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# Higher-Order Effects in Resonant Multiphoton Processes

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The multiphoton transitions in the hydrogen atom are investigated by taking into account the higher-order terms arising from the $S$-operator expansion. These higher-order effects originate induced shifts and induced breadths on the virtual-atomic-energy levels. It is shown that these perturbations play an important role for resonant multiphoton processes. In this case, the theory predicts a particular flux dependence of the transition rates. Furthermore, the well-known dispersion curves are strongly modified in the neighborhood of any resonance. Thus the transition rates can exhibit several peaks. Numerical results are given for a three-photon bound-free transition from the $2S$ state of hydrogen.

## I. INTRODUCTION

The multiphoton-absorption processes are conveniently handled with the help of time-dependent perturbation theory.[1–7] Within this framework one usually considers the lowest-order nonvanishing term which describes the phenomenon of interest. It seems that this approximation is not sufficient for a good description of resonant processes and when high-intensity radiation sources are involved.[8,9] In fact, among the higher-order terms arising from the $S$-operator expansion, some of them give a contribution to the cross section which may no longer by negligible. It is easily shown that the main contribution is brought by higher-order terms involving photons of the radiation field.

This paper is devoted to the analysis of higher-order effects occurring in resonant bound-bound and bound-free multiphoton transitions. It is shown how these effects perturb atomic energy levels. These perturbations caused by the Hamiltonian which produces the transitions originate induced shift and induced breadth of intermediate levels. As a consequence we find that the rate of any process can be calculated by the use of two equivalent methods. Firstly, one may consider the contribution coming from all the relevant terms of the $S$-operator expansion, each of them exhibiting unperturbed atomic energies. In this case, the calculations are done in the so-called undisplaced energy picture. Secondly, one may keep only the lowest-order term in which the perturbed atomic energies are substituted to the unperturbed ones. The calculations are then worked out in the displaced energy picture. The advantage of working in the undisplaced energy picture is that the rate of any multiphoton transition may be numerically calculated, up to an arbitrary order, with great accuracy. This is because, in this case, the implicit technique of Schwartz and Tieman[10–12] may be applied for the evaluation of the sums over the intermediate states. It is not so in the displaced energy picture, but in this scheme the expression of the transition rate has a particularly compact form which can be easily discussed.

In Sec. II we consider bound-free and bound-bound multiphoton transition rates as deduced from the lowest-order term of the $S$-operator expansion. Radiation corrections are taken into account through the electron propagator discussed by Low.[13]

The higher-order effects due to the radiation field are introduced in Sec. III. We find that the dominant contribution is brought by the terms which involve the repeated occurrence of absorption followed by emission, and emission followed by absorption, of a photon of the incoming light.[18–20] These effects are summarized into a perturbed propagation operator which is subjected to a careful analysis. We show how the higher-order terms originate a shift and a breadth of the atomic levels.

The aim of this paper is to investigate the effects of an intense radiation field on resonances occurring in intermediate states. Then, for shortness, we have overlooked an unessential treatment of perturbations on initial and final atomic states. For further discussion the reader is referred to the paper of Sen Gupta[14] and to references quoted in. The case of resonance is discussed in the displaced energy picture. It is found that the order of nonlinearity (as defined in Ref. 9), of any multiphoton process varies rapidly with regard to the wavelength of the radiation. When damping phenomena are taken into account, the values of the order of nonlinearity exhibit typical variations about the ones previously found from unperturbed theories. This fact has been confirmed by recent experiments.[15–17] In addition, these variations can explain the discrepancies which exist between theoretical[1–7] and experimental results.[8,9]

In Sec. IV, the procedure for calculating the $N$th-order matrix element is discussed and some numerical results are given in Sec. V.

## II. LOWEST-ORDER MULTIPHOTON PROCESSES

For brevity we put $\hbar = c = 1$, but we restore $\hbar$ and $c$ in the computational formula. In previous papers,[6,7] we have calculated the cross section for some multiphoton processes occurring when a hydrogen atom is irradiated by an intense light source. It has been seen that, when the energy corresponding to one or several absorbed photons approaches that of a transition line, the cross section becomes infinite; then the theory fails at exact resonances. This difficulty can be avoided by using the electron-propagation operator derived by Low[13]:

$$S_F(x_1, x_2) = \frac{1}{2\pi i} \sum_n \int d\omega \, \frac{e^{i\omega(t_1 - t_2)}}{\omega_n + \omega - w_{nn}(\omega)} \, \psi_n(\vec{x}_1) \bar{\psi}_n(\vec{x}_2). \tag{2.1}$$

In Eq. (2.1) $w_{nn}(\omega)$ is expressed in terms of $\delta_{nn}(\omega)$ and $\gamma_{nn}(\omega)$, which denote the natural level shift and the natural level width, respectively,

$$w_{nn}(\omega) = \delta_{nn}(\omega) + \tfrac{1}{2} i \gamma_{nn}(\omega) \quad . \tag{2.2}$$

In the nonrelativistic dipole approximation it is found that

$$\delta_{nn}(\omega) = \frac{e^2}{m^2 V} \sum_\lambda \sum_i \int \frac{d\vec{k}}{2|\vec{k}|} \, \frac{|[\vec{\epsilon}^{\lambda}(\vec{k}) \cdot \vec{p}]_{in}|^2}{\omega_i + \omega - |\vec{k}|} \, , \tag{2.3a}$$

$$\gamma_{nn}(\omega) = \frac{\pi e^2}{m^2 V} \sum_\lambda \sum_i \int \frac{d\vec{k}}{2|\vec{k}|} \, |[\vec{\epsilon}^{\lambda}(\vec{k}) \cdot \vec{p}]_{in}|^2 \delta(\omega + \omega_i - |\vec{k}|) \; ; \tag{2.3b}$$

$\vec{p}$ is the electron momentum and $\epsilon^{\lambda}(\vec{k})$ is the photon polarization vector. Then, the natural breadth of the resonant line acts as a damping factor which makes finite the corresponding cross section. The $S$-matrix element describing the $N$-photon bound-free transition is

$$S_{N,0}^{(N)\mathrm{BF}} = (-ie)^N \int dx_N \int dx_{N-1} \cdots \int dx_1 \langle n_p - N| \otimes \langle i_N(x_N)| A\!\!\!/^{+}(x_N) \\ \times S_F(x_N, x_{N-1}) A\!\!\!/^{+}(x_{N-1}) \cdots A\!\!\!/^{+}(x_2) S_F(x_2, x_1) A\!\!\!/^{+}(x_1) | i_0(x_1)\rangle \otimes |n_p\rangle \quad . \tag{2.4a}$$

For the $N$-photon bound-bound transition the matrix element is

$$S_{N,0}^{(N)\mathrm{BB}} = (-ie)^N \sum_{m=1}^{N} \int dx_N \int dx_{N-1} \cdots \int dx_1 \langle n_p - N + 1, 1_s| \otimes \langle i_N(x_N)| \mathcal{A}\!\!\!/_m (x_N) \\ \times S_F(x_N, x_{N-1}) \mathcal{A}\!\!\!/_m(x_{N-1}) \cdots \mathcal{A}\!\!\!/_m(x_2) S_F(x_2, x_1) \mathcal{A}\!\!\!/_m(x_1) | i_0(x_1)\rangle \otimes |n_p\rangle \quad , \tag{2.4b}$$

where

$$\mathcal{A}\!\!\!/_i(x_j) = [1 - \delta(i,j)] A\!\!\!/^{+}(x_j) + \delta(i,j) A\!\!\!/^{-}(x_j) \quad , \tag{2.5}$$

$\delta(i,j)$ is the Kronecker symbol, and the notation $A\!\!\!/^{\pm}$ is used for $\sum_i \gamma_i A_i^{\pm}(x)$, where the $\gamma_i$ are the Dirac matrices. The positive- and negative-energy parts of the em potential $A^{\pm}(x)$ are given by

$$A^{+}(x) = [A^{-}(x)]^{+} = V^{-1/2} \sum_\lambda \int \frac{a_\lambda(\vec{k})}{(2|\vec{k}|)^{1/2}} \, \vec{\epsilon}_\lambda(\vec{k}) e^{ikx} d\vec{k} \quad . \tag{2.6}$$

In Eqs. (2.4), the radiation field is described in terms of the occupation number state $|n\rangle$; $|i_0(x_1)\rangle$ and $|i_N(x_N)\rangle$ are used for the initial and final atomic states, respectively.

