Correlation and relativistic effects on the X-ray spectra of highly charged few electron ions

L.NATARAJAN and A.NATARAJAN

DEPARTMENT OF PHYSICS,

UNIVERISTY OF MUMBAI,

MUMBAI

Outline

1. Low lying levels of some selected configurations in Li-like ions that pose problems in unique identification of quantum states

   - entanglement of angular momentum coupled levels

2. Effects of relaxed and frozen spin orbitals of the initial and final states on the correlation sensitive radiative rates

3. Investigate the effects of relativity and QED on the radiative decay rates

THEORETICAL BACKGROUND

Single configuration Dirac-Fock model (relativistic SCF method)

Dirac- Fock -Coulomb equation is

$$\hat{H}_{DC}\left|\Psi\right\rangle = \left[\sum_{i=1}^{N}\hat{h}_D^i + \sum_{i=1}^{N-1}\sum_{j=i+1}^{N}\frac{1}{\vec{r}_{ij}}\right]\left|\Psi\right\rangle \qquad (1)$$

$\hat{h}_D^i$ - Dirac one –electron Hamiltonian

$$\hat{H}_D = c\hat{\alpha}.\hat{p} + (\beta - 1)c^2 + V_{nuc}(\bar{r})$$

Ψ - one-electron four-component Dirac spin- orbitals

- α is a 4x 4 vector matrix and β is a 4x4 scalar matrix

Limitations of single configuration Dirac - Fock model

- Ignores many body effects

  → e-e correlation

  → can not explain transitions due exclusively to correlation like two-electron one-photon transitions.

  (to be taken up in this talk)

* In DF, potential is non-relativistic

Combining many body effects , relativistic potential and quantum electrodynamics

Many body effects

Electron Correlation

$$E_{DFC}^{Corr} = E_{DFC}^{Exact} - E_{DFC}$$

Involves many body effects

Lindgren " Relativistic many body theory", Springer –Verlag, 2010

Evaluation of correlation functions :

Multi Configuration Dirac Fock -MCDF model

- Generation of additional configurations to the DF wave functions
- Construct ϕ (ΓJP) , configuration state functions (CSFs)
  - linear combination of Slater determinants of the Dirac orbitals belonging to a certain J, M and parity (same *n* and l)

- Construct atomic state functions (ASF)
  - linear combination of CSFs

$$\Psi_i(J^P) = \sum_{\alpha=i}^{n_{csf}} c_{i\alpha}\phi(\Gamma_\alpha J^P)$$

c's are mixing coefficients

K.G. Dyall et al Comput. Phys. Commun.55,425, 1989

P.Jonsson, X.He, C.F. Fischer and I.P.Grant, Comput. Phys. Commun. 177, 597, 2007 .

P.Jonsson et al. Comput. Phys. Commun,184,2197,2013

Relativistic corrections to Potential

Using optimized correlated functions in RCI calculations

1. Breit interaction – transverse component of e^- e^- interaction
   → consists of magnetic interaction and retardation

$$H_{Breit} = -\sum_{i<j}^{N} [\frac{\hat{\alpha}_i . \ddot{\alpha}_j \cos(\omega_{ij} r_{ij})}{r_{ij}} + (\hat{\alpha}_i . \overline{\nabla}_i)(\hat{\alpha}_j . \overline{\nabla}_j) \frac{\cos(\omega_{ij} r_{ij}) - 1}{\omega_{ij}^2 r_{ij}}]$$

P.Jonsson etal Comput. Phys. Commun. 177,597,2007

Effects beyond Dirac-Coulomb- Breit referred to as QED effects include vacuum polarization and self energy corrections

2 QED effects:

Comprises of self energy and vacuum polarization

Self energy – shift in the binding energy of an electron due to exchange of virtual photons with the nucleus.

Self energy in a hydrogenic system with a rough estimate of electron screening using

$$E_{SE} = \frac{\alpha (Z\alpha)^4}{\Pi n^3} F(Z\alpha)$$

Vacuum polarization –Shift in binding energy of an electron due to creation and annihilation of virtual e^+e^-pair in vacuum

included as a perturbation correction

Fully relativistic treatment (Relativistic configuration Interaction) includes

1.Relativistic wavefunctions include spin-orbit interaction

=> **Varies as $(Z\alpha)^2$**

2. Relativistic potential considers Breit interaction

=> **Varies as $(Z\alpha)^3$**

3. Self energy and vacuum polarization- second quantization

=> **Varies as $(Z\alpha)^4$**

4. Finite nuclear size effects -Extended Nucleus with Fermi Charge distribution

Revisiting energy level diagrams for Li-like ions through correlation and higher order corrections

* 1s2s3p and 1s2p3s configurations
* Enhanced configuration mixing leads to level crossing and entanglement of quantum states
* Difficulty in unique identification of levels
* Leads to irregularities and sharp discontinuity in the X-ray rates

I. Level structure of 1s2s3p states of ions with Z=38 to 43

1s2s3p

Mono-configuration

* 3x3 sub matrix from the 3 J = 1/2 , 3x3 sub matrix from the 3 J = 3/2 and 1x1 submatrix from the single J =5/2 states

* Concentrating on J=1/2 states, in jj coupling

$1s_{1/2}\ 2s_{1/2}\ [J'=0]\ 3p_{1/2}\ [J=1/2]$

$1s_{1/2}\ 2s_{1/2}\ [J'=1]\ 3p_{1/2}\ [J=1/2]$

$1s_{1/2}\ 2s_{1/2}\ [J'=1]\ 3p_{3/2}\ [J=1/2]$

* In LSJ coupling, these 3 J=1/2 states are

$1s2s[^1S_0]3p_{1/2}$ $[^2P_{1/2}]$,

$1s2s[^3S_1]3p_{3/2}$ $[^2P_{1/2}]$

$1s2s[^3S_1]3p_{1/2}$ $[^4P_{1/2}]$

valid for high/low Z

* Identify the eigen states with the dominant configuration mixing coefficients

  - in terms of J, parity and energy ordering

  (LSJ and jj coupling schemes)

* In intermediate coupling, additional quantum numbers – seniority number and coefficient of fractional parentage are used.

Multi-configuration -correlation

Inclusion of correlation functions from 1s2p3s leads to 6 eigen states with J =1/2.

* In case of strong mixing unambiguous identification even with energy ordering

* Major contributions to the J=1/2 states of $1s2s(^1S_0)3p_{1/2}$, $1s2s(^3S_1)3p_{1/2}$ for all ions nearly the same.

Breakdown of dominant contributions to $1s2s(^3S_1)3p_{3/2}$ $^2P_{1/2}$ state for mid Z ions

* For ions with Z=38 to 43, major contribution to $1s2s(^3S_1)3p_{3/2}$ comes from

1)$1s2p_{1/2}(^3P_0)3s$ (J=1/2) or from

2) $1s2p_{1/2}(^3P_1)3s$ (J=1/2)

* $1s2s(^3S_1)3p_{3/2}$ becomes the second major contributor in both the cases . This gives rise to two states with the same LSJ notation with marginal energy difference .

