

Excitation of the Sr and Sr⁺ resonance lines by electron impact on Sr atoms*

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The relative optical excitation functions and polarizations of the Sr resonance line (4607 Å) and of the Sr⁺ resonance lines (4078 and 4216 Å) have been measured, using crossed electron and strontium-atom beams, for electron energies from the excitation thresholds to 1497 eV. The electron-beam energy resolution was ~0.22 eV for energies below 13 eV, and the atom beam was optically thin. The excitation functions of the ionic lines were measured relative to that of the atomic resonance line at fixed energies near the maxima of the excitation functions. Using spontaneous-emission branching ratios, this yields the ratios of total excitation cross sections for the atomic and ionic resonance levels (including cascades). The Sr 5¹P excitation function has been normalized to Born theory taking account of cascade at 1497 eV, where the energy dependence of the excitation function has converged to the theoretical behavior. The resulting normalized cross sections have been compared with available theoretical calculations and other measurements. The 4607-Å polarization function is consistent with the theoretical threshold limit within experimental uncertainty.

I. INTRODUCTION

Electron-impact excitation functions and polarizations of the sodium, calcium, lithium, and magnesium resonance lines have been reported in previous papers from this laboratory.¹⁻⁴ It is interesting to look for the systematic behavior of the excitation of alkaline-earth elements by electron impact. In this paper we present our measured optical excitation functions and polarizations for the Sr 5¹P → 5¹S resonance line (4607 Å) and for the Sr⁺ 5²P → 5²S resonance lines (4078 Å and 4216 Å) excited by electron impact on neutral strontium atoms.

Previous measurements of these excitation functions have been reported⁵ with ±30% uncertainty in their absolute cross-section scale. Theoretical calculations of the Sr 5¹P excitation cross section have been performed,⁶⁻⁸ using either the Born approximation⁶ or close coupling.^{7,8} None of these papers report the polarization function.

We have measured the optical excitation functions and polarizations of the Sr resonance line and of the Sr⁺ resonance lines for electron-impact energies from the thresholds to 1497 eV. We used crossed low-density beams of electrons and strontium atoms, thereby minimizing space-charge and optical-depth problems. Furthermore, by going to sufficiently high energy we are able to normalize the 4607-Å excitation function using the Born-approximation cross section for the Sr resonance transition.⁶ We normalized the excitation functions of the ionic resonance lines to that of the atomic line by comparing the radiation intensities at fixed impact energies near the maxima of the excitation functions, using known branching ratios for each level. This yields the excitation cross sections for the Sr 5¹P and Sr⁺ 5²P_{3/2} and 5²P_{1/2} levels.

II. MEASUREMENTS AND CORRECTIONS

The apparatus used in this experiment has been described in detail in Refs. 1 and 2. A beam of atoms from an oven intersects an electron beam at right angles and the resonance radiation is detected in a cone along the third orthogonal axis. The electron gun produces a focused electron beam with a divergence half-angle of ~0.08 rad. The *f*/2.8 detection optics uses a lens to make the rays parallel as the light passes through a linear polarization analyzer and an interference filter (typical half-bandwidth ~30 Å) for the spectral line under study; then a second lens focuses the light onto the end of a light pipe leading to a cooled photomultiplier tube.

For the first time, in this laboratory, a mini-computer has been employed to handle and acquire data. The computer procedure was to set the electron-beam energy, and then switch the polarization analyzer at 5-sec intervals, so that its polarization axis is alternately parallel to and perpendicular to the electron-beam axis. At each polarizer orientation, the photoelectrons are recorded by a gated counter, while the electron-beam current is repeatedly read and averaged over the counting period. The radiation intensities for the two different polarizer orientations are then obtained by dividing the photon counts by averaged current; they are stored separately in the computer memory. The electron-beam energies were scanned up and down several times, so that we were able to check systematic uncertainties and have better statistical uncertainties as well.

