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Atomic-level shifts in a squeezed vacuum

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The energy-level shifts for a multilevel atom interacting with a squeezed vacuum are calculated. The level shifts are made up of two contributions: (i) the ordinary Lamb shift and (ii) a shift due to the squeezed-vacuum intensity spectrum (similar to the blackbody-radiation shifts).

I. INTRODUCTION

The interaction of a bound electron with the vacuum state of the field leads to a shift in the atomic energy levels, the Lamb shift. A similar calculation determines the level shifts that result when the atom interacts with a blackbody radiation field, i.e., a nonzero temperature "vacuum".[1,2] In both these cases the radiation has time stationary field statistics, equivalently that the modes of the field are uncorrelated. Recently there has been considerable interest in states of the field which do not have this property, the squeezed states.[3] Squeezed states are characterized not by time stationary field statistics but rather by time stationary quadrature phase statistics.[4,5] This implies that for such fields there exist mode-mode correlations. It thus becomes of interest to inquire how the atomic-level shifts are modified in the presence of a squeezed field. Squeezed states have recently been observed in four-wave mixing[6] in confirmation of the predictions of Reid and Walls.[7]

In this paper we calculate the atomic-level shifts due to the interaction of an atom with a multimode squeezed state of zero mean amplitude, i.e., a squeezed vacuum. The problem has some similarities with blackbody radiation induced level shifts. There is an important difference however. The bulk of the blackbody spectrum is usually very far from resonance. In the case of squeezed light the situation is different. Squeezed states may be generated by parametric optical processes coupling a strong field at frequency Ω, the pump frequency, to two side bands at $\Omega\pm\epsilon$. The noise spectrum of the light at the output of such a device is concentrated near the frequency of the pump and this frequency may be close to the atomic transitions of interest.

In the first part of this paper we present a simple model for a squeezed vacuum. The statistics of the field is constructed in such a way as to model the output of an ideal parametric amplifier. We then calculate the level shifts due to the interaction of such a field with a multilevel atomic system, using the method of Louisell.[8] In this treatment the field modes are treated as a quantum reservoir to which the atomic system is coupled. We work in the dipole approximation but do not make the rotating-wave approximation.

II. SQUEEZED-VACUUM STATES

The prototypical squeezed state generator is the parametric amplifier. In Ref. 9 the output statistics for such a device are calculated. We can model these results as follows. Let us write the electric field in terms of its quadrature amplitudes at frequency Ω (the carrier frequency),

$$E(t)=\left[\frac{\hbar\Omega^3}{16\pi^3\epsilon_0c^3}\right]^{1/2}[X_1(t)\cos(\Omega t)+X_2(t)\sin(\Omega t)] \; . \tag{1}$$

In terms of the field amplitude operators $a(t)$ and $a^\dagger(t)$ (the positive and negative frequency components, respectively),

$$X_1(t)=\tfrac{1}{2}[a(t)e^{i\Omega t}+a^\dagger(t)e^{-i\Omega t}] \; , \tag{2}$$

$$X_2(t)=(1/2i)[a(t)e^{i\Omega t}-a^\dagger(t)e^{-i\Omega t}] \; . \tag{3}$$

The field amplitude operators may be written as

$$a(t)=\int_0^\infty d\omega\, D(\omega)a(\omega)e^{-i\omega t} \; , \tag{4}$$

where $D(\omega)$ is a density of states factor. We now define the ideal squeezed state by

$$|0^{(s)}\rangle=U|0\rangle \; , \tag{5}$$

where $|0\rangle$ is the usual field vacuum state and U is a unitary operator defined by

$$U^\dagger a(\omega)U=\mu(\omega)a(\omega)+\nu(\omega)a^\dagger(2\Omega-\omega) \; , \tag{6}$$

$$U^\dagger a^\dagger(\omega)U=\mu^*(\omega)a^\dagger(\omega)+\nu^*(\omega)a(2\Omega-\omega) \; , \tag{7}$$

where

$$|\mu(\omega)|^2-|\nu(\omega)|^2=1 \; . \tag{8}$$

This unitary transformation models the coupling of a field mode of frequency ω to its image sideband at frequency $2\Omega-\omega$ with respect to the carrier frequency Ω.

