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Effect of a Non-Resonant Irradiation on Atomic Energy Levels — Application to Light-Shifts in Two-Photon Spectroscopy and to Perturbation of Rydberg States

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Abstract

Atomic or molecular energy levels are shifted by a non-resonant electromagnetic irradiation. These so-called "light-shifts" must be carefully taken into account in high resolution spectroscopy experiments since they perturb the atomic Bohr frequencies which one wants to measure or to use as frequency standards.

In this paper, we present a brief survey of various types of light-shifts corresponding to different physical situations. We use a dressed atom approach allowing simple physical discussions and simple calculations. Some new suggestions are made.

1. Light Shifts Corresponding to a Very Weak Light Intensity or to a Very Broad Spectral Width

Consider first an atom irradiated by a monochromatic laser light of frequency ω_L. Let g and e be the ground and excited state of the atom, $\omega_0 = E_e - E_g$ the unperturbed atomic frequency, Γ the natural width of e (see Fig. 1).

The parameter characterizing the coupling atom-laser is the Rabi nutation frequency $\omega_1 = dE$ proportional to the product of the atomic dipole moment d by the amplitude E of the laser electric field (we take $\hbar = c = 1$).

$$\omega_1 = dE \tag{1.1}$$

The weak intensity limit corresponds to

$$\omega_1 \ll \Gamma \tag{1.2}$$

In such a case, it is justified to treat first the effect of spontaneous emission, which is equivalent to replacing the energy ω_0 of e by the complex energy

$$\omega_0 \to \omega_0 - i\frac{\Gamma}{2} \tag{1.3}$$

(We suppose the Lamb-shifts of e and g reincluded in ω_0), and then consider the effect of the coupling with the laser mode. One is therefore led to a two-level problem: state $|g, 1\rangle$ corresponding to the atom in g with one laser photon (total energy ω_L) coupled to state $|e, 0\rangle$, i.e. atom in e with no laser photon (total complex energy $\omega_0 - i(\Gamma/2)$), the coupling between the two states being $\omega_1/2$ (see Fig. 2). This means that one has to diagonalize the following 2 × 2 non-hermitian matrix:

$$\begin{pmatrix} \omega_L & \omega_1/2 \\ \omega_1/2 & \omega_0 - i\frac{\Gamma}{2} \end{pmatrix} \tag{1.4}$$

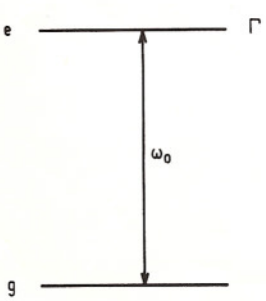

Fig. 1. Two-level atom e–g

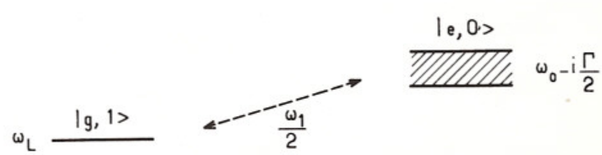

Fig. 2. Two-level problem corresponding to a weak intensity irradiation. One of the two levels, $|e, 0\rangle$, has a finite width $\Gamma/2$. The coupling is proportional to the Rabi frequency ω_1

Let $\omega_L + \Delta E' - i(\Gamma'/2)$ be the eigenvalue of (1-4) tending to ω_L when $\omega_1 \to 0$. $\Delta E'$ and Γ' represent the shift and the broadening of the ground state due to the light irradiation.

For a resonant irradiation ($\omega_L = \omega_0$), one easily finds from (1-4)

$$\omega_L = \omega_0 \Rightarrow \begin{cases} \Delta E' = 0 \\ \Gamma' = \Gamma\left(\dfrac{\omega_1}{\Gamma}\right)^2 \ll \Gamma \end{cases} \tag{1.5}$$

There is no light-shift at resonance. A small part of the instability of the excited state is transferred to the ground state. On the other hand, for a non-resonant irradiation ($|\omega_L - \omega_0| \gg \Gamma$), one gets

$$|\omega_L - \omega_0| \gg \Gamma \Rightarrow \begin{cases} \Delta E' = \dfrac{(\omega_1/2)^2}{\omega_L - \omega_0} \\ \Gamma' = \Gamma\left(\dfrac{\omega_1/2}{\omega_L - \omega_0}\right)^2 \end{cases} \tag{1.6}$$

Both $\Delta E'$ and Γ' are different from 0, but

$$\left|\frac{\Delta E'}{\Gamma'}\right| = \frac{|\omega_L - \omega_0|}{\Gamma} \gg 1 \tag{1.7}$$

Off resonance, the shift is much larger than the broadening and has the same sign as the detuning $\omega_L - \omega_0$.

