PHYSICAL REVIEW A VOLUME 17, NUMBER 2 FEBRUARY 1978

Calculation of resonant cross sections for multiphoton ionization using very-narrow-bandwidth sources

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(Received 23 May 1977)

We consider the infinite-order set of coupled, integral equations for the states of an atom in a very-narrow-bandwidth classical radiation field. In the vicinity of a one-photon resonance for two-photon ionization, this set is reduced to a pair of coupled equations describing the response of the atom to the radiation field. This set is derived by dropping terms of the original set whose lowest-order contributions to the radiation-induced shift and width of the resonance are quadratic in the intensity. In the absence of simultaneous absorption and emission, impossible except for two-photon processes from excited states, this pair can be decoupled. The remaining uncoupled integral equation describes the ac Stark effect based on contributions to the shift and width linear in the intensity. These contributions derive from radiative corrections to the intermediate atomic state and depend on the virtual processes of ionization from and recombination into, as well as emission from and reabsorption into, this state. A method for obtaining the exact numerical solution is discussed. An approximate solution is obtained by use of the single-eigenfunction approximation to the Green's function belonging to the response function. This Green's function has a pole at the static (unshifted) position of the resonance; thus, this approximation is quite accurate very near the pole. The order of nonlinearity is calculated as a function of the static detuning through the resonance and is shown to undergo rapid fluctuations about the nonresonance value of 2. The error inherent in the single-eigenfunction approximation to the Green's function is estimated and found to range from about 1% to 27% corresponding to static detunings ranging from about 1 to 25 Å, respectively.

I. INTRODUCTION

The recent experiments of Morellec, Normand, and Petite[1] cogently document the need to develop methods of calculating cross sections for multiphoton ionization through a region of one or more resonances. These experiments are performed using a laser bandwidth of about 50 MHz, of the same order as the free-atom natural linewidth; thus, the resonance absorption can be studied under single-mode conditions, eliminating the variation of the observed ion yields owing to photon statistics. This, in turn, enables the study of the rapid fluctuation of the order of nonlinearity [see Eq. (1) and Fig. 7 of Ref. 1] through the narrow region of frequency spanning the resonance.

To aid in the interpretation of their experiments, Morellec, Normand, and Petite[1] make use of a method of calculating the cross section for the three-photon ionization of H(2s) developed by Gontier and Trahin.[2(a)] These authors include the calculation of the radiation-induced shift and width of the intermediate level responsible for the resonance through use of a Schwinger-Dyson equation,[3] which allows a renormalized Green's function to be calculated. (For the use of ordinary Green's functions for "undressed states" see, for example, work of Rapoport *et al.*,[4] Klarsfeld,[5] and of the present author.[6]) In a more recent paper, Gontier *et al.*[7] criticize the work of Gontier and Trahin (which, according to these authors, derives from the quantum-electrodynamics method of Low[8]), on the grounds that certain contributions to the amplitude for the process (diagrams) are multiply counted or undercounted for "real photons." These defects are unfortunate because the form of the theory, which closely resembles that of ordinary perturbation theory, makes possible the interpretation of the radiation-induced shifting and broadening of the levels as a function of the laser flux and frequency. The numerical evaluation of the terms of the theory, however, is difficult, requiring that multiple summations be performed over complete sets of bound and continuum states of the target. These sets were truncated[2(a)] by omission of the continua, thus introducing a source of error. Gontier, Rahman, and Trahin[7] then introduce a new technique, valid for arbitrarily strong radiation fields, which is applied to a two-level atom. They remove the difficulty of the incorrect counting of diagrams. However, the mathematical structure of this theory is quite complicated, making the physical interpretation of the results perhaps not altogether transparent. For example, the continued fractions form of the theory does not explicitly exhibit expressions for the level shift.

It is the purpose of the present paper to introduce a method with the advantages of the renormalization Green's-function method,[2(a)] including a diagrammatic analysis displaying the leading radiative corrections, but without the theoretical flaw of the incorrect counting of diagrams or the great hardship of evaluating the terms by means of the direct-summation-over-states method. On the other hand, we must limit the validity of the present theory to field intensities (as measured by the ratio I/I_0, where $I_0 = 14.038 \times 10^{16}$ W cm^{-2} is twice the intensity corresponding to a root-mean-square field strength of 1 a.u. or 5.142×10^9 V cm^{-1}, such that $I/I_0 \to 1$ indicates the approach to the Coulombic limit) which are still weak enough for ordinary perturbation theory to be valid in the nonresonance region.

We accomplish this separation into a resonance and nonresonance region of validity by use of several steps. First, we write down the exact equations for the states of an atom coupled by the classical radiation field. We have cast the equations in integral form by use of the Coulomb Green's function. This enables us to pick out the Green's functions in a particular region of frequency which have poles. These are just the poles of the Γ function on the negative energy axis for the hydrogen atom[4,5] and for complex atoms as well.[6] In the complex-atoms case, additional integral terms exist, causing a shift relative to the hydrogenic pole through the interaction of the "optical" electron with the atomic field. Next, we truncate the infinite-order coupled set by keeping only the terms containing the "response" states constructed by use of the singular Green's functions. We then solve this reduced set of integral equations nonperturbatively and use the solutions ("response" states) to calculate the amplitude for the process. Obviously I/I_0 must be small enough so that (i) the ordinary validity criterion for perturbation theory holds in the nonresonance region, enabling the truncation of the infinite-order coupled set and (ii) the radiation-induced shifting and broadening of the levels is still small enough that the vicinities of the resonance lines are locatable by inspection of the unperturbed spectrum of atomic states. In other words, these criteria imply intensities at which a "spectroscopy," in the usual sense of resonance peaks superimposed on a smooth background, is still possible.

