Correlations in relativistic photoeffect

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Abstract

We develop a perturbative treatment of the effects beyond the independent particle approximation (IPA) in the relativistic photoionization of states with arbitrary values of the angular momenta ℓ. The dominant mechanism of IPA breaking is discussed. The dependence of IPA breaking contributions on the parameters $\frac{1}{Z}$ and $(\alpha Z)^2$ is analyzed. In the general case the amplitude is expressed as a linear combination of IPA amplitudes. The development of precise experiments, together with the realization that there is cancellation among the non-relativistic partial correlations in many cases, make a relativistic approach to the problem desirable even at relatively low values of photon energy.

We investigate the contributions beyond independent particle approximation (IPA) to the cross sections for relativistic photoionization of states with arbitrary orbital momenta ℓ. We limit ourselves to the case of not very heavy atoms $(\alpha Z)^2 \ll 1$, with $\alpha = \frac{1}{137}$ being the fine structure constant. Using a perturbative approach we obtain the expression for the amplitudes of ionization of the states of any angular momenta ℓ as a linear combination of IPA amplitudes for this process.

At first glance the correlations caused by the final state interactions (FSI) of the ionized electron with the bound electrons of the residual ion are of the same order as the perturbative contribution of this interaction to the final state wave function, i.e. of the order of the Sommerfeld parameter of this interaction,

$$\xi = \frac{\alpha E}{p}, \tag{1}$$

with p and $E = (p^2 + m^2)^{1/2}$ the momentum and the relativistic energy of the outgoing electron (we assume the system of units with $\hbar = c = 1$). At relativistic energies $\xi \sim \alpha$ and the correlation correction would seem to be as small as the radiative corrections, being less then one percent. However, there is a mechanism which enhances these correlations, as we now describe.

The photoeffect, being kinematically not allowed for free electrons, requires that a large momentum q, greatly exceeding the momentum of the bound electron $\tau_{n\ell}$, be transferred to the nucleus, when the ionization is caused by a photon with an energy greatly exceeding the energy of the bound electron. Thus the amplitude is determined by the behavior of the wave function at small $r \sim \frac{1}{q} \ll \tau_{n\ell}^{-1}$. In the lowest order of the expansion in powers of $(\alpha Z)^2$ the wave function is proportional to $(\tau_{n\ell} r)^\ell$ at these distances. The factor $\tau_{n\ell}$ comes from normalization of the bound state wave function. Hence the amplitude of ionization of a $n\ell$ state is $(\tau_{n\ell}/q)^\ell$ times smaller than that of ionization of a ns state. However, this quenching can be avoided if the photon interacts directly with any of the s states. The ionized s-state electron can then push an electron with orbital momentum ℓ into the hole in the s state which was created by the photon. This interaction takes place at distances of the order of the size of the bound states involved, as we shall see further, and this electron transfer amplitude contains one factor ξ, defined by Eq (1), but it multiplies an s state IPA amplitude rather than the amplitude of ionization of the state with orbital momentum ℓ.

This mechanism, first suggested in [1] to explain $2p$ photoionization of Ne, is the main IPA breaking mechanism in photoionization for high nonrelativistic energies of the outgoing electrons [2]. Detailed calculations were carried out in [3, 4]. In the nonrelativistic case $q \approx p = (2m\omega)^{1/2}$. Thus the mechanism changes the asymptotic behavior of the cross sections from the IPA behavior $\sigma^0_{n\ell} \sim \omega^{-7/2-\ell}$ [5] to $\sigma_{n\ell} \sim \omega^{-9/2}$ for all $\ell \geq 1$. Thus for $\ell \geq 2$ the IPA breaking effects change the energy dependence of the cross section at high energy. For $\ell = 1$ the energy dependence is not changed by the mechanism, while its asymptotic coefficient is. This generalizes the earlier analysis carried out in [1], where the interchannel correlations were shown to be responsible for the discrepancy of experimental results and IPA calculations of L-shell ionization of neon by photons with the energies of about 1 keV.

