Manifestation of nonclassical properties of strong electromagnetic radiation in multiphoton spectroscopy

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We examine multiphoton spectroscopy of a two-level system with nonvanishing intrinsic dipole moments in a two-frequency (Ω and ω) electromagnetic field. We also consider above-threshold ionization of an electron bound by a short-range potential. There is a substantial difference between the absorption coefficient and scattering spectrum of a high-frequency photon Ω in the presence of a quantized (squeezed) field ω. The latter phenomenon furnishes information about the contribution of high-order correlation functions in the nonclassical properties of a quantized electromagnetic field.

1. INTRODUCTION

The absorption and scattering of light by quantum systems in the presence of an intense coherent wave is well understood, both theoretically and experimentally.[1] The statistical properties of an external electromagnetic field have a significant impact both on the size of optical cross sections affecting the "probe" radiation and on the frequency and intensity dependence of the parameters of the external laser beam. There has been a great deal of recent interest in the interaction of squeezed light with matter, with nonclassical properties of the electromagnetic field coming to the fore.[2–4] Bergou *et al.*[5] report on the feasibility of creating intense sources of squeezed light, including some at low frequencies (micromasers[6,7]), thereby making it possible to examine the effects of squeezed photons on optical cross sections. In a typical process, the influence of a photon $\hbar\Omega$ of probe radiation with field strength $\mathscr{F}$ is much greater than that of a squeezed photon $\hbar\omega$ with field strength F. For example, in a two-level atomic or molecular system with an electronic energy gap Δ_{21}, the absorption of radiation at frequency Ω is governed by

$$\hbar\Omega=\Delta_{21}+k\hbar\omega \tag{1}$$

($k=0,\ +1,...$). The intensity of this multiphoton absorption process for the kth satellite of ω-light can be calculated in terms of the dipole-interaction matrix elements. For a generalized two-level system with nonvanishing dipole moment d in the excited electronic state state (2), and, for simplicity a vanishing dipole moment in the electronic ground state state (1), the optical cross sections can depend nonmonotonically on the intensity of the squeezed light. This results in the lowest-order correlation functions not being sufficient to describe optical processes; complete information on the quantum statistical properties of squeezed light requires that one take account of higher-order correlation functions of the ω-field.

We show in the present paper that systematic quantum statistical calculations of optical cross sections lead to equations like (15), (25), and (32).

In the simplest case of the squeezed vacuum $|0\rangle_s$, we know that $N={}_s\langle 0|a^+a|0\rangle_s=\sinh^2 r$ (a^+ and (a) are creation and annihilation operators for quanta of the ω-field, and r is the squeezing parameter[6]), and optical cross sections $\sigma^{(\mathrm{sq})}$ for some process can be represented in the form

$$\sigma^{(\mathrm{sq})}=\int_{-\infty}^{+\infty}dx|\psi(x,0)|^2\sigma^{(\mathrm{cl})}(x). \tag{2}$$

Here $|\psi(x,0)|^2=(1/\sigma)\exp(-x^2/\sigma^2)$ is a wave packet at the initial time $t=0$ ($\sigma^2=2\sinh^2 r$), which determines the distribution of the field "coordinate" x, and $\sigma^{(\mathrm{cl})}(x)$ is the optical process cross section in the presence of a classical laser field of frequency ω with field strength x.

In the general squeezed-light case $|\beta\rangle_s$ [see (4)], $N={}_s\langle\beta|a^+a|\beta\rangle_2=N_0+\sinh^2 r,\ N_0=|\beta|^2$. but there is no simple interpretation of the various formulae obtained. It is important to note that the cross section for optical processes turns out to be a sensitive function of the squeezing parameters by the second or third satellites, even at relatively low squeezed-light intensities. In the present study, we also consider a problem closely related to multiphoton spectroscopy—above-threshold ionization of quantum systems (negative ions, for example[8]) in a two-frequency (Ω, ω) field (Ω is "blue" light responsible for a single-photon process, the ω-field, which produces above-threshold peaks) with ionization potential Δ. The above-threshold peaks in the photoelectron distribution in this case are exceedingly sensitive detectors of te optical characteristics of the squeezed light.

Below, we carry out a systematic quantum statistical calculation of optical cross sections for a two-frequency (Ω, ω) electromagnetic field, as part of a quantum study of the squeezed ω-field. For the sake of simplicity, the quantum properties of the frequency-Ω probe radiation are not considered, by virtue of its single-photon participation in the optical transitions.

