Electron-hydrogen scattering in a chaotic laser field

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Elastic scattering of electrons from hydrogen atoms in a chaotic laser field is considered. The necessary ensemble averages are evaluated assuming a field correlation function with an associated Lorentz spectrum. For nonzero bandwidths, the double differential cross sections exhibit peaks around the incident energy as well as around the atomic transition energies, all sidebands having a spacing of the mean laser energy. For zero bandwidth, the (single) differential cross sections reduce to those due to a coherent field, averaged over a Gaussian probability distribution. Some representative numerical results are presented and discussed.

I. INTRODUCTION

The influence of field fluctuations on laser-assisted charged-particle scattering has been the subject of some recent studies.[1-4] In a recent communication[5] we had summarized the key results for electron scattering from hydrogen atoms in a chaotic laser field. While Ref. 5 was mainly concerned with the new structures in the double differential cross sections and their relation to the laser line shape, this paper describes the ensemble-averaging procedures used in calculating the exact scattering cross sections, and also presents a general discussion of various results.

II. FORMULATION

Consider the elastic scattering of electrons by hydrogen atoms in a plane-polarized classical electromagnetic field, whose amplitude and phase undergo Gaussian fluctuations. In the Coulomb gauge and dipole approximation the incident electron of average momentum $\mathbf{k}$ is represented by

$$\chi_{\mathbf{k}}(\mathbf{r},t)=\exp\left[i\mathbf{k}\cdot\mathbf{r}-i\int_{-\infty}^{t}\left[\mathbf{k}-\frac{e\mathbf{A}(\tau)}{c}\right]^2\Big/2\,d\tau\right]. \tag{1}$$

where $\mathbf{A}$ is the vector potential. (We use atomic units throughout, with $e=-1$ for electrons.) For the hydrogen atom, first-order perturbation theory yields the ground-state wave function

$$\psi_0(\mathbf{r},t)=\left[e^{-i\omega_0 t}\phi_0(\mathbf{r})+i\sum_k M_{k0}e^{-(\gamma_k+i\omega_k)t}\phi_k(\mathbf{r})\int_{-\infty}^{t}e^{(\gamma_k+i\omega_{k0})t'}E(t')dt'\right]e^{ie\mathbf{A}\cdot\mathbf{r}/c}, \tag{2}$$

where E is the field strength, ϕ_k is an unperturbed atomic state of energy ω_k, γ_k is the linewidth for the transition $|k\rangle\rightarrow|0\rangle$, and

$$M_{k0}=\langle k\,|\,e\mathbf{r}\cdot\hat{\mathbf{E}}\,|\,0\rangle, \tag{3}$$

with $\hat{\mathbf{E}}$ denoting the direction of polarization of the field. The linewidths are of no real significance in most of the following calculations, and hence they will be retained only when their presence is essential for obtaining finite results (cf. Appendix B). Further, in order to reduce the complexity of some of the algebraic expressions, we shall sometimes make the reasonable approximation that the photon frequency $\omega \ll \omega_{k0}$.

The average first-Born-transition probability per unit time for elastic scattering (direct), in which the initial and final momenta of the electron are, respectively, $\mathbf{k}_i$ and $\mathbf{k}_f$, is given by

$$\langle W_{fi}\rangle=\left\langle\lim_{T\rightarrow\infty}\frac{1}{2T}\left|\int_{-T}^{T}dt\,\langle\chi_{\mathbf{k}_f}\psi_0\,|\,V\,|\,\psi_0\chi_{\mathbf{k}_i}\,|\,\rangle\right|^2\right\rangle, \tag{4}$$

where

$$V=\frac{1}{|\mathbf{r}_1-\mathbf{r}_2|}-\frac{1}{r_1}\,, \tag{5}$$

$\mathbf{r}_1$ and $\mathbf{r}_2$ being the coordinates of the incident and atomic electrons, respectively. (Both ensemble averages and inner products are denoted by angular brackets; the distinction is clear from the context.) From Eqs. (1)–(4) we get

