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Three-photon detachment of electrons from the fluorine negative ion

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Abstract. Absolute three-photon detachment cross sections are calculated for the fluorine negative ion within the lowest-order perturbation theory. The Dyson equation of the atomic many-body theory is used to obtain the ground-state 2p wavefunction with correct asymptotic behaviour, corresponding to the true (experimental) binding energy. We show that in accordance with the adiabatic theory (Gribakin and Kuchiev 1997 *Phys. Rev.* A **55** 3760) this is crucial for obtaining absolute values of the multiphoton cross sections. Comparisons with other calculations and experimental data are presented.

1. Introduction

Starting from the pioneering works of Hall *et al* (1965) and Robinson and Geltman (1967) the behaviour of negative ions in laser fields has been the subject of numerous studies for over thirty years. Nevertheless, up to now there have been very few firmly established results on the absolute values of the cross sections and photoelectron angular distributions in multiphoton processes.

This is true even for the simplest two-photon detachment processes. For example, the results of a number of experimental and theoretical works on the cross sections and photoelectron angular distributions in the negative halogen ions (see, e.g., van der Hart 1996, Gribakin *et al* 1999 and references therein) differ significantly from each other. A number of experimental works reported the cross sections and angular asymmetry parameters of the two-photon detachment from the halogen negative ions at selected photon energies (Trainham *et al* 1987, Blondel *et al* 1989a, 1992, Kwon *et al* 1989, Davidson *et al* 1992, Sturrus *et al* 1992, Blondel and Delsart 1993). These measurements were performed from the end of the 1980s to the beginning of the 1990s, and to the best of our knowledge no new experimental data on multiphoton detachment from the negative halogens have been published since.

On the theoretical side, a recent development in the study of multiphoton detachment from negative ions has been done within the adiabatic approach (Gribakin and Kuchiev 1997a, b).

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It has established that the electron escape from an atomic system in a low-frequency laser field takes place at large electron–atom separations,

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

where ω is the photon frequency, κ 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). Therefore, the asymptotic behaviour of the bound-state radial wavefunction

$$R(r) \simeq Ar^{-1}\mathrm{e}^{-\kappa r} \tag{2}$$

is crucial for obtaining correct absolute values of the probabilities of multiphoton processes. Direct calculations of two-photon detachment from halogen negative ions within the lowest-order perturbation theory (Gribakin *et al* 1998, 1999) with both the Hartree–Fock (HF) and the asymptotically correct valence np wavefunctions confirm this understanding. The point is that the HF wavefunctions are characterized by κ values generally exceeding the true experimental ones. As a result, when we use asymptotically correct wavefunctions the cross sections are significantly higher than those obtained with other methods which rely on the HF or similar ground-state wavefunctions (Crance 1987, 1988, Jiang and Starace 1988, Pan *et al* 1990, van der Hart 1996).

The strong dependence of multiphoton detachment cross sections on the binding energy was noticed earlier by Liu *et al* (1992). They evaluated it using the zero-range potential model and showed that the n-photon cross section for linear polarization in the near-threshold region changes as $\Delta\sigma_n/\sigma_n \simeq -2n\Delta\omega/\omega$ for even n, and $\Delta\sigma_n/\sigma_n \simeq -(2n+1)\Delta\omega/\omega$ for odd n. The value of $\Delta\omega$ is related to the change in the binding energy. The minus sign means that bound states with smaller binding energies have larger multiphoton detachment cross sections. A similar estimate also follows from Gribakin and Kuchiev (1997b). Moreover, equation (5) of the latter work can be used to estimate the cross section maximum at photoelectron momentum $p \sim \sqrt{\omega}$. In this case we obtain

$$\frac{\sigma_n'}{\sigma_n} = \left(\frac{A'}{A}\right)^2 \left(\frac{\omega}{\omega'}\right)^{2n-1/2}, \tag{3}$$

where the two cross sections σ_n and σ_n' describe n-photon detachment from the negative ions with different binding energies $|E_0|$ and $|E_0'|$, hence $\omega/\omega' = E_0/E_0' \neq 1$. Equation (3) also allows for different A in the asymptotic region (2). Of course for $n \gg 1$ the frequency factor in (3) is much more important than A'/A. Note that in the zero-range potential model the normalization is fixed by $A = \sqrt{2\kappa}$, and instead of equation (3) one simply obtains $\sigma_n'/\sigma_n = (\omega/\omega')^{2n-1}$. However, for not large n the normalization factor $(A'/A)^2$ may be important, as the results discussed below show.

