01 Dec 2004

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Recommended Citation

J. Evers et al., "Relativistic and Radiative Corrections to the Mollow Spectrum," *Physical Review A - Atomic, Molecular, and Optical Physics*, vol. 70, no. 6, pp. 062111-1-062111-20, American Physical Society (APS), Dec 2004.
The definitive version is available at https://doi.org/10.1103/PhysRevA.70.062111

Relativistic and radiative corrections to the Mollow spectrum

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(Received 21 March 2004; published 28 December 2004)

The incoherent inelastic part of the resonance fluorescence spectrum of a laser-driven atom is known as the Mollow spectrum [B. R. Mollow, Phys. Rev. **188**, 1969 (1969)]. Starting from this level of description, we discuss theoretical foundations of high-precision spectroscopy using the resonance fluorescence light of strongly laser-driven atoms. Specifically, we evaluate the leading relativistic and radiative corrections to the Mollow spectrum, up to the relative orders of $(Z\alpha)^2$ and $\alpha(Z\alpha)^2$, respectively, and Bloch-Siegert shifts as well as stimulated radiative corrections involving off-resonant virtual states. Complete results are provided for the hydrogen $1S$-$2P_{1/2}$ and $1S$-$2P_{3/2}$ transitions; these include all relevant correction terms up to the specified order of approximation and could directly be compared to experimental data. As an application, the outcome of such experiments would allow for a sensitive test of the validity of the dressed-state basis as the natural description of the combined atom-laser system.

DOI: 10.1103/PhysRevA.70.062111 PACS number(s): 12.20.Ds, 31.30.Jv, 06.20.Jr, 31.15.−p

I. INTRODUCTION

Experimental possibilities in high-precision spectroscopy have received a rather significant "boost" in recent years due to the availability of phase coherent regularly spaced frequency combs that may bridge large frequency intervals between frequency standards and optical transition frequencies [1]. In general terms, highly accurate spectroscopy may lead to an experimental verification of known theoretical models of the physical process under study. Precision measurements—in combination with theory—allow one to obtain accurate values for physical parameters or fundamental constants [2]. With increasing accuracy, one may even ask whether the so-called constants are in fact constant [3–10]. These results may as well be used as input to more applied physics as the creation of unit standards, e.g., for time and mass.

On the theoretical side, quantum electrodynamics is one of the most accurate theories known so far. In many previous studies, the S-matrix formalism has been used to obtain accurate predictions for experimental investigations. The S matrix relates the distant past to the distant future without referring to the dynamics of the intermediate times and leads effectively to a somewhat static description [11].

Thus, the S-matrix formalism cannot, *a priori*, lead to a satisfactory description of quantum electrodynamic corrections to dynamical processes, and the subject of this paper is to provide a first step in the direction of a high-precision theoretical description of dynamical processes including radiative corrections, using a laser-driven atom as a paradigmatic example. Obviously, the treatment of radiative corrections to a dynamically driven atomic transition requires input from two different areas, which are laser physics and quantum electrodynamics. While the two areas are related, there are a couple of subtle points to consider when a unified understanding of a specific problem is sought, whose nature inevitably requires concepts introduced within the context of either of the two areas. In particular, it is known that the description of dynamical processes requires considerable care in the treatment of the gauge dependence of amplitudes, and with regard to the physical interpretation of the wave functions used in the mathematical description [12–21].

A classic textbook example for a dynamical atom-laser system, well known in theoretical quantum optics [22], consists of the Jaynes-Cummings model of an atom that contains two relevant energy levels interacting with a single monochromatic laser-field mode [23]. Due to the driving of the laser field, the atomic population undergoes Rabi oscillations. The population is driven periodically from the upper to the lower state and vice versa. The emission spectrum of this process with a strong driving field is known as the Mollow spectrum [24]. This case of strong driving may easily be interpreted in terms of the so-called dressed states. Laser-dressed states are defined as the eigenstates of the combined system of atom and driving-laser field [25] and have proven to be useful in countless cases of both theory and experiment, one of which is the Autler-Townes splitting [26].

When evaluating radiative corrections to the Mollow spectrum, it is natural to start from the dressed-state basis, which consists of the natural eigenstates of the (strongly) coupled atom-laser system rather than the bare atomic states. It might be assumed that in order to fully treat the Lamb shift of laser-dressed states, it would be sufficient to simply correct the energies of the bare states that enter into the formulas for the generalized Rabi frequencies by the "bare-state" (i.e., the usual, ordinary) Lamb shift. Indeed, the first investigations on the problem [27] revealed corrections to the dressed-state "quasienergies" consistent with this assumption. However, recently, it was found that at nonvanishing detuning and Rabi frequency, the Lamb shift of dressed

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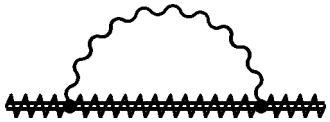

FIG. 1. Diagrammatic representation of a radiative self-energy correction to the laser-dressed atomic state. The double line corresponds to the electron bound by the nuclear Coulomb field. The jagged line denotes the additional dressing of the bound electron by the (strong) laser field. The self-energy of a laser-dressed Coulomb-bound electron is a quantum-field theoretic problem in the presence of two classical background fields.

states is nontrivially different from the bare-Lamb shift [28,29]. Thus the distinction between evaluations in terms of the bare- and the dressed-state basis in fact has to be made. In the limit of vanishing detuning, the coincidence of the bare and dressed-Lamb-shift effects on the detuning is obtained only after a summation of a specific series whose leading correction term may be obtained by carrying out the calculation to second order in the atom-field interaction. A diagrammatic representation of a radiative correction to the dressed state is shown in Fig. 1.

Thus in this paper we present a detailed and complete theoretical analysis of the leading nonrelativistic and relativistic corrections to the Mollow spectrum, up to the relative orders of $(Z\alpha)^2$ and $\alpha(Z\alpha)^2$, respectively, and of Bloch-Siegert shifts as well as stimulated radiative corrections involving off-resonant virtual states, and laser-field configuration-dependent corrections. The purpose is to enable a direct comparison between theory and experiment for a high-precision spectroscopic investigation involving laser-dressed states. Such a comparison to experimental data would allow one to address questions related to the physical reality of the dressed states (and their "quasienergy") on the one hand, and of the nature and the interpretation of the various radiative corrections on the other hand. As a promising candidate for the experiment, we identify the resonance fluorescence spectrum of a strongly driven hydrogen $1S$-$2P$ transition, which to lowest order may be described by the standard Mollow spectrum. A coherent Lyman-α source [30,31] has recently become available as a driving field, and we show that ionization into the continuum does not prohibit an experimental implementation. In particular, we discuss corrections which are due to resonant and off-resonant excitations as well as the Bloch–Siegert shift, and corrections to the transition dipole moment and to the secular approximation leading to the Mollow spectrum. As a result, we provide theoretical predictions which are directly comparable to possible experimental data.

The paper is organized as follows. In Sec. II we introduce our system of interest and provide the relevant theoretical background for the further analysis. In Sec. III we evaluate the corrections to the Mollow spectrum, which we divide into modifications of the detuning (Sec. III A) and the Rabi frequency (Sec. III B). The dominant relativistic corrections are of the order of $(Z\alpha)^2$, where Z is the nuclear charge number, and leading radiative effects lead to correction terms of the order of $\alpha(Z\alpha)^2 \ln[(Z\alpha)^{-2}]$. In Sec. IV we provide numerical data for the hydrogen $1S$-$2P_j$ transitions $\left(j=\frac{1}{2},\frac{3}{2}\right)$. Section V discusses and summarizes the results.

II. MOLLOW SPECTRUM

In this section, we introduce our system and recall results of previous studies which will serve as the basis of our analysis. Throughout the calculations, we adopt natural units with $\hbar=\epsilon_0=c=1$. The electron mass is denoted by m. We make use of the Einstein summation convention unless stated otherwise, and we employ the length gauge for all wave functions and operators as we deal with off-resonant excitations, as it is done in most of the literature, and in textbooks on the subject (see, e.g., [22]). The Mollow spectrum contains the incoherent, inelastic part of the atomic fluorescence, i.e., the fluorescence spectrum mediated by the many-photon processes whose intensity dominates over the elastic part in a strongly driven atom-laser system. In a purely quantum electrodynamic formalism, the description of many-photon processes would require perturbation theory in exceedingly high orders. However, as is well known, the description using dressed states [25] allows for a considerable simplification, as the formulas for the Mollow spectrum [24] follow rather naturally in terms of transitions among the dressed-atomic states which incorporate the atom-laser interaction to all orders in perturbation theory.

Before we now start with the discussion of the Mollow spectrum, a slight detour on questions related to gauge transformations of the laser-atom interaction is in order. The "length gauge" means that the laser-atom interaction is formulated in terms of the interaction $-q\boldsymbol{E}_{\mathbf{L}}\cdot\boldsymbol{x}$, where q is the physical electron charge and $\boldsymbol{E}_{\mathbf{L}}$ is the (gauge invariant, observable) laser-field strength. Instead, in the "velocity gauge," the interaction is formulated as $-q\boldsymbol{A}_{\mathbf{L}}\cdot\boldsymbol{p}/m$, where $\boldsymbol{A}_{\mathbf{L}}$ is a gauge-dependent suitable vector potential for the laser field. In the velocity gauge, of course, one also has to add the $\boldsymbol{A}_{\mathbf{L}}^2$ term, but dipole interactions are mediated exclusively by the "$\boldsymbol{A}_{\mathbf{L}}\cdot\boldsymbol{p}$" interaction. Due to gauge invariance, the two possible gauges are equivalent, *provided* that the gauge transformation of the wave function is properly taken into account [22,32]. In order to avoid confusion, we stress here the absolute necessity of considering the gauge transformation of the wave function in dynamical processes [12–21]. According to [13,21], the usual physical interpretation of a Schrödinger wave function is only conserved if the length gauge is used for the description of the atom-laser interaction. For dynamical processes, the velocity gauge leads to many more terms in intermediate steps of the calculation than the length-gauge formulation chosen here, due to the necessity of applying the gauge transformation to the wave function. Nevertheless, we would like to mention the possibility of an independent verification of our derivation, as presented here, in different gauges. In this case, the gauge transformation of the wave function should be applied already on the level of quantum mechanics (i.e., on the level of the Mollow spectrum as discussed in the current section), not just to the quantum electrodynamic corrections discussed in the following sections. This concludes our detour regarding gauge transformations.

We recall from [24,28,29] that the incoherent resonance fluorescence spectrum of atoms driven by a monochromatic coherent laser field may be expressed as

$$\mathcal{S}_{\rm inc}(\omega) = \frac{\Gamma}{\pi}\frac{2\Gamma^2+\Omega^2+2(\omega-\omega_{\rm L})^2}{\Gamma^2+2\Omega^2+4\Delta^2} \times \frac{4\Gamma\Omega^4}{X_0+X_2\Gamma^2+X_4\Gamma^4+X_6\Gamma^6}, \tag{2.1}$$

where

$$X_0 = 16[\Delta^2+\Omega^2-(\omega-\omega_{\rm L})^2]^2(\omega-\omega_{\rm L})^2, \tag{2.2a}$$

$$X_2 = 4[6(\omega-\omega_{\rm L})^4-2(3\Delta^2-\Omega^2)(\omega-\omega_{\rm L})^2+(2\Delta^2+\Omega^2)^2], \tag{2.2b}$$

$$X_4 = 8\Delta^2+4\Omega^2+9(\omega-\omega_{\rm L})^2, \tag{2.2c}$$

$$X_6 = 1. \tag{2.2d}$$

Here, Ω is the Rabi frequency

$$\Omega = -q\langle e|\boldsymbol{x}\cdot\boldsymbol{\epsilon}_{\rm L}|g\rangle\mathcal{E}_{\rm L} \tag{2.3}$$

of the driving-laser field $\boldsymbol{E}_{\rm L}(t)=\mathcal{E}_{\rm L}\boldsymbol{\epsilon}_{\rm L}\cos(\omega_{\rm L}t)$ with frequency $\omega_{\rm L}$, macroscopic classical amplitude $\mathcal{E}_{\rm L}$, and polarization $\boldsymbol{\epsilon}_{\rm L}$. $q=-|q|$ is the electron charge. $\Delta=\omega_{\rm L}-\omega_{eg}$ is the detuning of the laser-field frequency from the atomic transition frequency ω_{eg}, and Γ is the spontaneous decay rate of the atomic transition. The excited and the ground state of the laser-driven transition are denoted by $|e\rangle$ and $|g\rangle$, respectively, and $\boldsymbol{x}$ is the position operator vector. In secular approximation $\Omega\gg\Gamma$, this expression simplifies to

$$\mathcal{S}_{\rm inc}(\omega) \approx \frac{\Gamma}{\pi}\left[\frac{\Gamma_0 A_0}{(\omega-\omega_{\rm L})^2+\Gamma_0^2}+\frac{\Gamma_+ A_+}{(\omega-\omega_{\rm L}-\Omega_{\rm R})^2+\Gamma_+^2} + \frac{\Gamma_- A_-}{(\omega-\omega_{\rm L}+\Omega_{\rm R})^2+\Gamma_-^2}\right]. \tag{2.4}$$

Here, the separation of the Mollow spectrum into one central peak located at $\omega=\omega_{\rm L}$ and into two sidebands shifted by the generalized Rabi frequency $\Omega_{\rm R}=\sqrt{\Delta^2+\Omega^2}$ may easily be seen. The amplitudes and widths are given by

$$A_0 = \frac{\Omega^6}{4\Omega_{\rm R}^2(\Omega_{\rm R}^2+\Delta^2)^2}, \tag{2.5a}$$

$$A_\pm = \frac{\Omega^4}{8\Omega_{\rm R}^2(\Omega_{\rm R}^2+\Delta^2)}, \tag{2.5b}$$

$$\Gamma_0 = \Gamma\frac{\Omega^2+2\Delta^2}{2\Omega_{\rm R}^2}, \tag{2.5c}$$

$$\Gamma_\pm = \Gamma\frac{3\Omega^2+2\Delta^2}{4\Omega_{\rm R}^2}. \tag{2.5d}$$

The approximate form (2.4) does not represent the positions of the sideband peaks accurately in cases where $\Gamma/\Omega_{\rm R}$ is not small. Indeed, the position $\omega_\pm$ of the sideband peaks may be expanded in a series in powers of $\Gamma/\Omega_{\rm R}$ whose first terms read

$$\omega_\pm = \omega_{\rm L}\pm\Omega_{\rm R}\left[1-\frac{4+y^2}{8(1+y^2)}\left(\frac{\Gamma}{\Omega_{\rm R}}\right)^2-\frac{70+8y^2+y^4}{128(1+y^2)^2}\left(\frac{\Gamma}{\Omega_{\rm R}}\right)^4 + O\left(\frac{\Gamma}{\Omega_{\rm R}}\right)^6\right], \tag{2.6}$$

with $y=\Delta/\Omega$. For vanishing detuning $\Delta=0$, which implies $\Omega_{\rm R}=\Omega$, Eq. (2.6) specializes to

$$\omega_\pm = \omega_{\rm L}+\Omega\left[1-\frac{1}{2}\left(\frac{\Gamma}{\Omega}\right)^2-\frac{35}{64}\left(\frac{\Gamma}{\Omega}\right)^4+O\left(\frac{\Gamma}{\Omega}\right)^6\right]. \tag{2.7}$$

The correction terms move the sideband peaks closer to the central maximum.

