Squeezed States

Energy shifts for a multilevel atom in an intense squeezed radiation field

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We derive atomic energy-level shifts due to the presence of an electric field associated with an arbitrary quantum-mechanical state describing an intense, but otherwise arbitrary, radiation field, a particular example of such a state being squeezed radiation from an intense source. In another particular case, in which the radiation is classical and monochromatic, our result reduces to the well-known Townes–Schawlow result.

The Lamb shift[1] is the most well-known quantum-electrodynamic frequency shift; it is due to the interaction of an electron with the zero-point fluctuations of the electromagnetic field. In this case we are dealing with virtual photons. In the case of an external monochromatic classical electromagnetic field of frequency ω, the corresponding shift—the so-called ac Stark shift—has been derived by Townes and Schawlow.[2]

For an atom interacting with blackbody radiation, Ford *et al.*[3] used thermodynamic perturbation theory to calculate the corresponding shift—oftentimes referred to as the finite-temperature Lamb shift. A key feature of this work, as well as of previous exact work on the oscillator,[4] was the emphasis on thermodynamic concepts and the fact that the work done by external laser photons[5] corresponds to a change in free energy, as distinct from a change in energy.[6] The calculations of Ref. 3 are relatively complex because blackbody radiation is not a static source; instead, it interacts dynamically with the atom so that it can receive as well as give energy to the atom. In other words, the state of the blackbody radiation is affected by the presence of the atom. By contrast, radiation produced by an intense laser can be regarded as a source of great inertia that is unaffected by the presence of an atom. As a result, it is easier to calculate the atomic energy shifts in such a case.

In this paper, we consider the case of an atom interacting with an electric field that is due to an intense but otherwise *arbitrary* radiation field. Thus our treatment encompasses both pure and mixed states; it includes, as a *particular* case, the case of *squeezed* radiation[7,8] in which the associated electric field is a *pure* state.

In general, an arbitrary state is a multimode field. However, we start by asking a simpler question, viz., what is the result for the atomic energy shift of level a because of the presence of a monochromatic electric field of frequency ω? The answer is (see Appendix A for calculational details)

$$\delta\epsilon_a = \frac{2\pi e^2\omega}{V}\sum_b |\langle a|\mathbf{r}\cdot\hat{e}|b\rangle|^2\left[\frac{\langle n(\omega)\rangle}{\omega_{ab}+\omega}+\frac{\langle n(\omega)\rangle+1}{\omega_{ab}-\omega}+\frac{1}{\omega}\right]. \tag{1}$$

Here V denotes the total volume, $\omega_{ab} = \omega_a - \omega_b$, $\hat{e}$ refers to the electric-field unit polarization vector, and

$$\langle n(\omega)\rangle = \langle a^+a\rangle \tag{2}$$

is the average number of photons of frequency ω. In general, the average is taken with respect to an arbitrary (i.e., pure or mixed) quantum-mechanical state that is associated with the electric field in question.

The contribution corresponding to the case $\langle n\rangle = 0$ is that due to the zero-point fluctuations. In the classical limit, the latter is ignored and, in addition, the time average of the expectation value of the absolute square of the electric field is given by [see Appendix A, Eq. (A32)]

$$\overline{\langle|\mathbf{E}(t)|^2\rangle} \rightarrow (4\pi\hbar w/V)\langle n\rangle \equiv (E_0{}^2/2), \tag{3}$$

where the arrow refers to the classical limit and E_0 is defined as the amplitude of the classical field. It then follows that

$$\delta\epsilon_a \rightarrow \frac{e^2}{4\hbar}\sum_b |\langle a|\mathbf{E}_0\cdot\mathbf{r}|b\rangle|^2\left[\frac{1}{\omega_{ab}+\omega}+\frac{1}{\omega_{ab}-\omega}\right], \tag{4}$$

which is the well-known Townes–Schawlow result[2] for the energy shift of level a in a monochromatic classical electric field.

In the case of a multimode arbitrary radiation field, we obtain the corresponding energy shift by returning to our basic result [Eq. (1)], which we multiply by $V\rho(\omega)d\omega$ and then integrate over all frequencies, where

$$\rho(\omega) = \omega^2/\pi^2c^3 \tag{5}$$

is the density of states of the squeezed radiation field.

