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Impact-parameter dependence of $2p\sigma$ and $1s\sigma$ molecular-orbital x rays produced by Nb-Mo collisions at 143 MeV

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The impact-parameter dependence of molecular-orbital (MO) x rays has been measured as a function of x-ray energy for collisions of 143-MeV Nb ions with Mo. Different shapes for the impact-parameter dependence are found as a function of x-ray energy and are attributed to transitions to vacancies in the $2p\sigma$ and $1s\sigma$ orbitals, $C1$ and $C2$ continuum, respectively. $C1$ MO x-ray emission probabilities were calculated from separately determined $2p\sigma$ vacancy-production probabilities and agree well in magnitude and shape with the measured values.

I. INTRODUCTION

The study of the spectral shape, magnitude, and anisotropy of the quasimolecular (MO) radiation emitted in heavy-ion–atom collisions probes the structure of the transiently formed quasiatomic systems. Thus, even the spectroscopy of transitions in superheavy quasiatoms can be studied. Much interest focuses on the x rays with energies greater than the energy of the characteristic K x rays because this radiation is attributed to transitions to strongly bound transient states. Two types of MO x-ray continua have been observed[1-4]: the $C1$ assigned to radiative transitions into vacancies in the $2p\sigma$ orbital and the $C2$ assigned to transitions into the $1s\sigma$ orbital.

The molecular-orbital model is customarily used as a conceptual framework in which to discuss the ion-atom collision process. Much evidence has accumulated during recent years showing that the molecular-orbital model is not necessarily appropriate for describing systems in which large degrees of ionization are produced in a single collision. It is not surprising that details of the collision process calculated from wave functions of an essentially adiabatic model should fail to be particularly representative of the detailed dynamics of a violent collision. For the minimally perturbed innermost orbitals of concern here, however, the molecular-orbital approach can be useful as a qualitative guide to understanding the collision process.

Several experimental[1-4] and theoretical[5-10] studies have been performed to investigate the features of MO spectra in nearly symmetric heavy-ion–atom collisions, especially in the region of $50 \lesssim Z_1 + Z_2 \lesssim 100$. All the calculations[7-10] for this Z region use the $2p\pi$-$2p\sigma$ rotational coupling model to describe the vacancy-production process, but it has been shown that the theoretical predictions fail to reproduce the experimental K-vacancy-production probabilities $P_K(b)$.[11-13] This problem was not noticed previously because only comparisons between theory and experiment for total cross sections were made instead of the more critical comparison with impact-parameter (b) dependent MO x-ray emission probabilities.

Measurements of the impact-parameter dependence of MO x-ray production probability can be ambiguous.[14] Vacancies in the $1s\sigma$ and $2p\sigma$ orbitals can, in principle, be produced in either one- or two-step collision processes although reliable and convincing determinations of the relative importance of the two have not been made. However, it is plausible to argue that because of the short K-vacancy lifetime and the difference in the $2p\sigma$ and $1s\sigma$ vacancy-production probabilities, the $C1$ radiation is produced primarily in single collisions whereas the $C2$ radiation may be formed in additional two-step processes. For a two-step process, impact-parameter-dependent MO x-ray probabilities are the convolution of the two probabilities [$P_K(b)$ and $C2$], and the experimental results presently cannot be compared directly with theory. On the theoretical side it is necessary[5] to make fully dynamic calculations which take into account the time dependence of the vacancy-production amplitude and of the radiative transition matrix elements, before comparison with experiment is meaningful. To experimentally investigate the time-dependent radiative transition amplitude, a one-collision process should be investigated for which the impact-parameter dependence of the MO x-ray emission probability and the corresponding vacancy-production probability can be separately determined. This should be possible for the $C1$ MO x rays of an intermediate heavy system like Nb + Mo. Any two-collision process for $C1$ should be negligible[3] because of the short lifetime of a Nb K vacancy. The $2p\sigma$ vacancy-production probability, which is responsible for $C1$ radiation, can be determined separately from measurements of $P_K(b)$ for the characteristic $K\alpha$ and $K\beta$ radiation. As will be discussed in more detail in Sec. IV, the $2p\sigma$ occupation amplitude has reached its asymptotic value when the relevant $C1$ transitions occur, and this asymptotic value can be determined from the K-vacancy-production probability $P_K(b)$.[12]

In this paper we report experimental results of the impact-parameter-dependent MO x-ray emission probabilities for $C1$ $[P_{C1}(b, E_x)]$ and C_2 $[P_{C2}(b, E_x)]$ in 143-MeV Nb + Mo collisions, where the $2p\sigma$ vacancy-production probability $P_K(b)$ has been reported previously.[12] In using this $P_K(b)$ and scaled dipole matrix elements and transition energies,[10] absolute emission probabilities for $C1$ radiation $P_{2p\sigma}(b, E_x)$ were calculated, which agree well in shape and in magnitude with measured $P_{C1}(b, E_x)$. The experimentally determined probabilities for $C2$ radiation $P_{C2}(b, E_x)$ show a very different b dependence than $P_{C1}(b, E_x)$.

