Self-induced ac Stark shift in lasers

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The ac Stark effect is a well-known, well-established effect associated with the displacement of the atomic energy levels in the presence of an electromagnetic (e.m.) field. As the origin of the e.m. field is irrelevant, a self-induced ac Stark effect should be present in a laser. However, not only has this effect never been reported but it is not included in any laser model. Here, we show that a self-induced ac Stark shift does exist in the theoretical description of a two-level laser, provided that the reduced equations are derived not from the rotating-wave approximation, but from the full density matrix formulation. Interesting fundamental consequences are discussed and quantitative estimates of the self-induced frequency shift resulting from the laser field's ac Stark shift are obtained for a variety of lasers. We find that the effect is generally small but (at least in some cases) measurable and that the transfer of laser amplitude noise into frequency noise may play a role in future, ultrahigh-precision metrological applications.

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I. INTRODUCTION

The year 2010 marks the 50th anniversary of the experimental realization of the laser and the beginning of a very successful history. Nonetheless, lasers remain very complex systems, which entail the synchronization of a large number of oscillators under far-from-equilibrium conditions. Thus, describing their behavior has represented a challenge, met by a diversity of models each adapted to meet the corresponding level of complexity.

Models range from phenomenological rate equations to first-principle approaches based on Maxwell-Bloch equations [1] for unidirectional single longitudinal and transverse mode ring cavities, subsequently extended to multilongitudinal mode regimes [2], to descriptions based on synchronous boundary conditions [3], to the full spatial three-dimensional (3D) description [4], which opens the path to a wide range of investigations [5,6]. Adiabatic variable reductions have been used to simplify the mathematical description, thereby introducing a natural classification for the different kinds of lasers [7].

Approximations have obviously played a crucial role in model development: first and foremost the rotating-wave approximation (RWA) and the slowly varying envelope approximation (SVEA) [8]. Furthermore, modern mathematical techniques, which allow for a precise and rigorous evaluation of the manifolds over which the dynamics evolve, have improved the description of the laser threshold as a bifurcation. The so-called improved adiabatic elimination has identified the true directions in phase space corresponding to *fast* and *slow* dynamics in Class B lasers [7], thereby allowing for a better description of their dynamics through the Toda potential and providing analytical quantitative predictions for the oscillation frequency [9,10], as well as predicting anomalous frequency shifts in a laser with an injected signal [11].

Center manifold techniques [12,13] allow for a parallel to be drawn between threshold crossing in a spatially extended laser and fluids, superfluids, and magnetic systems, predicting the appearence of *optical vortices* [14] and thereby sparking a flurry of activity in a field which remains still very active [15]. These same techniques have been instrumental in establishing correct models for the long-term stability of spatiotemporal systems, avoiding spurious results and reducing the numerical computing time [16,17], or determining the stabilization conditions [18] for the formation of spatiotemporal patterns in large-aperture lasers (e.g., semiconductors).

One feature common to the various derivations of the reduced form of the laser equations through center manifold techniques is that the resulting Ginzburg-Landau equation predicts an optical frequency which does not depend on the amplitude of the laser field [14,18,19]. This result is quite peculiar because (1) it drastically differs from what is observed in all other nonlinear physical systems (starting from the simple pendulum in a gravitational field) and (2) a renormalization of the laser frequency is expected as a consequence of the atomic level displacement which should occur in the presence of an electromagnetic (e.m.) field. This physical mechanism is very well known in spectroscopy and atomic physics and takes the name of Rabi shift, light shift or ac Stark shift, and it is routinely calculated in first-order perturbation theory [20–22]. In lasers the complication is that the e.m. field is not externally imposed, but, rather, self-consistently generated, implying therefore a *self-induced ac Stark shift*, if present.

A question naturally arises: Is there a physical reason why lasers should possess such an uncommon independence of the e.m. field frequency on its amplitude, or may this result be due to a mathematical artefact introduced in all models which have been used so far for their description?

Historically, laser models have been based on the description of a two-level atom, which are grounded on the theory developed for spin systems. The problem of spin nutation was extensively studied starting from the 1930s, notably by Rabi and co-workers [23]. The first experimental measurements of nutation involved suitably tailored static magnetic fields, through which atoms moved with selected speed, thereby generating—in the reference frame of the atom—a variable magnetic (driving) field [24]. As an alternative, radio frequency (rf) generation offered a much more flexible and accurate control of the driving signal without the need for velocity selection in the atomic sample. However, the oscillating signal carried, as a complication, the appearance of two spectral Fourier components in the rf field.

From a mathematical point of view, the problem is solved in approximate form (the RWA) by choosing a reference frame in which one Fourier component is stationary and the other rotates at double its frequency. In this approximation, the latter component is neglected since its average rapidly tends to zero over the long time scales which characterize the now quasistationary component. This approximation, born within the framework of magnetic resonance, is extremely common in quantum optics and atomic and laser physics and is known to provide excellent results. Nevertheless, it remains an approximation [20]. This fact was first pointed out by Bloch and Siegert [25], and independently by Stevenson [26], who evaluated its impact on the nutation of nuclear spins and calculated the frequency shift resulting from the full account of the rf terms; they concluded that for nuclear spin nutation the correction was small (quantitatively of the order of a percent [25]). The origin of what is now known as the Bloch-Siegert (frequency) shift was thereby shown to reside in the neglected Fourier component of the rf field, a contribution absent in the original Rabi setup [24].

Consistently with the magnetic resonance observations [25,26], light shifts are ubiquitous in atomic physics and quantum optics, although, due to the much higher values of (the optical) frequency, their relative amplitude is much smaller than what is found in spin problems. Hence, one should expect the ac Stark shift to play a role in laser physics. From a computational point of view, however, its evaluation is more challenging, due to the requirement of a self-consistent calculation. Furthermore, the RWA, which fixes *a priori* the frequency of the wave to which the rotating reference system is pinned, represents an unsurmountable obstacle to a correct calculation of an eventual frequency shift; thus it needs to be eliminated.

The purpose of this paper is to show that an ac Stark shift does indeed occur in the theoretical description of a laser, based on two atomic levels actively participating in the quantum description (two more levels will be implicitly assumed to allow for pumping and efficient relaxation, as in all standard models). In order to obtain the ac Stark shift we avoid the RWA and restart from a full density matrix formulation of the problem. As an aside, we also obtain that the critical cavity detuning associated with the occurrence of a spatial transverse modulational instability is no longer strictly zero (as predicted by current models [17,18]) but slightly differs from zero. Both effects turn out to be very small, which explains their going unnoticed up until now.

II. DERIVATION OF THE MODEL WITHOUT THE RWA

The Schrödinger equation for a two-level system in interaction with a classical e.m. field takes the form

$$i\hbar\,\partial_t\psi = (\mathcal{H}_0 - \mu_{12}E)\psi, \tag{1}$$

where $\hbar$ is Planck's constant (divided by 2π), $\mathcal{H}_0$ is the unperturbed Hamiltonian, $-\mu_{12}$ is the dipole moment (with the sign taking explicitly into account the negative electron charge), and E is the electric field. Here and in the following the symbol ∂_t denotes the derivative with respect to time (the second derivative will be ∂_{tt}). Looking for a solution of Eq. (1) in the form

$$\psi = c_1(t)\phi_1(\overrightarrow{r})e^{-i\frac{\mathcal{E}_1}{\hbar}t} + c_2(t)\phi_2(\overrightarrow{r})e^{-i\frac{\mathcal{E}_2}{\hbar}t} \tag{2}$$

where $\phi_{1,2}(\overrightarrow{r},t)$ are eigenstates of $\mathcal{H}_0$, we get

$$\partial_t\rho_{11} = -\frac{i}{\hbar}E(\rho_{12}\mu_{12}^* - \rho_{12}^*\mu_{12}), \tag{3a}$$

$$\partial_t\rho_{22} = -\partial_t\rho_{11}, \tag{3b}$$

$$\partial_t\rho_{12} = \frac{i}{\hbar}[\rho_{12}(\mathcal{E}_2 - \mathcal{E}_1) + \mu_{12}E(\rho_{22} - \rho_{11})], \tag{3c}$$

where $\rho_{11} = c_1c_1^*$ and $\rho_{22} = c_2c_2^*$ are real and satisfy $\rho_{11} + \rho_{22} = 1$, while $\rho_{12} = c_1c_2^*$ is complex. By introducing the two real atomic variables

$$p = \rho_{12}\mu_{12}^* + \rho_{12}^*\mu_{12}, \quad q = i(\rho_{12}\mu_{12}^* - \rho_{12}^*\mu_{12}), \tag{4}$$