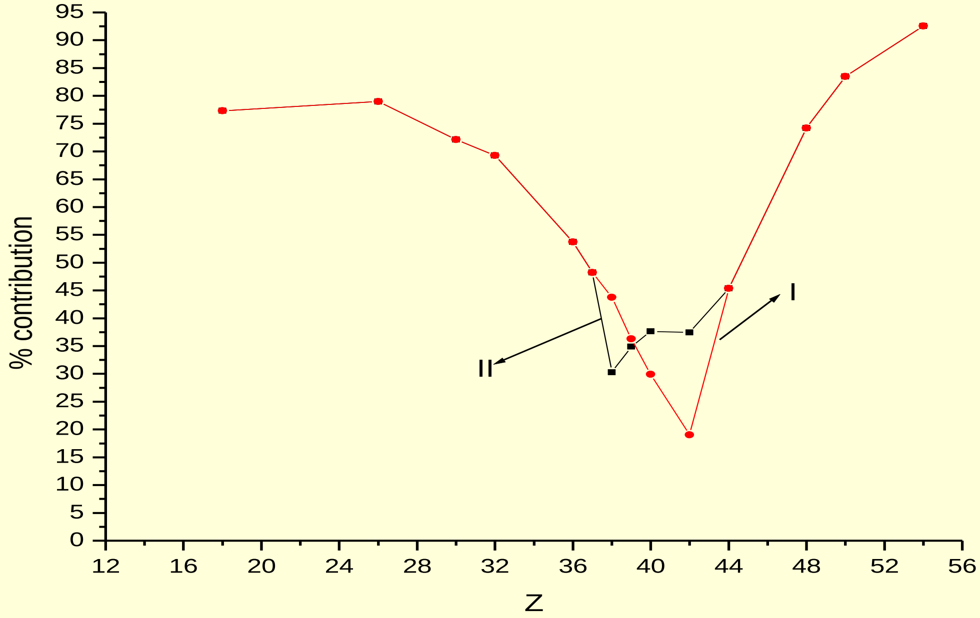
Z	16 CSFs	7353 CSFs.	with Breit+QED .	
36	$(^1S_0)\,^2P_{3/2}$	$(^1S_0)\,^2P_{3/2}$	$(^1S_0)\,^2P_{3/2}$	
	$(^1S_0)\,^2P_{1/2}$	$(^1S_0)\,^2P_{1/2}$	$(^1S_0)\,^2P_{1/2}$	
	$(^3S_1)\,^2P_{1/2}$	$(^3S_1)\,^2P_{3/2}$	$(^3S_1)\,^2P_{1/2}$	
	$(^3S_1)\,^2P_{3/2}$	$^4P_{5/2}$	$(^3S_1)\,^2P_{3/2}$	
	$^4P_{5/2}$	$(^3S_1)\,^2P_{1/2}$	$^4P_{5/2}$	
	$^4P_{3/2}$	$^4P_{3/2}$	$^4P_{3/2}$	
	$^4P_{1/2}$	$^4P_{1/2}$	$^4P_{1/2}$	
42	$(^1S_0)\,^2P_{3/2}$	$(^1S_0)\,^2P_{3/2}$	$(^1S_0)\,^2P_{3/2}$	
	$(^1S_0)\,^2P_{1/2}$	$(^1S_0)\,^2P_{1/2}$	$(^1S_0)\,^2P_{1/2}$	
	$(^3S_1)\,^2P_{1/2}$	$(^3S_1)\,^2P_{3/2}$	$(^3S_1)\,^2P_{3/2}$	
	$(^3S_1)^2P_{3/2}$	$(^3S_1)^2P_{1/2}$	$(^3S_1)^2P_{1/2}$	I
		$(^3S_1)^2P_{1/2}$	$(^3S_1)^2P_{1/2}$	II
	$^4P_{5/2}$	$^4P_{5/2}$	$^4P_{5/2}$	
	$^4P_{3/2}$	$^4P_{3/2}$	$^4P_{3/2}$	
	$^4P_{1/2}$	$^4P_{1/2}$	$^4P_{1/2}$	
54	$(^1S_0)\,^2P_{3/2}$	$(^1S_0)\,^2P_{3/2}$	$(^1S_0)\,^2P_{3/2}$	
	$(^3S_1)\,^2P_{1/2}$	$(^3S_1)\,^2P_{1/2}$	$(^3S_1)\,^2P_{1/2}$	
	$(^3S_1)\,^2P_{3/2}$	$(^3S_1)\,^2P_{3/2}$	$(^3S_1)\,^2P_{3/2}$	
	$^4P_{5/2}$	$^4P_{5/2}$	$^4P_{5/2}$	
	$(^1S_0)\,^2P_{1/2}$	$(^1S_0)\,^2P_{1//2}$	$(^1S_0)\,^2P_{1/2}$	
	$^4P_{3/2}$	$^4P_{3/2}$	$^4P_{1/2}$	
	$^4P_{1/2}$	$^4P_{1/2}$	$^4P_{3/2}$	

Sample data on % contribution to $1s2s(^3S_1)3p_{3/2}\ ^2P_{1/2}$ state

Z		contributions from		
		$1s2s(^3S_1)3p_{3/2}$	$1s2p_{1/2}(^3P_0)3s$	$1s2p_{1/2}(^3P_1)3s$
26		**79**	11	2.5
36		**53**	42.5	-
38	**I**	**44**	**52**	-
	II	30	30	**37**
39	**I**	**36**	**59**	-
	II	35	22	40
40	I	30	**66**	
	II	**38**	16	**40**
42	I	20	**75**	-
	II	**38**	**7**	**50**
44		**46**	15	37
46		**70**	13	26
54		**93**		3

Green – 1st major contribution Red- second major contribution

mixing coefficient contribution from 1s2s($^3S_1$)3p$_{3/2}$ state for various Z



I,II correspond to the levels as in the previous slide

1s2s3p -1s^{2}3s transition rates

L.Natarajan Phy. Rev.A, 88,052522,2013， M.H.Chen At.Data Nucl.Data Tables,34,301,1986

Z		$(^1S_0)\,^2P_{1/2}$	$(^1S_0)\,^2P_{3/2}$	$(^3S_1)\,^2P_{1/2}$	$(^3S_1)\,^2P_{3/2}$	$^4P_{1/2}$	$^4P_{3/2}$
36		6.494(13)	1.089(12)	1.223(12)	1.155(14)	2.419(11)	7.551(12)
	Ref. [21]	2.66(13)	3.71(11)	7.92(11)	1.04(14)	2.31(11)	6.76(12)
37		7.800(13)	6.033(11)	1.368(12)	1.491(14)	3.689(11)	8.525(12)
38	I	6.849(13)	1.823(11)	9.265(11)	1.835(14)	5.778(11)	8.664(12)
	II			1.933(14)			
39	I	7.815(13)	6.542(10)	3.613(12)	2.497(14)	7.053(11)	1.078(13)
	II			2.348(14)			
40	I	8.041(13)	3.492(14)	7.141(12)	3.248(14)	9.036(11)	1.236(13)
	II			2.894(14)			
42	I	4.294(13)	1.309(11)	2.568(13)	3.480(14)	1.340(12)	1.602(13)
	II			4.276(14)			
	Ref. [21]	1.26(13)	4.53(11)	4.09(14)	3.62(14)	1.18(12)	1.44(13)
44		4.667(12)	4.337(11)	5.405(14)	3.004(14)	2.007(12)	2.032(13)

Contribution from Breit+QED to transtion rates

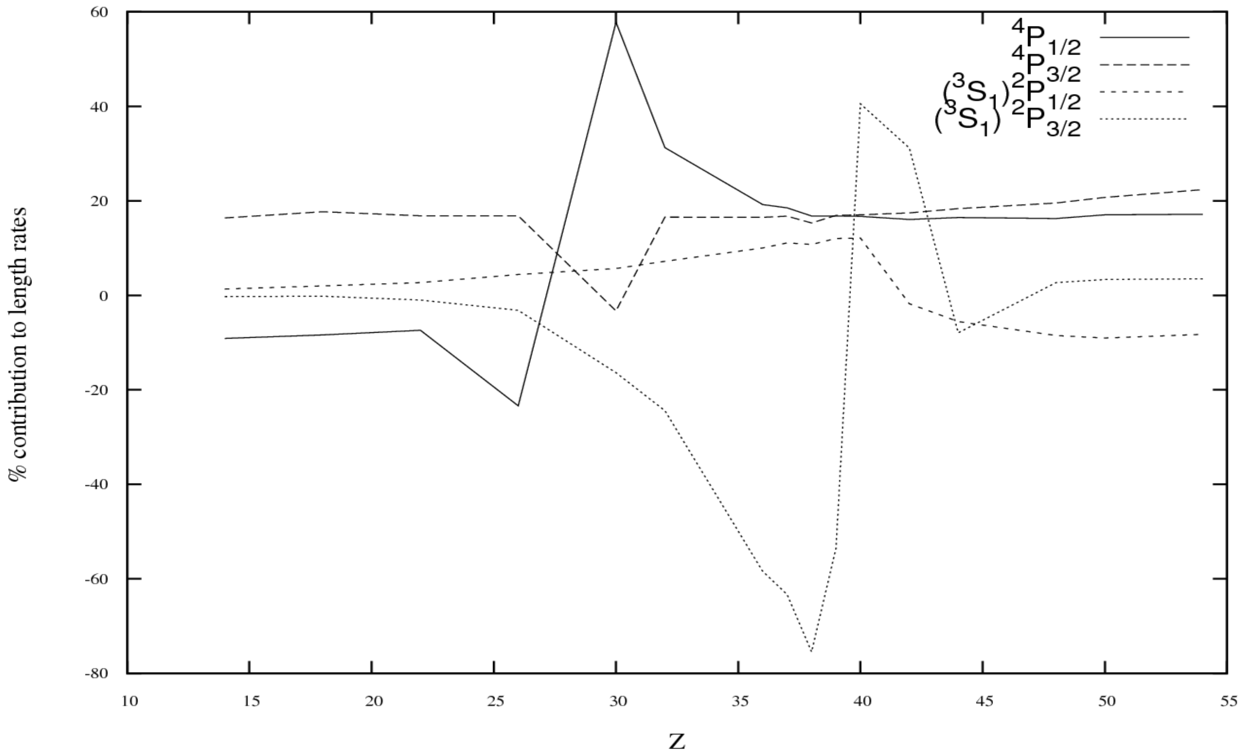


FIG.4 : % contribution from Breit +QED corrections to length rates of various 1s2s3p-1s^{2}3s transitions