$$\langle W_{fi}\rangle=\left\langle \lim_{T\to\infty}\frac{1}{2T}\left|\int_{-T}^{T}dt\exp\left[-i\left[E_{if}t-\Omega\int_{-\infty}^{t}A(t')dt'\right]\right.\right.\right.$$
$$\left.\left.\left.\times\left[\mathscr{V}_{00}+i\sum_k \mathscr{V}_{0k}M_{k0}F_k(t)-i\sum_k \mathscr{V}_{k0}M_{0k}F_k^*(t)\right]\right]\right|^2\right\rangle, \tag{6}$$

where

$$\Omega=e\mathbf{q}\cdot\mathbf{E}/c,\quad \mathbf{q}=\mathbf{k}_i-\mathbf{k}_f,\quad E_{if}=(k_i^2-k_f^2)/2\,,$$
$$\mathscr{V}_{nk}=\int\langle n\,|\,e^{i\mathbf{q}\cdot\mathbf{r}_1}V\,|\,k\rangle d\mathbf{r}_2\,, \tag{7}$$

and

$$F_k(t)=\int e^{-i\omega_{k0}(t-t')}E(t')dt'\,. \tag{8}$$

Noting that, because of the dipole selection rules, $\mathscr{V}_{0k}=\mathscr{V}_{k0}=-\mathscr{V}_{0k}^*$ and M_{0k} is real, the various terms in Eq. (6) can be appropriately grouped to yield

$$\langle W_{fi}\rangle=\left\langle\hat P\left[\mathscr{V}_{00}^2+i\mathscr{V}_{00}\sum_k\mathscr{V}_{0k}M_{0k}[F_k(t_2)-F_k^*(t_2)-F_k(t_1)+F_k^*(t_1)]\right.\right.$$
$$\left.\left.-\sum_k\sum_n\mathscr{V}_{0k}M_{k0}\mathscr{V}_{0n}M_{n0}[F_k(t_2)F_n^*(t_1)-F_k(t_2)F_n(t_1)-F_k^*(t_2)F_n^*(t_1)+F_k^*(t_2)F_n(t_1)]\right]\right\rangle, \tag{9}$$

where the operator $\hat P$ is defined as

$$\hat P\equiv\lim_{T\to\infty}\frac{1}{2T}\int_{-T}^{T}dt_1\int_{-T}^{T}dt_2\exp\left[iE_{if}(t_1-t_2)+i\Omega\int_{t_1}^{t_2}A(u)du\right]. \tag{10}$$

Evaluation of the various ensemble averages figuring in Eq. (9) may now be carried out, as described in Sec. III.

III. CALCULATION OF ENSEMBLE AVERAGES

Basically three types of ensemble averages appear in Eq. (9). The simplest of these is

$$\langle\hat P\rangle=\lim_{T\to\infty}\frac{1}{2T}\int_{-T}^{T}dt_1\int_{-T}^{T}dt_2\exp\left[E_{if}(t_1-t_2)-\frac{\Omega^2}{2}\int_{t_1}^{t_2}\int_{t_1}^{t_2}\langle A(u)A(v)\rangle du\,dv\right], \tag{11}$$

where the Gaussian property of A has been used. Trading the variable t_2 for $t=t_1-t_2$, Eq. (11) can be written as

$$\langle\hat P\rangle=H(0,E_{fi})\,, \tag{12}$$

where

$$H(x,y)=2\,\mathrm{Re}\int_0^\infty J(t)e^{-(x+iy)t}dt\,, \tag{13}$$

$$J(t)=\exp\left[\frac{-\Omega^2}{2}\int_0^t\int_0^t\langle A(u)A(v)\rangle du\,dv\right]. \tag{14}$$