## III. HIGHER-ORDER EFFECTS

The rates of bound-free and bound-bound transitions calculated from Eqs. (2.4) are the lowest-order approximations to the considered processes. In fact the $S$-operator expansion provides many higher-order terms. For a well-defined atomic transition involving absorbed and emitted quanta, only terms leading to the correct final occupation number state do not vanish. It may be easily verified that the remaining higher-order terms can be set up from the lowest-order one by the admixture of contributions owing to one or several processes which may be summarized in the following manner: The electron absorbs a photon of the radiation field and then emits the same quantum; or a photon of the field is emitted first and then absorbed.[18–20] These processes are both described by the operator $M_F$ defined as

$$M_F(x_1, x_2) = (ie)^2 [A\!\!\!/^{-}(x_1) S_F(x_1, x_2) A\!\!\!/^{+}(x_2) + A\!\!\!/^{+}(x_1) S_F(x_1, x_2) A\!\!\!/^{-}(x_2)] \quad . \tag{3.1}$$

Such an operator can act at any stage of the $N$th-order multiphoton transition. Then, all elements of the $S$-operator expansion may be rearranged to put forward particular sets of terms which are summed. In using a standard technique of resummation of diagrams,[21] one observes that the expression of transition rate obtained by this way can be deduced from Eqs. (2.4) in replacing the electron-propagation operator $S_F$ by the perturbed propagator $S_F'$ given by the integral equation

$$S_F'(x_1, x_2) = S_F(x_1, x_2) + \int\int dx'dx''S_F(x_1, x')M_F(x', x'')S_F'(x'', x_2) \ . \tag{3.2}$$

Then, the $N$-photon bound-free and bound-bound transition rates including higher-order effects are to be calculated from the matrix elements expressed in Eqs. (3.3a) and (3.3b), respectively:

$$\begin{aligned}\tilde{S}_{N,0}^{(N)\mathrm{BF}} = (-ie)^N \int dx_N \int dx_{N-1} \cdots \int dx_1 \langle n_p - N| \otimes \langle i_N(x_N)| \\ \times \not{A}^+(x_N)S_F'(x_N, x_{N-1})\not{A}^+(x_{N-1}) \cdots \not{A}^+(x_2)S_F'(x_2, x_1)\not{A}^+(x_1)|i_0(x_1)\rangle \ |n_p\rangle \ ,\end{aligned} \tag{3.3a}$$

$$\begin{aligned}\tilde{S}_{N,0}^{(N)\mathrm{BB}} = (-ie)^N \sum_{m=1}^{N} \int dx_N \int dx_{N-1} \cdots \int dx_1 \langle n_p - N + 1, 1_s| \otimes \langle i_N(x_N)| \not{\epsilon}_m(x_N) \\ \times S_F'(x_N, x_{N-1})\not{\epsilon}_m(x_{N-1}) \cdots \not{\epsilon}_m(x_2)S_F'(x_2, x_1)\not{\epsilon}_m(x_1)|i_0(x_1)\rangle \otimes |n_p\rangle \ .\end{aligned} \tag{3.3b}$$

In Eqs. (3.3) the functions $S_F'$ lying near the initial and final states give some contributions which are the same as the ones arising from corrections to external electron lines. Such contributions will be ignored throughout this paper.

### A. Calculation of $S_F'$

Following Low[15] we set

$$S_F'(x_1, x_2) = \frac{1}{2\pi i} \sum_{k,l} \int d\omega \, O_{kl}(\omega) \, e^{i\omega(t_1 - t_2)} \, \psi_k(\vec{x}_1)\bar{\psi}_l(\vec{x}_2) \ , \tag{3.4}$$

where $O$ is an operator acting on the radiation-field states. The summations run on the complete set of atomic states, discrete plus continuum. Substituting Eqs. (2.1) and (3.4) into Eq. (3.2) and taking the inner product with $\bar{\psi}_m(\vec{x}_1)\beta \cdots \beta\psi_n(\vec{x}_2)$ we find

$$\begin{aligned}\int d\omega \, O_{mn}(\omega) \, e^{i\omega(t_1-t_2)} = \int d\omega \, \frac{e^{i\omega(t_1-t_2)}}{\omega_m + \omega - w_{mm}(\omega)} \, \delta_{mn} \\ + \frac{1}{2\pi i} \int\int dx_3 \, dx_4 \int d\omega_1 \frac{e^{i\omega_1(t_1-t_3)}}{\omega_m + \omega_1 - w_{mm}(\omega_1)} \, \bar{\psi}_m(\vec{x}_3)M_F(x_3, x_4) \\ \times \sum_r \int d\omega_2 \, \psi_r(\vec{x}_4)O_{rn}(\omega_2) \, e^{i\omega_2(t_4-t_2)} \ .\end{aligned} \tag{3.5}$$

Then, we introduce the operator $\Omega$ defined as

$$\Omega_{mn}(\omega) = \Omega_{mn}^+(\omega) + \Omega_{mn}^-(\omega) \ , \tag{3.6}$$

with

$$\Omega_{mn}^+(\omega) = \sum_\lambda \sum_i \int d\vec{k} \, \frac{h_{mi}^\lambda(\vec{k})h_{in}^{\lambda*}(\vec{k})}{\omega_i + \omega + |\vec{k}| - w_{ii}(\omega)} \, \frac{a_\lambda(\vec{k})a_\lambda^\dagger(\vec{k})}{V} \ , \tag{3.7}$$

$$\Omega_{mn}^-(\omega) = \sum_\lambda \sum_i \int d\vec{k} \, \frac{h_{mi}^{\lambda*}(\vec{k})h_{in}^\lambda(\vec{k})}{\omega_i + \omega - |\vec{k}| - w_{ii}(\omega)} \, \frac{a_\lambda^\dagger(\mathrm{k})a_\lambda(\vec{k})}{V} \ , \tag{3.8}$$

and

$$h_{mn}^\lambda(\vec{k}) = \frac{e}{(2|\vec{k}|)^{1/2}} \langle m|\vec{\alpha} \cdot \vec{\epsilon}_\lambda(\vec{k}) \, e^{i\vec{k}\cdot\vec{x}}|n\rangle \ . \tag{3.9}$$

It should be pointed out that $a^\dagger a/V$ is the flux operator in natural units.

After some algebraic manipulations one finds that $O_{mn}$ is given by the equation

$$O_{mn}(\omega) = \frac{\delta_{mn}}{\omega_m + \omega - w_{mm}(\omega)} + \sum_i \frac{\Omega_{mi}(\omega)O_{in}(\omega)}{\omega_m + \omega - w_{mm}(\omega)} \ , \tag{3.10}$$

which yields

$$O_{nn}(\omega) = \frac{1}{\omega_n + \omega - w_{nn}(\omega)} + \frac{\Omega_{nn}(\omega)O_{nn}(\omega)}{\omega_n + \omega - w_{nn}(\omega)} + \sum_{i\neq n} \frac{\Omega_{ni}(\omega)O_{in}(\omega)}{\omega_n + \omega - w_{nn}(\omega)} \ , \tag{3.11a}$$

$$O_{mn}(\omega)=\frac{\Omega_{mn}(\omega)O_{nn}(\omega)}{\omega_m+\omega-w_{mm}(\omega)}+\sum_{i\neq n}\frac{\Omega_{mi}(\omega)O_{in}(\omega)}{\omega_m+\omega-w_{mm}(\omega)}\quad(m\neq n)\ . \tag{3.11b}$$

Let the operator $U(\omega)$ be such that[22]

$$U_{mn}(\omega)=0\qquad\text{if } m=n\ , \tag{3.12a}$$

$$O_{mn}(\omega)=U_{mn}(\omega)O_{nn}(\omega)\qquad\text{if } m\neq n\ . \tag{3.12b}$$

Then from Eqs. (3.11a) and (3.12b) one finds that the diagonal part $O_{nn}(\omega)$ can be expressed in the form

$$O_{nn}(\omega)=\frac{1}{\omega_n+\omega-w_{nn}(\omega)-W_{nn}^{\mathrm{op}}(\omega)}\ , \tag{3.13}$$

where

$$W_{nn}^{\mathrm{op}}(\omega)=\Omega_{nn}(\omega)+\sum_{i\neq n}\Omega_{ni}(\omega)U_{in}(\omega) \tag{3.14}$$

and

$$U_{mn}(\omega)=\frac{\Omega_{mn}(\omega)}{\omega_m+\omega-w_{mm}(\omega)}+\sum_{i\neq n}\frac{\Omega_{mi}(\omega)U_{in}(\omega)}{\omega_m+\omega-w_{mm}(\omega)}\ , \tag{3.15}$$

while the nondiagonal part $O_{mn}$ is deduced from Eqs. (3.12b), (3.13), and (3.15). Substituting Eqs. (3.12b) and (3.13) into Eq. (3.4) one finds that $S_F'$ is given by

$$S_F'(x_1,x_2)=\frac{1}{2\pi i}\sum_n\int d\omega\,\frac{\psi_n(\vec{\mathrm{x}}_1)\bar{\psi}_n(\vec{\mathrm{x}}_2)}{\omega_n+\omega-w_{nn}-W_{nn}^{\mathrm{op}}(\omega)}\,e^{i\omega(t_1-t_2)}$$
$$+\frac{1}{2\pi i}\sum_{\substack{m,n\\ m\neq n}}\int d\omega\,\frac{\psi_m(\vec{\mathrm{x}}_1)\bar{\psi}_n(\vec{\mathrm{x}}_2)}{\omega_n+\omega-w_{nn}(\omega)-W_{nn}^{\mathrm{op}}(\omega)}\,U_{mn}(\omega)\,e^{i\omega(t_1-t_2)}\ . \tag{3.16}$$

In the future the off-diagonal part of $S_F'$ will be neglected because it is of higher order in $e^2$ than the diagonal one.