Adiabatic theory reveals that this sensitivity is caused by the role of the exponential tail of the bound-state wavefunction. It is common in multiphoton calculations to use experimental values of the binding energies. However, if this is not accompanied by correcting the asymptotic behaviour of the bound state, a large error can be introduced. Moreover, the use of the ground-state wavefunctions with correct asymptotic behaviour in multiphoton detachment calculations is often more important than other effects of electron correlations.

As far as three-photon detachment from negative ions is concerned, the experimental and theoretical results are more scarce than those on the two-photon detachment. Thus, there have been only two experimental measurements of the cross section for F^- at a single photon energy performed by Blondel *et al* (1989b) and Kwon *et al* (1989), and a few theoretical values obtained in the early calculations by Crance (1987, 1988). Recently, van der Hart (1996) applied an R-matrix Floquet approach to study the photodetachment from F^- and Cl^- for $n = 1$, 2 and 3.

The aim of this work is to perform direct numerical calculations of the three-photon detachment cross section for the negative fluorine ion using an asymptotically correct ground-state wavefunction and compare the results with the available theoretical and experimental data. As in our previous two-photon calculations (Gribakin *et al* 1999) the correct 2p wavefunction is obtained within the many-body Dyson equation method. Section 2 outlines briefly the method of calculation. A discussion of our results and comparisons with other calculations and experimental data are presented in section 3.

2. Three-photon detachment cross section

The total cross section of three-photon detachment of the n_0l_0 electron from an atomic system by a linearly polarized light of frequency ω can be written as

$$\sigma_3(\omega) = \sum_{l_f,L} \sigma_{l_f L} = \frac{32\pi^4\omega^3}{c^3} \sum_{l_f,L} |B^{(L)}_{l_0,l_f}(\omega)|^2. \tag{4}$$

In this sum above the partial cross sections $\sigma_{l_f L}$ are characterized by the orbital momentum l_f of the final-state photoelectron coupled with the atomic residue into the total orbital momentum L. The second equality assumes that the continuous-spectrum wavefunction of the photoelectron in the matrix element $B^{(L)}_{l_0l_f}(\omega)$ is normalized to the δ-function of energy. After absorption of three dipole photons by an outer np electron in a halogen negative ion $n\mathrm{p}^6\ {}^1\mathrm{S}$, the final-state photoelectron can leave the system in the s-, d- or g-waves. So, the possible final states are: $l_f = 0$ (^{1}P), $l_f = 2$ (^{1}P and ^{1}F) and $l_f = 4$ (^{1}F).

In the lowest perturbation-theory order the three-photon amplitude $B^{(L)}_{l_0l_f}(\omega)$ is characterized by the following sequence of electronic states, $n_0l_0(L_0) \to n_1l_1(L_1) \to n_2l_2(L_2) \to E_fl_f(L)$, produced by successive absorption of three photons. This amplitude may be presented as

$$B^{(L)}_{l_0l_f} = \sum_{L_2l_2} \sqrt{(2L_2+1)(2L+1)} \begin{pmatrix} 1 & L & L_2 \\ 0 & 0 & 0 \end{pmatrix} \begin{Bmatrix} 1 & L & L_2 \\ l_0 & l_2 & l_f \end{Bmatrix} \times \sum_{E_2} \frac{\langle \varepsilon_f l_f \| \hat{d} \| n_2 l_2 \rangle A^{L_2}_{l_0l_2}(\omega, E_0, E_2)}{2\omega - E_2 + E_0 + \mathrm{i}\delta}, \tag{5}$$

where n_2l_2 is the intermediate electron state after the absorption of the second photon, l_2 is the electron's orbital momentum and L_2 is the total orbital momentum of the system in the intermediate state. For a halogen negative ion $l_2 = 1$ with $L_2 = 0, 2$ and $l_2 = 3$ with $L_2 = 2$. In equation (5) and below E_0, E_1, E_2, and E_f are energies of the corresponding electron states. The amplitude $A^{L_2}_{l_0l_2}(\omega, E_0, E_2)$ in equation (5) is the two-photon amplitude (cf Pan *et al* 1990, Gribakin *et al* 1999),

