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New Photon-Correlation Effects in Near-Resonant Multiphoton Ionization

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It is shown that in near-resonant N-photon ionization, chaotic light can be more efficient than purely coherent light by more than the usual factor of $N!$ (N factorial). This occurs when the resonant intermediate state interferes with another nearby state. Specific results are presented for two-photon ionization.

Multiphoton processes depend on the photon correlation (coherence) properties of the radiation and not simply on the average intensity, as single-photon processes do.[1-4] In short,[5,6] the yield of nonresonant N-photon processes with chaotic (incoherent) radiation is larger by a factor of $N!$ than with purely coherent (Glauber state) radiation. More generally, the process is proportional to the Nth-order correlation function of the radiation, a result intimately related to the nonresonant nature of the process which leads to the factorization of the field-correlation function from the atomic parameters. As shown in two recent theoretical papers,[7,8] in the presence of a resonant intermediate atomic state, this dependence on photon correlations changes considerably. For example, in two-photon ionization via one intermediate state, all dependence on photon correlations disappears when the transition from the initial to the (resonant) intermediate atomic state is completely saturated, which occurs in the limit of large field intensities. Well below saturation, one has the usual factor of 2! between chaotic and purely coherent radiation. Similar results are obtained for resonant processes of higher order. Under saturation conditions the difference between the effect of chaotic and coherent light diminishes. Thus the factor $N!$ has so far been considered as the maximum ratio between chaotic and coherent radiation for multiphoton transitions into a continuum.

In this Letter, we report a surprising new result. We show that under near-resonance conditions, the ratio of the yields for chaotic to that for purely coherent radiation can be significantly larger than $N!$ (N factorial). This occurs when a near-resonant intermediate state interferes with another nearby state or even with a background due to more than one distant nonresonant level.

The problem can be formulated in more than one way. We have chosen the resolvent-operator[9] formalism which has been used in a number of recent papers on resonance processes. Consider an initial atomic state $|g\rangle$ and a single-mode photon state $|n\rangle$ of frequency ω. The initial state of the system "atom plus field" is $|I\rangle = |g\rangle|n\rangle$. Consider in addition the system states $|A\rangle = |a\rangle|n-1\rangle$ and $|B\rangle = |b\rangle|n-1\rangle$, where the atomic states $|a\rangle$ and $|b\rangle$ are both connected to the state $|g\rangle$ by a dipole single-photon transition. Let $|F\rangle = |f\rangle|n-2\rangle$ be the final state for two-photon ionization, where the atomic state $|f\rangle$, assumed here to be in the continuum, is connected to both $|a\rangle$ and $|b\rangle$ by a single-photon electric dipole transition. The energies of the system states are denoted by ω_I, ω_A, ω_B, and ω_F and are measured in inverse seconds, as all Hamiltonians have been divided by $\hbar$. In terms of atomic and field energies we have $\omega_I = \omega_g + n\omega$, $\omega_A = \omega_a + (n-1)\omega$, $\omega_B = \omega_b + (n-1)\omega$, and $\omega_F = \omega_f + (n-2)\omega$. We shall be interested in photon frequencies near and around the resonance frequency $\omega_{ag} \equiv \omega_a - \omega_g$. It is rather straightforward to write a set of equations governing the relevant matrix elements of the resolvent operator $G(z)$. These equations as obtained from the fundamental relation,[9] $(z-H^0)G=1+VG$, are

$$(z-\omega_I)G_{II}=1+V_{IA}G_{AI}+V_{IB}G_{BI}, \tag{1a}$$

$$(z-\tilde{\omega}_A)G_{AI}=V_{AI}G_{II}+\textstyle\sum_F V_{AF}G_{FI}, \tag{1b}$$

$$(z-\tilde{\omega}_B)G_{BI}=V_{BI}G_{II}+\textstyle\sum_F V_{BF}G_{FI}, \tag{1c}$$

$$(z-\omega_F)G_{FI}=V_{FA}G_{AI}+V_{FB}G_{BI}, \tag{1d}$$

where H^0 is the unperturbed Hamiltonian of the system and V the interaction coupling the electron to the field. A typical matrix element of V is, for example,

$$V_{AI}=i(2\pi e^2/\hbar)^{1/2}L^{-3/2}\omega^{1/2}n^{1/2}\langle a|\vec{\epsilon}\cdot\vec{r}|g\rangle, \tag{2}$$