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Hence it follows that

$$\delta\epsilon_a = \delta\epsilon_a^{(0)} + \delta\epsilon_a^{(A)}, \tag{6}$$

where

$$\delta\epsilon_a^{(A)} = \frac{2\pi e^2}{3}\sum_b |\langle a|\mathbf{r}\cdot\hat{e}|b\rangle|^2 \times \int_0^\infty \omega\rho(\omega)\langle n(\omega)\rangle\left(\frac{1}{\omega_{ab}+\omega}+\frac{1}{\omega_{ab}-\omega}\right)\mathrm{d}\omega$$

$$= \frac{2e^2}{3\pi c^3}\sum_b |\langle a|\mathbf{r}\cdot\hat{e}|b\rangle|^2 \times \int_0^\infty \omega^3\langle n(\omega)\rangle\left(\frac{1}{\omega_{ab}+\omega}+\frac{1}{\omega_{ab}-\omega}\right)\mathrm{d}\omega \tag{7}$$

and

$$\delta\epsilon_a^{(0)} = \frac{2e^2}{3\pi c^3}\sum_b |\langle a|\mathbf{r}\cdot\hat{\mathbf{e}}|b\rangle|^2 \int_0^\infty \frac{\omega^2\omega_{ab}}{\omega_{ab}-\omega}\mathrm{d}\omega. \tag{8}$$

This is the desired result for the energy shift of level a that is due to a multimode arbitrary radiation field, where $\langle n(\omega)\rangle$ denotes the average number of photons of frequency ω in the arbitrary quantum state. The superscript A indicates the arbitrary nature of the intense field, and the superscript 0 denotes the contribution due to the zero-point fluctuations of this field. However, we emphasize that this formula is not applicable to the case of blackbody radiation; if one replaces $\langle n\rangle$ in Eq. (7) by the familiar expression $[\exp(\hbar\omega/kT) - 1]^{-1}$, where T is the temperature, then the result obtained is not the energy shift but the free-energy shift [see Eq. (42) of Ref. 3 and perform some algebra[9]].

Milburn[10] calculated atomic-level shifts in the particular case of a squeezed vacuum (based on techniques developed by Louisell[11]), and we can compare his Eqs. (24) and (25) with our Eqs. (6) to (8) above, if in the latter equations we take the average of $n(\omega)$ with respect to the particular case of a (pure) squeezed-vacuum state. We find agreement for the $\delta\epsilon_a^{(A)}$ part of $\delta\epsilon_a$ but not for the $\delta\epsilon_a^{(0)}$ part. Milburn's expression corresponding to $\delta\epsilon_a^{(0)}$ is too large by a factor of 3/2, and, in addition, he obtained the integrand $\omega^3/(\omega_{ab} - \omega)$ instead of the correct value of $\omega^2\omega_{ab}/(\omega_{ab} - \omega)$ appearing in Eq. (8). Apart from the factor 3/2, the reason for this discrepancy is that in our opinion Milburn (incorrectly) did not include the H_2 term appearing in Eq. (A32) of Appendix A, which would have given him an additional ω^2 term in the integrand. Our result agrees exactly with the corresponding result that we obtained for the case of blackbody radiation in Ref. 3 [see Eq. (23) and use Eq. (50)], the latter being obtained by use of the $\mathbf{A}\cdot\mathbf{p}$ gauge. Thus, as a by-product of our calculation, we have in effect emphasized a point often forgotten by many investigators, viz., that the $-\boldsymbol{\mu}\cdot\mathbf{E}$ term alone is not equivalent to the use of the $(2e/c)\mathbf{p}\cdot\mathbf{A} + (e^2/c^2)A^2$ interaction terms but must be supplemented by the $|\hat{e}\cdot\boldsymbol{\mu}|^2$ term given in Eq. (A32). Perhaps it should also be emphasized that, in contrast to the contribution from the A^2 term, the contribution from the $|\hat{e}\cdot\boldsymbol{\mu}|^2$ term is not the same for all levels, and, for that reason, its inclusion is an essential element in the calculation.

