

Solutions to Assignment #5

1. Optical traps and scattering.

a) The AC Stark shift

We derived in class a perturbative expression for the wavefunction and thus the polarizability of an atom in an oscillating electric field. The result we obtained was straightforward except perhaps for the neglect of a “-1” term. This term was dropped because we chose to neglect transient effects, that is we sought to describe the atom after a long exposure to monochromatic light. Another way to view our neglect of transient terms is to consider that rather than suddenly switching on the the light field, we had ramped it on “adiabatically,” meaning that the atom would be oscillating in a steady state manner. Other than an overall phase factor, this would yield exactly the result that we had obtained in class, namely that if we consider an electric field $\epsilon(t) = \epsilon \hat{\mathbf{z}} \cos(\omega t)$ which couples the ground state $|g\rangle$ to a single excited state $|e\rangle$, and represent our wavefunction in the form

$$|\psi(t)\rangle = a_g |g\rangle + a_e e^{-i\omega_0 t} |e\rangle \quad (1)$$

then

$$a_e(t) = \frac{\epsilon}{2\hbar} \langle e | D_z | g \rangle \left(\frac{e^{i(\omega_0 - \omega)t}}{\omega_0 - \omega} + \frac{e^{i(\omega_0 + \omega)t}}{\omega_0 + \omega} \right) \quad (2)$$

The atomic dipole moment of the atom is given by

$$\mathbf{d}(t) = \frac{1}{\hbar} |\langle e | D_z | g \rangle|^2 \left(\frac{1}{\omega_0 - \omega} + \frac{1}{\omega_0 + \omega} \right) \epsilon(t) \quad (3)$$

$$= \alpha(\omega) \epsilon(t). \quad (4)$$

From the fact that the oscillator strength is unity we obtain

$$|\langle e | D_z | g \rangle|^2 = \frac{\hbar e^2}{2m\omega_0} \quad (5)$$

and thus,

$$\alpha(\omega) = \frac{e^2}{m} \frac{1}{\omega_0^2 - \omega^2} \quad (6)$$

The AC Stark shift is then given by

$$U_{AC}(i) = -\frac{1}{2} \alpha(\omega) \epsilon^2 \overline{\cos^2 \omega t} \quad (7)$$

$$= -\frac{1}{4} \frac{e^2}{m} \frac{1}{\omega_0^2 - \omega^2} \epsilon^2 \quad (8)$$

$$\boxed{U_{AC}(i) = -\frac{2\pi e^2 I}{mc(\omega_0^2 - \omega^2)}} \quad (9)$$

where the intensity I is given by $I = c\epsilon^2/8\pi$.

The AC Stark shift can also be obtained by nonperturbatively solving the time-dependent Schrödinger equation for the time-dependent eigenstates of the atom and field combined. This approach is suited for describing near-resonant driving of a two-level atom and is known as the *dressed atom picture*.

In this approach, we make the rotating wave approximation. This requires us to solve the following Hamiltonian (see Homework 1, problem 2):

$$H = \frac{\hbar}{2} \begin{pmatrix} -\omega_0 & \omega_R e^{i\omega t} \\ \omega_R e^{i\omega t} & \omega_0 \end{pmatrix} \quad (10)$$

where ω_0 is the frequency separation between the S and P states, ω_R is the Rabi frequency, and ω is the radiation frequency, as usual. The time-dependent eigenstates are

$$|+\rangle = e^{-i(\omega'_R - \omega)t/2} [\cos\theta |S\rangle + \sin\theta e^{-i\omega t} |P\rangle] \quad (11)$$

$$|-\rangle = e^{-i(-\omega'_R + \omega)t/2} [-\sin\theta e^{i\omega t} |S\rangle + \cos\theta |P\rangle] \quad (12)$$

up to a shared, global (unimportant) phase. We again use the conventions $\delta = \omega - \omega_0$, $\omega'_R = \sqrt{\omega_R^2 + \delta^2}$, $\sin\theta = \sqrt{\frac{\omega'_R + \delta}{2\omega'_R}}$, and $\cos\theta = \sqrt{\frac{\omega'_R - \delta}{2\omega'_R}}$.

