yond the atomic field in order to observe a departure from the multiphoton process in the ionization of an atom with very high laser intensity. We hope that these experimental results will stimulate further theoretical work in this direction, especially now that with high power lasers developed for laser-fusion experiments, $E/E_0 > 1$ can be attained.

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Photon-Correlation Effects in Resonant Two-Photon Ionization

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The effect of photon correlations (coherence) on the transition probability of resonant two-photon processes is discussed. It is shown that coherence influences not only the transition probability but also the linewidth of the resonance. The implications for higher-order multiphoton ionization are discussed.

Two relatively recent experiments[1,2] have brought again to the forefront one of the most interesting aspects of multiphoton processes; the dependence of the total transition rate on the coherence (correlation) properties of the light source. Krasinski *et al.*[1] have shown that the nonresonant two-photon absorption rate with a chaotic (incoherent) source is about 1.52–1.86 times higher than with an approximately coherent (Glauber state) source. Theoretically it should have been larger by a factor of 2, but the difference is readily understood since what was assumed to be a pure coherent state in fact was not.[3] In addition, a single-photon component may have existed in the signal. This experiment resolves an ambiguity stemming from contradictory results of two earlier experiments[4,5] performed under essentially identical conditions. The pulsed lasers used in those experiments were in all probability in a more or less chaotic state (because of the presence of many modes), and thus further randomization should have produced no additional effect. Therefore, the result obtained by Carussotto, Polacco, and Vaselli[5] is what one should have expected.

The most dramatic demonstration of coherence effects was provided by a beautiful experiment[2] by Lecompte *et al.* who showed that eleven-photon nonresonant ionization of Xe atoms with a multimode laser (100 modes ≈ chaotic state) is more efficient by a factor of $10^{6.9 \pm 0.3}$ than with a single-mode laser. Theoretically, the difference between perfectly incoherent (chaotic) and purely coherent light is $11! = 3.99 \times 10^7$.

The theoretical prediction of these effects has been in print[6-15] for about ten years now, but it is only recently that improvements in laser technology have rendered their observation reasonably accessible. Their influence, of course, has always been present, causing perhaps the most serious uncertainty in the measurement of multiphoton-ionization generalized cross sections. In addition to their influence on multiphoton processes, these effects provide a rather unique way of measuring field fluctuations over very short time scales (of the order of 10^{-14} sec) which are electronically inaccessible.

One of the aspects of this problem that has received essentially no attention so far is the modification of photon-correlation effects by the presence of resonant intermediate states. In view of the recent interest in the study of resonance multiphoton processes, this would seem to be an appropriate time to address this important question. Even from the viewpoint of measuring field correlation functions through multiphoton ionization, resonance processes are important because they enhance the transition probability significantly. It is the purpose of this note to discuss the dependence of two-photon resonance absorption on the photon-correlation properties of the light source. The discussion will be specifically concerned with ionization but the general conclusions have wider applicability.

We consider a single mode of the radiation field with frequency ω. The probability of N-photon off-resonance ionization is proportional to the Nth moment of the photon probability distribution p_{nn} in the initial state; i.e., to the quantity

$$\langle n^N \rangle \equiv \sum_n p_{nn} n^N. \tag{1}$$

(It is here assumed that $\langle n\rangle \gg N$ which also implies $\langle n \rangle \gg 1$.) It is this quantity that gives the enhancement factor of $N!$ for chaotic light as compared to purely coherent light. The above quantity enters because, to lowest nonvanishing order in perturbation theory, N-photon off-resonance absorption is proportional to $|V|^{2N}$, where V is the interaction between field and atom, and $|V|^2$ is linear in the photon number n. In the presence of resonance(s) with intermediate atomic states, field-dependent quantities appear also in the denominator, and the process is no longer proportional to just $\langle n^N\rangle$. The average of a more complicated function of n must then be calculated.

In the case of two-photon ionization with a single-mode radiation field, the probability of ionization is proportional to the quantity[16]

$$G \equiv \langle n(n-1)/(n+\Delta)\rangle \tag{2}$$

where Δ is proportional to the square of the detuning from resonance (spontaneous emission is assumed to be negligible). We have lumped all parameters into Δ so as to deal with the simplest possible form of G. Equation (2) is obtained from Eq. (18) of Ref. 16a by neglecting the effects of nonresonant states which are included in that calculation. These nonresonant processes are included in Ref. 16 in order to obtain equations which are valid both near to and relatively far away from the resonance. This is not our purpose here, since, as shall be clear from the results below, the effect presently under study is one which occurs in a region very close to the resonance. For an actual atom and an optical-frequency monochromatic light source (linewidth less than 10^9 Hz), the nonresonant contribution to the present process is negligible, since the effects of the nonresonant processes will appear only far out on the wings of the resonance.[16,17] It should also be noted that Eq. (2) is valid only if the coupling between ground and intermediate states is much stronger than the coupling between the intermediate state and the continuum. This should be the case for two-photon ionization of real atoms using realistic laser power levels.[16]

