Nonlinearity and temperature dependence of drive-induced shifts in a thermal environment

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Drive-induced shifts, such as ac Stark shift and Bloch-Siegert shift, are routinely used in various spectroscopies. These shifts are experimentally known to show dispersive Lorentzian behavior as a function of its characteristic frequencies in optical pumping experiments. However, the drive-induced Stark shifts, as calculated using Floquet or dressed atom approaches, do not show the above nonlinear behavior. To address this, we theoretically investigated the drive-induced shifts using a previously reported fluctuation-regulated quantum master equation [A. Chakrabarti and R. Bhattacharyya, Phys. Rev. A **97**, 063837 (2018)]. The shifts are obtained as closed-form expressions over the entire detuning range of the drive. The predicted shifts match satisfactorily with the known experimental data of the light shifts. We show that the calculated shifts are a Kramers-Kronig pair of the drive-induced dissipation in conformity with experimental findings. Moreover, we show that at low temperatures, i.e., for less thermal fluctuations, our results asymptotically match with the known theoretical form of the shifts. In the high-temperature regime, we predict that the shifts decrease in magnitude and are inversely proportional to the square of the temperature.

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I. INTRODUCTION

Driven-dissipative quantum systems play an important role in the fields of quantum information processing [1–5], quantum metrology [6–8], quantum sensing [9–11], and others. An accurate description of a driven-dissipative system requires that all relevant interactions be taken into account at least up to the second order. The observed resonance frequency of such a system appears to be shifted when compared to an isolated system, and a part of the shift is induced by the external drive. The drive-induced shifts (DIS) could be Bloch-Siegert shift originating from the counter-rotating (CR) part of the drive [12,13], or ac Stark shift arising from the detuned corotating part of the drive [14–18]. These shifts find specific utilities in the study of the sidebands in the fluorescence spectrum [19–22], the asymmetry in the Autler-Townes profile [23], enhancement of the atomic interactions in Rydberg atoms [24], the nonlinear Faraday effect [25], etc. Recently, Hung *et al.* showed that, using the Bloch-Siegert shifts of a proton, a power calibration can be done for the rf channels of the low gyromagnetic ratio nuclei [26].

Both of these shifts are estimated as corrections to the Zeeman Hamiltonian (or an effective field along the static Zeeman field). To estimate the DIS, one may adopt the Floquet formalism developed by Shirley for classical fields [27], the method of frame transformations by Pegg and others [15,28–30], or the method of continued fractions by Autler, Townes, and Stenholm [14,31,32]. The usual practice for quantum fields is to employ the dressed-atom approach [33,34]. These theoretical attempts estimate the shifts as a power series in drive amplitude (ω_1). To the leading order, it has been found that the shifts are proportional to the square of the drive amplitude and proportional to the inverse characteristic frequency (ω_s; for Bloch-Siegert shift the characteristic frequency ω_s is the CR frequency while for Stark shifts it is the detuning frequency) [12,17,33]. As such, the drive-induced shifts (DIS) are proportional to ω_1^2/ω_s barring some numerical factors.

The shift of the resonance frequency is also observed during optical pumping experiments [35,36]. The pump beam, aligned along the Zeeman field, has a transverse component of oscillating fields. The oscillating fields give rise to an apparent shift in the resonance frequency, known as the light shift. The origin of the light shift was attributed to two separate mechanisms, namely, (i) light shift due to real transition and (ii) light shift due to virtual transition [37,38]. In one of the original experiments of optical pumping on ^{199}Hg vapor, it was found that the ground state coherence, upon the absorption of a pump photon, was converted to an excited state coherence having different frequencies [35]. The subsequent spontaneous emission to the ground state leaves the original coherence to be phase shifted. The repeated occurrence of this process results in a shift of the resonance frequency, known as the light shift due to real transition. The process has a Kramers-Kronig pair, which is the light narrowing. The mechanism of this type of light shift has been discussed in detail by Kastler, Mathur, Happer, and others [37,39–42]. The light shift due to the virtual transition was explained as ac Stark shift caused by the oscillating electric field of the light beam [37,40]. In the original reports, experiments were performed using circularly polarized light, and hence the Bloch-Siegert shift was not observed [35,36]. Later, the Bloch-Siegert shifts were observed during optical pumping by Arimondo and others [43]. It is interesting that the theoretical estimations of the light shift due to the virtual transitions (LSVT) do not match with that of the

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regular Stark shift. The experiments show that LSVT have a dispersive Lorentzian dependence on the offset frequency. The shift is accompanied by an absorptive light broadening part, which is the Kramers-Kronig pair [37,38,40–42]. We note that the regular Stark shift, as described in the previous paragraph, has no Kramer-Kronig pair, i.e., no absorptive component. It is expected that the theoretical estimation of LSVT should match with the ac Stark shift, since both the effects originate for a magnetic dipole (spin-$1/2$ particle) subjected to transverse oscillating fields. Since LSVT, Bloch-Siegert, and the ac Stark shifts are induced by an externally applied oscillating (or rotating in the case of a circularly polarized field) drive, we broadly classify these shifts as drive-induced shifts (DIS). It is clear from the experimental cue on LSVT that the DIS should ideally be accompanied by a drive-induced dissipative (DID) term as its Kramers-Kronig pair. DID terms require that we treat our system as a driven-dissipative system and not as an isolated quantum system subjected to an external drive.

