FIELD THEORETICAL EFFECTS IN X-RAY SPECTRA

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ABSTRACT

The L_I–L_{II} screening doublet separation in the relativistic approximation up to Z^{20} has been compared with that obtained experimentally by the transitions $M_{IV}L_{II}$ ($L\beta_1$) and $M_{IV}L_I$ ($L\ \beta_{10}$). The experimental and theoretical values deviate considerably. If one retains terms up to Z^4, an apparently close fit with the experimental values is obtained. It is shown that such a fit disappears on applying the field theoretical corrections and this stresses the need of retaining the terms up to Z^{20}. The justification of neglecting the still higher powers of Z lies in the smallness of their magnitude when compared to the experimental errors. The field theoretical corrections modify the relativistic curve that contains terms up to Z^{20} in such a way that one again gets a good fit with the experimental values. This shows how the various field theoretical corrections and the contributions from relativistic terms with powers of Z greater than four compensate each other.

INTRODUCTION

THE possibility of Lamb shift in the X-ray L_I and L_{II} levels was pointed out by Bethe (1947) who stressed the necessity of performing an exact calculation of the screening constants. Several authors (*e.g.*, Shacklett and DuMond, 1957 and Shacklett, 1958 and Lindenmeier, 1960) have given an estimate of the effect for the spin-relativity doublet $L_{II}L_{III}$ but not for the screening doublet L_IL_{II}. Since L_{II} and L_{III} both are *p*-levels the sign of Lamb shift for both will be the same. However, for L_I and L_{II} levels the shift will be in opposite directions, the former being the *s*-level. Thus the theoretical splitting $\triangle L_IL_{II}$ given by the uncorrected relativistic formula alone should show deviations from the experimental value. Further, apart from the Lamb shift one has to take into account other field theoretical effects as listed by Novick, Lipworth and Yergin (1955). All these effects have been calculated for elements Z = 50 to 92. When the total correction due to all the effects is applied to the theoretical values obtained from the relativistic formula calculated to the necessary power of Z, a curve is obtained that runs closely parallel to the experimental curve.

Theoretical and the Experimental $L_I L_{II}$ Splitting

The X-ray levels arise out of configurations lacking one electron for completion (L_I from $2s^1$, $L_{II}L_{III}$ from $2p^5$, etc., etc.). The spectroscopic states are therefore the same as those due to an one-electron system with the same value of Z, but are inverted. It is, therefore, not very much unjustified to use the one-electron formula for an estimate of the L_I and L_{II} levels. In fact Shacklett has done the same for the L_{II} and L_{III} levels while estimating the effect of the nuclear size on the splitting. We have estimated L_I and L_{II} levels separately using the usual screening constants. The relativistic formula has to be expanded up to Z^{20} in order to give the required accuracy to the second place of decimal in ν/R units for a comparison with the experimental values. This was done by the standard method due to Sommerfeld (1934) and the various terms are shown in Table I. Individual contributions to $\triangle L_I L_{II}$ from terms with various powers of Z are shown in Figs. 1 *a* and 1 *b* as functions of Z. Fig. 1 *a* compares the terms from Z^2 to Z^{10} and Fig. 1*b* shows the relative magnitudes of the terms from Z^{10} to Z^{20}. The Z^{20}-term for uranium is only 0·007 ν/R units. Since the maximum experimental accuracy in the available data is only 0·01 ν/R, one need not calculate the higher powers of Z for the present. Figure 2 shows how the $L_I L_{II}$ separation varies when the terms with various powers of Z are included. The curve (*a*) shows the variation of Z^2 term only, (*b*) shows the resulting curve when Z^4-term is included while (*c*) results from the addition of the Z^6-term, (*d*) from Z^8-term and finally (*f*) from the terms up to Z^{20}. Curve (E) is the experimental curve. The vertical lines represent limits of error in the experimental evaluation of $\triangle L_I L_{II}$ which is calculated as a difference in the ν/R values of the lines $L\beta_1$ (M_{IV}–L_{II}) and $L\beta_{10}$ (M_{IV}–L_I). Data were taken from Sandstrom's tables (Sandstrom, 1957) and the corresponding errors were evaluated by reference to original literature. Limits of errors for those elements for which the authors do not mention probable errors could not be plotted. A glance at Fig. 2 shows the considerable deviation of the experimental curve (E) from the theoretical curve (*f*). The deviation increases regularly with Z. It is interesting to note that the curve (*b*) runs closer to the experimental curve than other uncorrected curves. This shows that apparently one could retain terms only up to Z^4 in order to get the closest fit with the experimental curve within the latter's limits of errors. If, however, we now introduce the various field theoretical effects this apparent agreement