This mechanism is quite general, and it works at all photon energies greatly exceeding the binding energies of the electrons participating in the process. However in the ultrarelativistic limit the momentum q which determines the value of the cross section becomes a constant, of the order of the electron mass, which corresponds to the minimum kinematically allowed q. We can assume that the bound state momentum $\tau_{n\ell} = m\alpha Z a_{n\ell}/n^3$, with $a_{n\ell}$ a numerical coefficient of the order of unity. The IPA amplitude of ionization of a state with nonzero ℓ thus contains an additional factor of the order $(\tau_{n\ell}/q)^\ell$ with respect to the amplitude of ionization of a ns state.

A perturbative expression for the amplitudes, also valid in the relativistic region, can be obtained with the perturbative treatment of FSI worked out in [6]. The amplitude F_i for ionization of a state with quantum numbers $i = n, \ell, \ell_z$, including this mechanism, can be represented as a linear combination of the IPA amplitudes F^0_j of ionization of atomic states j with the quantum numbers $j = n', 0, 0$

$$F_i = F^0_i + \sum_j F^0_j \Lambda_{j,i}, \tag{2}$$

with

$$\Lambda_{j,i} = \int \frac{d^3f}{(2\pi)^3} \frac{2E}{f^2 + 2(\mathbf{p}\cdot\mathbf{f}) + 2E\delta_{j,i}} V_{j,i}(f). \tag{3}$$

Here $\delta_{j,i} = I_j - I_i$ is the difference between the binding energies ($I_{i,j} > 0$), while

$$V_{j,i}(f) = v(f)\langle j|e^{i(\mathbf{f}\cdot\mathbf{r})}|i\rangle. \tag{4}$$

Here $\langle i|$ and $\langle j|$ are the single-electron states in the atom. The interaction $V(f)$ can be treated as the perturbation which is suffered by the wave function of the outgoing electron. In Eq.(4) $v(f)$ is the Fourier transform of the interaction between the outgoing and the bound electrons.

In further estimations we assume that the sizes of the bound states i and j are of the same order of magnitude (the effect is quenched if these values differ strongly), and thus $\tau_{n\ell} \sim \tau_{n'0}$. The matrix element in Eq.(4) is determined at distances r of the order of the size of the bound states. Thus the integral over f on the right hand side of Eq.(3) is determined at small $f \sim \tau_{n\ell}$. This enables us to obtain in the lowest order of $\frac{\tau_{n\ell}}{E}$

$$\Lambda_{j,i} = i\xi S_{j,i}. \tag{5}$$

The matrix elements $S_{j,i}$ describe the transfer of an electron from the state i to fill the hole in the state of the positive ion. Taking the direction of the outgoing electron momentum as the axis of quantization of angular momentum, we find that the mechanism works only for i states with states with $\ell_z = 0$. Note that Eqs.(2)-(4) are valid for all energies $\omega \gg \frac{m(\alpha Z)^2}{2}$. Of course, in the relativistic case all s states j contribute. One can use Eq.(4) to include the influence of the states j with nonzero values of ℓ also, but they contribute beyond the asymptotics. In the ultrarelativistic limit we must put $E = \omega$ and $\xi = \alpha$.

Note that Eq.(4) contains the relativistic IPA amplitudes F_i^0 and F_j^0. The first factor in the integrand of Eq.(3) describes the propagation of the free relativistic electron. However, since the charge Z is not too large and $(\alpha Z)^2 \ll 1$, the averaged velocities of the bound electrons $(\alpha Z/n)^2 \ll 1$. Thus, the electrons in the states $\langle i|$ and $\langle j|$ in the transfer matrix element (4) can be described by nonrelativistic functions. All relativistic effects in Eq.(5) are contained in the relativistic parameter ξ defined by Eq.(1).

In this approximation the interaction $v(f)$ in Eq (4) is just the Fourier transform of the static Coulomb interaction, i.e. we can set $v(f) = \frac{4\pi\alpha}{f^2}$. Since $v(f)$ does not depend on the spin variables, the electrons in the states $\langle i|$ and $\langle j|$ should have the same spin projection, thus forming a spin triplet state.