2. ABSORPTION OF LIGHT BY A GENERALIZED TWO-LEVEL SYSTEM IN A SQUEEZED LIGHT FIELD

Consider the absorption of probe radiation at frequency Ω by a generalized two-level system in an intense field of squeezed light at frequency ω. The cross section for such a multiphoton process is normally related to the differential probability $W_{21}^{(sq)}(\Omega)$ of absorbing probe light:

$$W_{21}^{(sq)}(\Omega)=\lim_{t\to\infty}\frac{1}{t}\langle|(S-1)_{21}|^2\rangle_{sq}. \tag{3}$$

Here $(S-1)_{21}$ is the S-matrix element between the initial (1) and final (2) states of the generalized system in the two-frequency (Ω,ω) field, and $\langle...\rangle_{sq}$ denotes series averaging with the ω-field squeezed light density operator $\rho=|\beta\rangle_{ss}\langle\beta|$. The vector $|\beta\rangle_s$ is given by[9]

$$|\beta\rangle_s=\hat{D}(\beta)\hat{S}(r)|0\rangle, \tag{4}$$

where the displacement operator $\hat{D}(\beta)$ and rotation operator $\hat{S}(r)$ take the form

$$\hat{D}(\beta)=\exp(\beta a^+-\beta^*a), \tag{5}$$

$$\hat{S}(r)=\exp\left[\frac{1}{2}r(a^2e^{-i\varphi_r}-a^{+2}e^{i\varphi_r})\right]. \tag{6}$$

Here $\beta=\sqrt{N_0}\,e^{i\varphi_\beta}$, and φ_β and φ_r are arbitrary phases. $\hat{D}(\beta)$ and $\hat{S}(r)$ have the following effects on the photon creation (a^+) and annihilation (a) operators of the electromagnetic field:

$$\begin{aligned}\hat{S}^+(r)\hat{D}^+(\beta)a\hat{D}(\beta)\hat{S}(r)&=\mu a-\nu a^++\beta,\\ \hat{S}^+(r)\hat{D}^+(\beta)a^+\hat{D}(\beta)\hat{S}(r)&=\mu a^+-\nu^*a+\beta^*,\end{aligned} \tag{7}$$

where $\mu=\cosh r$ and $\nu=\sinh r\cdot e^{i\varphi_r}$.

To lowest order in the probe-field intensity (frequency Ω), and with electron interactions with the ω-field allowed for exactly, we have from (3)

$$W_{21}^{(sq)}(\Omega)=\lim_{t\to\infty}\frac{V_{21}^2}{\hbar^2\Omega}2\,\mathrm{Re}\int_0^\infty d\tau \times\exp\left[\frac{i}{\hbar}(\tilde{\Delta}_{21}-\hbar\Omega)\tau\right]I_t(\tau), \tag{8}$$

$$I_t(\tau)=\langle G(t,t-\tau)\rangle_{sq}. \tag{9}$$

In these equations, $v_{21}=\mathscr{F}d_{21}$, $\tilde{\Delta}_{21}$ is the electron energy-level gap including the Stark shift, $G(t,t_0)$ is the evolution operator defined by

$$\begin{aligned}&i\hbar\dot{G}(t,t_0)=[g(t)a+g^*(t)a^+]G(t,t_0),\\ &G(t_0,t_0)=1,\quad g(t)=ive^{-i\omega t},\quad v=d\left(\frac{2\pi\hbar\omega}{V}\right)^{1/2},\end{aligned} \tag{10}$$

d is the dipole moment in electron state (2); $d\sim 10e_0\alpha_0$ for the level with principal quantum number $n=3$ (α_0 is the Bohr radius, e_0 is the charge on the electron, V is the normalization volume). The solution of Eq. (10) is[10]

$$G(t,t_0)=e^{A(t,t_0)}e^{-B^*(t,t_0)a^+}e^{B(t,t_0)a}, \tag{11}$$

where $A(t,t_0)$ and $B(t,t_0)$ are coefficient functions:

$$\begin{aligned}A(t,t_0)&=-\hbar^{-2}\int_{t_0}^t d\tau_1 g(\tau_1)\int_{t_0}^{\tau_1}d\tau_2 g^*(\tau_2),\\ B(t,t_0)&=-\frac{i}{\hbar}\int_{t_0}^t d\tau g(\tau).\end{aligned} \tag{12}$$

Making use of (4) and (7), the generating function in (9), $I_t(\tau)$, can be calculated immediately:

$$I_t(\tau)=\exp\Big[-|B(t,t_0)|^2\sinh^2 r -\frac{1}{2}\sinh r\cosh r(B^{*2}(t,t_0)e^{-i\varphi_r}+B^2(t,t_0)e^{i\varphi_r}) -B^*(t,t_0)\beta^*+B(t,t_0)\beta\Big],\quad t_0=t-\tau. \tag{13}$$

