

Empirical Regularities for Excitation Cross-Sections of Lead Atom (Radiative Transitions from Even Levels)

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Abstract

Excitation of even levels of lead atoms was studied using the method of extended crossing beams with recording of optical signal emitted by excited atoms. Forty-eight excitation cross sections have been measured at an incident electron energy of 50 eV. Four optical excitation functions (OEFs) have been recorded under varying electron energies in the range $E = 0\text{--}200$ eV. The cross-sections obtained have been used to examination of empirical regularities in the behavior of excitation cross-sections, including those of perturbed series.

1 Introduction

Lead is one of the elements of interest to pure sciences and applied research alike. An important scientific area is astrophysics where recent decades have seen intense research into optical spectra of stars with peculiar chemical compositions. This researches provides a crucial insight into chemical and dynamic evolution of the Local Group of galaxies and they are essential for understanding physical processes inside stars. Objects of studies include stars of different classes. For instance, publications [5, 12] report on spectra of HD 101065 and HD 213637 roAp stars (cool magnetic chemically peculiar stars) respectively, even though no lines were discovered in these stars that could be reliably identified as belonging lead. A similar situation is reported in a study concerned with chemically peculiar stars spectra of Late-B and Early-A classes of the main sequence (HgMn stars) [1]. The latter study focuses on the content of heavy elements in HgMn stars, with

the superheavy element sequence appearing as: Pt, Au, Hg, Tl, and Bi. They follow immediately in the periodic table by radioactive elements. Lead, taking the place between thallium and bismuth, is reported in [1] to be completely absent. The absence of lead lines in the spectra of objects mentioned above can be most likely attributed to peculiarities of its spectrum (i.e. by accidental blending of sensitive lead lines with lines of other elements and partially with molecular band spectra). Nevertheless, discovery of actual lead lines in spectra of peculiar stars remains a problem of high relevance.

Lead is one of the oldest elements known to mankind. Featuring an uncommon combination of physical and chemical properties (high density, high plasticity, low melting point, the ability of molten lead to dissolve gold, etc.), lead enjoyed a multitude of diverse applications long ago in the pre-scientific era. However, accelerated scientific advancement from 19th century onward was able to expand this range of applications even further into chemical industry, electrical engineering, radiation shielding, medicine etc.

Gas-discharge lasers employing lead atom and ion transitions have been developed. Laser generation using PbI transition at wavelength $\lambda = 722.9$ nm was pioneered in [7]. However estimates in [10] show that generation using other transitions originating at the same level, $6p7s(1/2,1/2)^o_1$ (or $6p7s\ ^3P^o_1$, using the *LS* notation), should be just as efficient. The absence of generation using $6p7s(1/2,1/2)^o_1 \rightarrow 6p^2(3/2,1/2)_2$ and $6p7s(1/2,1/2)^o_1 \rightarrow 6p^2(3/2,1/2)_1$ transitions in [7] was explained in [10] to be the result of the electric current pulse front being insufficiently steep. With steeper pulse front in [10], generation was obtained for three new laser lines of PbI, supporting the authors' supposition in [10] on the role of pulse front steepness. A later study of electron-beam excited lead vapor laser [3] concluded that the efficiency and generation power of self-limited metal vapor lasers could be improved substantially if electron beam excitation were used. However all development of these devices had so far to rely on empirical approaches due to the lack of reference data on elementary processes in the working medium of such lasers.

At the same time there is ongoing research into spectral and collision properties of the lead atom. The most detailed research of the PbI spectrum and low-lying energy levels has been undertaken in [18] listing 370 classified lines in the wavelength range $\lambda = 173$ – 1256 nm, as well as 59 even and 58 odd levels. Absorption spectrum studies in the vacuum UV range in [4] yielded extensive information on odd PbI levels. Completely studied are *s* series up to $n = 59$ and *d* series up to $n = 77$. Three-photon excitation of even levels of Rydberg series was studied later in [19, 6, 9]. There are instances where findings of referenced papers diverge severely, in addition to falling out of agreement with findings reported in [18] (see below).