The above procedure yields, after minor corrections, the parallel and perpendicular polarized components $I_{||}$ and I_{\perp} of the radiation intensity propagating at right angles to the electron-beam axis. The corrections are discussed below. The polar-

ization P is then $(I_{\parallel} - I_{\perp}) / (I_{\parallel} + I_{\perp})$, and the total cross section Q_T for emission of a given line is proportional to $I_{\parallel} + 2I_{\perp}$.

In our experiment the strontium-atom beam was optically thin (density $\leq 5 \times 10^8$ atoms/cm³), and the electron-beam current was always less than 20% of the space-charge limit for the 0.1 radial cone angle, and also small enough so that space-charge depression of the electron energy was less than 1% of the energy. In spite of the low densities of electron and strontium beams, typical signals from the photomultiplier were 10^3 – 10^4 counts/sec for electron energies above 13 eV, while the background was about 5 counts/sec. Using a retarding-potential analyzer, we measured the electron-beam energy distribution and found that the energy spread of the electron beam used in this experiment was about 0.22 eV full width at half-maximum (FWHM) for energies below 13 eV; it increased slightly at higher energies. We adjusted the energy scales by reference to the excitation thresholds. This adjustment agreed with that given by the retarding-potential analyzer within 0.1 eV.

Our experimental results for the polarizations and excitation functions have been corrected for finite electron-beam and optical solid angles, imperfect polarization analyzer, and minor radiation entrapment (for the atomic resonance line only). All these corrections accounted for less than 2% for the atomic resonance line, less than 6% for the ionic resonance line, and introduced negligible uncertainties to our over-all experimental uncertainties. The polarization of the Sr resonance line was observed to increase by about $(0.2 \pm 0.2)\%$ when the strontium-beam density was reduced to $\frac{1}{3}$ of its typical operating value. We attributed this small density dependence of polarization to entrapment of the radiation, and corrected for it by extrapolating P linearly to zero atom-beam density. No significant variation to our relative excitation function was observed when the atom-beam density was changed as mentioned above. This indicates that optical-depth problems contribute only insignificant effects to our measurements. No correction was made for the instrumental polarization since it was found to be $(0.0 \pm 0.1)\%$ for the same apparatus in previous experiments.⁴

We have estimated the branching ratio for $5^1P - 5^1S$ versus $5^1P - 4^1D$ using the known optical oscillator strength for the former and the Coulomb approximation⁹ for the latter. We concluded that the transition rate from the 5^1P to the 4^1D level is less than 0.01% of that to the 5^1S level. The $(5^1P - 5^1S)$ -line emission cross section is thus effectively identical to the cross section for popu-

lating the 5^1P level, including cascade contribution. The branching ratios (B) for $5^2P_{3/2} - 5^2S$ versus $5^2P_{3/2} - 4^2D$ and for $5^2P_{1/2} - 5^2S$ versus $5^2P_{1/2} - 4^2D$ of Sr⁺ have been measured¹⁰ to be 14.8 ± 2.5 and 13.4 ± 2 , respectively. We have used these to obtain the level excitation cross sections, given by $(B+1)/B$ times the line emission cross section. Finally, we measured the ratios of the ionizing excitation functions at their maxima to the maximum of the atomic-resonance-line excitation function, and filter transmission for each individual line. These measurements, photomultiplier manufacturer's typical quantum efficiency curve (type "S" photocathode), and the known branching ratio for each line, enabled us to normalize the excitation functions of the ionic levels to that of the atomic 5^1P level. The above normalization procedure had about $\pm 8\%$ uncertainty, primarily from uncertainty in the relative filter transmissions and the wavelength dependence of the photomultiplier's quantum efficiency.

