8.421 Atomic Physics I — *Spring 2006*

Prof. Wolfgang Ketterle

Problem Set 10 – Two-Photon Processes
Due Friday, May 5th, 2006

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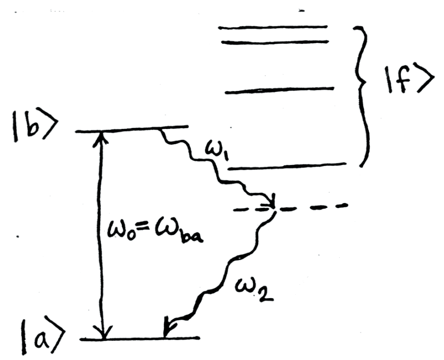
Recitation: Tuesday, May 2nd @ 7pm in 26-201

Problem 1. Spontaneous Two-Photon Emission



In this problem you will first derive a very general formula for calculating the rate and fluorescence spectrum of an atomic transition from an upper state $|b\rangle$ to a lower state $|a\rangle$ by spontaneous emission of two photons. Next, you will apply this formula to estimate the lifetime of the metastable $2S$ state of hydrogen. Throughout this problem, difference frequencies are labeled according to the convention $\omega_{mn} \equiv \omega_m - \omega_n$.

(a) The Göppert-Mayer formula.

In order to find $A(\omega_1)\mathrm{d}\omega_1$, the rate for spontaneous emission of two photons with one photon having frequency between ω_1 and $\omega_1 + \mathrm{d}\omega_1$, we can follow an approach similar to the one used in class for calculating the single-photon spontaneous emission rate. We will start with the two-photon excitation rate from $|a\rangle$ to $|b\rangle$ for monochromatic beams with frequencies ω_1 and ω_2 (as in section 9.2 of the notes).

If we neglect absorption of two photons with the same frequency, then second order perturbation theory gives the following amplitude for state $|b\rangle$ after excitation for a time t (see Eq. 9.11):

$$a_b^{[2]} = \frac{1}{4\hbar^2}\sum_f \left\{ \frac{H_{bf,2}H_{fa,1}}{\omega_1 - \omega_{fa}} \frac{e^{i(\omega_{ba}-\omega_1-\omega_2)t} - 1}{\omega_{ba} - \omega_1 - \omega_2} + \frac{H_{bf,1}H_{fa,2}}{\omega_2 - \omega_{fa}} \frac{e^{i(\omega_{ba}-\omega_1-\omega_2)t} - 1}{\omega_{ba} - \omega_1 - \omega_2} \right\}$$

(As will be clear later, the contribution to the spontaneous emission rate from processes involving two photons of the same frequency is completely negligible because there are so many other probable ways to emit two photons.)

$\triangleright$ Find the excitation rate $\Gamma_{ab}(\omega_1)$ when one beam has frequency ω_1. Your expression should include a resonance condition which constrains ω_2.

▷ Show that your expression is mathematically equivalent to

$$\Gamma_{ab}(\omega_1) = \Gamma_{ba}(\omega_1)$$
$$= \frac{\pi E_1^2 E_2^2 e^4}{8\hbar^4} \left| \sum_f \left\{ \frac{\langle a|\mathbf{r}\cdot\hat{\mathbf{e}}_1|f\rangle\langle f|\mathbf{r}\cdot\hat{\mathbf{e}}_2|b\rangle}{\omega_1+\omega_{fb}} + \frac{\langle a|\mathbf{r}\cdot\hat{\mathbf{e}}_2|f\rangle\langle f|\mathbf{r}\cdot\hat{\mathbf{e}}_1|b\rangle}{\omega_2+\omega_{fb}} \right\} \right|^2$$
$$\times\, \delta(\omega_{ba} - (\omega_1+\omega_2)).$$

Note that we can also obtain this expression by considering two-photon *emission*—one needs only to swap a and b and change the signs of ω_1 and ω_2 in the expression for the absorption rate.

▷ Give a physical interpretation for the terms in the sum.