with L the dimension of the quantization box and $\vec{\epsilon}$ the polarization vector of the radiation mode. Since we are working in the single-mode formalism, a more useful expression for V_{AI} in terms of more conventional and easier to determine parameters is

$$V_{AI}=(2\pi\alpha F\omega|\langle a|\vec{\epsilon}\cdot\vec{r}|g\rangle|^2)^{1/2}, \tag{3}$$

where F is the total photon flux in number of photons/cm$^2\cdot$sec and α is the fine-structure constant. In Eqs. (1b) and (1c) the energies $\tilde{\omega}_A$ and $\tilde{\omega}_B$ are complex given by $\tilde{\omega}_A=\omega_A-i\gamma_a$ and $\tilde{\omega}_B=\omega_B-i\gamma_b$, where γ_a and γ_b are the widths of states $|a\rangle$ and $|b\rangle$, respectively. These widths may contain natural as well as induced (intensity-dependent) contributions. There are also shifts which are not exhibited explicitly here as they are not essential to the argument. Such widths and shifts are rigorously derived in the development of the equations and are not introduced phonomenologically, although the result would be the same. If one wishes to include the effect of more than one state $|b\rangle$, Eqs. (1a) and (1d) are replaced by

$$(z-\omega_I)G_{II}=1+V_{IA}G_{AI}+\textstyle\sum_B V_{IB}G_{BI}, \tag{1a$'$}$$

$$(z-\omega_F)G_{FI}=V_{FA}G_{AI}+\textstyle\sum_B V_{FB}G_{BI}, \tag{1d$'$}$$

where the summation can in principle be over a complete spectrum of states $|b\rangle$. In that case, such states provide a background that interferes with the contribution of state $|a\rangle$. This background often depends on the photon frequency to an extent that cannot be neglected, depending on the detuning $\omega-\omega_a+\omega_g$.

Considering now two interfering resonances, we solve Eq. (1d) for G_{FI} and substitute into Eqs. (1b) and (1c) which become

$$\left(z-\tilde{\omega}_A-\sum_F\frac{|V_{FA}|^2}{z-\omega_F}\right)G_{AI}=V_{AI}G_{II}+\sum_F\frac{V_{AF}V_{FB}}{z-\omega_F}G_{BI}, \tag{4a}$$

$$\left(z-\tilde{\omega}_B-\sum_F\frac{|V_{FB}|^2}{z-\omega_F}\right)G_{BI}=V_{BI}G_{II}+\sum_F\frac{V_{BF}V_{FA}}{z-\omega_F}G_{AI}. \tag{4b}$$

The summations (integrations) on both sides of these equations can now be replaced by z-independent quantities by setting $z=\omega_A$ in Eq. (4a) and $z=\omega_B$ in Eq. (4b). Each of these terms gives rise to a real part and an imaginary part, in general. The imaginary parts of the terms inside the parentheses represent the ionization widths of states $|a\rangle$ and $|b\rangle$. The above approximation is completely justified for a situation in which we have a transition into a smooth continuum and intensities for which perturbation theory is valid. All of these conditions are satisfied here and our procedure is essentially the usual way ionization is introduced in the formalism. The summations in the right-hand side of the equations are terms that couple the states $|A\rangle$ and $|B\rangle$ mainly through transitions into the continuum and are very important in our problem. We introduce the notation $\Gamma_J\equiv-\mathrm{Im}\sum_F|V_{JF}|^2(\omega_J-\omega_F)^{-1}$ for $J=A, B$, $\mathcal{U}_{AB}\equiv\sum_F V_{AF}V_{FB}(\omega_A-\omega_F)^{-1}$, and $\mathcal{U}_{BA}\equiv\sum_F V_{BF}V_{FA}(\omega_B-\omega_F)^{-1}$. Because of the near-resonance conditions and the smooth continuum, the above-defined quantities are numerically comparable and often practically equal.