TABLE I

Contributions from the terms with various powers of Z to $\triangle L_I L_{II}$

(All values are in ν/R units)

Element	Z^2–term	Z^4–term	Z^6–term	Z^8–term	Z^{10}–term	Z^{12}–term	Z^{14}–term	Z^{16}–term	Z^{18}–term	Z^{20}–term	Total	Experimental values	Difference Cal.–Expt.
50	19·6251	2·6340	0·2468	0·0249	0·0026	0·0003	..	..	..	..	22·5337	22·737	−0·203
51	20·1153	2·8047	0·2740	0·0290	0·0032	0·0004	..	..	..	..	23·2262	23·429	−0·202
52	20·5998	2·9827	0·3036	0·0335	0·0039	0·0004	..	..	..	..	23·9240	24·084	−0·160
53	21·0900	3·1682	0·3357	0·0385	0·0046	0·0006	..	..	..	..	24·6376	24·726	−0·088
54	21·5802	3·3610	0·3704	0·0440	0·0055	0·0006	..	..	..	..	25·3617	..	..
55	22·0704	3·5616	0·4080	0·0506	0·0065	0·0009	..	..	..	..	26·0980	..	..
56	22·5549	3·7701	0·4485	0·0576	0·0077	0·0011	..	..	..	..	26·8399	27·024	−0·184
57	23·0451	3·9863	0·4922	0·0657	0·0091	0·0015	0·0001	..	..	..	27·6000	27·441	0·159
58	23·5353	4·2067	0·5389	0·0747	0·0108	0·0016	0·0001	..	..	..	28·3681	28·27	0·098
59	24·0198	4·4438	0·5899	0·0849	0·0126	0·0025	0·0001	..	..	..	29·1536	29·11	0·044
60	24·5100	4·6850	0·6442	0·0958	0·0148	0·0025	0·0001	..	..	..	29·9522	29·85	0·102
61	25·0002	4·9347	0·7025	0·1083	0·0173	0·0029	0·0001	..	..	..	30·7659	..	..
62	25·4904	5·1932	0·7649	0·1218	0·0202	0·0034	0·0001	..	..	..	31·5939	31·33	0·264
63	25·9749	5·4604	0·8316	0·1366	0·0235	0·0041	0·0001	..	..	..	32·4312	31·97	0·461
64	26·4651	5·7356	0·9029	0·1531	0·0271	0·0050	0·0001	..	..	..	33·2888	32·93	0·359
65	26·9553	6·0227	0·9791	0·1709	0·0313	0·0058	0·0002	..	..	..	34·1651	33·96	0·205
66	27·4398	6·3166	1·0602	0·1932	0·0364	0·0071	0·0003	..	..	..	35·0536	34·28	0·774
67	27·9300	6·6218	1·1467	0·2140	0·0415	0·0083	0·0003	..	..	..	35·9626	35·39	0·573
68	28·4202	6·9357	1·2374	0·2370	0·0476	0·0098	0·0004	0·0001	..	..	36·8882	35·948	0·940
69	28·9104	7·2596	1·3390	0·2670	0·0554	0·0118	0·0005	0·0002	..	..	37·8439	37·01	0·834
70	29·3949	7·5935	1·4407	0·2960	0·0631	0·0138	0·0007	0·0002	..	..	38·8029	37·43	1·333