To proceed further, the field correlation functions have to be specified. As in the work of Daniele, Faisal, and Ferrante[2] on potential scattering, we choose these to be

$$\langle E(t)E(t')\rangle\equiv L_E(t-t')$$
$$\equiv\frac{\mathscr{E}_0^2}{2}\cos[\omega(t-t')]\exp(-\Delta\omega\,|\,t-t'\,|\,) \tag{15}$$

and

$$\langle A(t)A(t')\rangle\equiv L_A(t-t')$$
$$=\frac{c^2\mathscr{E}_0^2}{2(\omega^2+\Delta\omega^2)}\cos[\omega(t-t')-\phi]$$
$$\times\exp(-\Delta\omega\,|\,t-t'\,|\,)\,, \tag{16}$$

where $\mathscr{E}_0^2$ is the variance of the field amplitude, $\Delta\omega$ is the bandwidth, and ϕ is given by

$$\tan\phi = \frac{2\omega\Delta\omega}{\omega^2-\Delta\omega^2} .$$

The function $H(0,y)$, with the above correlation functions, has been evaluated by Daniele, Faisal, and Ferrante.[2] We give below somewhat simpler expressions for $H(x,y)$ as well as its conjugate $G(x,y)$, equal to twice the imaginary part of the integral on the right-hand side (RHS) of Eq. (13), which we shall be needing later on

$$H(x,y)+iG(x,y) = 2\exp(-z\cos\phi) \times \sum_{n=-\infty}^{\infty}\sum_{k=0}^{\infty}\frac{(z/2)^{\nu+2k}}{k!(\nu+k)!} \times \frac{\Gamma_{\nu k}(x)+iE_n(y)}{[\Gamma_{\nu k}(x)]^2+[E_n(y)]^2}e^{-i2n\phi} , \quad (17)$$

where

$$z=\frac{\lambda_0^2}{2}\frac{\omega^4}{(\omega^2+\Delta\omega^2)^2} , \quad (17a)$$

$$\lambda_0=\frac{(e\mathbf{q}\cdot\hat{\mathbf{E}})\mathscr{E}_0}{\omega^2} , \quad (17b)$$

$$\Gamma_{\nu k}(x)=(\nu+2k)\Delta\omega+z(\Delta\omega\cos\phi+\omega\sin\phi)+x , \quad (17c)$$

$$E_n(y)=n\omega-y \quad (17d)$$

and $\nu=|n|$. For later use, we note that H and G obey the crossing relations

$$H(x,y)=H(x,-y) \quad (18a)$$

and

$$G(x,y)=-G(x,-y) . \quad (18b)$$

Next, we consider terms in the first square brackets of Eq. (9). Using the identities

$$\hat{P}F_k^*(t_2)=\hat{P}^*F_k^*(t_1) \quad (19a)$$

and

$$\hat{P}F_k^*(t_1)=\hat{P}^*F_k^*(t_2) , \quad (19b)$$

we get

$$\langle\hat{P}[F_k(t_2)-F_k^*(t_2)-F_k(t_1)+F_k^*(t_1)]\rangle = 2\,\mathrm{Re}[\langle\hat{P}F_k(t_2)\rangle-\langle\hat{P}F_k(t_1)\rangle] . \quad (20)$$

Using the results of Appendix A and invoking the relations (18), we finally get

$$2\,\mathrm{Re}[\langle\hat{P}F_k(t_2)-\langle\hat{P}F_k(t_1)\rangle]=-Q/\omega_{k0} , \quad (21)$$

$$Q=\frac{2K\Omega}{c}\{(\cos\phi)[H(\Delta\omega,E_{if}+\omega)+H(\Delta\omega,E_{if}-\omega)-2H(0,E_{if})]+(\sin\phi)[G(\Delta\omega,E_{if}-\omega)-G(\Delta\omega,E_{if}+\omega)]\} , \quad (22)$$

and K is defined by Eq. (A14).