In contrast to Eq. (3.2), Eq. (3.16) exhibits the expression of the propagation operator in the displaced energy picture.

### B. Resonances

We consider a radiation field of fixed amplitude with small random-frequency variations. We are led to phase diffusion, and no detailed treatment of this model will be presented here, since number of authors have used it for describing a well-stabilized laser beam.[23,24] As a result of heavy calculations, it appears that at each step $m$ of the interaction, the linewidth $b$ of the light gives rise to a damping factor $(im^2b)$ associated with the absorption of the $m$th photon. Then, multiphoton bound-free and bound-bound transition rates may be derived from Eqs. (3.3), by the use fo a propagation function which differs from Eq. (3.16) by the additional damping caused by the radiation field. For a linearly polarized field in a pure state, it is found that

$$\tilde{T}_{N,0}^{(N)\,\mathrm{BF}}=F^N\,|\tilde{K}_{N,0}^{(N)\,\mathrm{BF}}|^2\rho(N\omega_p)\ , \tag{3.17a}$$

$$\tilde{T}_{N,0}^{(N)\,\mathrm{BB}}=F^{N-1}\,|\tilde{K}_{N,0}^{(N)\,\mathrm{BB}}|^2 g\{(N-1)\omega_p-\omega_s\}\ . \tag{3.17b}$$

In Eqs. (3.17) $\rho(N\omega_p)$ is the density of final atomic states and $g\{(N-1)\omega_p-\omega_s\}$ is the atomic-line-shape function defined as

$$g\{N\omega_p\}=\frac{2(\frac{1}{2}\gamma_f+N^2b)}{(\omega_0+N\omega_p-\omega_f)^2+(\frac{1}{2}\,\partial\gamma_f+N^2b)^2}\ , \tag{3.18}$$

where $\gamma_f$ is the width of the final state and $\omega_0$ and $\omega_f$ are the unperturbed energies of the initial and final state, respectively.

The $N$th-order reduced transition matrix elements are given by

$$\tilde{K}_{N,0}^{(N)\,\mathrm{BF}}=\sum_{i_1,i_2,\ldots,i_{N-1}}h_{N,N-1}(\vec{\mathrm{k}}_p)S_{N-1,N-2}(\vec{\mathrm{k}}_p,\,-\omega_0-(N-1)\omega_p)$$
$$\times S_{N-2,N-3}(\vec{\mathrm{k}}_p,\,-\omega_0-(N-2)\omega_p)\cdots S_{1,0}(\vec{\mathrm{k}}_p,\,-\omega_0-\omega_p)\ , \tag{3.19a}$$

$$\tilde{K}_{N,0}^{(N)\,\mathrm{BB}}=\sum_{\nu=1}^{N}\ \sum_{i_1,i_2,\ldots,i_{N-1}}H_{N,N-1}^{(\nu)}(\vec{\mathrm{k}}_p,\vec{\mathrm{k}}_s)U_{N-1,N-2}^{(\nu)}(\vec{\mathrm{k}}_p,\vec{\mathrm{k}}_s,\,-\omega_0-\xi_{(N-1)}^{(\nu)})$$

$$\times U^{(\nu)}_{N-2,N-3}(\vec{k}_p, \vec{k}'_s, -\omega_0 - \xi^{(\nu)}_{(N-2)}) \cdots U^{(\nu)}_{1,0}(\vec{k}_p, \vec{k}_s, -\omega_0 - \xi^{(\nu)}_{(1)}) \ , \tag{3.19b}$$

with

$$S_{m,n}(\vec{k}, \omega) = \frac{h_{mn}(\vec{k})}{\omega + \omega_m - w_{mm}(\omega) - W_{mm}(\omega) - im^2 b} \ , \tag{3.20}$$

$$H^{(\nu)}_{mn}(\vec{k}, \vec{k}') = [1 - \delta(\nu, m)]h_{mn}(\vec{k}) + \delta(\nu, m) h^*_{mn}(\vec{k}') \ , \tag{3.21}$$

$$U^{(\nu)}_{mn}(\vec{k}, \vec{k}', \omega) = \frac{H^{(\nu)}_{mn}(\vec{k}, \vec{k}')}{\omega + \omega_m - w_{mm}(\omega) - W_{mm}(\omega) - i[m - \delta(\nu, m)]^2 b} \ , \tag{3.22}$$

$$\xi^{(\nu)}_{(m)} = \xi^{(\nu)}_{(m-1)} + \omega_p - \delta(\nu, m)(\omega_p + \omega_s) \quad (\xi^{(\nu)}_{(0)} = 0) \ . \tag{3.23}$$

$\vec{k}_p$ and $\vec{k}_s$ denote the photon momentum of the incoming and outgoing quanta, respectively. The function $W(\omega)$ deduced from Eq. (3.14) is given by the series

$$W_{mm}(\omega) = F D_{mm}(\omega) + F^2 \sum_{q \neq m} D_{mq}(\omega) \frac{1}{\omega + \omega_q - w_{qq}(\omega)} D_{qm}(\omega) + \cdots \ , \tag{3.24}$$

where

$$D_{nm}(\omega) = \sum_j \left( \frac{h_{nj}(\vec{k}_p) h^*_{jm}(\vec{k}_p)}{\omega + \omega_j + \omega_p - w_{jj}(\omega)} + \frac{h^*_{nj}(\vec{k}_p) h_{jm}(\vec{k}_p)}{\omega + \omega_j - \omega_p - w_{jj}(\omega)} \right) \ . \tag{3.25}$$

The polarization index has been dropped in the above equations since no confusion can exist. Furthermore for brevity, the intermediate states have been denoted by their lower subscripts, i.e., $i_1 \to 1$, $i_2 \to 2$, .... As is easily seen from Eqs. (3.17)–(3.25), the multiphoton absorption rates have a somewhat complicated flux dependence. This is due to the occurrence of the complex quantity $W(\omega)$, which has a simple meaning when it is split into its real and imaginary parts. Thus, Eq. (3.24) yields

$$W_{nn}(\omega) = \Delta_{nn}(\omega) + \tfrac{1}{2} i \Gamma_{nn}(\omega) \ , \tag{3.26}$$

where

$$\Delta_{nn}(\omega) = \Delta^{(1)}_{nn}(\omega) + \Delta^{(2)}_{nn}(\omega) + \cdots \ , \tag{3.27a}$$

$$\Gamma_{nn}(\omega) = \Gamma^{(1)}_{nn}(\omega) + \Gamma^{(2)}_{nn}(\omega) + \cdots \ . \tag{3.27b}$$

$\Delta^{(i)}$ and $\Gamma^{(i)}$, the $i$th approximations to $\Delta$ and $\Gamma$, are the real and the imaginary parts of the successive terms of the series Eq. (3.24), respectively. It is simple matter to interpret $\Delta(\omega)$ as the level shift and $\Gamma(\omega)$ as the level breadth originated by the radiation field on the virtual level of energy $\omega$. If the energy $\omega$ coincides with the energy $\omega_n$ of the level $n$, then one deals with the shift and breadth of the atomic level $n$. Recent experiment have confirmed such atomic perturbations.[25–27] Far from resonance the expressions for the corresponding quantities can be simplified. They are given in the Appendix.