The driven-dissipative systems are usually treated using quantum master equations (QMEs). In the usual QME treatments, one often considers the drive Hamiltonian in the first order and applies the usual perturbation techniques to the system-bath coupling Hamiltonian to get the Bloch equations [44–47]. However, such approaches fail to give an explanation of the DID from a coherently controlled drive, recently observed in experiments [48]. Also, the theoretical attempts like polaron [49], variational polaron [50], and Keldysh renormalization [51] approaches obtain drive-induced shifts that are identical (in the leading order) to the expressions obtained by Shirley and others [27–34].

Recently, Chakrabarti and Bhattacharyya developed an experimental scheme that showed the existence of drive-induced dissipation (DID) in a driven-dissipative system [48]. Since the known forms of the quantum master equation do not predict the DID, the authors proposed a theoretical framework known as the fluctuation-regulated quantum master equation (FRQME) to explain the observed behavior [52]. Their results (both theoretical and experimental) show an explicit dependence of the environmental correlation time to the drive-induced decay rates. The FRQME predicts that the DID to be an explicit function of the correlation time of the fluctuations (τ_c) of the environment. Since the DID and the corresponding shift appear as a Kramers-Kronig pair, it is expected that the shifts should also carry the signature of the environmental correlation timescale τ_c. Hence, in the present work, in order to mitigate the discrepancy in the understanding of these drive-induced shifts, we use the FRQME. We find a closed generic form of the shifts over the entire range of the characteristic frequencies (ω_s) and show that the calculated shifts are, in general, nonlinear functions of ω_s.

The present paper is arranged as follows. In Sec. II, we provide a brief derivation of the FRQME. In the next section (Sec. III), we describe the system of interest and construct the equations of motion of its density matrix elements in the presence of drive. In Sec. IV, we calculate the shifts from the constructed equations of motion of Sec. III. In Sec. V, various asymptotic limits are discussed where we tally our results with the known forms of the shifts. Subsequently, we analyze and compare our results with known experiments in two subsections A and B. We discuss the temperature dependence of shifts, which is one of the key results of the present paper. Finally, in Sec. VI, we summarize our results by pointing out the major findings of the present work and then briefly discuss its importance in the present understanding of the theory and experiments.

II. QUANTUM MASTER EQUATION AND REGULATION BY FLUCTUATIONS

The fluctuation-regulated quantum master equation (FRQME) was introduced to explain a variety of drive-induced phenomena of a driven-dissipative system [52]. The primary motivation to deviate from the standard quantum master equation (QME) is that the conventional framework does not predict the drive-induced dissipation or the non-Bloch decay recently witnessed in experiments [48]. The drive terms directly appear in the first order in the standard QME after a rotating wave approximation (RWA). On the other hand, the drive-induced shifts are calculated for the non-RWA part of the drive using Floquet or renormalization techniques [27,33,51]. Such shifts are included in the QME as a correction factor, but are not obtained from the derivation of the QME [53]. These calculated shifts show discrepancy with experiments as the detuning tends to vanish [54]. To address this, Chakrabarti *et al.* introduced a FRQME that can include the higher-order influences of drive in a driven-dissipative system [52]. When applied to small quantum systems, such a formalism naturally predicts the DID, whose existence was confirmed earlier by experiments [48]. Most recently, FRQME has been used in quantum information processing to achieve the optimality condition in the gate operation time in the case of both single and multiple qubit gates [55].

The fluctuation-regulated quantum master equation (FRQME) has been obtained using the concept of the regularization by the fluctuation. Since the FRQME is relatively new, we present a brief sketch of the derivation of the FRQME. We begin with the assumption that the thermal fluctuations are ubiquitous in a thermal reservoir and could be adequately represented by a suitably chosen Hamiltonian. As such, the complete Hamiltonian for the system and the local environment in the frequency unit may be written as

$$\mathscr{H}(t) = \mathscr{H}_\circ + \mathscr{H}_{\text{eff}}(t) + \mathscr{H}_{\text{L}}(t), \tag{1}$$

where $\mathscr{H}_\circ$ is the sum of the time-independent Hamiltonian of the system ($\mathscr{H}_{\text{S}}^\circ$) and the local environment ($\mathscr{H}_{\text{L}}^\circ$), $\mathscr{H}_{\text{eff}}$ contains (i) the coupling between the system and the local environment with strength ω_{SL} and (ii) the external drive applied to the system with an amplitude of ω_1. $\mathscr{H}_{\text{L}}(t)$ denotes the fluctuations in the local environment. Since the fluctuations must not destroy the equilibrium of the local environment, $\mathscr{H}_{\text{L}}(t)$ is chosen to be diagonal in the eigenbasis $\{|\phi_j\rangle\}$ of $\mathscr{H}_{\text{L}}^\circ$, represented by

