Evidence for Resonance States of Lead Decaying Into Two Exit Channels with Counterbalancing Contributions

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The analysis of the electrons emitted in the decay of the $6s^26p^2 + h\nu \rightarrow 6s6p^2(^4P_{1/2})np_{3/2}$ resonance series of Pb shows that negative contributions occur in the $6p^{-1}{}^2P_{1/2}$ channel and positive contributions in the $6p^{-1}{}^2P_{3/2}$ channel due to interference with the direct photoionization process. These contributions effectively cancel. Hence the series is difficult to observe in absorption, but can easily be delineated in emission.

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For the excitation of a $6s$ electron of lead, each member of the series, $6s^26p^2 \rightarrow 6s6p^2np$, $n \geq 7$, should consist of a doublet corresponding to the $np_{1/2}$ and the $np_{3/2}$ states. Such doublets were, indeed, observed by Connerade *et al.*[1] in photoabsorption measurements for two of the $6s$ series, those converging to the $^2D_{5/2}$ and $^2P_{3/2}$ limits, but not for the strong series converging to the $^4P_{1/2}$ limit. The doublet component which is missing in absorption can, however, be recovered, as we will show, in an emission experiment in which we monitored the electrons that arise from the decay of the excitation or resonance states. The $6s6p^2np$ states are imbedded in two continua as illustrated in Fig. 1 and we present evidence in this paper that the decay of the $np_{3/2}$ components into these two exit channels via autoionization will proceed in such a way that constructive interference occurs with the direct photoionization process in one channel and destructive interference in the other channel leading to effective cancellation of the overall intensity residing in the resonance. As a result, either no or very weak features occur in an absorption spectrum. However, the measurement of the partial widths for *each* of the two possible exit channels provides direct evidence of the phenomenon which undoubtedly is not restricted to the case of Pb, but may occur in other systems, atomic, molecular, nuclear, etc., whenever two or more channels[2] are available. In related work, Woodruff and Samson[3] previously invoked a cancellation effect in the case of doubly excited He on the basis of a comparison between the total cross section and the partial cross section measured for one channel only. Kobrin *et*

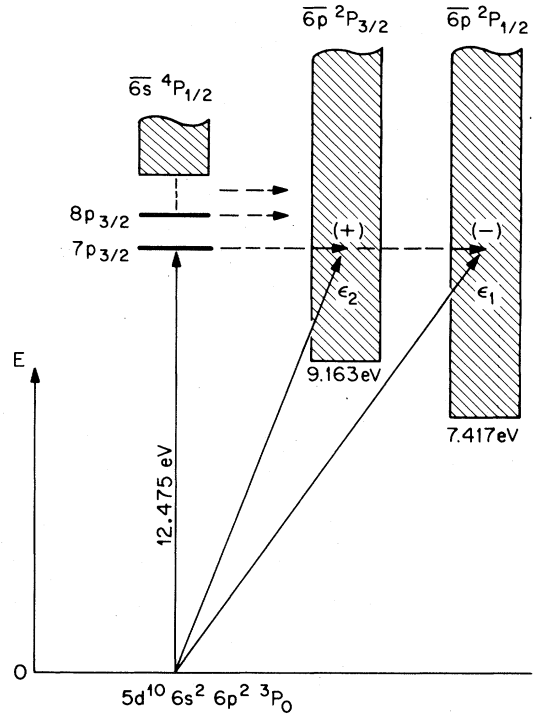

FIG. 1. Photoionization in the region of the $np_{3/2}$ resonance series of Pb. Decay into $^2P_{3/2}$ leads to positive contributions (plus) and into $^2P_{1/2}$ to negative contributions (minus).

al.[4] reported a pronounced window for the 4*d* subshell and weak peaks for 5*s* and 5*p* at the 588-Å resonance in Cd.