Evaluation of terms of the type $\langle\hat{P}F_k(t_2)F_n^*(t_1)\rangle$ is outlined in Appendix B. Expressions for the remaining ensemble averages in Eq. (9) may now be written down by inspection. Combining all these, we finally get

$$\langle\omega_{fi}\rangle=\mathscr{V}_{00}^2H(0,E_{if})+\mathscr{V}_{00}XQ+\mathscr{E}_0^2X^2[S(\Delta\omega,\omega)-U]+R , \quad (23)$$

where

$$X=\mathrm{Im}\sum_k\frac{\mathscr{V}_{0k}M_{k0}}{\omega_{k0}} , \quad (24)$$

$$S(x,y)=H(x,E_{if}+y)+H(x,E_{if}-y) , \quad (25)$$

$$T(x,y)=G(x,E_{if}+y)-G(x,E_{if}-y) , \quad (26)$$

$$U=\frac{z}{2}\{4(\cos^2\phi)[S(\Delta\omega,\omega)-H(0,E_{if})]-2(\sin2\phi)T(\Delta\omega,\omega)-(\cos\phi)S(2\Delta\omega,2\omega)+(\sin\phi)T(2\Delta\omega,2\omega)-2H(2\Delta\omega,E_{if})\} \quad (27)$$

and

$$R=\frac{3\Delta\omega\mathscr{E}_0^2c^3}{8}\sum_k\frac{|\mathscr{V}_{0k}|^2}{\omega_{k0}^s}S(\gamma_k,\omega_{k0}) . \quad (28)$$

In writing Eq. (28) we have made use of the fact that $\mathscr{V}_{0k}$ is pure imaginary while M_{0k} is real, and identified $2\gamma_k$ with the spontaneous transition probability per unit time τ_k, from $|k\rangle$ to $|0\rangle$, given by[6]

$$\tau_k=\frac{4e^2\omega_{k0}^3|M_{k0}|^2}{3\hbar c^3} .$$

The double differential cross sections may now be calculated from

$$\frac{d^2\sigma}{d\Omega\,dE_f}=\frac{k_f}{k_i}\left[\frac{1}{2\pi}\right]^3\langle W_{fi}\rangle . \quad (29)$$

IV. DISCUSSION AND NUMERICAL RESULTS

The first term on the RHS of Eq. (23) represents scattering by the static potential $\mathscr{V}_{00}$, while the remaining terms are due to the dressing of the atom by the field. To make contact with the single-mode results of Byron and Joachain,[7] we may study the limiting case of $\Delta\omega\to 0$, for which the term R vanishes. Further, we see from Eq. (17) that in this limit ϕ also vanishes, while[2]

$$H(0,y)\to 2\pi\sum_n I_n(\lambda_0^2/2)e^{-\lambda_0^2/2}\delta(y-n\omega)\ , \tag{30}$$

where I_n is a Bessel function of imaginary argument. Using these results, we get from Eqs. (23)—(29),

$$\left[\frac{d\sigma}{d\Omega}\right]_{\Delta\omega=0}=\int\left[\frac{d^2\sigma}{d\Omega\,dE_f}\right]_{\Delta\omega=0}dE_f$$
$$=\sum_n\frac{k_f(n)}{4\pi^2k_i}\{\mathscr{V}_{00}^2I_n+2\lambda_0\mathscr{E}_0\mathscr{V}_{00}X(I_n'-I_n)+\mathscr{E}_0^2X^2[2(1-\lambda_0^2)I_n'+\lambda_0^2(I_n+I_n'')]\}e^{-\lambda_0^2/2}\ , \tag{31}$$

where $k_f^2(n)=2(E_i-n\omega)$, and the argument of all Bessel functions is $\lambda_0^2/2$. (The primes on the Bessel functions denote differentiation.) Equation (31) is also the result of averaging the differential cross section for a coherent field $\mathbf{E}_0$, given (in the present notation) by[7]

$$\frac{d\sigma}{d\Omega}=\sum_n\frac{k_f(n)}{4\pi^2k_i}\,|\,\mathscr{V}_{00}J_n(\lambda)+2E_0XJ_n'(\lambda)\,|^2\ ,$$
$$\lambda=e\mathbf{q}\cdot\mathbf{E}_0/\omega^2\ , \tag{32}$$

over a Gaussian probability distribution

$$P(E_0)dE_0=\exp(-E_0^2/\mathscr{E}_0^2)d(E_0/\mathscr{E}_0)^2\ , \tag{33}$$

in agreement with what one would expect on the basis of similar results obtained in Refs. 2 and 3.