$$\mathscr{H}_{\text{L}}(t) = \sum_j f_j(t)|\phi_j\rangle\langle\phi_j|, \tag{2}$$

where $f_j(t)$'s are assumed to be independent, Gaussian, δ-correlated stochastic variables with zero mean and standard deviation κ, i.e., $\overline{f_j(t)} = 0$, $\overline{f_j(t_1)f_j(t_2)} = \kappa^2\delta(t_1 - t_2)$. We assume that a timescale separation exists between the characteristic time of evolution of the system and that of the fluctuations, the latter being much shorter. This assumption for a rapidly fluctuating local environment is equivalent to the assumption that the environment has a very short memory ($\tau_c \ll 1/\omega_{SL}, 1/\omega_1$). We note that such a short memory corroborates the Markovian nature of the FRQME [47].

The integrated form of the Liouville–von Neumann equation for a finite time interval t to $t + \Delta t$, for a single system in the interaction representation, is given by

$$\tilde{\rho}_S(t + \Delta t) = \tilde{\rho}_S(t) - i \int_t^{t+\Delta t} dt_1 \mathrm{Tr}_L[H_{\mathrm{eff}}(t_1), U(t_1, t)\tilde{\rho}(t)U^\dagger(t_1, t)], \quad (3)$$

where $\tilde{\rho}(t)$ denotes the density matrix of a single system-environment pair (prior to ensemble averaging) in the interaction representation, $H_{\mathrm{eff}}(t)$ is the time-dependent part of the Hamiltonian expressed in the interaction representation, $\tilde{\rho}_S(t)$ denotes the system density matrix obtained by taking a trace over the environment variables (denoted by Tr_L), and $U(t_1, t)$ denotes the propagator for system and the environment pair from time t to t_1 in the Hilbert space. The commutator involving $H_L(t_1)$ vanishes due to the partial trace over environmental degrees of freedom. The trailing t in the propagator is omitted for notational simplicity in the rest of the document. Starting from the Schrödinger equation, $U(t_1)$ can be expressed as

$$\begin{aligned} U(t_1) &= \mathbb{I} - i \int_t^{t_1} H(t_2)U(t_2)dt_2 \\ &= \mathbb{I} - i \int_t^{t_1} H_{\mathrm{eff}}(t_2)U(t_2)dt_2 \\ &\quad - i \int_t^{t_1} H_L(t_2)U(t_2)dt_2. \end{aligned} \quad (4)$$

The above propagator is explicitly linear in H_{eff}. We require a propagator having H_{eff} in the first order, hence all preceding evolution [$U(t_2)$] could be approximated to be solely due to H_L. The assumption of the timescale separation ensures that the evolution due to H_{eff} during the interval t to t_2 can be neglected; the density matrix evolves from t to t_2 solely under H_L with a propagator denoted by $U_L(t_2)$. With this approximation, the Eq. (4) can be written as

$$\begin{aligned} U(t_1) &\approx \mathbb{I} - i \int_t^{t_1} H_{\mathrm{eff}}(t_2)U_L(t_2)dt_2 - i \int_t^{t_1} H_L(t_2)U_L(t_2)dt_2 \\ &\approx U_L(t_1) - i \int_t^{t_1} H_{\mathrm{eff}}(t_2)U_L(t_2)dt_2, \end{aligned} \quad (5)$$

where $U_L(t_1) = \mathbb{I} - i \int_t^{t_1} H_L(t_2)U_L(t_2)dt_2$. We note that the above propagator is finite in H_L through the presence of U_L, but is infinitesimal in H_{eff}.

We substitute Eq. (5) in Eq. (3), and follow the steps outlined by Cohen-Tannoudji *et al.* [47]. So, we use the Born approximation, i.e., at the beginning of the coarse-graining interval, the density matrix could be factorized into that of the system and the environment under an ensemble averaging, and then we use the coarse-graining procedure [47]. Finally, we obtain the FRQME given in the following form:

$$\begin{aligned} \frac{d}{dt}\rho_S(t) = &-i\,\mathrm{Tr}_L\big[H_{\mathrm{eff}}(t), \rho_S(t) \otimes \rho_L^{\mathrm{eq}}\big]^{sec} \\ &- \int_0^\infty d\tau\, \mathrm{Tr}_L\big[H_{\mathrm{eff}}(t), \big[H_{\mathrm{eff}}(t-\tau), \\ &\qquad \rho_S(t) \otimes \rho_L^{\mathrm{eq}}\big]\big]^{sec} e^{-\frac{|\tau|}{\tau_c}}, \end{aligned} \quad (6)$$

where $\tau_c = 2/\kappa^2$ and the superscript "*sec*" stands for the secular approximation that involves ignoring the fast oscillating terms in the quantum master equation. One of the remarkable properties of Eq. (6), which makes it distinct from the other QMEs, is the appearance of an exponential kernel in the evolution of the density matrix. The kernel is often known as the regulator, as it plays an important role to regulate divergences of the physical observables. Such a regulator also ensures that the environmental coherences should vanish within a timescale τ_c. Since τ_c is much shorter than the system's timescale, FRQME essentially is Markovian.