The above results in secular approximation may easily be interpreted with the help of the so-called dressed states, which are defined as the eigenstates of the interaction part of the Hamiltonian. Under the influence of the external driving field, the atomic states are no longer eigenstates of the Hamiltonian, but rather have to be combined with the driving-laser field to give the new eigenstates. To show the precise composition of the dressed states, we use the quantum representations of the Rabi frequency

$$\Omega_n = 2\,g_{\rm L}\sqrt{n+1}, \tag{2.8}$$

the mixing angle θ_n defined by

$$\tan(2\theta_n) = -\Omega_n/\Delta, \tag{2.9}$$

and the generalized Rabi frequency

$$\Omega_{\rm R}^{(n)} = \sqrt{\Omega_n^2+\Delta^2} \tag{2.10}$$

rather than the corresponding classical entities. Here, n is the number of photons in the laser-field mode, and the coupling constant $g_{\rm L}$ for the interaction of the driving-laser field with the main atomic transition is defined by

$$g_{\rm L} = -q\langle g|\boldsymbol{\epsilon}_{\rm L}\cdot\boldsymbol{x}|e\rangle\mathcal{E}_{\rm L}^{(\gamma)}, \tag{2.11}$$

where $\mathcal{E}_{\rm L}^{(\gamma)}=\sqrt{\omega_{\rm L}/2V}$ is the electric laser field per photon and V is the quantization volume. The matching of the electric field per photon with the corresponding classical macroscopic electric field $\mathcal{E}_{\rm L}$ is given by

$$2\sqrt{n+1}\mathcal{E}_{\rm L}^{(\gamma)} \leftrightarrow \mathcal{E}_{\rm L}. \tag{2.12}$$

Throughout this paper, we will sometimes refer to the quantum description during the derivations, but use the classical entities in the final results. We may switch between the two descriptions as the driving laser field is assumed to be intense in our analysis. The matching of the quantum and the classical entities is possible with the help of the following list of replacements:

$$\Omega_n \leftrightarrow \Omega, \tag{2.13a}$$

$$\theta_n \leftrightarrow \theta, \tag{2.13b}$$

$$n+1 \approx n. \tag{2.13c}$$

Using this notation, the dressed states are given by

$$|(+,n)\rangle = \cos\,\theta_n|e,n\rangle + \sin\,\theta_n|g,n+1\rangle, \tag{2.14a}$$

$$|(-,n)\rangle = -\sin\,\theta_n|e,n\rangle + \cos\,\theta_n|g,n+1\rangle. \tag{2.14b}$$

Here, $|i,n\rangle$ ($i \in \{e,g\}$) are combined atom-field states where the atom is in state i with n photons in the driving-field mode. The energies of these dressed states are given by $E_{\pm,n}=\left(n+\frac{1}{2}\right)\omega_{\rm L}+\omega_{eg}/2\pm\Omega_{\rm R}^{(n)}/2$, where the splitting between the two dressed states $|(\pm,n)\rangle$ is known as the ac Stark shift. The various spectral components then arise from transitions $|(\pm,n)\rangle \to |(\pm,n-1)\rangle$. The transitions $+\to+$ and $-\to-$ yield the central Mollow component and the coherent elastic peak, while the transitions $+\to-$ and $-\to+$ yield the sidebands shifted to higher and lower frequencies, respectively.

III. CALCULATION OF RELATIVISTIC AND RADIATIVE CORRECTIONS TO THE MOLLOW SPECTRUM

In the following, we discuss corrections whose understanding is essential for the additional relativistic and radiative energy shifts received by the dressed states. First, we evaluate corrections which may be incorporated in a redefinition of the detuning of the driving-laser field to the atomic transition frequency; in the second part, we complete the analysis by considering corrections which effectively modify the Rabi frequency. Throughout the analysis, we focus on the hydrogen $1S$-$2P$ transition as a promising candidate for a possible experiment.

In Sec. II we have employed a purely nonrelativistic theory. Both the resonance frequency as well as the transition dipole moments are evaluated first for the nonrelativistic (Schrödinger) case. However, in order to resolve radiative effects, it is necessary to include the relativistic shifts of the transitions in a unified theory, and to analyze the fine structure. The nonrelativistic expressions for the transition dipole moments also change once we resolve the fine-structure levels, because the angular momentum algebra is augmented by the spin.

A. Corrections to the detuning

1. Relativistic corrections to the resonance frequency

The well-known relativistic correction to the hydrogen energy levels is given by

$$H_{\rm rel} = -\frac{\boldsymbol{p}^4}{8m^3} + \frac{\pi Z\alpha}{2m^2}\delta(\boldsymbol{r}) + \frac{Z\alpha}{4m^2r^3}\boldsymbol{\sigma}\cdot\boldsymbol{L}. \tag{3.1}$$

The effects are of the order of $(Z\alpha)^4 m$, whereas the Schrödinger energy is of the order of $(Z\alpha)^2 m$. The full expression for the Dirac energy of a hydrogenic level with quantum numbers n,j, is [[33], Eq. (2-87)]

$$E_{nj} = m - \frac{(Z\alpha)^2 m}{2n^2} - \frac{(Z\alpha)^4 m}{n^3}\left[\frac{1}{2j+1} - \frac{3}{8n}\right], \tag{3.2}$$

where we neglect terms of order $(Z\alpha)^6$. When evaluating the expectation values of $H_{\rm rel}$ on the dressed states (2.14) in first-order perturbation theory, the following expression results:

$$\delta\omega_{\pm,j}^{(\rm rel)} = \mp\frac{\Delta}{\sqrt{\Omega^2+\Delta^2}}E_{\rm rel}^{(j)}. \tag{3.3}$$

Here

$$E_{\rm rel}^{(1/2)} = \langle 2P_{1/2}|H_{\rm rel}|2P_{1/2}\rangle - \langle 1S_{1/2}|H_{\rm rel}|1S_{1/2}\rangle, \tag{3.4a}$$

and

$$E_{\rm rel}^{(3/2)} = \langle 2P_{3/2}|H_{\rm rel}|2P_{3/2}\rangle - \langle 1S_{1/2}|H_{\rm rel}|1S_{1/2}\rangle. \tag{3.4b}$$

The expression for $\delta\omega_{\pm,j}^{(\rm rel)}$ finds a natural interpretation as a first-order (in $E_{\rm rel}^{(j)}$) correction to the quantity

$$\sqrt{\Omega^2+(\Delta-E_{\rm rel}^{(j)})^2} = \sqrt{\Omega^2+\Delta^2} - \frac{\Delta}{\sqrt{\Omega^2+\Delta^2}}E_{\rm rel}^{(j)} + \cdots. \tag{3.5}$$

We can thus formally define a "summed" relativistic shift of the Mollow sidebands as

$$\Delta\overline{\omega}_{\pm,j}^{(\rm rel)} = \pm\left[\sqrt{\Omega^2+(\Delta-E_{\rm rel}^{(j)})^2} - \sqrt{\Omega^2+\Delta^2}\right]. \tag{3.6}$$

We recall that the detuning has been defined as $\Delta=\omega_{\rm L}-\omega_{eg}$ in Sec. II. If the fine structure is included, the resonance frequency becomes j dependent. The shift of the detuning as given in Eq. (3.6) is thus equivalent to a modification of the resonance frequency according to

$$\omega_{eg} \to \omega_{eg}^{(j)} \equiv \omega_{eg} + E_{\rm rel}^{(j)}, \tag{3.7a}$$

$$\Delta \to \Delta - E_{\rm rel}^{(j)}. \tag{3.7b}$$

Thus, the summed shift of the detuning due to the relativistic correction $E_{\rm rel}^{(j)}$, evaluated using the dressed-state basis, is equivalent to the shift of the detuning that would have been obtained if we had evaluated the detuning, right from the start, with a resonance frequency corrected by the relativistic effects. The "summation" implied by Eq. (3.6) thus finds a natural interpretation.

Throughout the calculations, we will refer to shifts of the Mollow sidebands $\omega_\pm$ due to first-order perturbations as $\delta\omega_\pm$ [see, e.g., Eq. (3.3)], whereas summed expressions like Eq. (3.6) will be denoted $\delta\overline{\omega}_\pm$.

2. Bare Lamb shift

In addition to the relativistic shifts, the positions of the sidebands have to be modified further if one desires a numerical accuracy as required to appropriately model current high-precision spectroscopy experiments. In [28,29], the second-order radiative self-energy corrections due to the interaction of the combined system of atom and driving-laser field with the surrounding nonlaser-field vacuum modes was analyzed. Taking into account both interactions of the atom-field system with resonant and off-resonant intermediate states, in the limit $\Delta,\Omega \ll \omega_{eg}$ (i.e., under the replacements $\omega_{\rm L}\to\omega_{eg}$, $\omega_{eg}-\omega_{\rm L}\pm\Omega_{\rm R}\to 0$, and $\omega_{\rm L}+\omega_{eg}\pm\Omega_{\rm R}\to 2\omega_{eg}$), we obtain corrections to the energy of the dressed states which yield an additional shift of the position of the sidebands given by

$$\delta\omega_{\pm,j}^{(\mathrm{Lamb})} = \mp \frac{\Delta}{\sqrt{\Omega^2+\Delta^2}} L_{\mathrm{bare}}^{(j)}. \tag{3.8}$$

Here, the prefactor arises from the mixing coefficients $\cos\theta$ and $\sin\theta$, and $L_{\mathrm{bare}}^{(j)}$ is the usual Lamb shift of the atomic bare-state transition frequency which for the hydrogen $1S$-$2P$ transition is given by

$$L_{\mathrm{bare}}^{(j)} = L_{2P_j} - L_{1S}, \tag{3.9}$$

where $j=\frac{1}{2},\frac{3}{2}$ is the total angular momentum quantum number of the excited state [for the definition of L_{nl_j} see also Eq. (3.16) below]. The Lamb shifts of the individual states are given by [34,35]

$$L_{1S} = 8172\,811(32)\ \mathrm{kHz}, \tag{3.10}$$

$$L_{2P_{1/2}} = -12\,835.99(8)\ \mathrm{kHz}, \tag{3.11}$$

$$L_{2P_{3/2}} = 12\,517.46(8)\ \mathrm{kHz}. \tag{3.12}$$

The correction may be interpreted physically by defining the dressed summed Lamb shift $\delta\overline{\omega}_{\pm,j}$ as

$$\delta\overline{\omega}_{\pm,j}^{(\mathrm{Lamb})} = \pm\left[\sqrt{\Omega^2+(\Delta-L_{\mathrm{bare}}^{(j)})^2} - \sqrt{\Omega^2+\Delta^2}\right], \tag{3.13}$$

where to first order in $L_{\mathrm{bare}}^{(j)}$, one recovers Eq. (3.8). Thus the correction $\delta\omega_{\pm,j}^{(\mathrm{Lamb})}$ effectively is a shift

$$\Delta \to \Delta - L_{\mathrm{bare}}^{(j)} \tag{3.14}$$

of the detuning [in analogy to Eq. (3.7b)].

With typical parameters (see Sec. IV), the summed expression $\delta\overline{\omega}_{\pm}^{(j)}$ yields results which significantly differ from the first-order expression $\delta\omega_{\pm,j}^{(\mathrm{Lamb})}$. The reason is that the bare Lamb shift is not small as compared to the detuning Δ, so that the higher-order terms of the series expansion are relevant. Nevertheless, we use the summed formula Eq. (3.13) instead of Eq. (3.8), as it is the expected result in the sense that the Lamb shift is naturally interpreted as a modification of the transition frequency and a corresponding alteration of the detuning.

3. Unified expressions for the relativistic and radiative shifts

Both the summed relativistic shift Eq. (3.6), as well as the summed Lamb shift Eq. (3.13), are effectively summarizing the corrections received by the detuning due to various shifts that go beyond the nonrelativistic treatment of the hydrogen (and Mollow) spectrum discussed in Sec. II. These effects would also be observable in low-intensity scattering of (laser) light off atoms, and are automatically included in the observable resonance frequency of the transition whose high-intensity behavior we are studying. The corrections can therefore be included into the formalism if we replace the detuning Δ by the detuning Δ_{expt} to the experimental transition frequency given by

$$\Delta_{\mathrm{expt}} = \omega_{\mathrm{L}} - \omega_{\mathrm{expt}}, \tag{3.15}$$

where ω_{expt} is the experimentally observable transition frequency as it would be obtained from low-intensity scattering [36]. In Sec. II, we have started from a nonrelativistic theory, and therefore the detuning $\Delta=\omega_{\mathrm{L}}-\omega_{eg}$ was calculated with regard to the inaccurate resonance frequency ω_{eg} as it follows from the Schrödinger theory that fails, as is well known, to describe even the relativistic effects that lead to the fine structure (let alone the Lamb shift). Thus, in practice, the bare Lamb shift modification to the detuning may be accounted for by replacing the resonance frequency ω_{eg} as it would be obtained from a nonrelativistic theory, by an experimental value for the atomic transition frequency as found in low-intensity scattering experiments [36].

The frequency ω_{expt} may not be known well enough for any given transition to lead to a meaningful comparison between theory and a conceivable high-accuracy measurement of the Mollow spectrum. This is because we are sensitive, in the measurement of the Mollow spectrum, to tiny differences between the laser frequency and the actual resonance frequency. It may therefore be useful to recall that for the Lamb shift L_{nl_j} of a hydrogenic energy level (spectroscopic notation nl_j), one may use the implicit definition (see, e.g., [34,37]),

$$E(nl_j) = m_r[f(n,j)-1] - \frac{m_r^2}{2(m+m_N)}[f(n,j)-1]^2 + L_{nl_j} + E_{\mathrm{hfs}}, \tag{3.16}$$

where E is the energy level of the two-body system, $f(n,j)$ is the dimensionless Dirac energy, m is the electron mass, m_r is the reduced mass of the system, and m_N is the nuclear mass. In very accurate experiments, one also has to include the hyperfine frequency shift E_{hfs} which depends on the quantum number m_F that includes the nuclear spin. Note, however, that the hyperfine structure does not contribute to the Lamb shift according to the definition Eq. (3.16).