Passing to the limit $t\to\infty$ in Eq. (13), we easily obtain for the generating function $I(\tau)=\lim_{t\to\infty}I_t(\tau)$

$$I(\tau)=e^{-a|\nu|^2|\lambda(\tau)|^2}\sum_{m=-\infty}^{\infty}(-1)^m I_m(a\mu|\nu||\lambda(\tau)|^2) \times J_{2m}(2\sqrt{a}|\lambda(\tau)|\sqrt{N_0})e^{im\psi}. \tag{14}$$

Here $a=v^2/(\hbar\omega)^2$, $\lambda(\tau)=e^{i\omega\tau}-1$; $I_m(x)$ and $J_n(x)$ are Bessel functions of real and imaginary argument, respectively, and $\psi=2\varphi_\beta-\varphi_r$. Inserting (14) into the expression (8) for the absorption probability of Ω-light, we have after some simple manipulations

$$W_{21}^{(sq)}(\Omega)=\int dF f_\psi(F)W_{21}^{(cl)}(\Omega;F). \tag{15}$$

Here $W_{21}^{(cl)}(\Omega;F)$ is the absorption probability for frequency-Ω probe light with specified field strength F:

$$W_{21}^{(cl)}(\Omega;F)=\frac{V_{21}^2}{\hbar^2\Omega}\sum_{p=-\infty}^{\infty}J_p^2(\rho_F)\delta(\tilde{\Delta}_{21}-\hbar\Omega-p\hbar\omega) \tag{16}$$

$(\rho_F=dF/\hbar\omega)$. The function $f_\psi(F)$ takes the form

$$f_\psi(F)=\frac{2}{\sqrt{\pi}\bar{\sigma}}\frac{F}{F_0}\exp\left[-\frac{1}{F_0^2\bar{\sigma}^2}(F^2-F_0^2\cos\psi)\right]\frac{\theta(F-F_0\cos\psi/2)}{\sqrt{F^2-F_0^2\cos^2\psi/2}} \times\cosh\left[2\sqrt{F^2-F_0^2\cos^2\psi/2}\,\frac{\sin\psi/2}{F_0\bar{\sigma}^2}\right], \tag{17}$$

where $F_0=[8\pi\hbar\omega(N_0/V)]^{1/2}$ and $\bar{\sigma}^2=\bar{\sigma}^2/N_0$.

We can simplify the expression (17) for $f_\psi(F)$ in the case of phase-squeezed light $(\psi=\pi)$

$$f_{\psi=\pi}(F)=\frac{1}{\sqrt{\pi}}\frac{1}{\bar{\sigma}F_0}\exp\left[-\frac{(F-F_0)^2}{F_0^2\bar{\sigma}^2}\right], \tag{18}$$

and for amplitude-squeezed light $(\psi=0)$

$$f_{\psi=0}(F)=\frac{2}{\sqrt{\pi}}\frac{1}{\bar{\sigma}}\frac{F}{F_0}\,\theta(F-F_0)\frac{1}{(F-F_0)^{1/2}}\times\exp\left[-\frac{F^2-F_0^2}{\bar{F}_0^2\bar{\sigma}^2}\right]. \tag{19}$$

For the squeezed vacuum and a low-intensity ω-field, $\rho_F \ll 1$, and we easily obtain from (15)

$$W_{21}^{(\mathrm{sq})}(\Omega)=(2n_0-1)!!W_{21}^{(\mathrm{cl})}(\Omega). \tag{20}$$

In this equation,

$$n_0=\left[\frac{\Delta_{21}-\hbar\Omega}{\hbar\omega}\right]. \tag{21}$$

($[A]$ here is the integer part of A). In general ($|\beta|^2 \neq 0$, arbitrary phase ψ), the integral in Eq. (15) must be calculated numerically.

