Lamb Shift of Laser-Dressed Atomic States

Ulrich D. Jentschura, Jörg Evers, Martin Haas, and Christoph H. Keitel
Fakultät für Mathematik und Physik der Albert-Ludwigs-Universität, Theoretische Quantendynamik,
Hermann-Herder-Straße 3, D-79104 Freiburg, Germany

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.

PACS numbers: 42.50.-p, 42.50.Ct, 12.20.Ds

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, 2, 3]. The bound-state self-energy as predicted by QED is the dominant radiative rate theories known so far [1, 2, 3]. The bound-state self-energy in atomic hydrogen [4]. QED correction in hydrogenlike systems and gives 98% of the energy as predicted by QED is the dominant radiative rate theories known so far [1, 2, 3]. The bound-state self-energy in atomic hydrogen [4].

Quantum electrodynamics and from quantum optics to analyze radiative corrections received by laser-dressed atomic 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.

We first incorporate the strong interaction with the laser field and treat the second-order shift due to the vacuum field in a second step of the calculation. 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 [5, Ch. 10]), the system Hamiltonian may be approximated as \(\hbar = c = \epsilon_0 = 1\)

\[
\hat{H}_{\text{RWA}} = \omega_g |g\rangle \langle g| + \omega_e |e\rangle \langle e| + \omega_L a_L^\dagger a_L
+ g_L \left( a_L^\dagger |g\rangle \langle e| + a_L |e\rangle \langle g| \right). \tag{1}
\]

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.

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, \(\Omega_n = 2 g_L \sqrt{n + 1}\) 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 $\pm \Omega_R^{(n)} = \pm \sqrt{\Omega^2 + \Delta^2}$ 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 $|\pm, n\rangle$ 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

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

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

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

$$
E_{\pm,n} = \left( n + \frac{1}{2} \right) \omega_L + \frac{1}{2} \omega_R \pm \frac{1}{2} \Omega_R^{(n)}. \quad (3)
$$

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,

$$
\mathcal{H}_R = -q \mathbf{r} \cdot \mathbf{E}_R, \quad \mathcal{H}_F = \sum_{k\lambda} \omega_k a^+_k a_k, \quad (4)
$$

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

$$
\mathbf{E}_R = \sum_{k\lambda \neq L} \sqrt{\frac{\omega_k}{2 V}} \varepsilon_\lambda(k) \begin{bmatrix} a_k & a^+_k \end{bmatrix}. \quad (5)
$$

$V$ is the quantization volume, $\varepsilon_\lambda(k)$ is a polarization vector, and $\omega_k$, $a_k$ and $a^+_k$ are the frequency, the annihilation and the creation operator of the vacuum mode with wave vector $k$ 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)

$$
\Delta L_{\pm,n}^{(1)} = \left\langle \pm, n \right| \mathcal{H}_R \left( \frac{1}{E_{\pm,n} - \mathcal{H}_{\text{res}}} \right) \mathcal{H}_R \left| \pm, n \right\rangle, \quad (6)
$$

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

$$
\Delta L_{\pm,n}^{(2)} = \left\langle \pm, n \right| \mathcal{H}_R \left( \frac{1}{E_{\pm,n} - \mathcal{H}_{\text{eff}}} \right) \mathcal{H}_R \left| \pm, n \right\rangle, \quad (7)
$$

where $\mathcal{H}_{\text{eff}}$ 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 $\delta \left(\delta E_{\text{Lamb}}\right)$. Indeed, neglecting the detuning and the Rabi frequency, the sum of the terms (9) and (7) leads to the following approximative (app) result

$$
\Delta L_{\pm,n}^{(\text{app})} = \frac{4\alpha}{3m^2} (Z\alpha) \ln[(Z\alpha)^{-2}] \times \left\{ \cos^2 \theta \langle |e\rangle \delta^{(3)}(r)|e\rangle + \sin^2 \theta \langle |g\rangle \delta^{(3)}(r)|g\rangle \right\}, \quad (8)
$$

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

$$
\Delta L_{\pm,n}^{(\text{app})} = \langle |\pm, n\rangle \mid \Delta V_{\text{Lamb}}(r) \mid |\pm, n\rangle \rangle \quad (9)
$$

with $\Delta V_{\text{Lamb}}(r) = 4\alpha (Z\alpha) \ln[(Z\alpha)^{-2}] \delta^{(3)}(r)/(3m^2)$. The expectation value vanishes of this approximative “effective Lamb shift” potential $\delta \left(\delta E_{\text{Lamb}}\right)$ vanishes for all states with angular momentum $l \geq 1 [15]$.