$$A^{L_2}_{l_0l_2}(\omega, E_0, E_2) = \sqrt{2L_2+1} \begin{pmatrix} 1 & L_2 & 1 \\ 0 & 0 & 0 \end{pmatrix} \sum_{l_1} (-1)^{l_1} \begin{Bmatrix} 1 & 1 & L_2 \\ l_2 & l_0 & l_1 \end{Bmatrix} M^{L_2}_{l_0l_1l_2}(\omega, E_0, E_2), \tag{6}$$

where the two-photon radial matrix element $M^{L_2}_{l_0l_1l_2}(\omega, E_0, E_2)$ is given by

$$M^{L_2}_{l_0l_1l_2}(\omega, E_0, E_2) = \sum_{E_1} \frac{\langle n_2 l_2 \| \hat{d} \| n_1 l_1 \rangle \langle n_1 l_1 \| \hat{d} \| n_0 l_0 \rangle}{\omega + E_0 - E_1 + \mathrm{i}\delta}. \tag{7}$$

The sums in equations (6) and (7) run over the intermediate electron states n_1l_1 populated after the absorption of the first photon ($l_1 = 0, 2$ with $L_1 = 1$ for the halogen negative ions). The

reduced dipole matrix elements are defined in the usual way, e.g. in the length form,

$$\langle nl\|\hat{d}\|n_0l_0\rangle = (-1)^{l_>}\sqrt{l_>}\int P_{nl}(r)\,P_{n_0l_0}(r)r\,\mathrm{d}r, \tag{8}$$

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

If one describes the initial state n_0l_0 in the HF approximation, the asymptotic behaviour of the corresponding radial wavefunction is incorrect. Namely, it is characterized by κ corresponding to the HF binding energy, rather than the exact (experimental) one. Thus, in F^- the HF value is $\kappa = 0.6$, whereas the true one is $\kappa = 0.5$. As we showed for the two-photon detachment (Gribakin *et al* 1998, 1999), it is very important to use asymptotically correct bound-state wavefunctions. In this paper we refine the bound-state wavefunction using the Dyson equation method in the same way as it is done in our two-photon calculations (Gribakin *et al* 1999). The bound-state orbital $P_{n_0l_0}(r)$ is found from

$$\hat{H}^{(0)}P_{n_0l_0}(r) + \int \Sigma_{E_0}(r, r')P_{n_0l_0}(r')\,\mathrm{d}r' = E_0P_{n_0l_0}(r), \tag{9}$$

where $H^{(0)}$ is the single-particle HF Hamiltonian of the negative ion, and $\Sigma_E(r, r')$ is the self-energy of the electron's single-particle Green function. This energy-dependent non-local operator describes electron correlation effects, and, if known exactly, produces exact bound-state energies and quasi-particle orbitals from equation (9). We calculate Σ_E using second-order perturbation theory. It improves the agreement between the calculated eigenvalue and the experimental binding energy, e.g. for F^- the Dyson equation gives $E_{2p} = -0.187$ Ryd, which is closer to the experimental $E_{2p}^{\mathrm{exp}} = -0.250$ Ryd, than the HF value $E_{2p}^{\mathrm{HF}} = -0.362$ Ryd. In order to further improve the wavefunction we introduce a scaling parameter in Σ_E (see Gribakin *et al* 1999) and choose it in such a way that equation (9) reproduces the experimental binding energy of the fluorine negative ion, $|E_{2p}| = 0.250$ Ryd (Hotop and Lineberger 1985).

The wavefunctions of the intermediate (n_1l_1, n_2l_2) and final (E_fl_f) states of the photoelectron are calculated in the HF field of the frozen neutral F-atom residue $2p^5$. The photoelectron is coupled to the atomic residue to form the total spin $S = 0$ and the angular momenta $L_1 = 1$ for the first intermediate s and d states ($l_1 = 0, 2$), $L_2 = 0, 2$ for the second intermediate p-wave state ($l_2 = 1$), and $L_2 = 2$ for the second intermediate f-wave state ($l_f = 3$). In the final state the photoelectron is coupled to the core with $L_f = 1$ for the s- and d-wave, and $L_f = 3$ for the d- and g-wave. The intermediate state continua are discretized and represented by a 70-state photoelectron momentum mesh with constant spacing Δk.