The dependence of the two-photon ionization on photon statistics can easily be found in two limiting cases. For large Δ, or for weak field, $\Delta \gg \langle n\rangle$ and one finds $G \sim \langle n(n-1)\rangle \cong \langle n^2\rangle$, which is the usual off-resonance case. For strong field, and on resonance, $\Delta \ll \langle n\rangle$ which leads to $G\sim\langle n-1\rangle \cong \langle n\rangle$, indicating that in this case (strong saturation) all dependence on photon coherence disappears. This result physically is to be expected; it is as if one is ionizing atoms from an excited state via single-photon ionization.

At low field strengths, therefore (i.e. well below saturation of the intermediate state), we have the factor of 2 between chaotic and purely coherent light, and well above saturation there is no difference between the two. To obtain the behavior for intermediate situations, which are the more interesting experimentally, the average in Eq. (2) must be calculated.

It is more convenient to write G as

$$G = \langle n\rangle - (\Delta+1) + \Delta(\Delta+1)\langle 1/(n+\Delta)\rangle, \tag{3a}$$

and introduce the notation

$$\langle 1/(n+\Delta)\rangle = \sum_n p_{nn}/(n+\Delta) \equiv g. \tag{3b}$$

One must now calculate g for chaotic light, in which case[6,18]

$$p_{nn} = \langle n\rangle^n/(1+\langle n\rangle)^{n+1}, \tag{4a}$$

and for purely coherent light (Glauber[18] state), for which[6,18]

$$p_{nn} = (e^{-\langle n\rangle}\langle n\rangle^n)/n!, \tag{4b}$$

where in both cases $\langle n\rangle \equiv \sum_n p_{nn} n$. Note that in the off-resonance case, the absence of the n in the denominator enables one to write G in terms of creation and annihilation operators and evaluate it quite simply by using the P representation of the density operator.[6] Here the situation is considerably more complicated.

Coherent state.—Setting for the moment $\langle n\rangle \equiv x$ one observes that $\sum_n x^{n+\Delta-1}/n! = x^{\Delta-1}e^x$. Using this, one can write

$$g = \sum_n \frac{x^n}{n!}\frac{e^{-x}}{n+\Delta} = x^{-\Delta}e^{-x}\int_0^x dx' e^{x'} x'^{\Delta-1}.$$

The integral in the right-hand side of the above equation is essentially the incomplete γ function[19] for which one can use known series expansions.

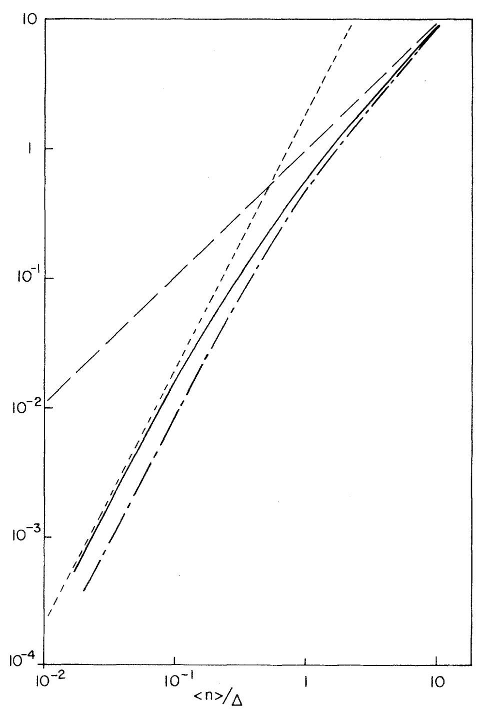

FIG. 1. G/Δ versus $\langle n\rangle/\Delta$ for a chaotic field (——), and for a coherent field (—— - ——). Also shown are the asymptotic curves $G = 2\langle n\rangle^2/\Delta$ (in the limit $\langle n\rangle/\Delta \ll 1$ for the chaotic field) (— — —), and $G = \langle n\rangle$ (in the limit $\langle n\rangle/\Delta \gg 1$ for both fields) (------).

