Electron-impact excitation of atoms in the presence of a nearly resonant laser field

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We have studied the electron-impact excitation of an atom, in the presence of a laser field whose photon energy is tuned close to the energy difference between two excited final states. Both the laser-projectile and the laser-target interactions are treated nonperturbatively, while the electron-atom interaction is treated within the first Born approximation. As an application, we have analyzed the resonant laser-assisted excitation of the $2\,^1S$ and $2\,^1P$ states of helium. The agreement between the present nonperturbative results and previous perturbative ones is excellent, except for very small detunings. The present nonperturbative treatment also shows that the results given by perturbation theory on both sides of the resonance, when plotted as a function of the laser frequency, correspond to the excitation of different Floquet pseudostates. This is related to the presence of avoided crossings in the diagram of the Floquet pseudoenergies, as a function of the laser frequency.

I. INTRODUCTION

It is now recognized that laser-assisted electron-atom collisions can be very sensitive to the dressing of the target by the external radiation field.[1-9] The formalism which has been developed to describe such laser-assisted collisions treats the laser-projectile interaction to all orders, while the laser-target interaction is usually treated by using first-order time-dependent perturbation theory.[1-3,5,7,8] This approach is certainly well justified even for strong laser fields, provided, however, that the electric field strength $\mathcal{E}_0$ remains much smaller than the atomic unit of field strength, $\mathcal{E}_0 \ll 5\times10^9$ V cm^{-1}. It should be noted that this condition will always be fulfilled in practice, for at laser intensities comparable to the atomic unit the atom would be ionized, so that electron-atom collisions would no longer be observed.

However, another limitation to the range of intensities that can be considered within this formalism arises from the possibility that the laser photon energy is close to the energy of an atomic transition between the initial or final states and an intermediate state of the collision process.[5] Indeed, in such a resonant situation, even rather moderate laser intensities can result in a strongly nonperturbative laser-atom interaction.

This problem is particularly acute in the case of electron-impact excitation, since such resonant processes can then occur at frequencies which are comparable to the frequencies of commonly used lasers. It has been shown[5,6] that perturbation theory then leads to a series of divergences.

The goal of the present work is to explore the limits of validity of first-order perturbation theory around such resonances. We have therefore developed a nonperturbative treatment of the laser-atom interaction, and we have applied it to the excitation of the $2\,^1S$ and $2\,^1P$ states of helium in the presence of a laser field whose photon energy $\hbar\omega$ is tuned close to the energy difference $E_{2\,^1P}-E_{2\,^1S}=0.602$ eV. The reason for choosing a helium target is that helium is the simplest atomic system with nondegenerate energy levels (the fine structure of the atom being neglected), a point which greatly simplifies the analysis of the results. In what follows, we will only consider fast incident electrons and small momentum transfer collisions, so that all scattering calculations can be performed in the first Born approximation and exchange effects can be neglected.

II. THEORY

Following our previous work,[5] we assume the laser field to be purely monochromatic, linearly polarized, and spatially homogeneous. Working in the Coulomb gauge we have, for the electric field, $\mathcal{E}(t)=\mathcal{E}_0\sin\omega t$, and for the corresponding vector potential, $\mathbf{A}(t)=\mathbf{A}_0\cos\omega t$ with $\mathbf{A}_0=c\mathcal{E}_0/\omega$. Working from now on in atomic units, the Volkov wave function describing the incident "unbound" electron embedded in the field reads (in the velocity gauge)

$$\chi_{\mathbf{k}}(\mathbf{r}_0,t)=(2\pi)^{-3/2}\exp[i(\mathbf{k}\cdot\mathbf{r}_0-\mathbf{k}\cdot\boldsymbol{\alpha}_0\sin\omega t-E_k t)]\ , \qquad (1)$$

where $\mathbf{r}_0$ is the projectile coordinate, $\mathbf{k}$ denotes the electron wave vector, $E_k=k^2/2$ is its kinetic energy, and $\boldsymbol{\alpha}_0=\mathcal{E}_0/\omega^2$.

Our main problem now consists in obtaining an expression for the "dressed" wave functions of the initial and final atomic target states in the laser field, valid to all orders in the resonant (or nearly resonant) interaction between the radiation field and the final states of the collision. We should, therefore, solve the time-dependent Schrödinger equation:

$$i\frac{\partial}{\partial t}\Phi_n(X,t)=[H_0+H_{A-F}(t)]\Phi_n(X,t) , \tag{2}$$

where X denotes the ensemble of the target electrons coordinates, H_0 is the target atom Hamiltonian in the absence of the external field, and H_{A-F} is the atom-field interaction Hamiltonian which reads, in the velocity gauge,

$$H_{A-F}(t)=-\frac{i}{c}\sum_{k=1}^{Z}\mathbf{A}(t)\cdot\nabla_{r_k} , \tag{3}$$

where Z is the atomic number of the target and $\mathbf{r}_k$ is the position vector of the kth target electron.