Eq. (3c) splits into

$$\partial_t p = \omega_g q, \quad \partial_t q = -\omega_g p - \tfrac{2|\mu_{12}|^2}{\hbar}E(\rho_{22} - \rho_{11}), \tag{5}$$

where $\hbar\omega_g = \mathcal{E}_2 - \mathcal{E}_1$. Finally, introducing the phenomenological coefficients required for the semiclassical description of the nonconservative processes we obtain

$$\begin{aligned} \partial_{tt}P &= -\nu\partial_t P - \omega_g^2 P - \frac{2|\mu_{12}|^2\omega_g}{\hbar}EN,\\ \partial_t N &= \gamma(N_p - N) + \frac{2E\partial_t P}{\hbar\omega_g} + d\nabla^2 N,\\ \partial_{tt}E &= c^2\nabla^2 E - \frac{1}{\epsilon_0}\partial_{tt}P - \frac{\sigma}{\epsilon_0}\partial_t E, \end{aligned} \tag{6}$$

where $N = N_0(\rho_{22} - \rho_{11})$ is the population inversion density (N_0 is the number of atoms per unit volume) and $P = N_0(\rho_{12}\mu_{12}^* + \rho_{12}^*\mu_{12})$ is the atomic polarization density. N_p stands for the pumping rate, d for the diffusion coefficient and γ for the population decay rate. Finally, ν is the polarization decay rate, σ the electric conductivity, ϵ_0 the vacuum permittivity, and c the speed of light in vacuum.

Note that performing the usual RWA and SVEA on Eqs. (6) yields the well-known Maxwell-Bloch equations, omnipresent in the laser literature for the past half-century [1].

With the following scalings:

$$\begin{aligned} &\partial_t = \omega_g\partial_{\tilde{t}}, && \nabla = \frac{\omega_g}{c}\tilde{\nabla}, && g = \frac{2|\mu_{12}|^2N_0}{\hbar\epsilon_0\omega_g},\\ &P = \epsilon_0 s\tilde{P}, && E = s\tilde{E}, && \sigma = \epsilon_0\omega_g\tilde{\sigma},\\ &s^2 = \frac{N_0\hbar\gamma}{2\epsilon_0}, && \nu = \omega_g\tilde{\nu}, && N = N_0\tilde{N},\\ &N_p = N_0\tilde{N}_p, && \gamma = \omega_g\tilde{\gamma}, && D = \frac{d\omega_g^2}{c^2\gamma} \end{aligned} \tag{7}$$

and dropping the superscript, Eqs. (6) become

$$\begin{aligned} \partial_{tt}P &= -\nu\partial_t P - P - gEN,\\ \frac{1}{\gamma}\partial_t N &= [N_p - N + E\partial_t P + D\nabla^2 N],\\ \partial_{tt}E &= \nabla^2 E - \partial_{tt}P - \sigma\partial_t E. \end{aligned} \tag{8}$$

Realistic boundary conditions are not involved in the existence of a self-induced Stark shift effect, which is a pure bulk physical mechanism. Therefore, for the sake of simplicity, a simple infinite geometry will be assumed for the amplifier medium description [Eqs. (8)]. Then elementary calculations show that $N = N_p$ and $E = P = 0$ is a stationary solution of Eqs. (8), which is stable if and only if $N_p < N_{pc} = \frac{\sigma\nu}{g}$. N_{pc} corresponds to the laser threshold. Above this value, linear stability analysis predicts that the traveling waves $e^{i(t\pm\overrightarrow{k}.\overrightarrow{r})}$ with $k^2 = 1$ are the unstable perturbations with the highest growth rate.

III. WEAKLY NONLINEAR MULTISCALE ANALYSIS

The following well-established analytical approach is rigorously justified from a mathematical point of view [12,13] and has been successfully used in numerous nonlinear problems (for nonlinear optics cf. [14,18,19]). It is valid only close enough to the laser threshold, $N_p - N_{pc} = \epsilon^2\mu$, where ϵ is a small parameter and μ is of order one. In this parameter region, the amplitudes of E and P are expected to saturate to a small value such that E and P can then be sought as a $O(1)$ power expansion in ϵ. In contrast, their phases are known to rapidly oscillate in space and time. The main idea behind the multiscale analysis is to describe the slow and fast behavior with independent variables. Hence, the time variable t is replaced by t_0 for the fast components, t_1 for the temporal evolution with $\frac{1}{\epsilon}$, t_2 for $\frac{1}{\epsilon^2}$ characteristic time scales, and so on.

The following multiscale Ansatz is therefore introduced:

$$\begin{aligned} N_p &= N_{pc} + \epsilon^2\mu, \quad \partial_t = \partial_{t_0} + \epsilon\partial_{t_1} + \epsilon^2\partial_{t_2} + \epsilon^3\partial_{t_3} + \cdots,\\ \partial_z &= \partial_{z_0} + \epsilon\partial_{z_1}, \quad \nabla_\perp = \sqrt{\epsilon}\nabla_{\perp_{\frac{1}{2}}},\\ E &= \epsilon E_1 + \epsilon^2 E_2 + \epsilon^3 E_3 + \cdots,\\ P &= \epsilon P_1 + \epsilon^2 P_2 + \epsilon^3 P_3 + \cdots,\\ N &= N_{pc} + \epsilon^2 N_2 + \epsilon^3 N_3 + \cdots, \end{aligned} \tag{9}$$

where ∂_s ($s = t_0, t_1, \ldots, z_0, z_1, \ldots$) stands for partial derivative with respect to s, and where $\nabla_\perp^2$ is the transverse Laplacian $(\partial_{xx} + \partial_{yy})$. For the space variables (e.g., z), we have not used the full expansion $\partial_z = \partial_{z_0} + \epsilon^1\partial_{z_1} + \epsilon^2\partial_{z_2} + \ldots$ but a truncated form [Eq. (9)] which yields the same result as the complete one.

Replacing Eq. (9) into Eq. (8) leads to a hierarchy of linear equations which can be successively solved provided that the solvability conditions are satisfied. After some manipulations, we find that the Taylor expansions

$$\begin{aligned} E_1 &= Fe^{i(t_0-z_0)} + Be^{i(t_0+z_0)} + \text{c.c.}.\\ E_2 &= 0, \qquad P_1 = \sigma(Fe^{i(t_0-z_0)} + Be^{i(t_0+z_0)}) + \text{c.c.},\\ P_2 &= \frac{\sigma(\nu+2i)}{\sigma+\nu}(\partial_{z_1}Fe^{i(t_0-z_0)} - \partial_{z_1}Be^{i(t_0+z_0)}) + \text{c.c.},\\ N_1 &= N_{pc},\\ N_2 &= \mu - \sigma\left[\frac{F^2}{R(2,-2)}e^{i(2t_0-2z_0)} + \frac{F^{*2}}{R(-2,2)}e^{i(-2t_0+2z_0)} + \frac{B^2}{R(2,2)}e^{i(2t_0+2z_0)} + \frac{B^{*2}}{R(-2,-2)}e^{i(-2t_0-2z_0)} + 2(|F|^2+|B|^2)\right.\\ &\quad \left. +2\left(\frac{FB}{R(2,0)}e^{i(2t_0)} + \frac{F^*B^*}{R(-2,0)}e^{i(-2t_0)}\right) + 2\left(\frac{FB^*}{R(0,-2)}e^{i(-2z_0)} + \frac{F^*B}{R(0,2)}e^{i(+2z_0)}\right)\right] \end{aligned} \tag{10}$$