For $\Delta\omega\neq 0$ the double differential cross sections are no longer δ functions, but exhibit peaks at energies corresponding to the exchange of an integral number of photons of energy $\hbar\omega$. This is illustrated in Fig. 1, which shows some typical peaks at $E_{if}=0$, ± 1, and -2 in the scattering of 100-eV electrons at an angle $\theta=0.25°$, for $\mathscr{E}_0=0.02$, $\omega=0.0735$, and $\Delta\omega=10^{-2}\omega$. These parameters, as well as the polarization, which is taken to be parallel to the change in momentum $\mathbf{q}$, are the same as in Ref. 7. In these computations, summations over the atomic states $|\,k\,\rangle$ were effected by assuming an average energy of $\frac{4}{9}$ au., and then applying closure, since this procedure has been shown to yield quite accurate results in the case of a single mode field,[7] and a chaotic field is the resultant of an infinite number of uncorrelated modes. In

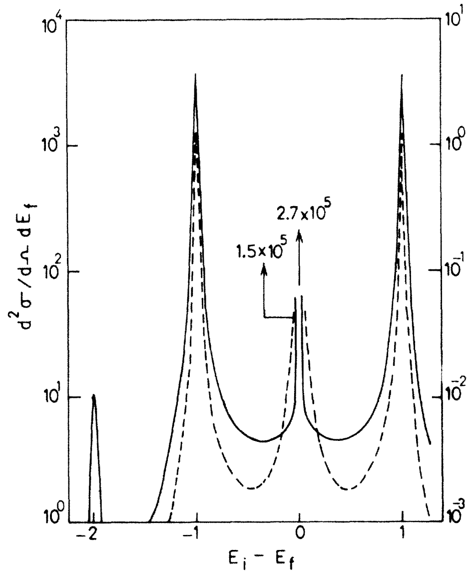

FIG. 1. Double differential cross sections (a.u.) for the scattering of electrons of energy $E_i=0.02$ a.u. from hydrogen at an angle of 0.25° in a laser field polarized parallel to the change in momentum. E_i-E_f is the change in the electron energy in units of the photon energy $\hbar\omega=0.0735$ a.u. The bandwidth $\Delta\omega=10^{-2}\omega$. The dashed curves refer to the undressed atom (scale to the right).

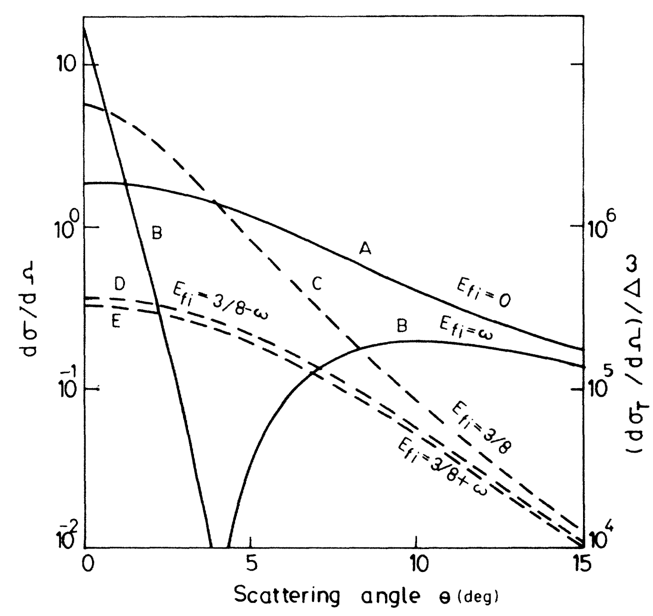

FIG. 2. Differential cross sections (a.u.) as a function of the scattering angle, for various values of the change in projectile energy E_{fi}. The scale to the left applies to the solid curves and that to the right applies to the dashed curves. The latter represent differential cross sections per unit bandwidth. The parameters are as in Fig. 1.

