KfK 4176
März 1987

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Relativistic Calculations of Level Energies of High-Z Atom

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ISSN 0303-4003

Abstract

Level energies are calculated using relativistic and non-relativistic Hartree-Fock-Slater (RHFS & HFS) models for an Au-atom in various charge states. The level splitting due to orbital angular momentum and electron spin is investigated in comparison to a simple analytical model. This model is based on shell model and is an extension of previous such models in that it takes overlap of orbitals into account. The model can satisfactorily replicate the results of RHFS and HFS calculations. This model can be applied to other atoms as well.

Relativistische Berechnung des Energie-Niveaus der Atome mit hoher Ladungszahl

Zusammenfassung

Niveau-Energien für Au-Atome in verschiedenem Ladungszustand werden mit Hilfe der relativistischen und der nicht relativistischen Hartree-Fock-Slater-Methode (RHFS & HFS) berechnet. Die Niveau-Aufspaltung durch Bahndrehimpuls und Elektronenspin wird im Vergleich zu einem einfachen analytischen Modell untersucht. Dieses Modell basiert auf dem Schalen-Modell und ist eine Erweiterung der bisherigen Modelle, indem die Überlappungen der Elektronen-Bahnen berücksichtigt werden. Dieses einfache Modell gibt die Ergebnisse der RHFS und HFS Rechnungen wieder. Dieses kann daher auch für andere Atome angewandt werden.

Recently, the non-LTE average-ion model coupled with a hydrodynamic code and radiation transports has been extensively used for interpreting soft X-ray spectra from laser-irradiated high-Z plasmas[1-4]. Although for level calculations only principal quantum number and a screened hydrogenic model were used, the model replicated the experimental results both qualitatively and quantitatively , except a few discrepancies. One of these discrepancies is probably attributed to the ionic charge distribution : the line locations of the fictitious " average ion" are different from those of "real ions" being in different charge states ; even if they are in the same charge state, the level electron population is different which causes the change of level energies[5]. In order to eliminate this defect we developed a hybrid-atom model[6], in which the charge state of ions and their excited state population can be simultaneously calculated. The calculation time is comparable to that of the average-ion model. We have applied this model to analyze X-ray spectrum from a laser-heated cavity plasma[7]. By introducing the charge state distribution and hence treating the "real ions" instead of an average ion, line transport can be correctly performed and the line locations in energy space are correctly calculated.

As pointed out in a previous paper[4], some of the discrepancies can also be attributed to the neglect of line splitting due to angular momentum and spin-orbit interaction. Although preliminary calculations show that introduction of orbital quantum numbers can make the line spectra shift towards the lower energy, its implementation into the hydrodynamic code

was until now beyond our reach, because a simple formula for energy levels and oscillator strenths of high-Z plasmas were not available. The aim of this paper is to provide one of such formula suitable for the average ion model as well as for hybrid atom model. For this purpose, we extend the simple shell model proposed by Parker[8] and compare the result with a relativistic Hartree-Fock-Slater model, which was used in interpreting the M-shell line spectra in laser-produced Au plasmas.[9]

Calculations have been done by peeling off electrons one by one from ground levels. In Fig.1, level energies of the ℓ=0 state are plotted for various n ; ℓ and n mean the orbital and principal quantum numbers, respectively. In Figs.2 and 3, level energies measured from ℓ=0, that is $E_{n,\ell}-E_{n,0}$ are plotted both for non-relativistic and relativistic cases, respectively. Surprisingly, these values remain constant over a wide range of the charge state although $E_{n,0}$ changes largely as seen from Fig.1.

In the followings, we try to find a simple scaling law of these level energies using Parker's shell model, which has been modified for open shells ; the electron density is written in terms of contributions from each shell,

$$\rho(r)=\sum_{p=1}^{\infty}\rho_p(r)=\sum_{p=1}^{\infty}P(p)\delta_p(r)/4\pi r^2 \qquad (1)$$

where P(p) is the electron population in the p-th shell and is $2p^2$ for a closed shell and $\rho_p(r)$ is the density of p-shell electron at radius r. Each $\delta_p(r)$ is a fucntion

which sharply peaks at $r_p = p^2/Z_p$ (atomic units are used). The effective charge Z_n can be calculated to be

$$Z_n = Z - P(n)/2 - \sum_{p=1}^{n-1} P(p) \qquad (2)$$

Then the level energy is given by

$$E_n = - Z_n^2/2n^2 + P(n)/2r_n + \sum_{p=n+1}^{\infty} P(p)/r_p + \Delta E_{NC} + \Delta E_{EX} + \Delta E_{RE}, \qquad (3)$$

where ΔE_{NC}, ΔE_{EX}, and ΔE_{RE} are the non-Coulombic, exchange, and relativistic corrections, respectively.

The exchange correction was given by Parker as follows;

$$\Delta E_{EX} = -1.2(3/2)^{4/3} Z_n/\pi^{5/6} n^{7/6}. \qquad (4)$$

An estimate of the non-Coulombic potential can be obtained by expanding the electron density around $r=r_n$ and thus[8]

$$\Delta V \simeq - 2\pi r_n \rho(r_n)(r-r_n)^2/r \qquad (5)$$

The density in the n-th state is approximated as

$$\rho_n(r)=A_n r^{2n-2} \exp(-2Z_n r/n), \qquad (6)$$

where the normalization constant A_n is

$$A_n = P(n)\ [4\pi\ (n/2Z_n)^{2n+1}(2n)!]^{-1}$$

This profile is found to be a good approximation to that obtained with RHFS. However, Parker used $\rho_n(r_n)$ for $\rho(r_n)$ assuming non-overlapping of orbits. Using this expression we obtain

$$\Delta E_{NC} = \Delta E_{n,\ell} = -Z_n P(n)/4\pi^{1/2} n^{7/2} \times [\ n^2 - \ell(\ell+1)\ ] \qquad (7)$$

This equation distinguishes from Parker's equation in that it is not confined to closed shell.