are the solution of Eq. (8) up to $O(\epsilon^3)$ provided that

$$\begin{aligned} \partial_t F &= a_0F - a_1\partial_z F + a_2\partial_{zz}F - (a_3|F|^2 + a_4|B|^2)F\\ &\quad +[a_5 + a_6\partial_z + a_7\nabla_\perp^2]\nabla_\perp^2 F,\\ \partial_t B &= a_0B + a_1\partial_z B + a_2\partial_{zz}B - (a_3|B|^2 + a_4|F|^2)B\\ &\quad +[a_5 - a_6\partial_z + a_7\nabla_\perp^2]\nabla_\perp^2 B, \end{aligned} \tag{11}$$

with

$$\begin{aligned} a_0 &= \frac{1}{2}\frac{g\mu}{\sigma+\nu}, \quad a_1 = \frac{\nu}{\sigma+\nu},\\ a_2 &= \frac{\sigma\nu}{2(\sigma+\nu)^3}[4 - i(\sigma+\nu)]\\ a_3 &= \frac{\sigma g}{2(\sigma+\nu)}\frac{3\gamma + 4i + 8D\gamma}{\gamma + 2i + 4D\gamma},\\ a_4 &= \frac{\sigma g}{(\sigma+\nu)}\frac{3\gamma + 4i + 8D(\gamma+i)}{(1+4D)(2i+\gamma)}, \quad a_5 = -\frac{i}{2}a_1,\\ a_6 &= -4i\,a_7, \quad a_7 = -\frac{\nu}{8(\sigma+\nu)^3}[4\sigma + i\nu(\sigma+\nu)], \end{aligned} \tag{12}$$

where F and B are the envelopes of the forward and backward electric field traveling wave solutions. They are the order parameters of the laser bifurcation and enter into the expression of all the physical variables, Eqs. (10). F and B depend on the slow variables t_1, t_2, t_3,...,z_1,$x_{\frac{1}{2}}$,$y_{\frac{1}{2}}$ even though this dependence is not explicit in Eqs. (11) due to the scale aggregation procedure. F^* (B^*) stands for the complex conjugate of F (B). $R(p,q) = \frac{ip}{\gamma} + 1 + Dq^2$ is an auxiliary function whose purpose is to strongly simplify the expression of N_2.

The following remarks are in order:

(1) The previous asymptotic analysis is always valid close enough to the laser threshold N_{pc} and is especially well suited to describe Class A laser dynamics [7]. For Class B and C lasers, a range of validity still exists close enough to the threshold, i.e., for $\mu \ll \gamma$ and $\mu \ll \nu$.

(2) Considering the population inversion as a critical mode (i.e., assuming $\gamma \simeq \epsilon$) extends the range of validity of the

previous description. The computations are more complex, but the conceptual results we are discussing here are not qualitatively modified.

(3) $a_{2r} = \mathrm{Re}(a_2) > 0$ so that diffusion prevents small longitudinal wavelength instabilities from occurring. Higher order spatial derivatives with respect to z are not required.

(4) $a_{7r} = \mathrm{Re}(a_7) < 0$, and thus spatial transverse instabilities at small wavelengths are damped.

(5) The coefficients a_3 and a_4 are given by

$$a_3 = \frac{\sigma g}{2(\nu+\sigma)} \frac{[8+\gamma^2(3+20D+32D^2)] - 2i\gamma}{4+\gamma^2(1+4D)^2} \tag{13}$$

and

$$a_4 = \frac{\sigma g}{(\nu+\sigma)} \frac{[8+3\gamma^2+16D+8D\gamma^2] - 2i\gamma(1+4D)}{(1+4D)(4+\gamma^2)}. \tag{14}$$

The real parts of a_3 and a_4 are always positive. Thus, the nonlinear cubic terms stop the linear exponential growth and there is no need for an expansion to order (ϵ^4).

(6) Up to $O(\epsilon^3)$, the Taylor expansion for N can be structured in the following way:

$$N = \underbrace{N_p - 2\sigma(|F|^2+|B|^2)}_{I}$$
$$\underbrace{-\sigma\left[\begin{array}{l} \frac{F^2}{R(2,-2)}e^{i(2t_0-2z_0)} + \frac{F^{*2}}{R(-2,2)}e^{i(-2t_0+2z_0)} \\ +\frac{B^2}{R(2,2)}e^{i(2t_0+2z_0)} + \frac{B^{*2}}{R(-2,-2)}e^{i(-2t_0-2z_0)} \\ +2\left(\frac{FB}{R(2,0)}e^{i(2t_0)} + \frac{F^*B^*}{R(-2,0)}e^{i(-2t_0)}\right) \\ +2\left(\frac{FB^*}{R(0,-2)}e^{i(-2z_0)} + \frac{F^*B}{R(0,2)}e^{i(+2z_0)}\right) \end{array}\right]}_{II}$$
$$+O[(N_p - N_{pc})^3]. \tag{15}$$

Part (I) only depends on the slow space and time coordinates. In contrast, part (II) does depend on the fast variables t_0 and z_0. In particular, the last terms in part (II) correspond to stationary spatial modulations at twice the wave vector and are responsible for spatial hole burning. As expected, the amplitude of this spatial grating decreases with D [through the linear dependence of $R(0,\pm 2)$ on D].

(7) The ratio between cross- and self-saturation (a_{4r}/a_{3r}), which controls the stability of the traveling waves relative to the standing waves, depends on the γ and D parameters. Easy computations show that, whatever the value of these parameters, $a_{4r} > a_{3r}$ and the traveling waves are definitely the most stable solution.

(8) For insulating active media, the electric conductivity σ is vanishingly small. Equations (8) are no longer self-consistent because of the lack of an energy dissipation mechanism. Hence, realistic boundary conditions with nonvanishing transmission coefficient become essential. Fortunately, they can be taken into account in the previous analysis by simply replacing $\frac{\epsilon_0}{\sigma}$ with the photon cavity lifetime.

A. Self-induced ac Stark shift

We now investigate some solutions of Eqs. (11). Homogeneous traveling waves (HTW) correspond to

$$\begin{array}{l} F = R_{tw}e^{i(\Omega t)}, \\ B = 0, \end{array} \quad \text{or} \quad \begin{array}{l} F = 0 \\ B = R_{tw}e^{i(\Omega t)} \end{array} \tag{16}$$

with

$$R_{tw}^2 = \tfrac{a_0}{a_1}, \quad \Omega = -a_{3i}R_{tw}^2, \tag{17}$$

where a_{3r} stands for the real part of a_3 and a_{3i} for its imaginary part. The linear stability analysis, not explicitly performed here, shows that the HTW solutions are always stable. Equations (17) clearly show that the frequency Ω of the HTW envelope does depend on the electric field intensity R_{tw}^2. Taking into account the relationship between the electric field E and the envelopes (F and B), Eqs. (10), as well as the scalings, Eqs. (7), we obtain for the angular frequency ω_e of the electric field in the HTW regime (in physical units):

$$\omega_e = \omega_g\left(1 + \frac{-a_{3i}}{s^2}|E|^2\right) = \omega_g + \Delta|E|^2, \tag{18a}$$

$$\Delta = \left[\frac{\sigma}{\sigma+\epsilon_0\nu}\right]\left[\frac{2}{4+\left(\frac{\gamma}{\omega_g}\right)^2\left(1+4\frac{d\omega_g^2}{c^2\gamma}\right)^2}\right]\Delta_s, \tag{18b}$$

$$\Delta_s = \frac{2|\mu_{12}|^2}{\hbar^2\omega_g}, \tag{18c}$$

where Δ_s is exactly the usual ac Stark shift. The fact that Δ is proportional to Δ_s but not equal to it follows from the self-induced nature of the physical mechanism. Indeed, the feedback of the atomic displacement onto the electric field, absent when the latter is an external parameter, introduces the prefactor of Δ_s found in the expression for Δ. Note that the frequency shift has only one sign (blue shift).

