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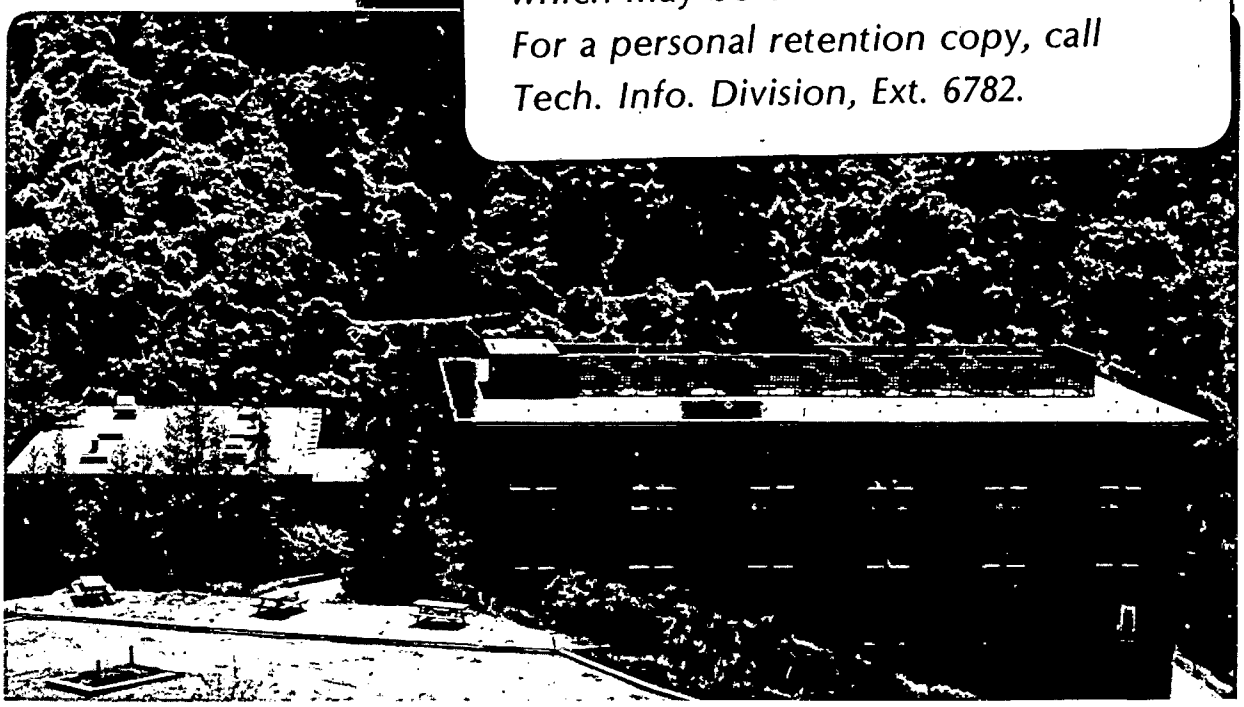
June 1983



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DISCLAIMER

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ABSTRACT

Photoelectron spectra of atomic barium have been recorded at several photon energies in the range 20 eV $\leq h\nu \leq$ 29 eV. The variations of the Ba^+ $5p^6nl$ (nl = 6s, 5d, 6p, 7s, and 6d) photoelectron peak intensities were measured in the 20-21 eV autoionization region. The results indicate that each autoionizing state decays to the various states of the ion in a characteristic way. Asymmetry parameters measured for the 6s, 5d, and 6p channels show strikingly different values that follow the general trend $\beta(6s) > \beta(6p) > \beta(5d)$. Above 21 eV, 5p ionization begins to dominate, and the Auger spectra were used to monitor the production of the various 5p-hole states. As the photon energy is scanned across the autoionizing resonances, these Auger spectra indicate that excitation of 5p-hole states decaying by two-step autoionization is the dominant mode in the production of very low energy photoelectrons ($\leq$2 eV) and their corresponding high-energy Auger electrons. Calculations suggest that a heretofore unobserved $5p^5 6p^2\ ^2P_{3/2}$ 5d autoionizing level is responsible for the Auger distribution measured at $h\nu$ = 28.9 eV.

I. INTRODUCTION

The creation and decay of 5p-hole states in atomic barium has received considerable attention during the past decade. Complex interactions among the various Ba I, Ba II, and Ba III states and their associated continua are involved (see Fig. 1). The complexity is increased by strong mixing of the 6s and 5d levels in this high-Z atom. Thus, the barium 5p shell provides a valuable system for studying electron-correlation phenomena. To place the present work in perspective, related work is briefly reviewed in this section.

The predicted $5p \rightarrow 5d$ giant resonance (Fano 1973; Wendin 1974) was found to be spread over many lines because of Ba I final-state mixing (Ederer 1975; Connerade 1979a). The creation of a 5p-hole state in Ba I lowers the energy of the 5d subshell relative to the 6s subshell. The $(5p)^{-1}$ part of the Ba II spectrum has been examined by Ba II absorption (Roig 1976), Auger spectroscopy (Mehlhorn 1977; Rassi 1980; Rosenberg 1980), He II photoelectron spectroscopy (Potts 1979), Penning ionization electron spectroscopy (Gerard 1977), and He^+ and Ne^+ collision electron spectroscopy (Hultzsch 1979). Multiconfigurational Dirac-Fock calculations have been used to assign most of the observed Ba II states (Rose 1980).