This model suggests that $\Delta E_{n,\ell} - \Delta E_{n,0} = Z_n P(n)\ell(\ell+1)/4\pi^{1/2} n^{7/2}$ remains constant for fixed n and ℓ if $Z_n P(n)$ is constant. As is easily seen, this is not the case. $Z_n P(n)$ can change by an order of magnitude from a closed shell to an open shell. The result with this model(Eqs.(3) and (7)) is shown in Figs.1, 2, and 3 by the dashed lines and is compared with calculations with a non-relativistic Hartree-Fock-Slater (HFS) model. Although Eq.(7) and HFS agree with each other for the lower charge state, the difference becomes significant when the number of electron in the shell in which the energy is being calculated decreases. This is due to inadequacy of the shell model as shown below. In order to clarify this, we take the ratio of electron densities in the n-th and (n-1)-th shells at $r=r_n$;

$$\frac{\rho_{n-1}}{\rho_n}\Big|_{r=r_n} = \frac{P(n-1)}{P(n)}\left[\frac{Z_{n-1}{}^n}{Z_n(n-1)}\right]^{2n-1}{}^{n(n-1/2)} \times \exp\left\{-2n\left[\frac{Z_{n-1}{}^n}{Z_n(n-1)}-1\right]\right\}.$$

If we assume $Z_n \simeq Z_{n-1}$ and n=4, then $\rho_3/\rho_4 \simeq 7.3\ P(3)/P(4)$ and hence the density of the third shell cannot be neglected even at $r=r_4$. This behavior is easily understood if the orbit radius calculated by $r_n=n^2/Z_n$ and Eq.(2) is plotted for various ionic charge states as in Fig.4 ; as the charge state becomes higher, all the shells come closer, and the density profile of various shells overlap with each other.

We find that this overlap can be easily taken into account by modifying the density to give;

$$\Delta E_{n,\ell} = -\pi(n/Z_n)^2[n^2-\ell(\ell+1)] \left[\rho_n(r_n)+\Sigma\rho_k(r^*_n)\right], \qquad (8)$$

where the contribution from different shells is included in the last term in Eq.(8), where densities are estimated at $r=r^*_n$ which is taken to be somewhat smaller than r_n because of the finite width of the distribution Eq.(6). We find that

$$r^*_n = r_n(A-Br_n)$$

where

$A=1.0$, $B=0.93333$ for $r_n<0.15$,

$A=0.9275$, $B=0.45$ for $0.15<r_n<0.45$,

$A=0.725$, $B=0.$ for $0.45<r_n<0.8$,

$A=0.8797$, $B=0.1934$ for $0.8<r_n<2.17$,

$A=0.46$, $B=0.$ for $2.17<r_n$,

can reasonably describe the behavior obtained with HFS. This comparison is given in Figs.1 and 2 ; the results with the above model Eq.(8) are shown by the solid lines.

As clear from Figs.2 and 3, a significant difference exists between RHFS and HFS, particularly in the inner shells. If the effective charge Z_n from Eq.(2) is used in the expression of the relativistic correction ;

$$\Delta E_{RE}=-Z_n{}^4\alpha^2/2n^3[1/(j+1/2)-3/4n], \quad (10)$$

the large difference can not be explained. Instead of trying to find an alternative expression, we try to adjust the effective charge Z^*_n to fill this difference with Eq.(10). Z^*_n is calculated so that $\Delta E_{RE}|_{j^*} - \Delta E_{RE}|_{j=1/2}$ fits to the corresponding difference between RHFS and HFS ; j^* is 3/2, 5/2, and 7/2 for n = 3, 4, and 5, respectively, and other levels are calculated with the corresponding same Z^*_n and Eq.(10). As is shown in Fig.3, this modification can satisfactorily replicate the RHFS results. The Z^*_n thus obtained is plotted in Fig.5, which shows Z^*_n is significantly larger than Z_n except for the lower n (≤ 3). a reasonable explanation of this large deviation of

Z^*_n from Z_n has not yet been found. One possibility is the non-Coulombic potential ; in deriving Eq.(10), the Coulombic potential $Z_n e^2/r$ was used. If we extend it to include the non-Coulombic potential such as Eq.(5), the relativistic correction also changes.

In summary, we performed the relativistic and non-relativistic Hartree-Fock-Slater calculations for an Au atom and tried to get a simple scaling law for level energies. We found that for the estimation of the non-Coulombic potential the shell model should be modified to include the overlap of orbits. This aim was partly accomplished. Although it is somewhat empirical, we have derived a formula to incorporate orbit overlap in a simple form and furthermore obtained an empirical formula for the relativistic corrections. Further improvement of the formula is expected for this relativistic correction.

The authors would like to thank Mr.S.Kiyokawa at Nara Women's University for his help to make computer programs of RHFS and HFS and Dr.R.Fröhlich at KfK for discussion and encouragement. One of the authors (T.Y.) would also like to thank Professor G.Keßler and Dr.R.Fröhlich at KfK for giving him a chance to stay at KfK as a guest scientist.

