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**LETTER TO THE EDITOR**

# Strong dependence of multiphoton detachment rates on the asymptotic behaviour of the ground-state wavefunction

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**Abstract.** Two-photon detachment from the $F^-$ negative ion has been investigated within the lowest-order perturbation theory. We show that in accordance with the adiabatic theory a proper asymptotic behaviour of the 2p bound-state wavefunction is crucial for obtaining correct absolute values of the multiphoton detachment cross sections. We find that the latter are substantially higher than was believed previously.

In recent papers by Gribakin and Kuchiev (1997a, b) an adiabatic analytical theory of multiphoton detachment from negative ions has been developed, based on the Keldysh approach (Keldysh 1964). Simple analytical expressions obtained there for the differential and total $n$-photon detachment cross sections allow one to estimate them for any negative ion. One of the important points of that work is that the electron escape from the atomic system in a low-frequency laser field takes place at large distances,

$$r \sim 1/\sqrt{\omega} \sim \sqrt{2n}/\kappa \gg 1 \tag{1}$$

where $\omega$ is the photon frequency, $\kappa$ is related to the initial bound-state energy, $E_0 = -\kappa^2/2$, and $n$ is the number of quanta absorbed (atomic units are used throughout). Accordingly, the multiphoton detachment rates are determined by the long-range asymptotic behaviour of the bound-state wavefunction, namely by the asymptotic parameters $A$ and $\kappa$ of the bound-state radial wavefunction $R(r) \simeq Ar^{-1}e^{-\kappa r}$. This result has been obtained using the length form of the interaction with the laser field, which proved to be the most convenient for multiphoton processes.

The analytical adiabatic approach is valid for *multiphoton* detachment processes, i.e. strictly speaking, for $n \gg 1$. However, the calculations for $H^-$ and halogen negative ions indicate (Gribakin and Kuchiev 1997a, Kuchiev and Ostrovsky 1998) that the analytical formulae should give reasonable answers even for $n = 2$. The aim of the present work is to verify these conclusions by performing direct numerical calculations of the two-photon

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detachment cross sections. In particular, we examine the sensitivity of the photodetachment cross sections to the asymptotic behaviour of the ground-state wavefunction and show that it is indeed very strong. Thus, a 'small' 20% error in $\kappa$ present in the Hartree–Fock (HF) wavefunction of the fluorine negative ion results in a factor of three underestimation of the two-photon cross section. This emphasizes the need to use bound-state wavefunctions with correct asymptotic behaviour in calculations of multiphoton processes.

In this letter we calculate the two-photon detachment amplitudes, cross sections and photoelectron angular distribution using the lowest-order perturbation theory and compare the results obtained with different ground-state wavefunctions. We present and analyse the results for the $F^-$ negative ion, where the results of a few other theoretical calculations (Robinson and Geltman 1967, Crance 1987a, b, Pan *et al* 1990, Pan and Starace 1991, van der Hart 1996) as well as experimental data (Kwon *et al* 1989, Blondel *et al* 1992, Blondel and Delsart 1993) are known. Pan *et al* and Pan and Starace calculated the two-photon detachment cross section and photoelectron angular distribution in the HF approximation (similar to that used by Crance) and taking account of first-order electron correlation effects. Their results show that the correlation corrections are about 15% for the partial and total cross sections and almost negligible for the angular distribution parameters, when the dipole length form is used.

The total cross section of the two-photon detachment of an electron from an atomic system by a linearly polarized light of frequency $\omega$ is

$$\sigma = \sum_{l_f L} \sigma_{l_f L} = \frac{16\pi^3}{c^2}\omega^2 \sum_{l_f L} \left| A_{l_f L}(\omega) \right|^2 \tag{2}$$

where $\sigma_{l_f L}$ is the partial cross section for the detachment into the final state with the photoelectron orbital momentum $l_f$ and the total orbital momentum $L$, and the continuous spectrum wavefunction of the photoelectron is normalized to the $\delta$-function of energy. For the 2p electron detachment from $F^-$ $2p^6\ {}^1S$ the final state can be either ${}^1S$ ($L = 0$, $l_f = 1$) or ${}^1D$ ($L = 2$, $l_f = 1,\ 3$). The two-photon amplitude $A_{l_f L}(\omega)$ is determined by the following equations:

$$A_{l_f L}(\omega) = \sqrt{2L+1} \begin{pmatrix} 1 & 1 & L \\ 0 & 0 & 0 \end{pmatrix} \sum_l (-1)^l \begin{Bmatrix} 1 & 1 & L \\ l_f & l_0 & l \end{Bmatrix} M_{l_f l}(\omega) \tag{3}$$

$$M_{l_f l}(\omega) = \sum_\nu \frac{\langle \varepsilon_f l_f \| \hat{d} \| \nu l \rangle \langle \nu l \| \hat{d} \| n_0 l_0 \rangle}{E_0 + \omega - E_\nu + \mathrm{i}0} \tag{4}$$

where $\nu l$ is the intermediate electron state with the orbital momentum $l$ after absorbing the first photon ($l = 0, 2$ for $F^-$), and $n_0 l_0$ is the initial bound state. The reduced dipole matrix elements are defined in the usual way, for example, in the length form

$$\langle \nu l \| \hat{d} \| n_0 l_0 \rangle = (-1)^{l_>} \sqrt{l_>} \int P_{\nu l}(r)\, P_{n_0 l_0}(r) r\, \mathrm{d}r \tag{5}$$

where $l_> = \max\{l, l_0\}$ and $P$ are the radial wavefunctions.

The photoelectron angular distribution is described by the differential cross section

$$\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega} = \frac{\sigma}{4\pi} \sum_{j=0}^{2} \beta_{2j}(\omega) P_{2j}(\cos\theta) \tag{6}$$

where $\theta$ is measured with respect to the light polarization axis, and the asymmetry parameters $\beta_{2j}$ are defined in terms of the two-photon transition amplitudes $A_{l_f L}$ and scattering phases

of the photoelectron $\delta_{l_f}$:

$$\beta_{2j} = \frac{16\pi^3\omega^2}{c^2\sigma}(4j+1)\mathrm{Re}\Bigg[\sum_{l'_f L' l''_f L''}(-1)^{l_0+L'+L''}(-\mathrm{i})^{l'_f+l''_f}\exp\left[\mathrm{i}(\delta_{l'_f}-\delta_{l''_f})\right]\sqrt{[l'_f][L'][l''_f][L'']}$$
$$\times\begin{pmatrix} l'_f & 2j & l''_f \\ 0 & 0 & 0\end{pmatrix}\begin{pmatrix} L' & 2j & L'' \\ 0 & 0 & 0\end{pmatrix}\begin{Bmatrix} L' & L'' & 2j \\ l''_f & l'_f & l_0\end{Bmatrix}A_{l'_f L'}A^*_{l''_f L''}\Bigg] \qquad (7)$$

where $[l] \equiv 2l+1$ and $\beta_0 = 1$, so that the photoelectron angular distribution after a two-photon detachment is characterized by $\beta_2$ and $\beta_4$.

The self-consistent HF calculation of the $F^-$ ground state yields the 2p-electron energy $E_{2p}^{HF} = -0.362$ Ryd, which is much lower than its true value equal to the negative of the experimental electron affinity of F: $E_{2p}^{exp} = -0.250$ Ryd (Radtsig and Smirnov 1986). It is often assumed that the HF radial wavefunction is still a good starting point for calculations of multiphoton detachment, if the experimental binding energy is used in lieu of the HF value (Crance 1987a, b, Pan *et al* 1990). Pan *et al* (1990) showed that the two-photon detachment cross sections obtained with the dipole operator in the velocity form are very sensitive to the 2p-electron energy, while the length form results change little when the HF energy is replaced by the experimental one. However, one should use both the correct energy and, which is much more important, the bound-state wavefunction with the correct asymptotic behaviour†. The importance of large distances, where one can use the correct asymptotic form of the bound-state wavefunction, speaks strongly in favour of using the length form of the dipole operator (Gribakin and Kuchiev 1997a). To correct the 2p wavefunction we solved the HF equations for the $F^-$ ground state with an additional small repulsive potential $V(r) = \alpha/[2(r^2+a^2)^2]$. We chose $\alpha = 1$ and $a = 0.61$ au to ensure that the 2p energy was equal to the experimental value. The HF and corrected 2p radial wavefunctions $P(r) = R(r)/r$ are presented in figure 1(*a*). The difference between them appears to be small—it does not exceed 10% near the maximum. Their asymptotic behaviour $P(r) \simeq A\exp(-\kappa r)$ corresponds to $A = 0.94$ and 0.86, and $\kappa = 0.6$ and 0.5, respectively. The difference in $\kappa$ means that the two wavefunctions are, in fact, quite different at large distances.