Note, for $\delta < 0$ (red-detuning, as we are using here), in the weak field limit ($\omega_R = 0$), we identify the states

$$|+\rangle \rightarrow |P\rangle \quad (13)$$

$$|-\rangle \rightarrow |S\rangle \quad (14)$$

Therefore, if we again neglect transients, then the effect of slowly turning on the light is to place our once ground-state atom into the dressed state $|-\rangle$. The time-dependent phase factors in the equations above can be identified as energies. We obtain the AC Stark shift by considering the difference in energy with the light on and off, i.e.

$$U_{AC}(ii) = \hbar \left(\left. \frac{-\omega'_R}{2} \right|_{\omega_R} - \left. \frac{-\omega'_R}{2} \right|_{\omega_R=0} \right) \quad (15)$$

$$= -\hbar \frac{\sqrt{\omega_R^2 + \delta^2} - |\delta|}{2} \quad (16)$$

$$= -\frac{\hbar\omega_R^2}{4|\delta|} \quad (17)$$

Using the definition of the oscillator strength and the identification

$$\frac{\hbar\omega_R}{2} = \frac{\epsilon \cdot \mathbf{D}}{2} \quad (18)$$

we obtain

$$U_{AC}(ii) = -\frac{e^2}{8m\omega_0} \left(\frac{1}{\omega_0 - \omega} \right) \epsilon^2 = -\frac{\pi e^2 I}{m\omega_0(\omega_0 - \omega)}. \quad (19)$$

We can cast these results in more useful quantities by introducing the saturation intensity I_{SAT} which for our ideal two-level atom is

$$I_{SAT} = \frac{m\omega_0\Gamma^2}{8\pi\alpha}. \quad (20)$$

$\alpha = \frac{e^2}{\hbar c}$ is the fine structure constant. This gives

$$U_{AC}(i) = -\frac{1}{4} \frac{\omega_0\Gamma}{\omega_0^2 - \omega^2} \frac{I}{I_{SAT}} \hbar\Gamma. \quad (21)$$

and for small detunings, we get the dressed atom result, namely

$$U_{AC}(ii) = -\frac{1}{4} \frac{I}{I_{SAT}} \frac{1}{2|\delta|/\Gamma} \hbar\Gamma. \quad (22)$$

The ratio of results (i) and (ii) is

$$\boxed{\frac{\frac{1}{\omega_0 - \omega} + \frac{1}{\omega_0 + \omega}}{\frac{1}{\omega_0 - \omega}} = 1 + \frac{\omega_0 - \omega}{\omega_0 + \omega}} \quad (23)$$

On resonance, the two expressions are identical. At DC ($\omega = 0$), the perturbative (correct) result is twice the dressed atom result, since then the counter- and co-rotating terms give equal Stark shifts.

b) *Excited state fraction*

Using the above results, one finds the likelihood of finding the atom in the excited state in the perturbative approach is

$$P_e = |a_e^2| \quad (24)$$

$$= \frac{\epsilon^2}{4\hbar^2} |\langle e | D_z | g \rangle|^2 \left(\frac{1}{(\omega_0 - \omega)^2} + \frac{1}{(\omega_0 + \omega)^2} + \dots \right) \quad (25)$$

where the \dots refer to rapidly oscillating terms that we neglect. From the definition of the oscillator strength, we thus obtain

$$\boxed{P_e(i) = \frac{\epsilon^2 e^2}{8m\hbar\omega_0} \left(\frac{1}{(\omega_0 - \omega)^2} + \frac{1}{(\omega_0 + \omega)^2} \right) = \frac{\pi\alpha I}{m\omega_0} \left(\frac{1}{(\omega_0 - \omega)^2} + \frac{1}{(\omega_0 + \omega)^2} \right)}. \quad (26)$$

For the dressed atom result, we obtain

$$P_e(ii) = \cos^2 \theta \quad (27)$$

$$= \frac{1 + \cos 2\theta}{2} \quad (28)$$

$$= \frac{\omega'_R - |\delta|}{2\omega'_R} \quad (29)$$

$$\simeq \frac{\omega_R^2}{4\delta^2} \quad (30)$$

$$\boxed{P_e(ii) = \frac{\pi\alpha I}{m\omega_0} \frac{1}{(\omega_0 - \omega)^2}} \quad (31)$$

Equivalently, expressed in terms of the saturation intensity, we find

$$P_e = \frac{1}{2} \frac{I}{I_{SAT}} \frac{1}{(2|\delta|/\Gamma)^2}. \quad (32)$$

c) *Rayleigh scattering rate*

Rayleigh scattering can be calculated several ways. Let us use results of classical electromagnetism (for a fully quantum-mechanical derivation, look up Exercize 3 in Cohen-Tannoudji's *Atom-Photon Interactions*). The power emitted by an oscillating dipole of moment $\mathbf{d}(t) = \mathbf{d} \cos \omega t$ is [Jackson, Chapter 9.2]

$$P = \frac{ck^4}{3} |\mathbf{d}|^2. \quad (33)$$

Since each photon carries $\hbar\omega$ of energy, the photon scattering rate is

$$\Gamma_{scat} = \frac{P}{\hbar\omega} = \frac{\omega^3}{3\hbar c^3} |\mathbf{d}|^2. \quad (34)$$