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5. We remind that an average ion is an ion having in principle non-integral occupation numbers of electrons in a level. A plasma of N "average ions" represents N ions in different charge and excitation states, but conserves number of free and bound electrons. Number of bound electrons in a particular level is also conserved. For example an average ion of two ions with one neutral atom and one in charge state X will be in average charge state X/2 and all energy levels will belong to this charge state. In reality however, 50% of the levels correspond to a neutral atom and 50% to an atom in charge state X. The energies of these levels can be completely different than those of the average atom.
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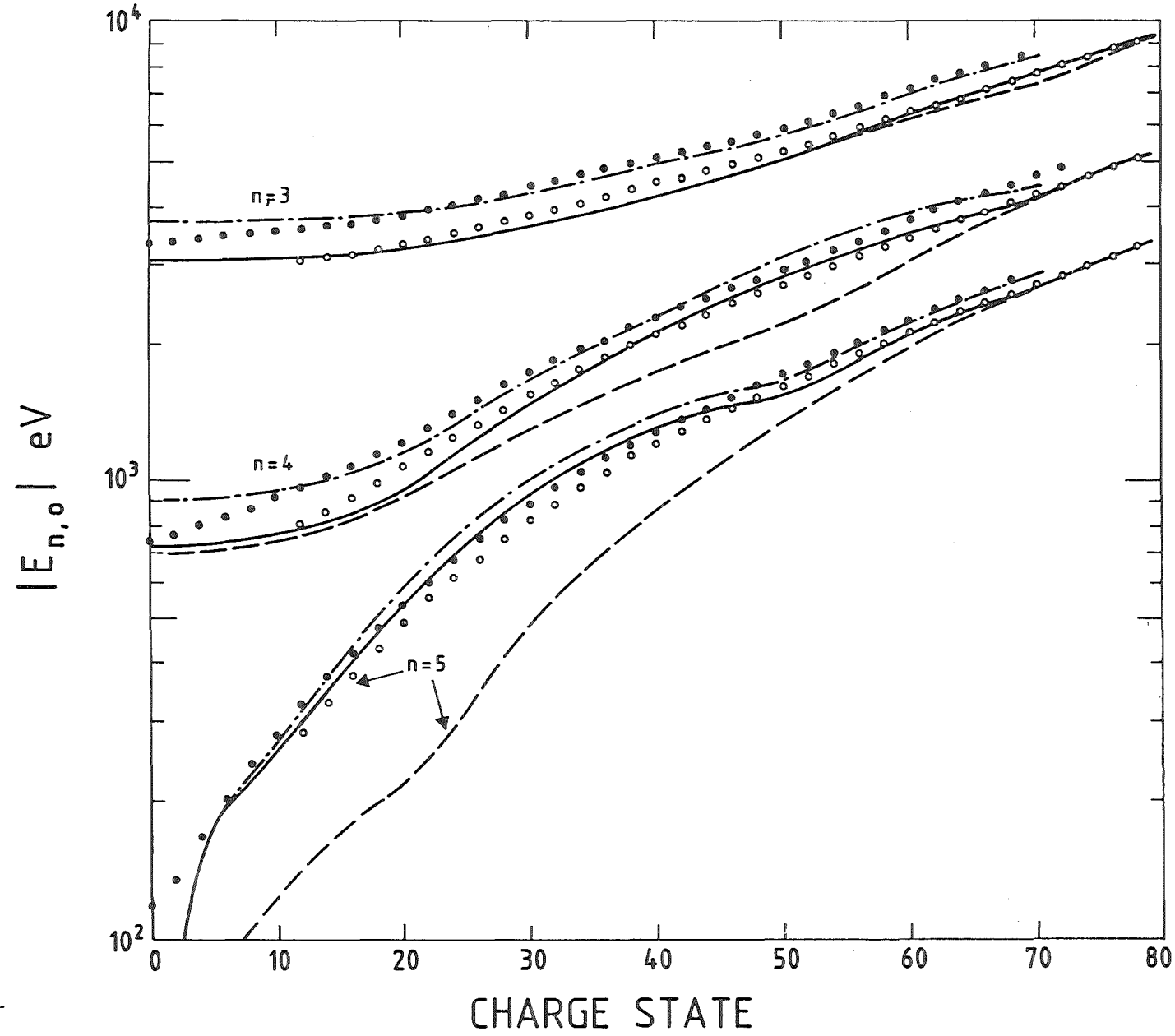


Fig. 1

Level energies of ℓ=0 state for various charge states. Filled circles: RHFS. Open circles: HFS. Dashed lines: modified Parker's model Eqs.(3),(4), and (7) without the relativistic correction ΔE_{RE}. Solid lines: the present model Eqs.(3),(4), and (8) without ΔE_{RE}. Dash-dot lines : the present model with ΔE_{RE}.