The wavefunctions of the intermediate ($\nu l$) and final ($\varepsilon_f l_f$) states of the photoelectron are calculated in the HF field of the frozen neutral $2p^5$ core. The photoelectron is coupled to the core to form the total spin $S = 0$ and angular momentum $L$: $L = 1$ for the intermediate $l = 0$, 2 states, $L = 0$, 2 for $l_f = 1$, and $L = 2$ for $l_f = 3$ final states. The intermediate states continua are discretized and represented by a 70-state momentum grid with constant spacing $\Delta p$.

There are two ways of calculating the two-photon amplitudes $M_{l_f l}$ of equation (4). The first is by direct summation over the intermediate states. It involves a non-trivial evaluation of the free–free dipole matrix elements together with the accurate treatment of pole- and $\delta$-type singularities (Korol 1994, 1997). Another way of calculating such sums is by solving an inhomogeneous Schrödinger-type equation for the effective radial function of the intermediate state

$$P_\omega(r) = \sum_\nu \frac{P_{\nu l}(r)\langle \nu l\|\hat{d}\|n_0 l_0\rangle}{E_0+\omega-E_\nu+\mathrm{i}0} \qquad (8)$$

† The need for an asymptotically correct wavefunction is clearly illustrated by the adiabatic hyperspherical calculation of multiphoton detachment from $H^-$ by Liu *et al* (1992), where a 3.4% change of $\kappa$ results in a 25% change of the two-photon cross section.