Now Eq.(2) can be written as

$$F_i = F_i^0 + i\xi\sum_j F_j^0 S_{j,i}. \tag{6}$$

Here the factor ξ comes from a perturbative treatment of the FSI matrix element.In $S_{j,i}$ the angular dependence is separated out in a straightforward way, and the dependence on the IPA wave functions enters through the overlaps of the radial parts $\langle\psi^{(r)}_{n'0}|\psi^{(r)}_{n\ell}\rangle$. The calculation [3] provides

$$S_{j,i} = \langle j|ln(1-t)|i\rangle, \tag{7}$$

$t = \frac{(\mathbf{p}\cdot\ \mathbf{r})}{pr}$ and $\mathbf{p}$ is the momentum of the outgoing electron.

From Eq.(2) we have

$$|F_i|^2 - |F^0_i|^2 = 2Re(i\xi\sum_j F^0_j S_{j,i} F^0_i) + \xi^2|\sum_j S_{j,i}F^0_j|^2. \tag{8}$$

This approach in our case includes contributions through terms of order ξ^2. In the general case, with arbitrary values for the orbital momenta of the states i and j, terms beyond Eq.(6) which involve two interactions in the final state also provide contributions of the order ξ^2 [6]. However, in our case with different angular momenta, one of them being zero, such terms only contribute beyond the leading asymptotic behavior [4].

In the nonrelativistic limit the first term on the rhs of Eq.(8) is zero for even values of ℓ since the corresponding IPA amplitudes are purely real, if we use a conventional definition, in which the bound state wave functions with $\ell_z = 0$ are real. The relativistic amplitudes contain both real and imaginary parts of the same order of magnitude for all values of ℓ. Neither part can be neglected without a more detailed analysis.

Now we estimate the magnitude of the terms on the rhs of Eq.(8) for the relativistic case. The relativistic IPA cross sections take a simple form in the ultrarelativistic low Z limit [5]

$$\sigma^0_{n\ell}(\omega) = \frac{\alpha(\alpha Z)^{2\ell+5}}{m\omega}c_{n\ell}, \tag{9}$$

with $c_{n\ell}$ coefficients of the order of unity. The corrections to this IPA equation are of the order of $\frac{m}{\omega}$ and of αZ. The energy dependence in Eq (9) is the same for all ℓ, unlike in nonrelativistic IPA. The dependence on ℓ manifests itself in the factors $(\alpha Z)^{2\ell}$, which come from the normalization factors of the wave functions of the bound states. Including IPA breaking terms does not alter the IPA dependence on the photon energy (for all ℓ), and one rather obtains the corrections from the two terms of Eq.(8) as of the order

$$\frac{\sigma_{n\ell}(\omega) - \ \sigma^0_{n\ell}(\omega)}{\sigma^0_{n\ell}(\omega)} \sim \frac{\alpha}{(\alpha Z)^{\ell}}\ ;\ \frac{\alpha^2}{(\alpha Z)^{2\ell}} \quad for\ \ell \geq 1, \tag{10}$$

with neglected terms of order $(\alpha Z)^2$. The magnitudes $\frac{\alpha}{(\alpha Z)}$ and $\frac{\alpha^2}{(\alpha Z)^2}$ which follow from the analysis presented above, are just the estimates of the first and second terms on the rhs of Eq.(8).

Note that the first term of the rhs of Eq.(8) does not provide contributions of the order $\frac{\alpha^2}{(\alpha Z)^2}$, since the dependence on the charge Z enters only through the parameter αZ, and dependence on $1/Z$ can be viewed as coming from the parameter $\xi = \frac{\xi Z}{Z}$, with the first factor depending on the product αZ.

For $\ell = 1$ the first term dominates if we assume $Z \gg 1$. In this case the IPA breaking effects are of the order $\frac{1}{Z}$. In the ionization of s states the correlations provide a small contribution of order α.