71	29·8851	7·9370	1·5509	0·3260	0·0716	0·0161	0·0009	0·0002	..	..	39·7878	38·615	1·173
72	30·3753	8·2911	1·6679	0·3620	0·0820	0·0190	0·0010	0·0003	..	..	40·7986	38·88	1·919
73	30·8598	8·6556	1·7909	0·4020	0·0938	0·0223	0·0013	0·0003	0·0001	..	41·8261	40·341	1·485
74	31·3500	9·0306	1·9231	0·4410	0·1055	0·0259	0·0016	0·0004	0·0001	..	42·8782	40·922	1·956
75	31·8402	9·4157	2·0619	0·4860	0·1199	0·0302	0·0021	0·0005	0·0001	..	43·9566	41·807	2·150
76	32·3304	9·8125	2·2084	0·5370	0·1360	0·0352	0·0025	0·0007	0·0002	..	45·0629	42·898	2·165
77	32·8149	10·2195	2·3634	0·5930	0·1543	0·0411	0·0030	0·0009	0·0003	..	46·1904	43·657	2·533
78	33·3051	10·6382	2·5269	0·6440	0·1723	0·0471	0·0037	0·0011	0·0003	..	47·3387	44·529	2·810
79	33·7953	11·0677	2·6992	0·7160	0·1961	0·0551	0·0046	0·0013	0·0003	..	48·5356	45·583	2·953
80	34·2798	11·5092	2·8811	0·7780	0·2190	0·0631	0·0185	0·0055	0·0017	0·0005	49·7564	46·381	3·315
81	34·7700	11·9618	3·0723	0·8500	0·2450	0·0727	0·0219	0·0066	0·0020	0·0006	51 0029	47·678	3·325
82	35·2602	12·4262	3·2737	0·9290	0·2770	0·0836	0·0258	0·0081	0·0025	0·0007	52·2868	48·57	3·717
83	35·7504	12·9025	3·4855	1·0150	0·3090	0·0960	0·0304	0·0097	0·0031	0·0009	53·6015	49·59	4·012
84	36·2349	13·3910	3·7082	1·1070	0·3440	0·1100	0·0357	0·0117	0·0039	0·0012	54·9476	..	..
85	36·7251	13·8913	3·9419	1·2060	0·3850	0·1250	0·0419	0·0141	0·0048	0·0015	56·3375	..	..
86	37·2153	14·4041	4·1874	1·3120	0·4290	0·1438	0·0490	0·0168	0·0059	0·0019	57·7652	..	..
87	37·6998	14·9294	4·4451	1·4303	0·4770	0·1635	0·0570	0·0201	0·0072	0·0024	59·2318	..	..
88	38·1900	15·4674	4·7152	1·5538	0·5300	0·1860	0·0664	0·0240	0·0087	0·0030	60·7445	55·4	5·345
89	38·6802	16·0183	4·9982	1·6860	0·5870	0·2110	0·0771	0·0284	0·0106	0·0038	62·3006	..	..
90	39·1704	16·5816	5·2947	1·8276	0·6600	0·2420	0·0907	0·0343	0·0130	0·0047	63·9190	57·1	6·819
91	39·6549	17·1583	5·6052	1·9793	0·7290	0·2750	0·1049	0·0406	0·0159	0·0058	65·5689	58·2	7·369
92	40·1451	17·7483	5·9300	2·1418	0·8040	9·3080	0·1210	0·0478	0·0190	0·0073	67·2723	59·5	7·772