III. NORMALIZATION AND CASCADES

Our measured 4607-Å relative excitation function has been normalized to Born theory at high electron impact energy to obtain an absolute excitation cross section. Substituting the experimental oscillator strength¹¹ into the high-energy analytic expression of the Born cross section Q_B for

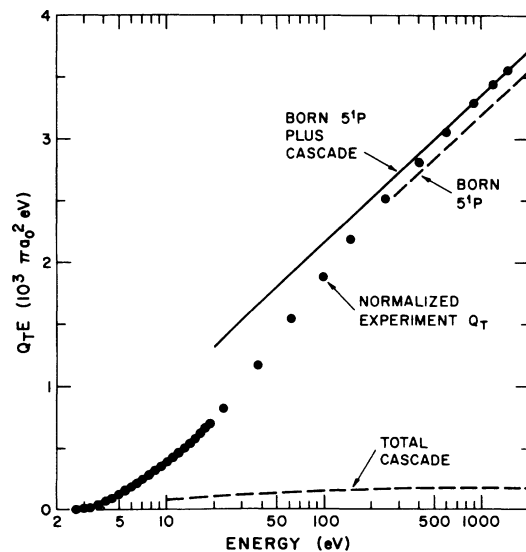


FIG. 1. Method of normalizing the relative cross section for the 5^1P level. The Born-approximation calculation by Kim and Bagus (Ref. 6) has been used for the 5^1P -level direct excitation cross section. Cascade contribution has been estimated based on experimental data of Ref. 5. The present total cross section Q_T (dots) is normalized to the sum of the Born 5^1P plus estimated total cascade cross sections.

TABLE I. Experimental results for electron-impact excitation of the Sr 5¹P resonance level, including cascade contributions.

Energy ^a (eV)	Polarization (%)	Q _f ^b (πa ₀ ²)
2.85(1) ^c	85.2(23) ^c	1.25(4) ^c
2.90(1)	82.6(17)	1.57(4)
3.00(1)	78.3(12)	2.35(4)
3.30(1)	65.0(8)	5.02(8)
3.50(1)	55.9(7)	6.97(10)
3.60(1)	48.7(7)	8.33(11)
3.80(1)	29.6(6)	11.34(14)
3.95(1)	22.8(5)	14.66(18)
4.10(1)	29.6(5)	16.22(19)
4.25(1)	34.4(5)	15.52(18)
4.50(1)	29.4(5)	17.65(21)
4.71(1)	32.6(5)	19.55(23)
5.00(1)	30.7(4)	23.34(26)
6.00(2)	27.5(3)	30.61(34)
7.00(2)	25.0(2)	34.79(37)
10.00(3)	20.4(2)	38.70(42)
13.06(3)	17.2(2)	39.12(43)
15.02(5)	15.4(1)	39.05(25)
22.71(6)	7.6(1)	36.36(22)
37.7(2)	-1.2(1)	31.08(19)
62.5(2)	-9.9(1)	24.84(15)
98.3(2)	-16.9(1)	19.23(12)
149.0(2)	-22.4(1)	14.70(9)
248.0(2)	-28.5(1)	10.16(6)
399.5(2)	-33.5(2)	7.041(44)
600.5(3)	-37.2(3)	5.096(33)
897.5(6)	-41.0(2)	3.663(24)
1198.4(8)	-43.1(2)	2.873(20)
1497.1(10)	-44.7(2)	2.373(15)

^a Mean energy of the incident electrons, corrected by reference to the 5¹P excitation threshold (2.689 eV). The electron energy resolution was ≈0.22 eV FWHM for energies below 13 eV. Besides the uncertainty given for each point, there is an additional uncertainty of ±0.05 eV in the energy scale, discussed in Sec. IV.

^b Q_f is the normalized excitation function including cascade contributions. The uncertainties given in this column do not include the uncertainty of ±4% to our normalized cross-section scale.