The distribution of ionic states produced by direct pnoto-ionization (process B in Fig. 1) is governed by dipole matrix elements that vary slowly with photon energy. The matrix elements for producing the $5p^6nl$ ($nl \neq 6s$) satellite lines are relatively small, and these lines are weak in the Ne I photoelectron spectrum (Lee

1977). Direct photoionization to the 5p-hole states, as in the He II photoelectron spectrum (Potts 1979), also involves slowly varying matrix elements, but the intensity is more evenly distributed among several lines because of strong ionic-state mixing.

Using resonance lamps, it was found that He I radiation at 21.22 eV excited an autoionizing transition which produced both excited Ba II states and an unexpectedly large Ba^{++}/Ba^{+} ratio (Brehm 1974; Brehm 1975; Hotop 1975; Lee 1977). Explanations of these observations and identification of the Ba I and Ba II states involved have been given (Hansen 1975; Wendin 1978; Connerade 1979b).

Photoexcitation at 21.22 eV coincides with a highly excited 5p-hole state of neutral Ba (process A in Fig. 1). This state subsequently autoionizes, producing $5p^6nl$ and $5p^5nln'l'$ states of Ba^{+} (e_1 and e_2 in Fig. 1). The distribution among the various $5p^6nl$ states in the Ba^{+} ions is very different from that following direct ionization from the barium ground state. Most of the $5p^5nln'l'$ ions ionize further, producing Auger electrons (e_3 in Fig. 1) and the ground-state Ba^{++} ion. This process has been called "two-step autoionization." The branching ratio of the e_2 to e_1 autoionization channels thus controls the Ba^{++}/Ba^{+} ratio at energies for which direct photoionization is negligible.

Examination of the outgoing channels from photoexcitation with tunable radiation was begun by Rosenberg et al. (1979b) who measured photoelectron spectra at two autoionizing energies and by Holland and Codling (1980) who measured the Ba^{++}/Ba^{+} ratio in the 5p-excitation

region. Higher resolution Ba^{++}/Ba^{+} measurements have been made more recently (Holland 1981; Lewandowski 1981).

Photoelectron spectroscopy with tunable radiation is capable of providing uniquely definitive information about the photoexcitation of barium 5p-hole states and their subsequent decay. To extend our knowledge of these processes, we have made quantitative measurements of the variation of $5p^6nl$ partial cross sections and photoelectron angular distributions over part of the autoionizing region. We have also measured Auger spectra at several photon energies between 21 and 29 eV, and found that two-step autoionization is frequently responsible for the distribution of observed peaks. In the following section we briefly describe the experimental arrangement, and in Sec. III we present and discuss the results.

II. EXPERIMENTAL

Radiation from SPEAR at the Stanford Synchrotron Radiation Laboratory (SSRL) was monochromatized (2.5Å FWHM) by a Seya-Namioka normal-incidence monochromator. An effusive beam of atomic Ba was produced by a non-inductively wound, resistively heated oven (Rosenberg 1979a; Kobrin 1982; Kobrin 1983a). The photoelectrons were analyzed by the double-angle time-of-flight (DATOF) method (White 1979; Southworth 1982).

The spectra reported here were recorded during three widely separated experimental runs. The SSRL monochromator energy calibration may have changed between runs, but this ±1Å uncertainty does not affect any of the conclusions presented below. The time-to-energy converted electron energy scales also differ between runs, but the energies of most of the strong Auger lines are well known, and were used to calibrate the electron energy scales internally.

III. RESULTS AND DISCUSSION

The Ba^{++} threshold energy is at 15.21 eV, but most of the Ba^{++} ions detected below 21 eV photon energy are produced by a direct double-ionization process. Above 21 eV, double ionization through Ba^{+} intermediate states becomes dominant. Figure 2 shows a scan of the total electron yield between 596Å and 630Å (19.7 - 20.8 eV). Only electrons with kinetic energies greater than ~4 eV were collected. The photon intensity was not monitored, but changes during this short scan were probably small. In Fig. 2 we also reproduce the relevant portion of the Ba^{+} and Ba^{++} total ion yield spectrum of Lewandowski et al. (1982) taken with a 0.9Å bandpass. The agreement of the two spectra is very good for some lines, with other lines being unresolved in the electron yield. The ionization between 596Å and 630Å is almost entirely due to autoionization (process A then channel e_1 in Fig. 1) with a very small contribution from direct ionization, because of the high density of absorption lines in this region.

Several photoelectron spectra were taken in the energy range of Fig. 2. Almost all of the electron intensity fell in the $5p^6nl$ peaks with nl=6s, 5d, 6p, 7s, and 6d. The partial cross sections for these channels are shown in Fig. 3. The total electron yield from Fig. 2 is reproduced in Fig. 3 for ease of comparison. The sum of the five partial cross sections is equal to the total electron yield at each energy.

Inspection of Fig. 3 shows that the autoionizing resonances

enhance the five nl states by different amounts. For instance, the largest resonance at 617.8Å, labeled as $(5p^5 6s^2)^2P_{3/2}$ 5d by Connerade et al. (1979a), is found predominantly to enhance the $5p^6 6s$ state of the ion. The resonance near 622.9Å, which receives most of its intensity from the $[(5p^5 5d)^3D\ 6s\ ^2D_{3/2}]5d$ state, shows more enhancement of the $5p^6 5d$ channel than of the $5p^6 6s$ channel. At most photon energies the enhancements of the smaller satellite lines (nl = 6p, 7s, and 6d) follow those of the two largest lines (5d and 6s). An exception occurs near 608Å, where the $5p^6 7s$ line alone shows a substantial resonance. This hints at the underlying complexity of the autoionization process in barium and documents the need for photoelectron experiments with higher photon resolution.