II. THEORY

A. Derivation of the equations describing resonant absorption induced by very-narrow-bandwidth radiation

We consider the case for the two-photon ionization of H(1s), in which a series of $1s \to np$ ($n \geq 2$) resonances occur.[9] The structure of the equations for the series $2s \to ns + nd$, considered by Gontier and Trahin[2(a)] would be more complicated owing to the coupling of the equations for the s and d response functions through their mutual coupling with the p response function, but no new difficulties are introduced in principle. The set of radial coupled equations describing H(1s) in a radiation field of arbitrary intensity is given by

$$\chi_l(r,\omega)=\psi_{1s}(r)\delta_{l0}\delta(\omega-\omega_{1s})+\sum_{l'}\int_{-\infty}^{\infty}d\omega'\int_0^{\infty}dr'\,r'^2 g_l(r,r';\omega)\{\delta[\omega-(\omega'+\omega_p)]+\delta[\omega-(\omega'-\omega_p)]\}$$
$$\times U_{ll'}(r')\chi_{l'}(r',\omega'), \tag{1a}$$

$$U_{ll'}=2\mathrm{i}^{l'-l+1}\left(\frac{I}{I_0\omega_p^2}\right)^{1/2}[2l+1)(2l'+1)]^{1/2}$$
$$\times\left[\begin{pmatrix} l & 1 & l' \\ 0 & 0 & 0\end{pmatrix}^2\frac{\partial}{\partial r}-\sqrt{2}\,(1-\delta_{l'0})\left(\frac{(l'+1)!}{(l'-1)!}\right)^{1/2}\begin{pmatrix} l & 1 & l' \\ 0 & 0 & 0\end{pmatrix}\begin{pmatrix} l & 1 & l' \\ 0 & 1 & -1\end{pmatrix}r^{-1}\right]. \tag{1b}$$

This set is the Fourier transform of the time-dependent solution $\Psi(\vec{r},t)$ of the Schrödinger equation for the atom plus the classical radiation field. The pair of Fourier transforms are defined by

$$\Psi(\vec{r},t)=\int_{-\infty}^{\infty}d\omega\,e^{-i\omega t}\Phi(\vec{r},\omega), \tag{2}$$

where the four-dimensional function $\Phi(\vec{r},\omega)$ is defined by the integral equation

$$\Phi(\vec{r},\omega)=\Psi_{1s}(\vec{r})\delta(\omega-\omega_{1s})$$
$$+\int_{-\infty}^{\infty}d\omega'\int d\vec{r}'\,G_c(\vec{r},\vec{r}';\omega)$$
$$\times U(\vec{r}',\omega,\omega')\Phi(r',\omega'), \tag{3a}$$

$$U(\vec{r},\omega,\omega')=2i(I/I_0\omega_p^2)^{1/2}\{\delta[\omega-(\omega'+\omega_p)]$$
$$+\delta[\omega-(\omega'-\omega_p)]\}$$
$$\times\left(\cos\theta\frac{\partial}{\partial r}-\frac{\sin\theta}{r}\frac{\partial}{\partial\theta}\right), \tag{3b}$$

where we have used the definition that $2\pi F\alpha\omega_p$ is I/I_0 in atomic units, where F is the flux and α the fine-structure constant. The kernel of Eq. (3a) is defined by the Coulomb Green's function[4-6,10] G_c. The set (1a) is derived by a partial wave resolution based on the expansions

$$G_c(\vec{r},\vec{r}\,';\omega)=\sum_l g_l(r,r';\omega)Y_{lm}(\theta_r,\phi_{r'})\,,$$
$$\times Y^*_{lm}(\theta_{r'},\phi_{r'})\,, \qquad (4a)$$

$$\Phi(\vec{r},\omega)=\sum_l i^l\chi_l(r,\omega)Y_{l0}(\theta_r,\phi_r)\,. \qquad (4b)$$

[See the Appendix for complete definitions of $g_l(r,r';\omega)$.] The light is assumed polarized along the polar axis in the laboratory frame and to be incident on spherically symmetric targets; this condition enables use of the same axis for the spatial quantization of the target states and the suppression of the aximuthal quantum number in Eq. (4b). All frequencies, ω, ω', ω_{1s}, and ω_p, are in dimensionless multiples of atomic units.

The intensity in Eqs. (1b) or (3b) can be treated as a parameter rather than as a function of frequency ω as a result of the physical assumption that the laser bandwidth is very narrow. Thus we are treating the intensity (and therefore the amplitude of the classical field) as a constant over this interval. This is in contrast to the assumption by others[11(a)–11(b)] of a pulse shape function depending on time which, for example,[11(b)] can exhibit a maximum at some time t_0 proportional to our $(I/I_0)^{1/2}$ and then decay on either side of this point. Under these conditions, appropriate for strong pulsed-laser fields, the line shape through the resonance will depend critically on the pulse width and its assumed shape. These experimental conditions are in sharp contrast to those described by Morellec, Normand, and Petite,[1] in which the lines for resonance absorption are very narrow with shapes governed by the ac Stark shifts. We note here that it is this assumption of a constant field amplitude during the duration of the absorption which makes the simple δ-function behavior of the Fourier transform $U(\vec{r},\omega,\omega')$ possible. In turn, as we shall see in Sec. III D, the δ-function behavior, appropriate for absorption in very-narrow-bandwidth fields, leads to a method for obtaining the nonperturbative solution to a truncated version of the infinite-order coupled set given by Eq. (1a).

B. Recovering the results of perturbation theory from the iterative solution of Eq. (1a)

The form of the coupled equations (1a), with δ function interactions in the frequencies ω and ω', is transparent with respect to the development of a perturbation theory. Recognizing that I/I_0 in Eq. (1b) is small except for fields whose strength approaches the Coulombic limit, an iterative solution of Eq. (1a) can be developed. On the first iteration we obtain

$$\chi_l(r,\omega)=\psi_{1s}(r)\delta_{l0}\delta(\omega-\omega_{1s})+\int_0^\infty dr'\,r'^2 g_l(r,r';\omega)\{\delta[\omega-(\omega_{1s}+\omega_p)]+\delta[\omega-(\omega_{1s}-\omega_p)]\}U_{l0}(r')\,\psi_{1s}(r')\,. \qquad (5)$$

We note that the ω' dependence of the integrands is completely specified on the first iteration through the product of the terms in curly brackets in Eq. (1a) and $\delta(\omega-\omega_{1s})$, appropriate for the 1*s* eigenfunction. Hence the integration over ω' can be performed immediately, causing ω' in the interaction [Eqs. (1a) or (3b)] to be replaced by ω_{1s}, so that Eq. (5) emerges. The existence properties of the 3*j* symbols in Eq. (1b) restrict l to $l=1$. Use of Eqs. (2) and (4) then gives the well-known response function[4,5] for the atom plus one virtual photon

$$\Psi(\vec{r},t)-\Psi_{1s}(\vec{r})\,e^{-i\omega_{1s}t}=2i\left(\frac{I}{I_0\omega_p^2}\right)^{1/2}\left[\left(\int_0^\infty dr'r'^2g_1(r,r';\omega_{1s}+\omega_p)\frac{\partial}{\partial r'}\psi_{1s}(r')\right)\exp-i(\omega_{1s}+\omega_p)t\right.$$
$$\left.+\left(\int_0^\infty dr'r'^2g_1(r,r';\omega_{1s}-\omega_p)\frac{\partial}{\partial r'}\psi_{1s}(r')\right)\exp-i(\omega_{1s}-\omega_p)t\right]\frac{\cos\theta_r}{(4\pi)^{1/2}}\,. \qquad (6)$$