disappears and only the theoretical curve (f) properly corrected for these effects gives a satisfactory fit with the experimental curve.

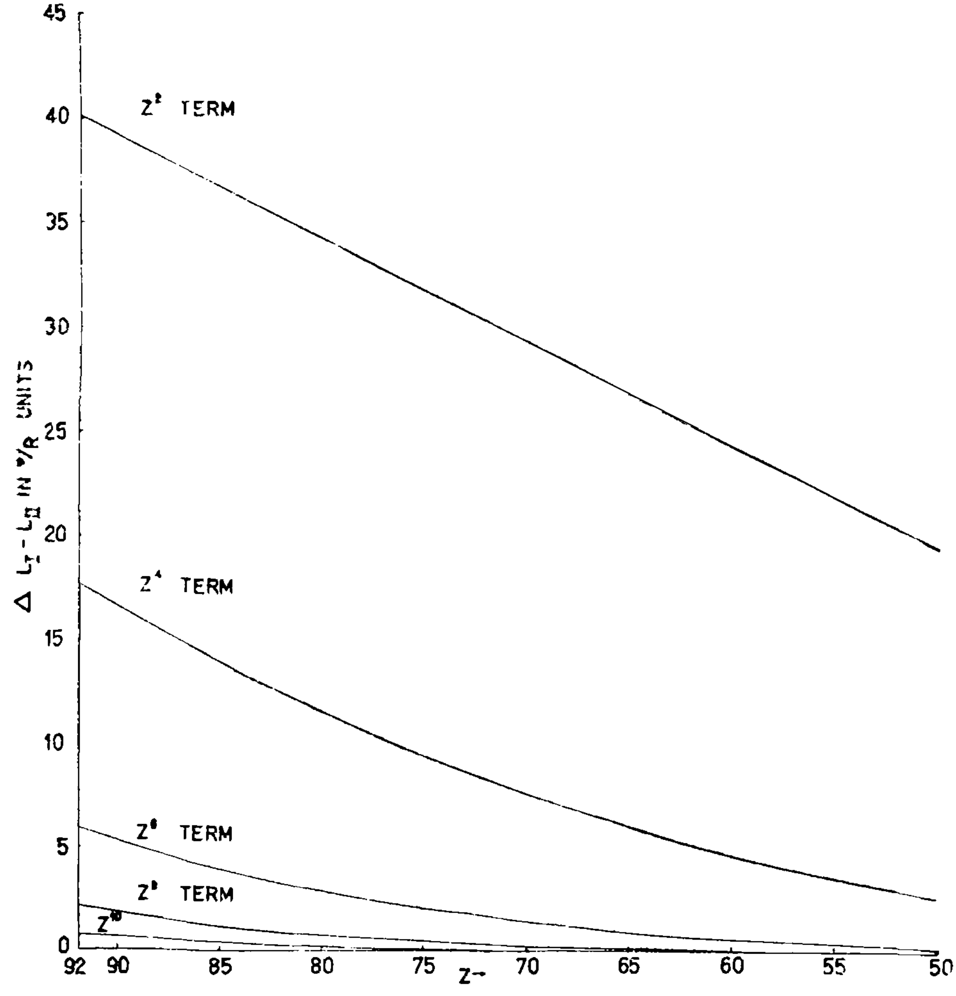

FIG. 1 (*a*). Variation of the magnitude of terms with Various powers of Z as a function of Z. The range Z^2 to Z^{10}.

Field-theoretical Corrections to the Relativistic L_I–L_{II} Splitting

Novick *et al.* have listed the following field theoretical corrections to the relativistic separation $2S_{\frac{1}{2}}-2P_{\frac{1}{2}}$ in megacycles per second:

(1) The Lamb shift.

$$L\left(1-\frac{3m}{M}\right)\left(2ln\frac{1}{a}+\frac{m}{M}-2lnZ+\frac{11}{24}-ln\frac{16\cdot 646}{0\cdot 9704}\right)Z^4;$$

(2) Second-order vacuum polarisation

$$L\left(1-\frac{3m}{M}\right)\left(-\frac{1}{5}\right)Z^4;$$

(3) Second-order magnetic moment

$$L\left(1-2\cdot75\frac{m}{M}\right)\left(\frac{1}{2}\right)Z^4;$$

(4) Second-order relativistic shift

$$L\left(1-\frac{3m}{M}\right)\left(3\pi\alpha\right)\left(1+\frac{11}{128}-\frac{1}{2}\,ln\,2+\frac{5}{192}\right)Z^6;$$

(5) Fourth-order radiative shift

$$L\left(\frac{3\alpha}{2\pi}\right)\left(0\cdot52\right)Z^4;$$

(6) Fourth-order vacuum polarisation

$$L\left(-\frac{41\alpha}{54\pi}\right)Z^4;$$

(7) Fourth-order magnetic moment

$$L\left(-2\cdot973\frac{\alpha}{\pi}\right)Z^4;$$

(8) Finite mass effect

$$L\left(\frac{m}{M}\right)\left(5\cdot3684-\frac{1}{2}\,ln\,Z\right)Z^5; \text{ and}$$

(9) Finite size effect

$$\frac{1}{24}\,\pi\,\frac{e^2}{a\hbar}\left(\frac{a_n}{a_0}\right)^2 Z^4,$$

where, L is the Lamb constant $= \alpha^3 R_\infty c/3\pi$.