The expression Eq. (3.16) can be used to make a theoretical prediction ω_{th} for the transition frequency by forming the difference of this expression for the two states involved in the atomic transition whose high-intensity behavior we are studying. The detuning can then alternatively be evaluated as $\Delta_{\mathrm{th}}=\omega_{\mathrm{L}}-\omega_{\mathrm{th}}$. Assuming $\omega_{\mathrm{th}}=\omega_{\mathrm{expt}}$, one then has $\Delta_{\mathrm{exp}}=\Delta_{\mathrm{th}}$. For some recent data on Lamb shifts, we refer to [35].

4. Bloch–Siegert shifts

The Mollow spectrum also receives corrections due to so-called counterrotating interactions of the driving-laser field with the atom [38]. These correspond to an excitation of the atom simultaneously with a creation of a laser photon or the vice versa process. The first-order perturbation vanishes, and the second-order expression is given by [29]

$$\begin{aligned}\Delta E_{\pm}^{(\mathrm{BS})} &= \pm\frac{\Omega^2}{\omega_{\mathrm{L}}}\frac{8\cos(2\theta)-(\Omega_{\mathrm{R}}/\omega_{\mathrm{L}})[3+\cos(4\theta)]}{64-16(\Omega_{\mathrm{R}}/\omega_{\mathrm{L}})^2}\\ &= \pm\frac{1}{8}\frac{\Omega^2}{\sqrt{\Omega^2+\Delta^2}}\frac{2\Delta^2+\Omega^2+4\Delta\omega_{\mathrm{L}}}{\Delta^2+\Omega^2-4\omega_{\mathrm{L}}^2}.\end{aligned} \tag{3.17}$$

The correction $\delta\omega_{\pm}^{(\mathrm{BS})}$ of the Mollow sidebands due to the Bloch-Siegert shift is thus given by

$$\delta\omega_{\pm}^{(\mathrm{BS})} = \Delta E_{\pm}^{(\mathrm{BS})} - \Delta E_{\mp}^{(\mathrm{BS})} \tag{3.18}$$

$$= \mp \frac{\Delta}{\sqrt{\Omega^2+\Delta^2}}[\mathcal{B}\Omega^2]. \tag{3.19}$$

Here, the parameter $[\mathcal{B}\Omega^2]$ depends on the laser-field intensity, which reflects the fact that the Bloch–Siegert shifts are stimulated processes. Assuming $\Omega,\Delta \ll \omega_{\rm L}$ as a typical range of parameters, one has $\omega_{\rm L} \approx \omega_{\rm R}$ and thus

$$\mathcal{B} = \frac{1}{4\omega_{\rm R}} + O(\Delta/\omega_{\rm L}^2, \Omega/\omega_{\rm L}^2). \tag{3.20}$$

Here, one should note that the energy shift Eq. (3.19) with $\mathcal{B}$ as in Eq. (3.20) vanishes for $\Delta=0$. This is consistent with the analysis in [29,39], where it was found that the Bloch-Siegert shift is suppressed by an additional power of $\Omega/\omega_{\rm L}$ for vanishing detuning. We also define a summed Bloch-Siegert shift in analogy with (3.6) and (3.13) as

$$\delta\bar{\omega}_{\pm}^{(\rm BS)} = \pm[\sqrt{\Omega^2+(\Delta-\mathcal{B}\,\Omega^2)^2} - \sqrt{\Omega^2+\Delta^2}]. \tag{3.21}$$

Effectively, the Bloch–Siegert shift may be accounted for by the replacement $\Delta \to \Delta - \mathcal{B}\Omega^2$. This correction to the detuning is proportional to Ω^2, i.e., proportional to the laser intensity.

5. Off-resonant radiative corrections stimulated by the atom-laser interaction

For these corrections, we restrict the atom-field interaction to the laser mode, but take into account the off-resonant (OR) atomic levels $|j\rangle$ (i.e., $|j\rangle \neq |e\rangle, |g\rangle$). The leading effect is the second-order perturbation

$$\Delta E_{\pm,n}^{(\rm OR)} = \left\langle (\pm,n) \left| \mathcal{H}_{\rm L} \frac{1}{E_{\pm,n} - (\mathcal{H}_{\rm M}+\mathcal{H}_{\rm F})} \mathcal{H}_{\rm L} \right| (\pm,n) \right\rangle. \tag{3.22}$$

Here, we have defined the Schrödinger-picture Hamiltonian $\mathcal{H}_{\rm L}$ describing the interaction of the atom with the driving-laser field, the free energy of the nonresonant atomic states $\mathcal{H}_{\rm M}$, and the free energy of the electromagnetic fields $\mathcal{H}_{\rm F}$ as

$$\mathcal{H}_{\rm L} = -q\boldsymbol{x}\cdot\boldsymbol{E}_{\rm L}, \tag{3.23a}$$

$$\mathcal{H}_{\rm M} = \sum_{j\neq g,e} \omega_j |j\rangle\langle j|, \tag{3.23b}$$

$$\mathcal{H}_{\rm F} = \sum_{\mathbf{k}\lambda} \omega_{\mathbf{k}} a_{\mathbf{k}\lambda}^{\dagger} a_{\mathbf{k}\lambda}, \tag{3.23c}$$

respectively, where $a_{\mathbf{k}\lambda}$ and $a_{\mathbf{k}\lambda}^{+}$ are annihilation and creation operators for photons with wave vector $\mathbf{k}$, frequency $\omega_{\mathbf{k}}$, and polarization λ, and ω_j $(j\neq g,e)$ are the energies of the nonresonant intermediate states. The sum in Eq. (3.23c) extends over all possible vacuum-field modes, and $\boldsymbol{E}_{\rm L}$ is the field operator for the laser mode,

$$\boldsymbol{E}_{\rm L} = \sqrt{\frac{\omega_{\rm L}}{2V}} \boldsymbol{\epsilon}_{\rm L}[a_{\rm L} + a_{\rm L}^{+}]. \tag{3.24}$$

Here, $\boldsymbol{\epsilon}_{\rm L}$ is the polarization vector for the laser mode. As the laser mode is highly populated with an occupation number $n \gg 1$, both the field annihilation and creation operators in $\mathcal{H}_{\rm L}$ contribute [see also Eq. (2.12)]. The resulting expression for the energy shift of the dressed state $|(+,n)\rangle$ is given by

$$\Delta E_{+,n}^{(\rm OR)} = \sum_{j\neq e,g} \left\{ |g_{ej}|^2 \cos^2\theta \left(\frac{n+1}{-\omega_j + E_1} + \frac{n}{-\omega_j + E_2} \right) + |g_{gj}|^2 \sin^2\theta \left(\frac{n+1}{-\omega_j + E_3} + \frac{n}{-\omega_j + E_4} \right) \right\}, \tag{3.25}$$

with

$$E_1 = \omega_g - \frac{1}{2}\Delta + \frac{1}{2}\Omega_{\rm R} \approx \omega_g, \tag{3.26a}$$

$$E_2 = \omega_g + 2\omega_{\rm R} + \frac{3}{2}\Delta + \frac{1}{2}\Omega_{\rm R} \approx \omega_g + 2\omega_{\rm R}, \tag{3.26b}$$

$$E_3 = \omega_g - \omega_{\rm R} - \frac{3}{2}\Delta + \frac{1}{2}\Omega_{\rm R} \approx \omega_g - \omega_{\rm R}, \tag{3.26c}$$

$$E_4 = \omega_g + \omega_{\rm R} + \frac{1}{2}\Delta + \frac{1}{2}\Omega_{\rm R} \approx \omega_g + \omega_{\rm R}. \tag{3.26d}$$

For the dressed state $|(-,n)\rangle$, we have

$$\Delta E_{-,n}^{(\rm OR)} = \sum_{j\neq e,g} \left\{ |g_{ej}|^2 \sin^2\theta \left(\frac{n+1}{-\omega_j + E_1} + \frac{n}{-\omega_j + E_2} \right) + |g_{gj}|^2 \cos^2\theta \left(\frac{n+1}{-\omega_j + E_3} + \frac{n}{-\omega_j + E_4} \right) \right\}. \tag{3.27}$$

In calculating these expressions, we may carry out the semiclassical approximation $n+1 \approx n$ and replace the above Rabi frequency by its semiclassical counterpart. The coupling g_{ij} is defined by

$$g_{ij} = -q\langle i|\boldsymbol{\epsilon}_{\rm L}\cdot\boldsymbol{x}|j\rangle \sqrt{\frac{\omega_{\rm L}}{2V}}, \tag{3.28}$$

and is of the same order of magnitude as $g_{\rm L}$. We therefore obtain as the second-order shift due to the off-resonant energy levels,

$$\delta\omega_{\pm}^{(\rm OR)} = \Delta E_{\pm,n}^{(\rm OR)} - \Delta E_{\mp,n}^{(\rm OR)} = \pm\mathcal{D}\cos(2\theta)\Omega^2 = \mp\frac{\Delta}{\sqrt{\Omega^2+\Delta^2}}[\mathcal{D}\Omega^2], \tag{3.29}$$

where Δ is the detuning and

$$\mathcal{D} = \frac{1}{4g_{\rm L}^2} \sum_{j\neq e,g} \left\{ |g_{ej}|^2 \left(\frac{1}{-\omega_j + E_1} + \frac{1}{-\omega_j + E_2} \right) - |g_{gj}|^2 \left(\frac{1}{-\omega_j + E_3} + \frac{1}{-\omega_j + E_4} \right) \right\} \tag{3.30}$$

depends again on the laser intensity. The energies E_i $(i=1,\ldots,4)$ are defined in Eq. (3.26). Therefore this additional shift is a stimulated radiative correction in the same sense as

the corrections discussed in the previous section [32].

To further evaluate the parameter $\mathcal{D}$, it is important to note that the virtual states are coupled to the initial states by the driving-laser field rather than by the vacuum as for example in Lamb-shift calculations. Thus, the polarization of the coupling field mode is fixed. For the off-resonant corrections, it is sufficient to evaluate the relevant matrix elements in the nonrelativistic approximation. So, if we assume the atoms to be in the $1S$ ground state initially ($m=0$, of course), then we have a situation in which for a given polarization of the laser field not all magnetic sublevels of the $2P$ states are coupled. In the following, we assume the driving-laser field to be z polarized [$\epsilon_{\mathrm{L}}=(0,0,1)$], so that only the $m=0$ sublevels of the $1S$ and the $2P$ ground and excited state are occupied. Then, the parameter $\mathcal{D}$ may be rewritten as [see Eq. (3.26)]

$$\mathcal{D}=\frac{M_g(E_3)+M_g(E_4)-M_e(E_1)-M_e(E_2)}{4|\langle g|z|e\rangle|^2} \tag{3.31}$$

in terms of the two matrix elements

$$M_g(\zeta)=\langle g|zG''(\zeta)z|g\rangle, \tag{3.32}$$

$$M_e(\zeta)=\langle e|zG''(\zeta)z|e\rangle, \tag{3.33}$$

where the propagator is given by

$$G(\zeta)=\frac{1}{H-\zeta}, \tag{3.34}$$

and where the double prime means that *both* resonant states $|g\rangle=|1S,m=0\rangle$ and $|e\rangle=|2P,m=0\rangle$ are *excluded* from the sum over intermediate states in the Green function. The evaluation of $M_e(\zeta)$ requires special care, as there are both S (angular quantum number $l=0$) and D ($l=2$) states as intermediate states. Due to the fixed polarization of the coupling field, the angular parts of these two contributions have different proportionality factors relative to the angular parts of the "standard" matrix element

$$\sum_{i=1}^{3}\langle e|x^iG''(\zeta)x^i|e\rangle \tag{3.35}$$

and thus have to be calculated separately. The above standard matrix element may however be recovered from the matrix elements for definite initial state and coupling field polarization by averaging appropriately. This is discussed in Appendix B.

In the propagator $G(\zeta)$, the energy is parametrized by the (in general complex) parameters

$$\zeta\equiv\zeta(t)=-\frac{(Z\alpha)^2m}{2n^2t^2}, \tag{3.36a}$$

$$t\equiv t(\zeta)=\frac{Z\alpha}{n}\sqrt{-\frac{m}{2\zeta}}. \tag{3.36b}$$

Usually, one has $\zeta=E-\omega$ where E is the bound-state energy, and we may write

$$\zeta(t)=E-\omega(t)=-\frac{(Z\alpha)^2m}{2n^2}-\omega(t). \tag{3.37}$$

The parameters ω and t are related by the equations

$$\omega=\frac{(Z\alpha)^2m}{2n^2}\frac{1-t^2}{t^2}, \tag{3.38a}$$

$$t_{nl}(\omega)=\left(1+\frac{2n^2\omega}{m(Z\alpha)^2}\right)^{-1/2}, \tag{3.38b}$$

where n,l are the principal and the angular momentum quantum number of the quantum state for which the relevant matrix elements are to be evaluated. In the following, we will also use common spectroscopic notation for the level characterized by n and l, i.e., for example, $t_{2P}(\omega)\equiv t_{21}(\omega)$. For the energies E_i ($i\in\{1,\ldots,4\}$) and $n=1,2$, we thus obtain

$$t_{nl}(E_i)=\sqrt{\frac{E_n}{E_i}}. \tag{3.39}$$

The above matrix elements Eqs. (3.32) and (3.33) without the double primes, i.e., including resonant intermediate states, may then be expressed in terms of the standard hypergeometric function [34,40,41]

$$\Phi(n,t)={}_2F_1\left(1,-nt,1-nt,\left(\frac{1-t}{1+t}\right)^2\right). \tag{3.40}$$

as (see Appendix B)

$$\overline{M}_g(\zeta)=\langle g|zG(\zeta)z|g\rangle=ma_{\mathrm{B}}^4\left[\frac{2t^2\chi_g(t)}{3(t-1)^5(t+1)^4}-\frac{256t^9}{3(t-1)^5(t+1)^5}\Phi(1,t)\right], \tag{3.41a}$$

$$\overline{M}_e(\zeta)=\langle e|zG(\zeta)z|e\rangle=ma_{\mathrm{B}}^4\left[\frac{16t^2\chi_e(t)}{15(t-1)^7(t+1)^5}-\frac{2^{14}t^{11}(23t^2-7)}{15(t-1)^7(t+1)^7}\Phi(2,t)\right], \tag{3.41b}$$

where

$$\chi_g(t)=38t^7+26t^6+19t^5-19t^4-12t^3+12t^2+3t-3, \tag{3.42}$$

$$\chi_e(t)=6739t^{10}-1702t^9-231t^8-1420t^7-262t^6+1944t^5-402t^4-1140t^3+435t^2+270t-135. \tag{3.43}$$

Here, the Bohr radius scaled by the nuclear charge number Z, in our units, is

$$a_{\mathrm{B}}=\frac{1}{Z\alpha m}, \tag{3.44}$$

where α is the fine-structure constant and m is the electron mass.