These results are summarized in Figure 3 where the unperturbed energies of $|g, 1\rangle$ and $|e, 0\rangle$ are represented in dotted lines as a function of ω_L ($|e, 0\rangle$ has a width $\Gamma/2$). The perturbed level associated with $|g, 1\rangle$ is represented in full lines. The shift from the unperturbed position varies as a dispersion curve with $\omega_L - \omega_0$. The broadening of the level, suggested by the thickness of the line, is maximum for $\omega_L = \omega_0$ and varies as an absorption curve with $\omega_L - \omega_0$. The width of these absorption and dispersion curves is of the order of Γ. The level $|e, 0\rangle$ is also perturbed, but by an amount much smaller than Γ (the perturbed $|e, 0\rangle$ state is not represented).

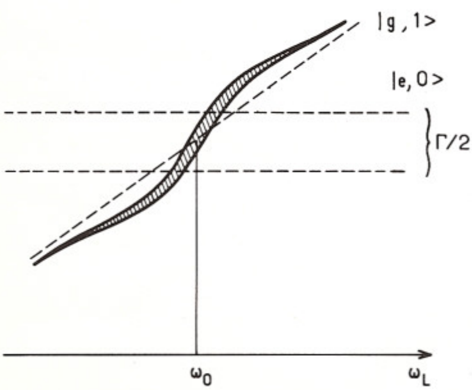

Fig. 3. Variations with ω_L of the energies of the unperturbed states $|e, 0\rangle$ and $|g, 1\rangle$ (dotted lines) and of the perturbed state corresponding to $|g, 1\rangle$ (full lines; the thickness of the line corresponds to the width of the perturbed state)

Similar results are found when, instead of a monochromatic excitation, one uses a broad line excitation with a spectral width $\Delta\nu \gg \Gamma$, provided that the mean Rabi nutation frequency

$$[\overline{\omega_1^2}]^{1/2} = [d^2 \int d\nu E^2(\nu)]^{1/2}$$

proportional to the square root of the total light intensity is small compared to $\Delta\nu$. The shift and the broadening of the ground state vary with $\overline{\omega}_L - \omega_0$ as dispersion and absorption curves with a width $\Delta\nu$ instead of Γ (see Refs. [1] and [2]). Expressions (1.5) and (1.6) remain valid provided ω_1^2 is replaced by $\overline{\omega_1^2}$ and Γ by $\Delta\nu$.

Such a situation occurs with ordinary thermal light sources which have a broad spectral width and a weak intensity. The shift of the ground state is generally much smaller than Γ, of the order of a few Hz to a few kHz. Such light-shifts have, however, been observed before the advent of lasers and have been studied in great detail (see Refs. [2] to [7]). They were measured not on optical transitions but on RF or microwave transitions connecting two ground-state sub-levels having a very small width. If the two sub-levels undergo different light-shifts, which can be achieved by a convenient choice of the light polarization, the corresponding RF or microwave transition is shifted by an amount which can be larger than the linewidth. Let us also mention a recent study of the light-shift of the 0–0 microwave transition of the ground state of ^{133}Cs induced by a CW tunable GaAs laser [8].

2. Light-Shifts Corresponding to a High Intensity

From now on we will consider a monochromatic laser excitation at frequency ω_L.

This laser light interacts with a 3-level atom a, b, c with energies E_a, E_b, E_c and two allowed electric dipole transitions ω_0, ω_0' (see Fig. 4).

The laser frequency ω_L is supposed close to ω_0, so that mainly levels a and b are perturbed. To study this perturbation, one looks at the modification of the absorption of a second weak probe laser beam with a frequency close to ω_0' (the perturbation associated with the probe will be neglected).