In Fig. 4 we show values of the photoelectron angular distribution asymmetry parameter, β, for the 6s, 5d, and 6p states, which were measured simultaneously with the partial cross-section data using the DATOF method (Southworth 1982). These measurements of β over such an unresolved autoionizing region are not amenable to quantitative interpretation. In addition, because they are the first photoelectron angular-distribution measurements from barium, we cannot make any comparisons to off-resonance values. The off-resonance value of β for an s electron from a closed-shell atom would ordinarily be 2.0 (Walker 1973). At autoionizing resonances, β will still be approximately 2.0 if the remaining s electron and the outgoing p-wave are coupled to a singlet. This has recently been observed at the

$3p \rightarrow 3d$ giant resonance in Mn (Kobrin 1983b). That we observe a β value appreciably less than 2.0 for the 6s state in barium may be due to the triplet nature of most of the autoionizing levels.

We note that a definitive study of β in this region would require a photon bandwidth much narrower than the resonance lines themselves, as was the case for cadmium (Kobrin 1982). We shall therefore make only one general observation about the β values displayed in Fig. 4. They tend to fall in the order $\beta(6s) > \beta(6p) > \beta(5d)$. In fact, the β values tend to lie in the ranges $1 < \beta(6s) < 2$, $0 < \beta(6p) < 1$, and $-1 < \beta(5d) < 0$. This is a remarkable result for which an explanation should be sought if it holds up in higher-resolution experiments.

Above 21 eV, Auger electrons may be observed following decay of the Ba^+ 5p-hole states, which can be created both by direct ionization (process B in Fig. 1) and by autoionization (process A then e_2). Potts et al. (1979) showed that the peak intensity distribution pattern in the He II photoelectron spectrum could be approximately accounted for by final-state configuration-interaction calculations. This shows that the intensity produced by direct photoionization is a result of mixing with the primary Ba^+ $5p^5 6s^2$ configuration.

Figures 5 and 6 show Auger spectra taken during two separate experiments, with spectra at $h\nu = 23.7$ eV included in both. The spectra in Fig. 6 were taken with the detector at 54.7° relative to the photon polarization direction, and are therefore independent of angular distribution effects. Those in Fig. 5 were taken with the analyzer at 0° to the polarization direction. We include the latter

spectra because the 0° analyzer provided better resolution than the 54.7° analyzer, and the branching ratios measured by each analyzer were not noticeably different. This implies that the angular distributions of the various Auger peaks are approximately the same. The differences between the two $h\nu$ = 23.7 eV spectra are caused by differences in resolution, transmission, and probably photon-energy calibration, as discussed in Sec. II.

Most of the Auger peaks in Figs. 5 and 6 have been observed in electron impact ionization, and their assignments are given in Table I. In the $h\nu$ = 28.9 eV spectrum, we apparently also observe the primary photoelectrons at 4.2, 5.4, and 6.2 eV from the three states of Ba^+ that produce the 9.5, 8.3, and 7.5 eV Auger electrons, respectively.

As the photon energy is increased in Figs. 5 and 6, new 5p-hole states become energetically accessible and new Auger peaks appear. Many of the Auger peaks become negligibly small at higher energies and are very small in the 500 eV electron-impact Auger spectrum (Rassi 1980). This is the case for most of the peaks observed at photon energies 21.4 and 22.8 eV. The two highest kinetic energy Auger peaks in the 28.9 eV spectrum also appear as small peaks in the 500 eV electron-impact spectrum (Rassi 1980) and in the low-energy electron-impact spectrum (Rosenberg 1980).

The unusually large intensity of the two high kinetic energy Auger peaks in the 28.9 eV spectrum must come from two-step autoionization, but no absorption spectra have been reported in this

energy range. The absorption spectrum of Connerade (1979a) extends in energy only up to 27.5 eV. Therefore, to determine whether any absorption features are likely to be found near 28.9 eV, we have performed multiconfigurational Hartree Fock (MCHF) calculations (with a relativistic correction) using the code of Cowan (1974) on the Ba^+ $5p^5(6s^2 \times 5d6s \times 5d^2 \times 6p^2)$ manifold. In the J=3/2 sub-manifold, two states of primarily $5p^56p^2$ character were calculated to have binding energies of 31.4 and 31.6 eV and to have 3% admixtures (squared CI coefficient) of the $5p^56s^2$ configuration. In the J=1/2 sub-manifold one state of primarily $5p^56p^2$ character was calculated to have a binding energy of 33.5 eV and to have a 7% admixture of the $5p^56s^2$ configuration. In addition to this final-state mixing with the Ba^+ $5p^56s^2$ configuration, the ground state of barium has been calculated to have 6% $5p^66p^2$ character (Rose 1978). This initial-state mixing should add further intensity to a $5p^56p^2nl$ absorption series. The absolute energies of the ionic states were obtained by adjusting the MCHF energies to those from multiconfigurational Dirac-Fock (MCDF) calculations of the $5p^5(6s^2 \times 5d6s \times 5d^2)$ manifold (Rose 1980). The relative spacings of the levels in our J=3/2 MCHF calculation agreed with those from the MCDF calculation to within ~0.3 eV (standard deviation). The first members of the three strong J=3/2 nd series observed by Connerade lie 2.6 ±0.3 eV from their respective limits. Based on these numbers, it is likely that the first members of nd series leading to the 31.4 and 31.6 eV $5p^56p^2$ J=3/2 thresholds would lie

at 28.8 ±0.4 and 29.0 ±0.4 eV. Thus the spectrum at 28.9 eV with a 2.5Å (=0.17 eV) monochromator bandwidth may well overlap an autoionizing resonance, producing Auger electrons by two-step autoionization. This would explain why the two high-energy Auger peaks are observed with such high intensities in the $h\nu$ = 28.9 eV spectrum.

