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## Lamb Shift of Laser-Dressed Atomic States

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We discuss radiative corrections to an atomic two-level system subject to an intense driving laser field. It is shown that the Lamb shift of the laser-dressed states, which are the natural state basis of the combined atom-laser system, cannot be explained in terms of the Lamb shift received by the atomic bare states which is usually observed in spectroscopic experiments. In the final part, we propose an experimental scheme to measure these corrections based on the incoherent resonance fluorescence spectrum of the driven atom.

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The interaction of coherent light with matter is of cardinal interest both from a theoretical point of view as well as for applications. Thus it is not surprising that different approaches to this problem have been proposed and successfully applied. At the most fundamental level, quantum electrodynamics (QED) is one of the most accurate theories known so far [1–3]. The bound-state self-energy as predicted by QED is the dominant radiative correction in hydrogenlike systems and gives 98% of the ground-state Lamb shift in atomic hydrogen [4]. QED radiative corrections are usually evaluated with the adiabatic  $S$ -matrix formalism [5,6]. A complementary approach to the matter-light interaction based on the same foundations is quantum optics [7,8], which is especially suited for the description of time-dependent processes for which the adiabatic  $S$  matrix is of limited use. Within the quantum optical formalism, the atom-laser interaction may intuitively be understood with the help of the so-called dressed states which are defined as the eigenstates of the interacting system of light and matter [9]. A textbook application is the incoherent, inelastic resonance fluorescence spectrum of an atomic system subject to a driving laser field (see [7], Chap. 10). It is well known that the peaks of the incoherent spectrum may easily be interpreted with the help of dressed states. The incoherent fluorescence has received considerable attention in the past, both theoretically and experimentally, as it may be modified by external influences to a great extent (see [7], Chap. 10, and references therein).

In this Letter, we take advantage of ideas from quantum electrodynamics and from quantum optics to analyze radiative corrections received by laser-dressed atomic states. This constitutes the rather fundamental field-theoretic problem of the electron-to-vacuum interaction with the electron being bound to an atomic nucleus and being driven simultaneously by an additional strong external laser field [10]. From the viewpoint of QED, our setup corresponds to the strong-coupling regime of the atom to the laser field. This should be distinguished from the strong binding (Coulomb) field limit of the Lamb shift usually found in high- $Z$  ions [6], from the radiative

shifts of Volkov states [11], and from radiative corrections in modified vacuum structures such as in photonic crystals [12]. The dominant interaction in the system is the coupling of the atom to the driving laser field which gives rise to the atomic laser-dressed states. This interaction is taken into account to all orders in the atom-laser coupling within the rotating-wave approximation (RWA). Starting from the natural dressed-state basis of the system, we perturbatively calculate the self-energy corrections (as we focus on the self-energy, we will use the terms “Lamb shift” and “self-energy” interchangeably). Thus we first incorporate the strong interaction with the laser and treat the second-order shift due to the vacuum field in a second step of the calculation. In this way we find that the self-energy shift of the laser-dressed states clearly deviates in a nontrivial manner from the usual  $S$ -matrix results for atomic bare states. We further point out situations where the modified radiative corrections are also of practical relevance.

The system under consideration is a monochromatic laser field which couples near-resonantly to an electric-dipole allowed transition  $|e\rangle \leftrightarrow |g\rangle$  of a single atom. In a typical quantum optical treatment in two-level, dipole, and RWA (see [7], Chap. 10), the system Hamiltonian may be approximated as ( $\hbar = c = \epsilon_0 = 1$ )

$$\begin{aligned} \mathcal{H}_{\text{RWA}} = & \omega_g |g\rangle\langle g| + \omega_e |e\rangle\langle e| + \omega_L a_L^\dagger a_L \\ & + g_L (a_L^\dagger |g\rangle\langle e| + a_L |e\rangle\langle g|). \end{aligned} \quad (1)$$

The  $\omega_i$  ( $i = e, g$ ) are the energies of the respective atomic states,  $\omega_L$  is the frequency of the laser field,  $a_L$  ( $a_L^\dagger$ ) are photon annihilation (creation) operators for the laser field mode, and  $g_L$  is a coupling constant which includes known relativistic corrections [see [13], Eq. (27) or [14], Eq. (18)].