The intensity at ω_L is supposed sufficiently large so that the Rabi nutation frequencies

$$\omega_1 = dE \quad \omega_1' = d'E \tag{2.1}$$

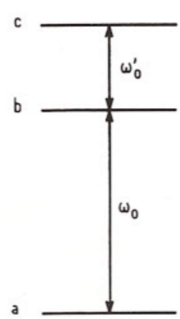

Fig. 4. Three-level atom a–b–c

where d and d' are the dipole moments of transitions ab and bc and E the ω_L laser electric field, are large compared to the natural widths of a, b, c

$$\omega_1, \omega_1' \gg \Gamma_a, \Gamma_b, \Gamma_c \tag{2.2}$$

We will first consider motionless atoms and then take into account the Doppler effect.

It will be useful to introduce energy diagrams analogous to that of Figure 3. The dotted lines of Figure 5 represent some unperturbed levels of the total system atom + ω_L photons as a function of ω_L. Only energy differences are important so that one can take as a zero of energy the energy of any of these states, $|a, n\rangle$ for example (atom in a in presence of n photons ω_L). Levels $|b, n\rangle$ and $|c, n\rangle$ are parallel to $|a, n\rangle$ and separated from $|a, n\rangle$ by distances ω_0 and $\omega_0 + \omega_0'$. Level $|a, n+1\rangle$, which contains one ω_L photon more than $|a, n\rangle$, has a slope + 1 and intersects $|a, n\rangle$ for $\omega_L = 0$.

When the coupling is neglected, the absorption frequencies of the probe are the Bohr frequencies of the diagram of Figure 5 which correspond to pairs of unperturbed levels between which the atomic dipole moment operator D has a non-zero matrix element. Since D cannot change n, one gets only the frequency ω_0' corresponding to the transition $|b, n\rangle \to |c, n\rangle$ (segment B′C′ of Fig. 5).

Levels $|a, n+1\rangle$ and $|b, n\rangle$ cross at point I for $\omega_L = \omega_0$. Since the atom in a can absorb a photon and jump to b, these two states are coupled. The coupling between them is characterized by the Rabi frequency ω_1 (see (2.1)). Since we suppose $\omega_1 \gg \Gamma_a, \Gamma_b$, the two perturbed states originating from $|a, n+1\rangle$ and $|b, n\rangle$ will not cross as in Figure 3, but will repel each

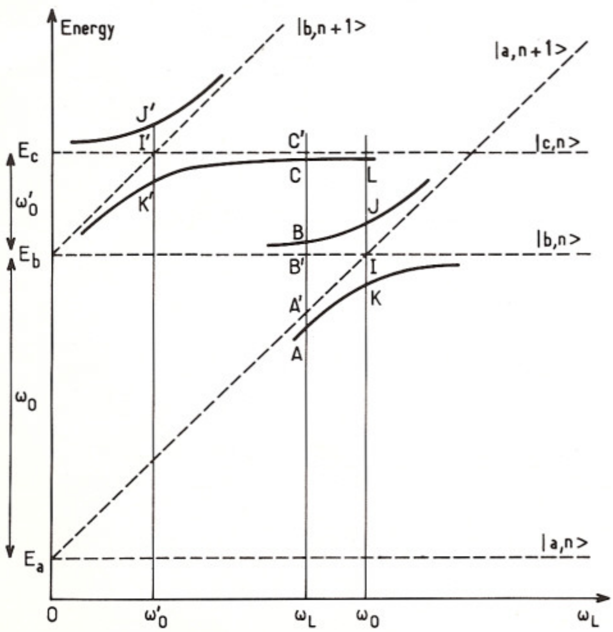

Fig. 5. Variations with ω_L of the energies of some unperturbed states of the total system atom + laser mode (dotted lines) and of some perturbed states of this system (full lines)

other and form an "anticrossing" (full lines of Fig. 5). The minimum distance KJ between the two branches of the hyperbola is nothing but ω_1 (we have not represented the width of the levels which is smaller than ω_1, according to (2.2)). Similarly, the perturbed levels originating from $|b, n+1\rangle$ and $|c, n\rangle$, which cross in I′ for $\omega_L = \omega_0'$, form an anticrossing with a minimum distance K′J′ equal to ω_1'.