2. Low lying level structure of 1s2p3s states of ions with Z=37 to 43

Level crossing

In LSJ coupling , the 3 J=1/2 states

$1s2p[^1P_1]3s \quad [^2P_{1/2}]$ } correlation CSFs same for all Z

$1s2p[^3P_0]3s \quad [^4P_{1/2}]$ }

$1s2p[^3P_1]3s \quad [^2P_{1/2}]$ → varies for Z = 37 to 43

Inter complex correlation affects transition rates of ions around Z = 26

Intra complex correlation leads to level crossing around Z =37 to 43

II. Mixing coefficients contributions to the $1s2p_{1/2}(^3P_1)3s$ J=1/2 state

Z	$1s2p_{1/2}(^3P_1)3s$	$1s2s(^1S_0)3p_{1/2}$	$1s2p_{1/2}(^3P_0)3s$	$1s2s(^3S_1)3p_{3/2}$	$1s2p_{3/2}((^1P_1)3s$
36	0.7233	-0.4191	0.4201	-0.3431	
37 I	0.6023		-0.5699	0.5339	-0.1585
II	0.7242	-0.3922	0.4221	-0.3693	
38 I	0.6229		-0.5115	0.5687	-0.1561
II	0.7249	-0.3522	0.4244	-0.4040	
40 I	0.6691	0.0661	-0.3787	0.6166	-0.2495
II	0.7100	-0.2709	0.4238	-0.4853	
42 I	0.7241	0.1216	-0.2406	0.6175	-0.1401
II	0.6605	-0.1776	0.4134	-0.5881	
44	0.7823	0.1880	0.1159	0.5660	-0.1279

Mixing coefficients contribution to the main $1s2p(^3P_1)3s\ ^2P_{1/2}$ state

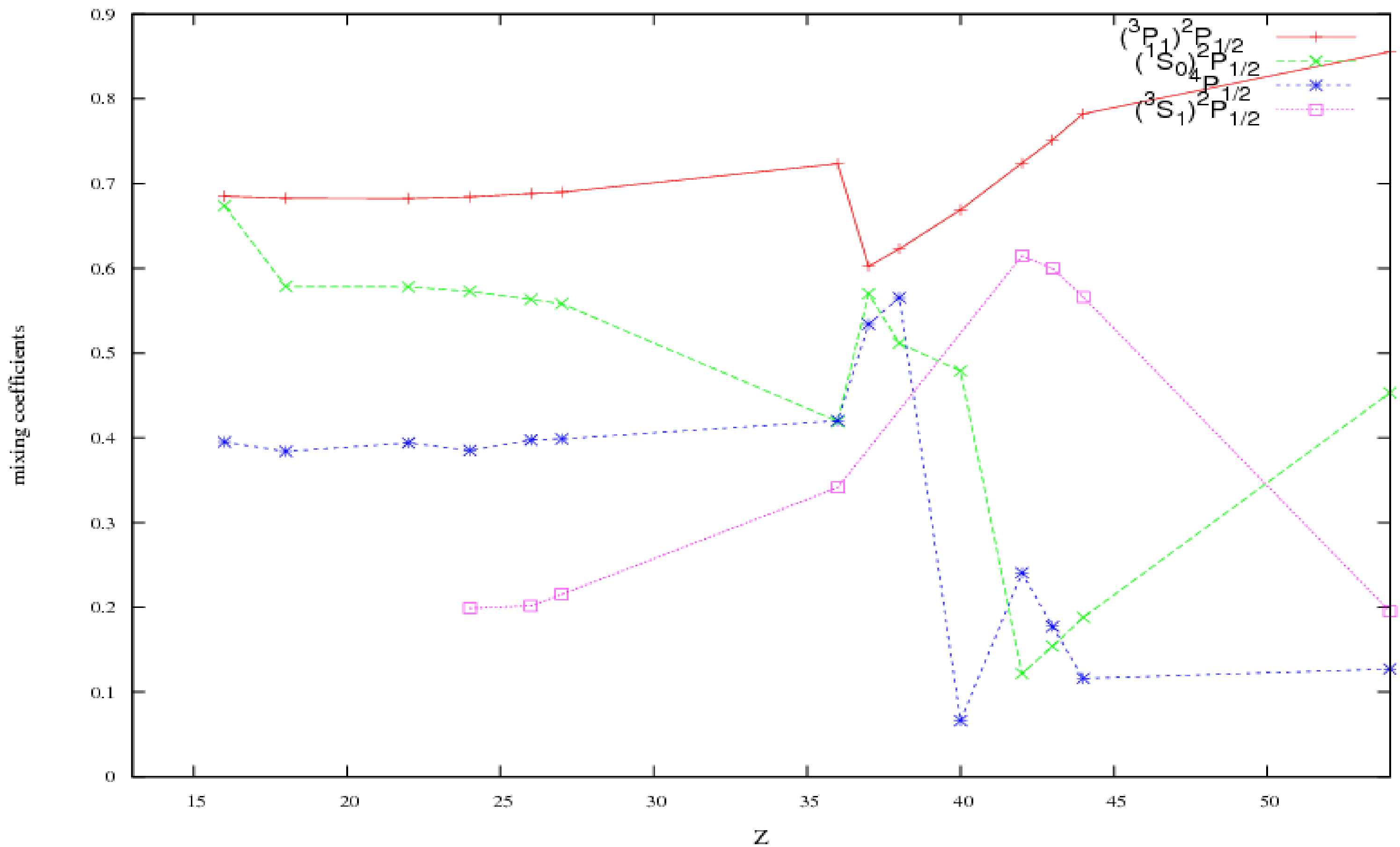


FIG.1 : mixing coefficient admixture to the main $1s2p(^3P_1)3s\ ^2P_{1/2}$ state (level I)