Photoionization above 21 eV does not always lead to double ionization. Mass spectrometry studies (Holland 1981) show a Ba^{++}/Ba^{+} ratio that increases at each major threshold. In addition to these increases there is considerable structure. This structure appears because the discrete autoionizing resonances each have different decay characteristics. Some decay predominantly to $5p^6nl$ states of Ba^{+} (e_1 in Fig. 1) while others decay by two-step autoionization to Ba^{++} (e_2 and e_3 in Fig. 1).

Using the 54.7° spectra obtained at the photon energies given in Fig. 5, we have compared the total intensity of the $5p^6nl$ peaks from Ba^{+} to the total intensity of the Auger peaks from Ba^{++}. Our Ba^{++}/Ba^{+} ratios are listed in Table II along with the ratio at the He I energy and the mass spectrometry values. Our ratios are accurate only to within a factor of two because of uncertainties in the analyzer transmission, but we note that better measurements of this type could provide information about the Ba^{++}/Ba^{+} ratio that is not obtainable by other means. If the Ba^{++}/Ba^{+} ratio from the photoelectron spectrum could be determined accurately enough, it would be possible to obtain, by difference, the amount of Ba^{++} that is

formed directly through the emission of two electrons. These electrons should have a continuum of energies and thus would not appear as peaks in the photoelectron spectrum.

In assuming that each 5p-hole state undergoes Auger decay to Ba^{++}, we have assumed that deexcitation by fluorescence in Ba^{+} is negligible by comparison. This assumption is supported by the Auger and x-ray emission calculations of Chen and Crasemann (1981), which were done with non-relativistic Hartree-Fock wavefunctions and included the effects of initial-state configuration interaction.

Regarding the Ba^{++}/Ba^{+} ratio, Connerade et al. (1980) have proposed that the two channels leading to production of Ba^{+} states, direct photoionization and autoionization, may show an interference effect. The possibility of interference exists for both the $5p^6nl$ states and the $5p^5nln'l'$ states. This interference could produce asymmetric autoionizing lineshapes in the absorption and partial cross-section spectra as well as in the Ba^{+} and Ba^{++} yield spectra. We note, however, that no asymmetric peaks are observed in the high-resolution absorption spectrum (Connerade 1979a) or in the high-resolution Ba^{++} yield spectrum (Lewandowski 1981). Therefore any apparently asymmetric features in the low-resolution Ba^{++}/Ba^{+} spectrum must result from the superposition of unresolved features.

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Table I. Kinetic Energies in eV for Observed Auger Lines

Line No.[a]	Kinetic Energy[a]
1	5.80
2	5.85
3	5.93
4, 5	6.09, 6.10
7	6.43
8	6.57
9 or 10	6.76 or 6.81
12	7.06
13	7.29
14, 15	7.48, 7.58
16	8.31
22	9.55
29	12.00[b]
34	13.54[b]

[a]From Rosenberg et al. (1980).

[b]In the MCDF Mn II calculations of Rose et al. (1980), the 10th and 12th levels in the J=1/2 sub-manifold include 13% and 3% admixtures of the $5p^5 6s^2$ configuration and have calculated Auger energies of 12.0 and 13.5 eV, respectively.

Table II. Ba^{++}/Ba^{+} Ratios

hν	PES	Mass Spectroscopy	
		Holland[c]	Lewandowski[d]
21.22	0.65[a]	1.05	0.7
21.4	0.3 - 0.65[b]	1.0	0.7
22.8	3.4 - 5.6[b]	3.5	1.9
23.7	7.8 - 11.7[b]	6.5	

[a]Lee (1977).

[b]This work. A correction has been made for the analyzer transmission. The uncertainty in this correction is responsible for the uncertainty in these ratios.

[c]From Holland et al. (1981) with a 2.0Å bandwidth. Note that Lewandowski et al. (1981) state that the monochromator calibration in the work of Holland et al. may be off by 2Å.

[d]From Lewandowski et al. (1981) with a 0.9Å bandwidth.

FIGURE CAPTIONS

Fig. 1 Energy-level diagram for barium, including the various excitation and decay channels.

