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Effects of alignment and interference in resonant transfer and excitation for F^{6+} and O^{5+} collisions with H_2 in 0° Auger measurements

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We have renormalized our previously reported 0° cross sections for resonant transfer excitation followed by Auger decay (RTEA), $d\sigma_{\rm RTEA}(0°)/d\Omega$ obtained in 0.25–2 Mev/u collisions of F^{6+} and O^{5+} ions with H_2 by normalizing to calculated binary encounter electron yields rather than the usual Ne K-Auger yields. The renormalized data are found to be in good agreement with recent angular dependent impulse approximation calculations of RTE, showing the importance of alignment and the small influence of interference between RTEA and elastic electron scattering for 0° observation.

Resonant transfer excitation[1,2] (RTE) in energetic ion-atom collisions is a correlated two-electron process, mediated by the electron-electron interaction, involving the transfer of a target electron to the projectile with the simultaneous excitation of a projectile electron, giving rise to doubly excited states. The study of RTE has received considerable attention in the last few years,[3] since it can provide direct information on electron correlation phenomena[4] presently of great interest in atomic physics.

Theoretically, RTE has been described within the impulse approximation[2,5] (IA) and as viewed from the projectile frame, has been considered[2] to be analogous to the time-reversed Auger electron process, in the limit where the loosely bound target electron to be captured can be considered to be free and having the speed of the projectile. In this way, one can relate RTE, an ion-atom collision process, to that of radiationless capture (RC), an ion-electron collision process.[2,3,6–8] Furthermore, the production of the doubly excited intermediate states (d) from the ground state (g) of the ion by an electron can be essentially calculated only from knowledge of the Auger rates (time-reversed $d \rightarrow g$) without any reference to the dynamics of the collision process itself.

The most stringent tests, to date, of any RTE calculation, have been supplied by state-selective studies[9–18] performed using high-resolution Auger electron spectroscopy[19,20] (RTEA). In these measurements, state-selective differential cross sections (SSDCS), $d\sigma(\theta_L)/d\Omega$, are determined by detecting the Auger electrons ejected, at a laboratory angle θ_L with respect to the beam direction, upon the decay of the intermediate projectile states (d) formed by RTE. Good ***absolute*** agreement between theory and experiment is expected since the absolute values of the RTEA SSDCS within the IA, depend[2] only on the various Auger rates. However, since no calculated SSDCS for the expected RTE angular distributions of the ejected electrons have been available, until recently,[21] *total* RTE cross sections were usually obtained from the measured SSDCS by assuming *isotropic* emission.[11,15,17,18] Such a comparison with total RTE cross sections, assuming isotropic emission, was recently reported by the authors.[16,17] It was found that the experimentally determined cross sections were larger than the calculated total RTEA cross sections by factors ranging between 2 and 3.3.[17]

In this communication, we have revised (see below) our previously reported RTEA SSDCS,[16,17] obtained at zero degrees, and compare them to calculated SSDCS using the recent theoretical treatment of Bhalla,[21] where the alignment of the intermediate doubly excited states is included, as well as the effects of interference between the RTEA and elastic scattering channel (binary-encounter electron peak).[22]

In a recent independent study[22] of binary-encounter electron (BEE) production, we reported excellent systematic agreement between the IA theory of BEE production and our data for 1-2-MeV/u bare projectiles ranging from protons to F^{9+}. Here, we use the theoretical BEE production formulas developed in Ref. 22 to obtain a direct and accurate *in situ* absolute efficiency normalization (calibration) of our electron spectrometer in the electron energy range of 1–5 keV. This way we do not have to extrapolate the spectrometer efficiency by normalizing to the usual Ne target K Auger electron cross sections[23] from proton impact on Ne as done in Ref. 17. The Ne K Auger lines have an energy around 800 eV, while most of the RTEA lines are measured at laboratory electron energies between 1 and 3 keV for which the spectrometer efficiency could drop by as much as 50%. The values of the efficiencies following the two different methods are shown in Fig. 1. The discrepancy between the H^{+}+Ne normalization and the BEE normalization methods is not understood. We have used the new absolute spectrometer efficiency to renormalize our recently reported[16,17] RTEA cross sections. The new efficiency, depending on laboratory electron energy, results in an overall 35–55% increase in the values of the renormalized RTEA cross sections.