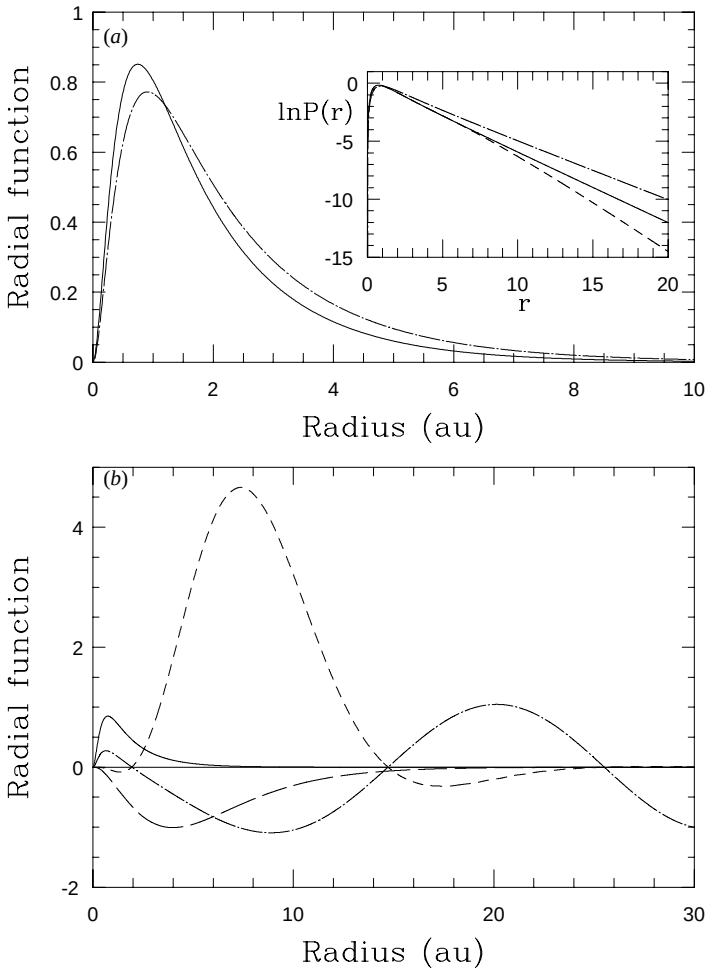


**Figure 1.** Wavefunctions of the $F^-$ ground state, effective intermediate state and final state of the photoelectron. (*a*) Radial wavefunction of the 2p subshell of $F^-$ in the HF approximation (——, $E_{2p}^{HF} = -0.362$ Ryd), and that with a model potential added to reproduce the experimental energy (— · —, $E_{2p} = -0.250$ Ryd). The inset shows the same wavefunctions on the logarithmic scale, together with the radial Roothaan-HF 2p radial wavefunction of $F^-$ from Clementi and Roetti (1974), - - - -. (*b*) ——, HF 2p wavefunction; — — —, effective wavefunction $P_\omega(r)$, equation (8), of the intermediate d state at $\omega = 0.226$ Ryd; — · —, final state p ($^1$D) wavefunction, $\varepsilon = 0.09$ Ryd; - - - -, integrand of equation (9) for the two-photon amplitude $M_{pd}$.

(Sternheimer 1951, Dalgarno and Lewis 1955). This wavefunction describes the amplitude of finding the electron at different distances from the atom after absorption of the first quantum. After calculation of $P_\omega(r)$ the amplitude is obtained from the radial integral as

$$M_{l_f l}(\omega) = (-1)^{l_>}\sqrt{l_>}\int P_{\varepsilon_f l_f}(r)\, P_\omega(r)\, r\, \mathrm{d}r \qquad \left(l_> = \max\{l_f, l\}\right). \tag{9}$$

In the present work we calculate the two-photon amplitudes using both techniques. The second one is especially simple below the single-photon detachment threshold, $\omega < |E_0|$, where $P_\omega(r)$ drops exponentially at large distances: $P_\omega(r) \propto \exp(-K_\omega r)$, where $K_\omega =$

$[2(|E_0|-\omega)]^{1/2}$. At finite distances $r < 1/\Delta p$ it can be computed easily by direct numerical summation in equation (8).

It is instructive to look at the shape of the effective wavefunction $P_\omega(r)$. As an example, figure 1(*b*) shows this function calculated for the intermediate d electron at $\omega = 0.226$ Ryd for the HF ground state. The maximum of $P_\omega(r)$ is shifted towards large radii, compared to the maximum of the ground-state wavefunction. Also shown in figure 1(*b*) are the radial wavefunction of the final p-wave electron ($\varepsilon_f = 2\omega + E_{2p}^{HF} = 0.09$ Ryd) and the integrand $P_{\varepsilon_f l_f}(r)\, P_\omega(r)\, r$ of equation (9). These plots illustrate the point that the two-photon amplitude $M_{l_f l_n}(\omega)$ is indeed determined by large electron–atom separations (1). Accordingly, the correct asymptotic behaviour of the ground-state wavefunction is crucial.

In this work we compare the cross sections and angular asymmetry parameters calculated in different approximations with both the HF and corrected 2p wavefunctions (figure 1(*a*)). Let us first discuss the results obtained with the HF energy of the 2p electron. It corresponds to the two-photon threshold $\omega = 0.181$ Ryd. The cross section calculated from equations (2)–(4) using the HF functions of the initial, intermediate and final states are shown in figure 2 by a short-broken curve. It is very similar to the dipole length lowest-order HF results of Pan *et al* (1990), although the latter are about 10% lower than ours. What