Fig. 1 the dashed lines refer to the "undressed" atom, i.e., only the first term in the RHS of Eq. (23) is taken into account. Now, the results of Byron and Joachain[7] shows that the differential cross sections for $\theta \lesssim 1°$ is higher for the $n=\pm 1$ process than for the $n=0$ process, because of the dressing of the atom, whereas the peak at $n=0$ is much higher than those at $n=\pm 1$. However, the areas under these peaks, which are well represented by Eq. (31), exhibit the opposite behavior, as shown in Fig. 2 (solid lines). Computations at smaller values of $\Delta\omega$ showed that the peak height is inversely proportional to $\Delta\omega$ (as one would expect from the limiting form of H), so that the ratio of the peak heights is unaltered.

As described in Ref. 5, for nonzero bandwidths, the term R [Eq. (28)] gives rise to a new series of peaks involving energy changes corresponding to the atomic transition energies, but with side bands at intervals of ω. The contribution to the differential cross section from these resonance peaks can be written as

$$\frac{d\sigma_r}{d\Omega}=\sum_k \left[\left[\frac{d\sigma_r}{d\Omega}\right]_k^+ + \left[\frac{d\sigma_r}{d\Omega}\right]_k^- \right], \quad (34)$$

where

$$\left[\frac{d\sigma_r}{d\Omega}\right]_k^{\pm} = \alpha_k^{\pm}\Delta\omega , \quad (35)$$

$$\alpha_k^{\pm}=\frac{3\mathscr{E}_0^2 c^3}{32\pi^2}\sum_n \frac{k_f^{\pm}(n)}{k_i}\frac{|\mathscr{V}_{0k}|^2}{\omega_{k0}^5}e^{-\lambda_0^2/2}I_n(\lambda_0^2/2) , \quad (36)$$

with $(k_f^{\pm})^2=2(E_i\pm\omega_{k0}-n\omega)$. These results are obtained under the approximation of small $\Delta\omega$ and γ_k, so that Eq. (30) may be used in Eq. (28). For hydrogen, since the dominant contribution comes from the $2p$ state, the contributions from the central peak (curve C) and the two nearest side bands (curves D and E) are plotted against the scattering angle in Fig. 2. As has already been discussed,[5] the present calculations are for a Lorentzian laser spectrum [implied by Eqs. (15) and (16)], and therefore, in a more realistic case where the spectrum has steeper wings,[8] Eqs. (34)–(36) would generally overestimate $d\sigma_r/d\Omega$.

APPENDIX A: EVALUATION OF $\langle \hat{P}F_k(t_2)\rangle$ AND $\langle \hat{P}F_k(t_1)\rangle$

We write $\langle \hat{P}F_k(t_2)\rangle$ as

$$\langle \hat{P}F_k(t_2)\rangle = -i\lim_{T\to\infty}\frac{1}{2T}\int_{-T}^{T}dt_1\int_{-T}^{T}dt_2 e^{iE_{if}(t_1-t_2)} \times\int_{-\infty}^{t_2}dt' e^{-i\omega_{k0}(t_2-t')}\left[\frac{\partial}{\partial x}\left\langle \exp\left[ixE(t')+i\Omega\int_{t_1}^{t_2}A(u)du\right]\right\rangle\right]\Bigg|_{x=0} . \quad (A1)$$