Note that the importance of large distances in multiphoton problems speaks in favour of the length form of the photon dipole operator (Gribakin and Kuchiev 1997a, b). This is in agreement with the results of Pan *et al* (1990) who showed that the two-photon detachment cross sections obtained with the dipole operator in the velocity form are much more sensitive to the shift of the photodetachment threshold and correlation corrections. On the other hand, electron correlations have a much weaker effect on the calculations with the length form, and the corresponding results are more robust, and hence, more reliable.

The two-photon $A_{l_0l_2}^{L_2}(\omega, E_0, E_2)$ (6) and three-photon $B_{l_0l_f}^{(L)}(\omega)$ (5) amplitudes are calculated by direct summation over the intermediate states. This method involves accurate evaluation of the free–free dipole matrix elements, and special attention is paid to pole- and δ-type singularities of the integrand (Korol 1994, 1997).

3. Results

This paper highlights the effect of the asymptotic behaviour of the bound-state wavefunction. In what follows we compare the results obtained using the HF 2p wavefunction ($E_{2p}^{\mathrm{HF}} =$

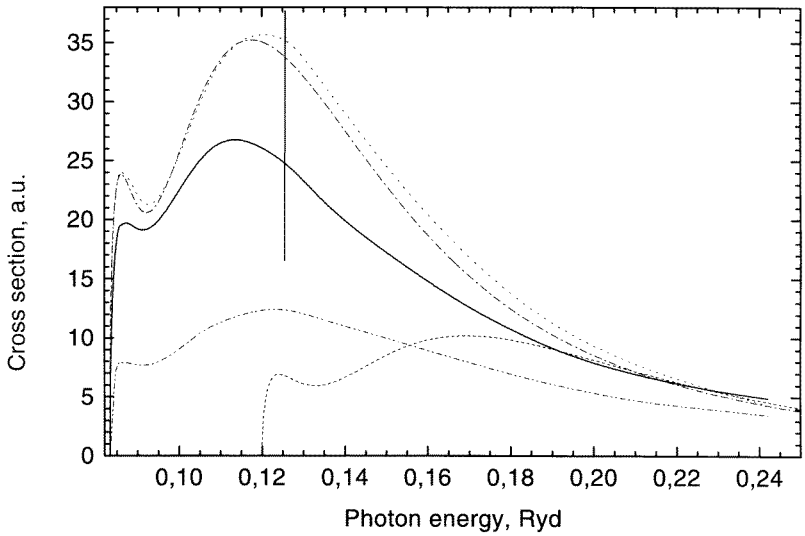

Figure 1. Three-photon detachment cross sections of F^-. Present calculations: - - - - and — · —, adiabatic theory, equation (5) of Gribakin and Kuchiev (1997b), with parameters corresponding to the HF 2p wavefunction and to the corrected 2p wavefunction, respectively; — · · —, direct calculation using the HF wavefunctions of the 2p, intermediate and final states and experimental 2p energy; ——, same with the 2p wavefunction from the Dyson equation; · · · · · ·, 2p wavefunction from the Dyson equation and plane waves for the intermediate and final states. The vertical line shows the position of the two-photon detachment threshold.

-0.362 Ryd), with those based on the 2p wavefunction from the Dyson equation, which has correct (experimental) energy $E_{2p}^{exp} = -0.250$ Ryd and asymptotic behaviour. The Dyson orbital is in fact very close to the HF wavefunction inside the atom, whereas for $r > 2$ au it has larger values than the HF solution, due to smaller binding energy and κ. The asymptotic behaviour of the Dyson 2p orbital is characterized by $\kappa = 0.50$ and $A = 0.64$, while the HF orbital has $\kappa = 0.60$ and $A = 0.94$. For comparison we also calculate the cross sections within the plane-wave approximation and by using the adiabatic theory formula (Gribakin and Kuchiev 1997a, b).

In figure 1 we present three-photon detachment cross sections calculated for F^- using various approaches for the whole energy range studied. Figure 2 shows a comparison between our results and other theoretical and experimental results. In general, all calculations reveal a small near-threshold maximum due to the contribution of the final photoelectron s-wave, and a broad maximum at larger energies due to the d-wave contribution.