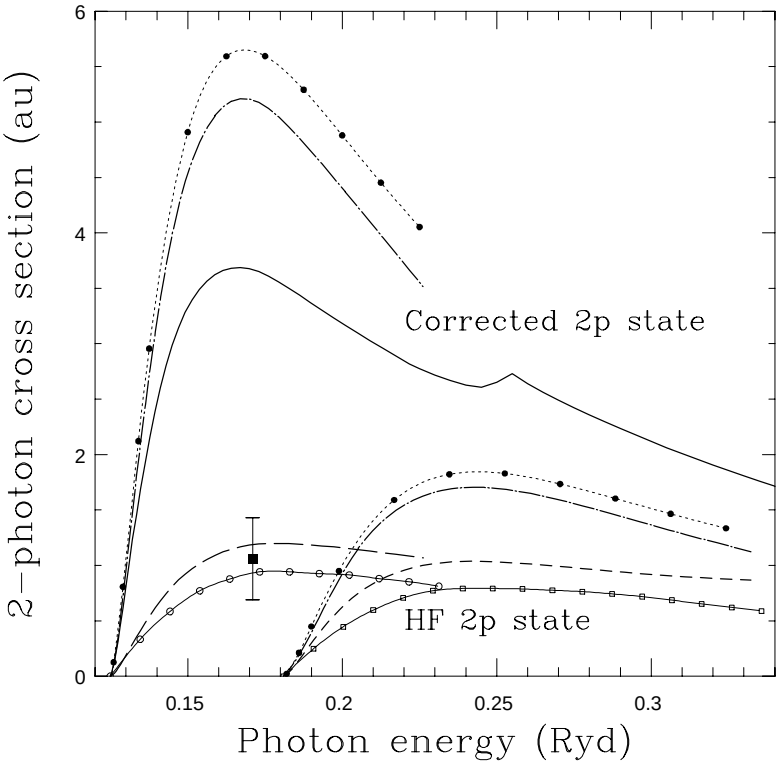


**Figure 2.** Two-photon detachment cross sections. Present calculations: - - - -, HF wavefunctions of the 2p, intermediate and final states; ——, same with the corrected 2p wavefunction; — · —, using plane waves in the intermediate and final state; — — —, HF wavefunctions combined with the experimental 2p energy; ●, adiabatic theory (equation (5) of Gribakin and Kuchiev 1997b). Other results: □ and ○, calculations by Pan *et al* (1990) with the HF and experimental binding energies, respectively; ■, experiment (Kwon *et al* 1989).

is the source of this discrepancy? Pan *et al* used the Roothaan-HF expansion of the bound state. This form of the bound-state wavefunction has an incorrect asymptotic behaviour at $r > 7$, see figure 1($a$), inset. Because of the importance of large electron–atom separations in the multiphoton processes even a small error in the wavefunction could lead to some inaccuracies in the two-photon detachment amplitudes. In the work of Pan *et al* electron correlation effects were calculated. It was shown that they suppress the cross section in $F^-$ by about 20% at the maximum. These results shown in figure 2 by open squares are still close to the HF curve.

The asymptotic parameters of the HF 2p wavefunction are $\kappa = 0.6016$ and $A = 0.94$. We use them in the adiabatic theory formula (equation (5) of Gribakin and Kuchiev 1997b) and obtain the cross section shown in figure 2 by full circles. It reproduces the energy dependence of the HF cross section well, though overestimates its magnitude by a factor of two. This is a reasonable result, since the adiabatic theory should only be valid for $n \gg 1$. The calculations of Gribakin and Kuchiev (1997a) showed that for $H^-$ and $n = 3$ the analytical adiabatic results are already 20% accurate. There are two approximations made in the adiabatic theory: (i) the use of the Volkov wavefunction to describe the photoelectron and (ii) the saddle-point calculation of the integral over time, which enables one to express the amplitude in terms of the asymptotic parameters of the bound state. In the weak-field regime the use of the Volkov function is equivalent to the so-called 'free-electron' approximation (examined earlier by Crance). In this approximation the wavefunctions of the photoelectron in the intermediate and final states are described by plane waves. When we perform such a calculation for $F^-$ (chain curve in figure 2) the results turn out to be very close to those of the adiabatic theory. This means that the approximation (ii) of the adiabatic theory is, in fact, quite good even at $n = 2$.

When we use the experimental energy of the 2p electron together with the HF wavefunctions, the magnitude of the two-photon cross section changes very little (broken curve in figure 2), as seen earlier by Pan *et al* (1990) for both HF and correlated results (open circles). The HF results of Crance (1987a) are close to the above, and the cross section of van der Hart (1996) is also similar, with a maximum of 1.27 au at $\omega = 0.166$ Ryd.

However, when we use the corrected 2p wavefunction, the photodetachment cross section increases more than three times. It is shown by the full curve in figure 2, and we consider this to be the best evaluation of the cross section for $F^-$. The cusp on the curve corresponds to the single-photon threshold†. The same increase is also demonstrated by the adiabatic theory (with modified asymptotic parameters $\kappa = 0.4998$ and $A = 0.86$) and the plane-wave results. As we explained earlier, this 'surprising' sensitivity of the multiphoton detachment probabilities to the asymptotic form of the bound-state wavefunction is a direct consequence of the dominant role of large electron–atom separations in this problem (Gribakin and Kuchiev 1997a).

