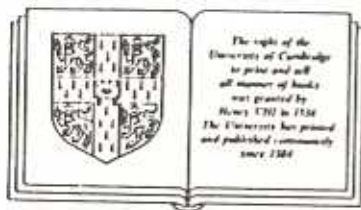


# Coherence, cooperation and fluctuations

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DISSIPATION AND MEMORY EFFECTS IN THE INTERACTION OF A  
BLACKBODY RADIATION HEAT BATH WITH MATTER

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Abstract. There exists a well-known perturbation result for the dynamic Stark shift, induced by blackbody radiation, of atomic energy levels. A similar derivation, for the simpler case of a quantum oscillator in a blackbody radiation field, leads to a result which, surprisingly, is at variance with the exact result, which was recently calculated by Ford, Lewis and O'Connell. The discrepancy is shown to result from the neglect of dissipation effects in the derivation of the perturbation result. The importance of memory effects is also emphasized.

INTRODUCTION

Considerable interest has been generated by the experiments of Gallagher and Cooke<sup>1</sup>, which indicate that blackbody radiation can cause significant reductions in radiative lifetimes. These authors also calculated the dynamic (a.c.) Stark shifts, induced by blackbody radiation, in the energy levels of high Rydberg states (Gallagher and Cooke<sup>1</sup>, Cooke and Gallagher<sup>2</sup>). The effect of blackbody radiation on the Lamb shift had been considered earlier by several authors (Auluck and Kothari<sup>3</sup>, Walsh<sup>4</sup>, Barton<sup>5</sup>, Knight<sup>6</sup>) and again more recently by others (Palanques-Mestre and Tarrach<sup>7</sup>, Cha and Yee<sup>8</sup>). Farley and Wing<sup>9</sup> made a more detailed calculation of the dynamic Stark shifts and obtained results which depend on the nature of the atomic species. In the high-temperature limit all these authors agree on the result for the energy shift:

$$(\pi e^2 / 3 \hbar m c^3) (k T)^2 . \quad (1)$$

Reports of measurements by Hollberg and Hall<sup>10</sup> of the blackbody shift in high Rydberg states of Rubidium appear to be consistent with this high-temperature formula.

In the more general case (i.e., no restriction to high Rydberg states), the Farley-Wing<sup>9</sup> derivation is based on the quantum-mechanical perturbation result for the energy shift of an electron in energy level  $n$  subject to an electric field (Townes and Schawlow<sup>11</sup>, Sobelman<sup>12</sup>):

$$\Delta E_n = \frac{e^2}{4\hbar} \sum_m \left\{ \frac{1}{\omega_{nm} - \omega} + \frac{1}{\omega_{nm} + \omega} \right\} |\vec{E}_0 \cdot \vec{r}_{nm}|^2 \quad (2)$$

where  $\vec{E}_0$  is the amplitude of the monochromatic (frequency  $\omega$ ) electric field,  $\vec{r}_{nm}$  is the matrix element of  $\vec{r}$  between the unperturbed energy states  $n$  and  $m$  and

$$\hbar \omega_{nm} = E_n - E_m. \quad (3)$$

The observed energy shift due to a laser beam (Lieberman *et al.*<sup>13</sup>) appears to be in reasonable - although not perfect - agreement with the result given by (2).

It should be emphasized that (2) neglects radiative damping and thus treats energy levels as arbitrarily sharp. However, as we will emphasize below, the damping is proportional to  $e^2$  (the coupling strength), from which it follows that the magnitude of  $\Delta E_n$  actually depends on the magnitude of the damping. Such an observation clearly demands a re-assessment of the assumptions underlying the derivation of (2) and, concomitantly, raises questions as to the validity of (2).