When we use the experimental threshold energy together with the HF 2p wavefunction (double-dot-dash curve in figure 1), the overall magnitude of the cross section remains close to that obtained with the HF threshold and wavefunction. On the other hand, when we use the Dyson orbital (solid curve) the cross section becomes substantially higher. This clearly demonstrates the effect of the asymptotic behaviour of the bound-state wavefunction. Moreover, the difference between the three-photon cross sections obtained with the HF and Dyson 2p wavefunctions is greater than that between the corresponding two-photon cross sections (Gribakin *et al* 1999), in accordance with equation (3). Physically this can be related to the fact that with the increase of n the range of important distances (1) increases, and the difference between the two bound-state wavefunctions becomes more significant. Note that if we use the parameters of the HF and Dyson orbitals quoted above, equation (3) gives a ratio

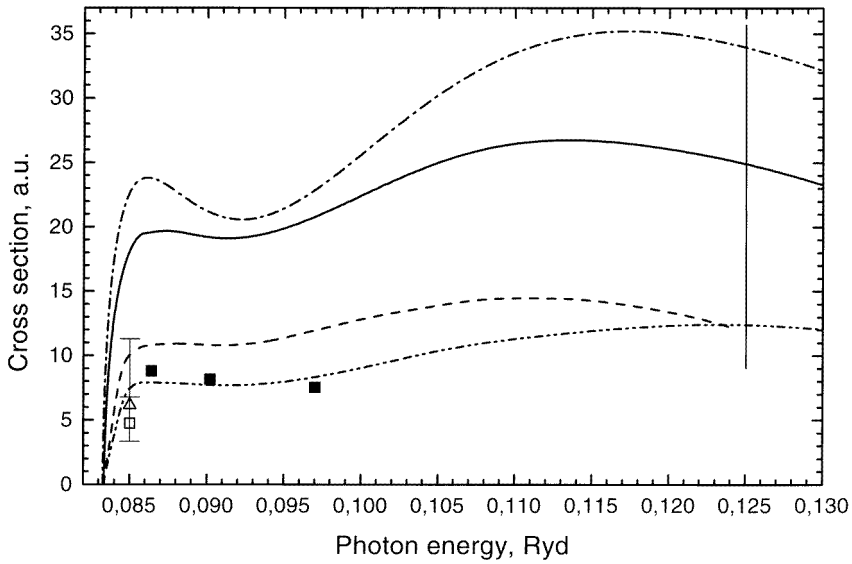

Figure 2. Three-photon detachment cross sections of F^- from different calculations and experiment. Present calculations: —·—, analytical adiabatic theory (Gribakin and Kuchiev 1997a, b) with parameters corresponding to the corrected 2p wavefunction; —··—, direct calculation using the HF wavefunctions of the 2p, intermediate and final states and experimental 2p energy; ——, same with the 2p wavefunction from the Dyson equation. Other results: ■, HF calculation of Crance (1987); - - - -, *R*-matrix Floquet approach (van der Hart 1996); □ and △, experiment of Blondel *et al* (1989b) and Kwon *et al* (1989), respectively. The vertical line shows the position of the two-photon detachment threshold.

of three, in agreement with the difference observed in figure 1.

The cross section obtained using the HF orbital together with the experimental energy (double-dot-dashed curves in the figures) shows a maximum of $\sigma_3 = 12.5$ au at $\omega = 0.125$ Ryd, near the two-photon detachment threshold. The HF results of Crance (1987) below the two-photon detachment threshold (solid squares in figure 2) are close to ours. The cross section of van der Hart (1996) obtained within the *R*-matrix Floquet approach is 20–30% higher (dashed curve in figure 2) with a maximum of $\sigma_3 = 14.5$ au at $\omega = 0.111$ Ryd. Note that a similar difference between the HF calculations with the experimental energy and the *R*-matrix Floquet approach was found for the two-photon detachment cross sections of F^- and Cl^- (Gribakin *et al* 1999). The F^- ground-state wavefunction used by van der Hart (1996) was obtained from a small-scale configuration-interaction (CI) calculation, which gave the binding energy of 0.22 Ryd. As discussed in the introduction, a weaker-bound ground state should have produced larger cross section values. However, the wavefunction from a small CI expansion does not have an appropriate asymptotic form. Moreover, because of the dominant role of the $2p^6$ configuration, the CI wavefunction at large distances is probably close to the HF wavefunction. This fact, together with a relatively small role played by other correlation effects, might explain the proximity of the cross section of van der Hart to the HF results. Experimental results are shown in figure 2 as open symbols. Measurements by Blondel *et al* (1989b) and Kwon *et al* (1989) at $\omega = 0.0856$ Ryd produced values of $\sigma_3 = 4.75(^{+2.02}_{-1.40})$ au and $\sigma_3 = 6.15(^{+5.14}_{-2.80})$ au, respectively. Within the error bars, the latter value is consistent with the HF and *R*-matrix Floquet calculation. However, bearing in mind the role of correct asymptotic behaviour, we believe that this agreement is probably fortuitous.