Solving Eq. (2) exactly would be a formidable task. In the present context we only need, however, to treat exactly the resonant part of the interaction. We therefore introduce orthogonal projection operators P,Q such that

$$P^2=P,Q^2=Q,P+Q=1,PQ=QP=0 , \tag{4}$$

where P projects onto the subspace $\mathcal{H}_P$ of the states which will be included exactly in the calculation. Note that this subspace should, at least, contain the initial and final states of the collision.

The full Schrödinger equation (2) is then approximated, in a first stage, by the simplified equation

$$i\frac{\partial}{\partial t}(P\Phi_n)=P(H_0+H_{A-F})P(P\Phi_n) , \tag{5}$$

in which only the few dominant states are coupled. Using the usual Floquet technique[9] we can now seek solutions of the form

$$P\Phi_n(X,t)=\exp(-i\varepsilon_n t)\exp(-i\mathbf{a}\cdot\mathbf{R})\times\sum_{m\in\mathcal{H}_P}\sum_{N=-\infty}^{+\infty}C_{mn}^{N}\exp(-iN\omega t)\psi_m(X) , \tag{6}$$

where we have defined $\mathbf{a}=c^{-1}\mathbf{A}$, $\mathbf{R}=\sum_{k=1}^{Z}\mathbf{r}_k$, and ψ_m is a target state of energy E_m in the absence of the external field. The Floquet coefficients C_{mn}^{N} and the pseudoenergies ε_n can be found by solving numerically the eigenvalue problem[9]

$$(E_m-N\omega)C_{mn}^{N}+\frac{i}{2}\sum_{m'\in\mathcal{H}_P}M_{mm'}(C_{m'n}^{N-1}-C_{m'n}^{N+1})=\varepsilon_n C_{mn}^{N} , \tag{7}$$

where we have introduced the dipole-coupling matrix elements

$$M_{mm'}=M_{m'm}^{*}=\mathcal{E}_0\cdot\langle\psi_m|\mathbf{R}|\psi_{m'}\rangle . \tag{8}$$

Finally, a first-order correction to the approximate wave function $P\Phi_n$ can be found by treating perturbatively the coupling to all the states which are not included in the subspace $\mathcal{H}_P$. We obtain

$$\Phi_n=P\Phi_n+Q\Phi_n , \tag{9}$$

where the first-order approximation to $Q\Phi_n$ is a solution of the equation

$$i\frac{\partial}{\partial t}(Q\Phi_n)=Q(H_0+H_{A-F})P(P\Phi_n) . \tag{10}$$

Explicitly, we have

$$Q\Phi_n(X,t)\simeq\exp(-i\varepsilon_n t)\exp(-i\mathbf{a}\cdot\mathbf{R})\frac{i}{2}\sum_{m\in\mathcal{H}_P}\sum_{m'\notin\mathcal{H}_P}M_{m'm}\sum_{N=-\infty}^{+\infty}\frac{(C_{mn}^{N+1}-C_{mn}^{N-1})}{E_{m'}-\varepsilon_n-N\omega}\exp(-iN\omega t)\psi_{m'}(X) . \tag{11}$$

In what follows we will denote by Φ_0 the dressed ground state in the presence of the field and by Φ_1,Φ_2 the dressed final states. It should be noted that since the Floquet pseudoenergies ε_n are only defined *modulo* the photon energy, the Floquet pseudostates are not unique. Our convention will be to define Φ_1,Φ_2 as the pseudostates whose pseudoenergies $\varepsilon_1,\varepsilon_2$ tend to the unperturbed eigenenergies E_1,E_2 in the limit $\mathcal{E}_0\to 0$.