II. EXPERIMENTAL METHOD

The probability for producing an x ray with energy E_x when an ion was scattered at an angle corresponding to an impact parameter b was determined from measurements of coincidences between the x rays and scattered particles. The probability for production of $C1$ or $C2$ radiation with an energy E_x at an impact parameter b can be extracted from these results and are defined as $P_{C1}(b, E_x)$ and $P_{C2}(b, E_x)$. A 143-MeV Nb beam from the Brookhaven National Laboratory MP Tandem Van de Graaff Facility was collimated to a spot of 1 mm^2 before striking a 100-$\mu g/cm^2$ Mo target which had been evaporated on a 20-$\mu g/cm^2$ carbon backing. A Ge(I) detector for x-ray detection was mounted at 90° with respect to the beam. The efficiency for detection of low-energy characteristic x rays was reduced by use of a 0.175 mm Cu absorber. A 16-ring parallel-plate avalanche detector[15] was used to detect particles scattered at 16 angles θ from 1° to 10°. The angular resolution of the rings was $\Delta\theta/\theta = \pm 5\%$. Rutherford scattering was assumed for the conversion between angle and impact parameter. The counter performed well at rates up to 3×10^6 particles/sec which made possible the measurement of probabilities as small as 10^{-8} photons/particle. As a consequence of this high particle counting rate, the random background in the coincidence spectrum decreased exponentially with time between events (inset in Fig. 3) rather than having the more familiar form of a constant independent of the time separation. The high rate and resulting exponential decay made it necessary to correct for a loss of true coincidences as well as correcting for the effect of random coincidences[16] to get the real number of true coincidences.

III. EXPERIMENTAL RESULTS

A singles spectrum without absorber correction is shown in Fig. 1 by a full line as a function of the x-ray energy E_x. Two different shapes for the continuum above the K lines are clearly visible. Following the notation of Gippner *et al.*[1] they are called $C1$ and $C2$. Possible contribution of radiative electron capture (REC) (Ref. 17) to the $C1$ continuum seems to be negligible. This can be deduced from the existence of the $C1$ continuum with the same spectral shape in the two systems: Kr on Nb (solid target) and Nb on Kr (gas target).[3] Any possible contribution of REC should vanish in the gas target. There is also nearly no dependence of the spectral shape on the shape of $C1$ in Nb on Nb for projectile energies from 67–200 MeV.[3,18] In contrast, the maximum of the REC peak should vary in proportion to the projectile energy.

The coincidence spectra were corrected channel by channel for the effect of random coincidences, as described above, to obtain the true coincidence spectra. Also shown in Fig. 1 (dashed line) is a true coincidence spectrum for an impact-parameter region $b = 180–210$ fm. By normalizing to the characteristic lines, different shapes in the high-energy tail ($C2$) can be observed. The $C2$ continuum is strongly enhanced at this small impact parameter, whereas the $C1$ intensity remains nearly

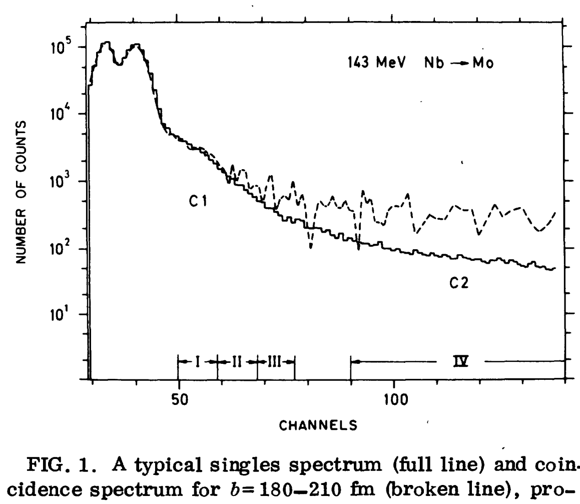

FIG. 1. A typical singles spectrum (full line) and coincidence spectrum for $b = 180–210$ fm (broken line), produced in collisions of 143-MeV Nb with Mo. The coincidence spectrum is normalized to the singles spectrum at the characteristic K x-ray lines.