Breit+QED corrections to 1s2p3s-1s^{2}3s wavelengths

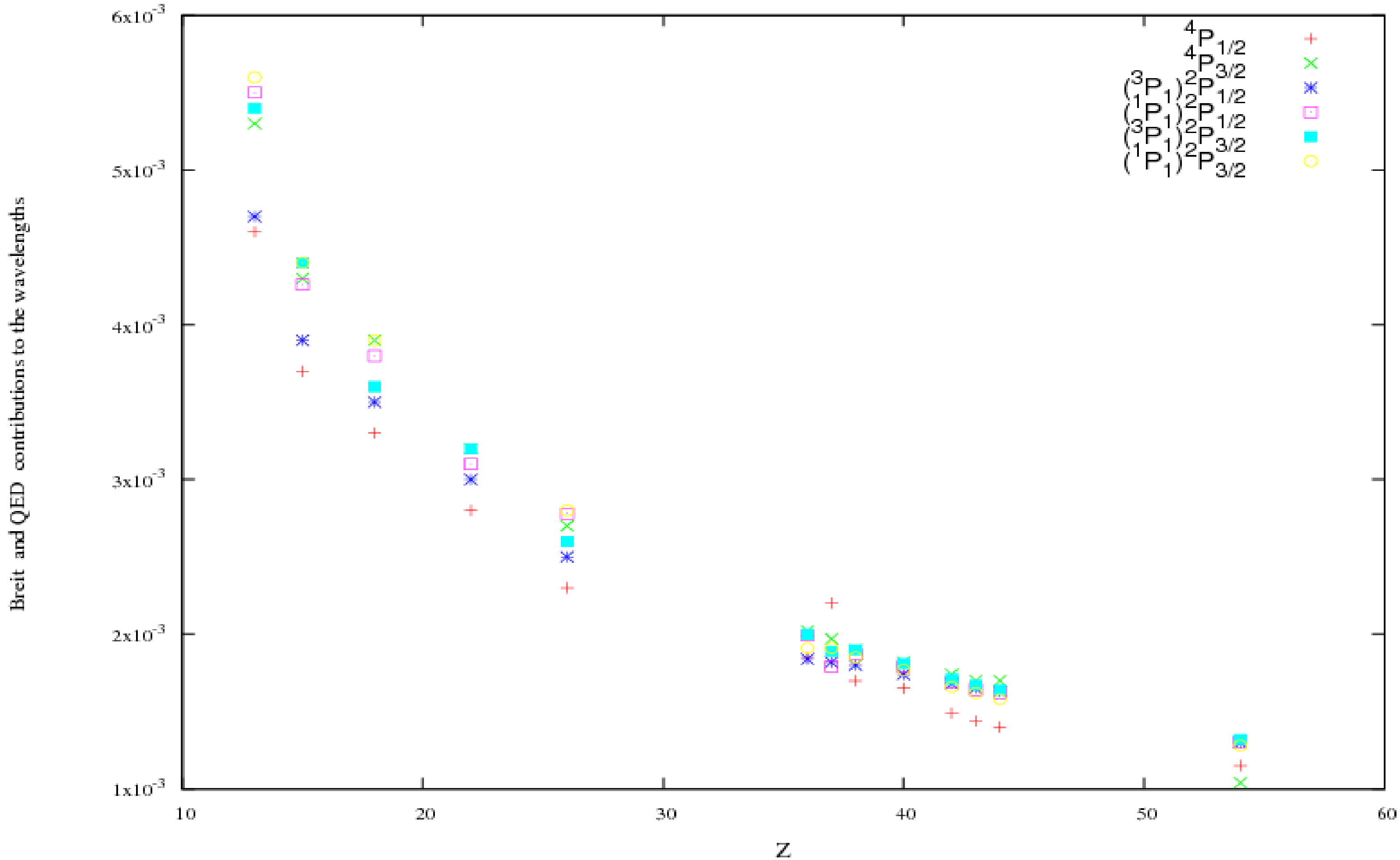


FIG.2 : Breit + QED contributions to the wavelengths in A^0 of the fine structure transitions from states of 1s2p3s to 1s^{2}3s ($^2S_{1/2}$)

$1s2p3s$-$1s^2 3s$ transition rates

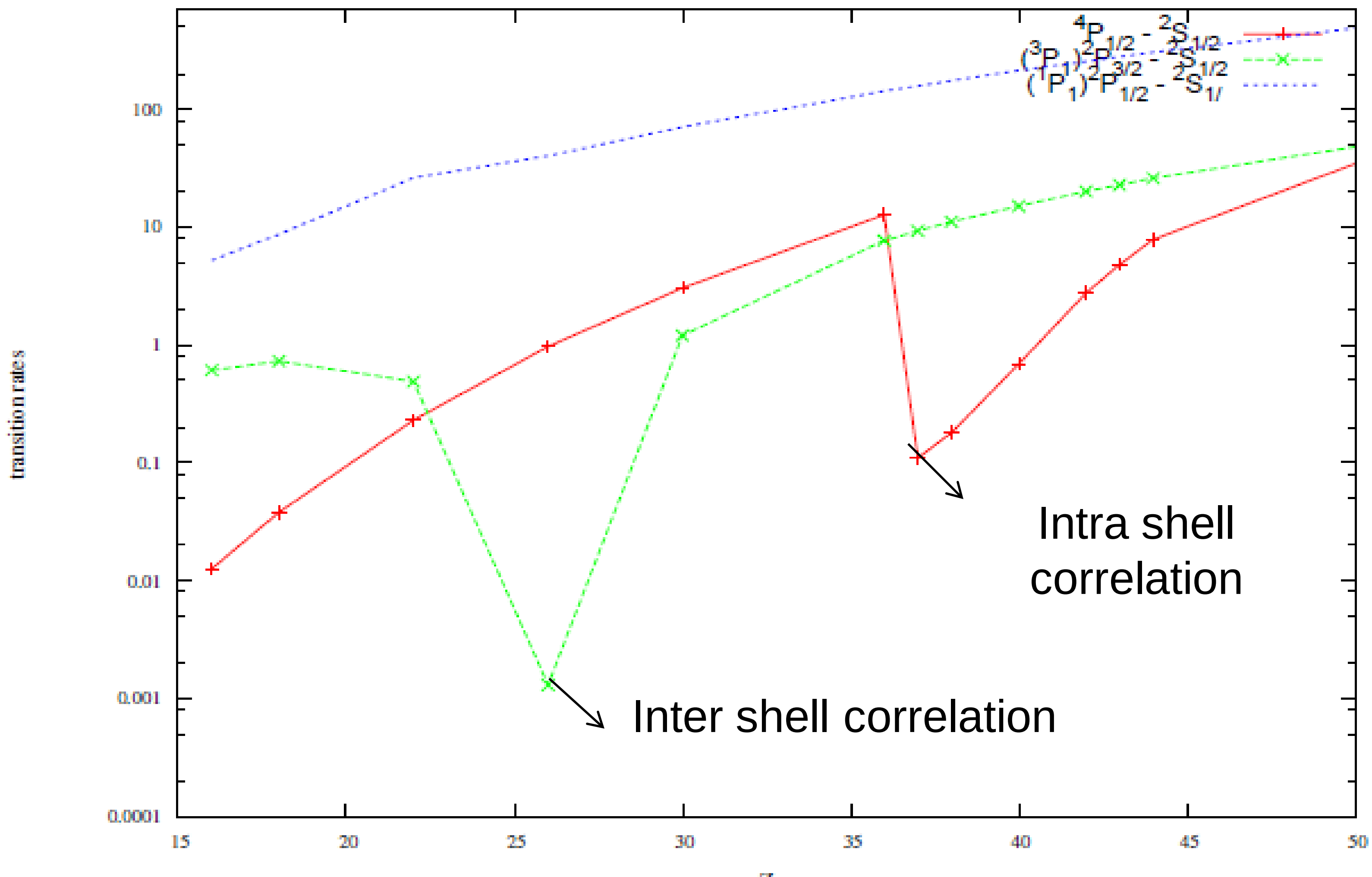


FIG.3 : transition rates in 10^(13)s^(-1)