3. SCATTERING OF LIGHT BY A GENERALIZED TWO-LEVEL SYSTEM IN A SQUEEZED RADIATION FIELD

The scattering of probe light at frequency Ω by a two-level atom or molecule in a squeezed electromagnetic field of frequency ω is described by

$$W^{(\mathrm{sq})}(\Omega,\Omega')=\lim_{t\to\infty}W_0 2\ \mathrm{Re}\int_0^\infty d\mu e^{i(\Omega'-\Omega)\mu}\times\int_0^\infty d\tau\int_0^\infty d\tau'\exp\left[-\frac{\Gamma}{2}(\tau+\tau')\right]\times\exp\left[i\left(\frac{\Delta_{21}}{\hbar}-\Omega\right)(\tau-\tau')\right]I_t(\mu,\tau,\tau'). \tag{22}$$

Here $W_0=(\bar{n}_0/\hbar^4)|M^{\kappa_0\lambda_0}M^{\kappa\lambda}|^2$, where $\bar{n}_0$ is the number of photons in the incident beam, κ and λ are the photon's wave vector and polarization, $M^{\kappa\lambda}$ is the dipole transition matrix element, Γ is a phenomenological damping constant for the excited electron state (2), and $I_t(\mu,\tau,\tau')$ is the generating function for the scattering process:

$$I_t(\mu,\tau,\tau')=\langle G(t-\tau,t)G(t-\mu,t-\mu-\tau')\rangle_{\mathrm{sq}}. \tag{23}$$

The operator G was defined by Eq. (11) of Sec. 1. The calculation of the generating function (23) proceeds in the same way as for (9). Lengthy but straightforward mathematics for the generating function $I(\mu,\tau,\tau')=\lim_{t\to\infty}I_t(\mu,\tau,\tau')$ yields an expression analogous to (14), in which the function $\lambda(\tau)$ must be replaced by $\lambda(\mu,\tau,\tau')$:

$$\lambda(\mu,\tau,\tau')=e^{i\omega\tau}-1+e^{i\omega\mu}-e^{i\omega(\mu+\tau')}. \tag{24}$$

We can thus rewrite (22) in the form

$$W^{(\mathrm{sq})}(\Omega,\Omega')=\int dF\ f_\psi(F)W^{(\mathrm{cl})}(F;\Omega,\Omega'). \tag{25}$$

Here $f_\psi(F)$ is the function defined by Eq. (17), and $W^{(\mathrm{cl})}\times(F;\Omega,\Omega')$ is the scattering probability for light of frequency Ω with the participation of photons from the classical ω-field with fixed field strength F:

$$W^{(\mathrm{cl})}(\Omega,\Omega';F)=2\pi W_0\sum_{s=-\infty}^{+\infty}\delta(\Omega'-\Omega-s\omega)\times\left|\sum_{m=-\infty}^{+\infty}J_{s+m}(\rho_F)J_m(\rho_F)\times\frac{1}{\tilde{\Delta}/\hbar-\Omega'-m\omega+i\Gamma/2}\right|^2. \tag{26}$$

For Rayleigh scattering ($\Omega=\Omega'$) in a squeezed vacuum ($|\beta|^2=0$) with low ω-field intensities ($\rho_F\ll 1$), Eq. (25) yields

$$W^{(\mathrm{sq})}(\Omega,\Omega')=(4n_0-1)!!W^{(\mathrm{cl})}(\Omega,\Omega). \tag{27}$$

We thus see that for Rayleigh scattering at the second harmonic of the scattered light, for example, the cross section including photons of the squeezed ω-field is $(4.2-1)!!=105$ times the scattering cross section for coherent (classical) photons of the ω-field! The integration in (25) can be carried out numerically for squeezed fields of arbitrary intensity (see below).

4. ABOVE-THRESHOLD IONIZATION IN A TWO-FREQUENCY (Ω,ω) ELECTROMAGNETIC FIELD WITH SQUEEZED ω RADIATION

Above-threshold ionization (ATI) was discovered experimentally by Agostini *et al.*[11] during an investigation of multiphoton atomic ionization. A vast literature has since grown up on the subject (see Ref. 12, for example). We limit our present analysis to above-threshold ionization in a two-frequency (Ω,ω) field with an electron bound by a short-range potential (negative ion).

The effect in a two-frequency classical field can be derived directly from the equations for above-threshold ionization in a single-frequency electromagnetic field obtained in the Keldysh model.[13,14] To do so, it is necessary to replace the ionization potential Δ by $\Delta-\hbar\Omega$ in the equations of Refs. 13 and 14. The corresponding probabilities of s-photon ionization take the form

$$W_{\mathrm{ion}}^{(\mathrm{cl})}(s)=\frac{V_{21}^2(s)}{\hbar^2\Omega}\left|\sum_{p=-\infty}^{+\infty}J_{s-2p}(\rho_s)J_p(\rho_F)\right|^2. \tag{28}$$