Turning now to the case of a blackbody radiation heat bath, Farley and Wing proceeded by identifying  $3 E_0^2/8\pi$  with  $u(\omega, T)$ , the energy density of the electromagnetic field. Substituting the Planck distribution

$$u(\omega, T) = (\hbar \omega^3/\pi^2 c^3)/(e^{\hbar\omega/kT} - 1), \quad (4)$$

they obtained the mean energy shift

$$\begin{aligned}
 U^{(FW)}(T) &= \int_0^{\infty} d\omega \Delta E_n(\omega) \\
 &= (2e^2/3\pi \hbar^3 c^3) (kT)^3 \sum_m \sum_i |\langle n | r_i | m \rangle|^2 F(y), \quad (5)
 \end{aligned}$$

where

$$F(y) = \int_0^{\infty} \left( \frac{1}{y-x} + \frac{1}{y+x} \right) \frac{x^3 dx}{e^x - 1} \quad (6)$$

$$y = (\hbar \omega_{nm} / kT), \quad x = (\hbar \omega / kT), \quad (7)$$

and the superscript "FW" denotes the Farley-Wing model. In the high-temperature (low-frequency) limit,  $|\hbar \omega_{nm}| \ll kT$ , it may be verified that (5) reduces to (1). In the more general case, Farley and Wing evaluated (5) for a variety of atoms.

We turn now to the simpler case where the electron is not in an atomic potential but, instead, is confined by a one-dimensional harmonic force  $Kx = m\omega_0^2 x$ . Then, in (2) and (5),  $r_i$  is replaced by  $x$  with the sum over  $i$  reducing to a single term and the states  $|n\rangle$  and  $|m\rangle$  now refer to oscillator states. In the latter case, it is well-known that the only non-zero matrix elements are those for which  $m = n \pm 1$ . Thus, we obtain

$$\Delta E_n^{(FW)} = \frac{e^2}{4m} \frac{E_0^2}{\omega^2 - \omega_0^2} \equiv \Delta E^{(FW)}(\omega), \quad (8a)$$

independent of  $n$ . However, as already noted in our discussion of (2), the derivation of (8) neglects radiative damping but yet the damping is actually proportional to the coupling constant  $e^2$ . In fact, anticipating results from the exact analysis given below, we use the expression (28) for the damping width  $\gamma$  (except that we use  $m$  instead of the renormalized mass  $M$  appearing in (28) and (29)) to write

$$\Delta E_n^{(FW)} = \frac{3c^3}{8} \gamma \frac{E_0^2}{\omega_0^2(\omega^2 - \omega_0^2)} \quad (8b)$$

Again, this result highlights the fact that whereas  $\gamma$  was taken to be zero in the derivation of (8), there is actually a one-to-one relationship between the result sought and the magnitude of  $\gamma$ .

As before, in proceeding from (2) to (5) - except that we take account of the fact that we are now dealing with a one-dimensional problem - we replace  $E_0^2/8\pi$  with  $u(\omega, T)$ . Then, using the fact that  $F(y) = -F(-y)$ , it follows that

$$\sum_m |\langle n | x | m \rangle|^2 F(y) = -\frac{\hbar}{2m\omega_0} F(y_0), \quad (9)$$

where  $y_0 = (\hbar\omega_0/kT)$ . Hence

$$\begin{aligned} U_0^{(FW)}(T) &= \int_0^\infty d\omega \Delta E^{(FW)}(\omega) \\ &= (2e^2/3\pi mc^3) \int_0^\infty d\omega \frac{\omega^2}{\omega^2 - \omega_0^2} \frac{\hbar\omega}{e^{\hbar\omega/kT} - 1}, \quad (10) \end{aligned}$$

where the subscript 0 denotes "oscillator". In the case where  $\hbar\omega_0 \ll kT$ , we find that  $U_0^{(FW)}(T)$  reduces to  $\frac{1}{3}$  of the result given by (1), the reduction by a factor of 3 reflecting the fact that we are treating a one-dimensional oscillator.

We now turn to a critique of (8) and (10). First of all, we note that they do not explicitly contain a damping term, reflecting the neglect of dissipation effects in the derivation of the perturbation result and made manifest by the fact that they display a pole at  $\omega = \omega_0$ .

Secondly, the perturbation result (10) does not contain what is in fact the leading and dominant term in the expression for the oscillator energy viz. the equipartition energy  $kT$ . Thus, we are motivated to introduce damping into our analysis of the problem. This was done in an exact manner by Ford *et al.*<sup>14</sup> but, before presenting an account of this work it is useful to consider, first of all, the usual manner in which damping is treated. This will serve to highlight the weakness of the latter method vis-à-vis the Ford *et al.* approach.