In part a) we calculated the dipole moments of the atoms in a monochromatic wave. So we obtain

$$\Gamma_{scat}(i) = \frac{\omega^3}{3\hbar c^3} \times \left(\frac{e^2}{m} \frac{1}{\omega_0^2 - \omega^2} \right)^2 \frac{8\pi I}{c}. \quad (35)$$

We can also recast this equation into the useful form

$$\Gamma_{scat} = \frac{|U_{AC}|}{\hbar} \times \frac{4}{3} \alpha^3 \frac{\omega}{\omega_A} \frac{\omega^2}{\omega_0^2 - \omega^2} \quad (36)$$

We obtain the dressed atom result with the substitution

$$\left(\frac{1}{\omega_0 - \omega} + \frac{1}{\omega_0 + \omega} \right) = \frac{2\omega_0}{\omega_0^2 - \omega^2} \rightarrow \frac{1}{\omega_0 - \omega} \quad (37)$$

so

$$\Gamma_{scat}(ii) = \frac{\omega^3}{3\hbar c^3} \times \left(\frac{e^2}{m} \frac{1}{2\omega_0} \frac{1}{\omega_0 - \omega} \right)^2 \frac{8\pi I}{c}. \quad (38)$$

We can relate this to the results of part b) by

$$\Gamma_{scat}(ii) = \left(\frac{\pi e^2 I}{\hbar c m \omega_0 \delta^2} \right) \times \left(\frac{4}{3} \frac{\omega_0^3}{c^3 \hbar} \frac{\hbar e^2}{2m\omega_0} \right) \times \left(\frac{\omega}{\omega_0} \right)^3 \quad (39)$$

$$= P_e \times \Gamma \times \left(\frac{\omega}{\omega_0} \right)^3. \quad (40)$$

Thus, the Rayleigh scattering rate is given by the probability of being in the excited state, times the spontaneous emission rate from that state, corrected by a phase-space factor which guarantees that an atom in a DC field does not radiate.

As one can see, when $|\omega - \omega_0| \ll \omega_0$, the scattered power is $\hbar\omega\Gamma_{scat}$ proportional to ω^4 – the blue-sky formula.

d) *Proper choice for an optical trap*

We use the following expressions

$$U_{AC}(i) = -\frac{1}{4} \frac{\omega_0 \Gamma}{\omega_0^2 - \omega^2} \frac{I}{I_{SAT}} \hbar \Gamma \quad (41)$$

and

$$\Gamma_{scat} = \frac{|U_{AC}|}{\hbar} \times \frac{4}{3} \alpha^3 \frac{\omega}{\omega_A} \frac{\omega^2}{\omega_0^2 - \omega^2} \quad (42)$$

Assuming an oscillator strength of unity for sodium gives

$$\Gamma = \frac{4}{3} \frac{\omega_0^3}{\hbar c^3} D^2 \quad (43)$$

$$= \frac{2}{3} \alpha^3 \frac{\omega_0}{\omega_A} \omega_0 \quad (44)$$

$$= 6.3 \times 10^7 s^{-1} \quad (45)$$

$$I_{SAT} = \frac{m\omega_0 \Gamma^2}{8\pi\alpha} = 6.2 \frac{\text{mW}}{\text{cm}^2} \quad (46)$$

For a 10 μK trap depth,

$$I = 0.52 \frac{\text{mW}}{\text{cm}^2} \times \frac{\omega_0^2 - \omega^2}{\omega_0 \Gamma} \quad (47)$$

and for a 6 μm spot size, this implies a power of

$$P = I \cdot 5.6 \times 10^{-7} \text{cm}^2 \quad (48)$$

$$\Gamma_{\text{scat}} = 0.68 \text{s}^{-1} \frac{\omega}{\omega_A} \frac{\omega^2}{\omega_0^2 - \omega^2} \quad (49)$$

Yellow Option: For the small detuning

$$I = 0.52 \frac{\text{mW}}{\text{cm}^2} \times \frac{2|\delta|}{\Gamma} = 175 \frac{\text{mW}}{\text{cm}^2} \quad (50)$$

$$\boxed{P = 9.6 \times 10^{-5} \text{mW}} \quad (51)$$

$$\boxed{\Gamma_{\text{scat}} = 7.9 \times 10^3 \text{s}^{-1}} \quad (52)$$

Red Option:

$$\omega = 3.0 \times 10^{15} \text{s}^{-1} \quad (53)$$

$$I = 3.2 \times 10^6 \frac{\text{mW}}{\text{cm}^2} \quad (54)$$

$$\boxed{P = 1.8 \text{mW}} \quad (55)$$

$$\boxed{\Gamma_{\text{scat}} = 0.36 \text{s}^{-1}} \quad (56)$$

Infrared Option:

$$\omega = 1.9 \times 10^{15} \text{s}^{-1} \quad (57)$$

$$I = 1.7 \times 10^7 \frac{\text{mW}}{\text{cm}^2} \quad (58)$$

$$\boxed{P = 9.6 \text{mW}} \quad (59)$$

$$\boxed{\Gamma_{\text{scat}} = 1.7 \times 10^{-2} \text{s}^{-1}} \quad (60)$$

Based on the above considerations, we chose to use infrared light — only handfults of milliwatts are needed, and the optical traps are extremely long lived. As a result, we have been able to study optically trapped condensates for as long as 20 \sim 30 seconds.

2. Gauge transformations.

See attached sheet from Weissbluth *Photon-Atom Interactions*.

3. Optical Transitions Driven by Blackbody Radiation.

a)

The average energy density is:

$$u = \frac{1}{V} \int_0^\infty \frac{\rho(\omega) \hbar \omega}{e^{\frac{\hbar \omega}{kT}} - 1} d\omega$$

The density of states is:

$$\rho(\omega) = \frac{\omega^2 V}{\pi^2 c^3}$$

Plugging this in we get:

$$\begin{aligned}
u &= \frac{\hbar}{\pi^2 c^3} \int_0^\infty \frac{\omega^3}{e^{\frac{\hbar\omega}{kT}} - 1} d\omega \\
&= \frac{(kT)^4}{\pi^2 (\hbar c)^3} \int_0^\infty \frac{x^3}{e^{x^3} - 1} dx
\end{aligned}$$

$$\boxed{u = \frac{\pi^2 (kT)^4}{15 (\hbar c)^3}}$$

The average photon density is:

$$\begin{aligned}
n &= \frac{1}{V} \int_0^\infty \frac{\rho(\omega)}{e^{\frac{\hbar\omega}{kT}} - 1} d\omega \\
&= \frac{(kT)^3}{\pi^2 (\hbar c)^3} \int_0^\infty \frac{x^2}{e^x - 1} dx
\end{aligned}$$

$$\boxed{n = \frac{2.404}{\pi^2} \left(\frac{kT}{\hbar c} \right)^3}$$

b)

$$\begin{aligned}
u &= \frac{1}{8\pi} (E^2 + B^2) \\
&= \frac{1}{4\pi} E^2 \\
E^2 &= 4\pi u \\
E_{rms} &= \sqrt{4\pi u}
\end{aligned}$$

Plugging in u , we get

$$\boxed{E_{rms} = B_{rms} = \sqrt{\frac{4\pi^3 (kT)^4}{15 (\hbar c)^3}}}$$

c) The energy flow rate is simply given by the Poynting vector

$$\begin{aligned}
\vec{S} &= \frac{c}{4\pi} \vec{E} \times \vec{B} \\
|\vec{S}| &= uc
\end{aligned}$$

To get the intensity escaping through the hole, we just integrate this over a hemisphere (multiplied by a cosine factor which takes into account the vanishing size of the hole as viewed from steep angles).

$$\begin{aligned}
P &= cu \frac{\int \cos \theta d\Omega}{\int d\Omega} \\
&= \frac{1}{4} cu \\
&= \frac{\pi^2 (kT)^4}{60 \hbar^3 c^2} \\
&= \sigma T^4
\end{aligned}$$

- d) The intensity at the sun's surface can easily be found from the intensity at the earth. If r_{eO} is the radius of the earth's orbit, and r_{sun} is the radius of the sun, then

$$\begin{aligned}
I_{sun} &= I_{earth} \left(\frac{r_{eO}}{r_{sun}} \right)^2 \\
&= I_{earth} \left(\frac{1}{\sin \frac{\theta}{2}} \right)^2 \\
&= \frac{1.4 kW/m^2}{(4.45 \cdot 10^{-3})^2} \\
&= 70.7 MW/m^2 \\
T &= \left(\frac{I_{sun}}{\sigma} \right)^{\frac{1}{4}} \\
&= 5942 K
\end{aligned}$$

- e) The transition rate is related to the average number of photons within the linewidth of the transition (which is proportional to the spectral density \bar{I}).

