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Strong Correlation and Alignment near the Be 1s Photoionization Threshold

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Photoionization in atomic beryllium has been studied in detail near the 1s threshold. Multiple excitation to the $1s(2s2p\ ^{1,3}P)$ states, i.e., conjugate shakeup, is found to be unusually strong, 40% of single excitation. The 3P component rapidly loses strength with increasing photon energy, while the 1P component persists. The $1s2s2p$ states are highly aligned, as shown by the Auger-electron angular distribution. The A_2 value of -0.65 implies a d -wave contribution of 0.39. Our results indicate that resonances reported for the solid-state photoabsorption are of atomic origin.

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Following helium, beryllium is the most important closed-shell atom which is simple enough to be theoretically tractable in a rigorous manner. Unlike helium, it exhibits many-body properties of a system with more than two electrons arranged in different principal shells, and is a prototype for multiconfiguration interactions in the ground state.^{1,2} In the present work we emphasize the many-body effects, and their consequences, as they manifest themselves in two-electron excitations in the threshold region of 1s photoionization. In particular, these excitations involve P states, which, in contrast to He,³⁻⁵ are not degenerate with S states, and which, in contrast to Li,^{6,7} can be clearly separated from S states according to their multiplicity, namely 1P and 3P .

An important aspect of this study is its relationship to photoabsorption measurements of metallic Be. In that work,^{8,9} structure observed in the 1s threshold region has not yet been interpreted, and the present results can indicate those features which are due to atomic effects.

The experiments were performed at the Aladdin storage ring with our previously described electron spectrometer.¹⁰ Photons exiting from a grasshopper monochromator interacted in a source cell with Be atoms produced in a Ta oven held at 1160°C. The bandpass of the photon beam was 0.55(5) Å and the resolution $\Delta E/E$ of the electron analyzers was 1%. Electron spectra were recorded simultaneously in two analyzers set to 0° and 90°, or 55° and 145°, with respect to the major polarization axis ($P_s=0.88$). Both photoelectron and Auger spectra were recorded, but the threshold data were derived entirely from Auger spectra. Possible differences between photoelectron and Auger intensities due to higher-order radiation were found to be negligible, less than 3%.

The schematic of Fig. 1 depicts the major excitation and deexcitation channels for the 1s photoionization of Be at low excess energy. The left-hand channels repre-

sent the photoelectron lines with energies ϵ_1 to ϵ_3 for a given $h\nu$ and the resulting ionic states. The Auger-line energies E_A are given on the right, together with the two-electron continuum. As an example of an Auger spectrum, we show in Fig. 2 the intensities recorded at 0° and 90° for $h\nu=130$ eV, together with line contours of the Voigt fits to the data points. Shown are the main Auger line, $1s2s^2 \rightarrow 1s^2$, corresponding to the single-electron excitation, and the two clearly resolved Auger

FIG. 1. Schematic diagram of major 1s excitation and deexcitation processes in Be, showing ionization limits and electron continua. Energies listed are our measured values. Outer-shell excitations are not shown.

FIG. 2. Be K Auger spectrum showing the main Auger line and the two satellite lines associated with the $1s2s2p$ excitation states. Data and fitted contours are shown for spectra recorded at 0° (circles) and 90° (crosses). $h\nu = 130$ eV.

satellite lines corresponding to two-electron transitions which produce the intermediate states $1s(2s2p^3P)$ and $1s(2s2p^1P)$.

The threshold region was examined in detail by setting of the electron energy analyzers to the main and satellite peaks shown in Fig. 2 and variation of the photon energy in 0.20-eV steps from 122 to 142 eV. Data were obtained at 55° for each of the three components, and the results, averaged from two runs each, are plotted in Fig. 3. Each trace was corrected for background and normalized to photon flux, grating efficiency, and vapor density. A final normalization was made with the aid of Auger spectra taken at several fixed photon energies. Hence, Fig. 3 gives the partial cross sections, σ_i , for the major transitions on a relative scale.