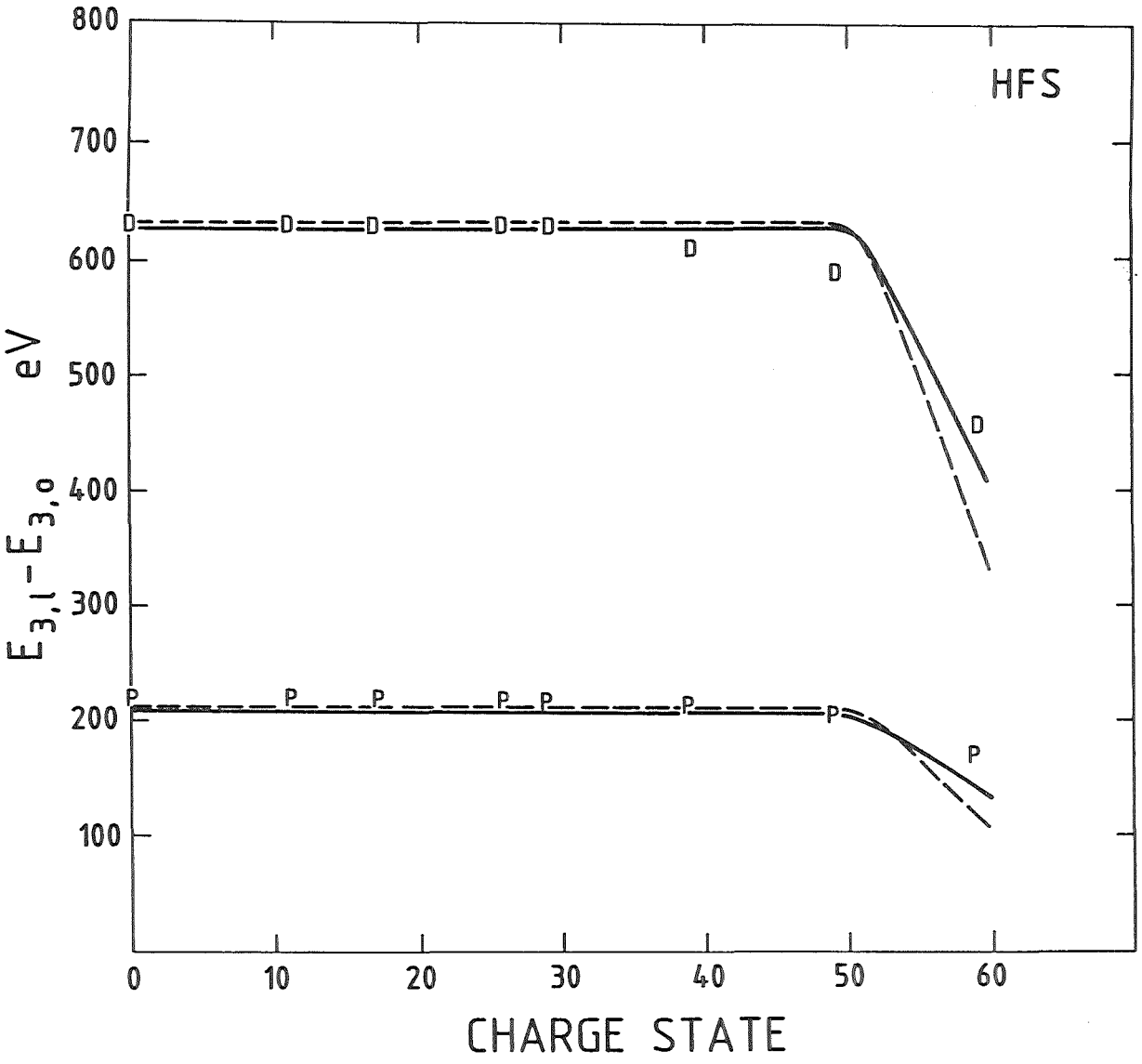


Fig 2(a)

Non-relativistic level energies measured from ℓ=0 state versus charge states for (a) n=3, (b) n=4, and (c) n=5. P,D, F, and G denote the results calculated with HFS for ℓ=1, 2,3, and 4, respectively. The solid and dashed lines represent the results with the present model and the modified Parker's model, repectively.