The corresponding matrix elements without resonant intermediate states may then be obtained by subtracting the respective contributions of the resonant intermediate states

$$M_g(\zeta) = \langle g|z\ G''(\zeta)z|g\rangle = \overline{M}_g(\zeta) - \frac{|\langle g|z|e\rangle|^2}{E_{2P} - \zeta}, \quad (3.45)$$

$$M_e(\zeta) = \langle e|z\ G''(\zeta)z|e\rangle = \overline{M}_e(\zeta) - \frac{|\langle g|z|e\rangle|^2}{E_{1S} - \zeta}. \quad (3.46)$$

We assume here that the Rabi frequency is not excessively large, which implies that it is small as compared to the optical transition frequency (i.e., $\Omega \ll \omega_R$). For a meaningful measurement of the Mollow spectrum, it is necessary, furthermore, to tune the laser close to the atomic resonance (which implies $\Delta \ll \omega_R$). In this case, we may carry out the following approximations [cf. Eq. (3.26)]

$$E_1 \to \omega_g \Rightarrow t_{2P}(E_1) \to 1/2, \quad (3.47a)$$

$$E_2 \to \omega_g + 2\omega_R \Rightarrow t_{2P}(E_2) \to \sqrt{-0.5}, \quad (3.47b)$$

$$E_3 \to \omega_g - \omega_R \Rightarrow t_{1S}(E_3) \to 2/\sqrt{7}, \quad (3.47c)$$

$$E_4 \to \omega_g + \omega_R \Rightarrow t_{1S}(E_4) \to 2. \quad (3.47d)$$

With these parameters, $\mathcal{D}$ evaluates to

$$\mathcal{D} = \frac{1}{(Z\alpha)^2 m}[6.2148(8) - 0.235\,32(2)\mathrm{i}]$$
$$= \frac{1}{\omega_R}[2.3305(3) - 0.088\,245(6)\mathrm{i}]. \quad (3.48)$$

The uncertainties are mainly due to the approximations carried out in Eq. (3.47) with respect to the energies E_i ($i=1, \ldots, 4$) originally defined in Eq. (3.26). This is possible because the off-resonant stimulated radiative correction amounts to a modification of the detuning which is of order $\Omega^2/\omega_R \ll \Omega$ [see also Eq. (3.49) below]. Therefore we may carry out the approximation Eq. (3.47), i.e., neglect the further corrections of order $\Omega^3/\omega_R^2 \ll \Omega^2/\omega_R$ and $\Omega^2\Delta/\omega_R^2 \ll \Omega^2/\omega_R$, which are beyond the scope of the current analysis. The real part $\mathcal{D}_R \equiv \mathrm{Re}(\mathcal{D})$ gives rise to a shift of the position of the Mollow sideband, while the imaginary part $\mathcal{D}_I \equiv \mathrm{Im}(\mathcal{D})$ describes the ionization into the continuum. This means that the imaginary part of the energy shifts $\Delta E_{\pm,n}^{(\mathrm{OR})}$ received by the two dressed states $|(\pm,n)\rangle$ must be negative, which is equivalent to a negative sign for the imaginary part $\mathcal{D}_I$.

In the numerical analysis in Sec. IV, it is shown that for typical parameters the system is sufficiently far from the ionization threshold [42] so that the ionization does not restrict the applicability of our scheme. The real part yields a correction to the detuning given by $\Delta \to \Delta - \mathcal{D}_R\Omega^2$, according to the summation [cf. Eq. (3.29)]

$$d\overline{\omega}_{\pm}^{(\mathrm{OR})} = \pm\left[\sqrt{\Omega^2 + (\Delta - \mathcal{D}_R\Omega^2)^2} - \sqrt{\Omega^2 + \Delta^2}\right] \quad (3.49)$$

with

$$\delta\overline{\omega}_{\pm}^{(\mathrm{OR})} \approx \delta\omega_{\pm}^{(\mathrm{OR})} = \mp \frac{\Delta}{\sqrt{\Omega^2 + \Delta^2}}[\mathcal{D}_R\Omega^2]. \quad (3.50)$$

As pointed out below in Sec. IV (see also Table I), the magnitude of the off-resonant correction is small as compared to the detuning for typical parameters so that there is no numerically significant difference between the first-order correction Eq. (3.29) and the summed form given in Eq. (3.49).

TABLE I. Summary of all individual energy shifts due to the various discussed corrections. All numbers are obtained for $h_j = 1000$ and $\Delta = 50\Gamma_j$ with $j=\frac{1}{2}$ ($j=\frac{3}{2}$) for the left (right) column using summation formulas such as Eq. (4.8). Here, $\delta\overline{\omega}_+^{(\mathrm{Lamb})}$ is the correction to the high-frequency Mollow sideband position related to the bare Lamb shift (cf. Sec. III A 2), the symbol (BS) denotes Bloch-Siegert shifts (Sec. III A 4), and the (OR) shifts are due to off-resonant excitations (Sec. III A 5). These shifts all may be interpreted as arising from a modified detuning Δ and are discussed in Sec. III A. The other five corrections are due to a modified Rabi frequency (Sec. III B). In particular, $\delta\overline{\omega}_+^{(\mathrm{R})}$ is discussed in Sec. III B 1 and refers to relativistic corrections, whereas $\delta\overline{\omega}_+^{(\mathrm{F})}$ (Sec. III B 2) is a field-configuration-dependent shift. The shift $\delta\overline{\omega}_+^{(\mathrm{TDM})}$ (Secs. III B 4 and III B 5) refers to radiative corrections to the transition dipole matrix element, and $\delta\overline{\omega}_+^{(\mathrm{C})}$ is a dynamic correction to the Rabi frequency (Secs. III B 3 and III B 5). Finally, $\delta\overline{\omega}_+^{(\mathrm{S})}$ is a shift due to corrections to the secular approximation (Sec. III B 6).

Shift	$1S_{1/2} \leftrightarrow 2P_{1/2}$ (kHz)	$1S_{1/2} \leftrightarrow 2P_{3/2}$ (kHz)
$\delta\overline{\omega}_{+,j}^{(\mathrm{Lamb})}$	741 599(4)	738 281(4)
$\delta\overline{\omega}_+^{(\mathrm{BS})}$	−50.30(5)	−50.30(5)
$\delta\overline{\omega}_+^{(\mathrm{OR})}$	−468.51(6)	−468.51(6)
$\delta\overline{\omega}_{+,j}^{(\mathrm{R})}$	−1842.1(1)	−1937.7(1)
$\delta\overline{\omega}_+^{(\mathrm{F})}$	−331.44(2)	−331.44(2)
$\delta\overline{\omega}_{+,j}^{(\mathrm{C})}$	−121(31)	−121(31)
$\delta\overline{\omega}_{+,j}^{(\mathrm{TDM})}$	374(25)	372(26)
$\delta\overline{\omega}_+^{(\mathrm{S})}$	−49.8(2)	−49.8(2)

The first-order imaginary contribution to the Mollow sidebands is

$$\delta\omega_{\pm}^{(\mathrm{Im})} = \mp \frac{\Delta}{\sqrt{\Omega^2 + \Delta^2}}[\mathrm{i}\mathcal{D}_I\Omega^2]. \quad (3.51)$$

This effect broadens the sidebands slightly, but its contribution is so small for typical parameters (see Sec. IV below), that it may be ignored on the current level of accuracy of the theoretical predictions. It is interesting to note that the stimulated off-resonant correction is small as compared to the relatively large effect mediated by the bare Lamb shift of the transition, which is discussed in Sec. III A 2.

B. Corrections to the Rabi frequency

In this section we discuss corrections to the incoherent fluorescence spectrum due to modifications of the Rabi frequency. In particular, we consider corrections to the transition dipole moment and to the spontaneous transition rate of the atomic transition, due to coupling of the driving-laser field to resonant and nonresonant atomic transitions, and corrections to the secular approximation.

1. Relativistic corrections to the transition dipole moment

In this section we discuss relativistic corrections to the fluorescence spectrum up to relative order $(Z\alpha)^2$. The corrections amount to a modification of the atomic transition frequency and of the transition dipole moments. The relativistic expressions for the state energies and the transition dipole moments depend on the total angular quantum numbers j of the involved states, which is the vectorial sum of the electron orbital angular momentum l and its spin s. Therefore, we specify the total angular momentum quantum number and thus the spin state of the atomic system in order to fix a specific experimental setup (see also Appendix A). We further assume the atom to be driven by a pure dipole laser field linearly polarized in the z direction, such that the laser field only couples states with equal magnetic quantum number. The situation of a pure dipole field has recently been studied in a related context in [43]. In the numerical analysis in Sec. IV, we consider a standing wave laser-field configuration where the atom is at a point of maximum electric-field intensity of the standing wave. As then, the magnetic field component of the driving laser may be neglected, it is not considered in the following analysis (corrections due to the variation of the electric field about its maximum are treated in Sec. III B 2).

The relativistic corrections to the energies of the atomic states and thus to the atomic transition frequencies effectively modify the detuning Δ and may be accounted for by choosing an experimental value for the atomic transition frequency as found in low-intensity scattering experiments (see Sec. III A 3). The corrections to the transition dipole moments may be evaluated with the help of the relativistic wave functions of the hydrogen atom as given in [44,45]. We denote the absolute relativistic correction to the nonrelativistic matrix element

$$\left\langle 1S_{1/2}, m=\pm\tfrac{1}{2}\middle|z\middle|2P_j, m=\pm\tfrac{1}{2}\right\rangle_{\mathrm{NR}}, \tag{3.52}$$

by $\delta d_j^{(\mathrm{R})}$, where $j=\frac{1}{2},\frac{3}{2}$ is the total angular momentum and $m=\pm\frac{1}{2}$ is the magnetic quantum number. In the following, we will omit the "$m=$" from the second parameter of the atomic state vectors. Then the relativistic matrix element (with subindex "R") gives rise to a relative $(Z\alpha)^2$ correction $\delta d_j^{(\mathrm{R})}/d_j$ with respect to the nonrelativistic (NR) expression which is given by

$$\frac{\left\langle 1S_{1/2}, \pm\frac{1}{2}\middle|z\middle|2P_j, \pm\frac{1}{2}\right\rangle_R}{\left\langle 1S_{1/2}, \pm\frac{1}{2}\middle|z\middle|2P_j, \pm\frac{1}{2}\right\rangle_{\mathrm{NR}}} = 1+\frac{\delta d_j^{(\mathrm{R})}}{d_j}, \tag{3.53}$$

where we ignore higher-order relativistic terms of order $(Z\alpha)^m$ with $m\geqslant 4$. The corresponding matrix elements where the "initial" and the "final" state have different magnetic quantum numbers vanish identically as the driving-laser field is assumed to be polarized in the z direction. Evaluating the relative corrections, one obtains

$$\frac{\delta d_{1/2}^{(\mathrm{R})}}{d_{1/2}} = -(Z\alpha)^2\left(\frac{13}{32}+\frac{3}{2}\ln 2-\ln 3\right), \tag{3.54a}$$

$$\frac{\delta d_{3/2}^{(\mathrm{R})}}{d_{3/2}} = -(Z\alpha)^2\left(\frac{31}{96}+\frac{5}{4}\ln 2-\frac{3}{4}\ln 3\right). \tag{3.54b}$$

The Rabi frequency and the transition dipole moment depend linearly on each other. Therefore, the relative correction to the Rabi frequency is identical to the relative correction to the transition dipole moment:

$$\frac{\delta\Omega_j^{(\mathrm{R})}}{\Omega} = \frac{\delta d_j^{(\mathrm{R})}}{d_j}. \tag{3.55}$$

Here, $\delta\Omega_j^{(\mathrm{R})}$ is the absolute correction to the Rabi frequency due to relativistic modifications to the transition dipole moment. The spin-dependent shift $\delta\bar{\omega}_{\pm,j}^{(\mathrm{R})}$ of the position of the Mollow sidebands due to the relativistic corrections of the matrix element is thus given by

$$\delta\bar{\omega}_{\pm,j}^{(\mathrm{R})} = \pm\left[\sqrt{(\Omega+\delta\Omega_j^{(\mathrm{R})})^2+\Delta^2}-\sqrt{\Omega^2+\Delta^2}\right], \tag{3.56}$$

where

$$\delta\bar{\omega}_{\pm,j}^{(\mathrm{R})} \approx \delta\omega_{\pm,j}^{(\mathrm{R})} = \mp\frac{\Omega^2}{\sqrt{\Omega^2+\Delta^2}}\mathcal{E}_j \tag{3.57}$$

and

$$\mathcal{E}_j = -\frac{\delta d_j^{(\mathrm{R})}}{d_j}+O\left(\frac{\delta d_j}{d_j}\right)^2. \tag{3.58}$$

With these definitions, the summed relativistic correction to the dipole moment effectively corresponds to a replacement $\Omega\rightarrow\Omega(1-\mathcal{E}_j)$.

2. Field-configuration-dependent correction to the Rabi frequency

It is well known that the magnetic component of plane-wave electromagnetic wave influences the transition current at relative order $(Z\alpha)^2$ (see, e.g., [34,46,47]). The radiation pressure due to the magnetic field could move the atom. Therefore, we propose a standing-wave field configuration, where the atom is placed at an antinode of the standing-wave electric field. In this setup, the influence of the magnetic field can be neglected to a very good approximation.

The analysis of the previous Sec. III B 1 is valid up to the order discussed [relative order $(Z\alpha)^2$] only for a pure dipole field which additionally has to be constant in any direction perpendicular to the polarization. However, for a standing-wave configuration, the z-polarized electric field of the laser is not constant in the propagation (x) direction. This leads to a further correction, which gives rise to a field-configuration-dependent shift of the Rabi frequency. In the following, this shift of relative order $(Z\alpha)^2$ is analyzed for the setup described above where the atom is at the maximum of the standing-wave electric field.

We start from the long-wavelength quantum electrodynamic (LWQED) interaction Hamiltonian [47]. The only relevant terms (in the context of our analysis) of the interaction part of this Hamiltonian are

$$H_I^{\mathrm{LW}} = -q\boldsymbol{x}\cdot\boldsymbol{E} - \frac{q}{2}x^i x^j E^i_{,j} - \frac{q}{6}x^i x^j x^k E^i_{,jk}. \tag{3.59}$$

Here, the x^i denotes the ith component of the position operator vector $\mathbf{x}$, and $E^i_{,j}$ is the partial derivative with respect to x^j of the ith component of the electric-field vector. The electric field of the standing wave is given by

$$\boldsymbol{E}(t,x) = \hat{e}_z \mathcal{E}_{\mathrm{SW}} \cos(\omega t)\cos(kx). \tag{3.60}$$

The term containing the first derivative of the electric field in Eq. (3.59) vanishes, and the last term gives

$$-\frac{q}{6}x^i x^j x^k E^i_{,jk} = -\frac{q}{6} z\, x^2 E^z_{,xx} = \frac{q}{6} z\, k^2 x^2 \mathcal{E}_{\mathrm{SW}} \cos(\omega t). \tag{3.61}$$

This result has to be distinguished from a simple expansion of the electric field around the maximum at $x=0$, which yields

$$-q\,\boldsymbol{r}\cdot\boldsymbol{E}(t,x) = -q\,z\,\mathcal{E}_{\mathrm{SW}}\cos(\omega t)\cos(kx) = -qz\mathcal{E}_{\mathrm{SW}}\cos(\omega t) \times\left(1 - \frac{(kx)^2}{2} + O(x^4)\right). \tag{3.62}$$

This naive expansion gives the wrong prefactor and is not applicable here.