unchanged. This effect can be seen more clearly in another representation of the data shown in Fig. 2. The absolute probabilities for $C1$ and $C2$ emission are integrated over E_x intervals (as indicated in Fig. 1) and then plotted as functions of the impact parameter. Also shown in Fig. 2 is the K-vacancy-production probability $P_K(b)$.[12] The similarities found in the comparison of the probabilities for $C1$ emission with those for $P_K(b)$ suggest that the two processes may originate from a common mechanism, such as the $2p\sigma$ vacancy-production process. The probability for $C2$ emission shows a completely different b dependence than $P_{C1}(b, E_x)$. The probability $P_{C2}(b, E_x)$ increases very strongly with decreasing impact parameter (nearly two orders of magnitude from $b = 1000$ to $b = 100$ fm). This will be discussed further in Sec. IV B.

IV. DISCUSSION

A. $2p\sigma$ MO radiation

In all previous calculations of $C1$ and $C2$ radiation done in the intermediate Z region[7-10] the vacancy-production process has been treated in the framework of $2p\pi$-$2p\sigma$ rotational coupling theory. However, large discrepancies have been found when theoretical predictions have been compared with experimental values of $P_K(b)$.[11-13] Here, we circumvent this difficulty by using not theoretical, but measured values for $P_K(b)$.

The emission probability for MO radiation to the $2p\sigma$ orbital, $P_{2p\sigma}(b, E_x)$, in units of photons per keV per scattered particle is given by the Fourier integral[5]

$$P_{2p\sigma}(b, E_x) = \frac{4}{3}\frac{e^2 E_x}{\hbar^3 C^3} \sum_K \left| \int_{-\infty}^{+\infty} dt\, a_{2p\sigma}(t) D^K(R(t)) \times \exp\left(\frac{i}{\hbar}\int_0^t (E_x - \Delta E^K(\tau))d\tau\right)\right|^2 . \quad (1)$$

Anholt[10] has calculated the dipole transition matrix element $D^K(R)$ as a function of the internuclear distance R and the transition energy ΔE^K for all possible dipole transitions K for Nb + Nb, which is very close to our system. These results show that dipole transitions to the $2p\sigma$ molecular orbital with energies above the K lines are produced for internuclear separations in the range from about 1500 to 15 000 fm. The maximum value of the transition rate and the x-ray energy (for $2p\sigma \to 3d\pi, 3d\sigma$) both occur at about $R = 5000$ fm. The $2p\sigma$ vacancy production is important only at much smaller R values since $P_K(b)$ is negligible for $b > 800$ fm.[12] Note that the maximum in $P_K(b)$ is found at an impact parameter more than a factor of 2 smaller than the value predicted by the $2p\pi$-$2p\sigma$ rotational coupling model. The $2p\sigma$ occupation amplitude has therefore reached its asymptotic value $a_{2p\sigma}(\infty)$ already in the R region where the relevant $C1$ MO transitions occur. These arguments are confirmed by O'Brien *et al.*[18] who show that $C1$ x rays are not in coincidence with the Nb K x rays and by Anholt and Meyerhof[9] who find the same spectral $C1$ intensity using either $[P_K(b)]^{1/2}$ or $a_{2p\sigma}(t)$ in their calculation. An influence of the time dependence of the vacancy-occupation amplitude on the continuum spectrum was generally found[9,10] to be important only for the very-high-energy part of the continuum. Therefore, to evaluate the Fourier integral, the amplitude $a_{2p\sigma}(R)$ can be taken as a constant $a_{2p\sigma}(\infty)$ for $R > 1000$ fm, and we can use $|a_{2p\sigma}(\infty)|^2 = P_K(b)$.