## DAMPED HARMONIC OSCILLATOR: PHENOMENOLOGICAL TREATMENT

The usual way in which damping is treated is phenomenologically, by writing down an equation of motion for a damped harmonic oscillator subject to a random force  $F(t)$ . It is actually easier to use the Fourier transform of this equation and, conforming to the notation of Ford, Lewis, and O'Connell<sup>14</sup>, who discuss an exact equation of motion, to be described below, we will use a tilde to denote a Fourier transform, e.g.  $\tilde{x}(\omega)$  and  $\tilde{F}(\omega)$  are the Fourier transforms of  $x(t)$  and  $F(t)$ , respectively. Thus

$$m \ddot{\tilde{x}} + m \gamma \dot{\tilde{x}} + m \omega_0^2 \tilde{x} = \tilde{F}(\omega) = -e E_0 e^{-i\omega t}, \quad (11)$$

where  $\gamma$  is a phenomenological damping constant (i.e. it is assumed to be independent of  $\omega$ ), and the random force is chosen to be the dipole interaction between the oscillator and the radiation field, with  $E_0$  representing the amplitude of the random electric field. The solution of this equation may be written in the form

$$\tilde{x}(\omega) = \alpha(\omega) \tilde{F}(\omega) \quad (12)$$

where

$$\alpha(\omega) = [m(\omega_0^2 - \omega^2 - i \omega \gamma)]^{-1}. \quad (13)$$

To proceed further and calculate energy shifts and mean energy shifts, we first of all calculate the free energy, using the "remarkable formula" for the free energy, in terms of  $\alpha(\omega)$ , obtained by Ford et al.<sup>14</sup>, which is valid regardless of the value of  $\alpha(\omega)$  chosen.

The system of oscillator coupled to the radiation field in thermal equilibrium at temperature  $T$  has a well defined free energy. The free energy ascribed to the oscillator,  $F_0(T)$ , is the free energy of this system minus the free energy of the radiation field in the absence of the oscillator. For this free energy Ford et al. obtained

$$F_0(T) = \frac{1}{\pi} \int_0^{\infty} d\omega f(\omega, T) \operatorname{Im} \left\{ \frac{d \ln \alpha(\omega)}{d\omega} \right\}, \quad (14)$$

where  $f(\omega, T)$  is the free energy of a single oscillator of frequency  $\omega$ , given by the familiar formula

$$f(\omega, T) = k T \ln [1 - \exp(-\hbar\omega/kT)]. \quad (15)$$

In (14) it is understood that  $\alpha(\omega)$  is the boundary value as  $\omega$  approaches the real axis from above. Note that we discard the  $T = 0$  contributions since our interest is in the temperature-dependent effects. Formula (14) is striking because it expresses the free energy of the interacting oscillator in terms of the susceptibility  $\alpha(\omega)$  alone.

For the purpose of accommodating models with different expressions for  $\alpha(\omega)$  it is convenient to write the result for  $\alpha(\omega)$  in the following general form:

$$\alpha(\omega) = -\frac{1}{\pi} \prod_n (\omega - \omega_n) \prod_i (\omega - \omega_i), \quad \begin{array}{l} i = 1, 2, 3, \dots \\ n = a, b, c, \dots \end{array} \quad (16)$$

It follows that

$$g(\omega) = \operatorname{Im} \left\{ \frac{d \ln \alpha(\omega)}{d\omega} \right\} = \sum_n \frac{\operatorname{Im} \omega_n}{|\omega - \omega_n|^2} - \sum_i \frac{\operatorname{Im} \omega_i}{|\omega - \omega_i|^2}. \quad (17)$$

In the case where  $\alpha(\omega)$  is given by (13), we see that the only roots are

$$\omega_{1,2} = -i(\gamma/2) \pm [\omega_0^2 - (\gamma/2)^2]^{1/2}. \quad (18)$$