^c Number in parentheses gives the uncertainty in the last places of the previous number. In columns 2 and 3, the quoted uncertainties include the observed statistical uncertainties (roughly 2σ) and estimated systematic uncertainties.

a dipole-allowed excitation given by Kim and Bagus,⁶ we obtain a value of 3387πa₀² eV for Q_BE at an electron energy of 1497 eV. The cascade contributions Q' to the 5¹P from higher levels have been estimated by extrapolating the measured emission cross sections in Ref. 5 for spectral lines terminating on the 5¹P level. This extrapolation indicated Q'E ≈ 166±60πa₀² eV for the total cascade contribution to the 5¹P level at ener-

gies above 300 eV, and Q' is ~5% of Q_B(5¹P) at 1497 eV. Our experimental total cross section has been normalized to the sum of the Born 5¹P cross section plus the estimated total cascade contributions, shown in Fig. 1. Note that our measured excitation function appears to converge uniformly to the Born energy dependence at high energies. We estimate that the uncertainty of the normalized cross-section scale for the 5¹P level is about ±4%, primarily owing to the uncertainties in the optical oscillator strength from which the Born 5¹P direct excitation cross section was obtained, and the estimated cascade contributions. Our measurement of the excitation functions of the Sr⁺ resonance lines relative to that of the Sr resonance line was uncertain to ±8% (Sec. II). The uncertainties in branching ratios are negligible, so adding the uncertainties in quadrature yields ±9% as the uncertainty of the cross-section scales for the Sr⁺ 5²P_{3/2} and 5²P_{1/2} levels.

IV. EXPERIMENTAL RESULTS AND DISCUSSION

A. Sr 5¹P → 5¹S line

The present results are given in Table I and in Figs. 1–3. All of the corrections mentioned in

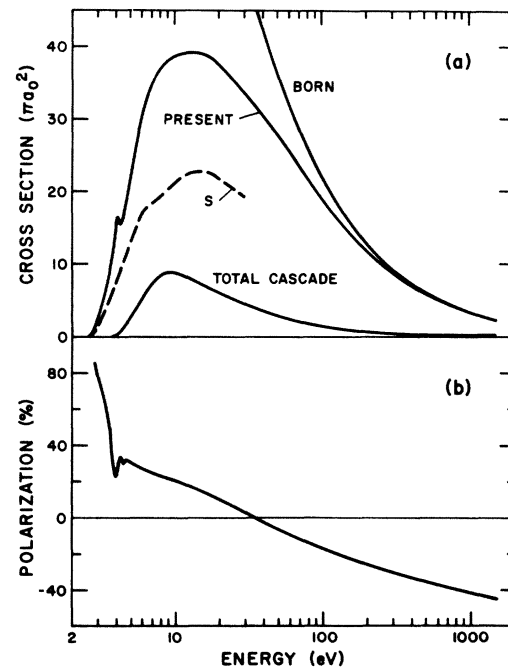


FIG. 2. Normalized total excitation cross section for the Sr 5¹P level and polarization of the 4607-Å resonance line (5¹P → 5¹S). The present cross section is compared with Born theory and previous measurements by Starodub *et al.* (S). All the experimental data include cascades, as does the Born cross section to which the present data are normalized. Estimated total cascade cross section is also given.

Sec. II have been taken into account. The uncertainty in the cross sections quoted in Table I includes uncertainty of the crossed beams's overlap and the observed statistical uncertainty. The uncertainties in the polarization value are mainly from the counting statistics and the uncertainty in the instrumental polarization.

The data were obtained at energy intervals of less than 0.1 eV below 20 eV, and at 11 energy