Our renormalized zero-degree RTEA SSDCS are shown in Fig. 2 for collisions of F^{6+} and O^{5+} with H_2. Details of the measurements can be found in Refs. 16, 17, and 24. Contributions from nonresonance transfer excitation[5,9,25–27] (NTE) were found to be quite small for H_2, allowing for a simple and direct comparison with RTEA calculations. The F^{6+} + H_2 data of Fig. 2 were fitted to the incoherent sum of RTEA and NTEA,

$$\frac{d\sigma}{d\Omega} = a_R \frac{d\sigma_{\rm RTEA}}{d\Omega} + a_N \frac{d\sigma_{\rm NTEA}}{d\Omega} \ .$$

Scaling factors a_R and a_N were obtained by scaling the H_2 RTEA (see below) and NTEA calculations[28] to the H_2 data. We obtained $a_R = 0.70\pm0.05$ and $a_N = 0.20\pm0.05$ for the ^3D and $a_R = 1.00\pm0.08$ and $a_N = 0.20\pm0.05$ for the ^1D states, respectively. This scaling was also found to be consistent with the O^{5+} + H_2 data also shown in Fig. 2. The RC strengths extracted from this comparison are listed in Table I. The uncertainty in the scaling factors is due to the uncertainty in the fit. The absolute un-

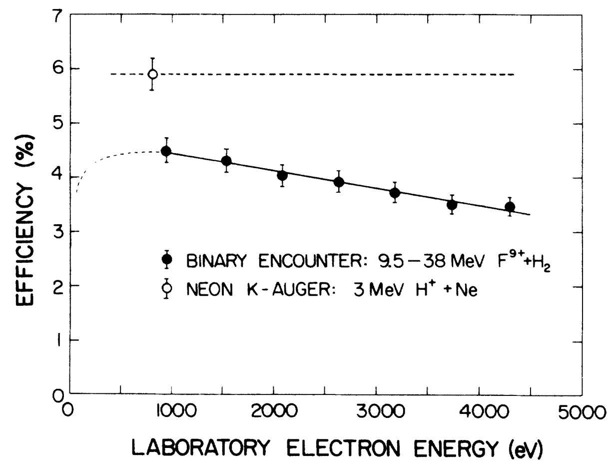

FIG. 1. Overall absolute spectrometer efficiency plotted as a function of the laboratory electron energy. The solid line was interpolated using the data points (solid circles) obtained by normalizing the F^{9+} + H_2 binary-encounter electron yields to the IA calculation as described in detail in Ref. 22. The open circle is the efficiency measured using the known Ne target K Auger cross section (Ref. 23) at 800 eV for 3-MeV H^{+} + Ne collisions. In Ref. 17 it was assumed that this efficiency could be extrapolated to 1–3 keV by assuming it to be a constant. The error bars are calculated from statistics alone. The Ne K Auger datum has an overall absolute uncertainty of 20 % (Ref. 23).