Here $V_{21}(s)$ is the dipole matrix element for interactions with the Ω radiation, which is a weak function of s, as in the Keldysh theory:

$$\rho_s=eF\left(\frac{s-s^*}{\hbar m\omega^3}\right)^{1/2} \tag{29}$$

(m is the mass of the electron). Here s labels the sth above-threshold peak in the photoelectron momentum distribution, reckoned from the ionization potential Δ with allowance for the Stark shift; s^* is given by

$$\hbar\omega s^*=\Delta-\hbar\Omega. \tag{30}$$

For $\rho_F\ll 1$ and $\rho_s\gtrsim 1$, Eq. (28) becomes

$$W_{\mathrm{ion}}^{(\mathrm{cl})}(s)\simeq\frac{V_{21}^2(s)}{\hbar^2\Omega}J_s^2(\rho_s). \tag{31}$$

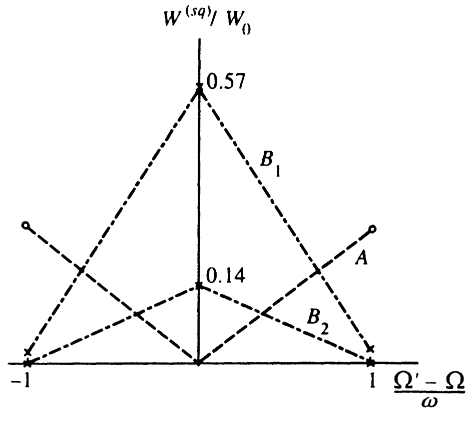

FIG. 1. Resonant scattering of light. A) Classical electromagnetic field; B_1) phase squeezing of light; B_2) amplitude squeezing of light ($\rho_F = 2$).

Now consider the ionization of an atom to which an electron is bound by a short-range potential in a two-frequency (Ω,ω) electromagnetic field, where the low-frequency ω radiation is nonclassical (squeezed). The calculation is largely the same as in Sec. 1. The probability of above-threshold ionization in the two-frequency field is, with squeezed ω radiation,

$$W_{\rm ion}^{(\rm sq)}(s) = \int dF\, f_\psi(F) W_{\rm ion}^{(\rm cl)}(F;s). \tag{32}$$

Here $f_\psi(F)$, as before, is the function defined in Eq. (17), and $W_{ion}^{(\rm cl)}(s)$ is the probability of above-threshold ionization in a classical field with fixed field strength F. Note also that if $\Omega = \omega$ and we let $s \rightarrow s+1$, then Eq. (32) is a generalization of the Keldysh formula to the case in which a bound electron interacts with a squeezed electromagnetic field.

5. DISCUSSION

The preceding examination of multiphoton and photoelectron spectroscopy of quantum systems demonstrates that in a two-frequency (Ω,ω) electromagnetic field, the photonic satellites of the ω-field depend heavily on the parameters of the squeezed ω radiation (squeezing parameter r, relative phase ψ), which is nonclassical by nature. In Figs. 1 and 2, we display the results of numerical calculations of the probability of optical multiphoton processes in a two-frequency (Ω,ω) field; for these calculations, we utilized Eqs. (15), (25), and (32) with various values of the intensity and phase of the ω-squeezed field. The figures clearly show that as the

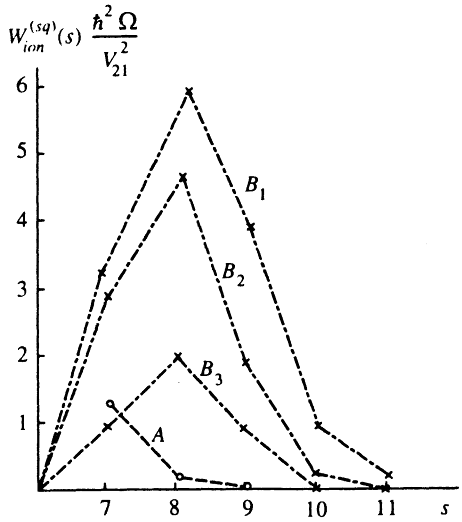

FIG. 2. Probability of an s-photon transition $W_{\rm ion}^{(\rm sq)}\hbar^2\Omega/V_{21}^2$. A) Classical electromagnetic field; $B_{1,2,3}$) squeezed electromagnetic field with various parameter values $\xi = N_0/\sinh^2 r$: $\xi_{B_1}=1$, $\xi_{B_2}=0.7$, $\xi_{B_3}=0.3$; ($A\times10^{-17}$, $B_1\times10^{-11}$, $B_2\times10^{-12}$, $B_3\times10^{-12}$).

intensity of the applied ω-field rises, the difference in magnitude of the scattering probability for the different squeezing methods (amplitude, phase) increases abruptly, particularly for amplitude-squeezed light.

Thus, as the intensity of the ω-field increases, its quantum properties become unmistakable.

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