Furthermore, Milburn's reference to $\delta\epsilon_a^{(0)}$ as the Lamb shift is not strictly correct; this term arises from the zero-point fluctuations of the electromagnetic field but the actual (observable) Lamb shift also incorporates subtractions common to all levels as well as renormalization contributions. This can be seen by writing our integrand as follows:

$$\frac{\omega^2\omega_{ab}}{\omega_{ab}-\omega} = \frac{1}{\omega_{ab}-\omega}\left[\omega_{ab}^3 + \omega_{ab}\omega(\omega_{ab}-\omega) - \omega_{ab}^2(\omega_{ab}-\omega)\right], \tag{9}$$

where, on the right-hand side, the first term corresponds to what Sakurai[1] refers to as the *observed* Lamb shift, the second term is due to the A^2 contribution, which is the same for all levels [see Ref. 3, especially Eq. (22)], and the third term arises from mass renormalization.[1]

In the case of a general squeezed state we have[8]

$$\langle \mathrm{n}\rangle = |\alpha|^2 + \sinh^2|z|, \tag{10}$$

where α is the eigenvalue of a for the coherent state $|\alpha|$ and $|z|$ is the modulus of the squeeze parameter. The Townes–Schawlow formula corresponds to the coherent state with $|z| = 0$, whereas the ideal squeezed vacuum considered by Milburn corresponds to $\alpha = 0$. The general case is given by the above formula, in conjunction with Eq. (7).

APPENDIX A: DERIVATION OF EQ. (1)

Consider an atom interacting with an electric field that is unaffected by the presence of the atom. Apart from the latter restriction (which implies that we are dealing with an intense source), the electric field may be associated with an *arbitrary* quantum-mechanical state of the *radiation field* (i.e., our treatment encompasses both pure and mixed states). In *particular*, an intense *squeezed* radiation is such a field; in fact, the associated electric field is a *pure* state. Schrödinger's equation for this arbitrary system is, to first order in μ,

$$i\hbar\frac{\partial\psi}{\partial t} = [H_0 - \boldsymbol{\mu}\cdot\mathbf{E}(t)]\psi \equiv [H_0 + H_1(t)]\psi, \tag{A1}$$

where H_0 is the noninteraction part of the total Hamiltonian, $\boldsymbol{\mu} = e\mathbf{r}$ is the dipole moment, and $\mathbf{E}(t)$ is the time-dependent electric-field operator, which for a single mode has the form

$$\mathbf{E}(t) = (2\pi\hbar\omega/V)^{1/2} i(ae^{-i\omega t} - a^+e^{i\omega t})\hat{e}, \tag{A2}$$

where ω is the frequency, V is the volume, and $\hat{e}$ is the unit polarization vector.

To find the perturbed energies, we go to the interaction picture, writing

$$\psi = e^{-iH_0/\hbar}\Phi. \tag{A3}$$

Then

$$i\hbar\frac{\partial\Phi}{\partial t} = -\boldsymbol{\mu}(t)\cdot\mathbf{E}(t)\Phi, \tag{A4}$$

where

$$\boldsymbol{\mu}(t) = \exp(iH_0t/\hbar)\boldsymbol{\mu}\exp(-iH_0t/\hbar). \tag{A5}$$

Now Φ will have fluctuations of two kinds: the first from fluctuations in the operator $\mathbf{E}$, the second from fluctuations in the time dependences of $\boldsymbol{\mu}(t)$ and $\mathbf{E}(t)$. To examine the first case of fluctuations in **E**, we write

$$\Phi = [1 + F^{(1)}(t) + F^{(2)}(t) + \ldots]\langle\Phi\rangle, \tag{A6}$$

where Φ is the mean and satisfies the fluctuation-free equation

$$i\hbar \frac{\partial\langle\Phi\rangle}{\partial t} = [\Omega^{(1)}(t) + \Omega^{(2)}(t) + \ldots]\langle\Phi\rangle, \tag{A7}$$

and the quantities F and Ω remain to be determined. Then we find, to first order,

$$\Omega^{(1)}(t) + i\hbar \frac{\partial F^{(1)}}{\partial t} = -\mu(t)\cdot\mathbf{E}(t), \tag{A8}$$

so that

$$\Omega^{(1)}(t) = -\mu(t)\cdot\langle\mathbf{E}(t)\rangle \tag{A9}$$

and

$$F^{(1)}(t) = -\frac{1}{i\hbar}\int_{-\infty}^{t} \mathrm{d}t_1 \mu(t_1)\cdot[\mathbf{E}(t_1) - \langle\mathbf{E}(t_1)\rangle]. \tag{A10}$$

In second order, we obtain

$$\Omega^{(2)}(t) + i\hbar\frac{\partial F^{(2)}}{\partial t} = -\,\mu(t)\cdot\mathbf{E}(t)F^{(1)}(t) - F^{(1)}(t)\Omega^{(1)}(t) \tag{A11}$$

so that

$$\begin{aligned}\Omega^{(2)}(t) &= -\langle\mu(t)\cdot\mathbf{E}(t)F^{(1)}(t)\rangle \\ &= \frac{1}{i\hbar}\int_{-\infty}^{t}\mathrm{d}t_1\mu_j(t)\mu_k(t_1)[\langle E_j(t)E_k(t_1)\rangle \\ &\quad - \langle E_j(t)\rangle\langle E_k(t_1)\rangle].\end{aligned} \tag{A12}$$