Suppose that a particular level $(n_r l_r m_r)$ is energetically distant from the inital one by an integer multiple $p$ of the photon energy $\omega_p$, i.e., $\omega_r - \omega_0 = \omega_{r,0} \simeq p\omega_p$. Such a resonance will cause important changes in the multiphoton process. In order to simplify the discussion, let us consider a resonance occurring for $p = N - 1$ in a $N$-photon bound-free transition. Before writing the expression for the resonant ionization cross section we define the following quantity:

$$W^{(k)}_j(\omega_0 - \omega_k + N\omega_p) = \frac{F}{F_0 \omega_p} \left( \frac{\omega_{0,k} + N\omega_p}{(\omega_{0,k} + N\omega_p)^2 + (N^2 b + \frac{1}{2}\gamma_k)^2} + i \frac{N^2 b + \frac{1}{2}\gamma_k}{(\omega_{0,k} + N\omega_p)^2 + (N^2 b + \frac{1}{2}\gamma_k)^2} \right) p_{jk} p_{kj} \ . \tag{3.28}$$

After summation over $k$ the real and the imaginary parts of $W^{(k)}_j$ occur in the expression giving the first approximation to the shift and breadth of the virtual level $j$ of energy $\omega_0 + N\omega_p$, respectively. In using Eq. (3.28) we have

$$\sigma^{(N)}_{\mathrm{BF}} = \frac{4\pi^2 r_0^2}{\alpha^3} \frac{1}{\omega_p} \left( \frac{F}{F_0 \omega_p} \right)^{N-1} .$$

$$\times \left| \sum_{i_{N-2}} \frac{h_{N,r} h_{r,N-2} K^{(N-2)}_{N-2,0}}{\omega_{0,N-2} + (N-2)\omega_p - \sum_j W^{(j)}_{N-2}(\omega_{0,j} + (N-3)\omega_p) - \sum_{j \neq i_r} W^{(j)}_{N-2}(\omega_{0,j} + (N-1)\omega_p) - W^{(r)}_{N-2}(\omega_{0,r} + (N-1)\omega_p) - i(N-2)^2 b} \right.$$

$$\left. \times \frac{1}{\omega_{0,r} + (N-1)\omega_p - \sum_k W^{(k)}_r(\omega_{0,k} + (N-2)\omega_p) - i(N-1)^2 b} \right|^2 \rho(N\omega_p) \ . \tag{3.29}$$

From Eq. (3.29) it is clear that the resonance occurring for the absorption of the $(N-1)$th photon modifies the behavior of the function $W_{N-2}^{(r)}(\omega_{0,r} + (N-1)\omega_p)$ related to the absorption of the $(N-2)$th photon. As the photon energy approaches the resonant value $\omega_p^{(R)} = \omega_{r,0}/(N-1)$, the argument of the function $W_{N-2}^{(r)}(\omega_{0,r} + (N-1)\omega_p)$ becomes more and more small. Then, if we plot the real part of this function, say $\mathrm{Re}\{W_{N-2}^{(r)}(\omega_{0,r} + (N-1)\omega_p)\}$, against the photon energy $\omega_p$, the curve presents two extrema at $\omega_p = \omega_p^{\pm} = \omega_p^{(R)} \pm [(N-1)^2 b + \frac{1}{2}\gamma_r]/(N-1)^2$ and is zero at exact resonance; i.e., when $\omega_p = \omega_p^{(R)}$. On the other hand, $\mathrm{Im}\{W_{N-2}^{(r)}(\omega_{0,r} + (N-1)\omega_p)\}$ takes its maximum value at exact resonance and is a rapidly decreasing function everywhere. We deduce that its dominant damping effect is not obtained when $\omega_p = \omega_p^{\pm}$. For these values of the photon energy the quantity $\omega_{0,N-1} + (N-2)\omega_p - \mathrm{Re}(W_{N-2}^{(r)})$ exhibits a maximum (minimum) and a minimum (maximum) successively. Then, one observes that a resonance occurs for one of the two particular values $\omega_p^{\pm}$. The amplitude of the corresponding peak on the dispersion curve has a weak dependence with regard to the damping caused by the induced breadth. The detuning of the resonance gives rise to the reverse phenomenon. The above energy difference becomes more and more large and reaches a maximum for which the dispersion curve exhibits a minimum. The extrema of the function $\mathrm{Re}(W_{N-2}^{(r)})$ do not coincide exactly with those pointed on the dispersion curve. The reason lies in the fact that the resonance peak varies both with the photon flux and with the photon energy. It is straightforward that, as the intensity of the radiation increases, the resonance (or the antiresonance) condition is satisfied for values of $\omega_p$ which depart more and more from $\omega_p^{\pm}$. When low-intensity radiation sources are involved ($I < 10^{10}$ W/cm$^2$), the position of the resonance peak essentially depends on the damping terms. Thus, for light with narrow linewidth (< 1 Å), the photon energy corresponding to the resonance peak is very close to $\omega_p^{(R)}$ and is practically centered on this value.

Finally, aside from the usual resonance peak, the dispersion curve shows an additional peak originated by the process discussed so far. It must be pointed that these resonances give rise to important changes for the flux dependence of the cross section. One observes that $\sigma_{\mathrm{BF}}^{(N)}$ can take very large values when the intensity of the radiation source is varied. The corresponding curves are distorted and resonant bumps appear. Thus for particular values of the radiation intensity, the cross section no longer varies as $(F/F_0)^{N-1}$ but rather as $(F/F_0)^{N'-1}$. $N'$ being the effective order of non linearity, $N' = N + \epsilon(\omega_p)$. The absolute value of $\epsilon(\omega_p)$ may be large and its upper bound is essentially determined by the damping terms. On the other hand $\epsilon(\omega_p)$ takes on positive as well as negative values and its sign depends on the photon energy. The quantitative analysis of Sec. V will clarify the above discussion.

## IV. INDUCED SHIFT AND TRANSITION-CROSS-SECTION CALCULATIONS

The high-order matrix element occurring in the multiphoton transition cross section and induced shift can be calculated by the use of two different methods. The more precise uses the summation technique developed by Schwartz and Tieman. In this case, the formula must be worked out in the undisplaced energy picture. Then, the cross sections are not calculated from the closed form given by Eq. (3.17), but rather in considering all the contributions to the process coming from all higher order terms of the $S$-operator expansion. Nevertheless, the computation cannot be achieved in a simple way when the natural shift and breadth of intermediate states are taken into account. These quantities must be ignored in the numerical evaluation. This approximation does not affect the numerical results in all the off-resonant cases. On the other hand, the damping brought by the linewidth of the radiation field has the dominant effect in the close neighborhood of resonances. Finally, by this method, the probability amplitude of any multiphoton process is the sum of the probability amplitudes corresponding to the set of diagrams shown in Fig. 1.

It must be pointed out that the calculation in the undisplaced energy picture allows accurate evaluation of each term of the series occurring in the expression of the transition rates. Far from resonances a good precision is obtained by taking into account the first few terms of the series, but it is not so in the presence of resonances. In this case, we observe that the convergence becomes more and more weak as the wavelength of the radiation is tuned to the resonant one. A good accuracy requires a prohibitive number of terms. In order to avoid an unessential expense of computer time, we have investigated the resonances by the second method. The calculations are made in the displaced energy picture. The sums over intermediate states are carried out over a sufficient number of discrete states to get a good stability for the numerical result. For processes involving a small number of photons this procedure leads to satisfactory results. The lacking terms corresponding to the continuum part of the spectrum originate the principal source of uncertainty. We have calculated their contributions for a three-photon ionization of hydrogen in the $2S$ state. The error slightly varies with regard to the wavelength but its average value is less than 16%. In this

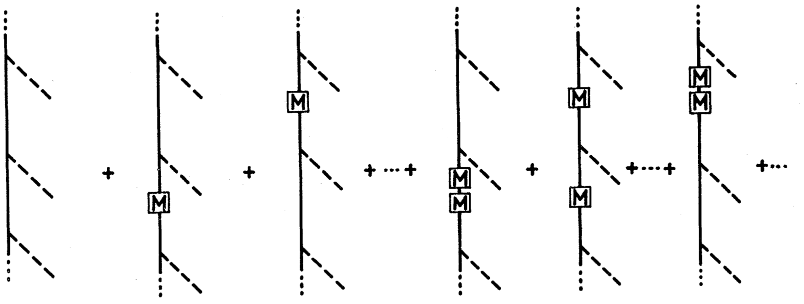

FIG. 1. Pictorial determination of higher-order diagrams contributing to a multiphoton process. M symbolizes the operator defined in Eq. (3.1).