Using the usual procedures,[4–6,9] we can derive the cross sections for two-photon absorption [to which only the first component of Eq. (6) contributes], for two-photon emission[12] (to which only the second component contributes), and for Raman or Rayleigh scattering[13,14] (to which both contribute coherently). These results are, of course, well known. What may not be so familiar (owing to the lack of theoretical development prior to the laser as a device for studying resonant multiphoton processes) is the set of coupled integral equations and, more important, how its noniterative (nonperturbative) solution would affect the cross sections for the above processes. We note that the familiar concepts of absorption or emission of a definite number of photons [being equal to the order of the iteration (perturbation)], the independence of the absorption and emission processes, and the relationship of the number of photons, K_0, absorbed or emitted to the parity of the atomic eigenstates, $K_0 \geq l$, all emerge from the perturbative solution. We note that Eq. (1a) is integral with respect to ω' as well as to r'; thus, the simplicity of the above conceptual framework disappears when we contemplate the noniterative solution of the equation.

C. Truncation of the infinite-order set

It is clear that the iterative solution just outlined will not be valid when one or more poles occur in the radial Green's functions.[6,10] These poles occur as ω sweeps through the excitation spectrum of the atom and are defined by zero and the set of negative integers of the Γ function (see the Appendix) $\Gamma(l+1-\nu)$ for $\nu=1, 2, 3, \ldots$, where ν is an effective principal quantum number related to the energy by $\nu = iZ/(2\omega)^{1/2}$ ($Z=1$ for hydrogen). Now the tuning to a particular intermediate resonance state restricts the number of Green's functions which can have poles at the same frequency ω_p to a very small number: one for a $1s \rightarrow np$ resonance, two for a $1s \rightarrow ns+nd$ resonance, two for a $1s \rightarrow np+nf$ resonance, three for a $1s \rightarrow ns+nd+ng$ resonance, and so on. Provided I/I_0 is reasonably small, the products of *nonsingular* Green's functions and powers of I/I_0 of higher order than the power multiplying the amplitude, namely $(I/I_0)^{(K_0-1)/2}$, can be dropped.

Let us develop a solution in the vicinity of the first resonance of the p series[9] for the two-photon ionization of H(1s), that is, in the vicinity of $\nu=2$, $l=1$. In this case it is clear that the large terms in Eq. (1a) are those corresponding to $l'=0$ and $l'=2$ in the summation on the right-hand side of Eq. (1a). Defining $\chi_0(r,\omega)=\psi_{1s}(r)\delta(\omega-\omega_{1s}) + \chi_0^{(S)}(r,\omega)$, we derive the reduced set for the coupled s, p, and d states,

$$\chi_0^{(S)}(r,\omega)=\int_{-\infty}^{\infty}d\omega'\int_0^{\infty}dr' r'^2 g_0(r,r';\omega)\{\delta[\omega-(\omega'+\omega_p)]+\delta[\omega-(\omega'-\omega_p)]\}U_{01}(r')\chi_1(r',\omega')\,, \tag{7a}$$

$$\begin{aligned}\chi_1(r,\omega)=&\int_0^{\infty}dr' r'^2 g_1(r,r';\omega)\{\delta[\omega-(\omega_{1s}+\omega_p)]+\delta[\omega-(\omega_{1s}-\omega_p)]\}U_{10}\psi_{1s}(r')\\&+\int_{-\infty}^{\infty}d\omega'\int_0^{\infty}dr' r'^2 g_1(r,r';\omega)\{\delta[\omega-(\omega'+\omega_p)]+\delta[\omega-(\omega'-\omega_p)]\}\\&\qquad\times[U_{10}(r')\chi_0^{(S)}(r',\omega')+U_{12}(r')\chi_2(r',\omega')]\,,\end{aligned} \tag{7b}$$

$$\chi_2(r,\omega)=\int_{-\infty}^{\infty}d\omega'\int_0^{\infty}dr' r'^2 g_2(r,r';\omega)\{\delta[\omega-(\omega'+\omega_p)]+\delta[\omega-(\omega'-\omega_p)]\}U_{21}(r')\chi_1(r',a')\,. \tag{7c}$$

It is clear that the first term on the right-hand side of Eq. (7b) is of order $(I/I_0)^{1/2}$, while the second term is at least of order $(I/I_0)^{3/2}$. The term on the right-hand side of Eqs. (7a) or (7c) is at least of order I/I_0. The rule $K_0 \geqslant l'$ can be used to obtain the lowest order of $(I/I_0)^{K_0/2}$ in a product of the form $U_{ll'}\chi_{l'}$, where $U_{ll'}$ always contributes $(I/I_0)^{1/2}$. Thus, far away from a pole of g_1, we can immediately recover the correct nonresonance result of first-order perturbation theory by keeping only terms of order $(I/I_0)^{1/2}$. Near the pole, we seek a nonperturbative solution to this set (Eqs. 7).

First, however, we wish to make several points. The first term on the right-hand side of Eq. (7b), of order $(I/I_0)^{1/2}$, is the source of the coupling to the radiation field. Without it all solutions vanish except $\chi_0(r,\omega)=\psi_{1s}(r)\delta(\omega-\omega_{1s})$. Second, the reduced set is based on the elimination of any response state $\chi_l(r,\omega)$ not coupled by the dipole interaction to the large response state $\chi_1(r,\omega)$. Since the source of Eq. (7b) is infinite at a pole of g_1, $\chi_1(r,\omega)$ can be finite only through the dipolar coupling to $\chi_0^{(S)}(r,\omega)$ and $\chi_2(r,\omega)$. Thus this set is irreducible. Further, analysis shows that the retention of response states for l greater than 2 [states not coupled directly to $\chi_1(r,\omega)$] contributes radiative corrections of lowest order $(I/I_0)^2$, while the radiative corrections deriving from Eqs. (7) are of lowest order I/I_0. Finally, we note that truncation of the infinite-order set [Eq.(1a)] is *not* equivalent to treatment of the atom as a finite-level system. This point becomes obvious if the radial Green's functions of Eqs. (7) are represented in the complete sets of atomic states having s (for g_0), p (for g_1), and d (for g_2) symmetries. Such sets, of course, have an infinite number of discrete bound states and a continuum of unbound states.