Next we turn to the time fluctuation. In the above equation for $\langle\Phi\rangle$, i.e., Eq. (A7), we put

$$\langle\Phi\rangle = [1 + G^{(1)}(t) + G^{(2)}(t) + \ldots]\overline{\langle\Phi\rangle}, \tag{A13}$$

where $\overline{\langle\Phi\rangle}$ is the time average of the mean and satisfies

$$i\hbar\frac{\partial\overline{\langle\Phi\rangle}}{\partial t} = [B^{(1)} + B^{(2)} + \ldots]\overline{\langle\Phi\rangle}, \tag{A14}$$

where the quantities B are time-independent operators. The bar denotes the time average, which can be written for an arbitrary function $f(t)$ as

$$\bar{f} = \lim_{\epsilon\to 0^+} \epsilon\int_0^{\infty}\mathrm{d}t e^{-\epsilon t}f(t). \tag{A15}$$

To first order, we obtain

$$B^{(1)} + i\hbar\frac{\partial G^{(1)}}{\partial t} = \Omega^{(1)}(t), \tag{A16}$$

so that

$$B^{(1)} = \overline{\Omega^{(1)}} \tag{A17}$$

and

$$G^{(1)}(t) = \frac{1}{i\hbar}\int_{-\infty}^{t}\mathrm{d}t_1[\Omega^{(1)}(t) - \bar{\Omega}^{(1)}]. \tag{A18}$$

In second order, we obtain

$$B^{(2)} + i\hbar\frac{\partial G^{(2)}}{\partial t} = \Omega^{(2)}(t) + \Omega^{(1)}(t)G^{(1)}(t) - G^{(1)}(t)B^{(1)}, \tag{A19}$$

so that

$$B^{(2)} = \overline{\Omega^{(2)}} + \overline{\Omega^{(1)}G^{(1)}}. \tag{A20}$$

Now in Eq. (A14) for $\overline{\langle\Phi\rangle}$ we expand

$$\overline{\langle\Phi\rangle} = \sum_a C_a\Phi_a, \tag{A21}$$

where Φ_a are the unperturbed eigenvectors of H_0:

$$H_0\Phi_a = \hbar\omega_a\Phi_a. \tag{A22}$$

Then, clearly the shift in the ath eigenvalue is

$$\delta\epsilon_a^{(1)} = [\Phi_a, B^{(1)}\Phi_a] + [\Phi_a, B^{(2)}\Phi_a] + \ldots, \tag{A23}$$

where the superscript 1 on the left-hand side indicates that the correction is due to the term in the Hamiltonian that is first order in μ. Putting in the above results, we get

$$\begin{aligned}\delta\epsilon_a^{(1)} = &-\overline{[\Phi_a, \mu(t)\Phi_a]\cdot\langle\mathbf{E}(t)\rangle} \\ &+ \frac{1}{i\hbar}\overline{\int_{-\infty}^{t}\mathrm{d}t_1\{\Phi_a, [\mu_j(t)\mu_k(t_1)\langle E_j(t)E_k(t_1)\rangle} \\ &- \mu_j(t)\langle E_j(t)\rangle\,\overline{\mu_k(t_1)\langle E_k(t_1)\rangle}]\Phi_a\}.\end{aligned} \tag{A24}$$

Now, if there is no static dipole moment, it follows that

$$[\Phi_a, \mu(t)\Phi_a] = (\Phi_a, \mu\Phi_a) = 0. \tag{A25}$$

Also, if $\omega \neq \omega_a - \omega_b$ for any a, b, then

$$\overline{\mu_k(t_1)\langle E_k(t_1)\rangle} = 0. \tag{A26}$$

Hence

$$\delta\epsilon_a^{(1)} = \frac{1}{i\hbar}\overline{\int_{-\infty}^{t}\mathrm{d}t_1[\Phi_a, \mu_j(t)\mu_k(t_1)\Phi_a]\langle E_j(t)E_k(t_1)\rangle}. \tag{A27}$$