The absorption frequencies of the probe laser beam now clearly appear on Figure 5.

Because of the perturbation associated with the irradiation at ω_L, one first sees that the absorption frequency $B'C' = \omega_0'$ is changed into BC. B′B and C′C are the light-shifts of b and c produced by the laser ω_L. Suppose ω_L near from ω_0 but $|\omega_L - \omega_0| \gg \omega_1$. The shift B′B can be easily calculated by perturbation theory and is found to be equal to

$$B'B = \frac{(\omega_1/2)^2}{\omega_0 - \omega_L} \tag{2.3}$$

The light-shift of b is proportional to the light intensity ($\sim\omega_1^2$) and has the same sign as $\omega_0 - \omega_L$. Similarly, one finds

$$C'C = \frac{(\omega_1'/2)^2}{\omega_0' - \omega_L} \tag{2.4}$$

which is much smaller than B′B since ω_L is nearer from ω_0 than from ω_0'. Strictly speaking, one should also take into account the coupling between $|b, n\rangle$ and $|c, n-1\rangle$ ($|c, n-1\rangle$ is not represented on Fig. 5), which slightly modifies (2.3) into

$$B'B = \frac{(\omega_1/2)^2}{\omega_0 - \omega_L} - \frac{(\omega_1'/2)^2}{\omega_0' - \omega_L} \tag{2.5}$$

Finally, the irradiation at ω_L changes the absorption frequency of the probe from ω_0' to ω_p where:

$$\omega_p = \omega_0' - \frac{(\omega_1/2)^2}{\omega_0 - \omega_L} + 2\,\frac{(\omega_1'/2)^2}{\omega_0' - \omega_L} \tag{2.6}$$

Several experiments, using high power lasers, have demonstrated the existence of light-shifts of optical transitions (Refs. 9 to 11).

One also sees on Figure 5 that a second absorption frequency appears, equal to AC. The perturbed level $|a, n+1\rangle$ contains a small admixture of $|b, n\rangle$, which allows a non-zero value of the dipole moment matrix element between $|a, n+1\rangle$ and $|c, n\rangle$. This new absorption frequency $\tilde{\omega}_p$, given by

$$\tilde{\omega}_p = \omega_0 + \omega_0' - \omega_L + \frac{(\omega_1/2)^2}{\omega_0 - \omega_L} + \frac{(\omega_1'/2)^2}{\omega_0' - \omega_L} \tag{2.7}$$

is close to $\omega_0 + \omega_0' - \omega_L$ and corresponds to a physical process where two photons, one from the laser ω_L, the other from the probe, are absorbed by the atom which jumps from a to c. The last two terms of (2.7) represent light-shift corrections to these two photon processes.

When ω_L approaches ω_0, one sees on Figure 5 that ω_p and $\tilde{\omega}_p$ transform continuously into JL and KL which are close to $\omega_0' - \omega_1/2$ and $\omega_0' + \omega_1/2$. This is the well-known Autler–Townes splitting [12] of the b–c transition due to the saturation of the a–b transition. There is therefore a close relation between light-shifts and Autler–Townes splittings [13–14]. If, $\omega_0 - \omega_L$ being fixed, one increases ω_1, the light-shift, which varies as ω_1^2 (proportional to the light intensity I) when $\omega_1 \ll |\omega_0 - \omega_L|$, varies as ω_1 (proportional to $\sqrt{I}$) when $\omega_1 \gg |\omega_0 - \omega_L|$. Such an effect has recently been observed experimentally [15].

Let us now introduce the Doppler effect. We will suppose in this section that $|\omega_0 - \omega_L| \gg \omega_1$, so that the slopes of the perturbed levels near C, B, A are respectively 0, 0, + 1. Consider an atom moving with velocity v towards the laser ω_L. In the rest frame of this atom, the laser appears to have the frequency $\omega_L + \omega_L(v/c)$ so that we have to make a translation $\omega_L(v/c)$ towards the right on Figure 5 (see also Fig. 6). The absorption frequencies of the atom in its rest frame are therefore given by the lengths of the segments B_1C_1 and A_1C_1 of Figure 6. Now, in order to calculate the absorption frequencies of the probe wave in the laboratory frame, we must correct B_1C_1 and A_1C_1 by the Doppler shifts which are, respectively, equal to $\pm v/c$ BC and $\pm v/c$ AC (the + sign must be taken if the probe and the ω_L laser are propagating in opposite directions, the − sign if they are propagating in the same direction).