D. Use of the δ-function form of the interaction to find a nonperturbative solution to the reduced set

The mathematical simplicity of the δ-function form of the atomic interaction with the narrow-bandwidth field makes it possible to find a nonperturbative solution to Eqs. (7). We note that the source, in addition to having the poles of g_1, has δ-function singularities at $\omega=\omega_{1s}+\omega_p$ and $\omega=\omega_{1s}-\omega_p$. We develop a solution of the form of a sum of products of rapidly and slowly varying parts,

$$\chi_1(r,\omega)=\delta[\omega-(\omega_{1s}+\omega_p)]\chi_1^{(a1)}(r,\omega)$$
$$+\delta[\omega-(\omega_{1s}-\omega_p)]\chi_1^{(e1)}(r,\omega)\,. \qquad (8)$$

It is important to note that it is the perturbative solution, found by retaining only the source, which is rapidly varying at $\omega=\omega_{1s}+\omega_p$ for absorption *or* at $\omega=\omega_{1s}-\omega_p$ for emission. The nonperturbative solutions $\chi_1^{(a1)}$ and $\chi_1^{(e1)}$ should be reasonably smooth relative to the rapid variation of one of the δ functions at either of these points. We substitute Eq. (8) into the right-hand sides of Eqs. (7a) and Eq. (7c) and perform the indicated integrations over ω', which are made possible by the condition that the integrands consist of a slowly varying part times the δ functions in Eq. (8). We note that a product of two δ functions occur in each integrand, but that one will vary rapidly while the other varies slowly at the point $\omega=\omega_{1s}+\omega_p$ or $\omega=\omega_{1s}-\omega_p$ of Eq. (8).

Thus, the validity of this evaluation depends on the smoothness of the unknown functions, $\chi_1^{(a1)}$ and $\chi_1^{(e1)}$ at these points. This is an important result because it enables us to solve the integral equations with respect to the variable, ω. From the result of this substitution, it is clear that $\chi_0^{(S)}$ and χ_2 can also be cast in a form analogous to Eq. (8),

$$\chi_j^{(S)}(r,\omega)=\delta[\omega-(\omega_{1s}+2\omega_p)]\chi_j^{(a2)}(r,\omega)$$
$$+\delta(\omega-\omega_{1s})\chi_j^{(0)}(r,\omega)$$
$$+\delta[\omega-(\omega_{1s}-2\omega_p)]\chi_j^{(e2)}(r,\omega)\,, \qquad (9)$$

for $j=0$ or 2, and $\chi_2^{(S)}=\chi_2$. We can derive an auxiliary set of coupled equations by equating the coefficients of the δ functions $\delta[\omega-(\omega_{1s}\pm n\omega_p)]$, on both sides of Eqs. (7), where n is 0, 1, or 2. We finally arrive at a pair of coupled equations for the absorption and emission response functions $\chi_1^{(a1)}$ and $\chi_1^{(e1)}$ of Eq. (8):

$$\chi_1^{(a1)}(r,\omega_{1s}+\omega_p)=\int_0^\infty dr'r'^2g_1(r,r';\omega_{1s}+\omega_p)U_{10}(r')\psi_{1s}(r')$$
$$+\sum_{j=0,2}^{2}\sum_{n=0,2}^{2}\int_0^\infty dr'r'^2g_1(r,r';\omega_{1s}+\omega_p)U_{1j1,n}^{(\mathrm{AEL})}(r')\chi_1^{(a1)}(r',\omega_{1s}+\omega_p)$$
$$+\sum_{j=0,2}^{2}\int_0^\infty dr'r'^2g_1(r,r';\omega_{1s}+\omega_p)U_{1j1}^{(\mathrm{C})}(r')\,\chi_1^{(e1)}(r',\omega_{1s}-\omega_p)\,, \qquad (10\mathrm{a})$$

$$\chi_1^{(e1)}(r,\omega_{1s}-\omega_p)=\int_0^\infty dr'r'^2g_1(r,r';\omega_{1s}-\omega_p)U_{10}(r')\psi_{1s}(r')$$
$$+\sum_{j=0,2}^{2}\sum_{n=0,2}^{2}\int_0^\infty dr'r'^2g_1(r,r';\omega_{1s}-\omega_p)U_{1j1,n}^{(\mathrm{EAL})}(r')\chi_1^{(e1)}(r',\omega_{1s}-\omega_p)$$
$$+\int_0^\infty dr'r'^2g_1(r,r';\omega_{1s}-\omega_p)U_{1j1}^{(\mathrm{C})}(r')\chi_1^{(a1)}(r',\omega_{1s}+\omega_p)\,, \qquad (10\mathrm{b})$$

$$U_{1j1,n}^{(\mathrm{AEL})}(r)=U_{1j}(r)\int_0^\infty dr'r'^2g_j(r,r');\omega_{1s}+n\omega_p)U_{j1}(r')\,, \qquad (10\mathrm{c})$$

$$U_{1j1,n}^{(\mathrm{EAL})}(r)=U_{1j}(r)\int_0^\infty dr'r'^2g_j(r,r';\omega_{1s}-n\omega_p)U_{j1}(r')\,, \qquad (10\mathrm{d})$$

$$U_{1j1}^{(\mathrm{C})}(r)=U_{1j}(r)\int_0^\infty dr'r'^2g_j(r,r';\omega_{1s})U_{j1}(r')\,. \qquad (10\mathrm{e})$$

We note that the response states $\chi_1^{(a1)}(r,\omega)$ and $\chi_1^{(e1)}(r,\omega)$ are defined at the points $\omega=\omega_{1s}+\omega_p$ and $\omega=\omega_{1s}-\omega_p$, respectively, where their δ function coefficients of Eq. (8) peak. This is the form of solution expected for very-narrow-bandwidth absorption. In fact, the equations rigorously describe absorption of infinitely-narrow-bandwidth radiation, based on the original assumption that the amplitude of the field is constant over this interval. The superscripts (AEL), (EAL), and (C) refer, respectively, to terms corresponding to absorption-emission loops, emission-absorption loops, and coupling loops. The reduced Coulomb Green's function $g_j^{(1)}(r,r')$, is required for propagator lines in which the frequency $\omega=\omega_{1s}$. This occurs, in an absorption loop, by the loss of all photons previously absorbed and, in an emission loop, by the reabsorption of all photons previously emitted. In Sec. III E we will present a diagrammatic analysis in which the physical interpretation of the radiative corrections of Eqs. (10) becomes clear.