The lower level for the $\lambda = 722.9$ nm laser line is $6p^2(3/2,3/2)_2$ ($6p^2\ ^1D_2$ in the *LS* notation). As the lifetime of the lower level τ affects the probability of obtaining inverse population on laser transition in a dramatic manner, an attempt to measure it was undertaken in [11]. Values of τ reported for metastable levels of lead and manganese atoms are equal within a measurement error margin. It was

additionally concluded that “direct electron impact is the principal mechanism of exciting upper laser levels in lead or manganese atoms from their ground state” [11, P. 927].

Both experimentally [2] and theoretically [14] studies of lead atom excitation by electron impact have been published. 24 excitation cross-sections of the lead atom have been measured with 50 and 150 eV incident electron energies, however only two of these represent excitation of even levels. In a theoretical paper [14] excitation cross-sections are calculated for eight spectral lines with the Born approximation; all these originate from odd upper levels. In addition, a later paper [16] explores excitation of several dozen odd levels in electron collisions with lead atoms and PbCl_2 molecules. Thus, extremely scarce information on excitation cross-sections for even levels of the lead atom has been published so far.

This paper is concerned with an experimental study of excitation cross-sections for even levels of the lead atom in collisions with slow electrons.

2 Main Experimental Conditions

This experiment based on the method of extended crossing beams described in greater detail in a number of past publications (e.g. [17]). We will only note the main conditions pertinent to the lead experiment proper.

Grade S-000 lead was used with the following maximum content of impurities: Cu, Ag, Fe, In, Tl, Cd, Mg – $1 \times 10^{-5}\%$ each; Al, Ca, Na, Bi, Zn, Hg, As, Sb – $5 \times 10^{-5}\%$ each. Lead was placed inside a molybdenum crucible that had its outer surface heated by a electron-ray gun beam to a temperature $T = 1060$ K. Concentration of lead atoms in the area of crossing between the atom beam and the extended electron beam produced by the low-voltage electron cathode gun reached $1.2 \times 10^{11} \text{ cm}^{-3}$. Electron beam current density never exceeded 1.0 mA/cm^2 throughout the entire working range of electron energies $E = 0\text{--}200$ eV. The experimental setup had a spectral resolution of about 0.1 nm within the wavelength range $\lambda = 190\text{--}600$ nm, deteriorating to about 0.2 nm at $\lambda = 600\text{--}850$ nm. As the ground level of the lead atom $6p^2(1/2,1/2)_0$ is separated from its nearest upper level $6p^2(3/2,1/2)_1$ by an energy interval $\Delta E = 7819 \text{ cm}^{-1}$, thermal population of the level $6p^2(3/2,1/2)_1$ was negligible at the vaporization temperature indicated above. Thus, even levels of interest to us were only excited from the ground state i.e. with parity unchanged.

3 Results and Discussion

Forty-eight excitation cross sections of radiative transitions from even levels, including a single blend, have been measured at incident electron energy of 50 eV. Four optical excitation functions (OEFs) have been recorded with electron energies varying in the range $E = 0\text{--}200$ eV.

Findings from measurements supplemented with necessary spectroscopic data are summarized in Tables 1 (transitions for which OEFs have been recorded)

Table 1. Excitation cross-sections of the lead atom even levels (with OEF's recorded)

λ nm	Transition	$J_{\text{low}}-J_{\text{up}}$	E_{low} cm ⁻¹	E_{up} cm ⁻¹	Q_{50} 10 ⁻¹⁸ cm ²	Q_{max} 10 ⁻¹⁸ cm ²	$E(Q_{\text{max}})$ eV	OEF
500.657	$6p7s(1/2,1/2)^{\circ}-6p9p(1/2,3/2)$	0-1	34959	54928	0.45	1.21	9.0	3
508.948	$6p7s(1/2,1/2)^{\circ}-6p9p(1/2,3/2)$	1-2	35287	54930	} 2.43	5.15 {	9.7	4
509.001	$6p7s(1/2,1/2)^{\circ}-6p9p(1/2,3/2)$	1-1	35287	54928			9.0	3
600.186	$6p7s(1/2,1/2)^{\circ}-6p8p(1/2,3/2)$	1-2	35287	51944	4.42	8.87	10	2
605.936	$6p7s(1/2,1/2)^{\circ}-6p8p(1/2,1/2)$	1-0	35287	51786	5.15	16.3	8.5	1

and Table 2 (transitions for which OEFs could not have been recorded enough reliably). Tables 1, 2 indicate wavelengths λ , transitions, internal quantum numbers for lower J_{low} and upper J_{up} levels, energies of lower E_{low} and upper E_{up} levels and excitation cross-section values Q_{50} at incident electron energy of 50 eV. Furthermore, Table 1 also provides cross-section values at OEF maxima, Q_{max} , and identifies positions of OEF maxima, $E(Q_{\text{max}})$. OEFs numbered to match curve numbers in Fig. 1. Shown in parentheses in Table 2 are three lines missing in [18]; they are classified in the present paper using data on PbI levels from [18].