Following now either the method of sections 7.5 and 7.6 or the method outlined in class for calculation of the single-photon spontaneous emission rate, express E_1^2 and E_2^2 in terms of the number of photons n_1, n_2 occupying some specific modes with frequencies ω_1, ω_2. Next, replace n_1, n_2 with the densities of modes at frequencies ω_1, ω_2. Proceed to calculate $A(\omega_1)\mathrm{d}\omega_1$, the two-photon spontaneous emission rate with one photon at frequency ω_1 and the other photon at frequency $\omega_2 = \omega_{ba} - \omega_1$. (By analogy to the one-photon case, if the two-photon absorption rate is $\bar{n}_1\bar{n}_2 R$, then the two-photon spontaneous emission rate is R.)

▷ Derive the following expression:

$$A(\omega_1)\mathrm{d}\omega_1 = \frac{8e^4}{\pi\hbar^2 c^6}\omega_1^3\omega_2^3 \left\langle \left| \sum_f \left\{ \frac{\langle a|\mathbf{r}\cdot\hat{\mathbf{e}}_1|f\rangle\langle f|\mathbf{r}\cdot\hat{\mathbf{e}}_2|b\rangle}{\omega_1+\omega_{fb}} + \frac{\langle a|\mathbf{r}\cdot\hat{\mathbf{e}}_2|f\rangle\langle f|\mathbf{r}\cdot\hat{\mathbf{e}}_1|b\rangle}{\omega_2+\omega_{fb}} \right\} \right|^2 \right\rangle_{\mathrm{avg}} \mathrm{d}\omega_1.$$

The angle brackets indicate an average over all possible polarizations and directions of propagation for the two photons. (Don't lose too much sleep if you can't get the factor of 8 in front.)

This result was first published in 1931 by Maria Göppert-Mayer, one of the first persons to investigate multi-photon processes using the new quantum mechanics.

You don't need to work this out, but the formula above can be simplified by evaluating the average and expressing the matrix elements in terms of the operator z:

$$A(\omega_1)\mathrm{d}\omega_1 = \frac{8e^4}{3\pi\hbar^2 c^6}\omega_1^3\omega_2^3 \left| \sum_f z_{af} z_{fb} \left(\frac{1}{\omega_1+\omega_{fb}} + \frac{1}{\omega_2+\omega_{fb}} \right) \right|^2 \mathrm{d}\omega_1.$$

(G. Breit and E. Teller, Astrophysical Journal **91**, 215, (1940).)

(b) $2S$ natural lifetime.

The $2S$ state in hydrogen decays almost exclusively by spontaneous two-photon emission. (FYI: The transition to the Lamb shifted $2\,{}^2P_{1/2}$ level is allowed by electric dipole radiation, but owing to the small value of the Lamb shift, the transition probability is negligibly small, equivalent to a lifetime of 163 years. The magnetic dipole transition to the $1\,{}^2S_{1/2}$

is forbidden in the non-relativistic approximation since the radial wavefunctions of the two states are orthogonal. However, after relativistic corrections, the matrix element is no longer exactly zero, corresponding to a lifetime of about five days (see Corney, Laser Spectroscopy)).

For an order of magnitude estimate of the hydrogen $2S$ state lifetime, we can use the Göppert-Mayer formula and make the following approximations:

(i) Only the $2P$ level contributes as an intermediate state.

(ii) Use the Bohr radius a_0 for both relevant matrix elements of z.

▷ Use the total spontaneous decay rate to obtain the lifetime τ of the $2S$ state:

$$\frac{1}{\tau} = A_\tau = \frac{1}{2}\int_0^{\omega_{ba}} A(\omega_1) d\omega_1.$$

The factor $1/2$ is included since the same photon pair occurs twice when integrating from 0 to ω_{ba}.