To obtain quantitative results one will need to sum over j in Eqs (6) and (8) . But in fact the general features of the sum have not yet been discussed for the nonrelativistic case. Therefore let us now discuss the non- relativistic sum over j, to establish the expected size of correlations at high energies. While the recent experiments on ionization of the outer shells of neon [1] and of argon [7] by photons with energies of about 1 keV demonstrated deviations from the IPA predictions, the non-relativistic account of correlations between shells [1, 7] improved the situation. Calculations for higher energies, for the outer shells of N and Ne [8], demonstrated the interplay of correlations within the L shell and with the electrons of the K shell, with strong cancellations. At higher nonrelativistic energies correlation effects were small.

However, it was at first not clear if there is a region where the nonrelativistic asymptotic analysis works, since the cross sections converge to this limit very slowly. The problem and its remedy can be understood [9, 10]. Corrections to the asymptotic results are of the order $1/p$ with rather large numerical coefficients. However it was shown in [3] that the cross sections ratios $R_{n\ell}(\omega) = \frac{\sigma_{n0}(\omega)}{\sigma_{n'}(\omega)}$ converge to the asymptotic limit much faster, mainly due to a common "generalized Stobbe factor", which contains the main slowly convergent energy dependence of the cross sections. This energy-dependent factor is common for the ionization amplitudes of all the bound states. The generalized Stobbe factor is present in all photoionization cross sections and it is primarily responsible for the slow convergence. This was supported by numerical calculations in the Hartree-Fock (HF) approximation. Thus one does expect to be able to predict $c_{n\ell}$ in a $1/\omega$ expansion at nonrelativistic energies.

In [3, 4] the perturbative equations (2)-(8) were used for the calculation of IPA breaking effects in the nonrelativistic case. The obtained relations are especially simple in the asymptotics [2, 4]. However, it is not necessary that the cross-sections have the asymptotic behavior. Somewhat more complicated expressions which are valid beyond the asymptotic region were found in [4] for the cases studied in [1, 7]. The results obtained in [4] eliminated or diminished strongly the difference between the experimental data for L and M shell ionization and the IPA calculations. As expected [11], the perturbative approach and a numerical random phase approximation calculation [9] gave similar results. It was also shown in [9] that at the energies considered in [5, 7] the correlations with the K shell give a minor contribution. For the case of $\ell = 1$, taking account of IPA breaking effects does not alter the linear law for the ratio , which we write as $R_{n1} = r_n\omega$. However IPA breaking effects change the IPA value $r_n = r_n^0$ to

$$r_n = \frac{r_n^0}{1+\kappa}; \quad \kappa = \sum_{n'} \kappa_{n'} + \sum_{n',n''} \eta_{n',n''}. \tag{11}$$

Here $\kappa_{n'}$ are the partial contribution of an s electron with the principal number n' to the first term of rhs of Eq.(8) and by the contributions with $n' = n''$ in the second term of rhs of Eq.(8), while $\eta_{n',n''}$ are cross terms involving products of the contributions of s electrons with the principal quantum numbers n' and n'', in the second term of the rhs of Eq.(8).

We use Eq.(8) to analyze the interplay of the partial contributions in the nonrelativistic asymptotics. Using the data on photoionization cross sections, normalized to data for the HF field [12], we find strong cancellations for the cases considered in [4]. Our calculations were carried out for ionization of $2p$ states by a photon of energy $\omega = 10\ keV$. For nitrogen we found $\kappa_1 = -0.29, \kappa_2 = 0.44$, $\eta_{1,2} = -0.23$, resulting in $\kappa = -0.08$. We found for neon that $\kappa_1 = -0.17$, $\kappa_2 = 0.18$, $\eta_{1,2} = -0.03$ leading to $\kappa = -0.02$. These results are in agreement with those of [8]. The cancellations in ionization of the $3p$ state of argon calculated for $\omega = 30\ keV$, appear to be just as strong: $\kappa_1 = -0.072, \kappa_2 = -0.010, \kappa_3 = 0.073$, $\eta_{1,2} = -0.001$, with the other correlations negligible small, leading to $\kappa = -0.010$. These cancellations take place on the amplitude level. There is a strong compensation between the contributions of the states with different values of j in the sums $\sum_j F_j^0 S_{j,i}$ in Eqs.(6) and (8). In other words, the correlations with different ns states compensate each other in the amplitude (6).