The experimental setup was described earlier,[5] and the experimental procedures were similar to those reported recently.[6] Briefly, Pb was evaporated from a Ta oven at 1150 K. A Seya monochromator at the Wisconsin Synchrotron Radiation Source provided a photon beam with a polarization of $p \approx 0.96$ and a bandpass of about 1.6 Å. Photoelectrons from the metal vapor were analyzed at the magic angle, $\varphi_m = 55°$ in respect to the polarization of the photon beam, so that the measured intensity was proportional to the cross section or width of the channel selected by the electron analyzer. Constant-ionic-state (CIS) spectra were recorded for the two channels that leave the ion in the $6p^{-1}{}^2P_{1/2}$ and $6p^{-1}{}^2P_{3/2}$ states by tracking the corresponding photoelectrons synchronously with the stepwise advance of the photon energy. These CIS spectra yielded relative partial widths as a function of photon energy following correction for detector background and normalization to photon flux and vapor density. Photoelectron spectra (PES) were also recorded at selected, fixed photon energies for reference purposes and, with an adjustment for the analyzer transmission, to quantitatively relate the CIS spectra to each other.

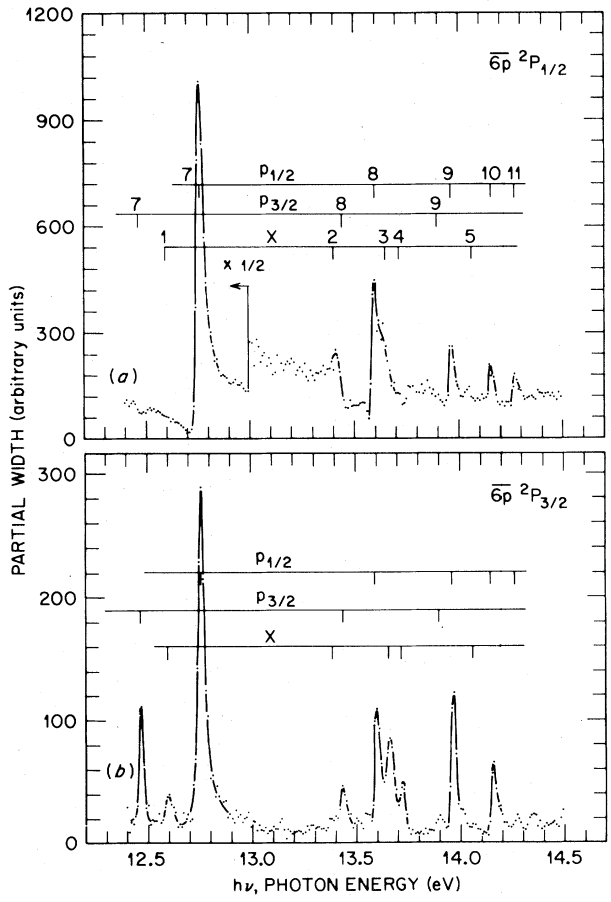

FIG. 2. Partial widths of the two exit channels in the $6s^2 6p^2 \rightarrow 6s6p^2(^4P_{1/2}), np(\frac{1}{2};\frac{3}{2})$ series of Pb. Photon bandpass was about 22 meV. Unidentified peaks are denoted by *X*.

The partial widths for the $6p^{-1}{}^2P_{1/2}$ and $6p^{-1}$-$^2P_{3/2}$ channels, the only channels into which the resonance states of the $6s^2 6p^2 \rightarrow 6s6p^2(^4P_{1/2})np$ series can decay, are displayed in Fig. 2. Two series and a number of unknown lines can be identified in both spectra. The $np_{1/2}$ series is identical with the absorption series reported by Connerade *et al.*[1] The "missing" $np_{3/2}$ series can be discerned in both spectra. The identification is based on the observation that the doublet intervals scale reasonably well with the $1/(n^*)^3$ rule, and that as seen from Table I, the effective quantum numbers n^* for the $np_{3/2}$ series relate well to those for the $np_{1/2}$ series. In addition, the n^* values of $np_{3/2}$ compare well with those found for the $6s^2 6p^2 \rightarrow 6s6p^2(^2D_{5/2})np_{3/2}$ series.[1]

The major point in the spectra of Fig. 2 is the

TABLE I. Energies, relative intensities, and effective quantum numbers of the resonances in the $6p^{-1}{}^2P_{1/2}$ and $6p^{-1}{}^2P_{3/2}$ exit channels over the $6s^{-1}[^4P_{1/2}] \rightarrow np(\frac{1}{2};\frac{3}{2})$ autoionization region.