Using Eqs. (6) and (7) one easily establishes the following squeezed-vacuum correlation functions:

$$\langle a(\omega)a(\omega')\rangle=\mu(\omega)\nu(\omega')\delta(2\Omega-\omega-\omega') , \quad (9)$$

$$\langle a(\omega)a^\dagger(\omega')\rangle=\mu(\omega)\mu^*(\omega')\delta(\omega-\omega') , \quad (10)$$

$$\langle a^\dagger(\omega)a(\omega')\rangle=\nu(\omega)\nu^*(\omega')\delta(\omega-\omega') . \quad (11)$$

$$\langle a^\dagger(\omega)a^\dagger(\omega')\rangle=\nu^*(\omega)\mu^*(\omega')\delta(2\Omega-\omega-\omega') . \quad (12)$$

One may regard these relations as an equivalent definition of a squeezed vacuum. For the case of the usual vacuum, $\nu(\omega)=0$ and $\mu(\omega)=1$.

Using Eqs. (9)–(12) and assuming Ω is much greater than the bandwidth of the squeezing [that is the bandwidth over which $\mu(\omega)$ and $\nu(\omega)$ are significantly different from their vacuum levels] one shows that the quadrature phase variances are time stationary and given by

$$\langle X_1(t)X_1(t')\rangle =\frac{1}{4}\int_{-\infty}^{\infty} d\epsilon[2N(\epsilon)+M(\epsilon)+M^*(\epsilon)+1]e^{-i\epsilon\tau} , \quad (13)$$

$$\langle X_2(t)X_2(t')\rangle =\frac{1}{4}\int_{-\infty}^{\infty} d\epsilon[2N(\epsilon)-M(\epsilon)-M^*(\epsilon)+1]e^{-i\epsilon\tau} , \quad (14)$$

where

$$N(\epsilon)=|\nu(\Omega+\epsilon)|^2 , \quad (15)$$

$$M(\epsilon)=\mu(\Omega+\epsilon)\nu(\Omega-\epsilon) \quad (16)$$

with $\tau=t-t'$. The results are similar to those given in Ref. 9 for an ideal squeezed-vacuum state generated by parametric amplification. Using heterodyne detection we may probe the normally ordered quadrature phase spectra defined by

$$S_1^N(\epsilon)\equiv\frac{1}{4}[2N(\epsilon)+M(\epsilon)+M^*(\epsilon)] , \quad (17)$$

$$S_2^N(\epsilon)\equiv\frac{1}{4}[2N(\epsilon)-M(\epsilon)-M^*(\epsilon)] . \quad (18)$$

Squeezing at frequency ϵ is said to occur if either of $S_1^N(\epsilon)$ or $S_2^N(\epsilon)$ drops below the vacuum level of zero.

It is important to note that a squeezed vacuum may have quite a high intensity. In fact one may show

$$\langle a^\dagger(t)a(t)\rangle=\frac{1}{\Omega^3}\int_0^\infty d\omega\,\omega^3|\nu(\omega)|^2 . \quad (19)$$

We may thus regard $|\nu(\omega)|^2$ as the intensity spectrum for a squeezed state.

III. ATOMIC-LEVEL SHIFTS

We now consider the interaction of a multilevel atom with the squeezed-vacuum state of the field. We follow the treatment of Louisell.[8] In the dipole approximation the field-atom interaction Hamiltonian is

$$H_I=\boldsymbol{\mu}\cdot\mathbf{E}=-i\sum_{\mathbf{k}}\left[\frac{\hbar\omega_{\mathbf{k}}}{2\epsilon_0 V}\right]^{1/2}(a_{\mathbf{k}}-a_{\mathbf{k}}^\dagger)\hat{\mathbf{e}}_{\mathbf{k}}\cdot\boldsymbol{\mu} , \quad (20)$$

where $\boldsymbol{\mu}$ is the atomic dipole operator, $\hat{\mathbf{e}}_{\mathbf{k}}$ is a unit polarization vector and the sum over k contains an implicit sum over polarizations. We now define the interaction picture field operators