$$\begin{aligned}
R &= \frac{\pi}{2} \sigma \frac{\bar{I}(\omega) \cdot \Gamma}{\hbar \omega} \\
&= \frac{\pi}{2} (6\pi \bar{\lambda}^2) \frac{\Gamma}{\hbar \omega} \left(\frac{\hbar \omega^3}{\pi^2 c^2} \frac{1}{e^{\frac{\hbar \omega}{kT}} - 1} \right) \\
&= \frac{3\Gamma}{e^{\frac{\hbar \omega}{kT}} - 1} \\
e^{\frac{\hbar \omega}{kT}} &= \frac{3\Gamma}{R} + 1 \\
\frac{\hbar \omega}{kT} &= \log \frac{3\Gamma}{R} + 1 \\
T &= \frac{\hbar \omega}{k \log \left(\frac{3\Gamma}{R} + 1 \right)} \\
&= \frac{2\pi \hbar c}{k \lambda \log \left(\frac{3\Gamma}{R} + 1 \right)}
\end{aligned}$$

We want to trap for a minute, so $R = (1 \text{ minute})^{-1} = 0.01667 \text{ Hz}$. That gives us a temperature of:

$$\boxed{T = 1055 K}$$

4. RF Transition Lifetimes and RF Blackbody Transitions.

a)

Since this transition is a magnetic dipole transition, as discussed in class, the transition rate should be down by a factor of α^2 compared to an electric dipole transition at the same frequency. In addition, since the lifetime scales with ω^{-3} , we expect another large factor compared to higher frequency optical transitions. So we expect to get a long lifetime for this state.

$$\begin{aligned}\Gamma &= \frac{4 \omega^3 \mu_B^2}{3 \hbar c^3} \\ &= 2.9 \cdot 10^{-15} Hz \\ \tau &= \frac{1}{\Gamma} = 3.4 \cdot 10^{14} \text{seconds} = 11 \text{millionyears}\end{aligned}$$

$$\boxed{\tau = 3.4 \cdot 10^{14} \text{seconds} = 11 \text{millionyears}}$$

b) We already have the formula for this from the last problem

$$R = \frac{3\Gamma}{e^{\frac{\hbar\omega}{kT}} - 1}$$

Since we know that λ is 21 cm, ω is 8.98 GHz. Plugging the numbers in we get:

$$\boxed{R(T = 4K) = 5 \cdot 10^{-13} Hz}$$

$$\boxed{R(T = 300K) = 3.8 \cdot 10^{-11} Hz}$$

In the same way,

$$(\Delta t_2)^2 = \frac{1}{2}. \quad (4.303)$$

From Eq. (4.299), however,

$$\langle \tau | t_1 | \tau \rangle = \langle \tau | a_1 | \tau \rangle e^r, \quad \langle \tau | t_1^2 | \tau \rangle = \langle \tau | a_1^2 | \tau \rangle e^{2r}, \quad (4.304a)$$

$$\langle \tau | t_2 | \tau \rangle = \langle \tau | a_2 | \tau \rangle e^{-r}, \quad \langle \tau | t_2^2 | \tau \rangle = \langle \tau | a_2^2 | \tau \rangle e^{-2r}. \quad (4.304b)$$

Consequently,

$$(\Delta a_1)_r^2 = (\Delta t_1)^2 e^{-2r} = \frac{1}{2} e^{-2r}, \quad (4.305a)$$

$$(\Delta a_2)_r^2 = (\Delta t_2)^2 e^{2r} = \frac{1}{2} e^{2r}, \quad (4.305b)$$

$$(\Delta a_1 \Delta a_2)_r = \frac{1}{2}. \quad (4.305c)$$

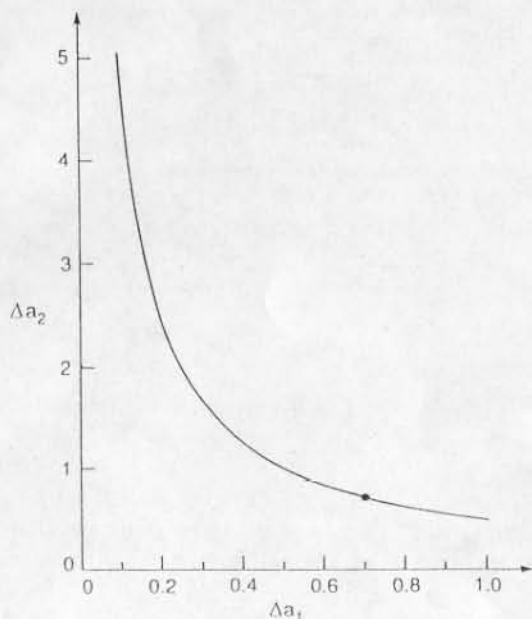


FIGURE 4.3 A plot of Δa_2 vs Δa_1 where $\Delta a_1 \Delta a_2 = 1/2$. The coherent state is the single point at $\Delta a_1 = \Delta a_2 = (1/2)^{1/2}$. Squeezed states correspond to other points on the curve where $\Delta a_1 \neq \Delta a_2$.