B. Occurrence of transverse patterns and cavity detuning threshold

Up until here, the longitudinal boundary conditions (cavity mirrors) have not been taken into account in the model. We now consider a forward traveling wave ($F \neq 0, B = 0$) in a ring cavity with length (in scaled units)

$$L = 2\pi(n+\delta) \quad \text{with} \quad n \in \mathcal{N} \quad \text{and} \quad -\tfrac{1}{2} < \delta < +\tfrac{1}{2}. \tag{19}$$

The z-periodic boundary conditions are accounted for by setting

$$F(t,x,y,z) = A(t,x,y)e^{-iQz} \quad \text{with} \quad Q = -\frac{\delta}{n+\delta}, \tag{20}$$

where A obeys

$$\partial_t A = \alpha A - a_3|A|^2A + \beta\nabla_\perp^2 A + a_7\nabla_\perp^4 A, \tag{21}$$

with

$$\begin{gathered} \alpha = a_0 + iQa_1 - Q^2a_2, \\ \beta = a_5 - iQa_6 = -\frac{i}{2}a_1 - 4Qa_7. \end{gathered} \tag{22}$$

Equation (21) admits a homogenous solution of the form

$$A = R_h e^{i\omega_h t} \qquad \text{with} \qquad R_h^2 = \frac{\alpha_r}{a_{3r}}, \quad \omega_h = \alpha_i - \beta_i R_h^2 . \tag{23}$$

Linear stability analysis provides the most unstable eigenvalue, associated with phase invariance symmetry:

$$\lambda = -\frac{\beta_r a_{3r} + \beta_i a_{3i}}{a_{3r}} k^2 + O(k^4), \tag{24}$$

where k is the wave vector of the transverse perturbation. The coefficient of k^2 in the previous expansion vanishes for

$$Q_c = -\frac{a_1 a_{3i}}{8(a_{7r} a_{3r} + a_{7i} a_{3i})} . \tag{25}$$

Note that Q_c vanishes with a_{3i}. Thus, in the usual mathematical description of the instability where frequency renormalization is absent ($a_{3i} = 0$), a spatially transverse structure develops as soon as the detuning δ changes its sign. Instead, in the presence of frequency renormalization, Q_c, as well as the critical cavity detuning, no longer vanishes. From a physical point of view, the picture is quite obvious. In the absence of frequency renormalization, the mismatch between cavity length and resonance condition, introduced by detuning, can only be counterbalanced by the growth of a transverse component in the wave vector when $\delta < 0$. When the frequency renormalization is taken into account, the self-adjustment of the frequency as a function of the field amplitude can compensate, in certain parameter ranges, for the mistuning from the cavity resonance. Thus, the appearance of transverse structures no longer coincides with the crossing of the perfect tuning condition ($\delta = 0$).

IV. PHYSICAL DISCUSSION

In this section we concentrate on a close examination of the laser frequency displacement, self-induced by the ac Stark shift (Sec. III A), and focus on quantitative estimates of their magnitude $\Delta\omega = \omega_e - \omega_g$ in various kinds of lasers. Thus, we specialize the discussion by introducing explicitly the cavity losses, K, and the dipole relaxation rate [or full width at half maximum (FWHM) linewidth of the transition], $\gamma_\perp$, which replaces the quantity ν used in the mathematical discussion.

The nonlinear multiscale analysis (Sec. III), an asymptotic form of expansion, rigorously holds only near threshold. However, we will extrapolate the predictions of Eq. (18a) in order to obtain order-of-magnitude estimates and to gauge the extent of the influence of the ac Stark shift on laser operation. Along the same line, we stretch somewhat the validity of the predictions (Sec. III A) obtained for the traveling wave solution (unidirectional emission in a ring laser) to obtain quantitative estimates in standing wave (Fabry-Perot) cavities. We justify this extension by analogy with a detailed investigation conducted in [27] showing a close correspondence between the dynamics of lasers (and other optical nonlinear systems) in different kinds of configurations (e.g., ring and Fabry-Perot cavities).

Close examination of Eqs. (18a)–(18c) shows that the self-induced term is composed by the usual, field-induced ac Stark shift—well known in atomic physics and quantum optics (Δ_s)—multiplied by two terms. The first can be recast using the substitution mentioned at the end of Sec. III: $K = \frac{\sigma}{\epsilon_0}$, which allows us to rewrite the first bracketed term in Eq. (18b) as $\frac{1}{1+\frac{\gamma_\perp}{K}}$. Its interpretation is immediate in the context of a laser: this factor introduces a *weight* coefficient defined as the fraction of the coherence time of the intracavity field (K^{-1}) over which the atomic dipole is excited ($\gamma_\perp^{-1}$). Contrary to what happens in the case of an atom excited by an externally generated e.m. field (where its coherence time is irrelevant as long as it is longer than the dipole's), the self-induced nature of the laser e.m. field requires the presence of a measure of its influence on the emitting (i.e., field-generating) dipole. Indeed, if on the one hand the e.m. coherence time is extended in a laser by the cavity action, nonetheless, its influence on each emitting dipole cannot exceed the latter's coherence time.

Another way of illustrating the influence of this prefactor rests on the relative widths of the cavity and emission resonance lines. In the usual derivation of the laser equations (not including the ac Stark effect) the oscillation frequency, in single mode operation, is the result of the so-called mode pulling [28]: the resonance linewidths of e.m. field and active medium, respectively, act as filters which pull the actual laser frequency toward their respective centers according to their relative inverse widths (quality factor). Frequency pulling is the result of the competition between the active medium and the cavity in the feedback mechanism leading to lasing. Thus, it is not surprising to find in the self-induced ac Stark shift a similar weighting factor, which can be recast in the first bracketed term of Eq. (18b) as $\frac{K}{\gamma_\perp}$ (if $K \ll \gamma_\perp$). Since lasers are characterized by a cavity whose resonance width is typically much smaller than that of the active medium, one expects the self-induced ac Stark shift to be weaker than the externally induced one [the first bracketed term in Eq. (18b)].

A second weighting factor appears in Eq. (18b): the second square bracketed term includes various laser parameters, such as the spontaneous linewidth (γ), the unperturbed emission frequency (ω_g), the diffusion coefficient d, and the speed of light (in vacuum). One should expect this term to affect the resulting Δ coefficient depending on the characteristics of the laser. Direct substitution of the physical parameters (cf. Table I) shows that the bracketed term is equal to 0.5 (with three significant figures) for all types of lasers considered in the table. In the calculations, we have used values for the diffusion coefficient ranging from $d \approx 10^{-5}$ m^2 s^{-1} for gases to $d \approx 10^{-2}$ m^2 s^{-1} for semiconductors. Thus, we can safely conclude that this term contributes only a factor $\frac{1}{2}$ to the self-induced ac Stark shift.