The driving of the external laser field gives rise to a resonance fluorescence spectrum which consists of an elastic scattering part centered at the frequency of the driving laser field and an incoherent part, which for  $\Omega_n \gg \Gamma$  (secular limit) splits up into three distinct peaks. Here,  $\Omega_n = 2g_L\sqrt{n+1}$  is the Rabi frequency of the

driven transition which depends on the number of photons  $n$  in the laser field mode and  $\Gamma$  is the decay rate of the transition. The main peak of this Mollow spectrum is again centered at the driving laser field frequency, while the two other peaks are shifted by the generalized Rabi frequency  $\pm\Omega_R^{(n)} = \pm\sqrt{\Omega_n^2 + \Delta^2}$  to higher and lower frequencies, respectively, with  $\Delta = \omega_L - \omega_R$  as the detuning of the driving laser field ( $\omega_R = \omega_e - \omega_g$ ).

The dressed states [9] are the eigenstates of the combined system of the atomic two-level system and the driving laser field in RWA and may be written as

$$|(+, n)\rangle = \cos\theta_n |e, n\rangle + \sin\theta_n |g, n+1\rangle, \quad (2a)$$

$$|(-, n)\rangle = -\sin\theta_n |e, n\rangle + \cos\theta_n |g, n+1\rangle. \quad (2b)$$

Here,  $|i, n\rangle$  ( $i \in \{e, g\}$ ) denotes the state where the atom is in the bare level  $i$  with  $n$  photons in the driving laser field mode, and  $\theta_n$  is the mixing angle defined by  $\tan(2\theta_n) = -\Omega_n/\Delta$ . The energies of these dressed states in RWA are given by  $E_{\pm, n} = (n+1/2)\omega_L + \omega_R/2 \pm \Omega_R^{(n)}/2$ . The Mollow spectrum may then be understood as originating from transitions  $|(\pm, n)\rangle \rightarrow |(\pm, n-1)\rangle$  among the dressed states. As the driving laser field discussed here is sufficiently intense, we replace  $\Omega_n$ ,  $\Omega_R^{(n)}$ , and  $\theta_n$  by their corresponding semiclassical entities  $\Omega$ ,  $\Omega_R$ , and  $\theta$  in the following discussion.

The Hamiltonian  $\mathcal{H}_R$  describes the interaction of the atom with all modes but the laser field mode, and  $\mathcal{H}_F$  describes the electromagnetic field,

$$\mathcal{H}_R = -qr \cdot \mathbf{E}_R, \quad \mathcal{H}_F = \sum_{k\lambda} \omega_k a_{k\lambda}^\dagger a_{k\lambda}. \quad (3)$$

Here,  $q$  is the physical charge of the electron ( $q^2 = 4\pi\alpha$  where  $\alpha$  is the fine-structure constant), and  $\mathbf{r}$  is the position operator. The electric field operator of the non-laser modes is given by

$$\mathbf{E}_R = \sum_{k\lambda \neq L} \sqrt{\omega_k/(2V)} \boldsymbol{\epsilon}_\lambda(\mathbf{k}) [a_{k\lambda} + a_{k\lambda}^\dagger]. \quad (4)$$

$V$  is the quantization volume,  $\boldsymbol{\epsilon}_\lambda(\mathbf{k})$  is a polarization vector, and  $\omega_k$ ,  $a_{k\lambda}$ , and  $a_{k\lambda}^\dagger$  are the frequency, the annihilation, and the creation operator of the vacuum mode with wave vector  $\mathbf{k}$  and polarization  $\lambda$ , respectively.