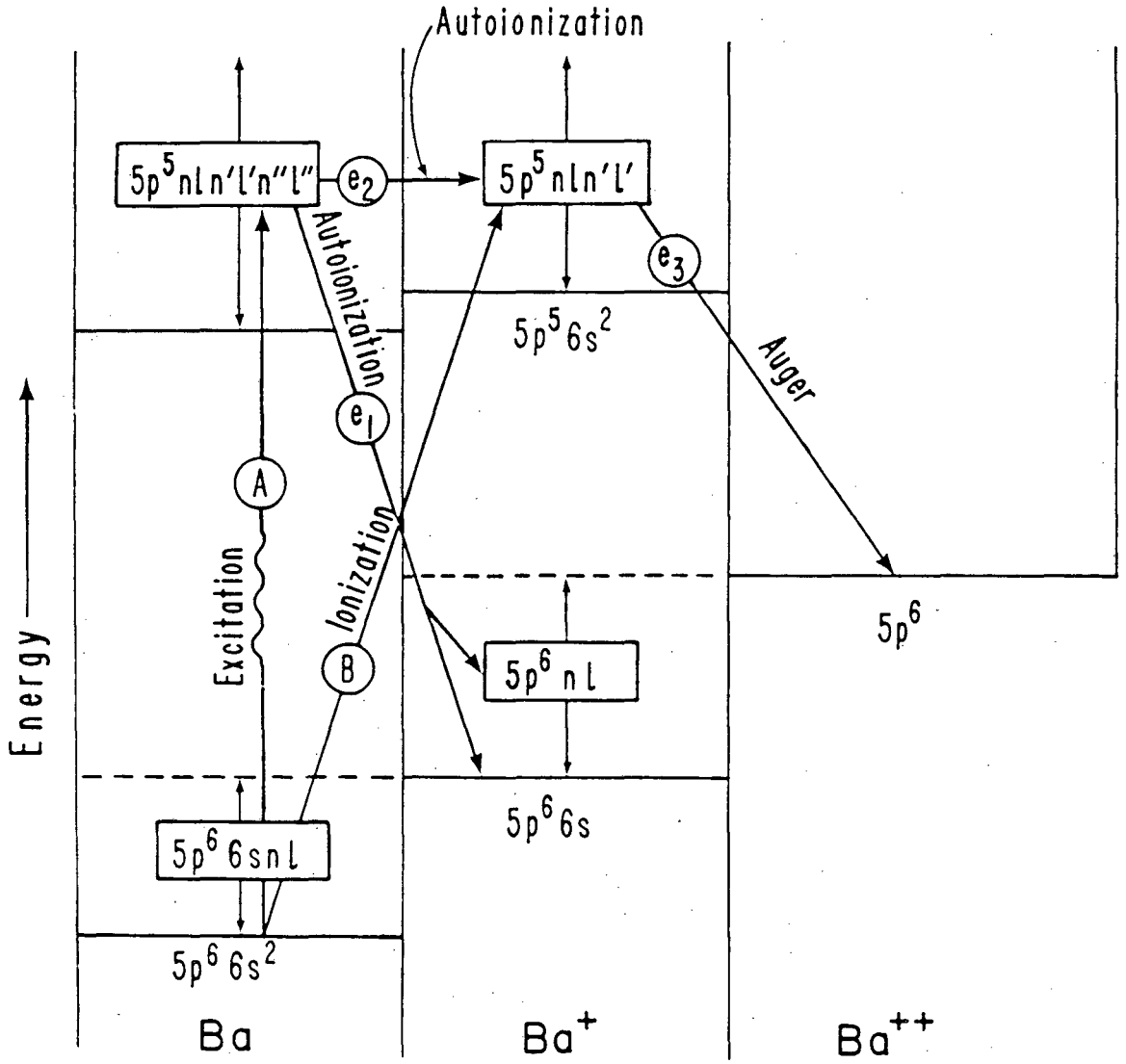
Fig. 2 The total electron yield spectrum from this work (bottom) and the Ba^{+} and Ba^{++} total ion yield spectrum of Lewandowski et al. (1982) (top). The ordinate scale applies only to the present results.

Fig. 3 The partial cross sections of the Ba^{+} $5p^{5}nl$ states where (nl = 6s, 5d, 6p, 7s, and 6d). The bottom panel shows the total electron yield as in Fig. 2. The units of the ordinate scales are the same, but arbitrary.

Fig. 4 The angular distribution asymmetry parameter, β, of the 6s, 5d and 6p photoelectrons. The error bars represent counting statistics only. The solid curve is the total electron yield, shown for comparison.

Fig. 5 Electron kinetic-energy spectra at three different photon energies, showing the Auger peak intensities. The peaks at kinetic energies above 6.6 eV in the $h\nu$ = 21.4 eV spectrum and at kinetic energies above 7.6 eV in the $h\nu$ = 22.8 eV spectrum are from Ba^{+} $5p^{6}nl$ photoelectrons, not Auger electrons.

Fig. 6 Electron kinetic energy spectra at three photon energies showing Auger peak intensities. Primary photoelectrons peaks (A, B and C) are also observed in the $h\nu$ = 28.9 eV spectrum, as discussed in the text. The corresponding Auger peaks are indicated as A', B', and C', respectively.