Let us now consider the first Born S-matrix element corresponding to the excitation of the dressed states Φ_1,Φ_2 from the dressed ground state Φ_0. It reads

$$S_{f,0}^{B1}=-i\int_{-\infty}^{+\infty}dt\,\langle\chi_{\mathbf{k}_f}(\mathbf{r}_0,t)\Phi_f(X,t)|V_d(\mathbf{r}_0,X)\times|\chi_{\mathbf{k}_i}(\mathbf{r}_0,t)\Phi_0(X,t)\rangle, \quad f=1,2 \tag{12}$$

where

$$V_d(\mathbf{r}_0,X)=-\frac{Z}{r_0}+\sum_{j=1}^{Z}\frac{1}{r_{0j}} \tag{13}$$

is the direct interaction potential, with $r_{0j}=|\mathbf{r}_0-\mathbf{r}_j|$. After integration on the time variable we have

$$S_{f,0}^{B1}=(2\pi)^{-1}i\sum_{L=-\infty}^{+\infty}\delta(E_{k_f}-E_{k_i}+\varepsilon_f-\varepsilon_0-L\omega)f_{f,0}^{B1,L} . \tag{14}$$

The first Born approximation to the inelastic scattering amplitude with the transfer of L photons, $f_{f,0}^{B1,L}$, can be written as

$$f_{f,0}^{B1,L}=f(\mathrm{I})+f(\mathrm{II})+f(\mathrm{III}), \quad f=1,2 \tag{15a}$$

with

$$f(\mathrm{I})=\sum_{N,N'=-\infty}^{+\infty}J_{L+N-N'}(\boldsymbol{\Delta}\cdot\boldsymbol{\alpha}_0)\sum_{m,m'\in\mathcal{H}_P}C_{m0}^{N}C_{m'f}^{N'*}f_{m'm}^{d,B1}(\boldsymbol{\Delta}) , \tag{15b}$$

$$f(\mathrm{II})=\frac{i}{2}\sum_{N,N'=-\infty}^{+\infty} J_{L+N-N'}(\Delta\cdot\alpha_0)\sum_{m,m'\in\mathcal{H}_P}\sum_{m''\notin\mathcal{H}_P} M_{m''m'}\frac{C_{fm}^{N'*}(C_{m'0}^{N-1}-C_{m'0}^{N+1})}{E_{m''}-\varepsilon_0+N\omega} f_{mm''}^{d,B1}(\Delta)\ , \tag{15c}$$

$$f(\mathrm{III})=-\frac{i}{2}\sum_{N,N'=-\infty}^{+\infty} J_{L+N-N'}(\Delta\cdot\alpha_0)\sum_{m,m'\in\mathcal{H}_P}\sum_{m''\notin\mathcal{H}_P} M_{m'm''}\frac{C_{0m}^{N'}(C_{m'f}^{N-1*}-C_{m'f}^{N+1*})}{E_{m''}-\varepsilon_f+N\omega} f_{m''m}^{d,B1}(\Delta)\ . \tag{15d}$$

In the above expressions $J_{L+N-N'}$ is an ordinary Bessel function of order $(L+N-N')$; $\Delta=\mathbf{k}_i-\mathbf{k}_f$ is the momentum transfer; and the quantities $f_{m'm}^{d,B1}(\Delta)$, $f_{mm''}^{d,B1}(\Delta)$, and $f_{m''m}^{d,B1}(\Delta)$ are the first Born amplitudes corresponding to the scattering events $m\to m'$, $m''\to m$, and $m\to m''$ in the absence of the laser field.

It should be stressed that, in agreement with our discussion in Ref. 5, the summation over the high-lying intermediate states m'' in the expressions (15c) and (15d) can safely be performed by using the closure approximation.

The perturbative counterpart of Eqs. (15) has been obtained in Ref. 5. It can conveniently be rewritten:

$$f_{f,0}^{B1,L}=J_L(\Delta\cdot\alpha_0)f_{f,0}^{d,B1}(\Delta)-\frac{i}{2}\sum_{N=\pm1}NJ_{L-N}(\Delta\cdot\alpha_0)\sum_m\left[\frac{M_{fm}f_{m0}^{d,B1}(\Delta)}{E_m-E_f+N\omega}+\frac{M_{m0}f_{fm}^{d,B1}(\Delta)}{E_m-E_0-N\omega}\right],\quad f=1,2\ . \tag{16}$$

Finally, the first Born differential cross section corresponding to the excitation process $0\to f$ accompanied by the transfer of L photons reads

$$\frac{d\sigma_{f,0}^{B1,L}}{d\Omega}=\frac{k_f}{k_i}|f_{f,0}^{B1,L}|^2\ . \tag{17}$$

III. RESULTS AND DISCUSSION

As an application of the method described above, we have studied the electron-impact excitation of the $2\,^1S$ and $2\,^1P$ states of helium in the presence of a laser field whose frequency is tuned close to the Bohr frequency associated with the $2\,^1S\to2\,^1P$ transition.