Breit contribution to transition rates

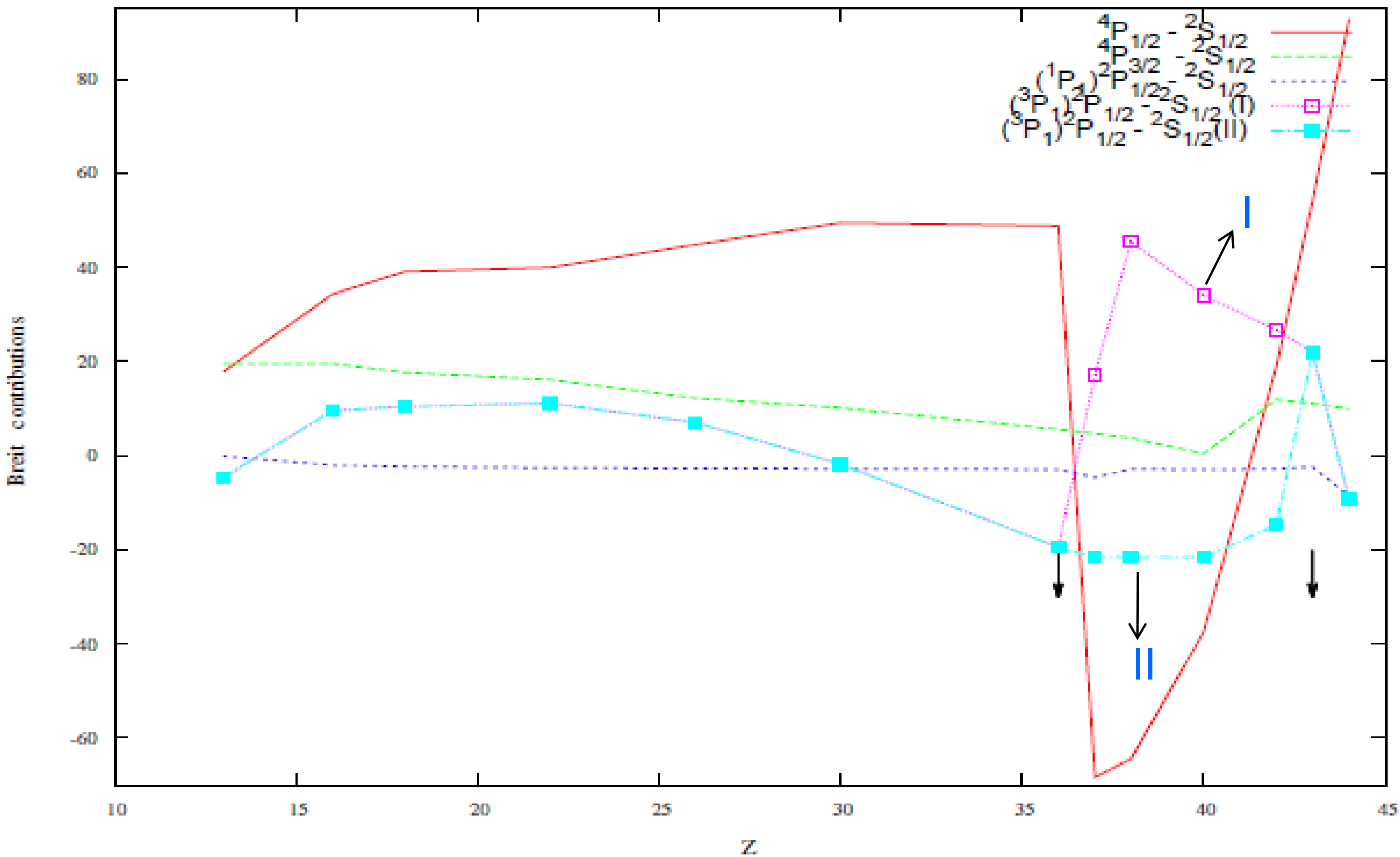


FIG.4 : Breit interaction contribution in percentage to transition rates

Essential features

1. For low and high Z ions ,LSJ /jj coupling scheme
2. For mid Z, due to level crossing ,unique identification becomes difficult.
3. Inter and intra correlation drastically affect the transition rates of certain ions
4. Breit interaction (relativistic component of e-e interaction) is

   a) Z dependent

   b) changes the rates of certain transitions drastically

   c) no influence on some transitions

.

Anomalously intense TEOP transitions from states of $2s^2$ and $1s2s^2$ configuration

In general - Usual mode of decay of

$2s^2\ (^1S_0)$ and $1s2s^2\ {}^2S_{1/2}$ to ground state :

: M1, weak transition

: arises due to relativistic effects

: K shell fluorescence yield zero for light and medium heavy ions.

Unusual TEOP decay mode : configuration mixing

: E1 possible

: anomalously intense

: K shell fluorescence yield is NOT zero

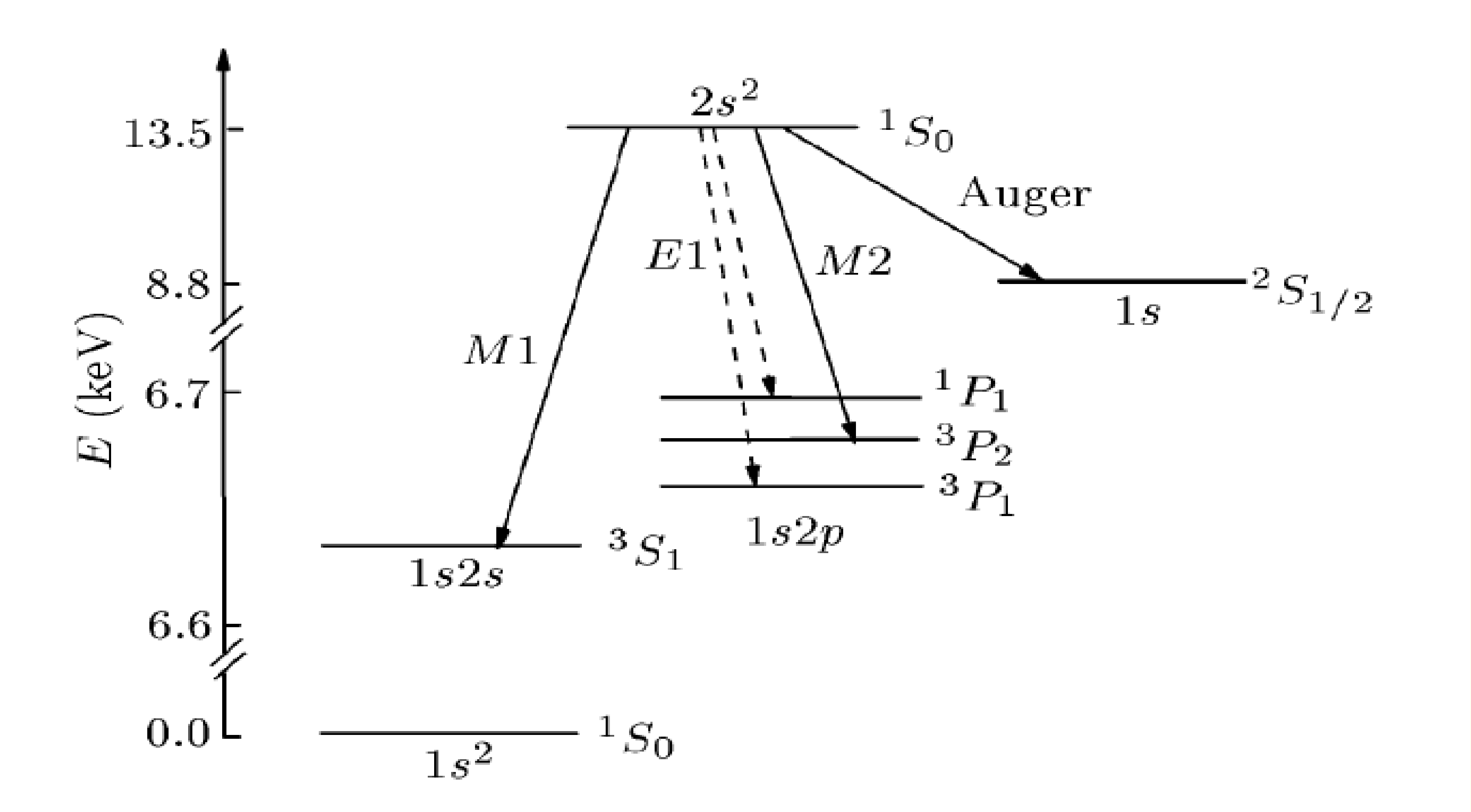
: atomic data needs modification

J.S.Kaastra and R.Mewe ,A & A,198,97,1993

TEOP and M1 transitions from $2S^2$ ($^1S_0$)

Decay channels of the $2s^2\ ^1S_0$ state in Fe^{24+}

Configuration mixing between $2s^2(^1S_0)$ and $2p^2(^1S_0, ^3P_0)$ make E1 mode possible