All the above effects but the last one (size effect) were calculated for elements $Z = 50$ to 92 and are shown in Table II. The reason for omitting the last term is that the X-ray spectra obtained are not from the isotopically separated elements. Since X-ray levels are inverted, and are positive with respect to the corresponding optical levels, the corrections are changed in sign before they are applied to the theoretical curve (f).

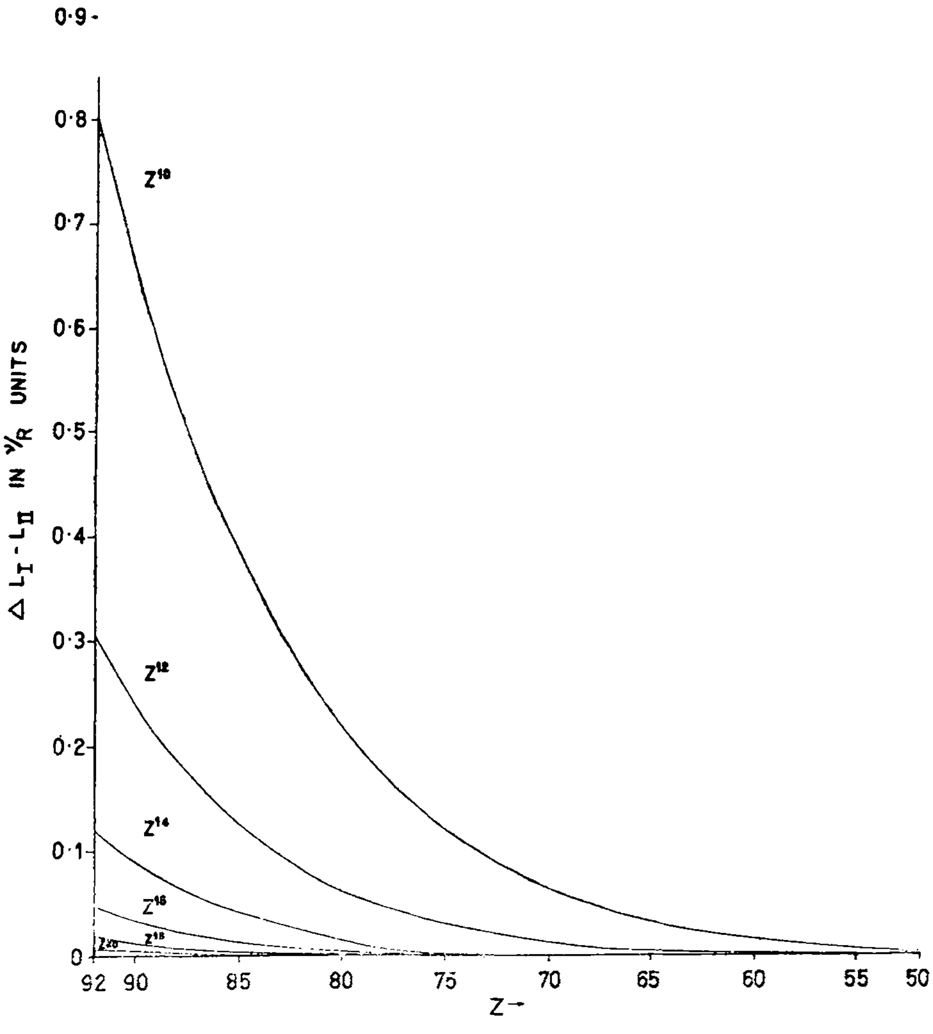

FIG. 1 (*b*). Variation of the magnitude of terms with various powers of Z as a function of Z. The range: Z^{10} to Z^{20}.