Thus, in the case $\ell = 1$, at least in the examples considered above, the IPA breaking effects appear to be rather large (of the order of about 20%), at intermediate energies which do not exceed strongly the binding energy of the K shell. At these energies the correlations with the K shell are small [3]. However, at asymptotic energies the correlations with the K shell become large, cancelling the total effect to a large extent. Ionization of a d electron of titanium ($Z = 22$), considered in nonrelativistic limit is one more illustration of this tendency. Only the second term of the rhs on Eq. (8) contributes in the nonrelativistic approximation. Calculations carried out at $\omega = 50\ keV$ give an IPA breaking effect of about 4%, while the individual contributions lead to magnitudes about 10 times larger.

In the framework of our approach it is clear that such cancellations do not take place in ions with holes in s states, since the corresponding partial correlation becomes smaller if fewer electrons are available to participate in correlations. The lack of cancellations in ions was noted first in [8].

The strong cancellation of the partial contributions to the correlations seen in these nonrelativistic calculations make taking of relativistic effects into account increasingly important. For example, the relativistic corrections appear to be important for the interplay of the correlations in the ionization of Ne and Ar at $\omega \sim 70\ keV$, and at even smaller energies in the case of ionization of the d states of Ti. In these cases the relativistic corrections to the partial contributions to the correlations of the electrons from the different shells become of the order of the total correlation effect, calculated in the nonrelativistic approximation. It is not clear if the cancellations obtained in nonrelativistic approximation still take place after taking account of relativistic effects. This could be studied experimentally: photon radiation in electron capture by a bare nuclei is to be contrasted with a situation in which IPA is exact and there are no correlations-see [13] and references therein.

In the relativistic case the IPA breaking correction to the square of the amplitude $|F_i|^2$ obtained by this approach is given by Eq (8). The first and the second terms contribute values of the order $\alpha/(\alpha Z)^\ell$ and $\alpha^2/(\alpha Z)^{2\ell}$ correspondingly. The second term is always positive. Thus for $\ell = 1$ the first term dominates, leading to a contribution of the order $1/Z$ as in Eq.(10). The second term provides a correction of the order $1/Z^2$. For higher values of ℓ one can not make a decisive conclusion about the relative role of the two terms. The relative role of the second term with respect to the first one can be described by the parameter $\mu_\ell = \frac{(\alpha Z)^{1-\ell}}{Z}$. We find $\mu_2 = 0.31$ and $\mu_3 = 0.10$ for the lightest atoms containing d and f electrons, with the charges of the nuclei being 21 and 57 correspondingly. Thus, as it stands now we do not see a situation where the second term on the rhs of Eq. (8) can be neglected.

Of course, inclusion of excited ions into the analysis would open more possibilities, since states with larger values of ℓ at small Z can be considered.

Unlike the nonrelativistic case, in the whole region where the ratio $\frac{\omega}{m}$ is not a small parameter the IPA breaking effects in the cross section can not simply be expressed in terms of the IPA cross sections. This is due to the complicated dependence of the amplitudes on the angular variables, which arises since the photon momentum can not be neglected with respect to the electron momentum any more. The amplitudes F_i^0 and $F_j^0 \Lambda_{i,j}$ have different angular dependence, and the integral of the product over the solid angle is not expressed in terms of the cross sections in a straightforward way, as was done in the nonrelativistic case [4].

The quantitative analysis of effects in the relativistic region requires knowledge of the relativistic amplitudes for the whole set of bound states of each atom. A relativistic approach is needed for the analysis of precision experiments on total cross section photoionization measurements. The strong cancellation of correlations at energies described by nonrelativistic approach also means that relativistic effects are of practical importance for more sophisticated analysis at the energies of present experiments.

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