Breit and QED contributions in eV to $2s^2$-1s2p transition energies

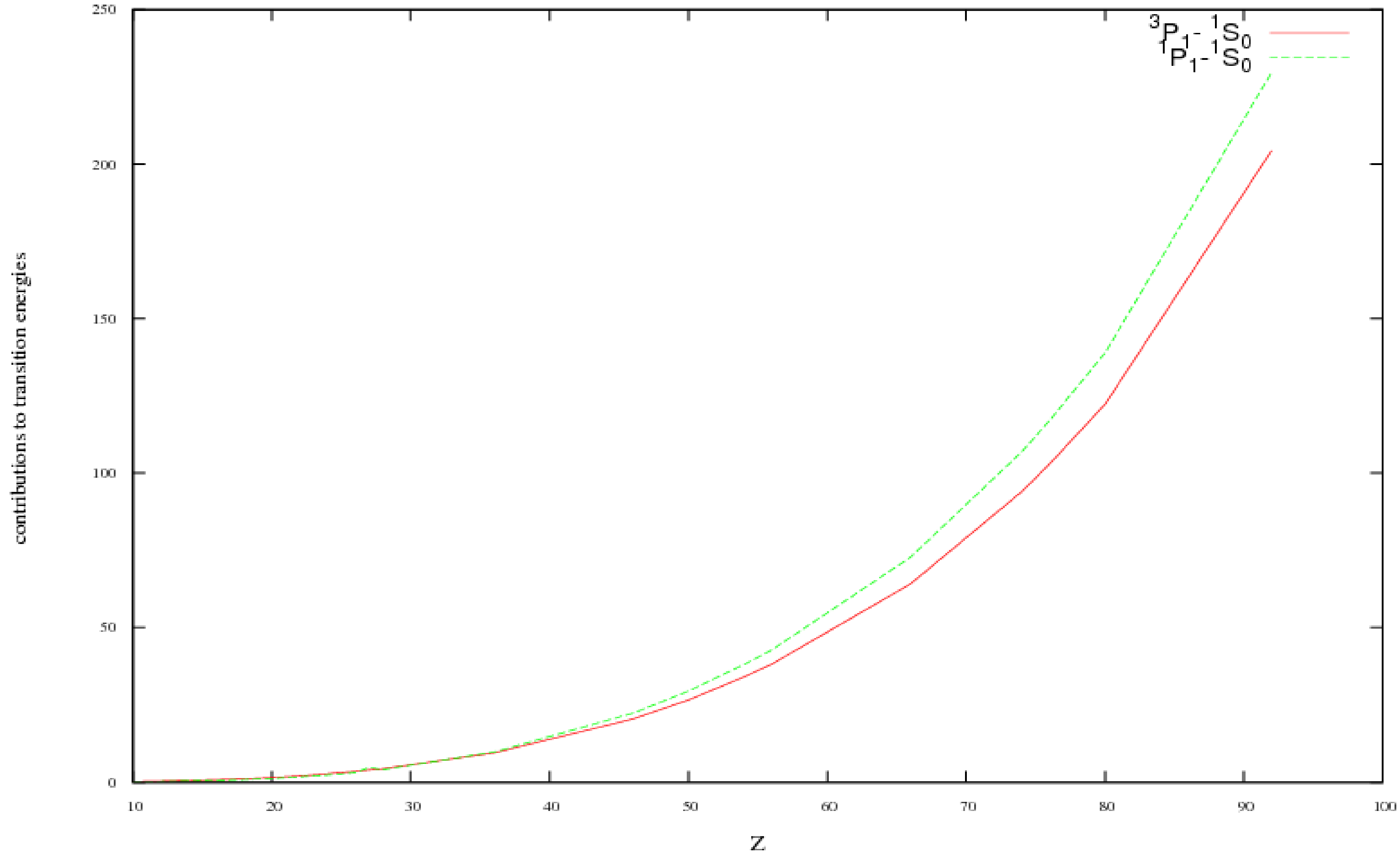


FIG.1 : energy contributions in eV from Breit +QED corrections to $2s^2$-1s2p transitions

E1 and M1 energies (eV) and rates (s^{-1}) from $2s^2$ ($^1S_0$) from Relaxed initial and final state orbitals

- Interplay between correlation and relativity

		$^1P_1$-$^1S_0$		$^3P_1$-$^1S_0$		$^3S_1$-$^1S_0$	
Z	Method	Energy	Rate	Energy	Rate	Energy	Rate
10	correlation	4026.26	3.253[13]	4046.21	5.853[12]	4068.30	3.906[
	Breit	4026.30	3.216[13]	4045.94	6.147[12]	4068.36	3.911[
	Breit+QED	4025.01	3.176[13]	4046.66	6.113[12]	4066.89	3.902[
27	correlation	7392.51	6.443[13]	7429.52	6.237[13]		
	Breit	7392.32	6.286[13]	7429.30	6.415[13]		
	Breit+QED	7388.91	6.267[13]	7425.58	6.743[13]		
28	correlation	7957.63	6.741[13]	7998.31	7.980[13]		
	Breit	7958.01	6.226[13]	7997.90	8.261[13]		
	Breit+QED	7953.55	6.526[13]	7993.92	8.634[13]		
46	correlation	21821.85	5.934[13]	22042.11	1.187[15]	22108.54	1.807[1
	Breit	21819.83	5.242[13]	22041.61	1.182[15]	22109.12	1.8179[
	Breit+QED	21799.49	5.114[13]	22021.66	1.359[15]	22085.23	1.741[1
54	correlation	30311.18	4.622[13]	30731.09	2.409[15]	30813.89	9.411[1
	Breit	30313.34	3.948[13]	30736.71	2.313[15]	30821.71	9.553[1
	Breit+QED	30273.09	3.720[13]	30697.07	2.898[15]	30774.55	8.862[1
92	correlation	93127.10	1.315[13]	97543.09	3.049[16]	97726.49	2.552[1
	Breit	93108.70	8.677[12]	97547.89	2.866[16]	97734.11	2.622[1
	Breit+QED	92897.39	4.342[12]	97338.71	5.005[16]	97482.21	1.742[1

Percentage contributions from Breit + QED to $2s^2$-1s2p E1 rates

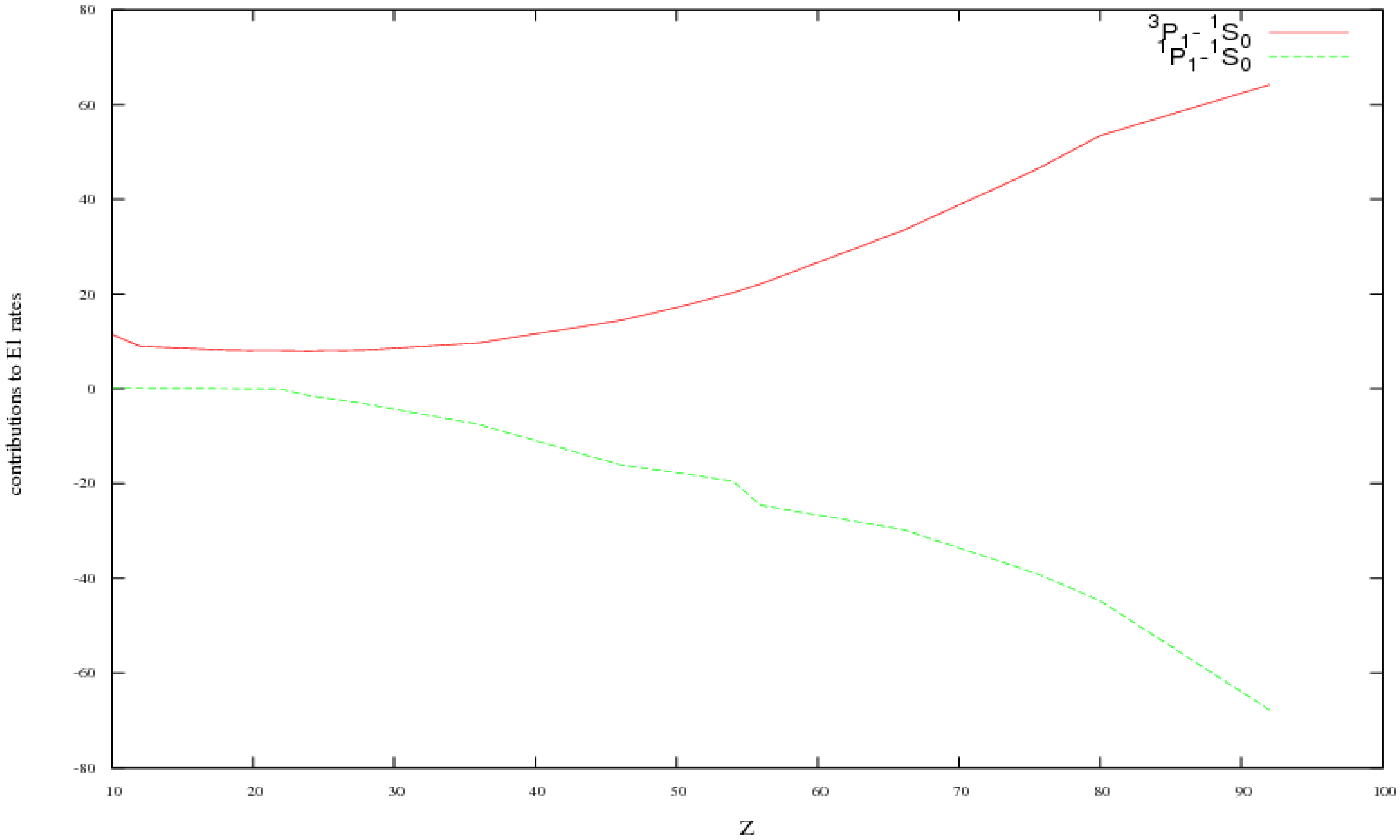


FIG.2 : percentage contributions from Breit +QED corrections to length gauge E1 rates