$$f_{lm}(t)=-\frac{i}{\hbar}\sum_k\left[\frac{\hbar\omega_k}{2\epsilon_0 V}\right]^{1/2}(a_k e^{-i\omega_k t}-a_k^\dagger e^{i\omega_k t})d_{lm}(k) \quad (21)$$

where $d_{lm}(k)\equiv\hat{\mathbf{e}}_k\cdot\langle l|\boldsymbol{\mu}|m\rangle$ and $|l\rangle,|m\rangle$ are atomic states. The many modes of the field may be treated as a reservoir and with the Markov approximation may be eliminated from the atomic dynamics. (This should be a reasonable approximation when the bandwidths of the atomic transitions are small compared to the bandwidth of squeezing.) This results both in dissipation and level shifts. For our purposes we write down only the shift in the transition frequencies ω_{ij}. The modification of the decay constants in a squeezed vacuum has been considered by Gardiner.[10] As shown in Louisell

$$\Delta\omega_{ij}=-\frac{\mathrm{Im}}{t}\left[\sum_l\int_0^t dt'\int_0^{t'}dt''[e^{i\omega_{jl}\tau}\langle f_{jl}(t')f_{lj}(t'')\rangle_R+e^{i\omega_{li}\tau}\langle f_{il}(t')f_{li}(t'')\rangle_R]\right] , \quad (22)$$

where Im indicates the imaginary part, $\langle\ \rangle_F$ indicates an average over the set of the field, and $\tau=t'-t''$.

To evaluate the reservoir correlation functions we assume that all modes are in the vacuum state except those parallel to a particular direction. Those modes with wave vectors parallel to this direction are in a squeezed-vacuum state defined in Sec. II. (That the squeezed-vacuum state should have a preferred wave vector is ultimately due to phase-matching conditions in the squeezed light source.) One then finds

$$\langle f_{jl}(t')f_{lj}(t'')\rangle=\frac{|d_{jl}|^2}{4\hbar\epsilon_0\pi^2c^3}\int_0^\infty d\omega\,\omega^3e^{-i\omega\tau} +\frac{|d_{jl}|^2}{6\hbar\epsilon_0\pi^2c^3}\left[\int_0^\infty d\omega\,\omega^3|\nu(\omega)|^2(e^{-i\omega\tau}+e^{i\omega\tau}) -\int_0^\infty d\omega\,\omega^2[\omega(2\Omega-\omega)]^{1/2}[\mu(\omega)\nu(2\Omega-\omega)e^{-i\omega\tau-2i\Omega t''}+\mu^*(2\Omega-\omega)\nu^*(\omega)e^{i\omega\tau+2i\Omega t''}]\right] , \quad (23)$$

where $|d_{jl}|^2=|\langle j|\mu|l\rangle|^2$.

The first term in Eq. (23) leads to the usual Lamb shift. The final phase-dependent terms average to zero in the time integrals due to the rapidly oscillating term at the carrier frequency.

Once these correlations are evaluated $\Delta\omega_{ij}$ may be found using Eq. (22). Writing $\Delta\omega_{ij}=\Delta\omega_i-\Delta\omega_j$, we find $\Delta\omega_i=\Delta\omega_i^{(L)}+\Delta\omega_i^{(S)}$, where the first term represents the usual Lamb shift and is given by

$$\Delta\omega_i^{(L)}=\frac{\mathrm{P}}{4\pi^2\hbar\epsilon_0 c^3}\sum_l |d_{il}|^2\int_0^\infty d\omega\,\omega^3\left[\frac{1}{\omega_{il}-\omega}\right], \tag{24}$$

(P refers to the Cauchy principal value), while $\Delta\omega_i^{(S)}$ represents the level shift due to the squeezed vacuum and is given by

$$\Delta\omega_i^{(S)}=\frac{\mathrm{P}}{6\pi^2\hbar\epsilon_0 c^3}\sum_l |d_{il}|^2\int_0^\infty d\omega\,\omega^3|\nu(\omega)|^2 \times\left[\frac{1}{\omega_{il}-\omega}+\frac{1}{\omega_{il}+\omega}\right]. \tag{25}$$