We note the presence of coupling between the response functions for absorption and emission, $\chi_1^{(a1)}$ and $\chi_1^{(e1)}$, respectively. This coupling occurs through the interference of each component of Eq. (8) with its opposite frequency counterpart in the δ-function part of the potential in Eqs. (7a) or (7c) on substituting Eq. (8) into the right-hand sides of these equations, e.g., through the occurrence of products of terms of the form

$$\int_{-\infty}^{\infty} d\omega' \,\delta[\omega-(\omega'+\omega_p)]\delta[\omega'-(\omega_{1s}-\omega_p)]\,\chi_1^{(e1)}(r,\omega') \tag{11}$$

and

$$\int_{-\infty}^{\infty} d\omega' \,\delta[\omega-(\omega'-\omega_p)]\delta[\omega'-(\omega_{1s}+\omega_p)]\chi_1^{(a1)}(r,\omega') \,. \tag{12}$$

Through these terms $\chi_j^{(0)}(r,\omega)$ is the only component of Eq. (9) which is coupled to both $\chi_1^{(a1)}(r,\omega)$ and $\chi_1^{(e1)}(r,\omega)$, and on substituting Eq. (9) into the right-hand side of Eq. (7b) and equating the coefficients of the δ functions, $\delta[\omega-(\omega_{1s}+\omega_p)]$ and $\delta[\omega-(\omega_{1s}-\omega_p)]$, respectively, the pair of coupled equations (10a) and (10b) results. We note, also, that this back substitution [Eq. (9) into Eq. (7b)] implies the existence of the response functions $\chi_1^{(a3)}$ and $\chi_1^{(e3)}$, corresponding to the absorption and emission, respectively, of three virtual photons; however, equations for these functions do not appear based on the original ansatz that response functions whose Green's functions $[g_1(r,r';\omega_{1s}+3\omega_p)$ and $g_1(r,r';\omega_{1s}-3\omega_p)$, respectively, for the above responses] are nonsingular for the ω_p of interest are dropped as producing radiative corrections whose lowest order is $(I/I_0)^2$

The form of the solution given by Eq. (8) makes it possible to derive the usual formula for the cross section for the process.[9] The differential cross section for two-photon ionization is

$$\frac{d\sigma}{d\Omega}=\frac{\alpha a_0^2}{2\pi}\frac{k}{\omega_p}|\langle\psi_f^{(-)}|\hat{p}\cdot\vec{\nabla}|\chi_1^{(a1)}\rangle|^2\,, \tag{13}$$

where α is the fine-structure constant, a_0 the Bohr radius, k the momentum of the ejected electron (in atomic units), $\psi_f^{(-)}$ the wave function for ionized hydrogen, $\hat{p}$ the unit vector of polarization of the light, and the other quantities have been defined previously. We note that this cross section will show a complicated dependence of I/I_0 through the dependence of $\chi_1^{(a1)}$ on I/I_0; this complicated dependence (see Fig. 5) occurs in the vicinity of the resonance[1,2(a)]; otherwise the cross section will be proportional to I/I_0 [or generally to $(I/I_0)^{K_0-1}$].

E. Diagrammatic representation and physical interpretation of the terms of Eqs. (10)

To aid in the physical interpretation of Eqs. (10), we make use of diagrammatic analysis. These equations can be represented as in Fig. 1. The dashed lines connected to the crosses represent interactions with the radiation field. The arrows stand for g_j (for $j=0$, 1, or 2) or for $\chi_1^{(a1)}$ or $\chi_1^{(e1)}$, consistent with parity conservation at the interaction lines. Since the arrows stand either for g_j, in which case they are conventional propagator lines, or for one of the response functions, the number of interaction lines does *not* give the order of the term, as in diagrammatic perturbation theory, except in the first diagram in each row which represents the source. Otherwise, the number of interaction lines, which is the same as the number of real or virtual photons K_0 gives the minimum order $(I/I_0)^{K_0/2}$ for the process. That is, if the equations were iterated once, then the number of interaction lines would be the same as the order for the process.

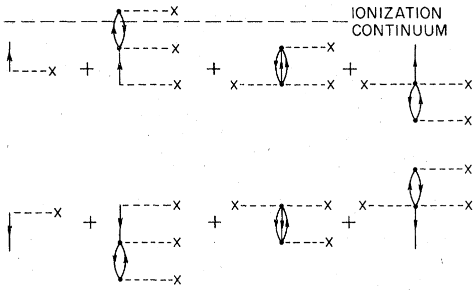

FIG. 1. Diagrams representing the coupled set, Eqs. (10). Definitions are given in the text.

Upward and downward arrows stand for absorbed and emitted photons, respectively. In row 1 the first diagram represents the source for absorption [first term in Eq. (10a)], the second and third diagrams represent radiative corrections to the intermediate $2p$ state, consisting of virtual ionization and recombination [second diagram, corresponding to the $n=2$ component in the second term of Eq. (10a)] and virtual emission and reabsorption [third diagram, corresponding to the $n=0$ component in the second term of Eq. (10a)]. The fourth diagram represents radiative corrections to the initial $1s$ state [the coupling term in Eq. (10a)]. It is the latter correction which is omitted in the analysis of Gontier and Trahin.[2(a)] This diagram is calculated knowing $\chi_1^{(e1)}$, given by row 2. In this row, the first diagram represents the source for emission [first term in Eq. (10b)], the second and third diagrams represent radiative corrections to a virtual level lying below the initial level, consisting of virtual emission and reabsorption [second diagram, corresponding to the $n=2$ component in the second term of Eq. (10b)] and virtual absorption and reemission [third diagram, corresponding to the $n=0$ component of the second term of Eq. (10b)]. Finally, the fourth diagram represents radiative corrections to the initial state [the coupling term in Eq. (10b)] and is calculated knowing $\chi_1^{(a1)}$ of row 1.