We now investigate the corrections to the shift of the high- and low-frequency Mollow sidebands $\Delta \omega_{\pm}$ due to Eq. (9) with respect to the lowest-order results

$$
\omega_+ = E_{+, n} - E_{-, n - 1}, \quad \omega_- = E_{-, n} - E_{+, n - 1}. \quad (10)
$$

We obtain

$$
\Delta \omega_+ = \Delta L_{+, n}^{(\text{app})} - \Delta L_{-, n - 1}^{(\text{app})} = -\frac{\Delta}{\sqrt{\Delta^2 + \Omega^2}} L_{\text{bare}} \quad (11)
$$

where $L_{\text{bare}} = \langle |e\rangle \mid \Delta V_{\text{Lamb}}(r) \mid |e\rangle - \langle g \mid \Delta V_{\text{Lamb}}(r) \mid g \rangle$ 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. (9) and (10), we obtain the following corrections $\Delta C_{\pm,n}$ to the leading-order shift of the dressed states $|\pm, n\rangle$ given in Eq. (10) [for a detailed derivation we refer the reader to [15]]:

$$
\Delta C_{+, n} = -\frac{\alpha}{\pi} \ln[(Z\alpha)^{-2}] \frac{1}{m^2} \times \left[ \cos^2 \theta \langle \mathbf{p}^2 \rangle_e (\Omega_R + \Delta) + \sin^2 \theta \langle \mathbf{p}^2 \rangle_g (\Omega_R - \Delta) \right] \quad (12a)
$$

$$
\Delta C_{-, n} = \frac{\alpha}{\pi} \ln[(Z\alpha)^{-2}] \frac{1}{m^2} \times \left[ \cos^2 \theta \langle \mathbf{p}^2 \rangle_g (\Omega_R + \Delta) + \sin^2 \theta \langle \mathbf{p}^2 \rangle_e (\Omega_R - \Delta) \right] \quad (12b)
$$
Here $\langle p \rangle_{ij} = \langle i | p | j \rangle$ is the dipole matrix element, and $\langle p^2 \rangle = \langle j | p^2 | j \rangle$ 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. (12a) and (12b), which we denote by $\delta \omega_\pm$ in contrast to $\Delta \omega_\pm$, may be simplified to 16

$$\delta \omega_\pm = \mp C \frac{\Omega^2}{\sqrt{\Omega^2 + \Delta^2}}, \quad C = \frac{\alpha}{\ell} \frac{\langle p^2 \rangle_g + \langle p^2 \rangle_e}{\ell^2}.$$  \hspace{1cm} (13)

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

The Mollow sidebands are thus Lamb shifted in total according to Eqs. (11) and (13) by

$$\omega_\pm \rightarrow \omega_\pm + \Delta \omega_\pm + \delta \omega_\pm$$ \hspace{1cm} (14)

$$= \omega_\pm + \sqrt{\Omega^2 + \Delta^2} = \frac{\Delta}{\sqrt{\Omega^2 + \Delta^2}} L_{\text{bare}} - C \frac{\Omega^2}{\sqrt{\Omega^2 + \Delta^2}}$$

$$= \omega_\pm + \sqrt{\Omega^2 (1 - C)^2 + (\Delta - L_{\text{bare}})^2} + O(\Omega^2, \Delta^2).$$

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, $(\omega_\text{R} \rightarrow \omega_\pm + L_{\text{bare}}) \Leftrightarrow (\Delta \rightarrow \Delta - L_{\text{bare}})$. This correction is therefore not a genuinely new effect; it can be obtained without invoking the dressed-state formalism and can be included after the fact in evaluating the theoretical value of the detuning with the “bare”, or in other words “usual”, Lamb shift being taken into account.