The second-order radiative self-energy shift arises from two terms. First, we have the non-laser-field radiation modes and resonant intermediate atomic states (treated as dressed states within the RWA)

$$\Delta C_{+, n} = -\frac{\alpha}{\pi} \ln[(Z\alpha)^{-2}] \frac{1}{m^2} [\cos^2\theta \langle \mathbf{p}^2 \rangle_e (\Omega_R + \Delta) + \sin^2\theta \langle \mathbf{p}^2 \rangle_g (\Omega_R - \Delta) + |\langle \mathbf{p} \rangle_{eg}|^2 (\Delta \cos(2\theta) + \Omega_R \cos^2(2\theta))], \quad (9a)$$

$$\Delta C_{-, n} = \frac{\alpha}{\pi} \ln[(Z\alpha)^{-2}] \frac{1}{m^2} [\cos^2\theta \langle \mathbf{p}^2 \rangle_g (\Omega_R + \Delta) + \sin^2\theta \langle \mathbf{p}^2 \rangle_e (\Omega_R - \Delta) + |\langle \mathbf{p} \rangle_{eg}|^2 (\Delta \cos(2\theta) + \Omega_R \cos^2(2\theta))]. \quad (9b)$$

Here  $\langle \mathbf{p} \rangle_{ij} = \langle i | \mathbf{p} | j \rangle$  is the dipole matrix element, and  $\langle \mathbf{p}^2 \rangle_j = \langle j | \mathbf{p}^2 | j \rangle$  is the expectation value of the square of the atomic momentum where  $|i\rangle$  and  $|j\rangle$  denote atomic bare states.

The additional shift to the high- and low-frequency Mollow sidebands  $\omega_\pm$  due to Eqs. (9a) and (9b), which we denote by  $\delta\omega_\pm$  in contrast to  $\Delta\omega_\pm$ , may be simplified to [17]

$$\Delta L_{\pm, n}^{(1)} = \langle (\pm, n) | \mathcal{H}_R (E_{\pm, n} - \mathcal{H}_{\text{res}})^{-1} \mathcal{H}_R | (\pm, n) \rangle, \quad (5a)$$

with  $\mathcal{H}_{\text{res}} = \mathcal{H}_{\text{RWA}} + \mathcal{H}_F$ . Second, we consider off-resonant intermediate states,

$$\Delta L_{\pm, n}^{(2)} = \langle (\pm, n) | \mathcal{H}_R (E_{\pm, n} - \mathcal{H}_{\text{off}})^{-1} \mathcal{H}_R | (\pm, n) \rangle, \quad (5b)$$

where  $\mathcal{H}_{\text{off}}$  is given by  $\mathcal{H}_{\text{res}}$  under the replacement  $\mathcal{H}_{\text{RWA}} \rightarrow \sum_{j \neq g, e} \omega_j |j\rangle \langle j|$ , excluding the resonant states  $|e\rangle, |g\rangle$ .

It is natural to assume that in the limit of vanishing laser intensity  $\Omega_R \rightarrow 0$  and vanishing detuning  $\Delta \rightarrow 0$ , the Lamb shift of the dressed states should be equal to the radiative shift we would expect from the usual bare-state treatment of the Lamb shift [3,6]. Indeed, neglecting the detuning and the Rabi frequency, the sum of the terms (5a) and (5b) leads to the following approximative (app) result

$$\Delta L_{+, n}^{(\text{app})} = \frac{4\alpha}{3m^2} (Z\alpha) \ln[(Z\alpha)^{-2}] \times \{\cos^2\theta \langle e | \delta^{(3)}(\mathbf{r}) | e \rangle + \sin^2\theta \langle g | \delta^{(3)}(\mathbf{r}) | g \rangle\}, \quad (6)$$

and the shift  $\Delta L_{-, n}^{(\text{app})}$  of  $\omega_-$  is obtained by replacing  $\sin\theta \leftrightarrow \cos\theta$  in the above formula ( $Z$  is the nuclear charge number, and  $m$  is the electron mass). This result may be rewritten as the expectation value  $\langle (\pm, n) | \Delta V_{\text{lamb}}(\mathbf{r}) | (\pm, n) \rangle$ , in a potential [15,16] given by  $\Delta V_{\text{lamb}}(\mathbf{r}) = 4\alpha(Z\alpha) \ln[(Z\alpha)^{-2}] \delta^{(3)}(\mathbf{r}) / (3m^2)$ .