The results presented below have been obtained by including explicitly the $1\,^1S$, $2\,^1S$, and $2\,^1P$ target states in the subspace $\mathcal{H}_P$. We have used for these states the same wave functions as in our previous work:[5] the $1\,^1S$ and $2\,^1S$ wave functions are those of Byron and Joachain,[10,11] while the $2\,^1P$ wave function is an Eckart wave function.[12]

Our results refer to an incident electron energy $E_{k_i}=500$ eV, a fixed scattering angle $\theta=5°$, and two typical electric field strengths $\mathcal{E}_0=10^6$ and 10^7 V cm^{-1}. Moreover, the polarization vector of the field is taken to be parallel to the momentum transfer Δ.

In Fig. 1, we display the cross sections corresponding to the excitation of the Floquet states $\Phi_{2\,^1S}$, with one photon emitted ($L=-1$), and $\Phi_{2\,^1P}$, without exchange of photon ($L=0$), as a function of the laser photon energy, for an electric field strength $\mathcal{E}_0=10^6$ V cm^{-1}. These results are compared with the cross section obtained by using first-order time-dependent perturbation theory, for the excitation of the $2\,^1S$ state with the emission of one photon ($L=-1$). As expected, the perturbative result exhibits a divergence at the photon energy $\hbar\omega=E_{2\,^1P}-E_{2\,^1S}=0.602$ eV, while the nonperturbative results do not. Surprisingly enough, however, it is seen that on the *left* of the resonance the perturbative curve

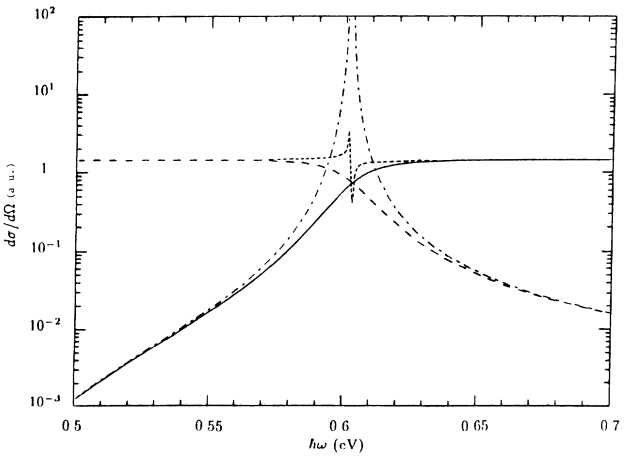

FIG. 1. First Born differential cross sections corresponding to the excitation of the Floquet state $\Phi_{2\,^1S}$ with the emission of one photon ($L=-1$) (solid line) and to the excitation of the Floquet state $\Phi_{2\,^1P}$ with no exchanged photon ($L=0$) (dashed line), as a function of the laser photon energy $\hbar\omega$. These results are compared to the perturbative cross sections describing the excitation of the $2\,^1S$ state, with the emission of one photon ($L=-1$) (dash-dotted curve), and the excitation of the $2\,^1P$ state, without exchanged photon ($L=0$) (dotted curve). The incident electron energy is $E_{k_i}=500$ eV, the scattering angle is $\theta=5°$, the electric field strength is $\mathcal{E}_0=10^6$ V cm^{-1}, and the polarization vector of the field is taken to be parallel to the momentum transfer Δ.

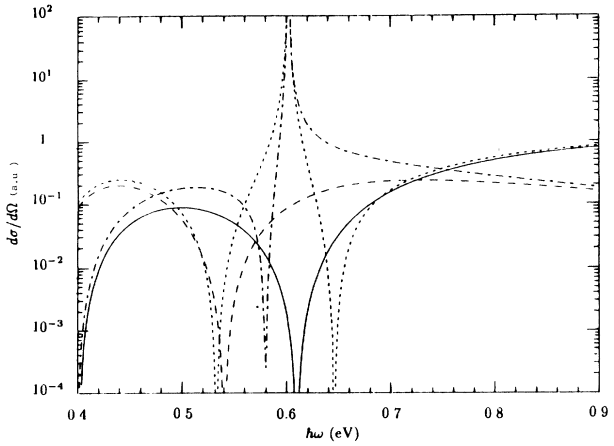

FIG. 2. Same as Fig. 1, but for an electric field strength $\mathcal{E}_0 = 10^7$ V cm^{-1}.