The term in Eq. (3.61) entails a spin-independent correction to the transition dipole moment. At resonance, one has

$$k = \frac{3}{8}(Z\alpha)^2 m. \tag{3.63}$$

The relative correction due to the additional contribution Eq. (3.61) to the interaction Hamiltonian is therefore

$$-\frac{k^2}{6}\frac{\langle 1S, m=0|z\,x^2|2P, m=0\rangle}{\langle 1S, m=0|z|2P, m=0\rangle} = -\frac{1}{16}(Z\alpha)^2. \tag{3.64}$$

Analogous to Eq. (3.53), this modification of the transition dipole moment, for a $1S$-$2P$ transition, gives rise to a correction to the Rabi frequency given by

$$\delta\Omega^{(\mathrm{F})} = -\frac{1}{16}(Z\alpha)^2\Omega. \tag{3.65}$$

The summed shift $\delta\bar{\omega}^{(\mathrm{F})}_{\pm}$ of the Mollow sidebands due to this modification of the Rabi frequency can be expressed as

$$\delta\bar{\omega}^{(\mathrm{F})}_{\pm} = \pm\left[\sqrt{(\Omega+\delta\Omega^{(\mathrm{F})})^2+\Delta^2} - \sqrt{\Omega^2+\Delta^2}\right], \tag{3.66}$$

where

$$\delta\bar{\omega}^{(\mathrm{F})}_{\pm} \approx \delta\omega^{(\mathrm{F})}_{\pm} = \mp\frac{\Omega^2}{\sqrt{\Omega^2+\Delta^2}}\mathcal{F} \tag{3.67}$$

and

$$\mathcal{F} = \frac{1}{16}(Z\alpha)^2. \tag{3.68}$$

With these definitions, the summed relativistic correction to the dipole moment effectively corresponds to a replacement $\Omega\to\Omega(1-\mathcal{F})$.

It is important to note that the long-wave QED correction to the interaction Hamiltonian Eq. (3.59) does not couple any unwanted magnetic quantum numbers to the laser-driven doublet.

3. Higher-order corrections (in Ω and Δ) to the self-energy of dressed states and corresponding correction to the Rabi frequency

The Lamb shift of dressed states is different from the Lamb shift of atomic bare states, as already discussed in Sec. I. In this section, we extend the analysis of Sec. III A 2 to the next-higher order. For this, we keep the terms linear in Ω and Δ in evaluating the energy shifts of the dressed states. As explained in detail in [28,29], we thereby obtain a further correction to the position of the fluorescence sidebands which may be expressed as

$$\delta\omega^{(C)}_{\pm} = \mp\,\mathcal{C}\frac{\Omega^2}{\sqrt{\Omega^2+\Delta^2}}, \tag{3.69}$$

where

$$\mathcal{C} = \frac{\alpha}{\pi}\ln[(Z\alpha)^{-2}]\frac{\langle \boldsymbol{p}^2\rangle_g + \langle \boldsymbol{p}^2\rangle_e}{m^2} \tag{3.70}$$

is a dimensionless constant. For the hydrogen $1S$-$2P$ transition, the leading logarithmic term is independent of the spin and given by

$$\mathcal{C} = \frac{5}{4\pi}\alpha(Z\alpha)^2\ln[(Z\alpha)^{-2}]. \tag{3.71}$$

This correction may be interpreted physically with a summation as used for the bare Lamb-shift correction

$$\delta\bar{\omega}^{(C)}_{\pm} = \pm\left[\sqrt{\Omega^2(1-\mathcal{C})^2+\Delta^2} - \sqrt{\Omega^2+\Delta^2}\right], \tag{3.72}$$

with $\delta\bar{\omega}^{(C)}_{\pm} \approx \delta\omega^{(C)}_{\pm}$ because of the smallness of the correction. Thus the additional shift $\delta\omega^{(C)}_{\pm}$ may be interpreted as a radiative correction $\Omega\to\Omega(1-\mathcal{C})$ of the Rabi frequency.

4. Leading logarithmic radiative corrections to the transition dipole moment (vertex corrections)

In "normal" bound-state quantum electrodynamics, vertex corrections are evaluated with respect to the interaction of the electron with the binding field of the atomic nucleus. In an effective treatment, and in leading logarithmic approximation, the effect of the self-energy may be accounted for by making use of an effective Lamb-shift potential [48]

$$\Delta V_{\mathrm{Lamb}}(\boldsymbol{r}) = \frac{4}{3}\alpha(Z\alpha)\ln[(Z\alpha)^{-2}]\frac{\delta^{(3)}(\boldsymbol{r})}{m^2}, \tag{3.73}$$

which modifies the Coulomb interaction according to

$$-\frac{Z\alpha}{r} \to -\frac{Z\alpha}{r} + \Delta V_{\mathrm{Lamb}}(\boldsymbol{r}). \tag{3.74}$$

Note that the potential (3.73) is really the consequence of a self-energy ("vertex") correction, not that of vacuum polarization. In many cases, vacuum polarization corrections may

also be accounted for by employing an effective potential, but the corresponding potential lacks the large logarithm $\ln[(Z\alpha)^{-2}]$. An accurate treatment of self-energy corrections requires the consideration of many more terms than the crude approximation (3.73).

Here, we evaluate the leading vertex corrections to the interaction of the bound electron with the driving-laser field, and so we have to consider both the Coulomb as well as the laser field. Nonrelativistically, the atom-laser interaction is given by the matrix element of the usual interaction Hamiltonian, which reads (length gauge, $1S \Leftrightarrow 2P$ transition) $-qE_{\rm L}\langle 1S|z|2P\rangle = -qE_{\rm L}d$ where $E_{\rm L}$ is the field strength of the (strong) laser field and the dipole moment is

$$d = \langle 2P|z|1S\rangle = \frac{2^7}{3^5}\sqrt{2}\frac{1}{m(Z\alpha)}. \tag{3.75}$$

Vertex corrections lead to modifications of the dipole moment given by $d \to d + \delta d$, where the vertex correction δd is considered below, and the radiative correction (in the length gauge) to the laser-atom interaction is effectively a replacement $-qE_{\rm L}d \to -qE_{\rm L}(d+\delta d)$. The large intensity of the driving-laser field is accounted for in this formalism because the electric laser-field strength $E_{\rm L}$ multiplies both the dipole moment matrix element of the interaction Hamiltonian d and the radiative correction δd.

Laser photons as well as the spontaneously emitted photons in the radiative decay of excited states are real rather than virtual. Consequently, the radiative corrections to the laser-atom interaction on the one hand and to the radiative decay rate on the other hand are related to each other. In the length gauge, the leading-order expression for the spontaneous emission decay rate is

$$\Gamma = \frac{4}{3}\alpha E^3 d^2 = \frac{2^8}{3^8}\alpha(Z\alpha)^4 m. \tag{3.76}$$

In order to obtain gauge-invariant results for the quantum electrodynamic corrections to Γ (while working in the length gauge), it is necessary to consider both radiative vertex corrections to the dipole moment d and corrections to the (bare) transition frequency (energy difference) E (see Refs. [48–50]). In our treatment, the vertex corrections to the dipole moment are given in Eqs. (3.81), (3.86a), and (3.86b), whereas the vertex corrections to the transition frequency enter into the radiative correction to the detuning in Eq. (4.10a).

In general, the vertex corrections to the laser-atom interaction enter at the relative order of $O(\alpha(Z\alpha)^2\ln[(Z\alpha)^{-2}])$. One may wonder why the corrections do not enter at the relative order $O(\alpha)$. The reason is that in interactions with real photons (the square of the four momentum being $q^2=0$), the otherwise dominant correction due to the F_1 Dirac form factor vanishes, and the remaining terms are then of higher order in the $Z\alpha$ expansion.

We now analyze the shift of the resonance frequency and the shift of the transition dipole moment induced by the Lamb-shift potential (3.73). The transition energy

$$E = E_{2P} - E_{1S} = \frac{3}{8}(Z\alpha)^2 m \tag{3.77}$$

is shifted by $V_{\rm Lamb}$ according to

$$\delta E^{(\log)} = -\langle 1S|\Delta V_{\rm Lamb}(\boldsymbol{r})|1S\rangle = -\frac{4m}{3\pi}\alpha(Z\alpha)^4\ln[(Z\alpha)^{-2}], \tag{3.78}$$

because the matrix element of the $2P$ state vanishes. This yields a relative shift of

$$\frac{\delta E^{(\log)}}{E} = -\frac{32}{9\pi}\alpha(Z\alpha)^2\ln[(Z\alpha)^{-2}]. \tag{3.79}$$

The modification of the matrix element due to the corrections to the $1S$-wave function amounts to

$$\delta d^{(\log)} = \left\langle 2P \left| z\left(\frac{1}{E-H}\right)' \Delta V_{\rm Lamb}(\boldsymbol{r}) \right| 1S \right\rangle = \sqrt{2}\frac{2^5\alpha(Z\alpha)}{3^7 m\pi}\ln[(Z\alpha)^{-2}]\left(48\ln\frac{4}{3}+131\right), \tag{3.80}$$

where the prime denotes the reduced Green function. Thus, the logarithmic relative correction is [49]

$$\frac{\delta d^{(\log)}}{d} = \frac{\alpha(Z\alpha)^2}{\pi}\ln[(Z\alpha)^{-2}]\left(\frac{4}{3}\ln\frac{4}{3}+\frac{131}{36}\right). \tag{3.81}$$

This leads to a correction to the Rabi frequency analogous to Eq. (3.55),

$$\frac{\delta\Omega^{(\log)}}{\Omega} = \frac{\delta d^{(\log)}}{d}, \tag{3.82}$$

where we may ignore the spin, in contrast to Eq. (3.54).

The interpretation of this shift is analogous to the relativistic corrections to the dipole matrix element in Sec. III B 1. The Mollow sidebands are shifted by the frequency

$$\delta\omega_{\pm}^{(\log)} = \pm\left[\sqrt{(\Omega+\delta\Omega^{(\log)})^2+\Delta^2} - \sqrt{\Omega^2+\Delta^2}\right], \tag{3.83}$$

where

$$\delta\omega_{\pm}^{(\log)} \approx \delta\omega_{\pm}^{(\log)} = \pm\frac{\Omega^2}{\sqrt{\Omega^2+\Delta^2}}\mathcal{A}, \tag{3.84}$$

and

$$\mathcal{A} = \frac{\delta d^{(\log)}}{d} + O\left(\frac{\delta d^{(\log)}}{d}\right)^2 \tag{3.85}$$

is a dimensionless constant. Then, these corrections may be accounted for by the replacement $\Omega \to \Omega(1+\mathcal{A})$. One should note that at this parametric order, the above results hold for both states $2P_{1/2}$ and $2P_{3/2}$ [49,50].

5. Nonlogarithmic vertex and vacuum polarization corrections to the transition dipole moment

The concurrence of the radiative shifts to the transition dipole matrix elements for the $1S$-$2P_{1/2}$ and the $1S$-$2P_{3/2}$

transitions, which was found in Sec. III B 4 for the effects of relative order $\alpha(Z\alpha)^2 \ln[(Z\alpha)^{-2}]$, is lifted on taking into account corrections of order $\alpha(Z\alpha)^2$ (no logarithms) due to the self-energy. Vacuum polarization corrections to the transition dipole matrix elements also enter at the relative order of $\alpha(Z\alpha)^2$.

The sum of the nonlogarithmic ("nlog") vertex and vacuum polarization corrections to the transition dipole moments of order $\alpha(Z\alpha)^2$ are given by [50]

$$\frac{\delta d_{1/2}^{(\mathrm{nlog})}}{d_{1/2}} = -\frac{\alpha(Z\alpha)^2}{\pi} 9.2(1.8) \tag{3.86a}$$

for the $2P_{1/2}$ state and by

$$\frac{\delta d_{3/2}^{(\mathrm{nlog})}}{d_{3/2}} = -\frac{\alpha(Z\alpha)^2}{\pi} 9.3(1.9) \tag{3.86b}$$

for the $2P_{3/2}$ state [50]. In between Eqs. (70) and (71) of [50], it is stated that currently, there is an internal discrepancy between the numerically obtained values for the radiative correction at $Z=5,10,\ldots$ on the one hand, and analytic results for the first terms of the $Z\alpha$ expansion (logarithm + constant) on the other hand. This discrepancy is of the order of 10% of the total constant term of order $\alpha(Z\alpha)^2$, and this limits the current status of the theory. Here, we employ an even more conservative error estimate and assign a 20% uncertainty to both of the numerical values in Eqs. (3.86a) and (3.86b). The corresponding correction to the Rabi frequency is again analogous to Eq. (3.55),

$$\frac{\delta\Omega_j^{(\mathrm{nlog})}}{\Omega} = \frac{\delta d_j^{(\mathrm{nlog})}}{d_j}. \tag{3.87}$$

One obtains the following total correction $\delta\bar{\omega}_{\pm,j}^{(\mathrm{TDM})}$ to the position of the sidebands due to logarithmic and nonlogarithmic correction to the transition dipole matrix element (TDM):

$$\delta\bar{\omega}_{\pm,j}^{(\mathrm{TDM})} = \pm\left[\sqrt{(\Omega+\delta\Omega^{(\mathrm{log})}+\delta\Omega_j^{(\mathrm{nlog})})^2+\Delta^2} - \sqrt{\Omega^2+\Delta^2}\right], \tag{3.88}$$

where

$$\delta\bar{\omega}_{\pm,j}^{(\mathrm{TDM})} \approx \delta\omega_{\pm,j}^{(\mathrm{TDM})} = \pm\frac{\Omega^2}{\sqrt{\Omega^2+\Delta^2}}\mathcal{A}_j \tag{3.89}$$

and

$$\mathcal{A}_j = \frac{\delta d^{(\mathrm{log})}}{d} + \frac{\delta d_j^{(\mathrm{nlog})}}{d_j} + O(\alpha(Z\alpha)^3). \tag{3.90}$$

$\mathcal{A}_j$ is a dimensionless constant and $j=\frac{1}{2},\frac{3}{2}$. Thus the corrections amount to a modification of the Rabi frequency given by $\Omega\to\Omega(1+\mathcal{A}_j)$.