We now investigate the corrections to the shift of the high- and low-frequency Mollow sidebands  $\Delta\omega_\pm$  due to Eq. (6) with respect to the lowest-order results

$$\omega_+ = E_{+, n} - E_{-, n-1}, \quad \omega_- = E_{-, n} - E_{+, n-1}. \quad (7)$$

We obtain

$$\Delta\omega_+ = \Delta L_{+, n}^{(\text{app})} - \Delta L_{-, n-1}^{(\text{app})} = -\frac{\Delta}{\sqrt{\Omega^2 + \Delta^2}} L_{\text{bare}}, \quad (8)$$

where  $L_{\text{bare}} = \langle e | \Delta V_{\text{lamb}} | e \rangle - \langle g | \Delta V_{\text{lamb}} | g \rangle$  is the effective Lamb shift acquired by the bare states. Also, we have  $\Delta\omega_- = -\Delta\omega_+$ .

When we keep the terms linear in  $\Omega_R$  and  $\Delta$  in evaluating the matrix elements in Eqs. (5a) and (5b), we obtain the following corrections  $\Delta C_{\pm, n}$  to the leading-order shift of the dressed states  $|(\pm, n)\rangle$  given in Eq. (6) (for a detailed derivation we refer the reader to [17]):

$$\delta\omega_{\pm} = \mp C \frac{\Omega^2}{\sqrt{\Omega^2 + \Delta^2}}, \quad C = \frac{\alpha}{\pi} \ell \frac{\langle p^2 \rangle_g + \langle p^2 \rangle_e}{m^2}. \quad (10)$$

Here,  $\ell = \ln[(Z\alpha)^{-2}]$ , and  $C$  is dimensionless.

The Mollow sidebands are thus Lamb shifted in total according to Eqs. (8) and (10) by

$$\begin{aligned} \omega_{\pm} &\rightarrow \omega_{\pm} + \Delta\omega_{\pm} + \delta\omega_{\pm} \\ &= \omega_L + \sqrt{\Omega^2 + \Delta^2} - \frac{\Delta}{\sqrt{\Omega^2 + \Delta^2}} L_{\text{bare}} - C \frac{\Omega^2}{\sqrt{\Omega^2 + \Delta^2}} \\ &= \omega_L + \sqrt{\Omega^2(1-C)^2 + (\Delta - L_{\text{bare}})^2} + \mathcal{O}(\Omega^2, \Delta^2). \end{aligned} \quad (11)$$

It is highly suggestive to interpret the approximative Lamb shift  $\Delta\omega_{\pm}$  as generating, under the square root, a specific term that effectively shifts the detuning  $\Delta$  by an amount that corresponds to the Lamb shift of the bare transition,  $(\omega_R \rightarrow \omega_R + L_{\text{bare}}) \Leftrightarrow (\Delta \rightarrow \Delta - L_{\text{bare}})$ . The appearance of the  $L_{\text{bare}}$  term is nontrivial within the dressed-state formalism, although its presence could be conjectured based on the evaluation of the detuning with allowance for the “bare” (the “usual”) Lamb shift of the transition. The shift mediated by the  $C$  term in Eq. (11) is effectively a radiative modification of the Rabi frequency.

We are led to define the *fully dressed Lamb shift* of the two Mollow sidebands as

$$\begin{aligned} \Delta^{(\text{full})} \mathcal{L}_{\pm} &= \pm \left( \sqrt{\Omega^2(1-C)^2 + (\Delta - L_{\text{bare}})^2} - \sqrt{\Omega^2 + \Delta^2} \right) \\ &\approx \Delta\omega_{\pm} + \delta\omega_{\pm}. \end{aligned} \quad (12)$$

We now turn to the experimental verification of the radiative corrections to the Mollow spectrum. A precision measurement of the Mollow spectrum is required. The atomic system under study should be described to very good accuracy by the two-level approximation. Otherwise, considerable further complications due to a multi-level formalism would arise. A further prerequisite is a frequency- and intensity-stabilized continuous-wave (cw) laser tuned to the atomic resonance to allow the system to evolve into the steady state.