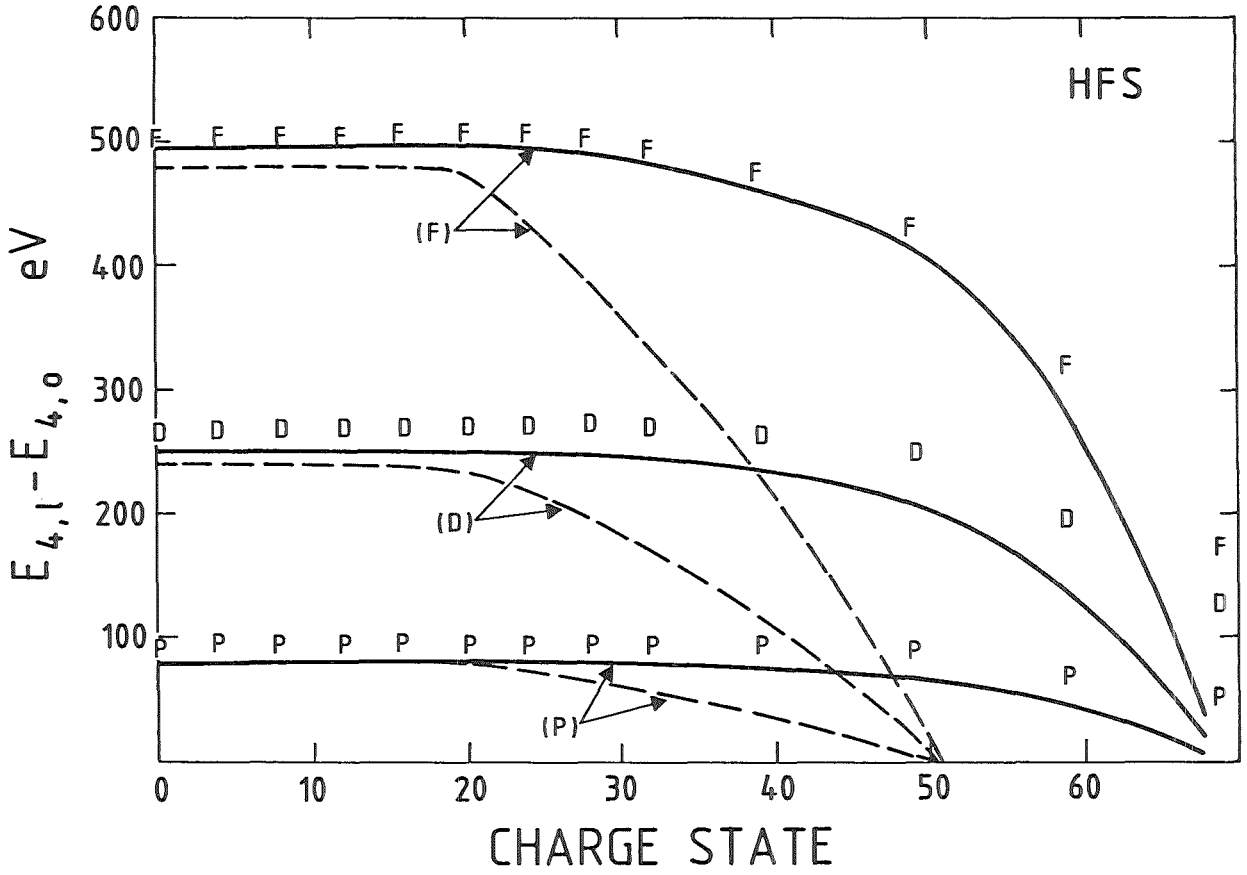


Fig 2(b)

see Figure Caption 2(a)

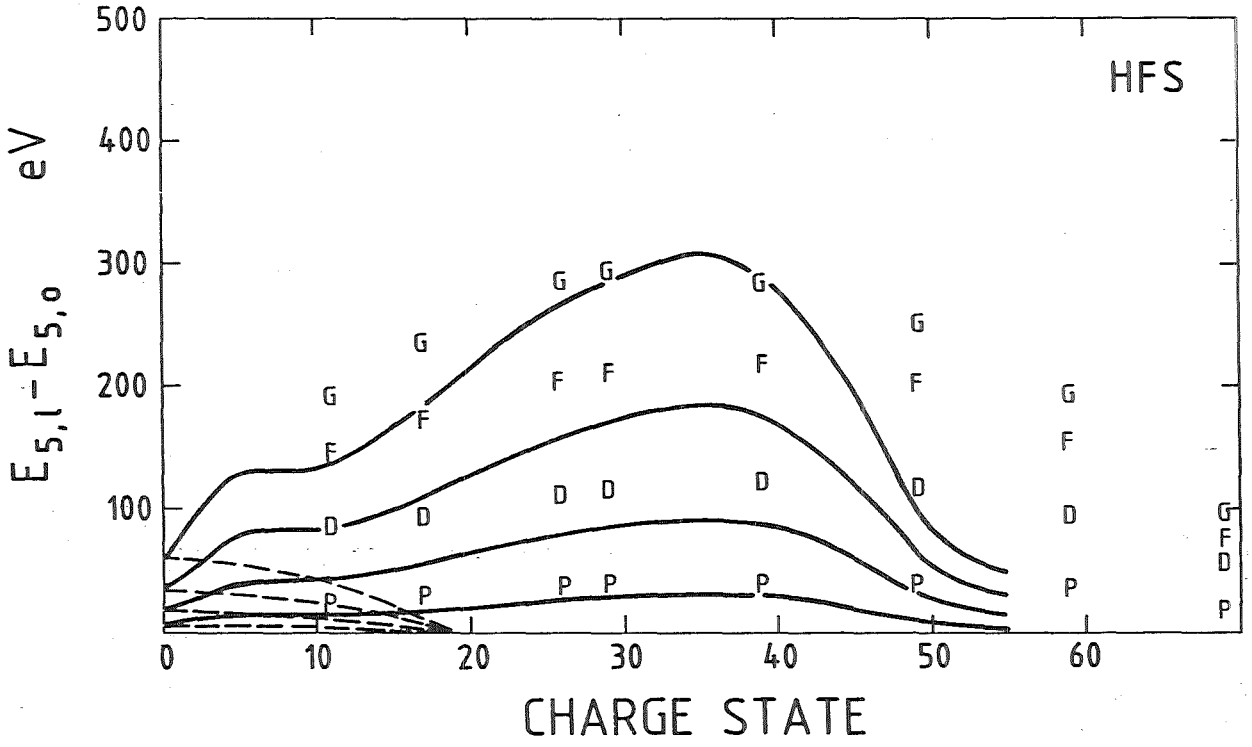


Fig 2(c)

see Figure Caption 2(a)