Curves obtained from (f) after applying the various corrections have been drawn in dashed lines and numbered numerically in Fig. 2. The largest magnitude of correction comes from the second-order relativistic shift. The curve (f) falls to the dashed curve (1). The sign of the correction due to the second-order magnetic moment is also the same as above, but its magnitude is smaller. Curve (1) now falls down to curve (2). On applying the next correction which is the Lamb shift, curve (2) is raised to (3) and finally we

TABLE II

Field theoretical corrections to $2P_{\frac{1}{2}}$–$2S_{\frac{1}{2}}$ splitting in ν/R units

(All signs are reversed for X-Ray levels)

Z	Individual Corrections (σ=3·5)								Total corrections (−) with σ		
	2nd order relativistic shift (−)	2nd order mag. mom. (−)	Lamb shift (+)	2nd order vac. pol. (+)	4th order mag. mom. (+)	4th order rad. shift (−)	4th order vac. pol. (+)	Finite mass corr. (−)	=3·5,	=2·0,	=0
50	0·472	0·096	0·071	0·039	0·0013	0·0003	0·0003	0·0001	0·458	0·538	0·659
51	0·525	0·105	0·085	0·042	0·0014	0·0004	0·0004	0·0002	0·501	0·588	0·717
52	0·582	0·114	0·101	0·046	0·0016	0·0004	0·0004	0·0002	0·548	0·659	0·779
53	0·645	0·124	0·119	0·049	0·0017	0·0004	0·0004	0·0002	0·598	0·717	0·846
54	0·713	0·134	0·139	0·054	0·0018	0·0005	0·0005	0·0002	0·652	0·779	0·917
55	0·786	0·145	0·161	0·058	0·0020	0·0005	0·0005	0·0002	0·710	0·846	0·993
56	0·865	0·157	0·186	0·063	0·0022	0·0006	0·0006	0·0002	0·772	0·917	1·074
57	0·951	0·169	0·212	0·068	0·0023	0·0006	0·0006	0·0002	0·838	0·993	1·160
58	1·043	0·182	0·241	0·073	0·0025	0·0007	0·0006	0·0003	0·909	1·074	1·251
59	1·142	0·195	0·272	0·078	0·0027	0·0007	0·0007	0·0003	0·985	1·160	1·350
60	1·249	0·210	0·307	0·084	0·0029	0·0008	0·0007	0·0003	1·066	1·251	1·454
61	1·364	0·225	0·344	0·090	0·0031	0·0008	0·0008	0·0003	1·152	1·350	1·564
62	1·486	0·241	0·384	0·097	0·0033	0·0009	0·0009	0·0003	1·244	1·454	1·682
63	1·618	0·258	6·428	0·103	0·0036	0·0009	0·0009	0·0004	1·342	1·564	1·806
64	1·758	0·276	0·474	0·110	0·0038	0·0010	0·0010	0·0004	1·446	1·682	1·938