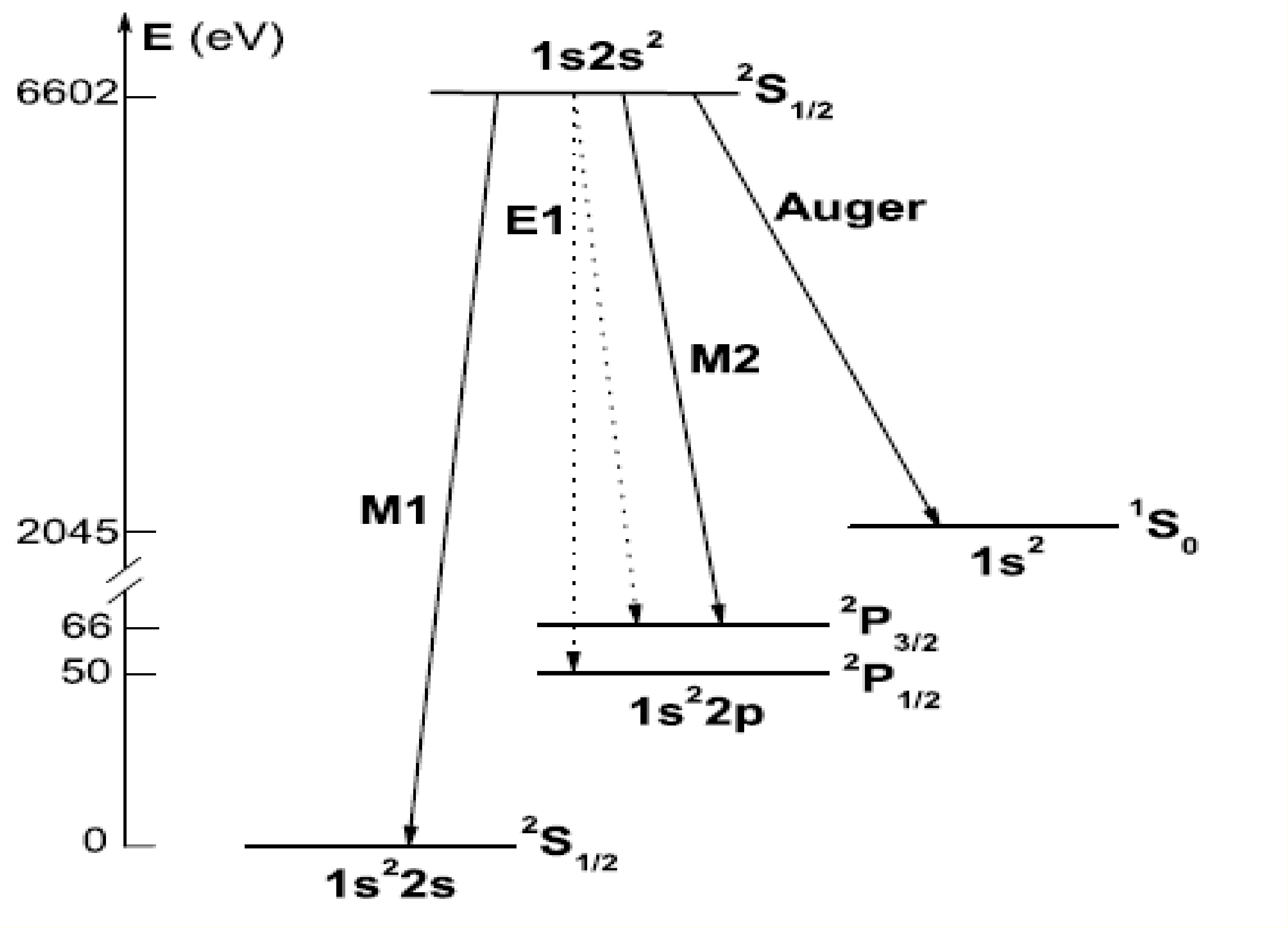
Biorthogonal(row 1) and common (row 2) orbital sets - E1 and M1 values for $2s^2$-1s 2p transitions

Z	$^1P_1-^1S_0$ RCI	$^1P_1-^1S_0$ Others	$^3P_1-^1S_0$ RCI	$^3P_1-^1S_0$ Others	$^3S_1-^1S_0$ RCI	$^3S_1-^1S_0$ Others
26	5.891(13)	5.9(13)	4.997(13)	4.9(13)	5.711(8)	5.46(8)
	5.837(13)		4.908(13)		5.651(8)	
27	6.267(13)	6.26(13)	6.743(13)	6.38(13)	1.186(9)	
	6.133(13)		6.661(13)			
46	5.114(13)	5.02(13)	1.359(15)	1.04(15)	1.74(11)	1.8(11)
	5.151(13)		1.156(15)		1.8(11)	
66	2.178(13)	2.37(13)	7.508(15)	4.07(15)	6.787(12)	7.79(12)
	2.469(13)		5.209(15)		7.203(12)	
92	4.342(12)	8.88(12)	5.005(16)	1.38(16)	1.742(14)	2.82(14)
	8.264(12)		2.345(16)		2.135(14)	

TEOP and M1 transitions from states of $1s2S^2$ configuration

Li-like Fe (Fe^{23+})

Level structure and decay channel of $1s2s^2\ {}^2S_{1/2}$ level of Fe^{23+}



$1s2s^2$-$1s^22p$ transition energies in eV and rates in s^{-1}

Z	$^2S_{1/2}$-$^2P_{1/2}$ Energy	rate	$^2S_{1/2}$-$^2P_{3/2}$ Energy	rate
10	538,76	1.12(11)	538.69	2.19(11)
14	1796.51	5.32(11)	1795.44	9.60(11)
Expt[a]	1802.83		1795.13	
18	3047.1	1.71(12)	3043.9	2.69(12)
Expt[a]	3044 (±6)		3041(±6)	
24	5549.2	6.03(12)	5537.9	6.93(12)
26	6551.98	**8.53(12)**	6536.56	**8.41(12)**
Expt[b]	6552.47		6536.54	

a Zou etal PRA 67, 042743,2003

b P.Beiersdorfer etal APJ,409,846, 1993

Effect of correlation in flipping the intensity of fine structure K x-rays at Li-like Fe

	No.of CSFs	$^2P_{1/2}$-$^2S_{1/2}$	$^2P_{3/2}$-$^2S_{1/2}$
Ref {1s2s2p}	DF	0	0
{1s2s2p }	22	9.463(12)	9.727(12)
{1s2s2p3s3p}	102	**8.541(12)**	**8.497(12)**
{1s2s2p---4s4p}	246	8.537(12)	8.379(12)
{1s2s2p----5s5p}	454	8.531 (12)	8.311(12)

Influence of mixing coefficients from $1s2p^2_{1/2}$ and $1s2p^2_{3/2}$ states to the main $1s2s^2\ ^2S_{1/2}$ state with only Coulomb and Coulomb +Breit

$$1s2s^2 \quad -c_1 \quad ; \quad 1s2p^2_{1/2} \quad -c_2 \quad ; \quad 1s2p^2_{3/2} \quad -c_3$$