Hence, from (14), (17), and (18), we obtain the following expression for the oscillator free energy:

$$F_0^{(PH)}(T) = \frac{1}{\pi} \int_0^{\infty} d\omega f(\omega, T) \frac{\gamma(\omega_0^2 + \omega^2)}{[(\omega_0^2 - \omega^2)^2 + \omega^2 \gamma^2]}, \quad (19)$$

where the superscript "PH" denotes the phenomenological, constant  $\gamma$ , model. This should be recognized as the free energy of an oscillator with natural frequency  $\omega_0$  and width  $\gamma$ . It is enlightening to form the corresponding energy,  $U_0^{(PH)}(T)$ , using the familiar thermodynamic relation between energy,  $U$ , and free energy,  $F$ , viz.

$$U = F - T \frac{\partial F}{\partial T} . \quad (20)$$

In the weak coupling limit ( $\gamma \rightarrow 0$ )

$$U_0^{(PH)} \rightarrow \hbar \omega_0 [\exp(\hbar \omega_0 / k T) - 1]^{-1} . \quad (21)$$

This is just the Planck energy of the quantum oscillator, and, of course, for  $\hbar \omega_0 \ll k T$  (high-temperature, low-frequency), we obtain  $k T$ . Thus, the result obtained by including damping (even a very small amount) gives a result very different from the no-damping case. In particular, it gives the desired  $k T$  result. However, there is no  $T^2$  or other energy shift but, instead, the effect of a non-zero  $\gamma$  is to give a width to the energy levels. Thus the question remains as to whether or not there exists an energy shift in addition to the energy level width. The answer is yes, as we will not demonstrate.

#### DAMPED HARMONIC OSCILLATOR: EXACT TREATMENT

We turn now to a discussion of the exact calculation carried out by Ford *et al.*<sup>14</sup>. The essence of their approach is the realization that, since we are dealing with a heat bath, a natural starting-point is the quantum Langevin equation, which takes the general form:

$$m \ddot{x} + \int_{-\infty}^t dt' \mu(t-t') \dot{x}(t') + K x = F(t) . \quad (22)$$

This is an equation for the time-dependent Heisenberg operator  $x(t)$ . The coupling with the radiative field is described by two terms: the radiation reaction term characterized by the memory function  $\mu(t)$ , and the fluctuating term characterized by the operator-valued random force  $F(t)$ . For our purposes we need this equation only to extract the generalized



susceptibility, which is done by forming the Fourier transform of (22) and writing the result in the form

$$\tilde{x}(\omega) = \alpha(\omega)\tilde{F}(\omega) . \quad (23)$$

Here  $\alpha(\omega)$  is the generalized susceptibility (a c-number) given by

$$\alpha(\omega) = [-m\omega^2 + K - i\omega \tilde{\mu}(\omega)]^{-1} , \quad (24)$$

where

$$\tilde{\mu}(\omega) = \int_0^{\infty} dt \mu(t) e^{i\omega t} , \quad \text{Im } \omega > 0 , \quad (25)$$

is the Fourier transform of the memory function. Clearly  $\tilde{\mu}(\omega)$  is analytic in the upper half  $\omega$ -plane.

The next step is the determination of  $\tilde{\mu}(\omega)$ . The phenomenological theory, as we saw, assumes that  $\tilde{\mu}(\omega) = m\gamma$ , independent of  $\omega$ . However,  $\tilde{\mu}(\omega)$  can be calculated exactly by starting with the quantum Hamiltonian for the oscillator interacting with the radiation field. The Heisenberg equations of motion are then used to obtain equations of motion for the system and bath variables in terms of each other. It is then possible to eliminate the bath variables and write the system equation of motion in the form (22) with

$$\tilde{\mu}(\omega) = 2 e^2 \Omega^2 \omega / 3 c^3 (\omega + i\Omega) , \quad (26)$$

where  $\Omega$  is a large cutoff frequency. From the form of  $\tilde{\mu}(\omega)$  it is clear that we should not take  $\Omega \rightarrow \infty$  (point electron) at this stage.