One final physical illustration of the interplay between the Stark shift and the lasing process convincingly shows that a limiting action must take place for lasing to occur. When driving a sample (e.g., atomic vapor) with an external field, one can induce a frequency shift that can be as large as desired (the limitation coming from external factors such as available power, etc.). Thus, it is in principle possible to induce a light shift which exceeds the spontaneous emission linewidth. In the case of the laser the latter situation becomes problematic. In coherently pumped lasers (e.g., molecular ones) the issue of the pump-induced Stark shift is an important one since the latter may detune the dressed transition outside the absorption linewidth (if the pump frequency is not adjusted accordingly), thus reducing the effective pumping (see the brief discussion at the end of Sec. IV A). Thus, in order to maintain a high pumping efficiency in a molecular laser (the most sensitive to this perturbation due to the narrow linewidth of typical molecular transitions) one is forced to either limit the pump strength or to tune its frequency with increasing power. When the ac Stark shift is self-induced this is not possible, as the frequency and amplitude are coupled and self-determined by the feedback mechanism. Thus, one immediately sees that if the ac-Stark-shift-induced frequency deviations are not negligibly small compared to the linewidth, then an automatic limiting mechanism would kick in as the self-induced detuning would reduce the effectiveness of the lasing process. One should expect, in principle, to even find an asymmetry between red and blue detuning (laser frequency smaller or larger than the emission line center, respectively) in this limiting mechanism, since a red-detuned emission would be shifted toward resonance by the ac Stark shift (thus increasing the output power and the resulting frequency shift), while a blue-detuned one would be shifted away from it (thus reducing the output power and the resulting shift). In practice, as we will see in the following section, the frequency shifts are so small that the linewidth envelope can be considered to be constant over the frequency interval, thereby removing the potential asymmetry between red and blue detuning.

In conclusion, overall inspection of Eq. (18b) suggests that at least in most lasers the self-induced ac Stark shift should induce a frequency shift substantially smaller than what one expects for the equivalent effect in the corresponding atomic species. In the following section we examine a number of specific lasers and draw some pertinent conclusions.

TABLE I. Laser classes according to classification in [7]; λ, chosen laser wavelength; B, spontaneous emission probability (Einstein coefficient); μ, dipole moment for the lasing transition; $\gamma_\perp$, transition's homogeneous linewidth (FWHM); K, cavity loss rate [29]; Δ_s, ac Stark shift induced by an external e.m. field; Δ, self-induced ac Stark shift in a laser.

Class	Type	λ (μm)	B[a] (s^{-1})	μ_{12}[b] (C m)	$\gamma_\perp$ (rad s^{-1})	K (s^{-1})	Δ_s[c] (m^2 s^{-1} V^{-2})	Δ[d] (m^2 s^{-1} V^{-2})
A	He-Ne[e]	0.6328	1.40×10^6	2.05×10^{-30}	2×10^8	3×10^6	2.54×10^{-7}	1.88×10^{-9}
A	Dye (Rhodamine 6G)[f]	0.62	3×10^8	2.91×10^{-29}	1×10^{13}	3×10^7	5.01×10^{-5}	7.51×10^{-11}
A	Ar^+ [g]	0.514	2.90×10^9	6.82×10^{-29}	4.4×10^9	1.5×10^7	2.29×10^{-4}	3.89×10^{-7}
B	Nd:YAG[h]	1.06	4.35×10^3	2.49×10^{-31}	7.5×10^{11}	2×10^7	6.31×10^{-9}	8.41×10^{-14}
B	Nd:YAG microcavity[i]	1.06	4.35×10^3	2.49×10^{-31}	7.5×10^{11}	1×10^{10}	6.31×10^{-9}	4.15×10^{-11}
B	CO_2 [j]	10.6	8×10^2	3.36×10^{-30}	1.5×10^8	3×10^7	1.14×10^{-5}	9.5×10^{-7}
B	Semiconductor (GaAs)[k]	0.895	2.71×10^8	4.81×10^{-29}	1×10^{13}	8×10^{10}	1.98×10^{-4}	7.86×10^{-7}
B	Ti:sapphire[l]	0.78	4.14×10^5	1.52×10^{-30}	6.2×10^{14}	1×10^7	1.73×10^{-7}	1.40×10^{-15}
C	He-Ne[m]	1.15	4.4×10^6	8.93×10^{-30}	1×10^9	5×10^8	8.81×10^{-6}	1.47×10^{-6}
C	He-Ne[n]	3.39	9.60×10^5	2.11×10^{-29}	1.2×10^9	1×10^9	1.44×10^{-4}	3.27×10^{-5}
C	He-Xe[o]	3.51	4.6×10^6	4.85×10^{-29}	4.4×10^8	1×10^9	7.89×10^{-4}	2.74×10^{-4}
C	NH_3[p]	374	1.22×10^{-3}	8.69×10^{-31}	2×10^5	2×10^6	2.70×10^{-5}	1.23×10^{-5}

[a]Primary quantity obtained directly from cited sources or from the fluorescence or natural lifetime (cf. GaAs and NH_3 for only exceptions).
[b]Derived from Eq. (26) (except GaAs and NH_3, where it is obtained as a primary quantity from the cited source).
[c]Estimated from Eq. (18c).
[d]Estimated from Eq. (18b).
[e]Spontaneous emission probability B and $\gamma_\perp$ from [30]; K estimated for a 50-cm-long Fabry-Perot cavity with 1% outcoupling.
[f]λ : peak wavelength; B and $\gamma_\perp$ from [30]; K estimated for a 1-m-long ring cavity with 10% outcoupling.
[g]B from [31]; $\gamma_\perp$ estimated on the basis of [32]; K estimated for a 1-m-long Fabry-Perot cavity with 10% outcoupling.
[h]B from [30]; $\gamma_\perp$ from [28,33]; K estimated for a 75-cm-long Fabry-Perot cavity with 10% outcoupling.
[i]Same parameters as for bulk laser but with a 1.5-mm-long Fabry-Perot cavity.
[j]For a longitudinal discharge low-pressure ($\approx$20 Torr) CO_2 laser: B from [34]; $\gamma_\perp$ from [35]; K estimated assuming a 1-m-long Fabry-Perot cavity with mirror reflectivity of 80%.
[k]B derived by inverting Eq. (26); μ_{12} and $\gamma_\perp$ from [36]; K from [37]. Notice that the cavity losses are of the same order of magnitude for edge emitters (bulk lasers) or VCSELs: the cavity length is approximately three orders of magnitude smaller for the VCSEL, but the mirror reflectivity is three orders of magnitude larger. Thus the self-induced ac Stark shift is equivalent in the two kinds of lasers.
[l]λ: peak wavelength; B derived from [38]; $\gamma_\perp$ derived from [33]; K estimated assuming a 1-m-long ring cavity with $\approx$3% outcoupling.
[m]B from [30]; $\gamma_\perp$ assumed close to the value for the $\lambda = 3.39$ μm transition [39], in qualitative agreement with [28]; K assumed half the value as for $\lambda = 3.39$ μm, given the ratio between the oscillator strengths of the two transitions [30].
[n]B from [30]; $\gamma_\perp$ from [39]; K experimentally estimated in [39].
[o]B,$\gamma_\perp$, and K from [39].
[p]B derived by inverting Eq. (26); μ_{12} given for a different transition in [40], but since the lifetimes do not vary very strongly in the various branches [41], we safely use this value. Note that we used the conversion 1 debye = 3.34×10^{-30} C m; $\gamma_\perp$ from [42] at pressure $P = 2$ Pa; K derived from an intermediate value of cavity linewidth [42]. The outcoupling is of the order of 0.4% [42].

A. Analysis for various kinds of lasers

We have chosen a sample of different kinds of laser, representative of the various dynamical classes [7], selected to consider devices which are of very widespread use, as well as lasers which have niche applications or historical importance (Table I). At the same time, we have tried to cover a range of lasers going from extremely insensitive to relatively sensitive ones as far as the self-induced ac Stark shift is concerned.

The dynamical class [7] to which each laser belongs is listed in the first column of Table I, while the emission wavelength appears in column 3. For lasers emitting on several lines we have chosen the most representative one (or several interesting ones, as for He-Ne). For others we have opted for the emission peak (tunable lasers), for a typical wavelength (semiconductor or CO_2), or even a particularly interesting wavelength (as in the case of the NH_3 laser, where we have chosen a far infrared (FIR) line with sufficiently high gain to operate in the so-called bad-cavity limit [29]).

Equation (18b) requires knowledge of the spontaneous emission rate (γ), which we obtain either directly from the sources cited in Table I or using the spontaneous emission probability (Einstein coefficient B).