Thus, for the coherent state we have

$$g^{\mathrm{coh}} = \langle n\rangle^{-\Delta}e^{-\langle n\rangle}\int_0^{\langle n\rangle} dy\, e^y y^{\Delta-1}$$
$$= (-\langle n\rangle)^{-\Delta}e^{-\langle n\rangle}\gamma(\Delta, -\langle n\rangle). \tag{5}$$

Chaotic state.—Setting $\langle n\rangle \equiv z^{-1}$, we have

$$g = \frac{z}{z+1}\sum_n \frac{(1+z)^{-n}}{n+\Delta} \equiv \frac{z}{z+1}f(z),$$

which defines $f(z)$. Using the definition of f and taking the derivative with respect to z, a differential equation for f is obtained which is easily solved. The result is

$$f(z) = (1+z)^\Delta \int_0^\infty \frac{dz'}{z'(1+z')^\Delta}.$$

Then we have

$$g^{\mathrm{chaotic}} = \frac{(1+\langle n\rangle)^{\Delta-1}}{\langle n\rangle^\Delta}\int_0^{\langle n\rangle}\frac{dy\, y^{\Delta-1}}{(1+y)^\Delta}, \tag{6a}$$

where the integral is the hypergeometric function,[19] or

$$g^{\mathrm{chaotic}} = (1/\Delta)F(1,1,\Delta+1,-\langle n\rangle). \tag{6b}$$

We have calculated the dependence of G on light intensity, i.e., the average number of photons $\langle n\rangle$, with detuning (Δ) held constant. A typical result is given in Fig. 1 which shows how for large $\langle n\rangle/\Delta$ the dependence of G/Δ on photon statistics ceases to exist. These curves were obtained for $\Delta = 50$. We have also calculated $G/\langle n\rangle$ as a function of $\Delta/\langle n\rangle$ for constant light intensity (fixed $\langle n\rangle$). An illustrative result is shown in Fig. 2, where $\langle n\rangle = 30$. As expected, the factor of 2 is recaptured for large Δ (detuning).

This figure also shows that the observed width of the two-photon resonance absorption will depend on the correlation properties of the radia-

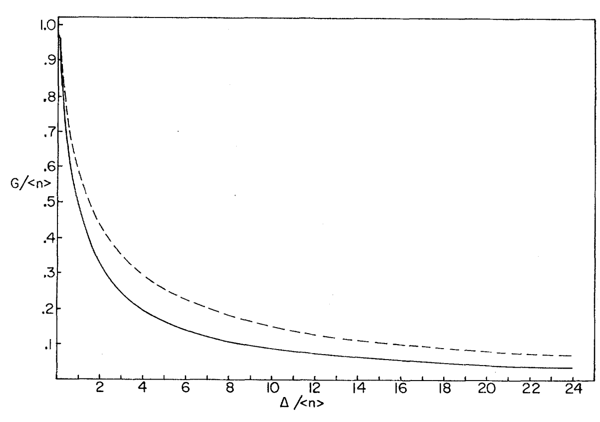

FIG. 2. $G/\langle n\rangle$ versus $\Delta/\langle n\rangle$ for a chaotic field (— — —), and for a coherent field (——).

tion field. In the particular example of Fig. 2, the linewidth for chaotic light is about 20% larger than for coherent light. It should be noted that the curve of $G/\langle n\rangle$ versus $\Delta/\langle n\rangle$ is relatively insensitive to the value of $\langle n\rangle$ and thus Fig. 2 can be regarded as indicative of the general, rather than a specific, result. The actual value of $\langle n\rangle$ chosen for the calculation of Fig. 2 is of the order of magnitude that corresponds to a laser power of about 1–10 mW, over a beam area of about 10^{-3} cm^2, and with a bandwidth of the order of 10–100 MHz. These numbers represent a situation usually met in experiments of this type.

The particular values chosen for the calculations represent two illustrative specific cases. Given an atom, the light intensity that corresponds to these cases will depend on the strength of the matrix element connecting the initial and intermediate states. It has been assumed here that this matrix element is much larger than that connecting the intermediate state with the continuum, which will usually be the case for two-photon ionization of atoms. For the sake of generality, it should be noted that for the case in which the coupling of the intermediate state to the continuum is stronger than its coupling to the ground state and still the effects of nonresonant processes are felt only in the wings of the resonance, one must calculate an average of the form $\langle n(n-1)/[(n-1)^2+\Delta]\rangle$ for two-photon ionization. This can be reduced to Eq. (2) by writing $(n-1)^2+\Delta=[(n-1)+i\sqrt{\Delta}][(n-1)-i\sqrt{\Delta}]$. This case is of particular interest in resonant N-photon ($N>2$) ionization with the last step being the resonant transition.[20] Then in many cases the coupling to the continuum would be stronger than the multiphoton coupling to the ground state. This leads to a transition probability of the form $\langle n^N/(n+D)-n^N/(n-D)\rangle$, which can be readily calculated using our results, in conjunction with the decomposition

$$\left\langle\frac{n^N}{n\pm D}\right\rangle=\left\langle\frac{n^N\pm D^N}{n\pm D}\right\rangle\mp D^N\left\langle\frac{1}{n\pm D}\right\rangle$$

and the binomial expansion to cancel the denominator in the first part of the right-hand side, thus reducing it to a linear combination of powers of n up to n^{N-1}.

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