Substituting (26) in (24), and introducing three new parameters,  $\Omega'$ ,  $\omega_0$ , and  $\gamma$ , we obtain

$$\alpha(\omega) = \frac{(\omega + i\Omega)}{m(\omega + i\Omega')(\omega_0^2 - \omega^2 - i\gamma\omega)} , \quad (27)$$

where, in the large cut-off limit ( $\Omega' \gg \gamma$ ),

$$\omega_0 = (K/M)^{1/2}, \quad \gamma = 2e^2\omega_0^2/3Mc^3, \quad \frac{1}{\Omega'} = \frac{1}{\Omega} + \frac{\gamma}{\omega_0^2}, \quad (28)$$

and where  $M$  is the renormalized (observed) electron mass,

$$M = m + 2e^2\Omega/3c^3. \quad (29)$$

In contrast to the phenomenological case (constant  $\tilde{\nu}(\omega)$ ), it is clear by comparison of (13) and (27) that  $\alpha(\omega)$  has an extra zero at  $\omega = \omega_\alpha = -i\Omega$  and an extra pole at  $\omega = \omega_3 = -i\Omega'$ . It follows that there is an extra contribution to  $g(\omega)$  and hence to  $F_0(T)$ . The result may be written as

$$F_0(T) = F_0(T) + \Delta F_0(T), \quad (30)$$

where

$$\Delta F_0(T) = -\frac{\gamma}{\omega_0^2\pi} \int_0^\infty d\omega f(\omega, T) = \frac{\pi e^2 (kT)^2}{9Mc^3}. \quad (31)$$

The term  $F_0(T)$  is identical to  $F_0^{(PH)}(T)$  given in (19) except that now  $\omega_0 = (K/M)^{1/2}$  i.e. we have the renormalized mass  $M$  instead of the bare mass  $m$ , and, in addition,  $\gamma$  is given precisely by the expression in (28). The additional term  $\Delta F_0(T)$  implies a corresponding energy level shift,

$$\Delta U_0(T) = -\frac{\pi e^2 (kT)^2}{9Mc^3} \quad (32)$$

which is negative. We emphasize that for the oscillator this is an exact result, not limited to high temperatures. Also noteworthy is the fact that it is the negative of the result obtained using the same formalism as that of Farley-Wing<sup>9</sup> which appears to be consistent with the experimental results. However, Ford et al. have argued, according to the principle that the minimum isothermal work is equal to the change in free energy, that the measurements of Hollberg and Hall<sup>10</sup> measure the shift in free energy and not of energy. In addition, since our calculated shift in the

photon energy, given by (31), turns out to be independent of the frequency of the oscillator, our result is equivalent to the result one would obtain for a nearly free electron. Also, highly excited Rydberg states are well approximated by nearly free electron states (the high-frequency limit). We would therefore argue that for such Rydberg states the temperature-dependent shift in free energy is given by (31), in accord with the experimental results (since there is good reason to believe that the low-lying state shifts much less than the Rydberg state).

We conclude by summarizing the key features of the Ford et al.<sup>14</sup> calculation.

- (a) It incorporates a precise calculation of the damping and demonstrates the inadequacy of the phenomenological damping theory. The latter theory is Markovian in nature (no memory) whereas in fact an essential feature of the exact calculation is the existence of memory effects.
- (b) It demonstrates the necessity of not assuming a point electron while working out the details of the calculation even though the final results are insensitive to the precise choice of electron form factor.
- (c) a clear and natural separation is effected between terms which contribute to a finite linewidth and those which contribute to an actual energy shift.
- (d) a remarkable formula, (14) above, is obtained for the free energy ascribed to the oscillator and, in addition, Ford et al. argue that the change in free energy is what is actually measured.
- (e) it is an enlightening example of how one can eliminate the degrees of freedom of a heat bath or reservoir and obtain an equation of motion for the system of interest, which contains a friction or damping term that has not been introduced in a phenomenological manner but, instead, has been derived

precisely in terms of the heat bath parameters and the coupling between the system and heat bath.

- (f) the expression for the damping term makes manifest the fact that there is a direct proportionality between the line width and the coupling strength. This highlights once more the need to incorporate dissipation in the analysis. This is made especially striking by an examination of the expression for  $\Delta F_0(T)$  given in (31) - it is zero if  $\gamma$  is taken to be zero.

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