Solutions to Problem 4.

4.10 Gauge Transformations

from Weissbluth, Photon-Atom Interactions 181

The conclusion, then, is that in a squeezed state the magnitude of the fluctuations or the noise level in one of the two quadrature components can be reduced below the noise level of a coherent state, which, we saw, corresponds to the vacuum limit. Clearly, the noise diminution in one quadrature is achieved at the expense of noise enhancement in the other quadrature. A plot of Eq. (4.305) is shown in Fig. 4.3.

Various nonlinear processes have been proposed for the production of squeezed states. Among them are parametric amplifiers [12], degenerate and nondegenerate four-wave mixers [13–17], resonance fluorescence [18, 19], and interaction of electromagnetic waves with plasmas [20]. Experimental verification employing phase-sensitive detection methods has been demonstrated by four-wave mixing [21–23] and by degenerate parametric down-conversion [24]. Applications of squeezed states to communication [25] and to gravitational radiation detectors [26] have been suggested.

4.10 Gauge Transformations

In classical electromagnetic theory, a gauge transformation on the scalar potential $\phi(\mathbf{r}, t)$ and the vector potential $\mathbf{A}(\mathbf{r}, t)$ is defined by the relations

$$\phi'(\mathbf{r}, t) = \phi(\mathbf{r}, t) - \frac{\partial}{\partial t} f(\mathbf{r}, t), \quad (4.306)$$

$$\mathbf{A}'(\mathbf{r}, t) = \mathbf{A}(\mathbf{r}, t) + \nabla f(\mathbf{r}, t),$$

where $f(\mathbf{r}, t)$ is an arbitrary differentiable scalar function of space and time. Since the fields are related to the potentials by

$$\mathbf{E}(\mathbf{r}, t) = -\nabla\phi(\mathbf{r}, t) - \frac{\partial}{\partial t} \mathbf{A}(\mathbf{r}, t), \quad (4.307)$$

$$\mathbf{B}(\mathbf{r}, t) = \nabla \times \mathbf{A}(\mathbf{r}, t),$$

a gauge transformation of the potentials has no effect on the fields; that is, when ϕ and \mathbf{A} are replaced by ϕ' and \mathbf{A}' , the fields are unchanged. Hence, the potentials are not determined uniquely.

It now will be shown that the transformations (4.306) also may arise in a quantum mechanical context. The fundamental Hamiltonian for the interaction of a nonrelativistic electron with an external electromagnetic field [1, 2] is

$$\mathcal{H}(\mathbf{r}, t) = \frac{1}{2m_e} [\mathbf{p} + e\mathbf{A}(\mathbf{r}, t)]^2 - e\phi(\mathbf{r}, t), \quad (4.308)$$

where m , e , and p are the mass, charge, and linear momentum, respectively, of the electron; $\phi(\mathbf{r}, t)$ and $\mathbf{A}(\mathbf{r}, t)$ are the scalar and vector potentials, respectively, of the external field. It is recalled (Section 3.2) that the unitary transformation

$$\psi'(\mathbf{r}, t) = O\psi(\mathbf{r}, t) \quad (4.309)$$

transforms the time-dependent Schrödinger equation

$$ih \frac{\partial}{\partial t} \psi(\mathbf{r}, t) = \mathcal{H} \psi(\mathbf{r}, t) \quad (4.310)$$

into

$$ih \frac{\partial}{\partial t} \psi'(\mathbf{r}, t) = \mathcal{H}' \psi'(\mathbf{r}, t), \quad (4.311)$$

where

$$\alpha) \quad \boxed{\mathcal{H}' = O \mathcal{H} O^\dagger + ih \frac{\partial O}{\partial t} O^\dagger} \quad (4.312)$$

Now, let

$$O = e^{-ief/\hbar}, \quad f \equiv f(\mathbf{r}, t). \quad (4.313)$$

With $p = -ih\nabla$,

$$pO^\dagger = eO^\dagger \nabla f + O^\dagger p, \quad (p + e\mathbf{A})O^\dagger = O^\dagger [p + e(\mathbf{A} + \nabla f)], \quad (4.314)$$

$$(p + e\mathbf{A})^2 O^\dagger = (p + e\mathbf{A})O^\dagger [p + e(\mathbf{A} + \nabla f)] = O^\dagger [p + e(\mathbf{A} + \nabla f)]^2, \quad (4.315)$$

we have

$$O(p + e\mathbf{A})^2 O^\dagger = [p + e(\mathbf{A} + \nabla f)]^2, \quad (4.316)$$

$$O\phi O^\dagger = \phi, \quad ih \frac{\partial O}{\partial t} O^\dagger = e \frac{\partial f}{\partial t}. \quad (4.317)$$