We can now solve the system of Eqs. (1a), (4a), and (4b) for G_{II}, G_{AI}, G_{BI}. This involves finding the roots of a third-degree algebraic equation in z. From the expressions for the G's we obtain $U_{II}(t)$, $U_{AI}(t)$, and $U_{BI}(t)$ through the inversion integral $U(t)=(2\pi i)^{-1}\int_c dze^{-izt}G(z)$. The fully-time-dependent ionization probability is given by

$$P(n;T)=1-|U_{II}(T)|^2-|U_{AI}(T)|^2-|U_{BI}(T)|^2, \tag{5}$$

where T is the interaction time and n indicates that P is a function of n since our initial state contained exactly n photons. The effect of photon correlations is obtained by calculating the average

$$\langle P(T)\rangle=\sum_{n=0}^{\infty}p_{nn}P(n;T), \tag{6}$$

where p_{nn} is the photon probability distribution in the initial state. Recall that for chaotic light $p_{nn}^{\text{cha}} = \langle n\rangle^n/(1+\langle n\rangle)^{n+1}$ while for a pure coherent state $p_{nn}^{\text{coh}} = e^{-\langle n\rangle}\langle n\rangle^n/n!$, where, in both cases, $\langle n\rangle = \sum_n p_{nn} n$ is the average number of photons (the intensity).

The above treatment of the problem of two intermediate resonances with ionization is essentially rigorous for the type of intensities that interest us in this paper. Under certain conditions, it is also possible to describe the process by an approximate transition probability per unit time (rate). Although less rigorous in general, it yields surprisingly good results in limiting cases; that is well below or well above saturation. It also turns out that it provides a very good estimate of the photon-correlation effects.

We give here a very brief outline of the derivation of such a rate. This is appropriate when the effect of one of the intermediate states, say $|b\rangle$, can be replaced by a contribution varying very slowly with the photon frequency; which implies frequencies around the other resonance $|a\rangle$. We solve Eq. (1c) for G_{BI} and substitute into Eqs. (1a) and (1d). In the resulting expression, we replace $z - \tilde{\omega}_B$ by $\omega_A - \tilde{\omega}_B \cong \omega_I - \tilde{\omega}_B$ since we shall confine ourselves to photon frequencies for which $\omega_A \cong \omega_I$, in the sense that $|\omega_A - \omega_I| \ll |\omega_B - \omega_I|$. The resulting expression for G_{FI} can then be solved and we can again make the replacement $z - \omega_F \cong \omega_I - \omega_F$. In the summation (integration) over F, the principal value is taken. Substituting now G_{FI} into Eqs. (1a) and (1b), we obtain a system of two linear algebraic equations for G_{II} and G_{AI} which can be solved exactly—in much the same way that the equations for two strongly coupled levels are solved, except that now the effect of the other level(s) is included. The complete derivation involves considerable algebraic detail which shall be presented elsewhere. The resulting transition probability per unit time is

$$W(n) = Cn(n-1)\frac{[1+q(n-1)]^2}{\delta^2 + An[1+q(n-1)]^2}, \qquad (7)$$

where $\delta = \omega - \omega_a + \omega_g$, $\Delta = \omega_a - \omega_b$, $A = r_{ag}^{\ 2}$, $q = \mathrm{P}\int d\omega_f \times \beta r_{bf} r_{fa} r_{gb}/r_{ag}(\Delta+\delta)\delta$, the coefficients C and β contain all constants, and P indicates the principal value. The parameter q is an approximate measure of the interference between the contributions of $|a\rangle$ and $|b\rangle$. For $q=0$ we recapture the special case of a single resonant intermediate state of Ref. 6. If one wishes to include the effect of more than one state $|b\rangle$, the derivation is similar but q will then involve a summation over b with Eq. (7) remaining the same. The derivation outlined here can also be obtained using Eqs. (1a), (4a), (4b), and (1d). Note also that, in general, δ should contain intensity-dependent terms[10] in Eq. (7). One can show, however, that they are negligible for the problem under consideration. The effect of photon correlations is again obtained by averaging $W(n)$ over the photon number distribution p_{nn}. We emphasize that this result is approximate but in our calculations we find it to be a very good approximation. One must, of course, always compare to the more rigorous time-dependent result of Eq. (5).

We have performed calculations of $\langle P(T)\rangle$ as well as $\langle W\rangle$ for chaotic and coherent light with atomic parameters corresponding to optical transitions in alkali atoms where $|a\rangle$ and $|b\rangle$ could be a $P_{1/2}$ and $P_{3/2}$ fine-structure pair. Typical results for the ratio ρ of the yields for chaotic light to coherent light are shown in Fig. 1. As