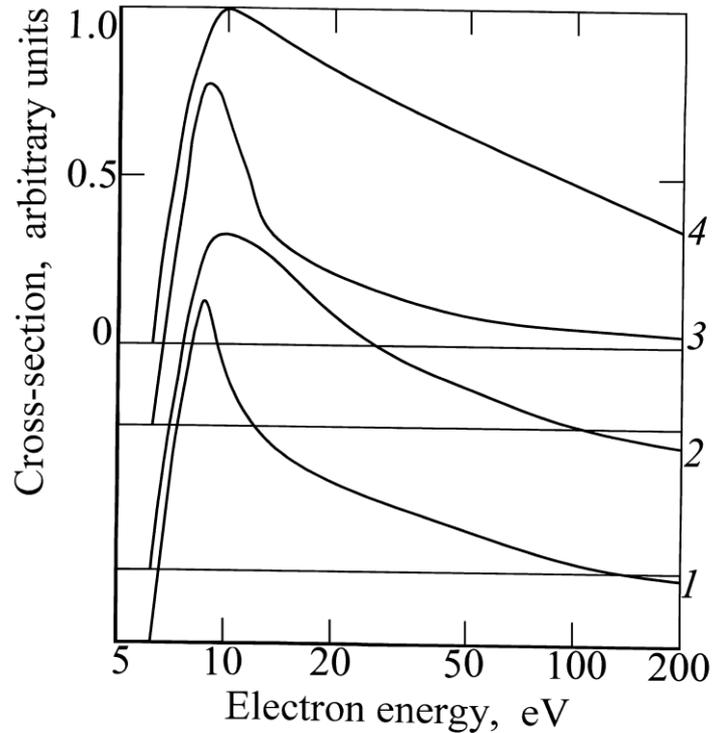


Figure 1: Optical excitation functions of lead atom

Table 2. Excitation cross-sections of the lead atom even levels (without OEF's recorded)