In contrast, the shift mediated by the $C$-term in Eq. (13), which is effectively a radiative modification of the Rabi frequency, cannot be obtained unless we use the dressed-state formalism. We are therefore led to define the fully dressed Lamb shift of the two Mollow sidebands as

$$\Delta^{(\text{full})} = \pm \left( \sqrt{\Omega^2 (1 - C)^2 + (\Delta - L_{\text{bare}})^2} - \sqrt{\Omega^2 + \Delta^2} \right) \approx \Delta \omega_\pm + \delta \omega_\pm.$$ \hspace{1cm} (15)

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

$$S_{\text{inc}}(\omega) \approx \frac{\Gamma}{\pi} \left[ \frac{\Gamma_0 A_{\text{inc}}^0}{(\omega - \omega)^2 + \Gamma_0^2} + \frac{\Gamma_+ A_+}{(\omega - \omega - \Omega R)^2 + \Gamma_+^2} + \frac{\Gamma_- A_-}{(\omega - \omega + \Omega R)^2 + \Gamma_-^2} \right].$$  \hspace{1cm} (16)

Corrections beyond the secular approximation may be expressed as a series in $\Gamma / \Omega R$. Modifications of the Mollow spectrum due to modified decay rates such as in a squeezed vacuum 17, via quantum interferences 18 as well as via modifications in strong driving fields with a Rabi frequency nonnegligible to that of the transition frequency 19 have been discussed in the literature.

The generalized Rabi frequency in this formula becomes

$$\Omega_R = \sqrt{\Omega^2 + \Delta^2} \rightarrow \sqrt{\Omega^2 (1 - C)^2 + (\Delta - L_{\text{bare}})^2},$$  \hspace{1cm} (17)

in order to take care of both the bare Lamb shift and the radiative shift of the Rabi frequency, and the parameters in 16 read:

$$A_{\text{inc}}^0 = \frac{\Omega^6}{4 \Omega_R^4 (\Omega_R^2 + \Delta^2)^2}, \quad A_\pm = \frac{\Omega^4}{8 \Omega_R^2 (\Omega_R^2 + \Delta^2)}, \quad \Gamma_0 = \Gamma \frac{\Omega^2 + 2 \Delta^2}{2 \Omega_R^2}, \quad \Gamma_\pm = \frac{3 \Omega^2 + 2 \Delta^2}{4 \Omega_R^2}.$$ \hspace{1cm} (18)

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 13. The radiative Rabi-frequency correction to the Mollow sidebands $\delta \omega_\pm$ is of the order of $C \Omega$ (see Eq. (13)). We compare $\delta \omega_\pm$ with the width of the Mollow sideband peak; this leads to the following order-of-magnitude estimate (“≈”) for the “shift-to-width” ratio $r_1$,

$$r_1 = \frac{\delta \omega_\pm}{\Gamma} \sim hC.$$ \hspace{1cm} (19)

The Bloch–Siegert shift $\delta_{\text{BS}} \omega_\pm$ (see 20) 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 $\Omega^2 / \omega_L^2$ 13 (a formula valid for arbitrary detuning is contained in 16). It is perhaps worth noting that according to 20, 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

$$r_2 = \frac{\delta \omega_\pm}{\delta_{\text{BS}} \omega_\pm} \sim \frac{C \Omega}{\Omega^2 / \omega_L} = \frac{\omega_L C}{\Omega^2} \frac{\ln[(Z\alpha)^{-2}]}{\alpha (Z\alpha)^2} h^{-2}.$$
We perform order-of-magnitude estimates based on the Zo-expansion [21]. The laser frequency (= atomic transition frequency) is \( \omega_L \sim (Z \alpha)^2 m \), the decay width is \( \Gamma \sim Z \alpha \), and \( C \sim Z \alpha^2 \ln[(Z \alpha)^2] \) is defined in Eq. (10). With \( h \approx 1000 \) and \( C \sim Z \alpha^2 \ln[(Z \alpha)^2] \sim 10^{-6} \) (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 \( 10^5 \) of its width is feasible as well as the theoretical description of the Bloch–Siegent shifts to the required accuracy [16].