Resonance state[a]	Energy (eV)[b]	Intensity (%) $6p^{-1}{}^2P_{1/2}$	Intensity (%) $6p^{-1}{}^2P_{3/2}$	n^*
$7p_{3/2}$	12.475(15)	dip	2.6(4)	2.53
$X1$	12.61(2)	···	1.4(3)	···
$7p_{1/2}$	12.765(10)	100.0[c]	14.5(1.1)	2.73
$X2$	13.42(2)	···	···	···
$8p_{3/2}$	13.45(2)	dip	1.9(3)	3.44
$8p_{1/2}$	13.610(10)		3.5(4)	3.71
$X3$	13.675(15)	15.0(1.5)	3.1(4)	···
$X4$	13.73(2)		1.1(2)	···
$9p_{3/2}$	13.91(2)	dip(?)	0.6(1)	4.45
$9p_{1/2}$	13.975(15)	4.3(5)	3.6(4)	4.68
$X5$	14.07(2)		0.3(2)	···
$10p_{1/2}$	14.170(15)	2.9(4)	2.4(4)	5.65
$11p_{1/2}$	14.28(2)	2.3(5)	0.8(3)	6.55
$12p_{1/2}$	14.38(2)	1.3(5)	1.0(3)	7.92

[a] $6s6p^2[^4P_{1/2}]np(\frac{1}{2};\frac{3}{2})$ and unidentified states, *X*.

[b] Average values from both exit channels; both sets of values agree within 5 meV; $Xe5p_{1/2} \rightarrow ns, nd$ resonances were used for calibration.

[c] Reference for the intensities of *both* exit channels.

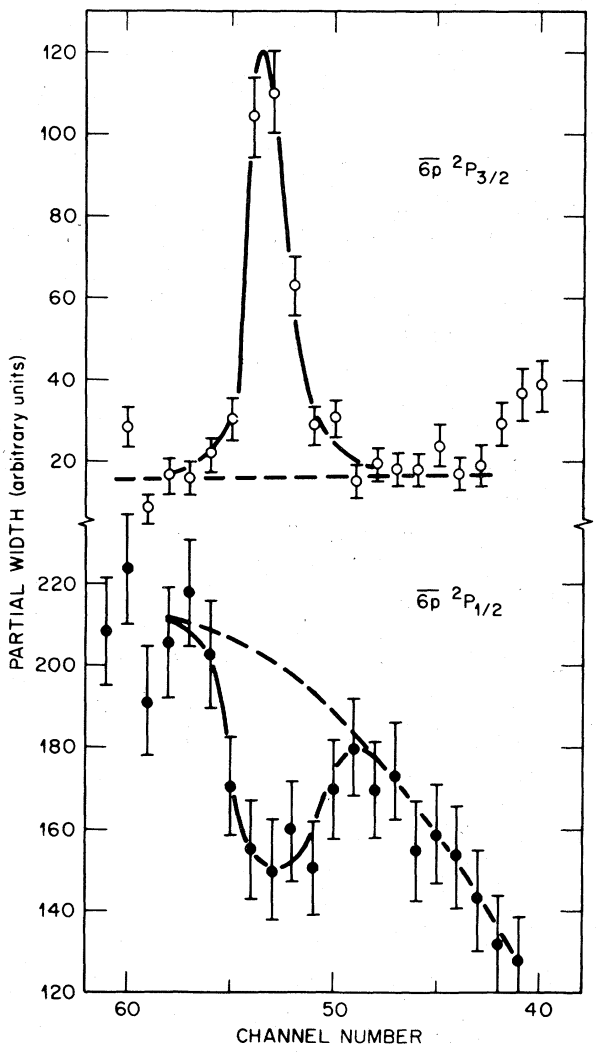

FIG. 3. Enlarged view of Fig. 2 around the $7p_{3/2}$ resonance state showing positive and negative contributions in the two available exit channels. Dashed line indicates estimated continuous widths.

window that occurs in the $6p^{-1}{}^2P_{1/2}$ channel at the $7p_{3/2}$ resonance and the higher resonances, as opposed to the "normal" line that appears in the $6p^{-1}{}^2P_{3/2}$ channel. The region around $7p_{3/2}$ is enlarged in Fig. 3; it shows that the window intensity counterbalances the line intensity, namely $-235(25)$ versus $+287(21)$ arbitrary units for the respective areas. Hence, an effective cancellation occurs in the sum spectrum, which in the absence of other transitions corresponds to the absorption spectrum. Actually a small and narrow positive contribution will remain in the sum spectrum, because the peak is somewhat narrower than the window.