section we give the general formula for the multiphoton transition cross section in the undisplaced energy picture. The induced level shifts, the bound-bound and bound-free multiphoton processes are treated on an equal footing with regard to the $N$th-order transition matrix element. The compact serial form for the relevant expressions can be easily handled for numerical analysis. All formulas are worked out in the atomic unit system. The high-order matrix element occurring in the calculation of level breadth, level shift, and multiphoton cross section is

$$K_{s,0}^{(s)}=\sum_{i_1 i_2\cdots i_{s-1}}\frac{\langle i_s|q_z|i_{s-1}\rangle\cdots\langle i_2|q_z|i_1\rangle\langle i_1|q_z|i_0\rangle}{[\omega_0-\omega_{s-1}+\epsilon(s-1)\omega_p-i\epsilon^2(s-1)b]\cdots[\omega_0-\omega_2+\epsilon(2)\omega_p-i\epsilon^2(2)b][\omega_0-\omega_1+\epsilon(1)\omega_p-i\epsilon^2(1)b]}\,, \tag{4.1}$$

where for convenience, we have introduced the parameters $\epsilon(1)$, $\epsilon(2)$, ..., $\epsilon(s-1)$. It is clear that the values taken by these parameters will determine the matrix element connected to the process we consider. We do not describe the method leading to the exact evaluation of $K_{s,0}^{(s)}$ since it has been discussed at length in previous papers.[6,7] Nevertheless, the functions $P$, $V$, and $y$ of Ref. 6 depend now on the set of parameters $\epsilon(1)$, $\epsilon(2)$, ..., $\epsilon(s-1)$. Finally, the set of equation which is to be solved may be written as

$$\begin{aligned}
&\mathcal{H}(\epsilon(1),l_1|p)\,y\left({}^{l_1,l_2,\ldots,l_{s-1},l_s}_{\epsilon(1),\epsilon(2),\ldots,\epsilon(s-1)}|p\right)=\mathfrak{D}(l_1,l_2|p)\,y\left({}^{l_2,l_3,\ldots,l_{s-1},l_s}_{\epsilon(2),\epsilon(3),\ldots,\epsilon(s-1)}|p\right)-\delta(\epsilon(1),0)S^{(l_1)}(n_0,l_1|p)P\left({}^{l_1,l_2,\ldots,l_{s-1},l_s}_{\epsilon(2),\ldots,\epsilon(s-1)}\right),\\
&\mathcal{H}(\epsilon(2),l_2|p)\,y\left({}^{l_2,l_3,\ldots,l_{s-1},l_s}_{\epsilon(2),\epsilon(3),\ldots,\epsilon(s-1)}|p\right)=\mathfrak{D}(l_2,l_3|p)\,y\left({}^{l_3,l_4,\ldots,l_{s-1},l_s}_{\epsilon(3),\epsilon(4),\ldots,\epsilon(s-1)}|p\right)-\delta(\epsilon(2),0)S^{(l_2)}(n_0,l_2|p)P\left({}^{l_2,l_3,\ldots,l_{s-1},l_s}_{\epsilon(3),\ldots,\epsilon(s-1)}\right)\\
&\qquad\vdots\\
&\mathcal{H}(\epsilon(s-1),l_s|p)\,y\left({}^{l_{s-1},l_s}_{\epsilon(s-1)}|p\right)=\mathfrak{D}(l_{s-1}l_s|p)\,y(l_s|p)-\delta(\epsilon(s-1),0)S^{(l_{s-1})}(n_0,l_{s-1})P(l_{s-1},l_s)\;.
\end{aligned} \tag{4.2}$$

$\mathcal{H}$, $\mathfrak{D}$, and $S$ are defined by

$$\mathcal{H}(\epsilon,l|p)\equiv[p^2+2(\omega_0+\epsilon\omega_p-i\epsilon^2b)]\frac{d}{dp}+2[(l+1)p-1]\;, \tag{4.3}$$

$$\mathfrak{D}(l_i,l_j|p)=(l_i-l_j+1)\left((2l_i+1)\frac{d}{dp}+p\frac{d^2}{dp^2}\right)+(l_j-l_i+1)p\;, \tag{4.4}$$

$$S_{(n,l|p)}^{(l)}=\frac{d^{l+2}}{dp^{l+2}}\,\mathcal{L}\{R(n,l|r)\}\;, \tag{4.5}$$

where $\mathcal{L}\{\,\}$ stands for the Laplace transform. The additional terms

$$\{\delta(\epsilon_j,0)S^{(l_j)}(n_0l_j|p)P\left({}^{l_j,l_{j+1},\ldots,l_{s-1},l_s}_{\epsilon(j+1),\ldots,\epsilon(s-1)}\right)\}$$

on the right-hand side of Eq. (4.2) arise from the exclusion of the state $(n_0,l_j,m_j)$, in the summations over intermediate states, when $\epsilon(j)=0$.

The induced shift of the $i$th level is given, up to the $N$th-order approximation, by

$$\Delta_{ii}(-\omega_i)=\sum_{s=1}^{N-1}\left(\frac{F}{\omega_pF_0}\right)^s\prod_{j=0}^{s-1}\,[\delta(\epsilon(2j+1),1)+\delta(\epsilon(2j+1),-1)]\,\delta(\epsilon(2j),0)\mathcal{R}_{ii}^{(0)}(\{\epsilon(2s-1)\})\;, \tag{4.6}$$

where the notation $\{\epsilon_i\}$ is used for the set $\epsilon(1)$, $\epsilon(2)$, ..., $\epsilon(i)$; $F$ is the photon flux in CGS units and $F_0=3.22\times10^{34}\ \mathrm{cm^{-2}\,sec^{-1}}$. The function $\mathcal{R}_{ii}$ is defined as

$$\mathcal{R}_{ij}^{(\nu)}(\{\epsilon(2s-1)\})=G_\nu(l_i,m_i;\ldots;l_{2s-1},m_{2s-1};l_j,m_j)P_\nu\left({}^{l_i,l_1,\ldots,l_k,\ldots,l_j}_{\epsilon(1),\epsilon(k),\ldots}\right)\;. \tag{4.7}$$

The angular function $G$ and the radial function $P$ have the same meaning as the ones used in Ref. 6.

For bound-bound and bound-free transitions, the values taken by the parameters $\epsilon(i)$ are determined through formulas which are somewhat more complicated than the preceding one. This is because we must take into account all the contributions arising from the repeated occurrence of processes involving absorption-emission and emission-absorption of photons. The values of the parameters $\epsilon$ are easily determined if we define the following functionals:

$$Y_\nu^{(m)}(j_m)=\delta'(\epsilon(\eta_m+2m-1),\ \xi'^{(\nu)}(\eta_m)+1)+\delta'(\epsilon(\eta_m+2m-1),\ \xi'^{(\nu)}(\eta_m)-1)\ , \tag{4.8}$$

$$Z_\nu^{(m)}(j_m)=\prod_{l=0}^{j_m}\ \delta'[\epsilon(\eta_{m-1}+l+2m-2),\ \xi'^{(\nu)}(\eta_{m-1}+l)]\ , \tag{4.9}$$

where $\xi'^{(\nu)}=\xi^{(\nu)}/\omega_p$, $\xi^{(\nu)}$ being given by Eq. (3.23). $\delta'(\alpha,\beta)$ is zero unless any number $\alpha$ coincides with $\beta$, in which case it equals unity. $\eta_m$ is defined in terms of the integer $j_m$ as

$$\eta_m=\eta_{m-1}+j_m\ ,\quad \eta_0=0\ . \tag{4.10}$$

In combining Eqs. (4.8) and (4.9) we define

$$W_\nu^{(k)}(\{j_{k+1}\})=\left(\prod_{m=1}^{k}\ Z_\nu^{(m)}(j_m)Y_\nu^{(m)}(j_m)\right)Z^{(k+1)}(j_{k+1})\ . \tag{4.11}$$

Therefore, the cross section for the multiphonon bound-bound transition which takes place between the states $(n_0l_0m_0)$ and $n_Nl_Nm_N)$ may be written as

$$\sigma_{\mathrm{BB}}^{(N)}=\frac{8\pi}{27}r_0^2\left(\frac{F}{F_0\omega_p}\right)^{N-2}\frac{\omega_s}{\omega_p}\prod_{\nu=1}^{N}\Bigg|Z_\nu^{(1)}(N-1)\mathcal{R}_{N,0}^{(\nu)}(\{\epsilon(N-1)\}) + \sum_{k=1}^{\infty}\left(\frac{F}{F_0\omega_p}\right)^{k}\sum_{\{j_k\}}W_\nu^{(k)}(\{j_{k+1}\})\mathcal{R}_{N,0}^{(\nu)}(\{\epsilon(N-1+2k)\})\Bigg|^2 g\{(N-1)\omega_p-\omega_s\}\ . \tag{4.12}$$

$r_0$ is the classical electron radius and the index $\nu$ is related to the emission of the single photon of energy $\omega_s$ in the Raman-like process we consider.