The recently developed continuous-wave (cw) Lyman-alpha source [22] was originally designed to cool antihydrogen. We propose a measurement on the hydrogen 1S–2P transition, with hydrogen being a standard system for Lamb shift measurements and the 1S–2P transition as a very 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), then a calculation shows that the required Lyman-alpha power of 340 \( \mu \)W for an \( h \)-parameter of 1000 is less than \( 10^5 \) times larger than the current maximum power of 20 \( \mu \)W [22]. Considerable progress (roughly a factor of 10 in power) might be achieved in the near future due to enhancement resonators that are resonant to the frequencies of all three incoming laser beams contributing to the four-wave mixing process which generates the coherent Lyman-alpha light in mercury vapor [22][23].

In summary, we find that our calculated Lamb shift of laser-dressed atomic states is nontrivially different from that via conventional approximate treatments where the perturbative quantum electrodynamic interaction is evaluated prior to the exact quantum optical coupling with the laser field. The corrections are present 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. The feasibility of a measurement on the hydrogenic 1S–2P transition is discussed. Other interesting questions are related to classical analogues of two-level quantum systems where the radiative corrections discussed necessarily assume a fundamentally different form [24]; these might be the subject of forthcoming investigations.

Financial support by the German Science Foundation (SFB 276) is gratefully acknowledged.

---

[1] C. Itzykson and J. B. Zuber, Quantum Field Theory (McGraw-Hill, New York, 1980).
[2] P. J. Mohr and B. N. Taylor, Rev. Mod. Phys. 72, 351 (2000).
[3] J. Sapirstein and D. R. Yennie, Quantum Electrodynamics (World Scientific, Singapore, 1990), pp. 560–672.
[4] U. D. Jentschura, P. J. Mohr, and G. Soff, Phys. Rev. Lett. 82, 53 (1999).
[5] M. Gell-Mann and F. Low, Phys. Rev. 84, 350 (1951).
[6] P. J. Mohr, G. Plunien, and G. Soff, Phys. Rep. 293, 227 (1998).
[7] M. O. Scully and M. S. Zubairy, Quantum Optics (Cambridge University Press, Cambridge, 1997).
[8] C. Cohen-Tannoudji, J. Dupont-Roc, and G. Grynberg, Photons and Atoms – Introduction to Quantum Electrodynamics (J. Wiley & Sons, New York, 1989).
[9] C. Cohen-Tannoudji, J. Dupont-Roc, and G. Grynberg, Atom–Photon Interactions (J. Wiley & Sons, New York, 1992).
[10] C. Cohen-Tannoudji, Atoms in Strong Resonant Fields (North-Holland, Amsterdam, 1975), pp. 4–104.
[11] J. H. Eberly and H. Reiss, Phys. Rev. 145, 1035 (1966).
[12] S.-Y. Zhu, Y. Yang, H. Chen, H. Zheng and M. S. Zubairy, Phys. Rev. Lett. 84, 2136 (2000).
[13] S. G. Karshenboim, J. Phys. B 29, L29 (1996).
[14] U. D. Jentschura and I. Nandori, Phys. Rev. A 66, 022114 (2002).
[15] The Lamb shift of the 2P state, being of the order of \(-13\) MHz only, may be neglected in comparison to the Lamb shift of the 1S state, which is of the order of \(+8000\) MHz.
[16] U. D. Jentschura and C. H. Keitel, in preparation (2003).
[17] R. Loudon and P. L. Knight, J. Mod. Opt. 34, 709 (1987); H. J. Carmichael, A. S. Lane, and D. F. Walls, Phys. Rev. Lett. 58, 2539 (1987).
[18] Z. Ficek and S. Swain, J. Mod. Opt. 49, 3 (2002); J. Evers and C. H. Keitel, Phys. Rev. Lett. 89, 163601 (2002).
[19] D. E. Browne and C. H. Keitel, J. Mod. Opt. 47, 1307 (2000).
[20] F. Bloch and A. Siegent, Phys. Rev. 57, 522 (1940).
[21] H. A. Bethe and E. E. Salpeter, Quantum Mechanics of One- and Two-Electron Atoms (Springer, Berlin, 1957).
[22] K. S. E. Elkema, J. Walz, and T. W. Hänsch, Phys. Rev. Lett. 86, 5679 (2001).
[23] J. Walz and A. Pahl, private communication (2003).
[24] M. W. Beijersbergen, R. J. C. Spreeuw, L. Allen, and J. P. Woerdman, Phys. Rev. A 45, 1810 (1992).