It is worthwhile to mention here that, in Eq. (6), since H_{eff} contains the drive term, the DIS and the DID originate from the double commutator within the integral in the equation. The above equation is in GKLS form, preserves the trace of the density matrix, and is completely positive. A complete derivation of the FRQME is given in [52] and the experimental confirmation of the existence of DID can be found in [48].

III. DRIVE-INDUCED SHIFTS AND THEIR KRAMERS-KRONIG PAIRS

We consider a two-level system (TLS) connected to its local environment, which is assumed to have a large number of degrees of freedom and is undergoing thermal fluctuations. We model such fluctuations as additive noise, as mentioned in the preceding section. Thus, the total Hamiltonian for the system and its local environment could be of the form given by Eq. (1). We assume the explicit form for $\mathscr{H}_S^\circ$ to be $\omega_\circ \sigma_z/2$ without any loss of generality, where σ indicates Pauli spin operators for spin-1/2 particles and $\omega_\circ$ is the splitting between energy levels of the TLS in the angular frequency unit. We make no specific choice for the bare Hamiltonian of the environment ($\mathscr{H}_L^\circ$), or for the coupling between the TLS and the environment ($\mathscr{H}_{SL}$). Since the light shift or the Bloch-Siegert shift does not depend on the coupling, a specific choice is not warranted. $\mathscr{H}_S(t)$ is a coherent time-dependent linearly polarized drive which acts only on the TLS and is of the form $H_S(t) = 2\omega_1 I_x \cos(\omega t)$, where ω_1 and ω are the amplitude and the frequency of the drive respectively and $I_x = \sigma_x/2$. The TLS is driven with a detuned drive with detuning frequency $\Delta\omega$ given by $\Delta\omega = \omega - \omega_\circ$ and CR frequency $\Omega = \omega + \omega_\circ$. We specifically choose a linearly polarized drive to include the effects of the counter-rotating part of the drive in the equation of motion in the form of the Bloch-Siegert shift.

Using the Hamiltonians stated above and the FRQME from Eq. (6), we obtain the following equations for the density matrix elements (ρ_{ij}, where $i, j \in 1, 2$ and 1,2 represent the energy eigenstates of the TLS). We note that the drive shows the usual Bloch like behavior in the first order but for the second order, due to the presence of drive-drive self terms, the equation of motion deviates from the usual Bloch form.

The set of equations for the density matrix in the interaction picture (only for the drive part) is given as follows:

$$\begin{aligned}
\dot{\rho_{11}} &= \frac{i}{2}\omega_1(e^{it\Delta\omega}\rho_{12} - e^{-it\Delta\omega}\rho_{21}) \\
&\quad - \omega_1^2[J(\Delta\omega) + J(\Omega) + \text{c.c.}](\rho_{11} - \rho_{22}), \\
\dot{\rho_{22}} &= -\frac{i}{2}\omega_1(e^{it\Delta\omega}\rho_{12} - e^{-it\Delta\omega}\rho_{21}) \\
&\quad - \omega_1^2[J(\Delta\omega) + J(\Omega) + \text{c.c.}](\rho_{22} - \rho_{11}), \\
\dot{\rho_{12}} &= \frac{i}{2}\omega_1(e^{-it\Delta\omega}\rho_{11} - e^{-it\Delta\omega}\rho_{22}) \\
&\quad - 2\omega_1^2[J(-\Delta\omega) + J(\Omega)]\rho_{12} \\
&\quad + 2\omega_1^2\rho_{21}\, J(\Delta\omega)\, e^{-2it\Delta\omega}, \\
\dot{\rho_{21}} &= -\frac{i}{2}\omega_1(e^{it\Delta\omega}\rho_{11} - e^{it\Delta\omega}\rho_{22}) \\
&\quad - 2\omega_1^2[J(\Delta\omega) + J(-\Omega)]\rho_{21} \\
&\quad + 2\omega_1^2\rho_{12}\, J(-\Delta\omega)\, e^{2it\Delta\omega},
\end{aligned} \tag{7}$$

where $J(\Delta\omega)$ and $J(\Omega)$ are the spectral densities corresponding to the rotating and counter-rotating frequencies of the drive, and c.c. denotes the complex conjugate of the preceding terms. The overhead dot represents the total time derivative of the density matrix which is assumed to be approximately equal to the coarse-grain time derivative within the coarse-graining timescale Δt. The functional form of J as a function of a generic frequency ω_s is given by

$$J(\omega_s) = \frac{1}{4}\int_0^\infty d\tau\, e^{i\omega_s\tau} e^{-|\tau|/\tau_c} \tag{8}$$

with the additional properties

$$J(-\omega_s) = J^\star(\omega_s). \tag{9}$$

In the equation of motion, the terms involving explicit time are the result of going to the interaction picture, which differs from the drive frequency by an amount $\Delta\omega = \omega - \omega_\circ$. Such time dependence could be easily removed by moving to a so-called rotating frame of reference where the observables can be defined as $M_\alpha(t) = e^{-iI_z\Delta\omega t}\, I_\alpha\, e^{iI_z\Delta\omega t}$ with $\alpha = x, y, z$.