The ionization cross section for the state $(n_0l_0m_0)$ is

$$\sigma_{\mathrm{BF}}^{(N)}=\frac{4\pi^2r_0^2}{\alpha^3}\frac{1}{\omega_p}\left(\frac{F}{F_0\omega_p}\right)^{N-1}\Bigg|Z_N^{(1)}(N-1)\mathcal{R}_{N,0}^{(0)}(\{\epsilon(N-1)\}) + \sum_{k=1}^{\infty}\left(\frac{F}{F_0\omega_p}\right)^{k}\sum_{\{j_k\}}W_N^{(k)}(\{j_{k+1}\})\mathcal{R}_{N,0}^{(0)}(\{\epsilon(N-1+2k)\})\Bigg|^2\rho(N\omega_p)\ , \tag{4.13}$$

where $\alpha$ is the fine-structure constant. In Eqs. (4.12) and (4.13) the sum over $\{j_k\}$ is achieved if we make use of the following relations:

$$j_1+j_2+\cdots+j_{k+1}=N-1\ ,\quad j_1\geq 1\ . \tag{4.14}$$

## V. NUMERICAL RESULTS

The induced shifts and induced breadths related to some energy levels of H are computed at 10 600 Å. The value of the level shift, calculated up to the third approximation, is to be deduced from

$$\Delta_{nl}=A_{nl}F+B_{nl}F^2+C_{nl}F^3\ . \tag{5.1}$$

In the first approximation, the level breadth is to be determined from

$$\Gamma_{nl}=D_{nl}F\ .$$

The coefficients $A_{nl}$, $B_{nl}$, $C_{nl}$, and $D_{nl}$ occurring in Eq. (5.1) are given in Table I where $F$ is expressed in CGS units and $\Delta_{nl}$ and $\Gamma_{nl}$ are in eV. As an example, in Fig. 2 the values of several level shifts are plotted against the photon flux $F$. Apart from the 3$S$ level, we are roughly concerned with linear flux dependence. On the other hand, the shift of the 1$S$ level is such that the ionization potential is slightly raised. From this last result, an important lowering of ionization potential cannot be invoked in order to explain the observed abnormal value of the order of nonlinearity for multiphoton ionization processes. A further remark is that the 1$S$ and 2$S$ levels have nonvanishing induced breadths with regard to electric dipole transitions.

The theory presented so far is applied to a typical three-photon ionization of hydrogen in the 2$S$ state. The calculations are worked out in the displaced energy picture. We have ignored the contributions due to corrections to external electron lines. We

TABLE I. Coefficients of Eqs. (5.1) and (5.2) for some hydrogen states. The number in parentheses indicates power of 10.

| $nl$ | $1s$ | $2S$ | $2P$ | $3S$ | $3P$ | $3D$ |
|---|---|---|---|---|---|---|
| $A_{nl}$ | +1.9795(−32) | +2.6225(−32) | +2.9567(−32) | −8.0383(−33) | −1.7472(−32) | −3.3609(−32) |
| $B_{nl}$ | −2.4248(−65) | −1.0405(−64) | −5.0932(−64) | −2.0225(−63) | −2.2191(−63) | −2.6586(−63) |
| $C_{nl}$ | +1.9928(−96) | +2.2211(−96) | +1.4412(−95) | +6.7477(−93) | −1.1769(−94) | −3.5059(−93) |
| $D_{nl}$ | +1.8174(−40) | +1.0777(−39) | +5.8063(−40) | +5.0239(−39) | +1.8044(−39) | +4.3055(−39) |

have limited ourselves to the first approximation for the induced shifts. The radiation wavelengths are chosen in such a way that the second absorbed photon originates a resonance with the $4S$ and $4D$ intermediate level. This happens when $\lambda_p \sim 9725.4$ Å. Simultaneously, one observes that a resonance is induced with the $3P$ level for the first absorbed photon.

Figure 3 shows the dispersion curves for different values of the radiation linewidth (solid lines). The dashed lines represent the energy difference $\Delta\mathcal{E}_{3P}(\omega_p) = \omega_{2S,3P} + \omega_p - \mathrm{Re}[W_{3P}^{4S}(\omega_{2S,4S} + 2\omega_p)]$ and the dotted lines represent the function $\mathrm{Im}[W_{3P}^{4S}(\omega_{2S,4S} + 2\omega_p)] + b$. It is straightforward that the first resonance peak at 9723 Å is due to the close neighborhood of the $3P$ level and the displaced virtual level of energy $\omega_{2S} + \omega_p$. At this wavelength the induced damping is small with regard to the radiation damping. Then, the amplitude of the peak depends on the radiation linewidth. As the wavelength approaches the value 9725.4 Å, this resonance is rapidly detuned while the induced breadth increases. When $\lambda_p \sim 9725$ Å the induced breadth reaches its maximum value and $\mathrm{Re}[W_{3P}^{4S}(\omega_{2S,4S} + 2\omega_p)]$ vanishes. Then $\Delta\mathcal{E}_{3P}(\omega_p)$ exhibits an additional minimum value and a distortion on the dispersion curve appears. Apart from this slight modification, it is clear that for $\lambda_p \simeq 9725$ Å the effect of the one-photon resonance is practically washed out. Then, in order to explain the second resonance peak appearing on the dispersion curves, one must refer to Fig. 4. Here, the dashed lines represent the energy difference

$$\Delta\mathcal{E}_{4S}(2\omega_p) = \omega_{2S,4S} + 2\omega_p - \mathrm{Re}\left(\sum_j W_{4S}^j(\omega_{2S,j} + \omega_p)\right) \quad ,$$

whereas the dotted lines represent the damping term

$$\mathrm{Im}\left(\sum_j W_{4S}^j(\omega_{2S,j} + \omega_p)\right) + 4b \quad .$$

For this two-photon resonance the induced shift and induced breadth have very small variations

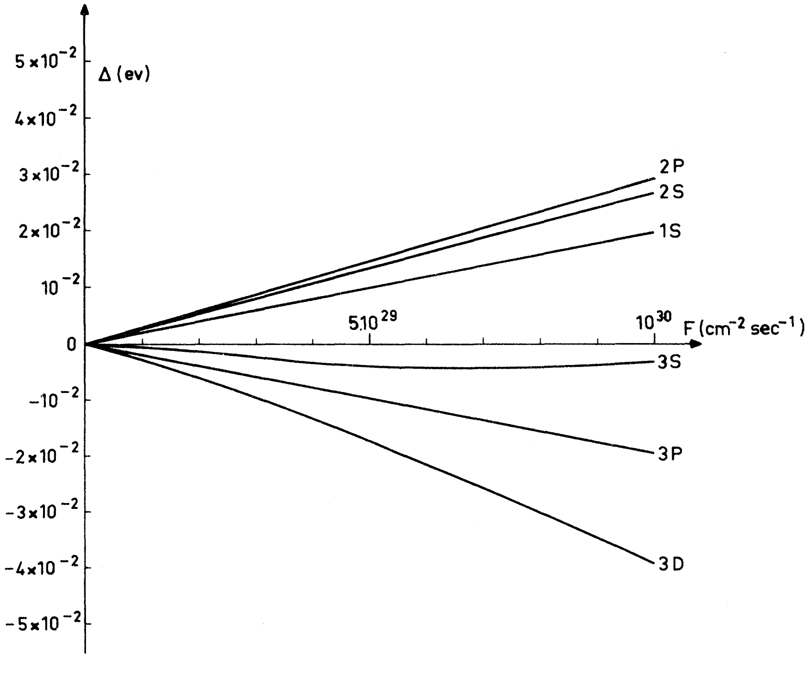

FIG. 2. Photon flux dependences of induced shifts for some energy levels of the hydrogen atom. The wavelength of the radiation is 1.06 $\mu$.

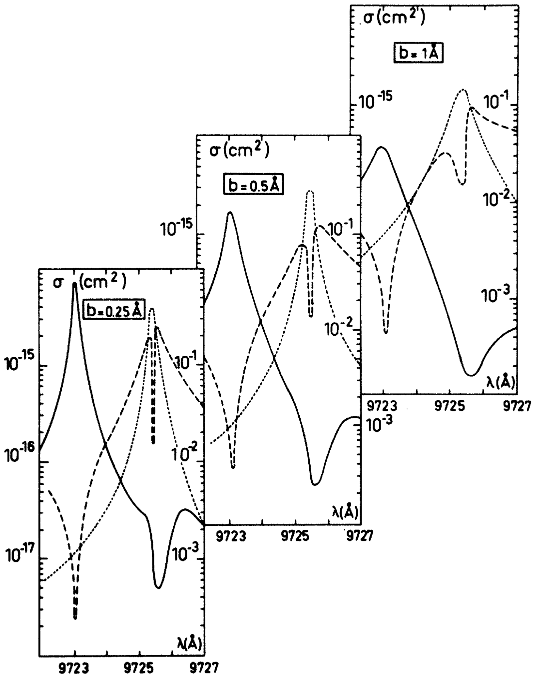

FIG. 3. Three-photon ionization cross section of H in the $2S$ state as a function of the wavelength (solid lines). The dashed lines represent the real part of the denominator related to the first absorbed photon. The dotted lines are the plots for the corresponding damping term. The values of these two last quantities are to be read on the right-hand-side scale. The photon flux is $10^{29}$ cm$^{-2}$ sec$^{-}$

with regard to the radiation wavelength and have small amplitude. In contrast to the preceding case, the resonance condition is obtained when the energy difference $\omega_{2S,4S}+2\omega_p$ is made as small as possible and comparable to $\mathrm{Re}\{\sum_j W^j_{4S}(\omega_{2S,j}+\omega_p)\}$. In practice, the one-photon and two-photon resonances cannot be identified without a careful analysis. One observes that the most sensitive effects are originated by the one-photon resonance. This is illustrated in Figs. 5 and 6 where we have plot the three-photon ionization cross section against the photon flux $F$. The wavelengths are close to that which induces a two-photon resonance on the $4S$ and $4D$. The two sets of curves correspond to equally spaced wavelengths located on both sides of 9725 Å. The radiation linewidth is equal to 0.25 Å.