The dipole moment, μ_{12}, for the transition can be obtained from the standard relation between the Einstein B coefficient (spontaneous emission) and its corresponding microscopic expression (cf., e.g., [22]):

$$\mu_{12} = \sqrt{\frac{\pi \epsilon_0 \hbar c^3 B}{\omega_g^3}}. \tag{26}$$

The laser linewidth (FWHM) values, $\gamma_\perp$, have been obtained from the cited sources while the cavity losses K have for the most part been estimated on the basis of the cavity characteristics specified in Table I.

Four kinds of lasers stand out for their particularly small self-induced ac Stark shift (Δ): the He-Ne laser emitting on the red line, the dye laser (for Rhodamine 6G, but in general all dyes have similar values the emission linewidth, $\gamma_\perp$), the Nd:YAG laser, and the Ti:sapphire laser. The last three are characterized by a particularly large value of $\gamma_\perp$, originating from the fact that the relaxation processes in a solid (and similarly in a liquid) are very broadband due to the strong contribution of lattice vibrations (phonons) to the linewidth. The very short coherence time of the oscillating dipole, compared to that of the intracavity field, very strongly suppresses the self-induced ac Stark shift, as is easily recognized by comparing the corresponding values of Δ_s and Δ: the latter is reduced by a factor ranging between five and eight orders of magnitude with respect to the former. The most spectacular reduction belongs to the Ti:sapphire laser, whose trademark is exactly one of the largest possible linewidhts [33], which make of it the workhorse of ultrashort pulses. In this setting, the microcavity Nd:YAG laser holds a special position and will be discussed later.

Notice that we have not explicitly considered the case of a Nd:glass laser, whose emission line, even broader than that of its counterpart based on a crystalline matrix, is inhomogeneously broadened. Indeed, for the present discussion the contribution to self-induced frequency shift comes from the homogeneous component of the linewidth, since the inhomogeneous one is basically responsible for multimode operation. This same consideration holds for the other kinds of lasers we have considered (He-Ne at $\lambda = 6328$ Å, or the laser line of the Ar^+ laser) whose overall linewidth is inhomogeneously broadened: in these cases we have indicated the homogeneous linewidths (FWHM).

At the opposite end of the range of values for the self-induced ac Stark shift (Δ) we find the near-IR to mid-IR lasers (He-Ne on the IR transitions and He-Xe) and the very FIR ammonia laser line. For the first three we remark that the reduction going from Δ_s to Δ is less than one order of magnitude, while for the latter it attains the theoretical minimum permitted by Eq. (18b): the factor $1/2$. This comes from the fact that this laser operates under the so-called bad-cavity regime [29], where the cavity losses are so high that the e.m. field coherence time is shorter than that of the active medium. The NH_3 laser wavelength confirms that we are nearly dealing with a maser, characterized by an active medium linewidth narrower than that of the cavity.

The other three lasers of the C class, however, maintain a self-induced ac Stark shift comparable to the atomic one, in agreement with their belonging to this dynamical class, where the time constants for intracavity e.m. field, atomic polarization, and population inversion are all of the same order of magnitude. Thus, one would expect these lasers to be most susceptible to the influence of the self-induced frequency shift.

The remaining three kinds of lasers (Ar^+, CO_2, and semiconductor) fall into an intermediate range of frequency shifts and will be examined in further detail in the following.

The actual frequency shift which may be attained in a laser depends not only on the coefficient Δ but also on the value of the electric laser field E present in the cavity. Thus, a realistic analysis must take into account typical (maximum) values of E field attainable to estimate the resulting frequency shift. For ease of comparison between the different laser types, we have chosen to quantify the electric field value necessary to obtain a frequency shift $\Delta\nu_{\rm acS} = \frac{\Delta\omega}{2\pi} = \frac{\omega_e - \omega_g}{2\pi} = 1$ MHz [from Eq. (18b)]. This arbitrary value is chosen of the order of the linewidth of a strong dipole transition, a quantity easily measurable and directly related to the induced ac Stark shift.

Table II repeats, in the first four columns, some basic information for the lasers we have considered in Table I. The fifth gives the value of the intracavity laser electric field E giving rise to a $\Delta\nu_{\rm acS} = 1$ MHz frequency shift, followed by the corresponding power density $\mathcal{P}$ (or intensity) (column 6), and by the power P necessary to obtain the desired frequency shift (column 7) assuming an intracavity beam radius $r = 200\ \mu$m (a value that does not make sense for semiconductor lasers but is kept for coherence in the table entries—see the following discussion in the text for semiconductor lasers). The last column gives the typical (maximum) value of the output power P_t which can be obtained from each of these lasers. These values are somewhat arbitrary and can be discussed and amended, but they are useful to identify those lasers for which measurable frequency shifts may occur.

Comparison of the values for P and P_t shows that for a number of these lasers the frequency shift is negligibly small and that in most cases it would be even not be measurable. This is the case of the red-emitting He-Ne laser, whose intracavity power $P_i \approx 10^2 P_t$ may at most reach a frequency shift of a couple of hertz for its highest power. In the same category fall the dye laser ($P_i \approx 10 P_t$, and thus $\Delta\nu_{acS} \leqslant 1$ Hz), the Nd:YAG laser ($P_i \approx 10 P_t$, and thus $\Delta\nu_{acS} < 1$ Hz for bulk), and the Ti:sapphire laser ($P_i \approx 30 P_t$, and thus $\Delta\nu_{acS} \leqslant 0.1$ mHz).

The case of the microchip (or microcavity) Nd:YAG needs a more detailed discussion. The typical spot size in microchip lasers gives a value for $r \approx 50\ \mu$m, which decreases by more than an order of magnitude its value for P given in Table II, while the typical reflectivities are rather of a few percent, giving another factor of 2 when converting from P_t to P. With this in mind, we obtain a self-induced frequency shift $\Delta\nu_{acS} \approx 2$ Hz, a value that is slightly larger than the one for the bulk laser. It must also be kept in mind that several different kinds of crystals (at different wavelengths) have been studied for microchip lasers, thus offering a wider palette of power density values and possible self-induced ac Stark shift; we also remark that these devices are still being developed and that power values larger than the one we have adopted may be found (cf., e.g., [43–45]).

With a intracavity enhancement factor of approximately 5, CO_2 lasers may attain a frequency shift of the order of 1 MHz at the highest power levels reported in Table II, for low-pressure laser configurations. As such, the shift should be easily measurable, even though it is understandable that it may have gone undetected if not looked for. One of the applications of these lasers has consisted in coherent pumping of molecular lasers, a use for which a frequency shift may be important, given the narrow linewidth of most molecular lines. However, the power levels involved (of the order of $P_{pump} \leqslant 10^1$ W) predict shifts $\Delta\nu_{acS} \approx 10$ kHz, thus reducing their potential impact on pump efficiency. In the case of frequency-stabilized CO_2 lasers (used to pump molecular lasers [42], but also for other precision applications) traces of frequency shifts may be present in the error signal. More details on the impact of the self-induced ac Stark shift on frequency-stabilized systems will be given (in the case of an IR He-Ne laser) in Sec. IV B.

For semiconductor lasers we need to separately consider two configurations and estimate anew the values of P: edge emitters and VCSELs. By assuming the same material properties, the typical cross-sectional surface of an edge emitter is of the order of $S \simeq 40\ \mu\text{m}^2$ (supposing a $2 \times 20\ \mu$m rectangular waveguide), thus providing a reduction in the power P, for a frequency shift $\Delta\nu_{acS} = 1$ MHz, relative to the value of P estimated in Table II, by a factor > 3000. Coupled to the minor cavity enhancement factor (≈ 1.5 considering $n_{\lambda=895\text{ nm}} \approx 3.6$, and thus a reflectivity value for a single crystal facet $R_m = 0.32$, with the second facet supposed high-reflection coated) the laser output power sufficient to induce $\Delta\nu_{acS} = 1$ MHz is of the order of $P_{out} \approx \mathcal{P} \times S \times (1 - R_m) \approx 2.9 \times 10^{-1}$ W. For a powerful edge emitter, $P_t \approx 1$ W, and we thus estimate $\Delta\nu_{acS} \approx 3$ MHz.