From the previous discussion, one deduces the following geometrical construction. From B_1 and A_1 one draws two straight lines with slopes respectively equal to $\pm$ BC/ω_L and $\pm$ AC/ω_L (+ : counterpropagating waves, − : copropagating waves). The absorption frequencies of the probe in the laboratory frame are B_2C and A_2C (counterpropagating waves) or B_3C and A_3C (copropagating waves). This construction must be done for every value of v (we suppose the Doppler width $\Delta\nu_D$ much smaller than $\omega_L - \omega_0$, so that the slope of the perturbed levels of Figures 5 and 6 remain constant over an interval $\Delta\nu_D$ around ω_L).

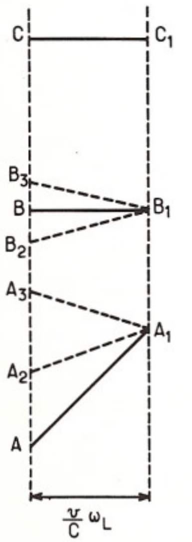

Fig. 6. Geometrical construction of the absorption frequencies of the probe wave for a given atomic velocity v

One sees on Figure 6 that the absorption frequency BC is always Doppler broadened. On the other hand, the absorption frequency A_2C (counterpropagating waves) can be Doppler free if the slope of A_2A_1 is equal to 1, in which case A_2 always coincides with A whatever v is. Since the slope of A_2A_1 is AC/ω_L, this is realized when the probe has the same frequency ω_L as the saturating laser, where ω_L is given, according to (2.7), by

$$2\omega_L = \omega_0 + \omega_0' + \frac{(\omega_1/2)^2}{\omega_0 - \omega_L} + \frac{(\omega_1'/2)^2}{\omega_0' - \omega_L} \qquad (2.8)$$

In this way we obtain the well-known principle of the Doppler-free two-photon spectroscopy [16, 17] which has recently received a great deal of attention [18]. The last two terms of (2.8) represent the light-shift corrections to the resonance frequency. These light-shifts have been studied experimentally by letting the saturating and probe frequencies be slightly different in order to make ω_L closer to ω_0 [19]. As mentioned in Ref. [17], the power broadening of the two-photon resonance is $\omega_1\omega_1'/4|\omega_0 - \omega_L|$, i.e. of the order of the light-shifts appearing in (2.8), unless one of the two Rabi frequencies ω_1 or ω_1', is much smaller than the other one. Therefore, when the two-photon resonance is not power broadened, one generally expects the light-shifts to be smaller than the width of the resonance.

We will not study the case where a, b, c are equidistant or nearly equidistant although the use of diagrams of the type of Figure 5 could be quite helpful for such a problem.

3. New Possibility of Suppressing Doppler Effect Using Ultra High Intensity Light-Shifts

For the geometrical construction of Figure 6 we have supposed that $|\omega_L - \omega_0| \gg \omega_1$, so that the slope in B is practically zero.

If $|\omega_L - \omega_0|$ is decreased, or if, $|\omega_L - \omega_0|$ being fixed, ω_1 is increased, the slope at the point B of Figure 5 is no longer zero and gets any value between 0 and 1. (Note, however, that since ω_L remains very far from ω_0', the slope in C is always zero.) Suppose now that the slope in B is adjusted to the value BC/$\omega_L \simeq \omega_0'/\omega_0$. This means that, on Figure 6, the slope of BB_1 is just equal to the slope of B_2B_1. The Doppler effect disappears on the transition bc probed by a weak laser beam counterpropagating with the laser ω_L. In physical terms, the variation of the light-shift of level b, due to the variation by Doppler effect of the detuning $\omega_L - \omega_0$, exactly cancels the Doppler effect on the probe beam.

Such a scheme supposes that one can neglect the curvature of the energy level near B over an interval of variation of ω_L of the order of the Doppler width $\Delta\nu_D$. It may be easily shown that such a condition implies for ω_1 to be of the order of, or larger than, $\Delta\nu_D$. Consequently, such a method would require ultra high light intensities which seem quite difficult to obtain at present.