On taking the ensemble average of the exponential and performing the differentiation, the term within the square brackets becomes

$$-\Omega J(|t_1-t_2|)\int_{t_1}^{t_2}\langle E(t')A(u)\rangle du . \quad (A2)$$

Since

$$\langle E(t')A(u)\rangle = -\frac{1}{c}\frac{\partial}{\partial t'}\langle A(t')A(u)\rangle = \frac{1}{c}\frac{\partial}{\partial u}L_A(|t'-u|) , \quad (A3)$$

$$\int_{t_1}^{t_2}\langle E(t')A(u)\rangle du = \frac{1}{c}[L_A(|t'-t_2|)-L_A(|t'-t_1|)] . \quad (A4)$$

Using (A2) and (A4) in (A1), we get

$$\langle \hat{P}F_k(t_2)\rangle = \frac{i\Omega}{c}[\mathscr{I}_1-\mathscr{I}_2(E_{if})-\mathscr{I}_3(E_{if})] \quad (A5)$$

where

$$\mathscr{I}_1=\int_{-\infty}^{\infty}dt\, e^{iE_{if}t}J(|t|)\int_{-\infty}^{t_2}e^{-i\omega_{k0}(t_2-t')} \times L_A(|t'-t_2|)dt' , \quad (A6)$$

$$\mathscr{I}_2(x)=\int_{-\infty}^{\infty}dt\, e^{ixt}J(|t|)\int_{-\infty}^{t_1}e^{-i\omega_{k0}(t_2-t')} \times L_A(|t'-t_1|)dt' , \quad (A7)$$

and

$$\mathscr{I}_3(x)=\int_{-\infty}^{\infty}dt\, e^{ixt}J(|t|)\int_{t_1}^{t_2}e^{-i\omega_{k0}(t_2-t')} \times L_A(|t'-t_1|)dt' . \quad (A8)$$

After simple changes of variables, the above integrals reduce to

$$\mathscr{I}_1=\psi(-\omega_{k0},\infty)H(0,E_{if}) , \quad (A9)$$

$$\mathscr{I}_2(x)=\psi(-\omega_{k0},\infty)H(0,x+\omega_{k0}) , \quad (A10)$$

$$\mathscr{I}_3(x)=2i\,\mathrm{Im}\int_0^{\infty}dt\, e^{-i(x+\omega_{k0})t}J(t)\psi(\omega_{k0},t) \quad (A11)$$

with

$$\psi(\xi,t)=\int_0^t e^{i\xi u}L_A(u)du, \quad t>0 . \quad (A12)$$

On carrying out the integration in Eq. (A12), and neglecting ω and $\Delta\omega$ in comparison with ω_{k0} in the denominators, we get

$$\psi(\xi,t)=K\left[\frac{e^{(i\xi-\Delta\omega)t}\cos(\omega t-\phi)-\cos\phi}{i\xi}\right], \tag{A13}$$

where

$$K=\frac{c^2\mathscr{E}_0^2}{2(\omega^2+\Delta\omega^2)}. \tag{A14}$$

Using Eq. (A13) in Eq. (A11), $\mathscr{I}_3$ can be expressed in terms of H and G. This completes the evaluation of $\langle \hat{P}F_k(t_2)\rangle$. $\langle \hat{P}F_k(t_1)\rangle$ may be calculated in an exactly similar manner, with the result that

$$\langle \hat{P}F_k(t_1)\rangle=\frac{-i\Omega}{c}[\mathscr{I}_1-\mathscr{I}_2(-E_{if})-\mathscr{I}_3(-E_{if})]. \tag{A15}$$