λ nm	Transition	$J_{\text{low}}-J_{\text{up}}$	E_{low} cm ⁻¹	E_{up} cm ⁻¹	Q_{50} 10 ⁻¹⁸ cm ²
270.262	$6p^2(3/2,1/2)-6p7p(1/2,3/2)$	1-2	7819	44809	0.32
(271.248	$6p^2(3/2,1/2)-6p7p(1/2,3/2)$	1-1	7819	44674)	0.12
292.663	$6p^2(3/2,1/2)-6p7p(1/2,3/2)$	2-2	10650	44809	0.10
293.819	$6p^2(3/2,1/2)-6p7p(1/2,3/2)$	2-1	10650	44674	0.10
426.358	$6p7s(1/2,1/2)^\circ-6p13p(1/2,3/2)$	0-1	34959	58407	0.17
(433.122	$6p7s(1/2,1/2)^\circ-6p13p(1/2,3/2)$	1-2	35287	58368)	0.38
439.905	$6p7s(1/2,1/2)^\circ-6p12p(1/2,3/2)$	1-2	35287	58013	0.51
440.199	$6p7s(1/2,1/2)^\circ-6p12p(1/2,3/2)$	1-1	35287	57997	0.32
440.696	$6p7s(1/2,1/2)^\circ-6p12p(1/2,1/2)$	1-0	35287	57972	0.68
448.078	$6p7s(1/2,1/2)^\circ-6p7p(3/2,1/2)$	1-2	35287	57598	0.49
450.109	$6p7s(1/2,1/2)^\circ-6p8f1/2[5/2]$	1-2	35287	57497	0.18
452.492	$6p7s(1/2,1/2)^\circ-6p11p(1/2,1/2)$	1-0	35287	57380	0.85
453.387	$6p7s(1/2,1/2)^\circ-6p7p(3/2,1/2)$	0-1	34959	57009	0.31
(454.968	$6p7s(1/2,1/2)^\circ-6p11p(1/2,3/2)$	1-2	35287	57260)	0.19
460.219	$6p7s(1/2,1/2)^\circ-6p7p(3/2,1/2)$	1-1	35287	57009	0.40
461.817	$6p^2(3/2,1/2)-6p^2(3/2,3/2)$	1-0	7819	29466	48.8*
464.657	$6p7s(1/2,1/2)^\circ-6p10p(1/2,3/2)$	0-1	34959	56475	0.31
466.449	$6p7s(1/2,1/2)^\circ-6p7f1/2[5/2]$	1-2	35287	56719	0.33
471.836	$6p7s(1/2,1/2)^\circ-6p10p(1/2,3/2)$	1-1	35287	56475	0.38
472.000	$6p7s(1/2,1/2)^\circ-6p10p(1/2,3/2)$	1-2	35287	56467	1.02
472.373	$6p7s(1/2,1/2)^\circ-6p10p(1/2,1/2)$	1-0	35287	56451	1.13
474.884	$6p7s(1/2,1/2)^\circ-6p10p(1/2,1/2)$	1-1	35287	56338	0.26
498.046	$6p7s(1/2,1/2)^\circ-6p6f1/2[5/2]$	1-2	35287	55360	0.43
507.635	$6p7s(1/2,1/2)^\circ-6p9p(1/2,1/2)$	0-1	34959	54653	0.52
510.724	$6p7s(1/2,1/2)^\circ-6p9p(1/2,1/2)$	1-0	35287	54861	2.28
516.211	$6p7s(1/2,1/2)^\circ-6p9p(1/2,1/2)$	1-1	35287	54653	0.41
531.302	$6p^2(3/2,1/2)-6p^2(3/2,3/2)$	2-0	10650	29466	12.7*
569.235	$6p7s(1/2,1/2)^\circ-6p5f1/2[5/2]$	1-2	35287	52849	0.56
589.562	$6p7s(1/2,1/2)^\circ-6p8p(1/2,3/2)$	0-1	34959	51916	1.14
601.167	$6p7s(1/2,1/2)^\circ-6p8p(1/2,3/2)$	1-1	35287	51916	1.63
611.052	$6p7s(1/2,1/2)^\circ-6p8p(1/2,1/2)$	0-1	34959	51320	0.56
623.527	$6p7s(1/2,1/2)^\circ-6p8p(1/2,1/2)$	1-1	35287	51320	0.95
733.015	$6p^2(3/2,1/2)-6p^2(3/2,3/2)$	1-2	7819	21457	121.*
(743.833	$6p7d1/2[3/2]^\circ-6p8p^3D$	2-3	52311	65751)	0.22
753.432	$6p6d1/2[5/2]^\circ-6p11f1/2[7/2]$	2-3	45443	58712	0.80
768.537	$6p6d1/2[5/2]^\circ-6p10f1/2[7/2]$	2-3	45443	58451	0.92
790.737	$6p6d1/2[5/2]^\circ-6p9f1/2[7/2]$	2-3	45443	58086	1.08

Table 2. (Continued): Excitation cross-sections of the lead atom even levels (without OEF's recorded)

810.192	$6p6d1/2[3/2]^{\circ}-6p13p(1/2,3/2)$	1-1	46068	58407	0.33
812.249	$6p6d1/2[3/2]^{\circ}-6p13p(1/2,3/2)$	2-2	46060	58368	0.52
815.023	$6p6d1/2[3/2]^{\circ}-6p7p(3/2,3/2)$	2-1	46060	58327	0.37
822.454	$6p6d1/2[5/2]^{\circ}-6p7p(3/2,1/2)$	2-2	45443	57598	0.48
824.605	$6p6d1/2[5/2]^{\circ}-6p10f1/2[7/2]$	3-4	46328	58452	0.87
825.561	$6p6d1/2[5/2]^{\circ}-6p8f1/2[7/2]$	2-3	45443	57552	1.40
829.331	$6p6d1/2[5/2]^{\circ}-6p8f1/2[5/2]$	2-2	45443	57497	0.92
841.897	$6p6d1/2[5/2]^{\circ}-6p11p(1/2,1/2)$	2-1	45443	57317	0.41
846.981	$6p6d1/2[5/2]^{\circ}-6p7p(3/2,3/2)$	3-3	46328	58132	0.63