Several observations can be made from examination of Fig. 3. First, the so-called conjugate shakeup process, in which the $1s$ electron goes into an ϵs -continuum channel and the excited $2s$ electron takes up the angular momentum ($2s \rightarrow 2p$), is very strong, up to 40% of single ionization. Until now, substantial, albeit smaller, strengths for this type of process have only been reported for He¹¹ and Li.^{6,7} Admixture of $2p^2$ into the single-particle $1s^2 2s^2$ ground state apparently facilitates transitions to the $1s2s2p$ states in Be⁺. The $1s2s2p$ states remain strongly populated at higher energies compared with the

FIG. 3. Partial cross sections of the three major ionization channels for $1s$ photoionization of Be, as obtained from K Auger spectra. The top trace (circles) corresponds to single- $1s$ -electron emission, $1s2s^2\epsilon l$, the middle trace (crosses) to $1s(2s2p^3P)\epsilon l$, and the bottom trace (squares) to $1s(2s2p^1P)\epsilon l$. All traces have a common ordinate with offsets as indicated. Line numbers refer to the states listed in Table I.

$1s2s3s$ states. At $h\nu \geq 160$ eV, we observe a value of about 0.5 for the ratio $I(2s2p)/I(2s3s)$. This is in contrast to He, where the analogous $2p/2s$ ratio becomes very small.¹¹ Secondly, there is a pronounced difference between $\sigma(^3P)$ and $\sigma(^1P)$. Following a high threshold value for both, $\sigma(^3P)$ drops off rapidly with energy, whereas $\sigma(^1P)$ is essentially constant. This first observation of the steep decrease of $\sigma(^3P)$ in photoionization with excitation processes parallels the behavior of triplet states in particle excitation. There, triplet states are seen to lose strength by 2 orders of magnitude more rapidly than singlet states.¹² Evidently, it is the spin exchange that dominates the behavior.

Third, structure appears in the σ curves of the various channels, some of it occurring in two or all three channels at the same photon energy. This suggests resonances due to quasiscrete multiple excitation states. In the absence of extensive energy calculations for the more highly excited states, only a few of the resonances can be assigned. The strongest resonance occurs at 125.1 eV, just above the $1s$ threshold, and displays a Fano-Beutler profile. It couples strongly with the $1s2s^2\epsilon p$ channel and

TABLE I. Energy levels of $1s$ excitation in beryllium.

Number ^a	Ionic state	Energy (eV)
1	$1s2s2p^{21,3}P$	122.3 ^b
2	$1s2s^2S$	123.56 ^c
3	$1s2s2p^4P$	125.3 ^b
4	$1s2p^{31,3}P$	125.9 ^b
5	$1s(2s2p^3P)^2P$	128.1 ^d
6	$1s(2s2p^1P)^2P$	130.3 ^d
7	$1s2p^2S$	135.1 ^d
8	$(1s2s^3S)3s^2S$	138.1 ^d
9	$(1s2s^3S)3p^2P$	138.7 ^d
10	$(1s2s^3S)3d^2D$	139.9 ^d
11	$(1s2s^1S)3s^2S$	140.5 ^d

^aStates numbered 1 to 4 arise from transitions $1s \rightarrow 2p$.^bCalculated energies of Ref. 14, referred to 123.56 eV.^cOur determination referred to $E(\text{Xe } 4d_{5/2}) = 67.54$ eV.^dEnergies are from Ref. 15.

contributes to both satellite channels below their thresholds, indicative of a resonance Raman effect.¹³

Although a number of excited states occur in the vicinity of the 125.1-eV resonance (see Table I) according to calculations¹⁴ and beam-foil experiments,¹⁵ we believe it to be identical with the absorption line reported previously in a plasma source,¹⁶ namely $1s(2s2p^3P)3s$. This two-electron excitation could then interfere with ionization of a $1s$ electron, giving rise to a slow photoelectron (1.5 eV) and the 96.0-eV Auger electron we observe. The only calculation¹⁷ of $1s$ photoionization places this resonance at slightly lower energy. The shoulder at about 127 eV can be assigned to the higher members of the $1s(2s2p^3P)ns$ (or nd), $n \geq 4$, series. Photoabsorption spectra^{8,9} of metallic Be display a resonance structure similar to the one we observed here for the atom. It appears persuasive to assign the resonance in the metal to the same two-electron process.