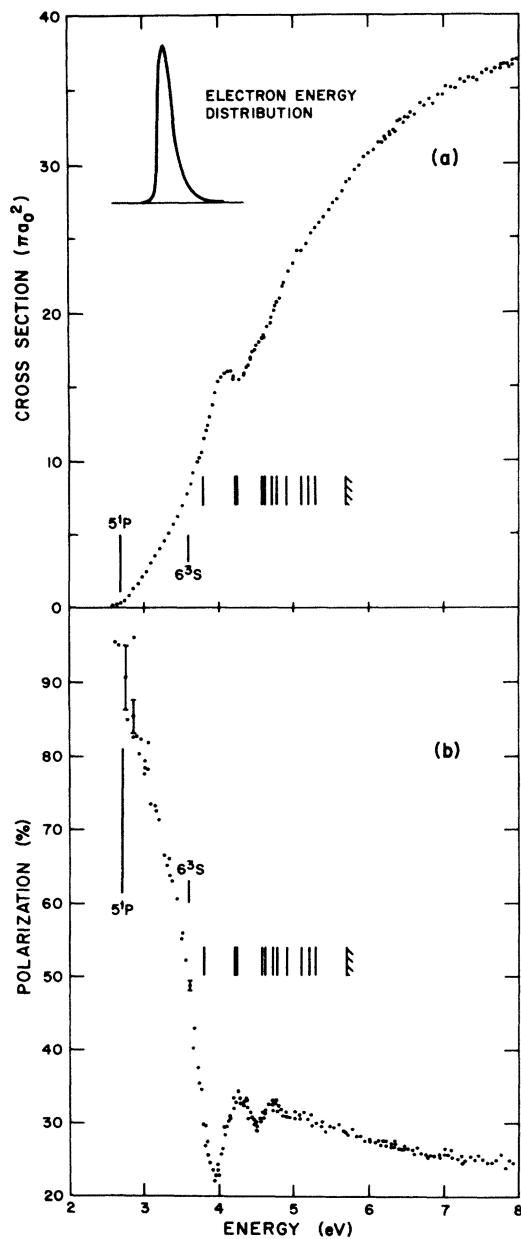


FIG. 3. Detailed low-energy experimental data (dots) for the 5^1P level including cascade contributions. Excitation thresholds for the 6^3S level and higher singlet levels are marked by bars.

values between 23 and 1497 eV. Table I contains all the data above 23 eV, and representative, averaged values below that. The averaged data below 8 eV can be seen in Fig. 3. Because of the smooth behavior of the high-energy Q_T data shown in Fig. 1, we have represented our data as a continuous curve in subsequent plots.

The threshold behavior of our cross-section and polarization data is obscured by the energy spread of the electron beam, as shown in Fig. 3(a). To find the threshold energy we have assumed that the cross section has the form $Q_T \propto (E - E_{th})^{1/2}$ or $Q_T \propto (E - E_{th})$ in the first 0.2 eV above threshold and we have convoluted it with the electron energy distribution. Both forms yield satisfactory fits to our data, while they reach zero cross section at different points about 0.045 eV apart along the energy axis. We have adopted the average of these two zero-cross-section points as the known excitation energy ($E_{th} = 2.689$ eV) of the 5^1P level and used it to correct our energy scale. From this convolution procedure we assume that our energy scale is uncertain to ± 0.05 eV.

The normalized excitation function and polarization are shown in Figs. 2 and 3. In Fig. 2 the present results are compared with earlier measurements by Starodub *et al.*⁵ Both results include cascade contributions. Starodub *et al.* used a crossed-beam apparatus and spectrometer to carry out their absolute measurements for the excitation function and quoted $\pm 30\%$ for their experimental uncertainty. Their excitation function was very different in shape, as well as in magnitude, from ours; we suspect that their measurements might be seriously affected by Sr resonance-radiation entrapment since they used a much larger atom-beam density than that used in the present work (≈ 200 times larger).

The only Sr excitation cross-section calculation other than Born approximation was made by Fabrikant.^{7,8} The calculations in Ref. 7 are 5^1S , 5^1P , 5^3P close coupling and results are much too high in the 3–6-eV region. In Ref. 8 Fabrikant noted that inclusion of 3^1D into an equivalent calculation for 4^1P of Ca yields a much lower 4^1P cross section. He did not give results for Sr, but apparently similar effects are expected. Consequently no comparison is made here with the results in Ref. 7.