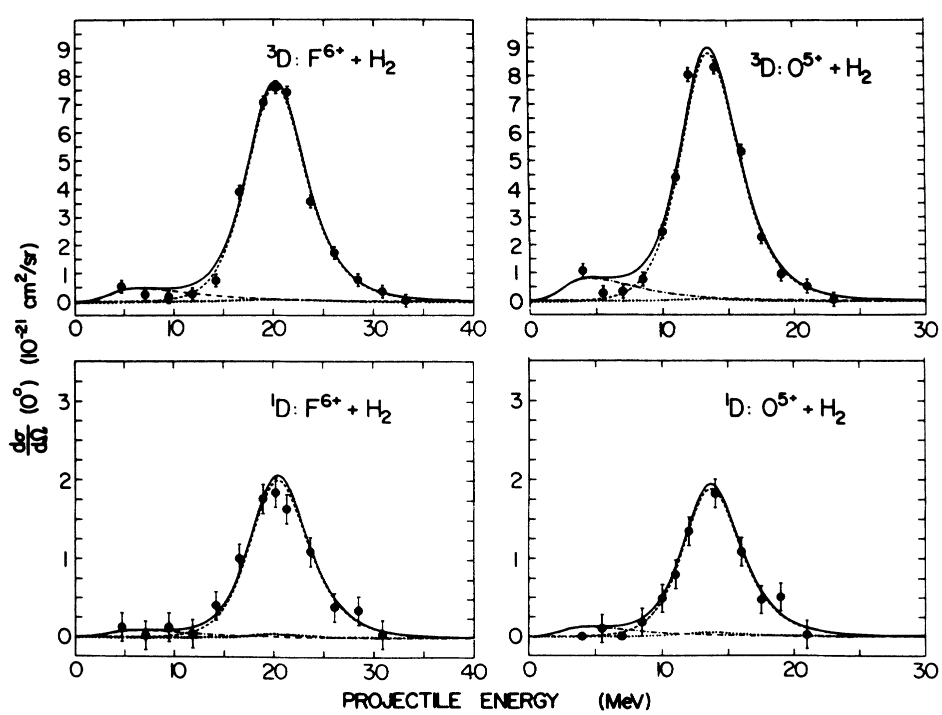

FIG. 2. Data: Absolute Auger electron SSDCS, $d\sigma(0°)/d\Omega$, for $(1s2s2p^2)\,^{3,1}D$ states produced in F^{6+} and O^{5+} $(1s^22s)$ + H_2 collisions vs projectile energy. The Auger electrons result from transitions back to the ground state. Only relative errors are shown. Solid line, sum of $a_R(d\sigma_{\rm RTEA}/d\Omega)$ $+a_N(d\sigma_{\rm NTEA}/d\Omega)$. Dashed line, a_R times RTEA [resonance term only, see Eq. (2)] (Refs. 2 and 21). Dash-dotted line, a_N times NTEA calculation (Ref. 28). Dotted line, a_R times RTEA [interference term only, see Eq. (1) and Ref. 21].

TABLE I. Calculated and experimental radiationless capture (Refs. 2, 17, and 29) strengths (Ω_{RC} in units of 10^{-19} cm^2 eV) for the production of $O^{4+}(1s2s2p^2)$ and $F^{5+}(1s2s2p^2)$ intermediate states. The initial and final states are assumed to be pure $1s^22s$ states. ε_A and ξ are the Auger energies (eV) and yields of the $(1s2s2p^2)\,^{3,1}D \rightarrow (1s^22s)\,^2S$ transition, respectively.

Ion	State	ε_A (eV)	ξ^a	Ω_{RC} ^{a}	$\Omega_{RC}^{expt}(H_2)$ ^{b}
O^{5+}	$1s2s(^3S)2p^2\,^3D$	448.0	0.899	36.3	$25.4 \pm .8$
O^{5+}	$1s2s(^1S)2p^2\,^1D$	453.0	0.409	12.1	$12.0 \pm .7$
F^{6+}	$1s2s(^3S)2p^2\,^3D$	567.84	0.89	35.8	$25.1 \pm .8$
F^{6+}	$1s2s(^1S)2p^2\,^1D$	576.24	0.50	11.7	$11.7 \pm .6$

[a]Calculation uncertainty ~ 15%.
[b]Quoted experimental errors are the relative errors. Total uncertainty (including uncertainty in calculation of ξ) ~ 20%.

certainty in the measured Auger SSDCS, using our new efficiency,[22] is about 10%.

The RTEA calculation (dashed lines in Fig. 2), was computed using the angular dependent IA treatment of RTE by Bhalla.[21] The theoretical SSDCS for the $(1s2s2p^2)\,^3D$ and ^1D states decaying to the $(1s^22s)$ ground state, emitting an Auger electron at an angle θ within the projectile rest frame can be described[21] within the LSM_LM_S-coupling scheme as follows (in units of cm^2/Sr):

$$\frac{d\sigma_{\rm RTEA}}{d\Omega} = \frac{J(Q)}{V_p}\frac{\Omega_{\rm RC}}{\varepsilon_0}[I_{\rm res}(\theta) + I_{\rm int}(\theta)]\ , \quad (1)$$

where $I_{\rm res}(\theta)$, the Auger angular distribution of the decaying RTE resonance, is given by[21]

$$I_{\rm res}(\theta) = \frac{\xi}{4\pi}[1 + \tfrac{10}{7}P_2(\cos\theta) + \tfrac{18}{7}P_4(\cos\theta)]\ . \quad (2)$$

The RC strength $\Omega_{\rm RC}$ is given (in cm^2 eV) by[17,21,30]

$$\Omega_{\rm RC} = 2.475 \times 10^{-30}\frac{(2L_d+1)(2S_d+1)}{(2L_g+1)(2S_g+1)}\frac{A_A(d \rightarrow g)}{\varepsilon_A}\ , \quad (3)$$

with the momentum transfer Q (in a.u.) given by

$$Q = \frac{\varepsilon_A + \varepsilon_I}{V_p\varepsilon_0} - \tfrac{1}{2}V_p\ . \quad (4)$$