65	1·909	0·295	0·525	0·118	0·0041	0·0011	0·0010	0·0004	1·557	1·806	2·078
66	2·069	0·314	0·579	0·126	0·0043	0·0011	0·0011	0·0004	1·675	1·938	2·226
67	2·240	0·335	0·637	0·134	0·0046	0·0012	0·0012	0·0005	1·709	2·078	2·383
68	2·422	0·357	0·699	0·143	0·0049	0·0013	0·0013	0·0005	1·932	2·226	2·549
69	2·615	0·379	0·766	0·152	0·0052	0·0014	0·0013	0·0905	2·072	2·383	2·724
70	2·821	0·403	0·837	0·161	0·0056	0·0015	0·0014	0·0005	2·221	2·549	2·909
71	3·040	0·428	0·913	0·171	0·0059	0·0015	0·0015	0·0006	2·379	2·724	3·106
72	3·272	0·454	0·993	0·181	0·0062	0·0016	0·0016	0·0006	2·545	2·909	3·311
73	3·518	0·481	1·079	0·192	0·0066	0·0017	0·0017	0·0007	2·721	3·106	3·528
74	3·778	0·509	1·170	0·204	0·0070	0·0018	0·0018	0·0007	2·907	3·311	3·756
75	4·054	0·538	1·267	0·215	0·0074	0·0020	0·0019	0·0007	3·103	3·528	3·997
76	4·345	0·569	1·369	0·228	0·0079	0·0021	0·0020	0·0008	3·310	3·756	4·251
77	4·654	0·601	1·478	0·240	0·0083	0·0022	0·0021	0·0008	3·529	3·997	4·517
78	4·979	0·635	1·593	0·254	9·0088	0·0023	0·0022	0·0008	3·759	4·251	4·798
79	5·322	0·669	1·714	0·268	0·0092	0·0024	0·0024	0·0009	4·001	4·517	5·092
80	5·684	0·706	1·842	0·282	0·0097	0·0026	0·0025	0·0009	4·256	4·798	5·403
81	6·065	0·743	1·978	0·297	0·0102	0·0027	0·0026	0·0010	4·524	5·092	5·726
82	6·467	0·782	2·120	0·313	0·0108	0·0028	0·0028	0·0010	4·807	5·403	6·066
83	6·889	0·823	2·270	0·329	0·0114	0·0030	0·0029	0·0011	5·103	5·726	6·423
84	7·334	0·864	2·428	0·346	0·0120	0·0031	0·0031	0·0011	5·413	6·066	6·798
85	7·801	0·909	2·594	0·364	0·0126	0·0033	0·0032	0·0012	5·741	6·423	7·189
86	8·291	0·954	2·768	0·382	0·0132	0·0035	0·0034	0·0012	6·084	6·798	7·599
87	8·806	1·001	2·951	0·401	0·0138	0·0036	0·0035	0·0013	6·444	7·189	8·029
88	9·346	1·050	3·143	0·420	0·0145	0·0038	0·0037	0·0013	6·820	7·599	8·348
89	9·912	1·101	3·344	0·440	0·0152	0·0040	0·0039	0·0014	7·215	8·029	8·947
90	10·506	1·153	3·555	0·461	0·0159	0·0042	0·0041	0·0015	7·629	8·348	9·439
91	11·127	1·208	3·775	0·483	0·0167	0·0044	0·0043	0·0016	8·061	8·947	9·951
92	11·778	1·264	4·006	0·505	0·0175	0·0046	0·0045	0·0016	8·514	9·439	10·487