Cascade contribution to the 5^1P level begins at 3.79 eV and may cause the structure discernible in the data of Fig. 3. The sharp feature near 4 eV, apparent on the cross-section and polarization curves, is probably caused by cascade contributions from the 6^1S level and the opening of new channels. Dips in the polarization curve occur

just above the thresholds for excitation to 6^1S , 6^1P , and 5^1D terms. Atoms excited to 6^1S and 6^1P (which cascades through 6^1S) will cascade into 5^1P and result in 4607-Å radiation with zero polarization. Thus the cascade contribution from these terms will tend to reduce the net polarization observed; this is precisely the effect we see.

In the simple case of a $^1S_0 \rightarrow ^1P_1$ excitation followed by $^1P \rightarrow ^1S_0$ decay with zero nuclear spin the threshold polarization predicted by Percival and Seaton¹² is +100% and the high-energy limit is -100%. This predicted threshold behavior has often been obscured by resonances in other elements.¹³ In the present experiment, the natural strontium used contains 7% of ^{87}Sr , which has nuclear spin of $\frac{3}{2}$. By interpolating between the hyperfine-structure data for the resonance levels of barium¹⁴ and calcium,¹⁵ we concluded that the hyperfine separation of the ^{87}Sr 5^1P level is much larger than the natural level width, and therefore the 4607-Å-line radiation from ^{87}Sr should be almost completely depolarized. The expected threshold polarization, observed at 90° to the electron beam, from natural Sr is thus about 95% consistent with the present data, as shown in Fig. 3(b), considering the electron energy spread and the large statistical experimental uncertainty for the small signals near threshold. At high energies, our polarization data are consistent with the expected logarithmic convergence.^{2,4} Neither theoretical calculations nor other measurements of polarization are available for comparison.

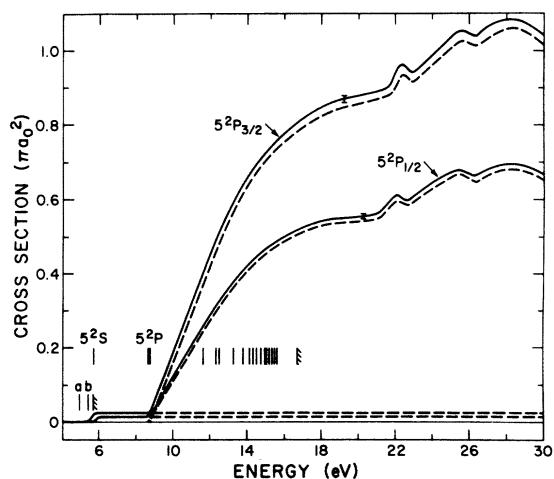


FIG. 4. Normalized total cross section for ionizing excitation of the Sr⁺ $5^2P_{3/2}$ and $5^2P_{1/2}$ levels (including cascades) from the ground state of Sr. Spectral leakages from nearby Sr lines were extrapolated above 9 eV on the basis of the data below the 5^2P thresholds, and subtracted from the raw data (solid curves), yielding the corrected cross sections (dashed curves). Thresholds for some cascade-producing terms are indicated by bars.

B. Sr⁺ $5^2P \rightarrow 5^2S$ lines

Our results for the ionic resonance doublet are shown in Figs. 4 and 5. The data were obtained at energy intervals of about 0.1 eV below 20 eV, at energy intervals of about 0.15 eV between 20 and 32 eV, and at the ten energy values between 38 and 1497 eV. To produce the results shown in Figs. 4 and 5, we plotted smooth curves through the Q_T data. The uncertainty of the relative excitation function is about ± 2 –3% for all energies.