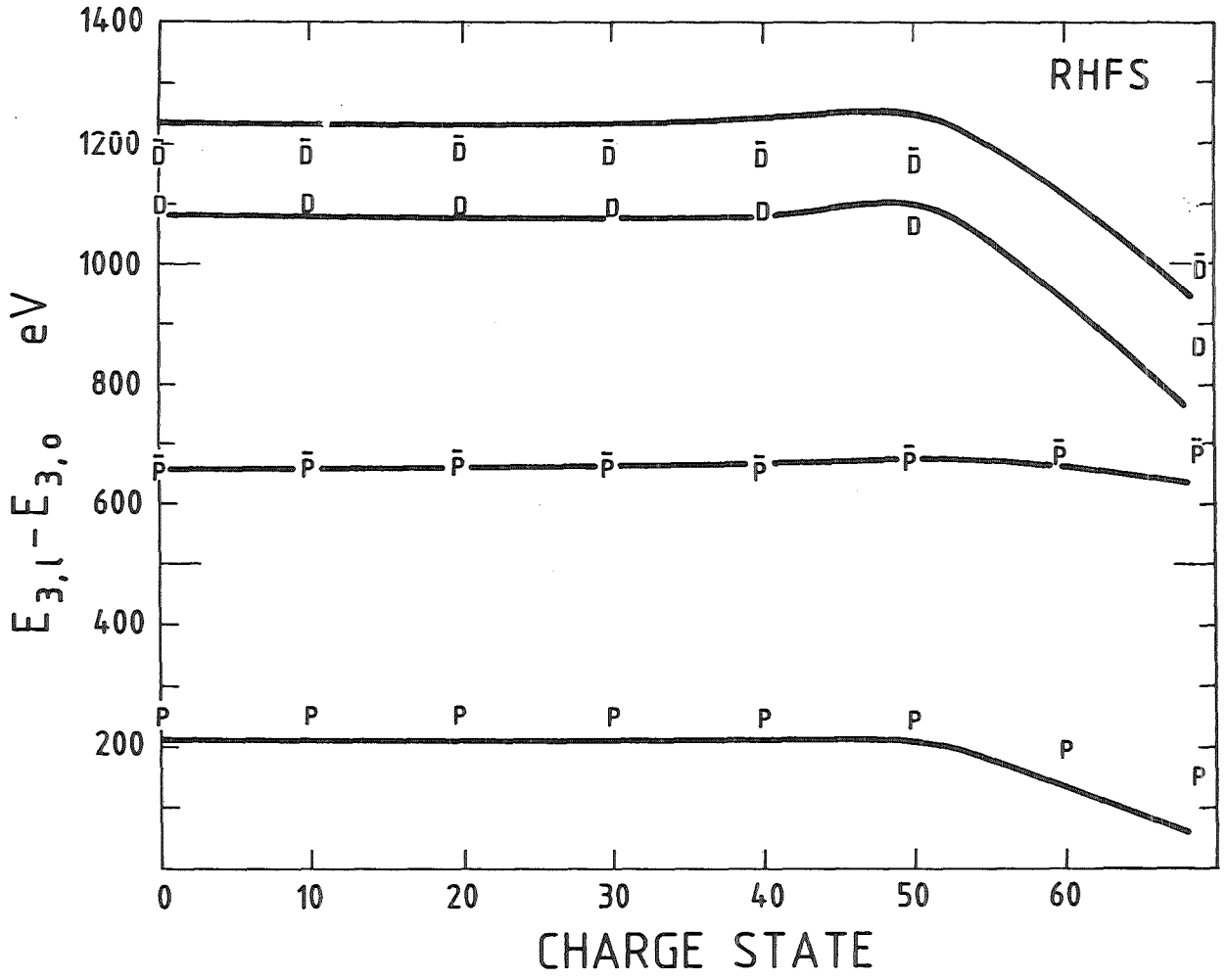


Fig. 3(n)

Relativistic level energies similar to Fig.2 P, $\bar{P}$, D, $\bar{D}$, F, and $\bar{F}$ denote the results with RHFS for $(\ell, j) = (1,1/2)$, $(1,3/2)$, $(2,3/2)$, $(2,5/2)$ $(3,5/2)$, and $(3,7/2)$, respectively. The solid lines are the results with the present model Eqs.(3), (4), (8), and (10); in Eq.(10) Z^*_n is used instead of Z_n. (a) n=3, (b) n=4.