Moreover, the oscillations in the off-diagonal elements of the density matrix with frequency $2\Delta\omega$ survive due to the secular approximation [47] that sets up a cutoff frequency $(1/\Delta t)$ over which all fast fluctuations are averaged out and the slow ones are retained.

The spectral densities calculated from the drive has a real as well as a complex part. The real part corresponds to the decay from the drive, that has been studied and experimentally verified in [48], and the imaginary part is the source of all kinds of shifts due to the presence of the drive in the system. In this paper, We only focus on the shift terms and analyze their behavior in the light of the FRQME.

IV. DRIVE-INDUCED SHIFTS FROM THE FRQME

The resonant shifts are calculated using off-diagonal elements of the above density matrix for the TLS. To this end, we define the density matrix elements in the following way:

$$\tilde{\rho}_{11} = \rho_{11}, \quad \tilde{\rho}_{22} = \rho_{22}, \tag{10a}$$

$$\tilde{\rho}_{12} = \rho_{12}\, e^{i\Delta\omega t}, \quad \tilde{\rho}_{21} = \rho_{21}\, e^{-i\Delta\omega t}, \tag{10b}$$

$$\tilde{\rho}_+ = \tilde{\rho}_{12} + \tilde{\rho}_{21}, \quad \tilde{\rho}_- = i\,(\tilde{\rho}_{12} - \tilde{\rho}_{21}). \tag{10c}$$

In terms of $\tilde{\rho}_+$ and $\tilde{\rho}_-$, Eq. (7) simplifies to

$$\begin{aligned}
\dot{\tilde{\rho}}_+ &= -\,\omega_{d+}\,\tilde{\rho}_+ + (\Delta\omega - \Omega_{\rm BS})\,\tilde{\rho}_-, \\
\dot{\tilde{\rho}}_- &= -\,\omega_{d-}\,\tilde{\rho}_- - (\Delta\omega - \Omega_{\rm BS} + \omega_{\rm LS})\,\tilde{\rho}_+ - \omega_1(\tilde{\rho}_{11} - \tilde{\rho}_{22}),
\end{aligned} \tag{11}$$

where

$$\omega_{d+} = \frac{\omega_1^2}{2}\left[\frac{\tau_c}{1 + \Omega^2\tau_c^2}\right], \tag{12a}$$

$$\omega_{d-} = \frac{\omega_1^2}{2}\left[\frac{\tau_c}{1 + \Omega^2\tau_c^2} + \frac{\tau_c}{1 + \Delta\omega^2\tau_c^2}\right], \tag{12b}$$

$$\Omega_{\rm BS} = \frac{\omega_1^2}{2}\left[\frac{\Omega\tau_c^2}{1 + (\Omega\tau_c)^2}\right], \tag{12c}$$

$$\omega_{\rm LS} = -\frac{\omega_1^2}{2}\left[\frac{\Delta\omega\tau_c^2}{1 + (\Delta\omega\tau_c)^2}\right]. \tag{12d}$$

In the above, ω_{d+} and ω_{d-} are two dissipative rates due to the external drive. $\omega_{\rm LS}$ and $\Omega_{\rm BS}$ represent the light shift and the Bloch-Siegert shift, respectively. So, it is clear from the above expressions that both the light shift and the Bloch-Siegert shift, i.e., DIS, could be generally expressed as

$$\omega_{\rm DIS} = \frac{\omega_1^2}{2}\left(\frac{\omega_s\tau_c^2}{1 + \omega_s^2\tau_c^2}\right), \tag{13}$$

where ω_s is a symbolic representation for the characteristic frequency of a DIS, e.g., for Bloch-Siegert shift $\omega_s = \Omega$ and for light shift $\omega_s = -\Delta\omega$.

V. RESULTS AND DISCUSSIONS

Figure 1 shows the behavior of light shifts ($\omega_{\rm LS}$) as a function of inverse detuning frequency. For an intuitive understanding, we consider the shift at two extreme limits of $\Delta\omega\tau_c$. (a) In the high detuning limit, i.e., $\Delta\omega \gg 1/\tau_c$, the shift can be written as $\omega_{\rm LS} \sim \omega_1^2/2\Delta\omega$ (shown by dashed straight lines in Fig. 1). This form exactly matches with the leading order shift term calculated for the ac Stark shift using Floquet or continued fraction methods for classical fields or from dressed states approaches for the quantum fields [27–34]. We note that the high detuning limit implies that a direct substitution $\Delta\omega = 0$ is not allowed in the above expression. (b) On the other hand, in the limit of near-resonant excitation, or low detuning, i.e., $\Delta\omega \ll 1/\tau_c$, the shift is obtained as $\omega_{\rm LS} \sim \omega_1^2\Delta\omega\tau_c^2/2$. Therefore, at low detuning, the light shift is proportional to the detuning frequency and vanishes for $\Delta\omega = 0$.