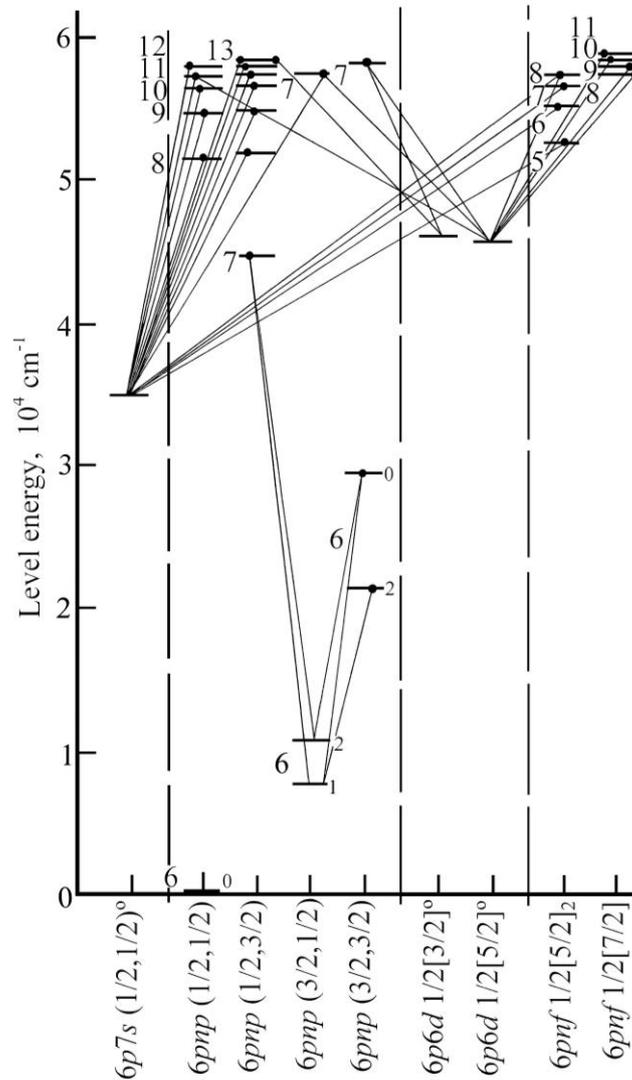


Figure 2: Partial state diagram of PbI

Wavelength data for spectral lines are cited according to [18] as [19, 6, 9] all only indicate positions of studied levels while providing no wavelength data. It should be noted that level energies match each other closely within 1 cm^{-1} for the majority of levels reported in all four papers. Nevertheless findings from [18] and [9] for $6p8p$ as well as $6p12p$ and a number of other levels diverge by $\Delta E = 4\text{--}6 \text{ cm}^{-1}$, with no discussion devoted to such a prominent discrepancy in [9]. Table 3 illustrates the nature of these discrepancies between [18, 19, 6, 9]. Lacking necessary data for detailed analysis, the author of the present paper has decided to use data from [18] exclusively however omitting transitions originating at the $6p13p(1/2,3/2)_2$ level with $E = 58368 \text{ cm}^{-1}$ as it is likely that interpretation of this level in [18] ran into a technical error.

Table 3. Experimental energies of lead atom $J=0$ and 2 even levels

Levels	Energy cm^{-1}					
	[6]	[18]	[19]	[9]	[9a]	[9b]
$6p10p(1/2,1/2)_0$	56450.6	56451.030		56450.77	56451.905	56455.161
$6p10p(1/2,3/2)_2$	56467.5	56467.714		56467.63	56467.69	
$6p7f1/2[5/2]_2$	56719.9	56719.794		57719.794*	56719.78	
$6p11p(1/2,3/2)_2$	57261.4	57260.668		57260.668*	57260.55	
$6p11p(1/2,1/2)_0$	57381.5	57380.876		57381.06	57381.021	57385.520
$6p8f1/2[5/2]_2$	57500.1	57497.795		57497.795*	57499.78	
$6p7p(3/2,1/2)_2$	57600.1	57598.573		57598.65	57599.63	
$6p12p(1/2,1/2)_0$	57972.0	57972.230		57968.05	57972.639	57976.674
$6p12p(1/2,3/2)_2$	58013.3	58013.016		58008.62	58013.13	
$6p9f1/2[5/2]_2$	58095.3		58095.0		58093.27	
$6p13p(1/2,1/2)_0$	58372.2		58372.4	58383.94	58372.429	58375.737
$6p13p(1/2,3/2)_2$	58395.0	58368.912		58397.77	58392.46	
$6p10f1/2[5/2]_2$	58455.6		58455.3		58453.75	
$6p14p(1/2,1/2)_0$	58655.5		58654.7	58654.15	58655.174	58657.834
$6p14p(1/2,3/2)_2$	58667.9		58637.9	58666.67	58662.57	
$6p15p(1/2,3/2)_2$	58849.8		58841.9		58853.96	
$6p15p(1/2,1/2)_0$	58862.7		58862.0	58861.90	58862.475	58864.617