XBL 794-1342A

Figure 1

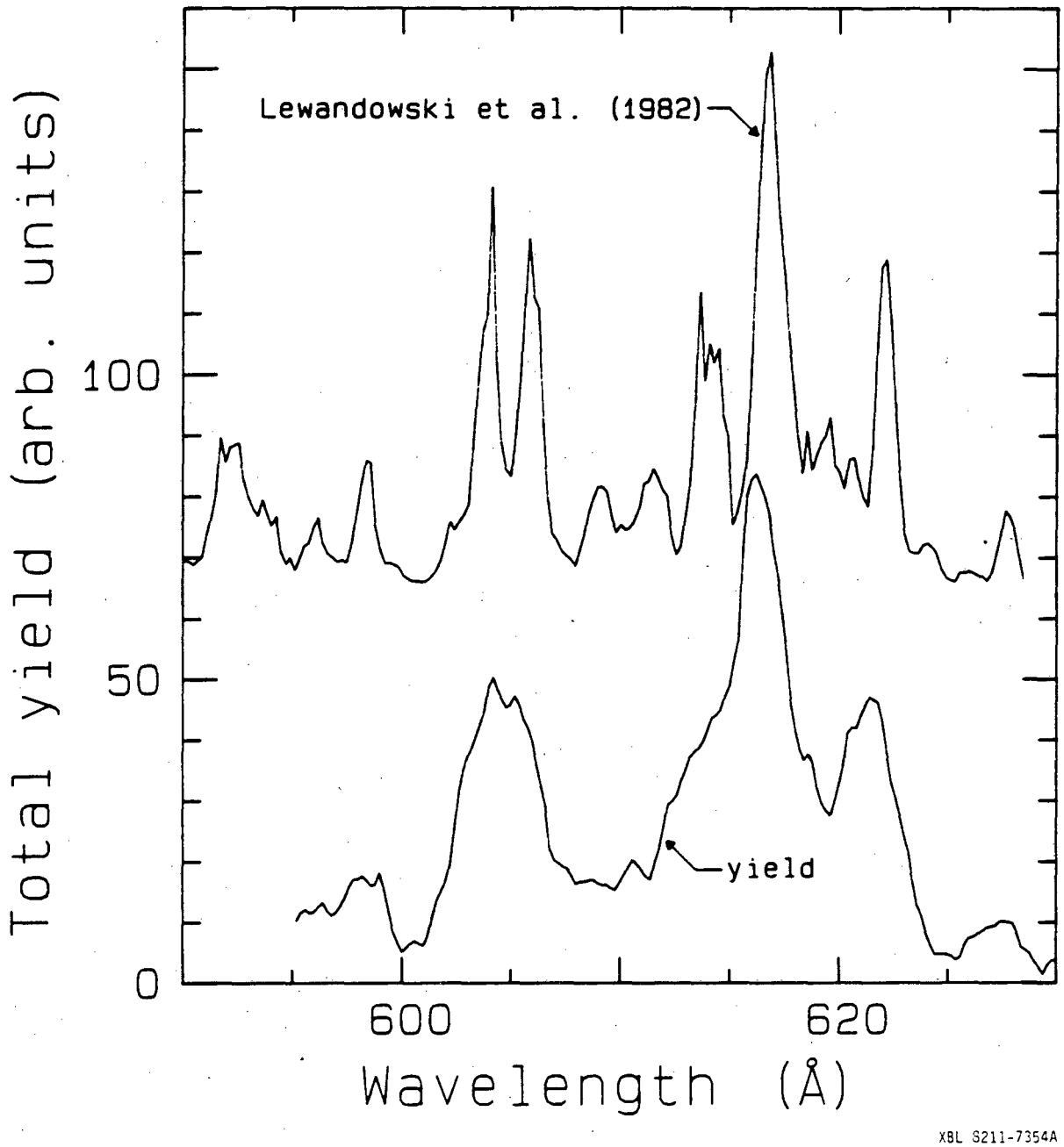
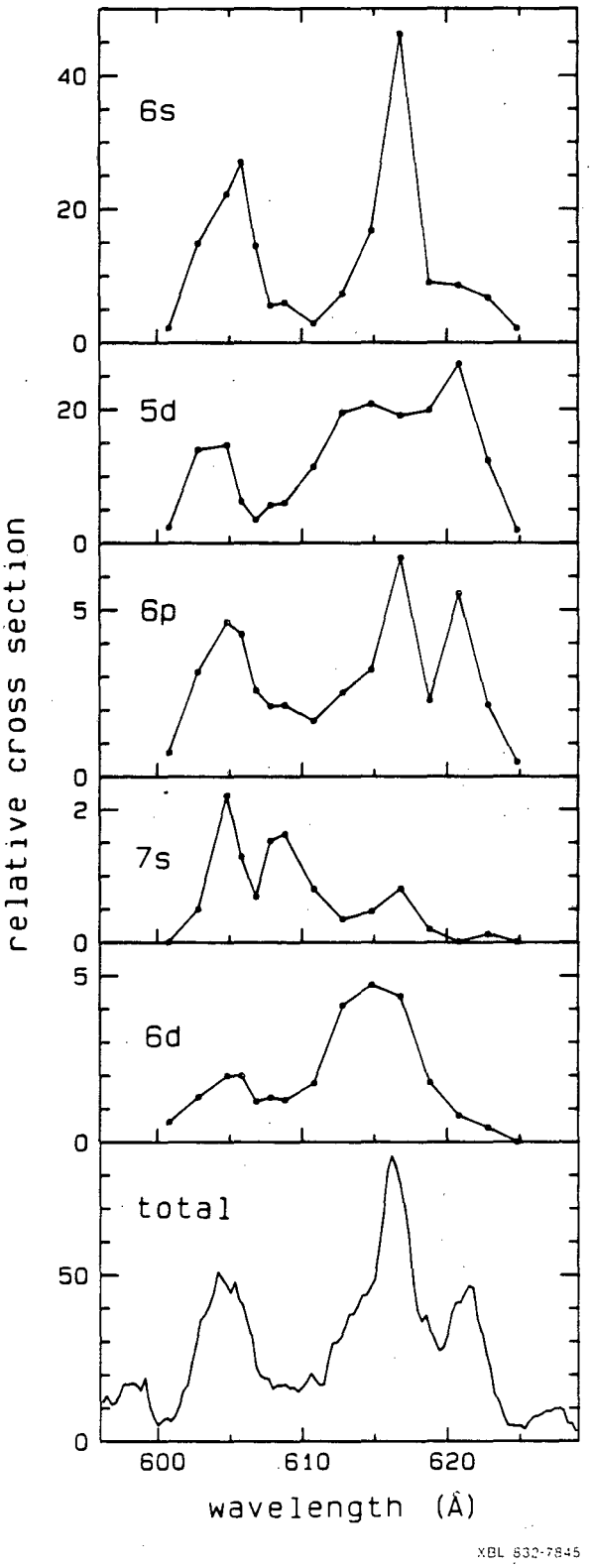

Lewandowski et al. (1982)
yield
Total yield (arb. units)
0
50
100
600
620
Wavelength (Å)
XBL 8211-7354A

Figure 2


6s
5d
6p
7s
6d
total
relative cross section
wavelength (Å)
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610
620
XBL 832-7845