APPENDIX B: EVALUATION OF $\langle \hat{P}F_k(t_2)F_n^*(t_1)\rangle$

As before, we first write

$$\langle \hat{P}F_k(t_2)F_n^*(t_1)\rangle = -\lim_{T\to\infty}\frac{1}{2T}\int_{-T}^{T}dt_1\int_{-T}^{T}dt_2 e^{iE_{if}(t_1-t_2)}\int_{-\infty}^{t_2}dt' e^{-i\omega_{k0}(t_2-t')} \times\int_{-\infty}^{t_1}dt'' e^{i\omega_{n0}(t_1-t'')} \times\left[\frac{\partial}{\partial x_1}\frac{\partial}{\partial x_2}\left\langle\exp\left[ix_1E(t')+ix_2E(t'')+i\Omega\int_{t_1}^{t_2}A(u)du\right]\right\rangle\right]\Bigg|_{x_1=x_2=0}. \tag{B1}$$

The terms in the square brackets reduce to

$$J(|t_1-t_2|)\{L_E(|t'-t''|)-\frac{\Omega^2}{c}[L_A(|t'-t_2|)-L_A(|t'-t_1|)]\times[L_A(|t''-t_2|)-L_A(|t''-t_1|)]\}. \tag{B2}$$

Substituting in Eq. (B1), the contribution from the first term in Eq. (B2) may be calculated to be

$$S_1=\frac{\mathscr{E}_0^2}{2}\int_0^\infty dt\, J(t)[e^{E_{if}t}\mathscr{I}(\omega_{n0},\omega_{k0},\omega,t)+e^{-iE_{if}t}\mathscr{I}(-\omega_{k0},-\omega_{n0},\omega,t)], \tag{B3}$$

where

$$\mathscr{I}(\omega_{n0},\omega_{k0},\omega,t)=[\mathscr{I}_1(\omega)+\mathscr{I}_1(-\omega)]/2, \tag{B4}$$

$$\mathscr{I}_1(\omega)=\frac{\exp[-(\Delta\omega+i\omega)t]}{(\omega_{n0}+\omega-i\Delta\omega)(\omega_{k0}+\omega-i\Delta\omega)}+\Delta, \tag{B5}$$

$$\Delta=\begin{cases}\dfrac{2i\Delta\omega e^{ixt}}{\omega_{nk}[(\omega_{n0}+\omega)^2+\Delta\omega^2]}, & n\neq k \quad \text{(B6)}\\[2ex] \dfrac{\Delta\omega\exp[(i\omega_{k0}-\gamma_k)t]}{\gamma_k[\Delta\omega^2+(\omega_{k0}+\omega+i\gamma_k)^2]}, & n=k. \quad \text{(B7)}\end{cases}$$

The expression (B7) is obtained taking due note of the presence of γ_k in the exact expression for ψ_0. It is clear that Δ can be significant only for $n=k$, and may be neglected when $n\neq k$. From Eqs. (B3)–(B7) we therefore get (after the usual simplification of the denominators)

$$S_1=\frac{\mathscr{E}_0^2}{4\omega_{k0}}\left[\frac{H(\Delta\omega,E_{if}+\omega)+H(\Delta\omega,E_{if}-\omega)}{\omega_{n0}}+\frac{\Delta\omega\delta_{kn}}{\omega_{k0}\gamma_k}H(\gamma_k,E_{if}+\omega_{k0})\right]. \tag{B8}$$

The contribution to Eq. (B1) from the remaining part of Eq. (B2) may also be evaluated in a similar fashion, and is given by

$$T_1=\frac{k^2}{\omega_{k0}\omega_{n0}}\left[(\cos^2\phi)H(0,E_{if})-4(\cos\phi)\mathrm{Re}\int_0^\infty e^{(iE_{if}-\Delta\omega)t}\cos(\omega t-\phi)J(t)dt+2\,\mathrm{Re}\int_0^\infty e^{(iE_{if}-2\Delta\omega)t}\cos^2(\omega t-\phi)J(t)dt\right]. \tag{B9}$$

Since Eq. (B9) is readily expressed in terms of H and G, the evaluation of $\langle \hat{P}F_k(t_2)F_n^*(t_1)\rangle=S_1+T_1$ is now complete.

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