As Tables 1 and 2 indicate, the majority of measured cross-sections are rather little, with just ten Q_{50} values exceeding $1 \times 10^{-18} \text{ cm}^2$. This should be viewed as a natural result as levels studied have identical parities with the initial (ground) level. Fig. 2 shows partial diagram of lead atom states with transitions examined by us. Vertical dashed lines are used to separate states of different parities in the diagram. Common properties of various levels are shown under the horizontal axis as far as possible. Shown in the plot area are values of n to the left of the respective levels. Values lying above $E = 30000 \text{ cm}^{-1}$ are shown without J -splitting which is rather insignificant for most of them. Levels having $E < 30000 \text{ cm}^{-1}$ are shown J -split with J values indicated to the right of levels.

Transitions terminating at $6p^2(3/2,1/2)_{1,2}$ levels are notable. As Fig. 2 shows, these transitions violate the electro-dipole forbiddenness, a parity selection

rule. Radiative lifetimes of $6p^2(3/2,3/2)_{0,2}$ levels – the upper levels for these transitions – have been determined by us using data from [8]. The respective values are $\tau = 9.73$ ms for the level with $J = 0$ and $\tau = 38.2$ ms for the level with $J = 2$. Both τ values are significantly greater than the time it takes for atoms to fly through the collision space in our experimental setup. As a consequence, a sizeable fraction of excited atoms does not exhibit radiation within the collision space; instead, the atoms lose their excitation energy in radiationless collisions with the atom collector.

This situation was examined by us repeatedly in earlier papers (e.g. [15]). A formula was obtained accounting for the departure of excited atoms out of the field of view of the optical system in our setup:

$$\chi = \frac{1}{1 - \frac{v\tau}{L} \left(1 - e^{-\frac{L}{v\tau}}\right)}, \quad (1)$$

where v is mean atom velocity, L is the flight distance by atoms within the field of view of the optical system. χ is the factor by which the actual excitation cross-section is greater than can be inferred from the number of lit atoms. At longer lifetimes the formula (1) is simplified to $\chi \approx 2v\tau/L$. At the conditions of our experiment, the factors are $\chi = 376$ for the level $J = 0$ and $\chi = 1470$ for the $J = 2$. Cross-section values adjusted appropriately are marked with asterisks in Table 2. For even higher lying levels $6p7p(1/2,3/2)_{1,2}$, adjustment for τ becomes unnecessary as forbidden transitions from these levels to $6p^2(3/2,1/2)_{1,2}$ compete with fully allowed transitions to the $6p7s(1/2,1/2)_{0,1}$ levels within the IR part of the spectrum at $\lambda > 1 \mu\text{m}$.

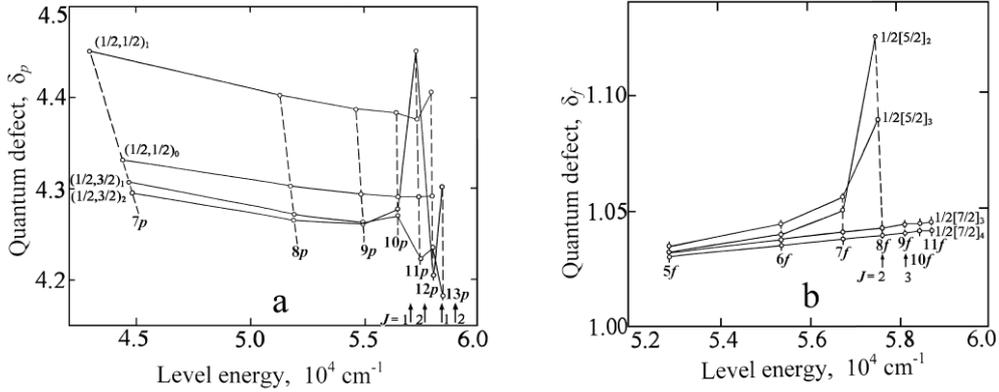


Figure 3: Dependence of quantum defect on the principal quantum number: a – $6pnp$ series; b – $6pnf$ series