It is easy to see that if ω_p is near to a pole of $g_1(r,r';\omega_{1s}+\omega_p)$, as in the present application, then the dominant term in row 2 is the fourth diagram, containing the coupling of the emission response function $\chi_1^{(e1)}$ to the *resonant* absorption response function $\chi_1^{(a1)}$. Substitution of $\chi_1^{(e1)}$, defined by keeping only this dominant term, into the fourth term of row 1 then gives an uncoupled equation for $\chi_1^{(a1)}$, where the decoupling has been accomplished by this distorted-wave Born approximation (DWBA) procedure. However, we now note that the second and third diagrams in row 1, are at least of order I/I_0, while the fourth diagram, after the decoupling, is at least of order $(I/I_0)^2$ relative to the source. Thus the fourth diagram would contribute radiative corrections of order $(I/I_0)^2$ and can be dropped.

Note that if the source for emission (row 2) had not been neglected relative to the fourth diagram (row 2), then the substitution of $\chi_1^{(e1)}$ into the term (fourth diagram of row 1) coupling it to $\chi_1^{(a1)}$ would have generated a second source term, of order $(I/I_0)^{1/2}$ relative to the original source. Since these terms originate from the radiative corrections to the initial state, we observe that Gontier and Trahin were correct, in the above sense, when they dropped the radiative corrections to the initial state as "unessential."

One must be careful, however, because under certain conditions the coupling terms in rows 1 and 2 can play an important role. This would occur in the two-photon ionization from an *excited* state of an atom such that a real state could exist below the initial level and the emission arrows of row two could reach this state. This would correspond to a region of ω_p in which $g_1(r,r';\omega_i-\omega_p)$ (ω_i, energy of the initial excited state), not $g_1(r,r';\omega_i+\omega_p)$, is near a pole (resonant emission). Now this channel is closed, assuming there are no states deeper in the atom which could be reached by two-photon emission, so that conservation of energy would require the reabsorption of this photon, giving rise to a resonant emission loop (fourth diagram, row 1), followed by arrows leading to the outside by additional absorptions, resulting in the production of electrons. Thus, in the absence of other competing processes there should be an amplification in the number of electrons, produced as a result of the resonant emission, followed by reabsorption, of a photon temporarily trapped in the interior of the atom. Thus, it may be possible to observe a resonance-type peak in a region of ω_p which is otherwise nonresonant, i.e., in which the laser is *not* tuned to an intermediate level *above* the initial level. This process has its analog in the resonance scattering of electrons[16] in which the incident electron can be temporarily captured by the target leading to the rapid rise of the scattering cross section to its maximum value over the narrow range of energy in which the capture can occur (resonance peaks super-imposed on a smooth background).

We note that only diagram 2 of row 1 has an imaginary component owing to the outgoing-scattering boundary conditions appropriate for the Green's function whose energy lies above the ionization threshold of the atom $\omega_{1s}+2\omega_p$ in this case (see the Appendix for further discussion). Thus the entire contribution to the width, of order I/I_0 relative to the source, arises from this term. The existence of a width must depend on the decay of the atom by electron emission; this electron is then recaptured to conserve energy. The other virtual processes involving deexcitation followed by excitation or vice versa contribute only to the shift.

F. Nonperturbative solution to the uncoupled integral equation for the absorption response function $\chi_1^{(a1)}(r,\omega_{1s}+\omega_p)$

Based on the above analysis we want to solve Eq. (10a) without the coupling term (fourth diagram, row 1) in the vicinity of the $1s\rightarrow 2p$ resonance. This is an uncoupled integral equation for $\chi_1^{(a1)}$. As pointed out by Chang and Poe,[17] in the case of the perturbative solution for complex atoms where the potential terms, other than the source, arise from the interaction of the electron absorbing the photon with the atomic field, a stable numerical solution is hard to obtain near a pole of the Green's function. This was also found to be the case by the present author.[6] In addition the noniterative method of solution presented in Ref. 6 fails in the present application owing to the infinite range of the radiative interactions.

Chang and Poe[17] have outlined a procedure which we propose as a method capable of handling the present equation. They use the well-known approximation[1] method of representing the Green's function very near a pole (vanishing energy denominator) by a single eigenfunction of the target (or by several eigenfunctions if there are several closely spaced poles). In the limit of coming very close to the pole this approximation becomes very accurate. They then suggest finding the exact solution to the integral equation (in their discussion, differential equation) sufficiently far enough on the wings of the resonance that numerical difficulties do not arise. The eigenfunction contribution is then subtracted from this solution, and the sum of the two components, the remainder, and the eigenfunction contribution is then continued as close as is numerically feasible for the integral equation, hopefully close enough so that the single eigenfunction representation gives the entire contribution. The contribution close to the pole in the present problem is

$$\chi_1^{(a1)} = \frac{\psi_{2p}(r)\int_0^\infty dr'\, r'^2 \psi_{2p}(r') U_{10}(r') \psi_{1s}(r')}{(\omega_{1s} - \omega_{2p}) + \omega_p - \Delta_+ - i\Gamma}\,, \tag{14a}$$

$$\Delta_+ = \mathrm{Re}\Bigg(\sum_{j=0,2}^{2} \sum_{n=0,2}^{2} \int_0^\infty \int_0^\infty dr\, dr'\, r^2 r'^2 \psi_{2p}(r) U_{1j}(r) \times g_j(r, r'; \omega_{1s} + n\omega_p) \times U_{j1}(r')\psi_{2p}(r') \Bigg)\,, \tag{14b}$$

$$\Gamma = \mathrm{Im} \sum_{j=0,2}^{2} \int \int dr\, dr'\, r^2 r'^2 \psi_{2p}(r) U_{ij}(r) \times g_j(r, r'; \omega_{1s} + 2\omega_p) U_{1j}(r')\psi_{2p}(r') \Bigg)\,. \tag{14c}$$

At this level of approximation this solution is exact. That is, if $g_1(r, r'; \omega_{1s} + \omega_p)$ of Eq. (10a) (with the coupling term omitted) can be represented by a single eigenfunction, then the exact noniterative solution given by Eqs. (14) results. In fact, if g_1 were to be represented by N such eigenfunctions (as in the case of N closely spaced poles), then the exact noniterative solution could still be constructed, requiring the inversion of an N-dimensional matrix. Thus, it is only in the one-eigenstate approximation that the solution is simple enough to be available in the analytic form given by Eqs. (14).