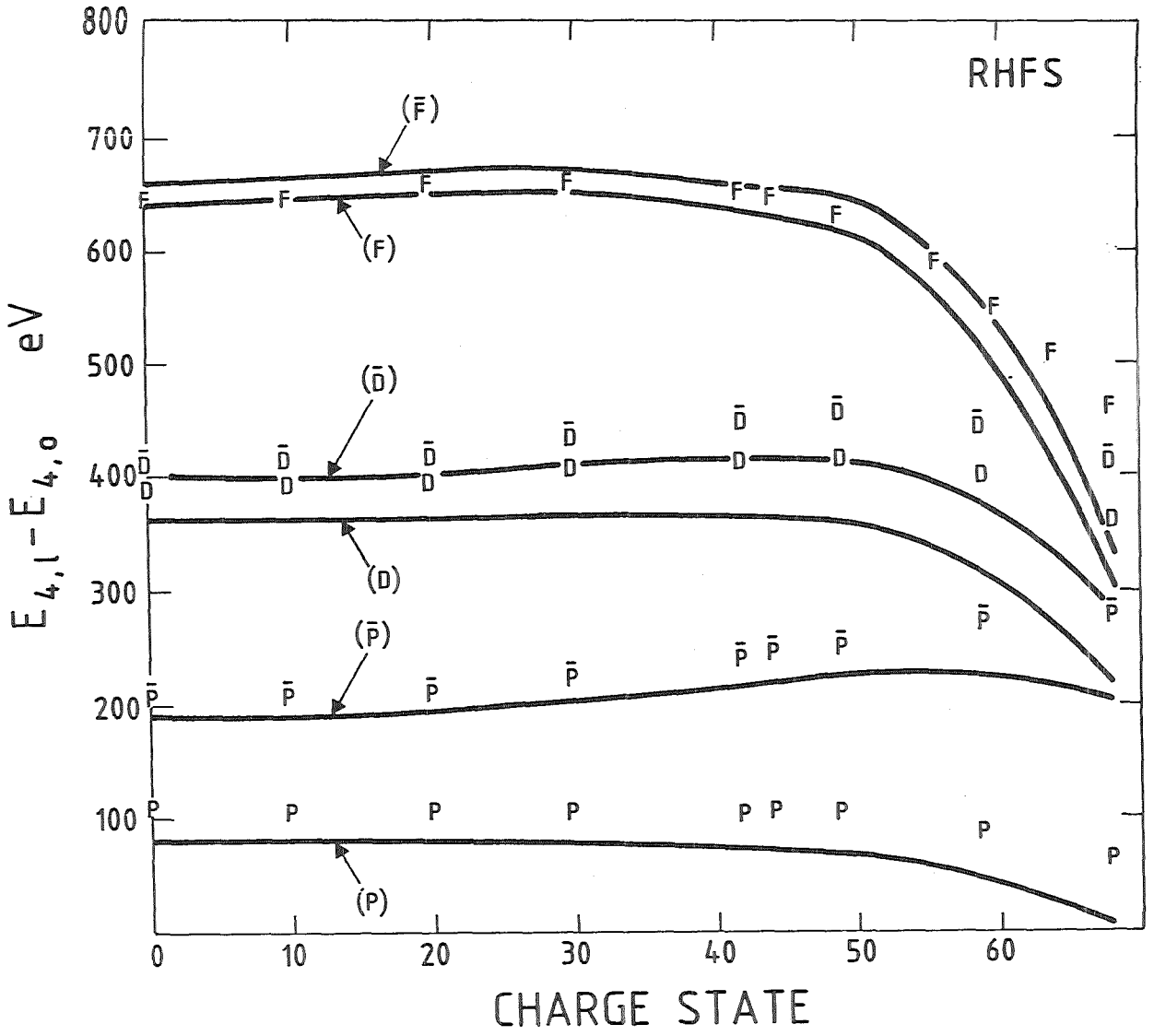


Fig. 3(b)

see Figure Caption 3(a)