Overall, the light shift shows a dispersive behavior, as was observed and explained originally by Cohen-Tannoudji *et al.* [37,38]. A comparison to LSVT shows that $1/\tau_c$ plays the

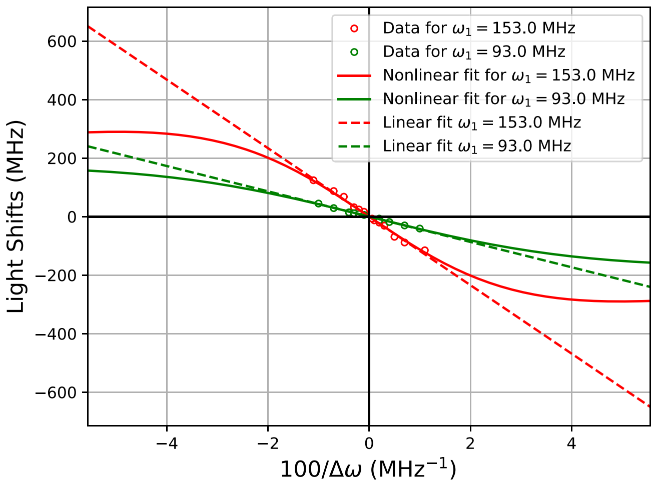

FIG. 1. Plots of the calculated shifts and the fitting of the earlier reported data of the light shift by Tamarat *et al.* [18]. The red and the green open circles represent the experimental light shift data for $\omega_1 = 153$ and 93 MHz, respectively, for two different molecules, as reported by Tamarat *et al.* [18]. We fit these data using the Eq. (13) and the fit is plotted with the solid line. For a comparison, the dashed lines are used to show the conventional linear fit of the data (also calculated by Tamarat *et al.* [18]). The fitting yields the values of τ_c as $\sim$ 50 and 77 ns. The range of τ_c values are in agreement with the range of values reported by Izeddin *et al.* [56]. We note that both the fits are completely equivalent within the experimental errors. But the fit with Eq. (13) in solid lines show remarkable departure from the linear behavior and completely matches with the dispersive Lorentzian behavior reported elsewhere (see Fig. 2).

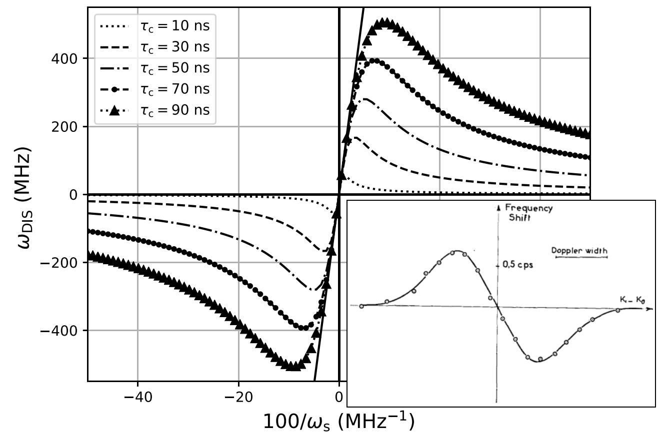

FIG. 2. Plot of the generic drive-induced shift $\omega_{\rm DIS}$ as a function of the inverse characteristic frequency for different values of τ_c. The values of τ_c are chosen in the nanosecond range, a typical timescale for quantum dots [56]. Also the drive amplitude is set to $\omega_1 = 150.0$ MHz as typically used in optical pumping experiments [18]. The solid straight line denotes the common asymptote of the plotted curves, which clearly shows that $\omega_{\rm DIS}$ deviates from linearity depending on the values of τ_c. The inset in the lower-right corner shows the complete LSVT behavior as depicted in one of the earliest reports by Kastler [37]. The similarity between Kastler's work and ours is clearly evident (we note that for light shift $\omega_s = -\Delta\omega$).

same role as the Γ factor in Eq. (17) of Kastler's paper or in the works of Cohen-Tannoudji *et al.* [35,37,38]. We note here that the calculated DIS follows the same dispersive behavior and is a nonlinear function of its characteristic frequency. To emphasize the nonlinearity of $\omega_{\rm DIS}$ and its dependence on the timescale of the correlation of the fluctuations, we plot the shift as a function of its characteristic frequency for different values of τ_c in the next figure.