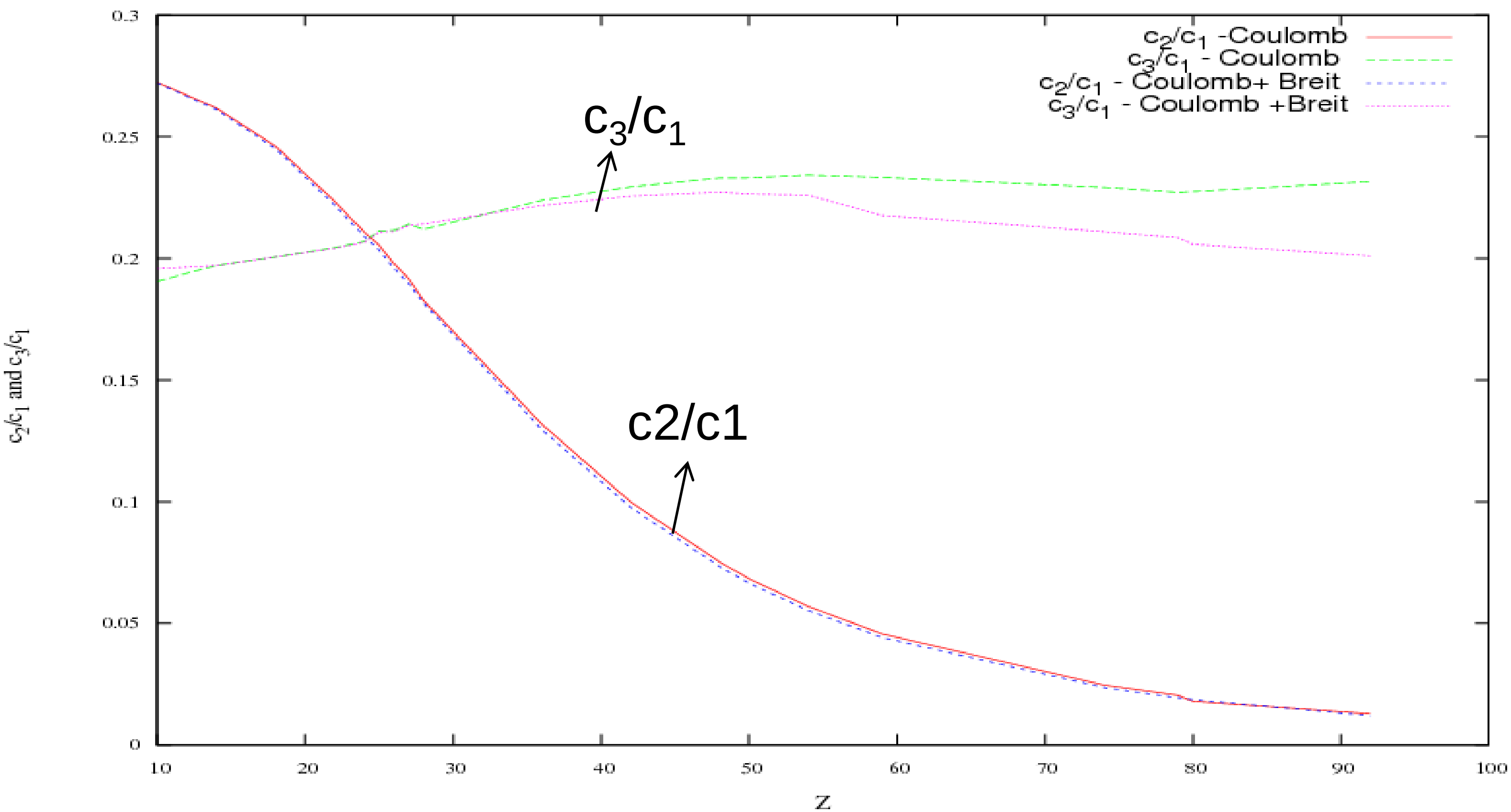


FIG.1 : ratios of mixing coefficients from $1s2p^2_{1/2}$ (c_2) and $1s2p^2_{3/2}$ (c_3) realtive to $1s2s^2$(c_1) with only Coulomb and Coulomb + Breit interactions

Breit interaction contribution to E1 and M1 rates

Either increases or decreases TEOP rates

M1 rates marginally affected

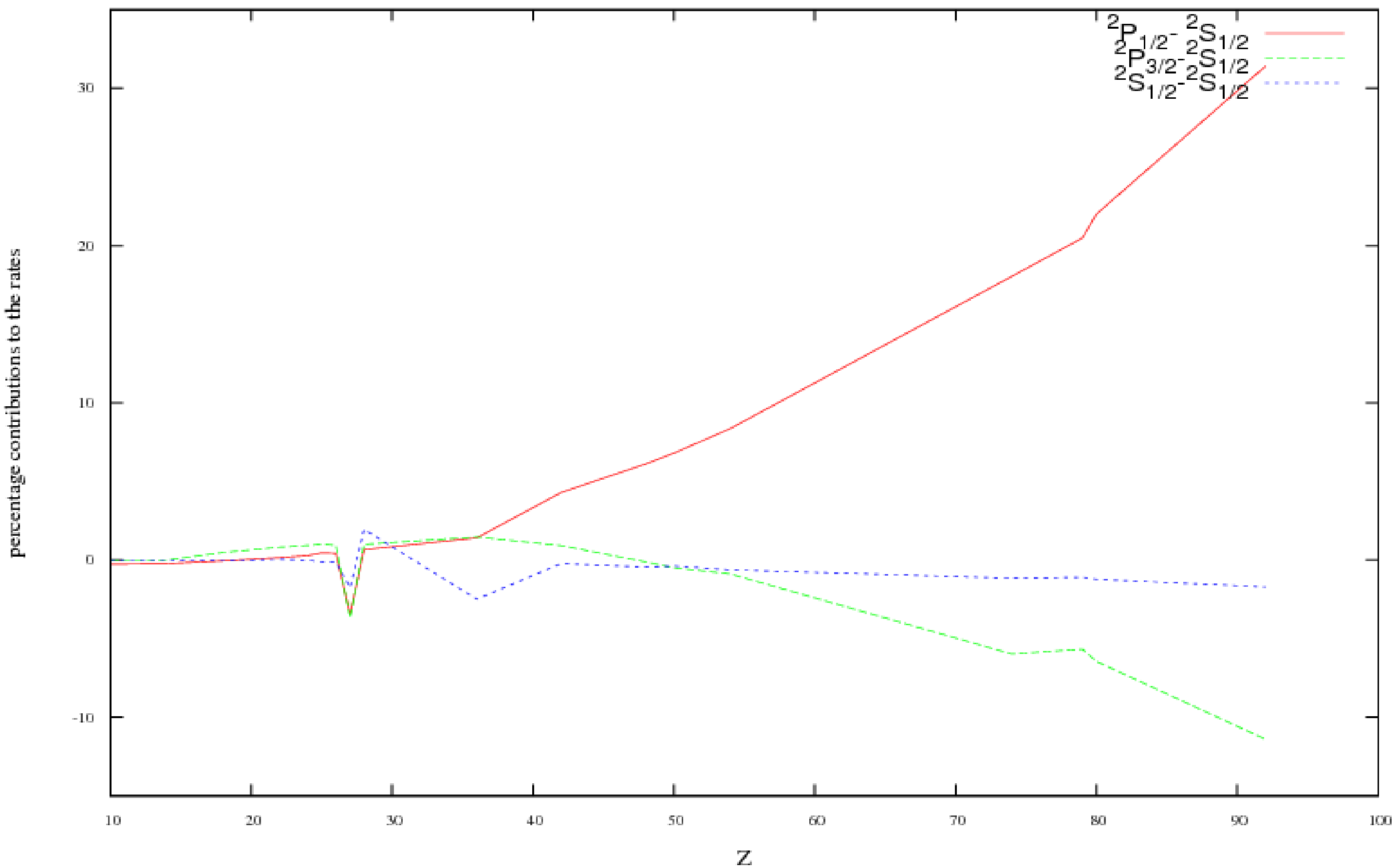


FIG.3 : Breit interaction contributions in percentage to the E1 and M1 transition rates

Essential features

1. Anomalous TEOP transitions are intense.
2. Existing data on the decay of $2s^2$ $(^1S_0)$ and $1s2s^2$ $^2S_{1/2}$ need modifications.

References

1. L.Natarajan,A.Natarajan and R.Kadrekar Phys.Rev.A, 82, 062514,(2010).
2. R. Kadrekar and L.Natarajan, Phys. Rev.A, **84**, 062506, (2011)
3. L.Natarajan Phys.Rev.A,85,2012 .
4. L.Natarajan Phys.Rev.A, 93,032516,2016,
5. L.Natarajan and R.Kadrekar,88,12501,2013
6. L.Natarajan Phys. Rev. A, 88,052522,2013
7. L.Natarajan Phys. Rev. A,90,032509,2014
8. L.Natarajan Phys. Rev.A, 92,012507,2015
9. L.Natarajan ADNDT,2016 (under communication)

Thank you