Let us finally describe a slight modification of the previous idea which gives the possibility of suppressing the net light-shift BB′ at point B while multiplying by 2 the slope in B. Suppose that one uses two intense laser beams, with the same intensity, the same direction of propagation and with frequencies $\omega_L^+ = \omega_0 + \delta$ and $\omega_L^- = \omega_0 - \delta$ symmetrically distributed with respect to ω_0. The energy diagram in the neighbourhood of B has the shape represented on Figure 7. The two dotted lines with slope +1 represent states where the atom in a is in the presence of $(n+1)$ photons ω_L^+ and n photons ω_L^- or n photons ω_L^+ and $(n+1)$ photons ω_L^-. The horizontal dotted line corresponds to the atom in b in the presence of n photons ω_L^+ and n photons ω_L^-. The perturbed energy levels are represented in full lines. Symmetry considerations show that B′ is on the intermediate energy level. This means that for an atom with zero velocity, the two light-shifts produced by ω_L^+ and ω_L^- cancel. But, as soon as $v \neq 0$, one of the two frequencies gets closer to ω_0, whereas the second one moves away, so that the two light-shifts do not cancel any longer, producing a modification of the atomic energy proportional to v, which can exactly compensate the Doppler effect on the probe transition b–c. Since the curvature of the energy level is 0 in B′ (the second-order derivative is zero by symmetry), the energy level remains closer to its tangent in B′ over a larger interval and one expects to get a symmetric narrow absorption line around ω_0' on the probe beam even if ω_1 is small compared to $\Delta\nu_D$. (If $\omega_1 < \Delta\nu_D$ two other narrow lines, symmetric with respect to ω_0' and corresponding to the two other points P and Q of the energy diagram of Figure 7 where the slope is equal to ω_0'/ω_0, would appear in the absorption spectrum.)

Note that the atoms do not need to be excited in b by the lasers ω_L^+ or ω_L^-. Any other excitation process, for example a discharge, would be convenient.

The slope s at point B′ is a function of ω_1 and of the detuning $\delta = \omega_L^+ - \omega_0$. By equating s to ω_0'/ω_0, one gets the condition between ω_1 and δ which must be fulfilled in order to suppress the Doppler effect on bc. When $\omega_1 < \delta$, one finds

$$s \simeq \frac{\omega_1^2}{2\delta^2} \tag{3.1}$$

When $\omega_1 > \delta$, it is necessary to take into account other states of the system, such as b, $(n+1)\omega_L^+$, $(n-1)\omega_L^- \ldots$ but it is still possible to derive analytical expressions for s which will be given in forthcoming publications.

4. Light Shifts of Rydberg States Produced by a High Frequency Irradiation

In the previous sections we have supposed ω_L to be close to an atomic frequency ω_0, so that it was justified to keep only a finite number of atomic levels in the calculations. This is no longer possible when ω_L is quite different from any atomic frequency, for example when atoms in highly excited states (Rydberg states) interact with an intense laser beam having a frequency much higher than the spacing and the ionization energy of these states. The light-shift of the Rydberg states is, in this case, given by an infinite series of terms representing the contribution of virtual transitions to all other atomic states including the continuum.

In Ref. [20], an approximate method is used to evaluate this series and leads to simple compact expressions for the light-shift. We would now like to discuss the physical meaning of some of the corresponding corrections which could be easily observed with the presently available laser intensities.

One first finds that all Rydberg states (including the continuum) are moved by an amount $\mathcal{E}_v$ given by

$$\mathcal{E}_v = \frac{e^2E^2}{2m\omega_L^2} \tag{4.1}$$

where e and m are the electron charge and mass, E and ω_L the amplitude and frequency of the laser electric field.