Figure 2 shows that DIS has a dispersive Lorentzian behavior with extrema at $\pm 1/\tau_c$, which can be calculated from Eq. (13). The distance between the extrema $(2/\tau_c)$ is but a measure of how the generic shift spreads out as a function of the inverse characteristic frequency ω_s. The figure shows that at very low τ_c, the departure of the shifts from linearity is prominent even for small values of inverse detuning frequency. On the other hand, at a high τ_c limit, it retains the linear shape for a larger range. Moreover, to study the explicit dependence of τ_c in the expression of shifts, a series expansion is performed in terms of $\omega_{\rm s}$ in different asymptotic limits. For the near-resonant excitation of TLS in magnetic resonance and in optical experiments, a physically meaningful limit is $\omega_{\rm s} \ll 1/\tau_c$, and in this limit the DIS can be expanded as

$$\omega_{\rm DIS} \approx \frac{\omega_1^2}{2}\left[\omega_{\rm s}\tau_c^2 - \omega_{\rm s}^3\tau_c^4 + \cdots\right]. \tag{14}$$

On the other hand, in optical experiments [e.g., terahertz (THz) spectroscopy, pump-probe spectroscopy, etc.] in the case of off-resonant detuned drive, the other limit $\omega_{\rm s} \gg 1/\tau_c$ is more realistic. In this limit, the expansion of DIS is given by

$$\omega_{\rm DIS} \approx \frac{\omega_1^2}{2}\left[\frac{1}{\omega_{\rm s}} - \frac{1}{\omega_{\rm s}^3\tau_c^2} + \cdots\right]. \tag{15}$$

We note that this expression has the usual form of ac Stark shift or Bloch-Siegert shift in the leading order in inverse characteristic frequency, with correction terms involving τ_c present in all higher orders [27–34].

It is important to note that the parameters responsible for the systematic expansion of the density matrix are the strength of the drive (ω_1) and the correlation time of the environment (τ_c). As long as their product is sufficiently smaller than one, i.e., $\omega_1\tau_c \ll 1$, the expansion is rapidly convergent. This condition originates from the timescale separation between the system and the environment beyond which the FRQME is no longer valid.

In the case of Bloch-Siegert shift, the expansion is performed only in the limit $\Omega\tau_c \gg 1$ because for the high CR frequencies the other limit becomes unimportant. A notable exception is liquid-state magnetic resonance experiments, where τ_c is typically in the picoseconds timescale [48], and hence with Larmor frequency in the range of hundreds of MHz it is quite possible to achieve $\Omega\tau_c \ll 1$. In the limit $\Omega\tau_c \gg 1$, the Bloch-Siegert shift recovers its usual form [12,13] in the first order in CR frequency, and eventually, the higher orders become τ_c dependent like the light shifts. The expressions are similar to Eq. (15) with $\Delta\omega$ is replaced by Ω. Unlike Shirley *et al.* [27] or Cohen-Tannoudji *et al.* [33], this analysis is restricted to the second-order contribution of the drive to the drive-induced shifts. However, even within a second-order theoretical framework, one can easily observe that the shifts deviate prominently from the standard expression of ω_1^2/ω_s as a function of ω_s.

A. Comparison with experiments

Our expressions of the dispersive nature of the light shift match fairly well with the original reports of LSVT [35]. In optical experiments, unless the drive is near resonance, the experiments are done mostly with high detuned frequency. Therefore, the drive-induced shift or the light shifts obtained there explicitly shows a linear dependence on the inverse detuning frequency, as observed in [18]. The experiments were not performed for a sufficiently wide range of $\Delta\omega$ to have a fair comparison with our expressions. However, later experiments by Appelt *et al.* on rubidium in a high-pressure optical pumping cell show the dispersive nature of the light shift with the variation of the optical pump wavelength [57]. We note that these experimental results are explained as an effect of the collision-based spin-exchange process, similar to a light shift due to a real transition. Subsequent experiments by Savukov *et al.* with K atoms in He-buffer gas cell also find the dispersive nature of the light shift as a function of the wavelength of the pump. Their work confirms that, at sufficiently small detuning of the pump laser, the shift becomes vanishingly small and eventually disappears at the center of resonance [54], which exactly matches with our calculation.

For a comparative study, a plot of the surroundings of the resonance and our result is given in Fig. 1. The experimental data are extracted from [18] in which the data of the light shift are given for the inverse detuning range −0.0125 to 0.0125 MHz^{-1}. Figure 1 shows that our results are in good agreement with the experimental data of the light shifts given for two different drive powers, namely 93 and 153 MHz. Moreover, as evident in Fig. 2, the calculated behavior of the light shift completely matches with the earlier reports of the dispersive Lorentzian-like behavior of LSVT by Kastler [37]. Therefore, the two figures complete the comparison of our results to the entire range of reported light shifts (linear as well as nonlinear regimes).