We observe that Eqs. (14) give a result implied by the renormalized Green's function method of Gontier and Trahin.[2(a)] This can be seen by inspection of Eq. (3) of Ref. 1, where the method is applied to the four-photon ionization of Cs, and the single $6F$ eigenfunction is used to represent the singular Green's function in the vicinity of the $6S \rightarrow 6F$ resonance. However, Morellec, Normand, and Petite[1] did not note that the quantities which they call the "shifts" will be complex, with real and imaginary parts corresponding to the shift and width, respectively, [as in Eqs. (14)] when the summations over the sets of intermediate bound and continuum states of the target are carried out explicitly, after first imposing the following boundary conditions: (i) outgoing-scattering boundary conditions if the number of photons absorbed causes the virtual energy to lie in the ionization continuum [as a virtual energy of $\omega_{1s} + 2\omega_p$ does in our case or as $\omega_{6S} + 4\omega_p$ does in the first term of Eq. (4c) in Ref. 1]; these boundary conditions produce the complex correction [see Eq. (A1) of the Appendix]; (ii) exponentially decaying boundary conditions if the virtual energy is less than the ionization threshold either by absorption of too few or by emission of one or more photons.

This last result can be obtained from the result of (11) above by analytic continuation below the energy axis accoring to $k = iZ/\nu$, where $\frac{1}{2}k^2 = \omega$ is the energy of the virtually ionized electron and ν is the effective principal quantum number defined previously (see the Appendix for greater detail). Analytic continuation of the scattering Green's function with incorrect boundary conditions will lead to a result below the axis having irregular (exponentially growing) behavior. This point can be easily understood if the energy denominators of the "shifts" of Ref. 1 are replaced by the appropriate resolvent operators and these represented according to the above prescriptions for the boundary conditions. The inspection of Eqs. (14c) and (A1) shows that Γ is proportional to the one-photon integral cross section for the ionization of the $2p$ state, which is just the width expected from elementary considerations. Finally, it is interesting that the renormalized Green's-function method of Ref. 2(a) and the single-eigenfunction approximation version of the present theory give the same result. This identification disappears when $\chi_1^{(a1)}$ is obtained from the numerical solution of the integral equation, constructed accoring to the method of Chang and Poe.[17]

III. NUMERICAL APPLICATION OF EQS. (14)

We present numerical results for the single-eigenfunction approximation to $\chi_1^{(a1)}$ in Figs. 2–5. The calculations of Fig. 2 give the error to be expected from this approximation to $g_1(r, r'; \omega_{1s} + \omega_p)$. This is seen to be extremely small in the range of wavelengths from 1210 to 1218 Å, but is about 27% at 1240 Å. These calculations were made in the perturbative limit (setting the shift and width equal to zero). The exact calculations of the present study were performed using the numerical representation of g_1 described in the Appendix of Ref. 6 (also see Refs. 4 and 5 for exact calculations for multiphoton processes using the Coulomb Green's functions). Also shown at 1200 and 1240 Å are the exact results of Chan and Tang,[9] using a different technique. The two calculations agree to within 1%; this is within the error tolerance of the single-eigenfunction approximation at about 1 Å away

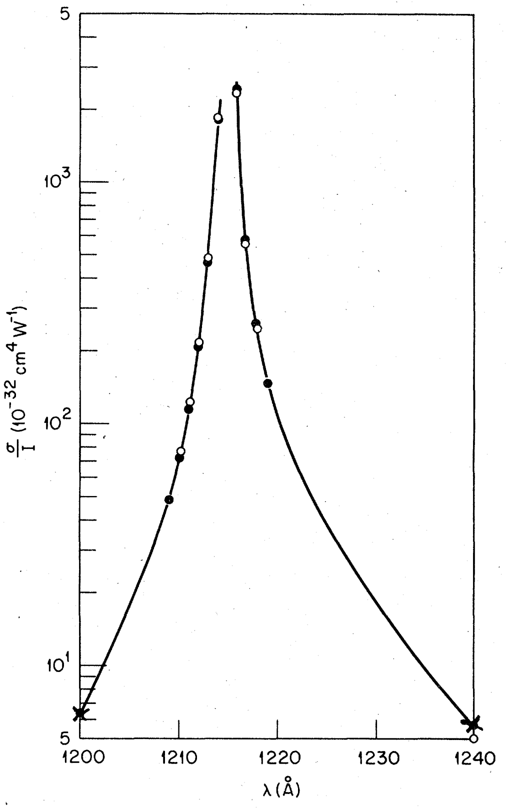

FIG. 2. Cross sections for the two-photon ionization of H(1*s*) using perturbation theory (all radiative corrections set equal to zero). ✕ : exact results of Chan and Tang, Ref. 9; ●: exact results of the present work; ○ : single-eigenfunction approximation of the present work.

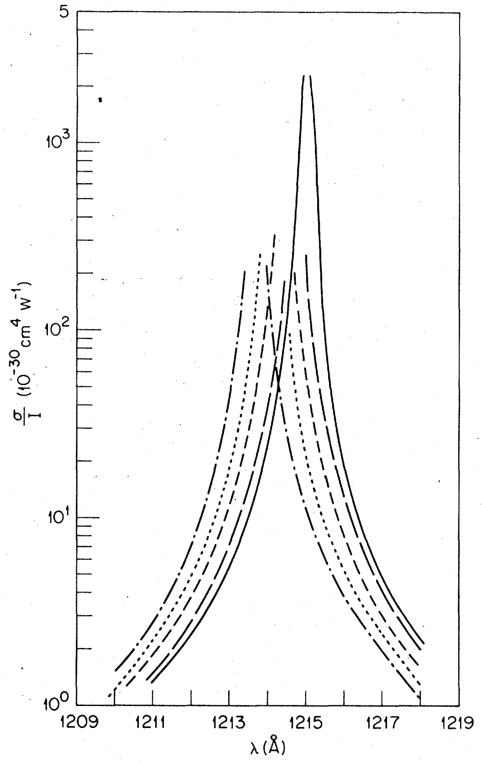

FIG. 3. Cross sections for the two-photon ionization of H(1*s*) from Eqs. (14). —, $I = 10^{12}$ W cm^{-2}; — —, $I = 2.5\times10^{12}$ W cm^{-2}; – – –, $I = 5\times10^{12}$ W cm^{-2}; ---, $I = 7.5\times10^{12}$ W cm^{-2}; – • –, $I = 10^{13}$ W cm^{-2}.

from the static position of the resonance 1214.7 Å.