The error induced by the use of an asymptotically incorrect wavefunction can be estimated within the adiabatic approach. It turns out that if one uses the experimental binding energy $E_0 = -\kappa^2/2$ together with an incorrect bound state $P(r) \propto \exp(-\kappa' r)$, the $n$-photon cross section acquires an error factor

$$\left(1 - \sqrt{\frac{\pi}{2}}\frac{\Delta\kappa}{\sqrt{\omega}}\right)^2 \tag{10}$$

where $\Delta\kappa = \kappa' - \kappa$. This equation implies that the relative error in the amplitude is $\sim \Delta\kappa R$, where $R = 1/\sqrt{\omega}$ is the large radius from equation (1). For $\kappa' > \kappa$ the error factor is

† This feature is a consequence of the Wigner threshold dependence $\sigma \propto \sqrt{\omega - E_0}$ of the s-wave single-photon detachment from $F^-$.

smaller than unity. Thus, using a stronger bound wavefunction leads to an underestimation of the cross section. For $F^-$ the factor (10) calculated for $\Delta\kappa = 0.1$ and $\omega = 0.085$ au near the cross section maximum, yields 0.33. This value agrees with the difference between the cross sections observed in figure 2. The only other work that used an asymptotically correct 2p wavefunction was the model potential calculation of Robinson and Geltman (1967), which produced a cross section twice as large as those of Crance, Pan *et al* and van der Hart.

To estimate the size of possible errors introduced by our way of correcting the 2p wavefunction we have examined the dependence of our cross section on the choice of $\alpha$ and $a$ in the repulsive potential. We find that as long as the asymptotic behaviour of the 2p state remains correct, the two-photon cross sections are always proportional to that shown by the full curve in figure 2. Different pairs of $\alpha$ and $a$ result in the variation of $A$, and the magnitude of the cross section is simply proportional to $A^2$. Our value of $A = 0.86$ is close to $A = 0.84$ from Radtsig and Smirnov (1986) and we are sure that our results are basically correct. Even a large 10% uncertainty in the value of $A$ would mean a maximal 20% error in the cross section. In any case the cross section will be much larger than those obtained with the HF 2p ground state.

The difference between experimental and HF values of the 2p energy is a manifestation of electron correlations. It influences the result via the asymptotic behaviour of the ground-state wavefunction. This is by far the most important correlation effect in multiphoton detachment. The use of the asymptotically correct 2p wavefunction changes the cross section by a factor of three, which is much greater than other correlation effects (Pan *et al* 1990). This fact distinguishes this problem from the single-photon processes, where other correlation effects are essential.

The angular asymmetry parameters $\beta_2$ and $\beta_4$ calculated using the experimental 2p energy are shown in figure 3, together with the correlated length results of Pan and Starace (1991) and experimental points of Blondel and Delsart (1993) at $\omega = 0.171$ Ryd. The asymmetry parameters (7) are relative quantities, and the results of different calculations are much closer for them than for the absolute values of the photodetachment cross sections. The adiabatic theory is again in good agreement with the plane-wave approximation, especially in $\beta_4$. It appears that this parameter is on the whole less sensitive to the details of the calculation, because it is simply proportional to the amplitude of f-wave emission, and there is no interference in the sum in equation (7) for $\beta_4$. The experimental values of $\beta$ for $F^-$ obtained in the earlier work of Blondel *et al* (1992) are close to those of Blondel and Delsart (1993). This is why $F^-$ serves as a good benchmark for angular asymmetry calculations. The perfect agreement between adiabatic theory and the experiment is probably fortuitous. Figure 3 indicates that for the experiment to be able to distinguish between various theoretical data one would wish to make measurements at higher photon energies, where the results of different approximations diverge.