In Fig. 5, each curve shows a slope which is greater than two, the value predicted by the lowest-order theory. For a particular value of the radiation intensity, a one-photon resonance with the $3P$ level appears. The corresponding resonance peak is displaced towards lower intensity values when the wavelength approaches the resonant one. For values of the wavelength exceeding 9725 Å the curves shown in Fig. 6 exhibit decreasing values for the order of nonlinearity; in varying the photon flux a one-photon resonance with the $2P$ level arises and the absolute value of the slope is rapidly increased. It must be noted that for a given wavelength the cross section shows several peaks as the photon flux is varied. Each of them corresponds to a particular level for which the resonance condition is satisfied. For example, in Fig. 5 the curve representing the plots of the cross section at 9725 Å has two resonance peaks. The first is connected to a one-photon resonance with the $3P$ level and the last involves the $5P$ level.

In Fig. 7 the order of nonlinearity for this three-photon resonant ionization is plotted against the wavelength (solid lines). We have considered the previous typical values for the radiation linewidth, but here the photon flux is $F=10^{28}$ cm$^{-2}$ sec$^{-1}$. A comparison with the corresponding dispersion curves (dashed lines), shows that the important variation in $N'$ are originated by the one-photon resonance.

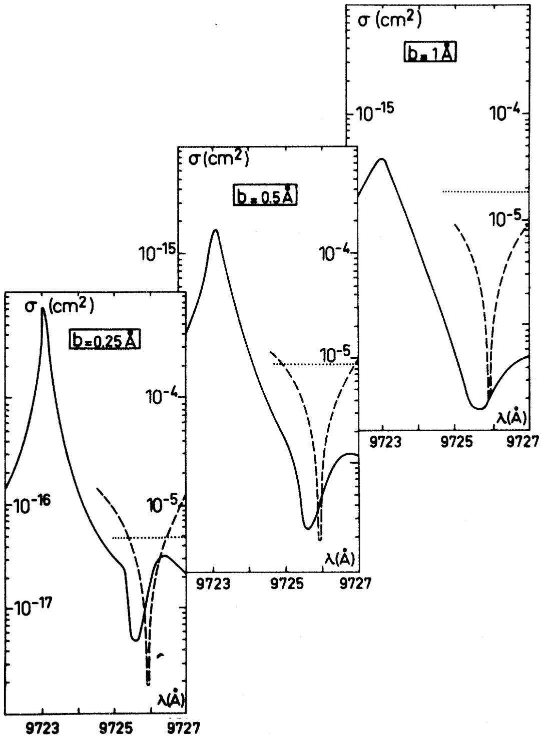

FIG. 4. Dispersion curves for the three-photon ionization cross section of H in the $2S$ state (solid lines). The real and imaginary parts of the denominator corresponding to the second absorbed photon are represented by the dashed lines and dotted lines, respectively. The right-hand-side scale refers to these two last quantities. The value for the photon flux is $10^{29}$ cm$^{-2}$ sec$^{-1}$.

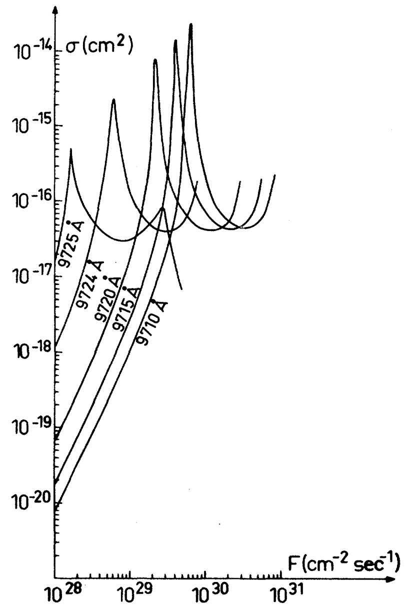

FIG. 5. Three-photon ionization cross section as function of the photon flux. The wavelengths considered are greater than 9725.4 Å. The first peaks observed in the curves are produced by a one-photon resonance with the 3*P* level. At 9725 Å the second peak corresponds to a one-photon resonance with the 5*P* level.

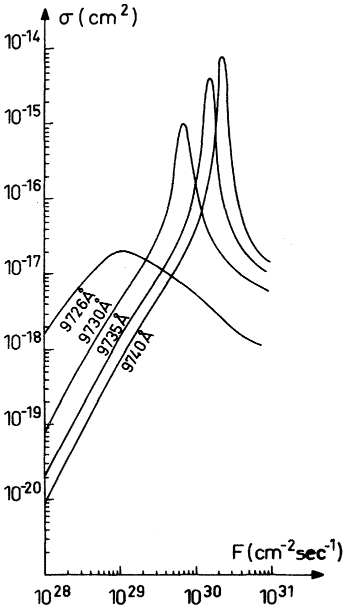

FIG. 6. Three-photon ionization cross section as a function of the photon flux. The wavelengths we consider are smaller than 9725.4 Å. The peaks shown in the curves are produced by the one-photon resonance with the 2*P* level.

## VI. CONCLUSION

This emphasizes the fundamental role played by the higher-order terms provided by the *S*-operator expansion. We have shown that they must be taken into account when a resonant multiphoton process is considered or when high-intensity sources are involved. In this last case a question arises about the validity of the perturbation theory.[28] Then, we have limited ourselves to small photon flux values. After summation of all nondivergent higher-order terms, the expression for the perturbed Green's function has been set up into a close form. It exhibits significant modifications concerning its energy denominator.[29] One observes that the virtual levels, occurring in any multiphoton process, are shifted and broadened. The calculation of the shift and breadth have been carried out when the energy of a virtual level coincides with the energy of a real level. We have seen that, in the hydrogen atom, the ionization potential is slightly raised. These perturbations in the energy of the virtual levels strongly modify the behavior of any resonant multiphoton process. The example we have considered in Sec. V has shown that a two-photon resonance induces a one-photon resonance in a three-photon absorption process. In taking into account the damping caused by the radiation, one observes that the one-photon resonance produces the dominant modifications. When the value for the radiation linewidth is increased, the two-photon resonance effects becomes negligible with regard to that induced by the one-photon resonance.

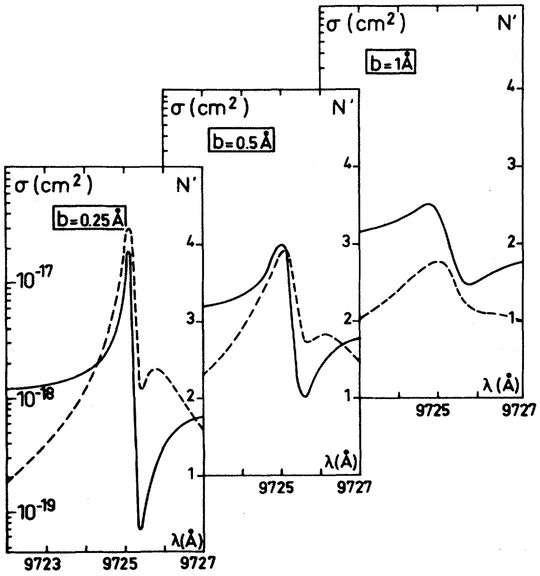

FIG. 7. Effective order of nonlinearity *N'* as a function of the wavelength (solid lines). The dashed lines represent the three-photon ionization cross section. The value for the photon flux is $10^{28}$ cm$^{-2}$ sec$^{-1}$.

A confusion can exist for the interpretation of the remaining peak in the dispersion curve and only a detailed analysis yields a correct identification.

It must be noted that the discussion can be extended to a more general case. One can imagine that, in a $N$-photon absorption process, the $(N-p)$th absorbed photon gives rise to a resonance. Then, one would observe the resonances induced by the $(N-p-1)$th and $(N-p+1)$th absorbed photon.