The Hamiltonian (Eq. (4.308)) is transformed then into

$$b) \quad \boxed{\mathcal{H}' = \frac{1}{2m} [p + e(\mathbf{A} + \nabla f)]^2 - e \left(\phi - \frac{\partial f}{\partial t} \right) = \frac{1}{2m} [p + e\mathbf{A}']^2 - e\phi'} \quad (4.318)$$

What has been shown is that the operator O defined by Eq. (4.313) acting through the transformation (4.312), generates a gauge transformation of the type of Eq. (4.306) when the Hamiltonian has the form of Eq. (4.308). This, then, is the quantum mechanical counterpart to the classical gauge transformation. Since O is a unitary operator, its effect on the wave function ψ is to alter the phase but not the absolute value. Consequently, the physical content of quantum mechanics cannot depend on the specific choice of the gauge function $f(\mathbf{r}, t)$.

Let us now consider the case of a electron interacting with an electromagnetic field that has no scalar potential ($\phi = 0$). The field then, is, described completely by the vector potential alone, as in the case of a radiation field. The Hamiltonian (Eq. (4.308)) reduces to

$$\mathcal{H} = \frac{1}{2m} [p + e\mathbf{A}(\mathbf{r}, t)]^2, \quad (4.319)$$

and the transformed Hamiltonian (Eq. (4.318)) reduces to

$$\mathcal{H}' = \frac{1}{2m} [p + e(\mathbf{A} + \nabla f)]^2 + e \frac{\partial f}{\partial t}. \quad (4.320)$$

We now choose a gauge function in the form

$$f(\mathbf{r}, t) = -\mathbf{A}(t) \cdot \mathbf{r}, \quad (4.321)$$

in which the vector potential is independent of spatial coordinates. Since we are concerned mainly with atomic or molecular systems, it is sufficient for \mathbf{A} (or the fields \mathbf{E} and \mathbf{B}) not to vary over a distance of a few Angstroms. In other words, the wavelengths associated with \mathbf{A} must be long in comparison with atomic dimensions, or,

$$\mathbf{k} \cdot \mathbf{r} \ll 1. \quad (4.322)$$

In the context of a multipole expansion, this condition is recognized as the approximation whereby all multipoles except the leading one—the dipole term—are ignored. Specifically, it is the electric dipole term ($E1$) that is of primary interest.

With the gauge function (Eq. (4.321)) we have

$$\nabla f = -\mathbf{A}(t), \quad \frac{\partial f}{\partial t} = -\mathbf{r} \cdot \frac{\partial \mathbf{A}}{\partial t} = \mathbf{r} \cdot \mathbf{E}(t), \quad (4.323)$$

and

$$c) \quad \boxed{\mathcal{H}' = \frac{p^2}{2m} + e\mathbf{r} \cdot \mathbf{E}(t)} \quad (4.324)$$

electric field is

Thus, the effect of the gauge transformation with the gauge function (Eq. (4.321)) has been to eliminate the vector potential from the Hamiltonian.

The Hamiltonian, Eq. (4.319), contains the terms $e(\mathbf{p} \cdot \mathbf{A} + \mathbf{A} \cdot \mathbf{p})$. If $g(\mathbf{r}, t)$ is an arbitrary function of position and time,

$$\mathbf{p} \cdot \mathbf{A}g(\mathbf{r}, t) = \mathbf{A} \cdot [pg(\mathbf{r}, t)] + g(\mathbf{r}, t)[\mathbf{p} \cdot \mathbf{A}]. \quad (4.325)$$

Since we are interested exclusively in radiation fields obeying the transversality condition, the vector potential is subject to the Coulomb gauge

(Section 4.2) $\mathbf{V} \cdot \mathbf{A} = 0$. Therefore,

$$(\mathbf{p} \cdot \mathbf{A}) = -i\hbar \mathbf{V} \cdot \mathbf{A} = 0, \quad (4.326)$$

and

$$\mathbf{p} \cdot \mathbf{A} = \mathbf{A} \cdot \mathbf{p}. \quad (4.327)$$

We see, then, that the interaction term in \mathcal{H} is

$$\frac{e}{m} \mathbf{p} \cdot \mathbf{A} + \left(\frac{e}{2m}\right)^2 A^2. \quad (4.328)$$

On the other hand, the interaction term in the gauge-transformed Hamiltonian (Eq. (4.324)) is

$$e\mathbf{r} \cdot \mathbf{E}(t) = -\mathbf{d} \cdot \mathbf{E}(t) \quad (4.329)$$

where $\mathbf{d} = -e\mathbf{r}$ is the electronic dipole moment operator. Thus, within the dipole approximation, the single term (Eq. (4.329)) is equivalent to the two terms (Eq. (4.328)).