V_p is the projectile velocity in a.u., $\varepsilon_0 = 27.212$ eV is the atomic unit of energy, and ε_I is the ionization potential of H_2 equal to 15.5 eV. The experimental Compton profile,[31] J(Q) (in a.u.), was used for H_2. L_d and S_d, as well as L_g and S_g, represent the orbital and spin angular momentum quantum numbers of states $|d\rangle$ and $|g\rangle$, respectively. The Auger rates A_A are in s^{-1}. The RC strength $\Omega_{\rm RC}$, the Auger energy ε_A in eV, and the Auger yields ξ, were calculated[21,29] in the intermediate coupling scheme with the Hartree-Fock atomic model and are given in Table I. We note that for zero-degree laboratory observation ($\theta_L = 0°$) $\theta = 180°$ in the projectile frame.[21] Thus Eq. (1) above becomes, for $\theta_L = 0°$

$$\frac{d\sigma_{\rm RTEA}(\theta_L = 0°)}{d\Omega} = \frac{J(Q)}{V_p}\frac{\Omega_{RC}}{\varepsilon_0}\left(\frac{\xi}{4\pi}5 + I_{\rm int}(\theta = 180°)\right)\ . \quad (5)$$

In previous analyses of RTEA with heavy ions, the Auger emission was assumed to be isotropic and therefore $d\sigma_{\rm RTEA}(\theta_L = 0°)/d\Omega$ was taken to be equal[11,17] to

$$\frac{J(Q)}{V_p}\frac{\Omega_{\rm RC}}{\varepsilon_0}\frac{\xi}{4\pi}.$$

It is clear from Eq. (5) that this assumption underestimates the resonant contribution at 0° by a factor of ≈ 5.

The angular distribution of the Auger electrons, $I_{\rm res}(\theta)$, due purely to the autoionization resonance $|d\rangle$ arises from a nonstatistical population of the magnetic substates. The amplitude for the formation of these states (i.e., ^3D and ^1D) is only nonzero for $M_{L_d} = 0$, when the axis of quantization is defined along the collision direction, and therefore the doubly excited state is collisionally aligned.[32] This seems to be corroborated by recent angular-dependence studies of RTEA in which only the total $M_{L_d} = 0$ substate was found to be populated.[14] We note that the nonstatistical population of magnetic substates (and therefore leading to a nonisotropic distribution of Auger electrons and x rays) has been previously reported for nonresonant processes, such as ionization of atoms by electrons and ions.[33]

The interference term, $I_{\rm int}(\theta)$, arising from interference between the elastic scattering amplitude and the resonance amplitude is given in Ref. 21. It depends sensitively on the relative phases of the two amplitudes and can be either positive or negative depending on the values of the phases and θ.[21] For the collision systems investigated here and at *zero* degrees, the interference (dotted lines in Fig. 2) was found to be smaller than ~ 3%.

In conclusion, we have modified the values of our previously measured[17] zero-degree SSDCS for RTEA, $d\sigma_{\rm RTEA}(0°)/d\Omega$, obtained in F^{6+} and O^{5+} collisions with H_2, in accordance with reevaluated electron detection efficiencies obtained by using the IA treatment of binary-encounter electron production as an absolute normalization standard.[22] This renormalization was found to increase the measured SSDCS by about 35 – 55%, depending on the laboratory electron energy. The modified SSDCS were compared to calculated values of $d\sigma_{\rm RTEA}(0°)/d\Omega$ obtained within the angular-dependent impulse-approximation treatment of RTE by Bhalla,[21] which considers explicitly the alignment of the doubly excited states and interference effects between RTEA and elastic electron scattering. The calculated interference between elastic electron scattering and RTEA for the collision systems investigated was too small to be observable at *zero* degrees. Extracted radiationless capture strengths $\Omega_{\rm RC}$ were found to be in agreement with theory in the case of the ^1D states and about 30% smaller than theory for the ^3D states, but probably within the overall theoretical and experimental uncertainty. The experimental ratio of $\Omega_{\rm RC}(^3D)/\Omega_{\rm RC}(^1D)$ was found to be equal to 2.1 ± 0.14 and 2.2 ± 0.13 for O^{5+} and F^{6+}, respectively, as compared to 3, the expected theoretical ratio.

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