As the region of $2p\pi$-$2p\sigma$ rotational coupling is well separated from the region of radiative $C1$

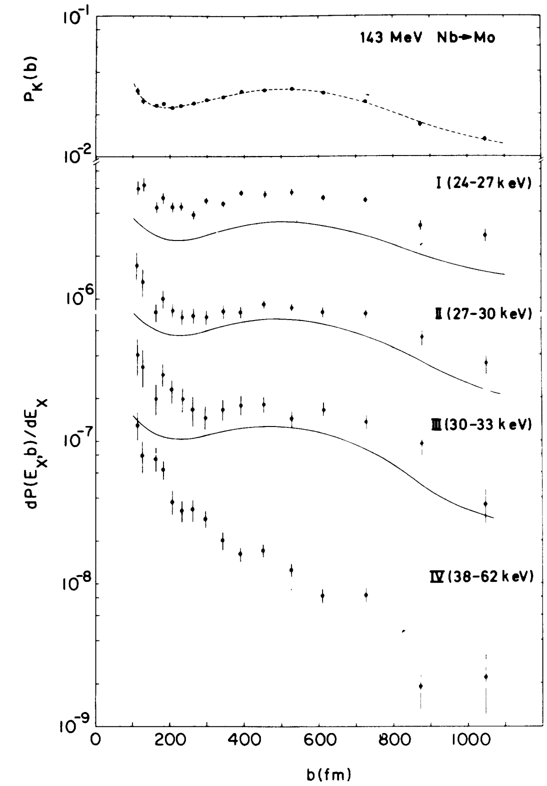

FIG. 2. The impact-parameter dependence of the absolute probabilities for K-vacancy production $P_K(b)$, taken from Ref. 12, is shown in the top curve. The $C1$- and $C2$-emission probabilities (in units of photons per scattered particle and photon energy interval in keV) are shown as functions of the impact parameter b and x-ray energy in curves I–IV. The solid curves are calculated probabilities as described in the text.

transition, the electron slip[10] from $2p\pi$-$2p\sigma$ coupling need not be included. Anholt[11] pointed out that electron slip influences the continuum spectrum only at very high x-ray energies roughly above the united atom K-shell energy E_{ua}.

The R-dependent dipole transition matrix elements $D^K(R)$ normalized to the united atom (u.a.) transition matrix elements $D^K(0)$, are taken from Ref. 10 for Nb+Nb. This normalization allows the scaling of the matrix elements from Nb+Nb to Nb+Mo (one Z difference) by use of united-atom transition rates Γ.[19] Also, the R dependence of the transition energy $\Delta E_K(R)$ can be taken from Ref. 11 for Nb+Nb and scaled to Nb+Mo by adjusting to the united-atom transition energy E_0 and the separated-atom transition energy. The correlation of the many-electron binding energies with the one-electron binding energies used here is discussed in Ref. 10. With these expressions we have

$$P_{2p\sigma}(b,E_x) = \frac{E_x}{E_0} P_K(b) \sum_K \Gamma_K/\hbar \left| \int_{R_1}^{\infty} \frac{1}{v_R} dR \frac{D^K(R)}{D^K(0)} \exp\left(\frac{i}{\hbar}\int_0^R [E_x - \Delta E^K(r)]dr\right)\right|^2 . \qquad (2)$$

We integrated this formula along Rutherford trajectories in steps of 500 fm from $R_1 = 10^3$ fm to 2×10^4 fm. Only the $3d\pi \to 2p\sigma$ and $3d\sigma \to 2p\sigma$ transitions have to be taken into account.

The absolute values of the calculated impact-parameter dependence of $P_{2p\sigma}(b, E_x)$ for fixed E_x intervals are shown in Fig. 2. The values of $P_K(b)$ used in the calculation are shown at the top of Fig. 2 by a dashed line. The shape of the impact-parameter dependence of $P_{C1}(b, E_x)$ is very well reproduced by the calculation, as is the steeper falloff with increasing b at higher values of E_x. The increase of $P_{C1}(b, E_x)$ at smaller b and larger E_x (above 30 keV) compared to the values of $P_{2p\sigma}(b, E_x)$ could possibly arise from a contribution of $C2$ radiation since a very strong increase for $P_{C2}(b, E_x)$ (38–62 keV) is observed for impact parameters less than 300 fm (see Fig. 2). The absolute values for theoretical and experimental probabilities differ by about 40%.

Systematic errors from uncertainties in the x-ray detector solid angle and efficiency are estimated to be about 30% but should be identical for all measurements. Any contribution of the $C2$ radiation at large E_x (above 30 keV) and of the tails from the characteristic K lines at small E_x have not been subtracted. Obviously, it is most likely that uncertainties in the calculation of Γ_K and $D_K(R)$ will largely account for the discrepancy. Anholt found deviations of 50% when calculated intensities were compared with measured total cross sections. However, he used the predictions of the $2p\pi$-$2p\sigma$ rotational coupling model for the $2p\sigma$ vacancy-production amplitude and compared total cross-section values. It should be emphasized that the calculated $P_{2p\sigma}(b, E_x)$ would deviate strongly from the measured values if the $2p\pi$-$2p\sigma$ rotational coupling model predictions, rather than experimental values had been used for $P_K(b)$.