We expect a similar nonlogarithmic correction of order $\alpha(Z\alpha)^2$ to supplement the $\mathcal{C}$ term discussed in Sec. III B 3. The logarithmic $\mathcal{C}$ term was found to be of the order of $\alpha(Z\alpha)^2\ln[(Z\alpha)^{-2}]$. The evaluation of the corresponding nonlogarithmic term is, however, beyond the scope of this work. Here, we only present a (conservative) estimate of the expected correction. To this end, we observe that the general form of the radiative corrections under discussion is

$$\alpha(Z\alpha)^2\ln[(Z\alpha)^{-2}]c_1 + \alpha(Z\alpha)^2 c_2, \tag{3.91}$$

where c_1 and c_2 are dimensionless constants. Based on experience with similar corrections (both for self-energy effects and well as radiative corrections to decay rates [37,50]), we assume the following relation with the corresponding uncertainty for the unknown parameter c_2:

$$c_2 = (-2\pm 2)c_1. \tag{3.92}$$

For example, we verify the validity of this estimate for the particular contribution to the nonlogarithmic part discussed above, i.e., for the terms that contribute to $\mathcal{A}_j$. For these terms, the estimate $(-2\pm2)c_1$ evaluates to -2.56 ± 2.56 for the constant term. In comparison, the values obtained above for c_2 by a direct numerical analysis are -2.92 for the correction $\mathcal{A}_{1/2}$, and -2.97 for the correction $\mathcal{A}_{3/2}$, which agrees to the estimate. For the $\mathcal{C}$-term correction, we thus obtain from Eqs. (3.71) and (3.92)

$$\mathcal{C}_j = \alpha(Z\alpha)^2\left(\frac{5}{4\pi}\ln[(Z\alpha)^{-2}] - \frac{5}{4\pi}(2\pm2)\right) \tag{3.93}$$

as the combination of the logarithmic and the estimated nonlogarithmic correction. We expect the nonlogarithmic correction to be spin dependent in analogy to Eq. (3.86a) and (3.86b). As for the correction to the matrix element, this shifts the Mollow sidebands by [see also Eq. (3.72)]

$$\delta\bar{\omega}_{\pm,j}^{(C)} = \pm\left[\sqrt{\Omega^2(1-\mathcal{C}_j)^2+\Delta^2} - \sqrt{\Omega^2+\Delta^2}\right] \approx \mp\frac{\Omega^2}{\sqrt{\Omega^2+\Delta^2}}\mathcal{C}_j, \tag{3.94}$$

such that the correction may be applied by the replacement $\Omega\to\Omega(1-\mathcal{C}_j)$.

6. Corrections to the secular approximation

In this section we transform the correction terms to the secular approximation in Eq. (2.6) such that they may be integrated into our correction scheme. Equation (2.6) may be rewritten as

$$\omega_\pm = \omega_\mathrm{L} \pm \Omega_\mathrm{R} \mp \frac{\Omega^2}{\sqrt{\Omega^2+\Delta^2}}\mathcal{S}, \tag{3.95}$$

$$\mathcal{S} = \frac{4+y^2}{8(1+y^2)}\left(\frac{\Gamma}{\Omega}\right)^2 + O\left(\frac{\Gamma}{\Omega}\right)^4. \tag{3.96}$$

For $\Delta\ll\Omega$, one may expand the leading contribution to $\mathcal{S}$ to give

$$\mathcal{S} = \frac{1}{2}\left(\frac{\Gamma}{\Omega}\right)^2 + O\left(\frac{\Delta^2\Gamma^2}{\Omega^4}\right). \tag{3.97}$$

With this definition, the corrections to the secular approximation may be accounted for in the final result with the replacement $\Omega\to\Omega(1-\mathcal{S})$, which results in a summed shift of

$$\delta\bar{\omega}_\pm^{(S)} = \pm\left[\sqrt{\Omega^2(1-\mathcal{S})^2+\Delta^2} - \sqrt{\Omega^2+\Delta^2}\right]. \tag{3.98}$$

IV. NUMERICAL DATA FOR THE HYDROGEN $1S$-$2P_j$ TRANSITIONS $\left(j=\frac{1}{2},\frac{3}{2}\right)$

In the preceding section, we have discussed both corrections to the detuning and to the Rabi frequency, which give rise to a modification of the positions of the sidebands in the Mollow spectrum. We start the evaluation of correction terms here from a point where we assume that all relativistic corrections to the transition frequency, as well as hyperfine-structure effects, have already been included in the bare transition frequency. This frequency corresponds to the prediction obtained using Eq. (3.16) by setting L_{nl_j} explicitly equal to zero. We therefore redefine the detuning Δ to be

$$\Delta = \omega_{\mathrm{L}} - \tilde{\omega}_{\mathrm{R}}, \tag{4.1}$$

where [cf. Eq. (3.16)]

$$\tilde{\omega}_{\mathrm{R}} = \tilde{E}(2P_j) - \tilde{E}(1S) \tag{4.2}$$

with

$$\tilde{E}(nl_j) = m_r[f(n,j)-1] - \frac{m_r^2}{2(m+m_N)}[f(n,j)-1]^2 + E_{\mathrm{hfs}}. \tag{4.3}$$

Of course, the full theoretical prediction $E(nl_j)$ is obtained as the sum of $\tilde{E}(nl_j)$ and L_{nl_j}. The modification of the Rabi frequency due to the bare Lamb shift

$$L^{(j)}_{\mathrm{bare}} = L_{2P_j} - L_{1S}, \tag{4.4}$$

as discussed in Sec. III A 2, finds a natural interpretation as a contribution to the *Lamb shift of the dressed states*. The various first-order correction terms to the lowest-order prediction for the generalized Mollow-sideband displacement

$$\pm\sqrt{\Omega^2+\Delta^2}, \tag{4.5}$$

starting from Eq. (4.1), may be summarized as follows:

$$\delta\omega^{(j)}_{\pm} = \mp\frac{\Delta}{\sqrt{\Omega^2+\Delta^2}}(\mathrm{L}^{(j)}_{\mathrm{bare}} + \mathcal{B}\,\Omega^2 + \mathcal{D}_{\mathrm{R}}\Omega^2) \pm \frac{\Omega^2}{\sqrt{\Omega^2+\Delta^2}}(\mathcal{A}_j - \mathcal{C}_j - \mathcal{E}_j - \mathcal{F} - \mathcal{S}), \tag{4.6}$$

where

$$L^{(1/2)}_{\mathrm{bare}} = -8185.647(34)\times 10^6\ \mathrm{Hz}, \tag{4.7a}$$

$$L^{(3/2)}_{\mathrm{bare}} = -8160.294(34)\times 10^6\ \mathrm{Hz}, \tag{4.7b}$$

$$\mathcal{B} = \frac{1}{4\omega_{\mathrm{R}}}, \tag{4.7c}$$

$$\mathcal{D} = \mathcal{D}_{\mathrm{R}} + \mathrm{i}\mathcal{D}_I = \frac{1}{\omega_{\mathrm{R}}}[2.3305(3) - 0.088\,245(6)\mathrm{i}], \tag{4.7d}$$

$$\mathcal{A} = \frac{\alpha(Z\alpha)^2}{\pi}\ln(Z\alpha)^{-2}\left(\frac{4}{3}\ln\frac{4}{3}+\frac{131}{36}\right), \tag{4.7e}$$

$$\mathcal{A}_{1/2} = \mathcal{A} - \frac{\alpha(Z\alpha)^2}{\pi}9.2(1.8), \tag{4.7f}$$

$$\mathcal{A}_{3/2} = \mathcal{A} - \frac{\alpha(Z\alpha)^2}{\pi}9.3(1.9), \tag{4.7g}$$

$$\mathcal{C}_j = \alpha(Z\alpha)^2\frac{5}{4\pi}\{\ln[(Z\alpha)^{-2}] - (2\pm 2)\}, \tag{4.7h}$$

$$\mathcal{E}_{1/2} = (Z\alpha)^2\left(\frac{13}{32}+\frac{3}{2}\ln 2 - \ln 3\right), \tag{4.7i}$$

$$\mathcal{E}_{3/2} = (Z\alpha)^2\left(\frac{31}{96}+\frac{5}{4}\ln 2 - \frac{3}{4}\ln 3\right), \tag{4.7j}$$

$$\mathcal{F} = \frac{1}{16}(Z\alpha)^2, \tag{4.7k}$$

$$\mathcal{S} = \frac{1}{2}\left(\frac{\Gamma}{\Omega}\right)^2. \tag{4.7l}$$

We have found that the bare Lamb shift $L^{(j)}_{\mathrm{bare}}$, the Bloch–Siegert shift ($\mathcal{B}$), and the off-resonant self-energy corrections ($\mathcal{D}_{\mathrm{R}}$) give rise to a modification of the detuning Δ in the expression for the Mollow spectrum. As the latter two effects are intensity dependent, also their correction to the detuning depends on the intensity of the incident laser field. The modifications to the transition dipole moment ($\mathcal{A}_j, \mathcal{E}_j, \mathcal{F}$) as well as the higher-order resonant self-energy shifts ($\mathcal{C}_j$) and the correction to the secular approximation ($\mathcal{S}$) may be interpreted as radiative corrections to the Rabi frequency (Ω). The interpretations as a modification of the detuning and the Rabi frequency may best be seen by using a summation of Eq. (4.6), which we have shown to be valid up to first order in the parameters in Eq. (4.7):

$$\delta\omega^{(j)}_{\pm} = \pm(\Omega^{(j)}_{\mathcal{C}} - \sqrt{\Omega^2+\Delta^2}) \tag{4.8}$$

with

$$\Omega^{(j)}_{\mathcal{C}} = \sqrt{\Omega^2(1+\hat{\Omega}^{(j)}_{\mathrm{rad}})^2 + (\Delta - \Delta^{(j)}_{\mathrm{rad}})^2}. \tag{4.9}$$

Here, the Rabi frequency and detuning are supplemented by the discussed relativistic and radiative corrections; these are given by

$$\Delta^{(j)}_{\mathrm{rad}} = \mathrm{L}^{(j)}_{\mathrm{bare}} + \mathcal{B}\Omega^2 + \mathcal{D}_{\mathrm{R}}\Omega^2, \tag{4.10a}$$

$$\hat{\Omega}^{(j)}_{\mathrm{rad}} = \mathcal{A}_j - \mathcal{C}_j - \mathcal{E}_j - \mathcal{F} - \mathcal{S}. \tag{4.10b}$$

This summation implied by Eq. (4.8) is motivated by Eqs. (3.6), (3.13), (3.21), (3.49), (3.56), (3.72), (3.88), (3.66), (3.94), and (3.98). In Eq. (4.10b), the symbol $\hat{\Omega}^{(j)}_{\mathrm{rad}}$ indicates a relative modification of the Rabi frequency, i.e., a dimensionless quantity.

We have thus summed all the radiative corrections as effective corrections to the Rabi frequency and the detuning. Of course, the mixing angle θ as defined in Eqs. (2.9) and

(2.13b) is changed by the radiative corrections. Indeed, one may evaluate the corrected θ by employing the relation $\tan(2\theta)=-\Omega_{\rm corr}/\Delta_{\rm corr}$, where $\Delta_{\rm corr}=\Delta-\Delta_{\rm rad}^{(j)}$ and $\Omega_{\rm corr}=\Omega\cdot(1+\hat{\Omega}_{\rm rad}^{(j)})$ are the relativistically and radiatively corrected Rabi frequency and the detuning, respectively [see Eqs. (4.10b) and (4.10a)]. Because all relativistic and radiative corrections find a natural interpretation as corrections to the Rabi frequency and the detuning, the corrected dressed states have the same structure as Eqs. (2.14a) and (2.14b), but with relativistic and radiative wave function corrections a corrected mixing angle.

All relativistic and radiative corrections to the Rabi frequency and the detuning have been evaluated here using the unperturbed mixing angle θ. The "corrections to the corrections" due to an evaluation of modifications to the Rabi frequency and the detuning in terms of the corrected mixing angle are of higher order than the terms relevant for the discussion in the current paper and may be neglected on the level of approximation employed in the current investigation.

In the following numerical analysis, we assume the atom to be located at an antinode of a laser field in standing-wave configuration. The atom is thus driven by two counterpropagating laser beams, whereas in the definitions of the electric field and the Rabi frequency in Sec. II and especially in the matching of the classical macroscopic field with the corresponding quantum counterpart in Eq. (2.12) a single-mode running-wave field was considered. However, all results of Sec. III also apply to a standing-wave field configuration if the macroscopic electric-field strength $\mathcal{E}_{\rm L}$ is taken to be the total field strength of both counterpropagating field modes at the position of the atom. In order to avoid confusions, from now on we thus denote the total electric-field strength of the standing wave as $\mathcal{E}_{\rm SW}$.

A. $1S_{1/2}\leftrightarrow 2P_{1/2}$

For the $|2P_{1/2},m=\pm 1/2\rangle$ state as upper state, the decay constant is given by $\Gamma_{1/2}=99.709\,42(1)\times 10^6$ Hz [50]. In order to account for the dependence on the laser-field intensity, we introduce the parameter $h_{1/2}=|\Omega|/\Gamma_{1/2}$. Then for $h_{1/2}=1000$ and $\Delta=50\Gamma_{1/2}$, one has $\omega_{\rm R}\gg\Omega\gg\Gamma_{1/2},\Delta$. Therefore, the relative corrections to the detuning and the Rabi frequency in Eqs. (4.10a) and (4.10b) become

$$\Delta_{\rm rad}^{(1/2)}=-8.175\,249(33)\times 10^9\ {\rm Hz}, \tag{4.11a}$$

$$\Omega_{\rm rad}^{(1/2)}=-19.78(56)\times 10^{-6}. \tag{4.11b}$$

Here, the parameter $h_{1/2}$ may be expressed in terms of the electric-field strength $\mathcal{E}_{\rm L}$ as

$$h_{1/2}=346.783\times 10^{-6}|\mathcal{E}_{\rm SW}\ ({\rm V/m})|. \tag{4.12}$$

Of course, [V/m] in this case means that the peak electric-field strength of the laser is assumed to be measured in volts per meter.

The absolute ionization rate $\mathcal{I}$ into the continuum due to the driving-laser field is given by $\mathcal{I}=\mathcal{D}_{\rm I}\Omega^2$. In an experiment, this ionization rate has to be much smaller than the Rabi frequency, such that on average the atom undergoes many fluorescence cycles before it is ionized. Thus, we define the ratio

$$\mathcal{I}_\Omega=\left|\frac{\mathcal{I}}{\Omega}\right|=|\mathcal{D}_{\rm I}|\Omega=|\mathcal{D}_{\rm I}|h_{1/2}\Gamma_{1/2}, \tag{4.13}$$

which has to be much smaller than unity ($\mathcal{I}_\Omega\ll 1$). For $h_{1/2}=1000$, one obtains

$$\mathcal{I}(h_{1/2}=1000)=356\ {\rm kHz}, \tag{4.14}$$

$$\mathcal{I}_\Omega(h_{1/2}=1000)=3.6\times 10^{-6}, \tag{4.15}$$

which means that the probability of one-photon ionization does not restrict the above measurement scheme.