The physical meaning of $\mathcal{E}_v$ is very simple. $\mathcal{E}_v$ represents the kinetic energy of the electron vibrating

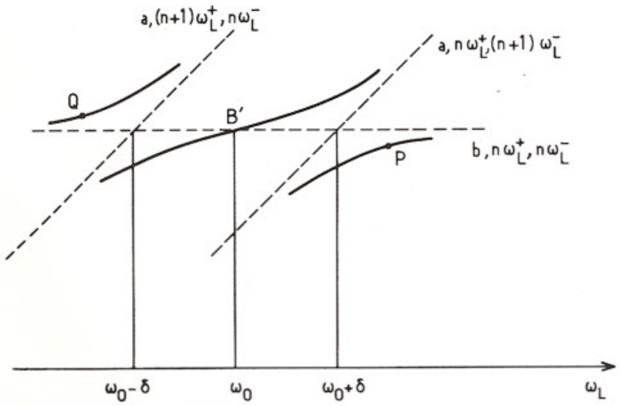

Fig. 7. Variations with ω_L of the energies of some unperturbed states of the total system atom + 2 laser modes ω_L^+, ω_L^- (dotted lines), and of the corresponding perturbed states (full lines)

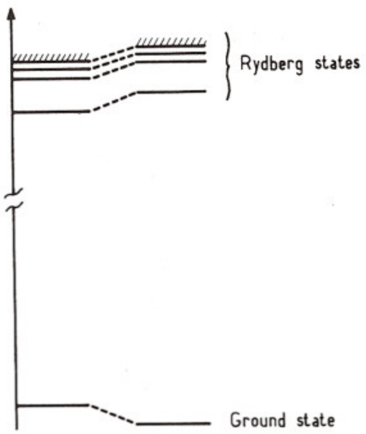

Fig. 8. Shift of the Rydberg states of an atom induced by a high frequency irradiation. The ground state is shifted by a different amount

at frequency ω_L in the laser wave. Note that such a picture of a vibrating electron is only valid for the Rydberg states for which the electron orbits around the nucleus with a frequency much lower than ω_L, so that it can be considered as quasi-free. For the ground state, this is no longer true since ω_L can be smaller than the resonance frequency, in which case the laser field polarizes the electron orbit and produces a negative light-shift (see Fig. 8).

The order of magnitude of $\mathcal{E}_v$ for focused N_2 laser delivering a flux of 1 Gw/cm^2 or for a focused CO_2 laser delivering a flux of 1 MW/cm^2 is 0.1 cm^{-1} or 3 GHz. Two-photon spectroscopy between the ground state and the Rydberg states could be used to measure the shift of the Rydberg states produced by such a laser irradiation, since $\mathcal{E}_v$ is expected to be much larger than the two-photon resonance linewidth.

The next correction given by the calculation may be written as:

$$\frac{\mathcal{E}_v}{3m\omega_L^2}\left[\frac{d^2v}{dr^2}+\frac{2}{r}\frac{dv}{dr}\right]+\frac{\mathcal{E}_v}{3m\omega_L^2}\left[\frac{d^2v}{dr^2}-\frac{1}{r}\frac{dv}{dr}\right] \times\left[\frac{3(\vec{\varepsilon}\cdot\vec{r})(\vec{\varepsilon}^*\cdot\vec{r})}{r^2}-1\right] \quad (4.2)$$

where v is the electrostatic potential which binds the electron, $\vec{\varepsilon}$ the polarization of the laser. This term has also a simple interpretation. The electron vibrates in the laser field with an amplitude $eE/m\omega_L^2$ so that is averages the electrostatic potential in a region with linear dimensions $eE/m\omega_L^2$. This explains the appearance of interaction terms proportional to the various derivatives of $v(r)$. The correction (4.2), which is split into its isotropic and anisotropic parts, can affect the relative position of the Rydberg states (different shifts for the n, l states) and remove the Zeeman degeneracy inside a given fine structure level. The order of magnitude of this correction for the laser intensities mentioned above is 40 MHz for the 10d state of Na.

Many other corrections (some of them being spin dependent) are derived in Ref. [20]. These we will not discuss but rather give the conditions of validity of the calculation.

(i) The binding energy of the Rydberg state must be much smaller than ω_L

$$|\mathcal{E}_{nl}| \ll \omega_L \quad (4.3)$$

(ii) Since ω_L is large compared to $|\mathcal{E}_{nl}|$, some photoionization processes can occur and broaden the Rydberg states. Such processes can be neglected if

$$l \gtrsim \sqrt{\frac{Ry}{\omega_L}} \quad (4.4)$$

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