If we write

$$\begin{aligned}[\Phi_a, \mu_j(t)\mu_k(t_1)\Phi_a] &= \sum_b[\Phi_a, \mu_j(t)\Phi_b][\Phi_b, \mu_k(t_1)\Phi_a] \\ &= \sum_b(\Phi_a, \mu_j\Phi_b)(\Phi_b, \mu_k\Phi_a) \\ &\quad\times\exp[i(\omega_a - \omega_b)(t - t_1)],\end{aligned} \tag{A28}$$

then

$$\begin{aligned}\delta\epsilon_a^{(1)} = &\frac{1}{i\hbar}\sum_b(\Phi_a, \mu_j\Phi_b)(\Phi_b, \mu_k\Phi_a) \\ &X\overline{\int_{-\infty}^{t}\mathrm{d}t_1\exp[i\omega_{ab}(t - t_1)]\langle E_j(t)E_k(t_1)\rangle},\end{aligned} \tag{A29}$$

where $\omega_{ab} \equiv \omega_a - \omega_b$.

From Eq. (A2) we obtain

$$\begin{aligned}\langle E_j(t)E_k(t_1)\rangle = &\hat{e}_j\hat{e}_k(2\pi\hbar\omega/V)\{-\langle a^2\rangle\exp[-i\omega(t + t_1)] \\ &- \langle a^{+^2}\rangle\exp[i\omega(t + t_1)] + \langle aa^+\rangle \\ &\times\exp[-i\omega(t - t_1)] + \langle a^+a\rangle\exp[i\omega(t - t_1)].\end{aligned} \tag{A30}$$

Substituting Eq. (A30) into Eq. (A29), the $\langle a^2\rangle$ and $\langle a^{+2}\rangle$ terms do not survive the time average, and we get

$$\delta\epsilon_a^{(1)} = (2\pi\omega/V)\sum_b |(\Phi_a, \hat{e}\cdot\mu\Phi_b)|^2 \times\left[\frac{\langle a^+a\rangle}{\omega_{ab}+\omega+i\epsilon}+\frac{\langle aa^+\rangle}{\omega_{ab}-\omega+i\epsilon}\right]. \quad (A31)$$

It should be noted that our result depends on the square of μ. Thus, for consistency, we should also consider the term of second order in μ in the Hamiltonian; this is the Power–Zienau term[12]

$$H_2 = \frac{2\pi}{V}|\hat{e}\cdot\mu|^2, \quad (A32)$$

and thus the corresponding energy shift is

$$\delta\epsilon_a^{(2)} = \frac{2\pi}{V}\sum_b |(\Phi_a, \hat{e}\cdot\mu\Phi_b)|^2. \quad (A33)$$

We add Eqs. (A31) and (A33) to get the total energy shift; the imaginary part is the width, and the real part is the principal value. Thus, by setting $\epsilon = 0$ in Eq. (A31), we obtain the total energy shift

$$\delta\epsilon_a = (2\pi\omega/V)\sum_b |(\Phi_a, \hat{e}\cdot\mu\Phi_b)|^2 \times\left(\frac{\langle a^+a\rangle}{\omega_{ab}+\omega}+\frac{\langle aa^+\rangle}{\omega_{ab}-\omega}+\frac{1}{\omega}\right), \quad (A34)$$

which is the same as Eq. (1). This is the key result of our paper. It is clear that it applies to an electric field associated with an arbitrary (i.e., pure or mixed) state of an intense, but otherwise arbitrary, monochromatic radiation field. Generalization to the multimode case is trivial, as we have shown above.

From Eq. (A30), it also follows that

$$\overline{\langle E_j(t)E_k(t)\rangle} = \hat{e}_j\hat{e}_k(2\pi\hbar\omega/V)(\langle aa^+\rangle + \langle a^+a\rangle) = \hat{e}_j\hat{e}_k(4\pi\hbar\omega/V)[\langle n(\omega)\rangle + 1/2], \quad (A35)$$

where $n \equiv a^+a$ and we have used the fact that $[a, a^+] = 1$. The classical limit of this equation appears as Eq. (3).

Finally, we emphasize again that we have assumed that the state of the radiation is unaffected by the presence of the atom (in contrast to the situation considered by Ford *et al.*[3] in their treatment of blackbody radiation). This implies that we are dealing with an intense source of radiation—yet not too intense, to ensure that the perturbation treatment that we have employed is valid.

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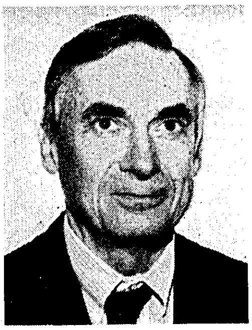

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