Figure 3

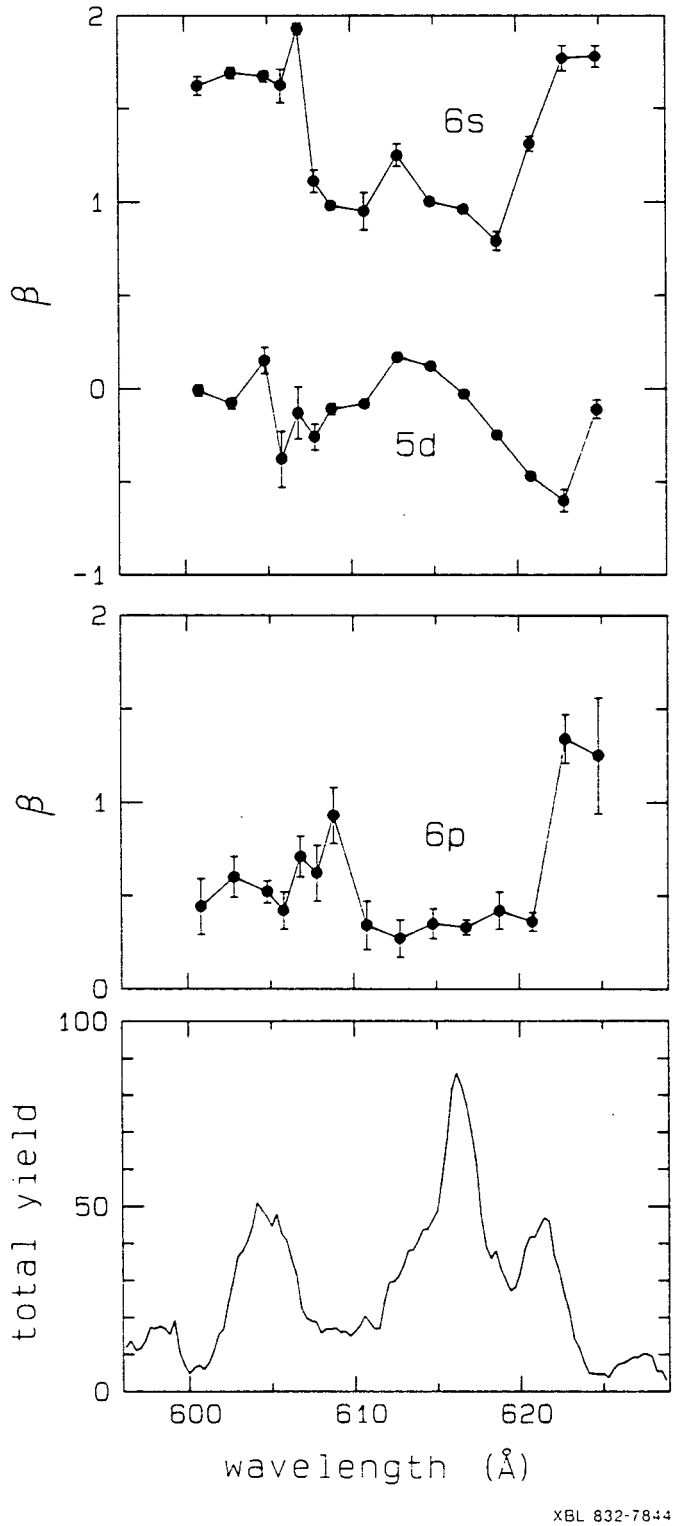

2
1
0
-1
β
6s
5d
2
1
0
β
6p
100
50
0
total yield
600
610
620
wavelength (Å)
XBL 832-7844