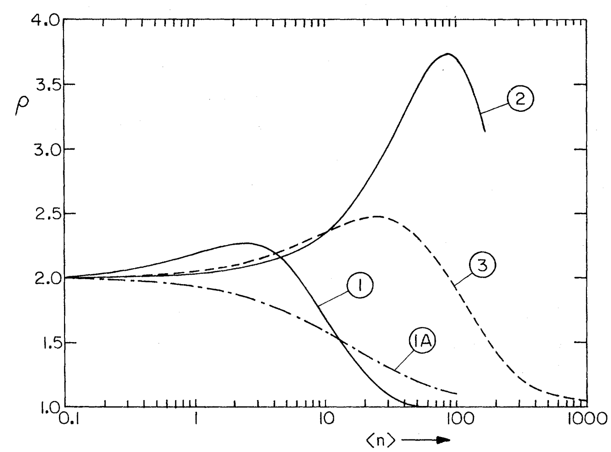

FIG. 1. Ratio of resonant two-photon ionization yields for chaotic light to that for coherent light. The variable $\langle n\rangle$ represents the average number of photons in the laser mode. The numerical results have been obtained for a scaled problem to facilitate numerical computation. In an actual atom, the effect would be observed for $\langle n\rangle$ about two orders of magnitude larger than shown here. Curve 1A corresponds to the results of Ref. 7 (single resonant intermediate state). Curves 1 and 2 correspond to two interfering resonances and have been obtained using the approximate equation (7). The parameters used in the calculation of the curves were $\delta = 3\times10^7$ Hz, $A = 0.695\times10^{15}$ Hz^2, $q = 0.0455$ for curve 1; $\delta = 3\times10^7$ Hz, $A = 0.695\times10^{15}$ Hz^2, $q = 0.0$ for curve 1A; $\delta = 3\times10^8$ Hz, $A = 0.695\times10^{15}$ Hz^2, $q = 0.0045$ for curve 2. Curve 3 has been obtained through the time-dependent expression of Eq. (5) for $T = 10$ nsec, $\delta = 1.2\times10^9$ Hz, and $\Delta = 3\times10^9$ Hz, with matrix elements corresponding to the $6S \to 7P \to \epsilon D, \epsilon S$ transitions in Cs. For this curve the scale of $\langle n\rangle$ should be multiplied by 10^{-2}.

mentioned earlier, $\rho=2$ for nonresonant processes as well as for a single resonant state well below saturation. In the limit of large intensity and a single resonance, ρ decreases monotomically to 1 (see curve 1*A* of Fig. 1). Clearly we have a drastically different behavior when the intermediate resonance interferes with other states. Again the weak- and strong-field limits are $\rho=2$ and $\rho=1$, respectively, as they should be. Between these two limits, however, there is an intensity regime where ρ reaches a maximum significantly larger than 2. Its exact value depends on the particular set of atomic parameters and the detuning. As seen in Fig. 1 we have obtained values as large as 3.75. Evidently, the intermediate virtual state resulting from the interference of $|a\rangle$ with other state(s) is more sensitive to the fluctuations of the light source thus giving a value of ρ significantly larger than 2. Stated somewhat differently, as a result of interference there are virtual transitions of "*apparent*" order higher than 2.

To facilitate the numerical calculation arising from the required summation in Eq. (6), we have scaled the parameters of the problem. Thus the effect represented by the curves of Fig. 1 would occur—in a real atomic transition—for $\langle n\rangle$ about two orders of magnitude larger than shown in the figure. These values of $\langle n\rangle$ correspond to cw-laser powers in the range of 10–100 mW and bandwidths of the order of 0.01–0.001 cm^{-1}. Although relatively weak by laser standards, these intensities are sufficiently large to saturate a typical bound-bound atomic transition. The physical picture that seems to be emerging is that the effect occurs when the light intensity is sufficiently large to populate the interfering excited states but not so large as to cause saturation of the ionization. Ionization is here the dominant damping mechanism for large intensities.

We have also considered three-photon ionization with a two-photon near-resonance between the initial and two bound excited states in which case we have found ratios as high as 9.35 instead of $3!=6$. It is evident that similar effects are more pronounced in higher-order resonant processes. Our results demonstrate that photon-correlation effects in multiphoton processes contain considerably more variety than hitherto suspected. In combination with resonant intermediate states, they presently constitute one of the most interesting and active areas of investigation.[3,11] If as we have shown the ratio ρ can have a maximum value larger than $N!$ and also depends on the laser duration, then experiments of the type reported in Refs. 3 and 11 acquire new significance.

A complete account of this work with further results will be published elsewhere. This work was supported by National Science Foundation Grant No. PHY 76-23163.

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