The atomic Sr spectrum contains a number of lines too close to the Sr⁺ resonance lines to be completely rejected by our ionic-line filters. Figure 4 shows how we corrected the ionizing excitation functions for leakages of these unwanted lines. It appears that the only Sr line that can account for the leakage to the 4078-Å ionic resonance line arises from the triplet term ($6^3D \rightarrow 5^3P$) marked *a* in Fig. 4, while the leakage to the 4216-Å line is due to the 4253-Å line ($7^1F \rightarrow 4^1D$, marked as *b*). We removed the effects of these leakages from the observed leakages below Sr⁺

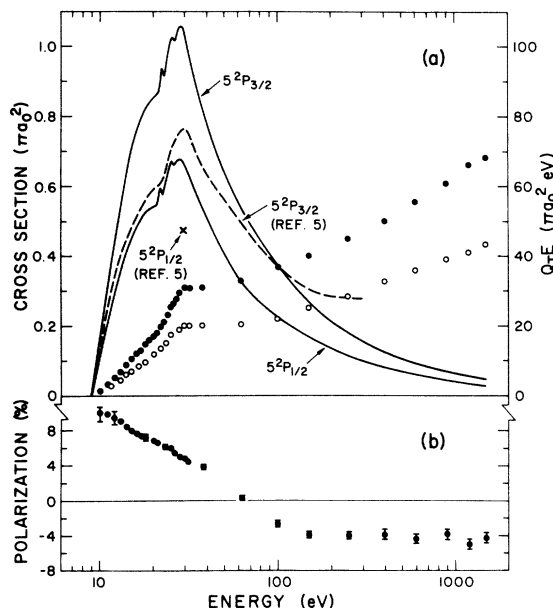


FIG. 5. Normalized total cross section for the Sr⁺ $5^2P_{3/2}$ and $5^2P_{1/2}$ levels and polarization for the 4078-Å line ($5^2P_{3/2} \rightarrow 5^2S_{1/2}$). The present cross section (solid curve) is compared with measurements of Starodub *et al.* (Ref. 5). These level excitation cross sections are obtained by the 4078- and 4216-Å-line excitation cross section times 1.067 and 1.074, respectively, due to branching effect discussed in Sec. II. All the experimental data include cascades. The present results have been corrected for spectral leakages of atomic Sr lines as indicated in Fig. 4. The present cross section times electron energy is also plotted against the $Q_T E$ scale on the right (dots for $5^2P_{3/2}$, circles for $5^2P_{1/2}$).

5^2P threshold energies shown in Fig. 4. For lack of sufficient information about these leakages, their effect on our observed ionic-line polarization has been ignored.

The present excitation functions are compared with previous measurements by Starodub *et al.*⁵ in Fig. 5. For this comparison, we have multiplied their line excitation cross sections by the $(1+B)/B$ factors in Sec. II (~ 1.07) to yield level excitation cross sections. We measured a ratio

$$\frac{\sum Q(\text{Sr } 5^1S_0 \rightarrow \text{Sr}^+ 5^2P_J)}{Q(\text{Sr } 5^1S_0 \rightarrow \text{Sr } 5^1P_1)} = 0.044 (\pm 8\%)$$

compared with a value of 0.055 from Ref. 5 data. The measured ratio of the $5^2P_{3/2}$ to $5^2P_{1/2}$ excitation cross sections was 1.56 in the present experiment and essentially independent of collision

energy, compared with 1.60 reported in Ref. 5 at one energy. LS coupling without variations of radial wave functions predicts¹² a ratio of 2:1.

As shown in Figs. 4 and 5, the shapes of the two ionic-level excitation functions are similar. The Q_{TE} versus $\ln E$ of the ionic levels are also plotted in Fig. 5(a). The present ionizing excitation functions appear to behave as $E^{-1} \ln E$ in the high-energy limit. Similar high-energy behavior has been reported in Ref. 5. One possible explanation for the $E^{-1} \ln E$ high-energy behavior, characteristic of dipole-allowed transitions, is that the Sr $5s^2(1S)$ ground-state wave function contains a substantial admixture of the $5p^2$ configuration,⁸ from which the $\text{Sr}^+ 5p(^2P)$ excited state can be reached by a single-electron transition. Similar ionizing-excitation effects in Ar have been discussed by Tan and McConkey.¹⁶

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