For $h_{1/2}=1000$ and $\Delta=50\Gamma_{1/2}$, the theoretical prediction for the shift of the Mollow sidebands relative to the central Mollow peak by the generalized corrected Rabi frequency is as follows:

$$\pm\Omega_{\mathcal{C}}^{(1/2)}=\pm 100.572\,258(60)\times 10^9\ {\rm Hz}. \tag{4.16}$$

This formula has been evaluated using the summation formula Eq. (4.8) and includes all corrections, in particular the $\mathcal{C}$-term evaluated in Sec. III B 3. For comparison, we also give here a theoretical prediction that would be obtained by ignoring the $\mathcal{C}$ term,

$$\pm\Omega_{{\rm no}\,\mathcal{C}}^{(1/2)}=\pm 100.572\,377(27)\times 10^9\ {\rm Hz}. \tag{4.17}$$

This result is obtained by explicitly setting $\mathcal{C}_j$ in Eq. (4.10b) equal to zero, but still using the full summation according to Eq. (4.9) for all other corrections. A comparison of Eq. (4.16) to Eq. (4.17) shows that the theoretical uncertainties of the two results do not overlap. Therefore, the current status of the theory would allow to discern the presence or absence of the $\mathcal{C}$-term corrections by means of an accurate experiment.

The principal uncertainty of the generalized Rabi frequency in Eq. (4.16) is due to the uncertainty in the estimate of the nonlogarithmic contribution to $\mathcal{C}_j$ and $\mathcal{A}_j$. As a reference, the bare shift without corrections is given by

$$\pm\sqrt{\Omega^2+\Delta^2}=\pm 99.833975\times 10^9\ {\rm Hz}. \tag{4.18}$$

This result has to be compared to the radiatively corrected result (4.16).

In Table I, the individual shifts due to the considered corrections are listed together with their respective uncertainties for both transitions $1S_{1/2}\leftrightarrow 2P_{1/2}$ and $1S_{1/2}\leftrightarrow 2P_{3/2}$. All shifts are evaluated in their "summed" form [see, e.g., Eq. (3.13)]. For the bare Lamb-shift corrections $\mathrm{L}_{\rm bare}^{(j)}$, the uncertainty in the shift is mainly due to the uncertainty in the numerical value for the Lamb shift of the hydrogen $1S$ state, see Eq. (3.10). The Bloch-Siegert shift acquires a numerical uncertainty due to neglected terms of relative order $O(\Omega/\omega_{\rm L})$ from the higher-order Bloch-Siegert-type shifts (higher-order perturbation theory in the counter rotating terms). There is a further source of uncertainty for the Bloch-Siegert shifts due to terms of relative order $O(\Delta/\omega_{\rm L},\Omega/\omega_{\rm L})$ from the expansion leading from Eq. (3.19) to Eq. (3.20). The shift due to off-resonant excitation $\mathcal{D}$ has an uncertainty because contributions to the energies Eq. (3.26) of relative order $\Delta/\omega_{\rm R}$ and

of order Ω/ω_R have been neglected in evaluating the t parameters in Eq. (3.47). There are also uncertainties of the Rabi-frequency shifts due to relativistic corrections to the transition dipole matrix elements ($\mathcal{E}_j$), which are due to neglected higher-order corrections of relative order $(Z\alpha)^2$ with respect to the leading corrections. The field-dependent corrections have an uncertainty due to higher-order effects of relative order $(Z\alpha)^2$. The main uncertainty of the $\mathcal{C}$-term and $\mathcal{A}_j$-term corrections are due to the uncertainty which we assign to the nonlogarithmic contribution of relative parametric order $\alpha(Z\alpha)^2$ [see Eqs. (3.86) and (3.92)]. The main uncertainty of the shift due to corrections to the secular approximation ($\mathcal{S}$) are due to higher-order terms of the expansion leading from Eq. (3.96) to Eq. (3.97) or relative order $O(\Delta^2/\Omega^2)$, and of fourth-order corrections to the secular approximation of relative order $O(\Gamma^2/\Omega^2)$. For the entries of Table I, the shifts have been evaluated individually according to Eqs. (3.6), (3.13), (3.21), (3.49), (3.56), (3.66), (3.72), (3.88), (3.94), and (3.98).

It is perhaps worthwhile to note that one cannot simply add the corrections to the quasienergy of dressed states in the same sense as corrections to the energy of bare atomic states. For the evaluation of a theoretical Lamb-shift prediction of a bare atomic state, the usual procedure is to list the various corrections and to simply add these in order to give a theoretical prediction for the total energy shift (see, e.g., Tables I and II of [34]). For the Lamb shift of laser-dressed states, the natural interpretation of the corrections implies modifications of the decisive physical parameters that determine the quasienergy of the dressed levels: These are the Rabi frequency Ω and the detuning Δ. This interpretation implies, however, summations of the expressions which agree well with the first-order results, so that at least for numerically small corrections, it is still permissible to simply add the correction terms in an approximative sense. The correspondence of first-order expressions and summed results holds approximately unless the correction is large compared to Ω and Δ; in this case, the summation yields a different result as compared to the first-order expression. This is the case for the numerically dominant effect referred to as $d\overline{\omega}_{+,j}^{(\mathrm{Lamb})}$ in Table I. As already discussed in Secs. III A 2 and III A 3, this summation is somewhat nontrivial in particular for the Lamb-shift corrections. Because fundamental symmetry properties prevent the radiative corrections from coupling S and P states (even in higher order), preference is given to the summed results. In principle, more explicit higher-order calculations outlined in Sec. III A 3 would be desirable to verify the summations for all radiative effects discussed here.

B. $1S_{1/2} \leftrightarrow 2P_{3/2}$

In this section, we repeat the above numerical analysis with $|1S_{1/2}, m=\pm\frac{1}{2}\rangle$ as the ground state and $|2P_{3/2}, m=\pm\frac{1}{2}\rangle$ as the excited state. The decay width of the $2P_{3/2}$ state is given by $\Gamma_{3/2}=99.709\,42(1)\times 10^6$ Hz [50]. In order to account for the dependence on the laser-field intensity, we introduce the parameter $h_{3/2}=|\Omega|/\Gamma_{3/2}$. Then for $h_{3/2}=1000$ and $\Delta=50\Gamma_{3/2}$, one has $\omega_R \gg \Omega \gg \Gamma_{3/2}, \Delta$ such that the corrections to the detuning and the Rabi frequency in Eqs. (4.10a) and (4.10b) become

$$\Delta_{\mathrm{rad}}^{(3/2)} = -\,8.149\,896(33)\times 10^9 \text{ Hz}, \tag{4.19a}$$

$$\Omega_{\mathrm{rad}}^{(3/2)} = -\,20.76(56)\times 10^{-6}. \tag{4.19b}$$

Here, the parameter $h_{3/2}$ may be expressed in terms of the electric-field strength $\mathcal{E}_L$ as

$$h_{3/2} = 490.425\times 10^{-6}\,|\mathcal{E}_{\mathrm{SW}}\ (\mathrm{V/m})|. \tag{4.20}$$

For this transition, the ionization to the continuum is again given by

$$\mathcal{I}(h_{3/2}=1000) = 356 \text{ kHz}, \tag{4.21}$$

$$\mathcal{I}_\Omega(h_{3/2}=1000) = 3.6\times 10^{-6}. \tag{4.22}$$

Thus the scheme is not restricted by ionization on this transition either.

With the above parameters for the Rabi frequency and the detuning, the positions of the Mollow sidebands relative to the center component with full corrections, without $\mathcal{C}$-term corrections and without any corrections are given by

$$\pm\Omega_{\mathcal{C}}^{(3/2)} = \pm\,100.568\,846(60)\times 10^9 \text{ Hz}, \tag{4.23}$$

$$\pm\Omega_{\mathrm{no}\ \mathcal{C}}^{(3/2)} = \pm\,100.568\,966(27)\times 10^9 \text{ Hz}, \tag{4.24}$$

$$\pm\sqrt{\Omega^2+\Delta^2} = \pm\,99.833\,975\times 10^9 \text{ Hz}. \tag{4.25}$$

Thus also in this case the theoretical uncertainties of the generalized corrected Rabi frequency in Eq. (4.23) and of the corresponding value in Eq. (4.24) obtained by ignoring the $\mathcal{C}$-term correction shift do not overlap. The individual shifts with their uncertainties are listed in Table I as discussed in Sec. IV A.

V. DISCUSSION AND SUMMARY

In this paper we have discussed radiative corrections to the usual quantum optical expression for the Mollow spectrum, i.e., the resonance fluorescence spectrum of an atom with two relevant energy levels driven by a strong coherent laser field. To lowest order, the Mollow spectrum consists of one main peak which is centered at the frequency of the driving-laser field and of two sidebands, which are shifted from the center by the generalized Rabi frequency $\Omega_R=\sqrt{\Omega^2+\Delta^2}$. For the analysis, we have used concepts introduced originally in two different fields: the dressed-state formalism of quantum optics and the renormalized radiative corrections which are treated in the formalism of quantum electrodynamics. Throughout the analysis, we have adopted the dressed-state basis as the natural starting point for our analysis of corrections to the quasienergies of the combined atom-laser system.

From the point of view of spectroscopy, the Mollow spectrum is attractive for several reasons. First of all, the radiative corrections manifest themselves in the shift of the Mollow sidebands with respect to the central Mollow peak. Thus

the tiny radiative corrections are measured relative to the generalized Rabi frequency, which for typical parameters of the driving-laser field is several orders of magnitude smaller than optical frequencies. Also, the Mollow spectrum is centered around the frequency of the driving-laser field. Thus it is a kind of a differential spectrum because the laser-field detuning is automatically subtracted.

As for quantum electrodynamics, radiative corrections to the Mollow spectrum are a quantum-field theoretic problem in the presence of two classical background fields, the laser field and the binding Coulomb field. Our analysis also differs from typical QED calculations relying on the *S*-matrix formalism, as the process under study is dynamical. In order to account for the quantum fluctuations of the dipole moment leading to the incoherent part of the Mollow spectrum, a static description is not sufficient.

The shifts of the Mollow sidebands may be interpreted as arising from corrections to either the detuning or the Rabi frequency. In particular, the detuning is modified by the bare Lamb-shift, Bloch-Siegert shifts, and virtual off-resonant excitations. The Rabi frequency is corrected by relativistic and radiative corrections to the transition dipole moment, by field-configuration-dependent corrections, by a dynamic correction, and by corrections to the secular approximation. Of particular interest is the dynamical correction to the Rabi frequency. This correction arises from an evaluation of the second-order radiative self-energy corrections of the combined system of atom and laser field in terms of the dressed states of this system. To lowest order of the limit $\Omega,\Delta \ll \omega_{eg}$, this yields a corrections which can be identified with the usual Lamb shift of the atomic bare states. The dynamic correction is then obtained by keeping terms linear in Ω, Δ in the above analysis and cannot be explained in terms of the bare state Lamb shift alone.

The corrections to the detuning may be incorporated into the analysis by the replacement $\left(j=\frac{1}{2},\frac{3}{2}\right)$

$$\Delta \to \Delta - \Delta^{(j)}_{\rm rad}, \tag{5.1}$$

where $\Delta^{(j)}_{\rm rad}$ is defined in Eq. (4.10a). Correspondingly, the corrections to the Rabi frequency are given by

$$\Omega \to \Omega(1+\hat{\Omega}^{(j)}_{\rm rad}). \tag{5.2}$$

The dimensionless quantity $\hat{\Omega}^{(j)}_{\rm rad}$ is defined in Eq. (4.10b). Then, the generalized Rabi frequency supplemented by the discussed relativistic and radiative corrections is [Eq. (4.9)]

$$\Omega^{(j)}_{\mathcal{C}} = \sqrt{\Omega^2(1+\hat{\Omega}^{(j)}_{\rm rad})^2 + (\Delta - \Delta^{(j)}_{\rm rad})^2}. \tag{5.3}$$

In a numerical analysis, we provide a theoretical analysis which is required in order to accurately resolve the dynamical shift. For this, we suppose the driving-laser fields to be in a standing-wave configuration. As a promising candidate for the experiment, we identify the hydrogen $1S_{1/2}\leftrightarrow 2P_{1/2}$ and $1S_{1/2}\leftrightarrow 2P_{3/2}$ transition. The results are discussed for a driving-laser field parameter set which is expected to be within reach of improvements of the currently available Lyman-α laser sources in the next few years.

For the $1S_{1/2}\leftrightarrow 2P_{1/2}$ transition and for $\Omega=1000\Gamma_{1/2}$, $\Delta=50\Gamma_{1/2}$, the Rabi frequency is shifted with respect to $\Omega_{\rm R}=\sqrt{\Omega^2+\Delta^2}$ by relativistic and radiative corrections as follows,

$$\pm(\Omega^{(1/2)}_{\mathcal{C}} - \Omega_{\rm R}) = \pm 738.282(60)\times 10^6\ {\rm Hz}. \tag{5.4}$$

The corresponding result for the $1S_{1/2}\leftrightarrow 2P_{3/2}$ transition with $\Omega=1000\Gamma_{3/2}$, $\Delta=50\Gamma_{3/2}$ is

$$\pm(\Omega^{(3/2)}_{\mathcal{C}} - \Omega_{\rm R}) = \pm 734.871(60)\times 10^6\ {\rm Hz}. \tag{5.5}$$

We note, however, that we are only concerned with theoretical issues. Thus uncertainties due to possible experimental issues such as a misalignment of the apparatus or due to additional trapping potentials have not been considered.

In summary, we have presented a detailed analysis of the leading nonrelativistic and relativistic corrections to the Mollow spectrum. The analysis includes the relativistic and nonrelativistic corrections up to relative orders of $(Z\alpha)^2$ and $\alpha(Z\alpha)^2$, respectively, and also includes Bloch–Siegert shifts, stimulated radiative corrections involving off-resonant virtual states, field-configuration-dependent corrections and corrections to the secular approximation. Based on these results, we provide a numerical analysis of the corrections of the Mollow spectrum of the hydrogen $1S$-$2P$ transition. By a comparison with experimental data, one may verify the presence of dynamical leading-logarithmic correction to the dressed-state radiative shift, which cannot be explained in terms of the bare Lamb shift (see Sec. III B 3). This allows us to address questions related to the physical reality of the dressed states. On the other hand, the comparison with experimental results could also be used to interpret the nature of the evaluated radiative corrections in the sense of the summation formulas which lead to the interpretation of the shifts as arising from relativistic and radiative corrections to the detuning and the Rabi frequency.

ACKNOWLEDGMENTS

Financial support by the German Science Foundation (SFB 276 and KE 721/1-1) is gratefully acknowledged. J.E. was supported by the German National Academic Foundation. U.D.J. acknowledges helpful conversations with Holger Gies and Wilhelm Becker regarding the choice of gauge in the description of dynamical processes.