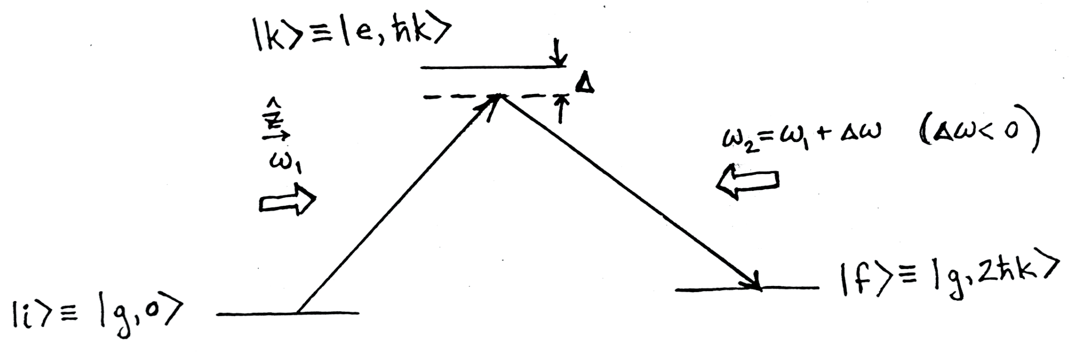
▷ Express the $2S$ lifetime in seconds.

▷ Plot the spectrum $A(\omega_1)$ over its entire range.

The actual $2S$ state lifetime is 0.122 s. The higher P states as well as the continuum contribute significantly to the $2S$ decay rate. However, it turns out to be an arduous calculation to sum over the intermediate states accurately, even for the hydrogen atom. It was nearly three decades after Göppert-Mayer did her pioneering work that the $2S$ lifetime was calculated to more than one significant digit! (J. Shapiro and G. Breit, Phys. Rev. **113**, 179 (1959).)

To my knowledge, the $2S$ lifetime has not been directly measured except in ultracold, magnetically trapped hydrogen. (C. L. Cesar *et al.*, Phys. Rev. Lett. **77**, 225 (1996).)

Problem 2. Bragg Scattering



Consider the above energy level diagram of an atom where the states are product states of an internal state and an external (momentum) state. If two counterpropagating lasers are tuned as indicated, recoil momentum will be transferred to the atoms by redistributing photons between the beams. We want to look at this “Bragg scattering” in two ways:

- by describing it as a two-photon stimulated Raman process
- by considering the mechanical effect of the AC Stark shift potential seen by an atom.

Such an arrangement is a grating for matter waves and is currently used in atom interferometers (See, for example, D. M. Giltner, R. W. McGowan, S. A. Lee, Phys. Rev. Lett. **75**, 2638 (1995)). The first observation of Bragg scattering of atoms by light was accomplished by a group at MIT: P. J. Martin, B. G. Oldaker, A. H. Miklich, D. E. Pritchard, Phys. Rev. Lett. **60**, 515 (1988).

(a) Calculate the two-photon Rabi frequency for the Raman process shown above. Assume the beams are counterpropagating along the z axis, have the same polarization, and can be expressed

$$\begin{aligned} E_1 &= E_o \cos(kz - \omega_1 t) \\ E_2 &= E_o \cos(-kz - (\omega_1 + \Delta\omega)t). \end{aligned}$$

Note that $|i\rangle$, $|k\rangle$, and $|f\rangle$, have external wavefunctions which are infinitely extended plane waves. The dipole matrix element of the internal states is D_{eg}. Also, note that only the excited state $|e, \hbar k\rangle = |k\rangle$ has nonvanishing matrix elements with the initial and final states.

$\triangleright$ What should $\Delta\omega$ be in order to realize the Raman resonance condition? It is possible to consider this an effective two level system involving only $|i\rangle$, $|f\rangle$, and the coupling between them.

$\triangleright$ If H' is the perturbation due to E_1 and E_2, what is $\langle i|H'|f\rangle$?

(b) Calculate the AC Stark shift $U(z,t)$ due to the total electric field $E_1 + E_2$. (Assume the weak-field limit holds, and average over the oscillation at optical frequencies.)

(c) What is the coupling $\langle i|U(z,t)|f\rangle$ due to the mechanical potential presented by the AC Stark shift? Compare this with the perturbation matrix element obtained in part (a).

This problem illustrates that forces due to the AC Stark effect, i.e., the stimulated light forces correspond in the photon picture to a stimulated Raman process which redistributes photons between laser beams.