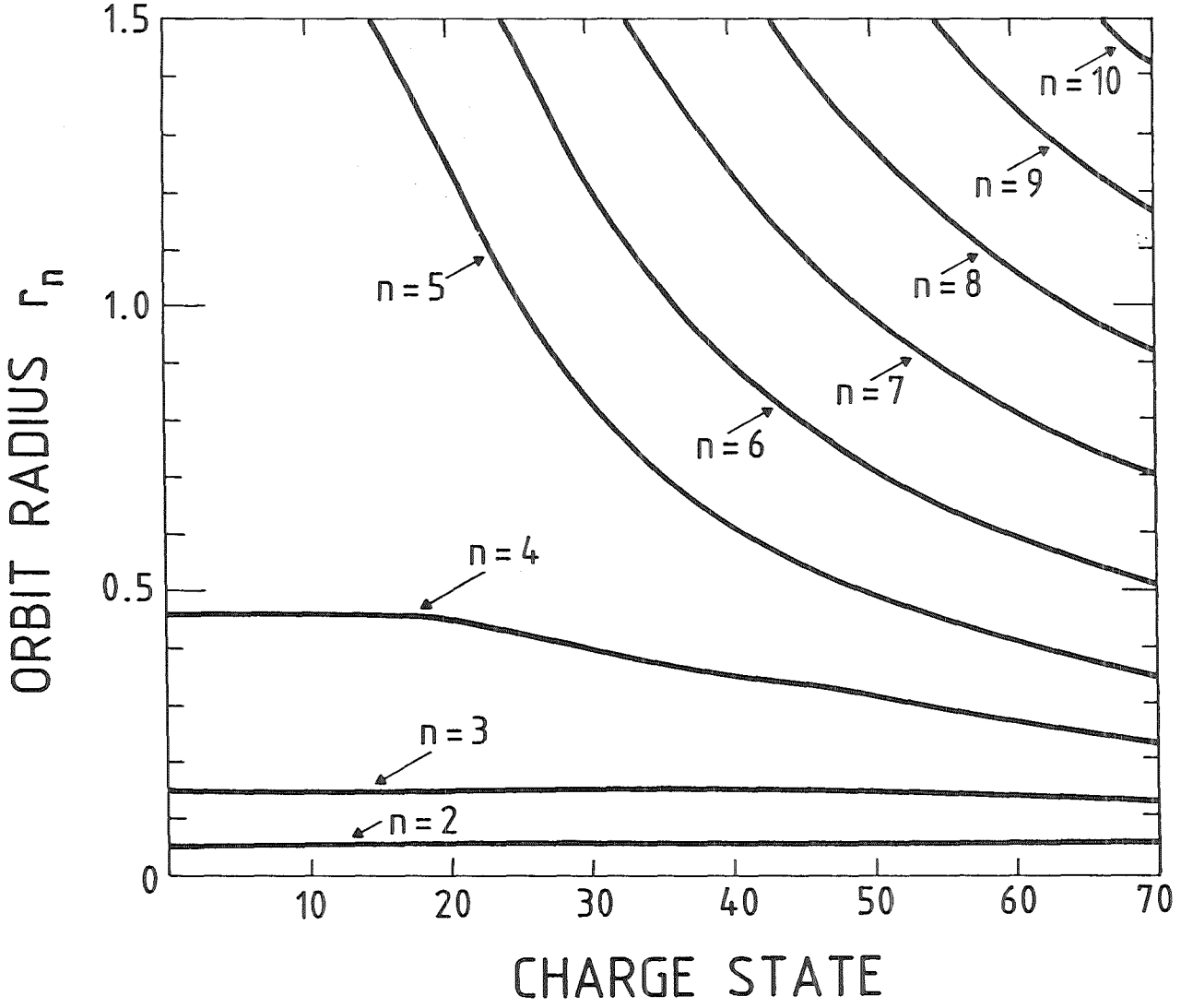
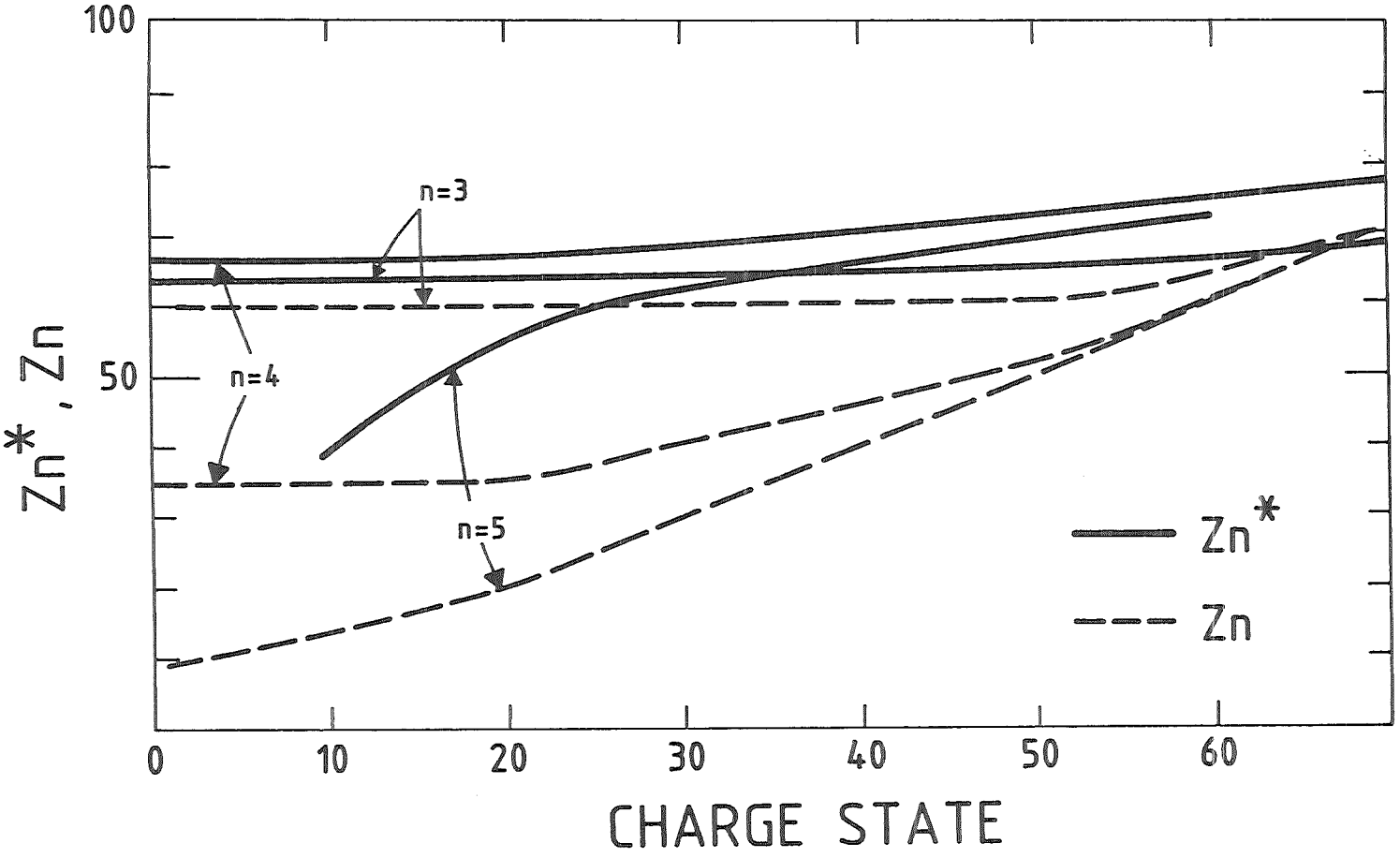


Fig 4

The orbit radii of various shells versus charge states calculated with $r_n = n^2/Z_n$ and Eq.(2).



The dashed lines: the effective charge Z_n calculated with Eq.(2). The solid lines: the effective charge Z^*_n required to get reasonable relativistic corrections with Eq.(10) by fitting it to the difference between RHFS and HFS.

Fig. 5