obtain curve (4) by applying corrections due to the remaining effects; fourth-order magnetic moment, second-order vacuum polarisation, etc. The magnitude of each of these remaining effects is too small to be separately represented in the figure. Thus our theoretical curve (*f*) is now corrected by various field theoretical effects to give the curve (4) which is running closely

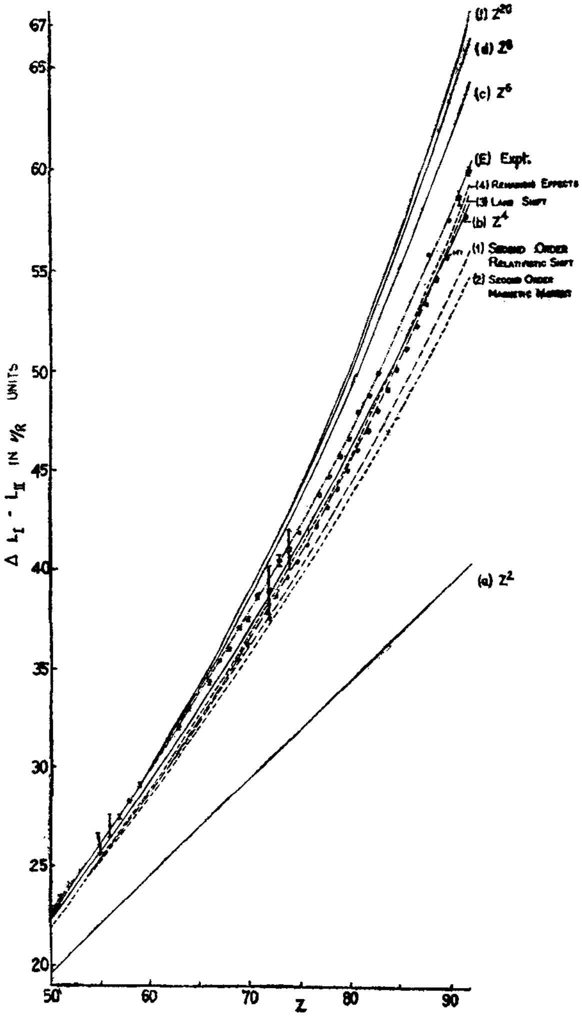

FIG. 2. Variations of $\Delta\, L_I L_{II}$ as a function of Z in successive approximations of the relativistic formula have been shown as curves (*a*), (*b*), (*c*), (*d*) and (*f*). Curve (E) is the experimental curve. Curves labelled numerically as (1), (2), (3) and (4) are obtained as the field theoretical corrections are applied successively. All these corrections have been calculated with $\sigma = 3 \cdot 5$. Curve (4′) is obtained with $\sigma = 2 \cdot 0$.

parallel to the experimental curve (E) and is still more close to the Z^4 curve (*b*). In case we apply the field theoretical corrections to the Z^4 curve (*b*) it is clear that the new curve will lie much below the experimental curve. This stresses the necessity of retaining contributions from terms with powers of Z higher than four. Also it may be remarked that the extremities of the errors in the experimental curve do not cross the new curve, *i.e.*, (4) everywhere and this might indicate the existence of a small discrepancy. However, the experimental data are not accurate enough for an estimation of this discrepancy to be made. To a rough order of magnitude this discrepancy is nearly 0·7 ν/R units or 9·5 eV. At this stage we wish to emphasize the importance of the screening constants in the calculation of the above corrections. On varying the value of screening constant σ by writing $(Z - \sigma)$ in place of Z in all the above field theoretical corrections the distance between the curve (4) and (E) is markedly affected. For $\sigma = 0$, this distance is greatest and as σ is given finite values the two curves get closer. Curve (4) has been drawn with $\sigma = 3 \cdot 5$, the value for the L_{II} level. In fact, the value of σ for both the levels should have been included but the formulae listed by Novick *et al.* represent directly the correction to the $L_I L_{II}$ separation rather than to the individual levels. If σ is put equal to 2·0 (the value for L_I level) we get the curve (4′) indicated by unjoined circles. The correct curve should involve appropriate weights attached to the two σ values and will lie somewhere between (4) and (4′). The very fact that the curve (4) and (E) run quite parallel to each other shows that their functional behaviour is the same. The closeness of the curve (*b*) Z^4 and (E) shows how the contributions from powers of Z higher than four compensate with the field theoretical corrections. In this connection we wish to mention the work of Bethe* and Cohen.* They have made self-consistent field calculations for Hg (80) and estimated the $\Delta L_I L_{II}$ value correct up to ± 3 ν/R and found apparently a fairly good agreement with the experimental values leaving no room for the field theoretical effects! Our analysis of the relativistic formula shows that such calculations are correct equivalently only up to Z^4 since the contribution for Hg (80) from Z^6-term is less than 3 ν/R (only 2·88) and the deviation of the experimental value from Z^6 is 2 ν/R and from Z^{20} it is 3 ν/R units. This clearly shows that in order to realise the necessity of the field theoretical effects, a self-consistent field calculation yielding energies correct up to 0·01 ν/R is necessary.

Finally, a few words about the isotopic shift. Wertheim and Igo (1955) have calculated the isotopic shifts in $1S_{\frac{1}{2}}$ level for Mo, $2S_{\frac{1}{2}}$ and $2P_{\frac{1}{2}}$

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* Private communications with Prof. H. Bethe and Dr. S. Cohen.

levels of uranium and the values are 0·084, 0·4 and 0·018 eV respectively. Since the maximum experimental accuracy so far for uranium $\triangle L_I L_{II}$ is nearly ± 0·12 ν/R, *i.e.*, nearly ± 1·6 eV; the question of observing such shifts does not arise at the moment. In a few recent publications (Merrill and DuMond, 1961), it has been suggested that for a transuranic element ^{237}Np there exists an experimental evidence for the nuclear hyperfine structure in the widths of some L-lines. However, the position of the levels and $\triangle L_I L_{II}$ appear to remain unaffected.

While concluding this paper we wish to make it clear that the limits of the agreement of the curve (4) with the experimental curve (E) are extremely sensitive to the screening constants. Thus the proposition of Bethe (1947) for evaluating accurately the screening constants has even today its original importance.

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