Figure 4

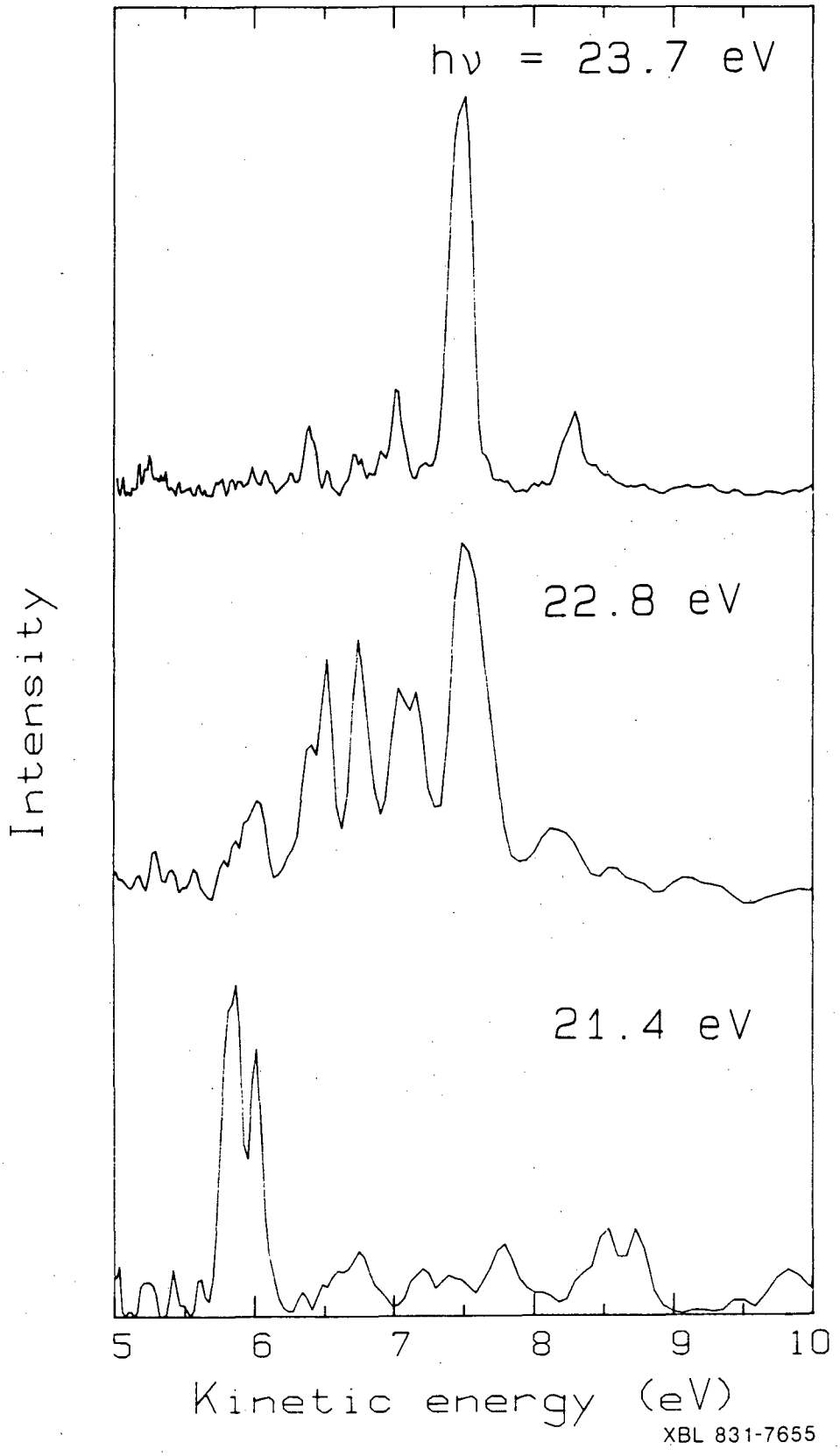
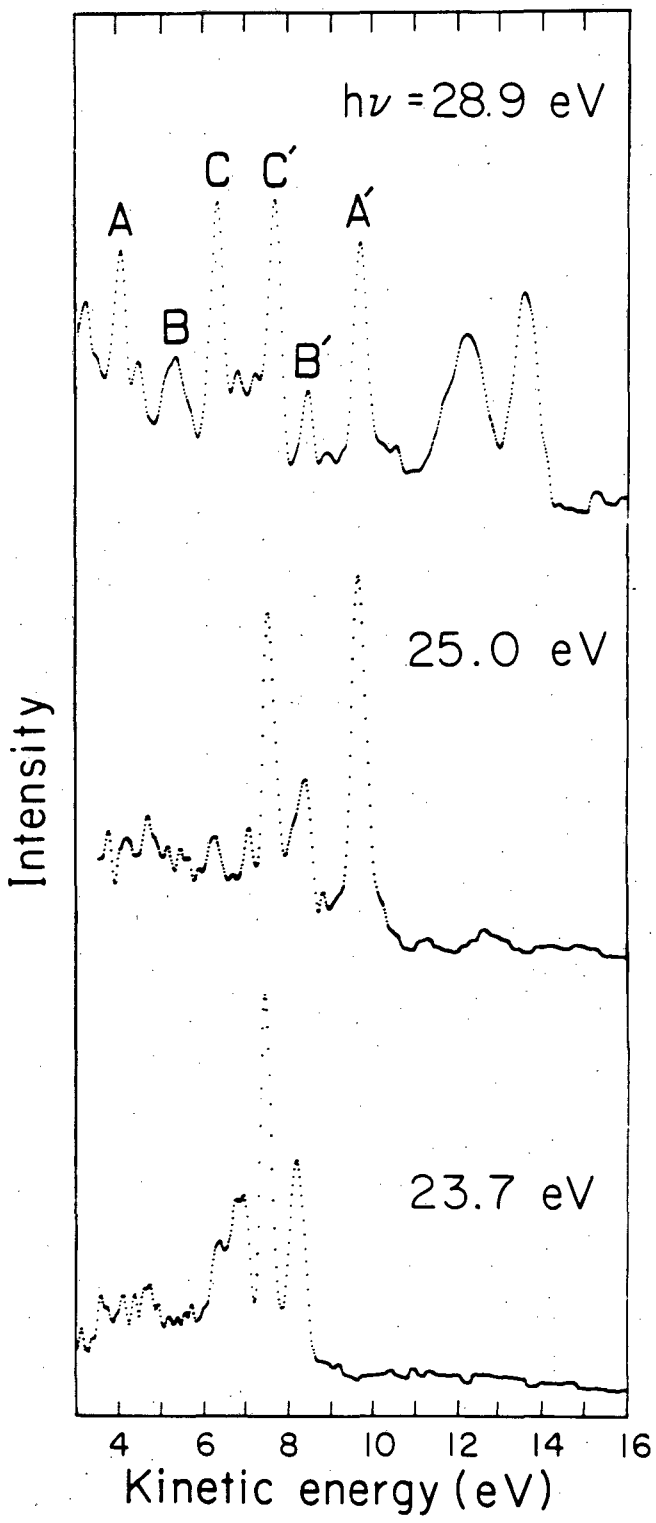

hν = 23.7 eV
22.8 eV
21.4 eV
Intensity
5
6
7
8
9
10
Kinetic energy (eV)
XBL 831-7655

Figure 5



XBL 797-2304B

Figure 6

TECHNICAL INFORMATION DEPARTMENT
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