fits very well the nonperturbative result corresponding to the excitation of the Floquet state $\Phi_{2\,^1S}$ with $L=-1$, while on the *right* of the resonance it fits the nonperturbative result corresponding to the excitation of the Floquet state $\Phi_{2\,^1P}$ with $L=0$. On this figure, we have also plotted the perturbative result corresponding to the excitation of the $2\,^1P$ state, without exchanged photon ($L=0$). Its behavior is seen to be opposite that obtained previously for the excitation of the $2\,^1S$ state with $L=-1$. Indeed, this cross section now coincides with the nonperturbative cross section describing the excitation of the Floquet state $\Phi_{2\,^1S}$ with $L=-1$ on the *right* of the resonance while it corresponds, on the *left* of the resonance, to the nonperturbative cross section describing the excitation of the Floquet state $\Phi_{2\,^1P}$ with $L=0$.

The same behavior is observed in Fig. 2, for a higher electric field strength $\mathcal{E}_0 = 10^7$ V cm^{-1}. The reason for this behavior can be understood by looking at the Floquet pseudoenergies $\varepsilon_{2\,^1S}, \varepsilon_{2\,^1P}$ as a function of the laser frequency. Indeed, as shown in Fig. 3, there is an *avoided crossing* between the Floquet pseudoenergies $\varepsilon_{2\,^1S}+\hbar\omega$ and $\varepsilon_{2\,^1P}$. For small frequencies, $\varepsilon_{2\,^1S}+\hbar\omega$ is close to the corresponding unperturbed value $E_{2\,^1S}+\hbar\omega$ and $\varepsilon_{2\,^1P}$ is close to the corresponding unperturbed value $E_{2\,^1P}$. When $\hbar\omega > E_{2\,^1P}-E_{2\,^1S}$, however, the situation is opposite, $\varepsilon_{2\,^1S}+\hbar\omega$ being close to $E_{2\,^1P}$ and $\varepsilon_{2\,^1P}$ being close to $E_{2\,^1S}+\hbar\omega$. This explains the nature of the difference in the cross sections: in perturbation theory the crossing between the levels is effective, while in the Floquet theory it is avoided.

This does not mean, of course, that perturbation theory is correct on one side of the resonance and in-

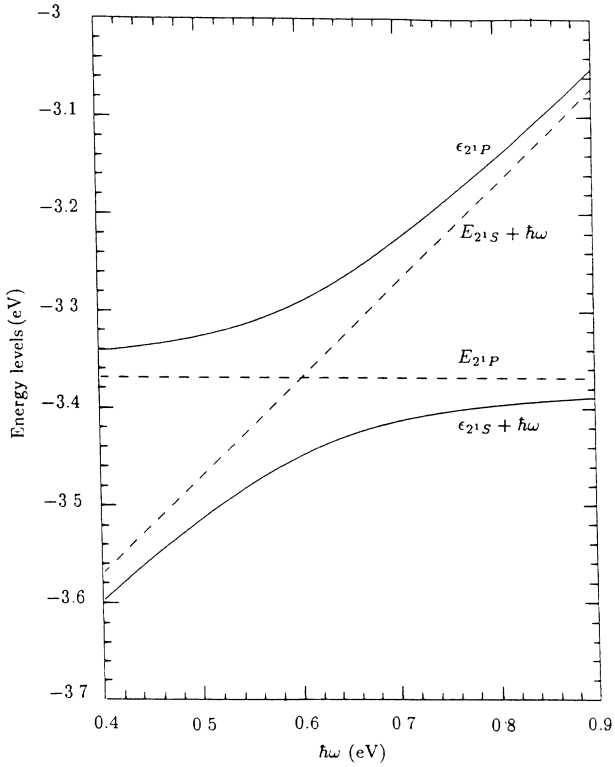

FIG. 3. Floquet pseudoenergies $\varepsilon_{2\,^1S}+\hbar\omega$ and $\varepsilon_{2\,^1P}$ (solid lines), compared to the unperturbed eigenenergies $E_{2\,^1S}+\hbar\omega$ and $E_{2\,^1P}$ (dashed lines). The electric field strength is $\mathcal{E}_0 = 10^7$ V cm^{-1}.