This frequency shift is similar to that causing blackbody radiation induced level shifts. This is to be expected in view of the role of $|\nu(\omega)|^2$ in determining the intensity spectrum for a squeezed vacuum. Changing the variable of intergration to $\epsilon=\omega-\Omega$ we have

$$\Delta\omega_i^{(S)}=\frac{\Omega^3}{6\pi^2\epsilon_0\hbar c^3}\mathrm{P}\sum_l|\tilde{d}_{il}|^2\int_{-\Omega}^\infty d\epsilon\left[\frac{N(\epsilon)}{\omega_{il}-\Omega-\epsilon}+\frac{N(\epsilon)}{\omega_{il}+\Omega+\epsilon}\right]. \tag{26}$$

We now consider the special case of a two-level atom.

IV. TWO-LEVEL ATOM

In the case of a two-level atom we may write

$$\Delta\omega^{(S)}=\frac{\gamma}{2\pi}\left[\frac{\Omega}{\omega_A}\right]^3 I(\Delta)\,, \tag{27}$$

where

$$\gamma=\frac{\omega_A^3|\mu_{12}|^2}{3\pi\hbar\epsilon_0 c^3} \tag{28}$$

is the spontaneous emmission rate and

$$I(\Delta)=\mathrm{P}\int_{-\Omega}^\infty d\epsilon\left[\frac{N(\epsilon)}{\Delta-\epsilon}+\frac{N(\epsilon)}{\Delta+2\Omega+\epsilon}\right] \tag{29}$$

with $\Delta=\omega_A-\Omega$.

To evaluate this integral we need to assume some form for the squeezing spectrum. We will take as a typical example the squeezing spectrum at the output of an ideal parametric amplifier:[8]

$$S_1^{(N)}(\epsilon)=\left[\frac{f}{4}\right]\frac{1}{(\epsilon/\gamma_T)^2+\mu^2}\,, \tag{30}$$

$$S_2^{(N)}(\epsilon)=-\left[\frac{f}{4}\right]\frac{1}{(\epsilon/\gamma_T)^2+1}\,. \tag{31}$$

In this case the carrier frequency Ω is equal to the cavity resonant frequency of the paramp. The parameter f is a measure of the squeezing, and is given by

$$f=\frac{\gamma_{\text{out}}}{\gamma_T}\,, \tag{32}$$

where γ_{out} is the loss constant for the output port of the device and γ_T is the total loss constant for the cavity. Clearly $0\le f\le 1$. Perfect squeezing at $\epsilon=0$ is obtained when $f=1$. The parameter μ is a small constant. When $f=1$ we expect $\mu=0$, which ensures $S_1^N(0)\to\infty$ when $S_2^N(0)\to-\frac{1}{4}$. The width of the squeezing spectrum is γ_T, that is, the same as the width of the paramp cavity resonance. Using Eqs. (17) and (18) we see that the form of the squeezing spectrum assumed here requires $M(\epsilon)=M(-\epsilon)$, that is, the squeezing spectrum is symmetric about the carrier frequency. In this case

$$I(\Delta)=\mathrm{P}\int_{-\Omega}^\infty d\epsilon[S_1^N(\epsilon)+S_2^N(\epsilon)]\left[\frac{1}{\Delta-\epsilon}+\frac{1}{\Delta+2\Omega+\epsilon}\right].$$

The assumption of small squeezing bandwidth with respect to the carrier frequency is equivalent to $\Omega/\gamma_T\gg 1$. With this assumption we find that the nonresonant second term in the integrands of Eqs. (34) and (35) makes a negligible contribution. Evaluating the integrals we find

$$\Delta\omega^{(S)}=\frac{\gamma f\delta}{8}\left[\frac{1}{\mu(\mu^2+\delta^2)}-\frac{1}{1+\delta^2}\right],$$

where $\delta=\Delta/\gamma_T$.

It is interesting that when the carrier frequency is resonant with the atomic transition no frequency shift is observed. Away from this resonance point we see the two quadratures shift the level in opposite directions. As the squeezing becomes large the fluctuations in the unsqueezed quadrature dominate and significant level shifts may result. However, it should be noted that we have implicitly assumed the intensity of the squeezed light is not too large in order that the perturbation used here to be valid. This means the analysis is restricted to the small squeezing regime.

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