One should be aware, however, that although Eq. (4.329) is used widely as the interaction Hamiltonian in quantum optics, more delicate considerations may need to be taken into account [27–29].

4.11 Density Matrix for Interactions with Monochromatic Fields

For later applications to nonlinear phenomena, we now shall evaluate the density matrix elements shown in Eqs. (2.274) and (2.275) for systems initially in thermal equilibrium when $V(t)$ is the dipole interaction operator

$$V(t) = -\mathbf{d} \cdot \mathbf{E}(t) \quad (4.330)$$

as in Eq. (4.329). Following the convention due to Shen [30], the complex classical field (with the spatial dependence omitted) is written in terms of its Fourier components.

$$\mathbf{E}(t) = \sum_r \mathbf{E}(\omega_r) e^{-i\omega_r t}. \quad (4.331)$$

Employing the definitions

$$v_{kl}(\omega_r) \equiv -\mathbf{d}_{kl} \cdot \mathbf{E}(\omega_r), \quad (4.332)$$

and the assumption that d_{kl} are real matrix elements, the matrix element of the interaction operator, Eq. (4.330), for a single monochromatic field of frequency ω , is

$$V_{kl}(t) = v_{kl}(\omega) e^{-i\omega t}, \quad (4.333)$$

and the matrix element (Eq. (2.274a)) becomes

$$\langle k | \rho_a^{(1)}(t) | l \rangle = -\frac{i}{\hbar} e^{-i\omega_a t} \rho_{ll}^0 v_{kl}(\omega) \int_{t_0}^t dt_1 e^{i(\omega_{kl} - \omega)t_1}. \quad (4.335)$$

The integral is readily evaluated, bearing in mind that $V(t) = 0$ for $t \leq t_0$ as discussed in Section 2.8. Thus,

$$\langle k | \rho_a^{(1)}(t) | l \rangle = -\frac{1}{\hbar} \rho_{ll}^0 \frac{v_{kl}(\omega) e^{-i\omega t}}{(\omega_{kl} - \omega)}. \quad (4.336)$$

Following the same procedure, the matrix element (Eq. (2.274b)) becomes

$$\langle k | \rho_b^{(1)}(t) | l \rangle = \frac{1}{\hbar} \rho_{kk}^0 \frac{v_{kl}(\omega) e^{-i\omega t}}{(\omega_{kl} - \omega)}. \quad (4.337)$$

Combining Eqs. (4.336) and (4.337),

$$\begin{aligned} \langle k | \rho^{(1)}(t) | l \rangle &= \langle k | \rho_a^{(1)}(t) | l \rangle + \langle k | \rho_b^{(1)}(t) | l \rangle \\ &= \frac{1}{\hbar} (\rho_{kk}^0 - \rho_{ll}^0) \frac{v_{kl}(\omega) e^{-i\omega t}}{(\omega_{kl} - \omega)}. \end{aligned} \quad (4.338)$$

When the field contains more than one frequency, it is necessary merely to write an expression like the one above for each frequency; the final matrix element is obtained by summing over all frequencies, i.e.,

$$\langle k | \rho^{(1)}(t) | l \rangle \equiv \rho_{kl}^{(1)}(t) = \frac{1}{\hbar} (\rho_{kk}^0 - \rho_{ll}^0) \sum_r \frac{v_{kl}(\omega_r) e^{-i\omega_r t}}{(\omega_{kl} - \omega_r)}. \quad (4.339)$$

It is clear from this expression that the individual frequencies do not mix with one another—each frequency behaves independently. Because of this property, $\rho^{(1)}(t)$ is said to be associated with linear effects.

Nonlinear effects begin to appear when we investigate $\rho^{(2)}(t)$. For this purpose, let

$$\mathbf{E}(t) = \mathbf{E}(\omega_1) e^{-i\omega_1 t} + \mathbf{E}(\omega_2) e^{-i\omega_2 t}. \quad (4.340)$$

The matrix element of the interaction operator then may be written

$$V_{kl}(t) = v_{kl}(\omega_1) e^{-i\omega_1 t} + v_{kl}(\omega_2) e^{-i\omega_2 t}. \quad (4.341)$$

With this interaction, we can generate a density matrix whose time-dependence is proportional to $\exp[-i(\omega_1 + \omega_2)t]$. Should we wish to generate a density matrix proportional to $\exp[-i(\omega_1 - \omega_2)t]$, the Fourier components

$$\mathbf{E}(t) = \mathbf{E}(\omega_1) e^{-i\omega_1 t} + \mathbf{E}^*(\omega_2) e^{i\omega_2 t}, \quad (4.342)$$