Certain levels of majority heavy atoms such as lead display characteristic perturbations, including relatively low-lying levels. Such perturbations are spectroscopically manifest as non-monotonous change in the quantum defect δ as

a function of principal quantum number n . The relationship $\delta = f(n)$ for p and f levels reproduced from [18] is shown in Figs. 3a, b. Positions of perturbing levels are indicated with arrows in the lower part of these plots. Strong excitation is thus apparent for $6pnp(1/2,3/2)_{1,2}$ levels at $n = 10-13$ and for $6pnf1/2[5/2]_{2,3}$ at $n = 8$. In cross-section behavior the perturbation of spectral series lead to several consequences [13], the most prominent of these being the deviation of the function $Q = f(n)$ from the power-law function

$$Q = A_i \times n^{-\alpha_i} \quad , \quad (2)$$

where A_i and α_i are constants having characteristic values for every series. Fig. 4 shows the relationship $Q = f(n)$ for five PbI series. Series having less than four members in Tables 1, 2 as well as series containing blends are excluded from consideration. In a log-log plot, any relationship similar to (2) will appear as a straight line. It can be seen that, with the exception of $6p6d1/2[5/2]_2-6pnf1/2[7/2]_3$ series following the relationship (2), all other series deviate from the power law to some measure. However, as reported in the study of long series [19], the greatest number of perturbations in series under discussion takes place in the area $n > 11$ which is beyond the scope of our study.

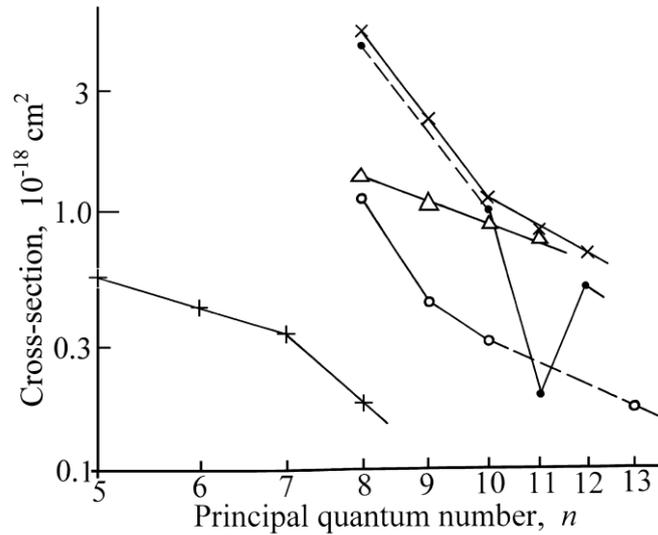


Figure 4: Dependence of cross-section on the principal quantum number for spectral series of PbI: \times $6p7s(1/2,1/2)^\circ_1-6pnp(1/2,1/2)_0$; o $6p7s(1/2,1/2)^\circ_0-6pnp(1/2,3/2)_1$; \bullet $6p7s(1/2,1/2)^\circ_1-6pnp(1/2,3/2)_2$; $+$ $6p7s(1/2,1/2)^\circ_1-6pnf1/2[5/2]_2$; Δ $6p6d1/2[5/2]_2-6pnf1/2[7/2]_3$

4 Conclusion

The study of empirical regularities in the behavior excitation cross-sections is of interest both for electron-atom collision physics and for the atom structure theory.

From a practical standpoint, empirical regularities can be useful as a potential method to obtaining required cross-section values by extrapolation. Unfortunately, the $Q = f(n)$ function cannot be extrapolated correctly for perturbed series. The behavior of the quantum defect as a function of n in the respective range of n values can be considered as criterion for determining whether extrapolation is valid.

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