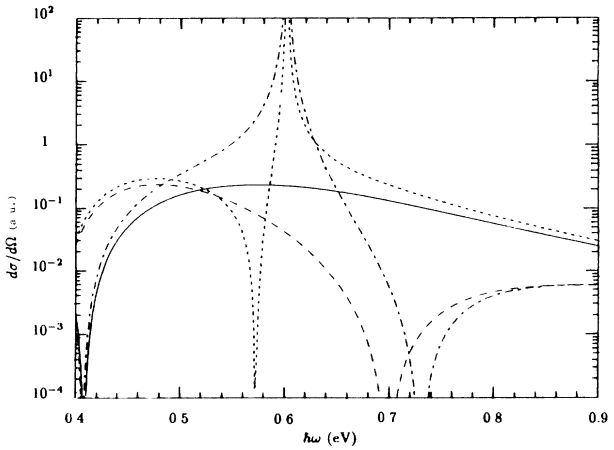

FIG. 4. First Born differential cross sections corresponding to the excitation of the Floquet state $\Phi_{2\,^1S}$ with the absorption of one photon ($L=+1$) (solid line) and to the excitation of the Floquet state $\Phi_{2\,^1P}$ with the absorption of two photons ($L=+2$) (dashed line), as a function of the laser photon energy $\hbar\omega$. These results are compared to the perturbative cross sections describing the excitation of the $2\,^1S$ state with the absorption of one photon ($L=+1$) (dash-dotted curve) and the excitation of the $2\,^1P$ state with the absorption of two photons ($L=+2$) (dotted line). The incident electron energy is $E_{k_i}=500$ eV, the scattering angle is $\theta=5°$, the electric field strength is $\mathcal{E}_0 = 10^7$ V cm^{-1}, and the polarization vector of the field is taken to be parallel to the momentum transfer Δ.

correct on the other: on both sides the coinciding perturbative and nonperturbative cross-sections correspond, except for the small dynamical Stark shift, to the same final energy. This final energy is of course the only quantity observable experimentally, the denomination of the atomic Floquet states being a question of convention.

The above conclusions drawn for the cases $L=\pm 1$ remain valid for other values of L. For instance, we compare, in Fig. 4, the perturbative results corresponding to the excitation of the $2\,^1S$ state with the absorption of one photon ($L=+1$) and the excitation of the $2\,^1P$ state with the absorption of two photons ($L=+2$), respectively, with the nonperturbative results corresponding to the excitation of the pseudostates $\Phi_{2\,^1S}$ with $L=+1$ and $\Phi_{2\,^1P}$ with $L=+2$. The electric field strength is $\mathcal{E}_0=10^7$ V cm^{-1}. Once again, except for very small detunings (less than, say, 0.1 eV), the agreement between the perturbative and nonperturbative results corresponding to nearly equivalent final energies is excellent. By comparing the results of Figs. 2 and 4, it is also seen that the asymmetry predicted by perturbation theory between the emission and the absorption of a given number of photons is well verified by our nonperturbative approach. Finally the perturbative and nonperturbative results differ in that the nonperturbative theory predicts *no* maximum of the cross sections at resonance, while the perturbative results diverge.

IV. CONCLUSIONS

We have elaborated a treatment of electron-atom inelastic collisions in the presence of a nearly resonant laser field. Our method treats to all orders the interaction of the field with the fast incident projectile, as well as the interaction of the field with the dominant (low-lying) atomic states, i.e., the initial state and the resonantly coupled final states. This latter interaction is treated by using the Floquet theory for multiphoton transitions. Finally, the coupling of the laser field with the high-lying states of the atomic spectrum is treated perturbatively.

This method has been applied to the excitation of the $2\,^1S$ and $2\,^1P$ states of helium, in the presence of a laser field whose frequency is close to the Bohr frequency associated with the $2\,^1S\rightarrow 2\,^1P$ transition. Our results have been compared with those obtained by treating the laser-atom interaction by first-order time-dependent perturbation theory. The agreement between both methods is good, except close to the resonance where the perturbative cross sections diverge, while the nonperturbative ones exhibit no maximum as a function of the laser frequency.

Another very interesting effect is that the presence of an avoided crossing of the Floquet pseudoenergies at resonance does not allow one to establish a one-to-one correspondence between the perturbative and nonperturbative cross sections corresponding to the excitation of a given final state. Indeed, it is the perturbative cross section corresponding to the excitation of the $2\,^1S$ state with the exchange of L photons, *together* with the perturbative cross section corresponding to the excitation of the $2\,^1P$ state with the exchange of $L+1$ photons, which have to be compared with their nonperturbative counterparts. This can be understood on physical grounds since, close to the resonance, both processes nearly correspond to the same final energy of the scattered electron and should, therefore, be difficult to distinguish experimentally.

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