A comparison of the theoretical and experimental spectral shapes is given in Fig. 3. The calculated probabilities $P_{2p\sigma}(b, E_x)$ at $b = 600$ fm were used to calculate counting rates and after correction for the energy-dependent attenuation of the copper absorber were compared to the measured coincidence spectrum for the ring where the impact parameter varied from 528 to 614 fm. The spectral shape and absolute value for theory and experiment agree very well between channel 50 and 78 (24–33 keV). Above and below these limits the $C2$ continuum and the K lines, respectively, dominate. The agreement in shape and absolute value at this impact parameter shows that the collision broadening responsible for the x-ray intensity above the united-atom limit at channel 48 ($E_{ua} = 22.6$ keV) is well treated in the calculation.

The absorber corrected coincidence spectra of $C1$ follows an exponential decay between 25 and 31 keV for all impact parameters measured, so that

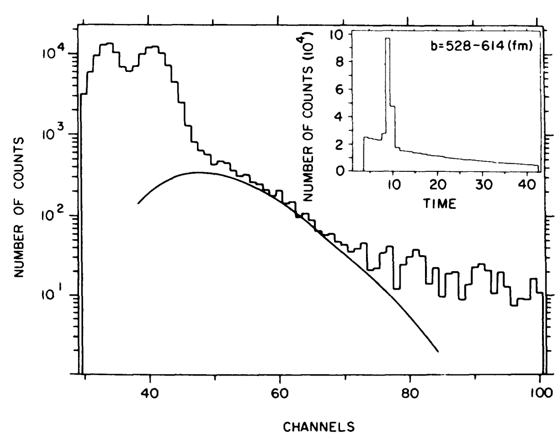

FIG. 3. Coincidence spectrum taken for impact parameters from 550 to 620 fm. The calculated absolute intensity for $2p\sigma$-MO radiation at $b = 600$ fm is indicated by the solid curve. A typical time to amplitude converter spectrum is shown in the inset.

collision broadening can be characterized by the half width H (Ref. 5) of this exponential decay. The value of H was determined for all measured coincidence spectra, and these values are shown in Fig. 4 as a function of impact parameter. The $H(b)$ obtained from the calculated spectral shape are represented by the solid line. The agreement in absolute value is remarkable. The decrease of $H(b)$ at large b seen in the data seems to be well reproduced by the theory. The discrepancy at very small b may again be due to a contribution from the $C2$ continuum.

B. $1s\sigma$ MO radiation ($C2$)

The MO radiation from transitions into the $1s\sigma$ orbital can be produced in a one-collision process or in a two-collision process.[2] In the case of a single collision process, the $1s\sigma$ vacancy is created by direct $1s\sigma$ excitation in the same collision as that in which the MO transition occurs. In a two-collision process, the K vacancy is created in a first collision and brought by the projectile into the second collision. In the case of $C1$ radiation, the two-collision process is negligible compared to the one-collision process, but it may be important for $C2$ radiation because direct $1s\sigma$ excitation occurs with a very low probability.

In the case of a two-collision process, the relation between scattering angle and impact parameter b is ambiguous.[14] The effect of the two-collision process has been assessed in a model calculation that considers all combinations of angles which could lead to a resulting angle θ. Two extreme cases were found to be important.

(1) The first collision with K-vacancy production occurs at very large impact parameters (small angles), and the angle θ is reached in the second collision.

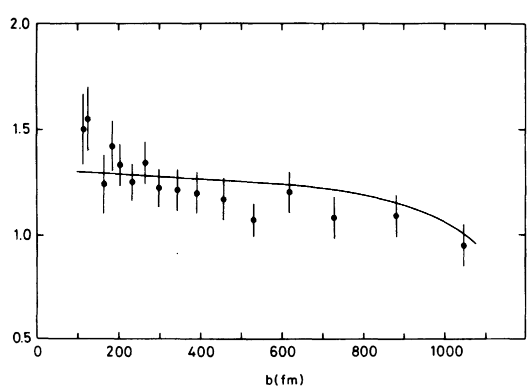

FIG. 4. The half width H of the exponential spectral decay is shown as a function of the impact parameter b. The full line represents H as predicted by the calculation described in the text.

