3}
	500	2×10^{-3}	1×10^{-3}
H_2	10	0.2	2
	20	0.1	1
	50	6×10^{-2}	0.7
	100	4×10^{-2}	0.3
	250	2×10^{-2}	0.2
	500	1×10^{-2}	0.05
O_2	10	7	2
	20	5	1
	50	0.6	0.2
	100	0.4	0.1
	250	0.2	7×10^{-2}
	500	0.2	5×10^{-2}
Ne	50	5×10^{-2}	2×10^{-2}
	100	4×10^{-2}	1×10^{-2}
	250	2×10^{-2}	5×10^{-3}
	500	2×10^{-2}	4×10^{-3}
	600	3×10^{-2}	4×10^{-3}

Note that the velocity of the incident ion does not appear in this expression. According to Eq. (5), only very-short-range He^{++}-e collisions can cause a spin flip, that is $b_e \approx 10^{-4}a_0$. Stated another way, for $b_e \lesssim 10^{-4}a_0$ the magnetic interaction term in the Hamiltonian, $\vec{\mu} \cdot \vec{B}_{external}$, dominates the electrostatic terms thus permitting spin nonconservation. In our experiments we have measured absolute emission cross sections[13] for the relevant atomic lines; Table II contains some of these data. The important observation is that the $3d \rightarrow 2p$ cross sections for He targets are considerably smaller than for H_2 targets. This suggests that only short-range encounters contribute to the cross section with He, but longer-range effects dominate in the He^{++}-H_2 system. Therefore, the higher relative yield of 587.5 nm radiation in the He^{++}-He system may be solely the result of very close He^{++}-e encounters for which the spin-orbit coupling is greatest. This then implies that the small amount of spin-orbit interaction observed in He^{++}-H_2 collisions arises from similar short-range effects. In this case, however, double capture to He($1s3d$) is also occurring at large impact parameters where virtually no spin-orbit interaction occurs. These long-range effects dominate so that the short-range spin-nonconserving effects appear relatively smaller. This point of view is supported by our observation of an increase in the triplet-singlet ratio in He^{++}-H_2 collisions at higher collision energy [compare spectra (c) and (d)] accompanied by a decrease in the total $3dD \rightarrow 2pP^0$ emission cross section which suggest increased relative importance of the short-range effects.

On the basis of the data presented here it may be concluded that spin-nonconserving processes can occur only when b_e is much smaller than atomic dimensions ($\sim a_0$). Such small He^{++}-e separations can occur only when the He^{++} actually penetrates the electron cloud of the target atom or molecule, causing, in accordance with Eq. (5), spin flip. As a consequence of the necessity to penetrate the electron cloud, it is expected that cross sections for spin-nonconserving processes would be extremely small as observed in this work. These conclusions are not restricted to the case of double electron transfer. Any process which requires a spin flip is of course subject to the same restriction on the magnitude of b_e. The He^{++} work reported here merely affords the opportunity to study this phenomenon under conditions which permit unambiguous interpretation of the data.

ACKNOWLEDGMENT

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