B. Temperature dependence of the shifts

The fluctuations in the local environment of the system are assumed to originate from the thermal processes. Also, experimental validations of drive-induced dissipations in liquid-state nuclear magnetic resonance (NMR) show a temperature dependence of the timescale of the fluctuations [48]. Therefore, we venture to use a scaling argument that the average energy of the fluctuations may be equated to the characteristic parameters (such as τ_c) of the fluctuations ($h/\tau_c \approx k_B T$), where the symbols have their usual meanings. As a consequence, the drive-induced shifts become temperature dependent through τ_c. A tentative plot of the DIS ($\omega_{\rm DIS}$) as a function of temperature is shown in Fig. 3. At low temperatures (i.e., high τ_c), DIS tends to $\approx \omega_1^2/\omega_s$, barring a numerical factor, as shown earlier. This form is independent of τ_c and hence does not depend on the temperature. In Fig. 3, this limit is indicated by the horizontal dashed line near the top of the figure. But for very low τ_c, which corresponds to higher fluctuations, i.e., high temperature, the temperature dependence of $\omega_{\rm DIS}$ is pronounced, and the leading behavior is given by $\omega_{\rm DIS} \propto \frac{1}{T^2}$. Also, for the near-resonant excitation $\Delta\omega \to 0$, the nutation frequency which depends on $\omega_{\rm DIS}$ becomes temperature dependent, even in the cases where Lamb shift could be neglected, i.e., for the weak system-environment coupling regime.

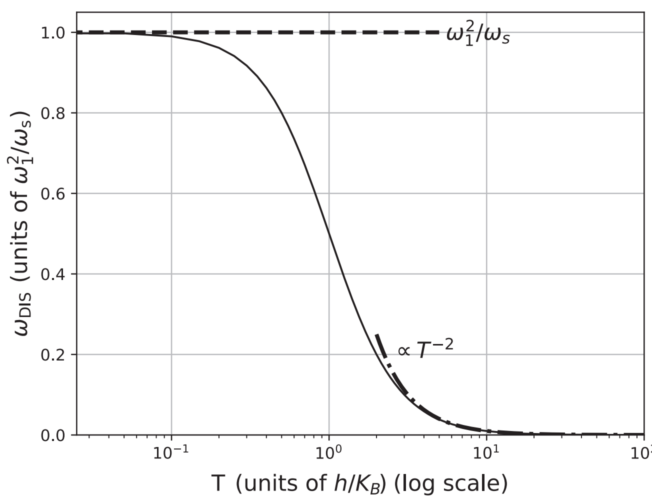

FIG. 3. Plot of the generic shift $\omega_{\rm DIS}$ as a function of the temperature. In this figure, the parameters are normalized to unity, i.e., $\omega_1 = \omega_s = 1.0$. The horizontal dashed line corresponds to the known form of ac Stark shift in the low-temperature regime [27–34]. The dashed-dot curve shows the shifts decay with the increase in temperature following $1/T^2$ behavior, where T is the temperature.

VI. CONCLUSION

We have used the fluctuation-regulated quantum master equation (FRMQE) to calculate an explicit closed-form expressions of drive-induced shifts (DIS), such as ac Stark shift and Bloch-Siegert shift. We obtained the generic drive-induced shift to be a dispersive Lorentzian function of its characteristic frequency, which at the appropriate limits matches with the hitherto known forms of ac Stark shift or Bloch-Siegert shift. The dispersive Lorentzian forms of DIS help to explain the observed dispersive nature of the light shift due to virtual transitions [35,37,38]. The DIS are found to be the Kramers-Kronig pair of experimentally observed drive-induced dissipation (DID) terms [48]. In line with the observed experimental temperature dependence of DID terms, we find DIS terms to be temperature dependent. At low temperatures, the DIS are shown to assume a limiting value independent of temperature and are identical to the hitherto known form of DIS. At high temperatures, the DIS are predicted to decrease as the inverse square of the temperature. We note that, although the analysis was performed with a TLS, FRQME could easily be extended to atoms with high spins or more than two levels. For two energy levels having a well-separated transition frequency from other levels in a multilevel system, TLS remains a fair approximation, and, as such, the calculated shifts could be used even for multilevel quantum systems. The closed form of DIS obtained by us results from a complex interplay between the drive, dynamical fluctuations of the environment, and the system of interest (i.e., TLS), which cannot be realized within the model framework of a simple system connected to its environment. Therefore, our analysis of such drive-induced shifts provides a deeper understanding of the theoretical formulation of the driven-dissipative system. Also, most recent studies on the spectrum of atoms near strong magnetic fields (magnetic fields from rotating stars, e.g., neutron stars) proposed the Bloch-Siegert shifts as a reason for the variations of the fundamental astrophysical constants of nature over time [58]. Therefore, in such a scenario our analysis of such shifts will provide additional insights into determining the variations of the fundamental constants of nature. Finally, we envisage that our results can be used to develop an experimental protocol for measuring the autocorrelation time (τ_c) of the fluctuations (or the strength of the fluctuations $\kappa^2 = 2/\tau_c$) by careful measurements of the drive-induced shifts in near-resonance conditions.

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