An important characteristic of these two-electron processes in Be is that they create an intermediate state that is aligned.¹⁸ The alignment pertains to an excited-electron state, rather than a hole state, which is produced according to

$$(1s^22s^2)^1S_0 + h\nu \rightarrow 1s(2s2p^{1,3}P)^2P + e^-(l=s, d).$$

Alignment results in an anisotropy in the angular distribution of the Auger electrons,¹⁹ given by

$$d\sigma/d\Omega = (\sigma/4\pi)[1 + (\beta_A/4)(1 + 3P_s \cos 2\theta)],$$

where θ is the angle measured with respect to the quantization axis, taken to be the direction of linear polarization of the radiation, and P_s is the polarization. The anisotropy is clearly seen in Fig. 2; our experimental values are summarized in Table II.

For Auger decay into a state 1S_0 , the parameter β_A is

TABLE II. Asymmetry parameters β_A for Auger satellite lines $1s^2 \rightarrow 1s(2s2p^{1,3}P)$. Numbers are referred to a value of $\beta_A = 0.0$ for the principal transition, $1s^2 \rightarrow 1s2s^2$.

$h\nu$ (eV)	3P	β_A	1P
128.6	1.3(2)	...	...
130.0	1.3(2)	...	1.4(2)
132.0	1.4(2)	...	1.1(2)
135.0	1.2(2)	...	1.3(2)
140.0	0.9(3)	...	1.3(2)
150.0	0.3(3)	...	0.6(2)
160.0	...	...	0.6(3)
175.0	...	...	0.2(3)

directly proportional to the alignment A_2 , defined by

$$A_2 = \frac{\sum_M \sigma(J, M)[3M^2 - J(J+1)]}{J(J+1) \sum_M \sigma(J, M)},$$

where $\sigma(J, M)$ is the population of the (J, M_J) magnetic sublevel, equal to the cross section for production of that level. From Table II the parameter A_2 is equal to -0.65 ± 0.06 at the lower energies.

Greene and Zare²⁰ have determined limits to the alignment in an analysis which includes double excitations. For our case, the calculation gives a limit of $A_2 = -1.0$, or a β_A value of 2.0. A comparison can be made between the $1s^22s^2 \rightarrow 1s2s2p\ell$ ionization/excitation in Be and the comparable $1s^2 \rightarrow 2p\ell$ ionization/excitation in helium measured by Jimenez-Mier, Caldwell, and Ederer.³ If we assume that the two electrons in Be that are not actually undergoing the transition remain passive, then the He analysis can be applied directly to this process. This implies that we can write the alignment as a function of the ratio

$$\xi = \sigma(2p, \epsilon d) / [\sigma(2p, \epsilon s) + \sigma(2p, \epsilon d)],$$

where $\sigma(2p, \ell)$ is the cross section for production of the ℓ th partial wave. Using this analysis, we obtain a ratio $\xi = 0.39 \pm 0.08$ for the contribution of the d wave near threshold. This is a much larger d -wave contribution than was observed in He,³ where $\xi = 0.25$. The alignment retains this high threshold value in the region where other excitation states may feed the decay. It decreases toward higher energies, where we expect the d -wave contribution to become even stronger.

In summary, this first study of atomic Be by use of electron spectrometry has shown the importance of the so-called conjugate shakeup processes in the $1s$ -threshold region and the strong alignment of the resulting intermediate states. Corresponding theoretical calculations are still lacking, but the relative simplicity of the system should allow one to identify the important interactions and obtain accurate results for comparison with experiment.

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