We recall the explicit familiar three-peak Mollow spectrum which describes the frequency-dependent intensity spectrum of the incoherent fluorescence (secular limit),

$$S_{\text{inc}}(\omega) \approx \frac{\Gamma}{\pi} \left[ \frac{\Gamma_0 A_0^{\text{inc}}}{(\omega - \omega_L)^2 + \Gamma_0^2} + \frac{\Gamma_+ A_+}{(\omega - \omega_L - \Omega_R)^2 + \Gamma_+^2} + \frac{\Gamma_- A_-}{(\omega - \omega_L + \Omega_R)^2 + \Gamma_-^2} \right]. \quad (13)$$

Corrections beyond the secular approximation may be expressed as a series in  $\Gamma/\Omega_R$  [17]. Modifications of the Mollow spectrum due to modified decay rates such as in a squeezed vacuum [18], via quantum interferences [19] as

well as via modifications in strong driving fields with a Rabi frequency nonnegligible to that of the transition frequency [20] have been discussed in the literature.

The generalized Rabi frequency in this formula becomes

$$\Omega_R = \sqrt{\Omega^2 + \Delta^2} \rightarrow \sqrt{\Omega^2(1-C)^2 + (\Delta - L_{\text{bare}})^2}, \quad (14)$$

in order to take care of both the bare Lamb shift and the radiative shift of the Rabi frequency, and the parameters in (13) read:

$$\begin{aligned} A_0^{\text{inc}} &= \frac{\Omega^6}{4\Omega_R^2(\Omega_R^2 + \Delta^2)^2}, & A_{\pm} &= \frac{\Omega^4}{8\Omega_R^2(\Omega_R^2 + \Delta^2)}, \\ \Gamma_0 &= \Gamma \frac{\Omega^2 + 2\Delta^2}{2\Omega_R^2}, & \Gamma_{\pm} &= \Gamma \frac{3\Omega^2 + 2\Delta^2}{4\Omega_R^2}. \end{aligned} \quad (15)$$

Here,  $\Gamma$  is the decay width of the upper atomic level  $|e\rangle$  which also determines the width of the Mollow sidebands. Let us consider a situation with vanishing detuning  $\Delta$  (this implies  $\Omega = \Omega_R$ ). Further, we define the ratio  $h = \Omega/\Gamma$ . The width of the Mollow sidebands  $\Gamma_{\pm}$  is of the order of  $\Gamma$  according to (15). The radiative Rabi-frequency correction to the Mollow sidebands  $\delta\omega_{\pm}$  is of the order of  $C\Omega$  [see Eq. (10)]. We compare  $\delta\omega_{\pm}$  with the width of the Mollow sideband peak; this leads to the following order-of-magnitude estimate (“ $\sim$ ”) for the “shift-to-width” ratio  $r_1$ :

$$r_1 = \frac{\delta\omega_{\pm}}{\Gamma} \sim hC. \quad (16)$$

The Bloch-Siegert shift  $\delta_{\text{BS}}\omega_{\pm}$  (see [21]) of the dressed states is a second-order effect in the atom-laser interaction which at  $\Delta = 0$  shifts the dressed states by a frequency of the order of  $\Omega^3/\omega_L^2$  [20] (a formula valid for arbitrary detuning is contained in [17]). It is perhaps worth noting that according to [8], the Bloch-Siegert correction could therefore be interpreted as a stimulated radiative correction. The ratio  $r_2$  of the radiative shift  $\delta\omega_{\pm}$  of the generalized Rabi frequency to the Bloch-Siegert shift is

$$r_2 = \frac{\delta\omega_{\pm}}{\delta_{\text{BS}}\omega_{\pm}} \sim \frac{C\Omega}{\Omega^3/\omega_L^2} = \frac{\omega_L^2 C}{\Omega^2} = \frac{\ln[(Z\alpha)^{-2}]}{\alpha(Z\alpha)^2} h^{-2}.$$