Figure 3 shows the behavior of the resonance peaks as a function of intensity from 10^{12} to 10^{13} W cm^{-2}. No attempt was made to locate the precise dynamic position of the resonance, since this would require a large number of points, each one of which is rather expensive in machine time owing to the need to calculate Δ_+ and Γ at each ω_p. Note the shift of the peak to smaller wavelengths as a function of increasing intensity.

Figure 4 gives a comparison of resonance peaks at 10^{13} W cm^{-2} for diagrams 2 and 3 of row 1 of Fig. 1 (virtual ionization followed by recombination and virtual emission followed by reabsorption, respectively), and for diagram 2 alone. We observe that the largest contribution to the shift comes from diagram 3, specifically from the $j = 2$ term of Eq. (14b) for $n = 0$. Hence, the quantitative description of the resonance peak would be incorrect if only one of the two diagrams were calculated.

Figure 5 shows the order of nonlinearity at

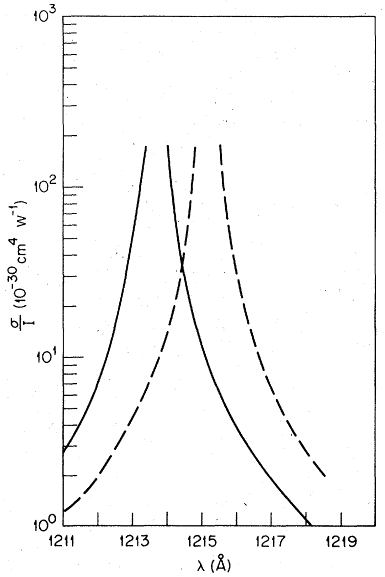

FIG. 4. Cross sections at $I = 10^{13}$ W cm^{-2} for the two-photon ionization of H(1*s*). — as in Fig. 3; – – result with terms of diagram 3, row 1 of Fig. 1 omitted.

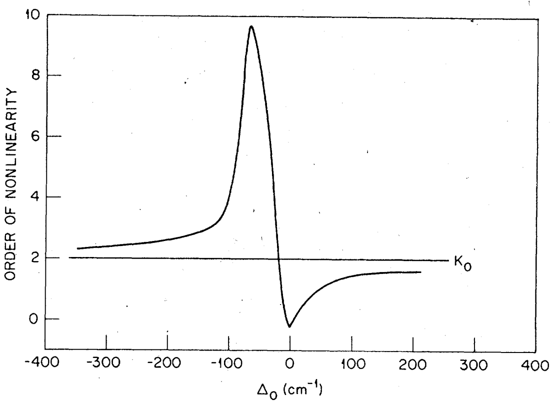

FIG. 5. Order of nonlinearity, i.e., derivative of the logarithm of the number of ions produced with respect to the logarithm of the intensity vs the static detuning at $I = 5\times10^{12}$ W cm^{-2}, where the static position is taken at 1215 Å.

5×10^{12} W cm^{-2} [see Eq. (1) of Ref. 1, where it is called K_{expt}, the experimental order of nonlinearity]. This curve is simply our estimate of the derivative of the logarithm of the counting rate for the number of ions produced (N_i of Ref. 1) with respect to the logarithm of the intensity versus the static detuning Δ_0. We note that it slowly approaches $K_0 = 2$ for large positive and negative Δ_0, where K_0 is the number of photons absorbed. Also, the shape closely resembles that of the measured order of nonlinearity for the four-photon ionization of Cs (Fig. 7 of Ref. 1). Since this quantity is a sensitive function of the dynamic detuning and radiation-induced linewidth, it should form the basis for detailed comparisons of theoretical and experimental results over a wide range of intensities.

ACKNOWLEDGMENTS

The author would like to thank Dr. M. G. Payne, Dr. G. S. Hurst, Dr. R. N. Compton, and Dr. W. R. Garrett of the Health Physics Divisions, Oak Ridge National Laboratory for their interest in this work and for helpful discussion concerning the physical interpretation of some of the results. He would also like to thank Verner Anderson for extremely able programming assistance. This research was sponsored by the U. S. Energy Research and Development Administration under contract with Union Carbide Corporation.

APPENDIX: COULOMB'S GREEN'S FUNCTIONS

The Coulomb Green's functions required for this problem have been extensively studied. They are as follows:

(i) Green's function for scattering boundary conditions,

$$g_l(r,r';\tfrac{1}{2}k^2) = \frac{1}{krr'}\,[G_l(kr_>)F_l(kr_<) + iF_l(kr)F_l(kr')]\,, \tag{A1}$$

for energy $\frac{1}{2}k^2$(a.u.). G_l and F_l are the irregular and regular Coulomb functions described, for example, in Mott and Massey.[18]

(ii) Green's functions for exponentially decaying boundary conditions. These functions can be obtained from those of Eq. (A1) by analytic continuation below the energy axis, according to $k = iZ/\nu$, where ν is an effective principal quantum number. These functions have been studied extensively in Ref. 10. Also see the Appendix of Ref. 6 for a description of their numerical evaluation. They are

$$g_l(r,r';\omega) = -\frac{\nu}{2Zrr'}\,\frac{\Gamma(l+1-\nu)}{\Gamma(2l+2)}\,M_{\nu;l+1/2}\left(\frac{2Zr_<}{\nu}\right) \times W_{\nu;l+1/2}\left(\frac{2Zr_>}{\nu}\right), \tag{A2}$$

where $M_{\nu;l+1/2}$ and $W_{\nu;l+1/2}$ are the Whittaker functions and $\nu = iZ(2\omega)^{-1/2}$, where ω is the energy on the negative axis in atomic units.

(iii) Reduced Green's functions. These are required to describe propagator lines in which all photons with energy ω_p have been lost; thus, they are appropriate for calculating the static Stark shifts. These are described in Ref. 15. They can be obtained from the Green's functions of (ii) above by the limiting procedure described in Ref. 15:

$$g_l^{(i)}(r,r') = \lim_{\omega\to\omega_i}\left(\frac{\partial}{\partial\omega}(\omega-\omega_i)g_l(r,r';\omega)\right). \tag{A3}$$

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