The best results of the present calculation, shown by a solid curve in figure 2, indicate that the cross section is substantially larger. Let us repeat once more that this increase of the cross section is due to the events which take place at large electron–atom separations, where all correlation corrections are controlled very well. Henceforth we believe that our calculations (solid curve) give the most accurate values for the cross section. Our cross section substantially exceeds, by a factor of two, the HF and R-matrix Floquet results. It has a maximum of $\sigma_3 = 27$ au at $\omega \approx 0.114$ Ryd. As is seen from figure 1, the difference between the cross sections obtained with the Dyson and HF orbitals decreases towards the one-photon detachment threshold ($\omega = 0.25$ Ryd). Indeed, with the increase of ω and the energy of the photoelectron, smaller distances become more important, see (1), and at these distances the two bound-state wavefunctions are quite close.

As noted above, the strong enhancement of the three-photon cross section due to the modified asymptotic behaviour of the wavefunction is in agreement with the two-photon calculations (Gribakin *et al* 1998, 1999) and with the conclusions of the analytical adiabatic theory. To make a direct comparison with this theory we calculate the cross section by using equation (5) of Gribakin and Kuchiev (1997b). The short-dashed curve (figure 1) is obtained using A and κ values of the HF 2p orbital. The corresponding cross section is rather close to the HF result (dashed curve) shifted to the HF threshold. When we use A and κ of the Dyson orbital, dot-dashed curves in figures 1 and 2, the cross section becomes much higher. It is about 30% greater than our direct perturbation-theory calculation with the Dyson orbital, which is a good accuracy for a simple analytical formula. If we describe the photoelectron in the intermediate and final states using plane waves instead of the HF wavefunctions the numerical results (dotted curve in figure 1) become very close to the predictions of the adiabatic theory. Therefore, we can attribute the discrepancy between the adiabatic theory and numerical calculations to the use of free-electron Volkov states in the theory. As seen from figure 1, this discrepancy is not large (and is expected to become smaller with the increase of the number of absorbed photons n).

We see that the use of the asymptotically correct 2p wavefunction changes the three-photon detachment cross section by a factor of two or more. This is similar to the two-photon detachment process, where the effect described above is greater than other correlation effects (Pan *et al* 1990, Gribakin *et al* 1999). There is no reason to expect that the role of such correlations in three-photon detachment is stronger than in two-photon detachment. Thus, we conclude that in *multiphoton* processes the error introduced by using a bound-state wavefunction with incorrect asymptotic behaviour could be much greater than the effects of electron correlations. For the sake of terminology we should mention that to describe correctly the asymptotic behaviour of the ground-state wavefunction, one needs to include many-electron correlations, e.g. through the Dyson equation. However, these correlations are very particular, their manifestation can be described as a simple shift of the electron binding energy. In contrast, conventionally, the term 'many-electron correlations' also includes processes which *cannot* be described in the single-electron picture. It seems though that they are *less important* in the problem considered.

4. Concluding remarks

In this paper we have performed direct numerical calculations of three-photon detachment from the fluorine negative ion, and paid special attention to the correct description of the initial ground-state wavefunction. We ensured that it has a correct asymptotic behaviour by using the many-body theory Dyson equation for calculation of the 2p electron orbital, and adjusting the non-local correlation potential to reproduce the experimental binding energy. Our calculations

demonstrate explicitly that the use of asymptotically correct initial state wavefunctions is very important for finding absolute values of multiphoton detachment cross sections. This confirms the conclusion of the adiabatic theory (Gribakin and Kuchiev 1997a, b, Gribakin *et al* 1999) about the significance of large electron–atom separations in multiphoton processes.

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