For VCSELs a typical cross-sectional surface is rather $S \simeq 7\ \mu\text{m}^2$ (assuming a 3-μm-diameter circular VCSEL). Since the cavity reflectivity is very high in these lasers ($R_m \approx 0.999$) and the factor Δ is the same as for edge emitters (cf. see Table I), we estimate $P_{out} \approx \mathcal{P} \times S \times (1 - R_m) \approx 7 \times 10^{-5}$ W for $\Delta\nu_{acS} = 1$ MHz. For a powerful VCSEL, $P_t \approx 0.1$ W, we expect a self-induced ac Stark frequency shift component $\Delta\nu_{acS} \approx 1.5$ GHz. A question arises as to whether this estimate is consistent with the cavity linewidth, since a dynamical (i.e., self-induced, power-dependent) shift which detunes the laser out of resonance would automatically limit the power output. From a reasonable estimate of the VCSEL's free spectral range (FSR $\approx 3 \times 10^{13}\ \text{s}^{-1}$ for a cavity length $l = 10\ \mu$m – roundtrip, optical path length) and finesse

TABLE II. E, electric field amplitude; $\mathcal{P}$, power density; and P, power to obtain an ac Stark frequency shift [cf. Eq. (18a)] with $\Delta\omega = \omega_e - \omega_g = 2\pi \times 1$ MHz. All quantities are given as intracavity values: the cavity enhancement factor must be taken into account to compare to the output power.

Class	Type	λ (μm)	Δ ($\text{m}^2\,\text{s}^{-1}\,\text{V}^{-2}$)	E ($\text{V}\,\text{m}^{-1}$)	$\mathcal{P}$[a] ($\text{W}\,\text{m}^{-2}$)	P[b] (W)	P_t[c] (W)
A	He-Ne	0.6328	1.88×10^{-9}	5.8×10^7	4.4×10^{12}	5.6×10^5	< 0.05
A	Dye (Rhodamine 6G)	0.62	7.51×10^{-11}	2.9×10^8	1.1×10^{14}	1.4×10^7	≈1
A	Ar^+	0.514	3.89×10^{-7}	4.0×10^6	2.1×10^{10}	2.7×10^3	≈10
B	Nd:YAG	1.06	8.41×10^{-14}	8.6×10^9	9.9×10^{16}	1.3×10^{10}	10^2–10^3
B	Nd:YAG microcavity	1.06	4.15×10^{-11}	3.9×10^8	2.0×10^{14}	2.5×10^7	≈0.25
B	CO_2	10.6	9.5×10^{-7}	2.6×10^6	8.8×10^9	1.1×10^3	10^2–10^3
B	Semiconductor (GaAs)[d]	0.895	7.86×10^{-7}	2.8×10^6	1.1×10^{10}	1.3×10^3	0.1–1
B	Ti:sapphire	0.78	1.40×10^{-15}	6.7×10^{10}	6.0×10^{18}	7.5×10^{11}	≈1
C	He-Ne	1.15	1.47×10^{-6}	2.1×10^6	5.7×10^9	7.1×10^2	< 0.1
C	He-Ne	3.39	3.27×10^{-5}	4.4×10^5	2.6×10^8	3.2×10^1	< 0.1
C	He-Xe	3.51	2.74×10^{-4}	1.5×10^5	3.0×10^7	3.8×10^0	< 0.1
C	NH_3	374	1.23×10^{-5}	7.2×10^5	6.8×10^8	8.5×10^1	≈0.1[e]

[a]$\mathcal{P}$: power density. The calculation is done supposing a top-hat intensity distribution [30] and $\mathcal{P} = \frac{1}{2}\epsilon_0 c |E|^2$.
[b]P: power on a laser beam (supposed cylindrical) of radius $r = 200\ \mu$m.
[c]Typical value of the maximum output power obtained from each kind of laser.
[d]For semiconductor lasers the mode volume is much smaller and the value of P given is not very significant.
[e]Indirectly deduced from [42], assuming an intracavity beam size $r \approx 1$ cm, thus output power in the hundreds of mW (and intracavity power approximately 200 times larger, due to the weak outcoupling [42]).

$\mathcal{F} \approx 1000$, we obtain a cavity linewidth $\Delta\nu_c \approx 30$ GHz. The predicted shift is more than one order of magnitude smaller and therefore does not pose difficulties in the interpretation of the data.

One last potentially interesting case is that of the so-called nanolaser. Currently, a large amount of work is being devoted to the realization and study of cavities with extremely high finesse (e.g., $\mathcal{F} > 15\,000$ [46]) and extremely small dimensions. Assuming a cavity with circular radius $r < 1\ \mu$m and the given finesse, one expects for a $P_{\rm out} \approx 2\ \mu$W laser a self-induced ac Stark frequency shift of the order of 1 MHz (using the cavity parameters from [46]). Given the very low power level assumed in this calculation, this shift, although not large in absolute terms, promises effects which may become sizeable with advances in nanolaser construction.

As a final comment on semiconductor lasers, we emphasize the fact that we are limiting our discussion to the analysis of the self-induced ac Stark shift based on the predictions of a two-level model. It is well known that the band structure of a semiconductor laser is much more complex and that numerous other effects (e.g., thermal) contribute to frequency shifts. Nonetheless, our approach highlights a possible contribution to the frequency shift in semiconductor lasers, which to our knowledge has not yet been taken into account. The values we obtain from our predictions suggest that at least for small-size systems (VCSELs and nanolasers) there may be some interesting physics to be further investigated, even though the quantitative relevance of the values we are giving here may be limited.

Class C lasers display the lowest power levels (due to the largest values of Δ in Table II) for achieving sizeable self-induced frequency shifts. In spite of this, however, a shift $\Delta\nu_{\rm acS} = 1$ MHz cannot be achieved in either He-Ne (both IR lines) or He-Xe laser. Indeed, coupled to high gain, these lasers possess a low saturation and cannot provide a very large output power. A comparatively large output power, $P_{\rm out} = 150$ mW, is reported for a He-Ne laser operating at $\lambda = 3.39\ \mu$m [47], but its cavity configuration (i.e., beam radius) is such that the electric field strength is smaller than what assumed in Table II for more conventional lasers. Commercial He-Ne lasers are also expected to display a relatively weak ($\Delta\nu_{\rm acS} \leqslant 200$ Hz) self-induced ac Stark frequency shift [48], with larger values appearing for the $\lambda = 3.39\ \mu$m, in agreement with Table II.

The ammonia laser operating at $\lambda = 374\ \mu$m holds a special place in this discussion. We have already seen that its large cavity losses render $\Delta \approx \frac{\Delta_s}{2}$, which in practice represents the upper limit for the self-induced ac Stark shift. This fact is not sufficient to compensate for a dipole moment strength which is nearly two orders of magnitude lower than that of the He-Xe laser, thus resulting in a required power P over ten times that of the He-Xe.

One interesting aspect of ammonia (and many FIR) lasers—as of many coherently (i.e., laser-) pumped devices—is that the issue of the Stark shift is often discussed. However, what is addressed there is not the self-induced effect that we are discussing in this paper, but the level shift induced by the pumping laser. Since the latter is often strong (stronger than the pumped laser line) it is important, in these lasers, to avoid inducing such a large Stark shift as to detune the transition out of resonance (cf., e.g., [42]). We remark that for the $\lambda = 374\ \mu$m wavelength of NH_3 the self-induced ac Stark shift may be of the order of a hundred kilohertz (cf. see Table I for cavity losses, i.e., conversion between output and intracavity power levels, and Table II for a laser power estimate). Other kinds of Stark shifts are discussed in the context of other lasers. This is the case of the Ar^+ laser, which displays a sizeable Stark shift originating from the high electron density in the very powerful electrical discharge [32]. Once again, this has nothing to do with the effect we are calculating here and should not be confused with our results.