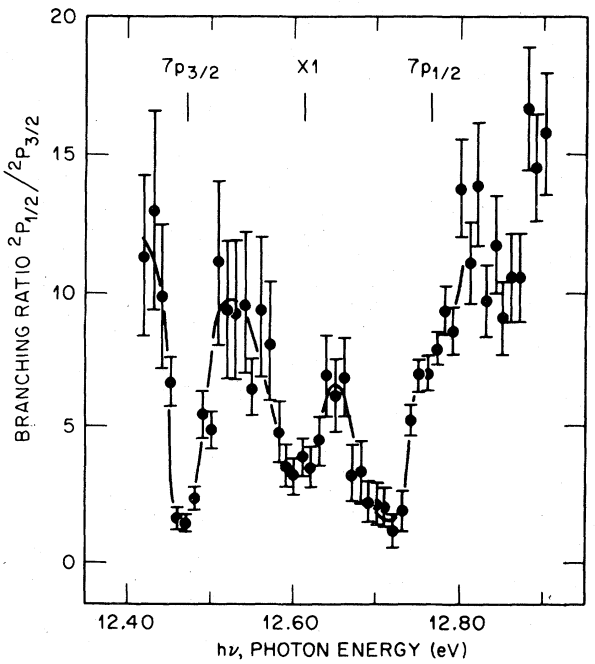

FIG. 4. Branching ratio of the partial widths for the $n=7$ resonance states and an unidentified line.

The fact that the $7p_{3/2}$ resonance as well as the higher members yield normal peak shapes with a very large profile parameter,[7] q, in the $6p^{-1}$-$^2P_{3/2}$ channel is not surprising, since the population of this channel is almost entirely indirect via the resonance states. As discussed by Süzer, Banna, and Shirley,[8] direct transitions to $6p^{-1}$-$^2P_{3/2}$ originate from the $(p_{3/2})^2$ state that is admixed to $(p_{1/2})^2$ giving a ground-state wave function

$$\psi = C_1(p_{1/2})^2 + C_2(p_{3/2})^2 . \quad (1)$$

However, the coefficient C_2 is small compared with C_1 resulting in a weak direct transition strength. On the other hand, the transitions from the $(p_{1/2})^2$ state, which populates the $6p^{-1}{}^2P_{1/2}$ channel, are comparatively strong so that the interactions with the indirect transitions via the resonance states lead to q parameters that should not be large. In the case of the $7p_{1/2}$ state, we find a reasonably good fit of the observed line shape[9] to the Fano resonance profile[7] using $q = 3.6(3)$, a width of $\Gamma = 32(5)$ meV, and $E_r = 12.765(10)$ eV.

As shown in Fig. 4, the branching ratio $^2P_{1/2}/^2P_{3/2}$ of the two continuum channels is subject to drastic variations in the neighborhood of the resonance states. In particular, a minimum occurs at a resonance that decays into channels

with negative and positive contributions. We note parenthetically that an attempt to estimate the ratio C_1/C_2 of the coefficients in Eq. (1) from the branching ratios determined in the region of resonance series would be futile. To relate the branching ratio to C_1/C_2 in a straightforward manner would require a measurement outside the resonance regions, where at the same time the strengths for the direct transitions could be assumed to be similar.

As seen from Table I and Fig. 2, the relative intensities within the $np_{1/2}$ and $np_{3/2}$ series generally deviate from the simple $1/(n^*)^3$ rule. The $7p_{1/2}$ intensity is found to be especially great. Our energy determinations agree satisfactorily with those reported from absorption data.[1] A number of unidentified lines are marked in Fig. 2. We suspect the lines $X3$ and $X4$ near $8p_{1/2}$ to be due to the two electron excitations $6p^2 \rightarrow 7s, 7p$ whose energies are estimated[10] to be 13.89 and 13.95 eV, which values are somewhat greater than the energies of the observed lines.

In conclusion, we presented evidence for resonances that make negative contributions to one exit channel and positive contributions to the other. By measuring the individual contributions directly in an electron emission experiment, in which the various channels are distinguished, we were able to quantitatively determine these contributions and recover an excitation series that could not be seen in a photoabsorption spectrum which, by its nature, does not differentiate between the various channels. This work serves to underline the need for studying the decay characteristics in order to be able to interpret anomalies observed in excitation experiments.

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