APPENDIX A: DIPOLE MOMENTS AND SPIN

The spontaneous emission decay rate of the population of an excited state $|i\rangle$ to a final state $|f\rangle$ is given by

$$\Gamma \propto \omega^3 |\langle f|\boldsymbol{x}|i\rangle|^2, \tag{A1}$$

where all elements of the position vector have to be considered in the coupling with the vacuum field. For the $2P\to 1S$ decay in atomic hydrogen, and in the nonrelativistic Schrödinger theory without spin, the squared modulus of the dipole moment vector is given by

$$\sum_{i=1}^{3} |\langle 1S|x^i|2P\rangle|^2 = \frac{2^{15}}{3^{10}}\frac{1}{(Z\alpha)^2 m^2}, \tag{A2}$$

independent of the magnetic quantum number $m_i \in \{-1,0,1\}$ of the initial P state (by the term final state we

will denote in this section the particular state that enters as a "bra-" in the Dirac notation, i.e., the $1S$ state in the above case). In the Schrödinger-Pauli theory with spin, one has different decay channels depending on the spin state of the initial and the final state. For the channel where the initial and the final state have the same magnetic quantum number $m_i=m_f=\pm\frac{1}{2}$, one obtains

$$\sum_{i=1}^{3}\left|\langle 1S_{1/2},m_i|x^i|2P_{1/2},m_i\rangle\right|^2=\frac{1}{3}\frac{2^{15}}{3^{10}}\frac{1}{(Z\alpha)^2m^2}. \tag{A3}$$

In contrast, the channel with opposite magnetic quantum number $m_i=-m_f=\pm\frac{1}{2}$ yields a contribution of

$$\sum_{i=1}^{3}\left|\langle 1S_{1/2},-m_i|x^i|2P_{1/2},m_i\rangle\right|^2=\frac{2}{3}\frac{2^{15}}{3^{10}}\frac{1}{(Z\alpha)^2m^2}. \tag{A4}$$

This calculation predicts that if one were to measure the electron spin polarization in the final state, then the $|2P_{1/2}\rangle$ state would be twice as likely to decay into a $|1S_{1/2}\rangle$ state with opposite total electron angular momentum than into a $|1S_{1/2}\rangle$ state with the same total electron angular momentum as the initial $|2P_{1/2}\rangle$ state. Adding the two decay channels to the final state, we obtain

$$\sum_{m_f=\pm 1/2}\sum_{i=1}^{3}\left|\langle 1S_{1/2},m_f|x^i|2P_{1/2},m_i\rangle\right|^2=\frac{2^{15}}{3^{10}}\frac{1}{(Z\alpha)^2m^2}, \tag{A5}$$

i.e., the same result as in the spinless case, as it should be. For the decay of the $|2P_{3/2},m_i=\pm\frac{1}{2}\rangle$ state, one obtains the same total decay rate, but here the decay with $m_f=m_i$ is twice as likely as the decay with $m_f=-m_i$.

These results for the dipole moments have to be reconsidered for excitation of an atom in the $|1S_{1/2},m_i\rangle$ ground state with a laser field which is linearly polarized in one direction, say the z direction. Then other than for the interaction with the vacuum field, only the z component of the dipole moment vector has to be considered. These matrix elements can be used to calculate the Rabi flopping frequency corresponding to the driving laser field. For the spinless case, one obtains

$$\langle 1S|z|2P\rangle=\frac{2^7}{3^5}\sqrt{2}\frac{1}{Z\alpha m}. \tag{A6}$$

We now include the spin and choose a definite initial state $|1S_{1/2},m_i=+\frac{1}{2}\rangle$. We obtain

$$\left\langle 1S_{1/2},\tfrac{1}{2}\middle|z\middle|2P_{1/2},\tfrac{1}{2}\right\rangle=-\frac{2^7}{3^5}\sqrt{\frac{2}{3}}\frac{1}{Z\alpha m}, \tag{A7}$$

where we have omitted the "$m_i=$" ("$m_f=$") from the initial (final) state vector. Furthermore, one has

$$\left\langle 1S_{1/2},\tfrac{1}{2}\middle|z\middle|2P_{1/2},-\tfrac{1}{2}\right\rangle=0, \tag{A8}$$

$$\left\langle 1S_{1/2},\tfrac{1}{2}\middle|x\middle|2P_{1/2},\tfrac{1}{2}\right\rangle=0, \tag{A9}$$

$$\left\langle 1S_{1/2},\tfrac{1}{2}\middle|y\middle|2P_{1/2},\tfrac{1}{2}\right\rangle=0, \tag{A10}$$

so that the z-polarized field only couples the $2P_{1/2}$ state with $m_i=m_f$ to the ground state, and this excited state is only one coupled to the ground state by z-polarized light. For the $|2P_{3/2},m_f\rangle$ upper state, the corresponding results are

$$\left\langle 1S_{1/2},\tfrac{1}{2}\middle|z\middle|2P_{3/2},\tfrac{1}{2}\right\rangle=\frac{2^8}{3^5}\sqrt{\frac{1}{3}}\frac{1}{Z\alpha m}, \tag{A11}$$

$$\left\langle 1S_{1/2},\tfrac{1}{2}\middle|z\middle|2P_{3/2},-\tfrac{1}{2}\right\rangle=0, \tag{A12}$$

$$\left\langle 1S_{1/2},\tfrac{1}{2}\middle|x\middle|2P_{3/2},\tfrac{1}{2}\right\rangle=0, \tag{A13}$$

$$\left\langle 1S_{1/2},\tfrac{1}{2}\middle|y\middle|2P_{3/2},\tfrac{1}{2}\right\rangle=0, \tag{A14}$$

$$\left\langle 1S_{1/2},\tfrac{1}{2}\middle|z\middle|2P_{3/2},\pm\tfrac{3}{2}\right\rangle=0. \tag{A15}$$

Thus also in this case only the upper state with same magnetic quantum number is coupled to the ground state, but with a matrix element which differs by a factor of $-\sqrt{2}$ in magnitude from the corresponding result for the $|2P_{1/2},\frac{1}{2}\rangle$ upper state. From these spin-resolved results, the corresponding matrix element without spin Eq. (A6) may be obtained by summing over the final states and averaging over the initial states.

APPENDIX B: EVALUATION OF THE MATRIX ELEMENTS

In this section, we demonstrate the evaluation of the matrix element in Eq. (3.32):

$$M_e=\langle e|zG''(\zeta)z|e\rangle, \tag{B1}$$

where $|e\rangle$ is the $2P$, $m=0$ state. We start by calculating the "unreduced" matrix element

$$\overline{M}_e(\zeta)=\langle e|zG(\zeta)z|e\rangle, \tag{B2}$$

where the full sum over intermediate states is employed in $G(\zeta)$, and the wave function of the excited state $\Phi_{2P,m=0}$ is given by a product of a radial and an angular contribution:

$$\Phi_{2P,m=0}(\vec{r})=R_{2P}(r)Y_{10}(\theta,\phi).$$

Here, (r,θ,ϕ) are spherical coordinates. In these coordinates, a representation of the Green function in position space is given by [Ref. [45], Eq. (2.2)]

$$\frac{1}{H-E}=\sum_{l,m}g_l(r_1,r_2,v)Y_{lm}(\theta_1,\phi_1)Y^*_{lm}(\theta_2,\phi_2),$$

where $g_n(r_1,r_2,v)$ is the radial component of the Schrödinger-Coulomb propagator

$$g_l(r_1,r_2,v) = 2m\left(\frac{2}{a_B v}\right)^{2l+1}(r_1 r_2)^l \exp\left(-\frac{r_1+r_2}{a_B v}\right) \times \sum_{k=0}^{\infty} \frac{k!\; L_k^{2l+1}\left(\frac{2r_1}{a_B v}\right) L_k^{2l+1}\left(\frac{2r_2}{a_B v}\right)}{(2l+1+k)!\,(l+1+k-v)} \quad \text{(B3)}$$

containing associated Laguerre polynomials $\mathrm{L}_a^b(r)$. The quantity $v=(a_B\sqrt{-2mE})^{-1}$ is an energy parameter which is related to the parameter t used in Sec. III A 5 by $v=nt$, where n is the principal quantum number of the initial bound state. The Bohr radius is defined in Eq. (3.44), and we evaluate all matrix elements here for the case $Z=1$ (atomic hydrogen). Thus, for the $2P$ state discussed here, we have $v=2t$. The index l is summed over all possible angular momentum numbers of the virtual intermediate states. Starting from a P state with $l=1$, both $S(l=0)$ and $D(l=2)$ states are possible as intermediate states. The integration may be further separated in angular and radial parts:

$$\overline{M}_e = \overline{M}_{l=0}^{\mathrm{ang}}\overline{M}_{l=0}^{\mathrm{rad}} + \overline{M}_{l=2}^{\mathrm{ang}}\overline{M}_{l=2}^{\mathrm{rad}}, \quad \text{(B4)}$$

where the angular integrations yield

$$\overline{M}_{l=0}^{\mathrm{ang}} = \frac{1}{3}, \quad \overline{M}_{l=2}^{\mathrm{ang}} = \frac{4}{15}. \quad \text{(B5)}$$

The radial parts may written explicitly as

$$\overline{M}_l^{\mathrm{rad}} = \int_0^\infty dr_1 dr_2 r_1^3 r_2^3 R_{2P}^*(r_1) R_{2P}(r_2) g_l(r_1,r_2,v), \quad \text{(B6)}$$

for $l=0,2$. Simplifying further, one obtains

$$\overline{M}_{l=0}^{\mathrm{rad}} = \frac{m}{12ta_B^6}\sum_{k=0}^{\infty}\frac{k!}{(1+k)!\,(1+k-2t)} I_0^2, \quad \text{(B7)}$$

$$I_0 = \int_0^\infty dr\; r^4 \mathrm{L}_k^1\left(\frac{r}{a_B t}\right) e^{[-(1+t)/2a_B t]r}. \quad \text{(B8)}$$

The integral in I_0 can be evaluated using (see [51], Sec. 6.10 and [52], Sec. 10.12)

$$\int_0^\infty dr \exp(-\lambda r) r^\gamma \mathrm{L}_n^\mu(r) = \frac{\lambda^{-1-\gamma}\Gamma(\gamma+1)}{n!\;\Gamma(\mu+1)}\Gamma(\mu+n+1)\,{}_2F_1(-n,\gamma+1,\mu+1,\lambda^{-1}) \quad \text{(B9)}$$

to give

$$I_0 = \frac{768a_B^5 t^5}{(1+t)^5}\frac{\Gamma(k+2)}{k!}\,{}_2F_1\left(-k,5,2;\frac{2}{1+t}\right). \quad \text{(B10)}$$

Using an explicit expression for the hypergeometric function ([51], Sec. 2.1.1), we obtain

$${}_2F_1(a,b,c;z) = \sum_{j=0}^{\infty}\frac{(a)_j (b)_j}{(c)_j}\frac{z^j}{j!}, \quad \text{(B11)}$$

where the Pochhammer symbols $(a)_j$ are given by

$$(a)_j = \frac{\Gamma(a+j)}{\Gamma(a)}. \quad \text{(B12)}$$

Contiguous relations for the hypergeometric function ([51], Sec. 2.8) then lead to

$$\overline{M}_{l=0}^{\mathrm{rad}} = ma_B^4\left(\frac{16t^2\mathcal{X}_0(t)}{3(t-1)^6(t+1)^4} - \frac{2^{14}t^{11}}{3(t^2-1)^6}\Phi(2,t)\right),$$

$$\mathcal{X}_0(t) = 45 - 90t - 84t^2 + 258t^3 + 18t^4 - 294t^5 + 148t^6 - 2t^7 + 257t^8,$$

where the hypergeometric function $\Phi(n,t)$ is defined in Sec. III A 5. A similar calculation for the intermediate $D(l=2)$ states yields

$$\overline{M}_{l=2}^{\mathrm{rad}} = ma_B^4\left(\frac{16t^2\mathcal{X}_2(t)}{3(t-1)^7(t+1)^5} - \frac{2^{16}t^{11}(4t^2-1)}{3(t^2-1)^7}\Phi(2,t)\right),$$

$$\mathcal{X}_2(t) = -45 + 90t + 165t^2 - 420t^3 - 174t^4 + 768t^5 - 34t^6 - 700t^7 - 37t^8 - 1274t^9 + 4733t^{10}.$$

Inserting in Eq. (B4) finally yields the expression in Eq. (3.41b). The reduced matrix element can then be obtained from this by subtracting the contributions of the two intermediate states $|e\rangle,|g\rangle$. The excited state contribution vanishes due to parity, and the ground state contribution is given by

$$\frac{|\langle g|z|e\rangle|^2}{E_{1S}-\zeta}. \quad \text{(B13)}$$

This term, which cancels the divergence as $\zeta\to E_{1S}$ in (3.46), may be verified by inserting the resonant $1S$ state as the intermediate state into the matrix element. Alternatively, the cancellation may be seen as follows: On setting the intermediate state energy to $E_{1S}+\epsilon$, the series expansion of the unreduced matrix element $\overline{M}_e(E_{1S})$ receives a contribution proportional to $1/\epsilon$ which diverges for $\epsilon\to 0$. This diverging part is canceled by the intermediate state contribution Eq. (B13) in the reduced matrix element $M_e(\zeta)$ to give a finite result.

If one compares this derivation with a similar calculation for the standard matrix element

$$\overline{\mathcal{M}} = \sum_{i=1}^{3}\langle e|x^i G''(\zeta) x^i|e\rangle \quad \text{(B14)}$$

$$= \bar{\mathcal{M}}^{\mathrm{ang}}_{l=0}\bar{\mathcal{M}}^{\mathrm{rad}}_{l=0} + \bar{\mathcal{M}}^{\mathrm{ang}}_{l=2}\bar{\mathcal{M}}^{\mathrm{rad}}_{l=2}, \qquad \text{(B15)}$$

where all polarization directions are considered, one findsthat the respective radial parts for $l=0$ and $l=2$ are identical to the ones in Eq. (B4):

$$\bar{\mathcal{M}}^{\mathrm{rad}}_{l=0} = \bar{M}^{\mathrm{rad}}_{l=0}, \quad \bar{\mathcal{M}}^{\mathrm{rad}}_{l=2} = \bar{M}^{\mathrm{rad}}_{l=2}. \qquad \text{(B16)}$$

For the angular parts, however, one finds

$$\bar{\mathcal{M}}^{\mathrm{ang}}_{l=0} = \bar{M}^{\mathrm{ang}}_{l=0}, \quad \bar{\mathcal{M}}^{\mathrm{ang}}_{l=2} = \frac{5}{2}\bar{M}^{\mathrm{ang}}_{l=2}. \qquad \text{(B17)}$$

The reason for this is that the fixed polarization of the driving laser field only allows to excite one of the magnetic sublevels of the intermediate *S* and *D* states. The sum over all polarizations in the standard matrix element still only gives one magnetic sublevel for the intermediate *S* states, but three possible virtual *D* states. Due to this asymmetry the desired matrix element Eq. (3.41b) cannot be calculated directly from the standard matrix element in Eq. (B14).

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