We have shown that in the presence of a resononance, the order of nonlinearity is a rapidly varying function of the radiation wavelength. Therefore, it is clear that this quantity no longer represents the number of absorbed quanta. These new effects predicted by this theory have been confirmed by recent experiments.[15-17] Nevertheless, their physical interpretation will need further careful investigations.

## ACKNOWLEDGMENTS

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## APPENDIX

We derive here the explicit forms of level shift and level breadth which are used in numerical computations. For this purpose, we split the complex perturbing term $W_{nn}(-\omega_n)$ given by Eq. (3.24) into its real and imaginary parts. When this is done one finds

$$W_{nn}(-\omega_n)=\Delta_{nn}^{(1)}(-\omega_n)+\Delta_{nn}^{(2)}(-\omega_n)+\cdots+\tfrac{1}{2}i[\Gamma_{nn}^{(1)}(-\omega_n)+\Gamma_{nn}^{(2)}(-\omega_n)+\cdots] \;, \tag{A1}$$

where $\Delta_{nn}^{(i)}(\omega_n)$ and $\Gamma_{nn}^{(i)}(\omega_n)$ are the $i$th approximation to the shift and breadth connected with the $n$th level, respectively.

In the nonrelativistic dipole approximation it is easily shown from Eqs. (3.24) and (3.25) that when no resonance occurs in $W_{nn}(-\omega_n)$, one gets

$$\begin{aligned}
\Delta_{nn}^{(1)}(-\omega_n) &\sim \frac{F}{F_0\omega_p}\sum_i\left(\frac{(p_z)_{ni}(p_z)_{in}}{\omega_i-\omega_n+\omega_p}+\frac{(p_z)_{ni}(p_z)_{in}}{\omega_i-\omega_n-\omega_p}\right) \;,\\
\Delta_{nn}^{(2)}(-\omega_n) &\sim \left(\frac{F}{F_0\omega_p}\right)^2\sum_i\sum_{j\neq n}\sum_k\left(\frac{(p_z)_{ni}(p_z)_{ij}}{\omega_i-\omega_n+\omega_p}+\frac{(p_z)_{ni}(p_z)_{ij}}{\omega_i-\omega_n-\omega_p}\right)\frac{1}{\omega_j-\omega_n}\left(\frac{(p_z)_{jk}(p_z)_{kn}}{\omega_k-\omega_n+\omega_p}+\frac{(p_z)_{jk}(p_z)_{kn}}{\omega_k-\omega_n-\omega_p}\right) \;,\\
&\vdots
\end{aligned} \tag{A2}$$

$$\begin{aligned}
\Gamma_{nn}^{(1)}(-\omega_n) &\sim \frac{F}{F_0\omega_p}\sum_i\left(\frac{(p_z)_{ni}(p_z)_{in}}{\omega_i-\omega_n+\omega_p}+\frac{(p_z)_{ni}(p_z)_{in}}{\omega_i-\omega_n-\omega_p}\right)\gamma_{ii}(-\omega_i) \;,\\
\Gamma_{nn}^{(2)}(-\omega_n) &\sim \sum_i\sum_{j\neq n}\sum_k\left(\frac{(p_z)_{ni}(p_z)_{ij}}{(\omega_i-\omega_n+\omega_p)^2}+\frac{(p_z)_{ni}(p_z)_{ij}}{(\omega_i-\omega_n-\omega_p)^2}\right)\frac{\gamma_{ii}\gamma_{jj}\gamma_{kk}}{(\omega_j-\omega_n)^2}\left(\frac{(p_z)_{jk}(p_z)_{kn}}{(\omega_k-\omega_n+\omega_p)^2}+\frac{(p_z)_{jk}(p_z)_{kn}}{(\omega_k-\omega_n-\omega_p)^2}\right) \;,\\
&\vdots
\end{aligned} \tag{A3}$$

where $p_z$ is the $z$ component of the electron momentum operator, i.e., the component of the momentum operator in the direction of the polarization of the radiation.

Apart from $F$ which is expressed in CGS units all quantities appearing in (A2) and (A3) are dimensionless multiples of atomic units.

Because of the smallness of the natural linewidth ($\gamma \leq 1.5\times 10^{-8}$ a.u.), we see that the dominant contribution to $\Gamma$ is brought by $\Gamma^{(1)}$, then the higher-order approximations to $\Gamma$ will be neglected.

It is not so for $\Delta$. Thus it may happen that with increasing values of the flux the term $\Delta^{(2)}$ brings out significant contributions and then acts as a smoothing factor which limits the growth of $\Delta$ with respect to $F$.

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[1] W. Zernik, Phys. Rev. **132**, 320 (1963); Phys. Rev. **133**, 117 (1964); Phys. Rev. **135**, 51 (1964); Phys. Rev. **172**, 420 (1968).

[2] H. B. Bebb and A. Gold, Phys. Rev. **143**, 1 (1966).

[3] H. B. Bebb, Phys. Rev. **149**, 25 (1966); Phys. Rev. **153**, 23 (1967).

[4] V. M. Morton, Proc. Phys. Soc. Lond. **92**, 301 (1967).

[5] B. A. Zon, N. L. Manakov, and L. P. Rapoport, Zh. Eksp. Teor. Fiz. **61**, 968 (1971) [Sov. Phys.-JETP **34**, 515 (1972)].

[6] Y. Gontier and M. Trahin, Phys. Rev. **172**, 83 (1968); Phys. Rev. A **4**, 1896 (1971).

[7] Y. Gontier and W. Trahin, Phys. Lett. **36**, 463 (1971).

[8] N. B. Delone and G. K. Piskova, Zh. Eksp. Teor. Fiz. **58**, 753 (1970) [Sov. Phys. JETP 31, 403 (1970)].

[9] P. Agostini *et al.*, Phys. Lett. **31**, 367 (1970); Proceedings of

the Sixth International Quantum Electronics Conference, 1970, p. 92 (unpublished).

[10] C. Schwartz, Ann. Phys. (N.Y.) **6**, 156 (1959).

[11] C. Schwartz and J. J. Tieman, Ann. Phys. (N.Y.) **6**, 178 (1959).

[12] A. Dalgarno and J. T. Lewis, Proc. Roy. Soc. Lond. **233**, 70 (1955).

[13] F. Low, Phys. Rev. **88**, 53 (1952).

[14] N. D. Sen Gupta, J. Phys. A **5**, 401 (1972).

[15] B. Held *et al.*, Phys. Rev. Lett. **30**, 423 (1973).

[16] J. Bakos *et al*., Phys. Lett. **41**, 163 (1972).

[17] G. A. Delone and N. B. Delone, Zh. Eksp. Teor. Fiz. Pis'ma Red. **10**, 413 (1969) [JETP Lett. **10**, 265 (1969)].

[18] L. P. Kotova and M. V. Terent'ev, Zh. Eksperim. Teor. Fiz. **52**, 732 (1967) [Sov. Phys.-JETP **25**, 481 (1967)]; V. I. Ritus, Zh. Eksperim. Teor. Fiz. **51**, 1544 (1966) [Sov. Phys.-JETP **24**, 1041 (1967)]; V. A. Davydkin, B. A. Zon, N. L. Manakov, and L. P. Rapoport, Zh. Eksperim. Teor. Fiz. **60**, 124 (1971) [Sov. Phys.-JETP **33**, 70 (1971)].

[19] P. Lambropoulos, Phys. Rev. **164**, 84 (1967).

[20] S. H. Autler and C. H. Townes, Phys. Rev. **100**, 730 (1955).

[21] C. Cohen-Tannoudji and S. Haroche, J. Phys. (Paris) **30**, 125 (1969).

[22] W. Heitler, *The Quantum Theory of Radiation,* 3rd ed. (Clarendon, Oxford, England, 1954), Chap. 4, Sec. 16.

[23] B. R. Mollow, Phys. Rev. **175**, 1555 (1968).

[24] G. S. Agarwal, Phys. Rev. A **1**, 1445 (1970).

[25] E. B. Aleksandrov *et al.,* Zh. Eksp. Teor. Fiz. Pis'ma Red. **3**, 85 (1966) [JETP Lett. **3**, 53 (1966)].

[26] P. Platz, Phys. Lett. **27**, 714 (1968); Appl. Phys. Lett. **14**, 168 (1969); Appl. Phys. Lett. **16**, 70 (1970).

[27] A. M. Bonch-Bruevich, H. N. Kostin, and V. A. Kodovoi, Usp. Fiz. Nauk **93**, 71 (1967) [Sov. Phys.-Usp. **10**, 637 (1968)].

[28] H. R. Reiss, Phys. Rev. A **1**, 803 (1970); Phys. Rev. D **4**, 3533 (1971); Phys. Rev. A **6**, 817 (1972).

[29] Y. Gontier and M. Trahin, C.R. Acad. Sci. (Paris) **266**, 1177 (1968).