From the theoretical point of view it seems that the total cross sections and the angular asymmetry parameters are determined by different physical features of the problem. The absolute size of the cross sections is very sensitive to the asymptotic behaviour of the bound-state wavefunction. This sensitivity *increases* for large-$n$ processes, when $\omega$ become smaller, as suggested by estimate (10). The cross sections also depend on the atomic potential which acts on the photoelectron, hence the difference between the results obtained with the HF and plane waves. This latter effect is *suppressed* for larger $n$, since this re-scattering of the photoelectron is inversely proportional to some power of large $R$. The angular asymmetry parameters are affected by the electron–atom potential via the scattering phaseshifts $\delta_{l_f}$. However, for large $n$ and small photoelectron energy $E \sim \omega$ the phaseshifts should be close

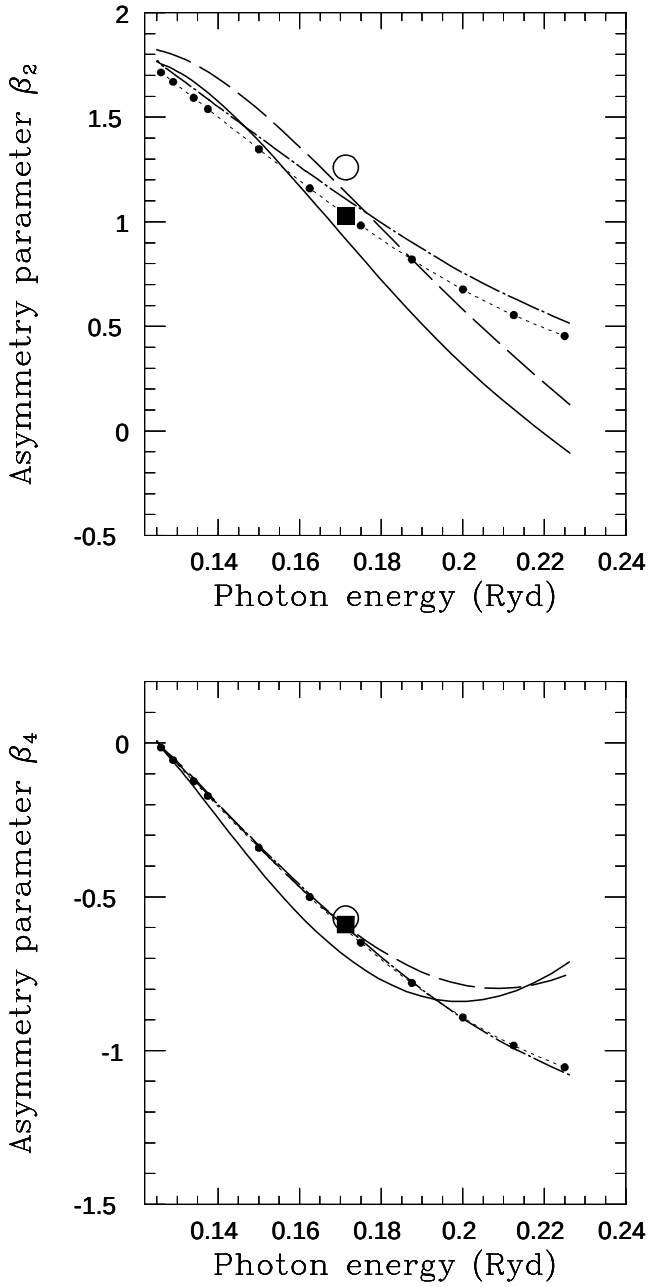


**Figure 3.** Photoelectron angular distribution parameters. Present calculation: — — —, HF wavefunctions of the initial, intermediate and final states, experimental 2p energy; ——, corrected 2p wavefunction, HF intermediate and final states; — · —, same with the plane wave in the intermediate and final states; ●, $\beta$ parameters obtained from the adiabatic theory (equations (3) and (4) of Gribakin and Kuchiev (1997b)). Other results: ○, correlated length results by Pan and Starace (1991); ■, experiment (Blondel and Delsart 1993).

to integer multiples of $\pi$. Also, contributions of higher partial waves become dominant. They are almost unaffected by the atomic potential and the adiabatic theory should become very accurate.

In summary, we have shown by direct numerical calculations that in agreement with the adiabatic theory, the multiphoton detachment rates are very sensitive to the asymptotic behaviour of the bound-state wavefunction. For fluorine this means that the true two-photon detachment cross sections are substantially higher than was believed previously. The discrepancy revealed is much greater than other electron correlation effects.

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