B. Metrological applications

The conclusion to be drawn from the previous section is that no matter which laser we consider, the self-induced ac Stark frequency shift is small, at least relatively to the intrinsic stability of each laser. In absolute terms, the estimates for VCSELs give values which are orders of magnitude larger than those of Class C lasers, but by taking into account the intrinsic sources of frequency shifts in semiconductors, it is not surprising that such shifts may not have been experimentally reported.

Considering, however, the precision required in metrological work, the small shifts we are predicting become relevant. Using the manufacturer's data from Jodon [48] for the $\lambda = 3.39\ \mu$m laser, on the basis of the discussion of the previous section we predict a differential frequency shift of $\frac{d\nu_{\rm acS}}{dP} \approx \frac{1\ \rm Hz}{130\ \mu\rm W}$, amounting to $\Delta\nu_{\rm acS} \approx 77$ Hz when going from the unperturbed atomic level spacing (zero field) to the typical output power $P_{\rm out} = 10$ mW of the smaller commercial laser model. Compared to the sensitivity of the absolute frequency measurement of a $\lambda = 3.39\ \mu$m He-Ne laser used as a reference for metrological standards (standard deviation $\sigma_\omega = 23$ Hz, e.g., [49]), one expects to be able to detect the frequency shift.

However, in metrological setups the laser power is typically not varied. What is important, in such a case, is the amount of coupled amplitude-frequency fluctuations which may result from the amplitude noise of the laser. Using again the manufacturer's data from Jodon, we find that after warmup the amplitude noise is 5%. Thus, considering $P_{\rm out} = 10$ mW, the resulting coupled amplitude-frequency fluctuations turn out to be $\Delta\nu_{\rm fluct} \approx 4$ Hz. This value is still smaller than the best estimate of the absolute laser frequency ($\sigma_\nu = 10$ Hz [50]) but is not entirely negligible and may contribute to the uncertainty. It is reasonable to suppose that the frequency-locking signal should contain detectable traces of the frequency corrections coupled to amplitude ones (especially in the case of slow amplitude drifts). We remark that $\Delta\nu_{\rm fluct} \approx 4$ Hz represents a 5×10^{-14} relative fluctuation on the He-Ne's absolute laser frequency [50].

The push towards better and better precision, aiming at relative values of the order of 10^{-18} [51] (down at least three orders of magnitude from current values), suggests that the coupled amplitude-frequency fluctuations self-induced by the laser's ac Stark shift may become important in the design of laser cavities for metrological applications. Indeed, the simple way of reducing their contribution consists in lowering the power density in the laser cavity by increasing the beam waist size. Notice that the self-induced ac Stark shift transforms amplitude into frequency noise, but that the converse is not true.

One specific domain where coupeld fluctuations may become relevant in the future is the detection of gravitational waves. The perspectives for the optical interferometer-based detection of gravitational waves (successor to the VIRGO project) set the goal in frequency stability at 1 Hz [52], for a third generation of instruments. The very low sensitivity to the self-induced ac Stark shift of Nd:YAG lasers (cf. Table II) makes their use a very good choice also from this point of view, and at the current stability level ($\approx$1 kHz) the effect we are predicting does not play any significant role. However, when the desired sensitivity approaches 1 Hz, coupled amplitude-frequency noise may start playing a role even for these lasers. The cavity design, as well as the choice of stabilization procedure (e.g., injection by smaller, low-power lasers) may play a role and reserve surprises if the self-induced ac Stark shift is not taken into account. As shown in Table II, a microcavity-microchip laser, which is certainly easier to stabilize as a master laser, is not necessarily more immune to the coupled amplitude-frequency noise originating from the ac Stark shift.

C. Pulsed lasers

Strictly speaking our treatment deals with the self-induced ac Stark shift in the continuous wave (cw) regime, and not with laser pulses. Nonetheless, we can use the results of Sec. III A for guidance. Laser pulses have been used since the very first lasers [53], which, indeed, did not operate in the cw regime. Because of the very high field amplitudes which are reached at the peak of a pulse, it is reasonable to expect sizeable self-induced ac Stark shifts, leading to a frequency chirp originating from other than the usual nonlinear effects due to the material response.

Current ultrashort (subfemtosecond) pulses promise large self-induced ac Stark shift values, but our treatment of the problem, based on a two-level atom description, in not adequate to extrapolate to these cases. Standard pulses provide rather modest frequency shifts. For instance, pulses in IR He-Ne lasers have been reported on the $\lambda = 3.39$ μm transition with peak power up to $P_{\text{peak}} = 100$ W [54]. The resulting induced frequency chirp is only $\Delta\nu_{\text{acS}} \approx 3$ MHz. Similarly, pulsed waveguide commercial CO_2 lasers [55] provide chirps of the order of 1 MHz. Even with the large peak power values that can nowadays be obtained with Nd:YAG lasers [56], the resulting values of $\Delta\nu_{\text{acS}}$ remain below 1 MHz.

The situation is quite different when considering mode-locked semiconductor lasers. There is a strong interest in obtaining very short pulses with high repetition rate and relatively large power. Peak power values of the order of 100W in the laser output can be currently obtained [57], thus raising the intracavity electric field to levels well beyond those used for the estimates of the self-induced ac Stark shift in Sec. IV A. The modulation, by the Stark effect, of the self-absorption in the saturable absorber used to obtain ultrafast mode-locking (up to 50 GHz) is being directly modeled without using the RWA [58], but under conditions [59] which are much simpler (Class A systems) than those discussed in the present paper. In this context, the intrinsic self-induced ac Stark may turn out to play an active role in the physics of fast pulses. This point is still under consideration.

V. CONCLUSIONS

The discussion of Secs. II and III has shown that the removal of the RWA and a self-consistent application of expansion techniques (multiscale analysis) allows for the solution of two conceptual problems which have affected the modeling of lasers for the past 50 years: the independence of the laser frequency with laser power and the appearance of transverse structures at $\delta = 0$. From a quantitative point of view, however, these corrections remain small and can be safely neglected in most practical cases.

The relevance of our treatment is to clarify the foundations of models, to offer a way of avoiding paradoxical predictions, and to provide correct quantitative estimates in those cases where a high precision may be demanded.

We have shown in Sec. IV, applying the results of the previous sections to various lasers with widely different properties, that a wide range of values for the self-induced ac Stark shift can be obtained and that although in most cases such a shift is small (relatively to the other sources of frequency shifts), for some lasers it ought to be measurable, or at the very least detectable in the error signal of the stabilization loop. We have also shown that the self-induced ac Stark frequency shift may be potentially important for future precision measurements because of the coupling that it introduces between amplitude and frequency noise.

An open avenue of development concerns the self-induced ac Stark shift in chirped pulses: on the one hand, mode-locked lasers are approaching regimes where the self-modulation of the laser field may become relevant; on the other hand, the very high power levels achieved in ultrashort pulses propagating through a medium may induce a self-modulation of its physical properties (e.g., bandgap). Notice that our analysis fully applies since (1) the electric field strength we are considering preserves the two-level approximation without requiring the simultaneous account for the interaction with additional levels, necessary for very strong fields, and (2) although the amplification of chirped pulses generally occurs in free propagation, the feedback phenomenon discussed in this paper does not require a laser cavity (cf. general expressions for the frequency shift in Sec. III A). Work is under way to further clarify the potential of this line of investigation.

In conclusion, we have shown that the RWA is responsible for an artefact in the description of laser operation, since it hides the true nonlinear dependence of the frequency on the e.m. field amplitude. Quantitatively, the consequences are minor as confirmed by the fact that the RWA has been quite successfully used in a variety of settings. Nonetheless, from a conceptual point of view our analysis clarifies some singular points of the theory and renders the description of laser dynamics consistent with that of standard out-of-equilibrium nonlinear dissipative systems.

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