We perform order-of-magnitude estimates based on the  $Z\alpha$  expansion [22]. The laser frequency (= atomic transition frequency) is  $\omega_L \sim (Z\alpha)^2 m$ , the decay width is  $\Gamma \sim \alpha(Z\alpha)^4 m$ , and  $C \sim \alpha(Z\alpha)^2 \ln[(Z\alpha)^{-2}]$  is defined in Eq. (10). With  $h \approx 1000$  and  $C \sim \alpha(Z\alpha)^2 \ln[(Z\alpha)^{-2}] \sim 10^{-6}$  (at  $Z = 1$ ), we obtain  $r_1 \sim 10^{-3}$  and  $r_2 \sim 10$ . A resolution of the peak of a Lorentzian to one part in  $10^3$  of its width is feasible as well as the theoretical description of the Bloch-Siegert shifts to the required accuracy [17]. Lineshape corrections [23] are much smaller in magnitude than the dressed radiative corrections themselves.

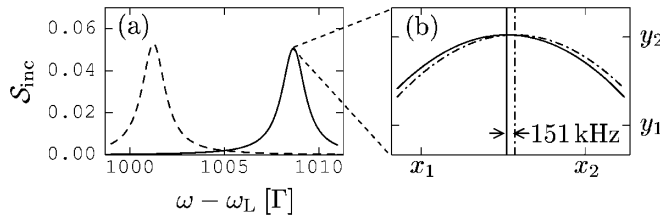


FIG. 1. One of the Mollow sidebands in Eq. (13) without any Lamb shifts (dashed line), corrected by the bare Lamb shift according to Eq. (6) (solid line), and corrected by the fully dressed Lamb shift according to Eq. (12) (dash-dotted line). (b) is a magnification of (a) with  $x_1 = 1008.64 \Gamma$ ,  $x_2 = 1008.67 \Gamma$ ,  $y_1 = 0.05062$ , and  $y_2 = 0.05068$ , where the vertical lines mark the line centers of the two Lorentzians. The parameters chosen are for the hydrogen  $1S-2P_{1/2}$  transition with  $h = 1000$ ,  $\Delta = 50\Gamma$ ,  $\Gamma = 99.8$  MHz, and  $L_{\text{bare}} = -8185.652$  MHz.

The recently developed continuous-wave (cw) Lyman- $\alpha$  source [24] was originally designed to cool antihydrogen. While not the only candidate for a prospective measurement of the laser-dressed shifts, we mention here hydrogen as a standard system for Lamb-shift measurements and the  $1S-2P$  transition as an example for a good realization of the two-level approximation. If we assume a tightly focused laser beam (limit on the beam waist is of the order of the laser wavelength  $\lambda$ ), then a calculation shows that the required Lyman- $\alpha$  power of  $340 \mu\text{W}$  for an  $h$  parameter of 1000 as in Fig. 1 is less than  $10^5$  times larger than the current maximum power of  $20 \text{ nW}$  [24]. Ionization rates are small: At intensity  $340 \mu\text{W}/(\pi\lambda^2)$ , the atom undergoes roughly  $5 \times 10^6$  Rabi oscillations before ionization [25]. Considerable progress toward the required laser power appears to be within reach for the near future [26]. These estimates are relevant for a measurement that strives to verify the radiative modification of the Rabi frequency [ $C$  term in Eq. (12)]; an experimental verification of the  $L_{\text{bare}}$  term, which requires far less experimental precision [see Fig. 1(a)], would be of considerable theoretical interest in its own right.

In summary, we find that our calculated Lamb shift of laser-dressed atomic states is nontrivially different from that obtained via conventional approximate treatments where the perturbative quantum electrodynamic interaction is evaluated prior to the exact quantum optical coupling with the laser field. The Lamb shift is modified [see Eq. (12)] even though the highly occupied laser mode, to a very good approximation (i.e., ignoring light-by-light scattering) does not interact with the other vacuum modes which are responsible for the Lamb shift. The radiative corrections amount to a change of the detuning corresponding to the Lamb shift of the bare transition, and a radiative modification of the Rabi frequency, both of which may be verified experimentally.

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