(2) The first collision with K-vacancy production occurs close to the final angle θ, and the second collision with $C2$ emission takes place at a very small angle.

The second case can be excluded because $P_{C2}(b, E_x)$ should then reflect the impact-parameter dependence of $P_K(b)$. The first case of the two-step process might be possible, but we think it is not very reasonable because we find a very strong increase of $P_{C2}(b, E_x)$ with decreasing impact parameter, especially for $b < 300$ fm, well within the united-atom K-shell radius of 640 fm. All calculations of dipole transition elements into the $1s\sigma$ molecular orbital do not predict such a strong impact-parameter dependence of $P_{C2}(b, E_x)$ for $b < R_K$. We conclude from this that for 143-MeV Nb + Mo collisions, $C2$ radiation is produced at small impact parameters ($b \lesssim 500$ fm) by a one-collision process.

The observed strong increase of $P_{C2}(b, E_x)$ with decreasing b could be due to an increase in the probability of $1s\sigma$ excitation $P_{1s\sigma}(b)$. Such a strong increase of $P_{1s\sigma}(b)$ at small b has been found both experimentally and theoretically; see, for example, the discussion by Liesen *et al.*[20] of the heavier asymmetric collision system of Xe + Au. The impact-parameter dependence of the one-collision $C2$ emission probability reflects at small b the features of $P_{1s\sigma}(b)$ and should therefore allow, since the radiative transition matrix elements are known (see Sec. IV A), the measurement of $P_{1s\sigma}(b)$ in symmetric collision systems.

At lower velocities other experiments[1-3] (e.g., 67-MeV Nb + Nb) show evidence from the total cross sections for the two-collision process. Also, good agreement between a scaling law for two-collision $1s\sigma$ Mo x rays and the measured total cross sections has been obtained by Anholt[7] for these types of collision systems. But the total cross sections are only sensitive to the probabilities at large impact parameters, where our results do not exclude a predominance of two-collision $C2$ radiation. Because of the very strong increase of $P_{1s\sigma}(b)$ at small b, it is important to distinguish between different impact-parameter regions when the importance of one- and two-collision processes is assessed.

For the exponentially decaying slope of the $C2$ continuum in the singles spectrum several formulas[6,21,22] for the half width H exist which can be compared with our experimental value of $H = 7 \pm 1$ keV. The value obtained from the formulas of Müller[6] and Betz *et al.*[21] are 4.7 and 4.9 keV, respectively; the semiempirical formula of Vincent *et al.*[22] gives 8.1 keV. The formulas of Müller and Betz *et al.* are only valid for the two-collision process. The discrepancy between their result and our experimental value may be due to the importance of a one-collision process in the $C2$ continuum for our collision system.

V. CONCLUSIONS

Absolute probabilities for $2p\sigma$ and $1s\sigma$ MO x-ray emission, $P_{C1}(b, E_x)$ and $P_{C2}(b, E_x)$, have been measured. Completely different impact-parameter dependences are found. $P_{C1}(b)$ has the same shape as the measured K-vacancy-production probability $P_K(b)$. This observation demonstrates that the $C1$ continuum is produced by transitions into the $2p\sigma$ orbital and that the $2p\sigma$-MO transition probability depends only weakly on impact parameter. $P_{C2}(b)$, however, rises steeply with decreasing b still far inside the united-atom K shell. Since the MO transition probability has a weak dependence on impact parameter, the steep rise of $P_{C2}(b)$ must be due to direct $1s\sigma$ excitation. Therefore, for the range of impact parameters studied here, $1s\sigma$ MO x rays are produced in a one-collision process.

Because $2p\sigma$ vacancy production and decay are well separated, the experimental $P_K(b)$ could be used to calculate absolute $2p\sigma$-MO x-ray emission probabilities. The results of such calculations are found to be in fair agreement with the measured $P_{C1}(b, E_x)$. This approach can be used to test calculated MO transition matrix elements and transition energies. If the predictions of $2p\pi$-$2p\sigma$ rotational coupling were used instead of $P_K(b)$, the calculated $P_{C1}(b, E_x)$ would deviate strongly from the measured values. Therefore, impact-parameter-dependent probabilities rather than total cross sections should be measured and compared with theoretical predictions because integrated probabilities may agree even though the shape of the impact-parameter dependence is completely different.

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