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We theoretically study nondestructive detection of ultracold molecules, using a Fabry-Pérot cavity. Specifically, we consider vacuum Rabi splitting where we demonstrate the use of collective strong coupling for detection of molecules with many participating energy levels. We also consider electromagnetically induced transparency and transient response of light for the molecules interacting with a Fabry-Pérot cavity mode, as a means for nondestructive detection. We identify the parameters required for the detection of molecules in the cavity using electromagnetically induced transparency. The theoretical analysis for these processes is parametrized with realistic values of both the molecule and the cavity. For each process, we quantify the state occupancy of the molecules interacting with the cavity and determine to what extent the population does not change during a detection cycle.

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I. INTRODUCTION

There are a number of motivations for creating ultracold molecules, such as quantum computation [1] and quantum simulation [2] with polar molecules, exotic phases of matter with long range interaction between particles [3], controlling chemical reactions [4], the study of few body dynamics of identical particles [5], etc. In addition, trapping of polar molecules will enable a study of potentials of the form 1/r^6 (rotating dipoles), 1/r^3 (static dipoles), and 1/r^2 (ion + static dipole) when co-trapped with ions. Here r is the distance between the interacting particles.

To meet these and other goals, great effort is geared towards developing techniques to create long-lived molecules at ultracold temperatures [6–16], particularly in their rovibrational ground state [6–13], and to detect them efficiently. In most experiments, detection is destructive and requires multiple cycles of system preparation and detection [7,17–22]. The techniques which are used to detect molecules in most experiments rely on (i) the ionization of the molecules and the detection of the resulting ions on an ion detector [17–19], or (ii) the conversion of the molecules back to atoms and the detection of the atoms [7,20–22], or (iii) photon shot-noise limited absorption imaging on a strong but open bound-bound molecular transition [23]. All these detection techniques result in destruction of the molecules at the end of the detection cycle. The development of a nondestructive technique to detect state-selected ultracold molecules would represent a major accomplishment for the study of ultracold molecules. This would enable repeated interrogation of the same molecular ensemble, building experimental statistics, and tracking of the dynamics of the molecular ensemble.

The goal of the present article is to devise a detection technique for ultracold molecules in a nondestructive manner, through the enhancement of their interaction with an electromagnetic field generated in an optical cavity. Two concave, reflecting mirrors form a Fabry-Pérot cavity, which can trap photons for a long duration. This trapping results in the confinement of electric field due to a single photon within a very small volume, which enhances the interaction time of the photon with the atoms, ions, or molecules coupled to the cavity. Such enhancement will be useful for not just ultracold molecules but also for organic dye molecules [24].

Unlike few-level atoms, detection of molecules using resonant light is not straightforward due to their large number of vibrational and rotational levels. When a ground-state molecule prepared in a particular rovibrational level absorbs a photon to populate a rovibrational level of an excited electronic state, there are multiple accessible ground-state rovibrational levels to which the molecule can relax by a single spontaneous emission. If a rovibrational level other than the initial one is populated due to emission the molecule is lost for further imaging by the light, which is resonant with the initial rovibrational transition. However, it may happen that there are a few molecules that can be detected using fluorescence imaging [14] if the excited level primarily decays to a small number of
ground-state rovibrational levels. However, such molecular species are rare and cooling them to ultracold temperatures requires elaborate techniques [15,16].

Here, we explore the possibility of detecting ultracold molecules through dispersive effects rather than absorption processes. As these dispersive effects are usually weaker than absorptive effects, we use a cavity to enhance them. We discuss various advantages and disadvantages of detecting molecules using a cavity. Recently, a nondestructive detection technique using Rydberg atoms was proposed for ultracold polar molecules [25]. However, the introduction of other interacting species, similar to [25] might result in the loss of ultracold molecules due to inelastic processes.

Dispersive effects have been used previously to detect two-level atoms nondestructively by recording changes in phase of light propagating through the atomic cloud [26,27]. However, for thin atomic clouds optimized detection through phase change has an exactly same signal-to-noise ratio as optimized detection through absorption of light, if the amount of absorbed light is kept the same for both cases [28]. Lye et al. [28] also showed that the use of an optical cavity greatly enhances the signal-to-noise ratio for detection through phase shift. However, they did not explore the collective effect of atoms on the cavity mode. Because of the collective coupling of molecules to a cavity, molecule detection in frequency space can be performed using few photons and a very slight destruction of the populations. In this article, we focus on cavity-based, nondestructive detection techniques for molecules, atoms, and ions possessing multiple levels, which use light as a measuring medium. Assuming that an ensemble of ultracold molecules in a single quantum state is prepared, we exploit in our proposal (i) the collective strong coupling to a cavity and the corresponding vacuum Rabi splitting (VRS) [29–33] and (ii) the electromagnetically induced transparency (EIT) [34–36] for the cavity mode. The effectiveness of these options as cavity-based nondestructive detection mechanisms and the requirements for being practically implemented are discussed in detail.

II. MOLECULAR TRANSITIONS FOR DETECTION

There are numerous experiments which have been successful in creating molecules in the rovibrational ground state [6–13]. For specificity and feasibility of molecular detection in a cavity, we choose the example of ultracold Rb$_2$ molecules. We assume that the Rb$_2$ molecules are all populated in the lowest rotational and vibrational level ($v_e = 0, J_e = 0$) of the electronic ground state $X^1 \Sigma_g^+$. For all the calculations in this article the $v_e = 1$ and $J_e = 1$ level of the excited electronic state $B^1 \Pi_u$ forms the excited state for all optical transitions. For the sake of the present investigation, the $B^1 \Pi_u$ state is considered as isolated, i.e., not coupled by spin-orbit interaction to other neighboring electronic states. The relevant parameters for the transitions from this excited state are shown in Table I.

The square of above mentioned transition dipole moments are equal to $S_{J'J} | \int \psi_{\nu e}^*(R) \times d(R) \times \psi_{\nu e}(R) \, dR|^2$ [37], where $\psi_{\nu e}(R)$ is the wave function of the vibrational level $\nu_e(v_e)$ as a function of distance between the atomic cores $R$ calculated using available potential energy curves [38,39]. $d(R)$ is $R$ dependent dipole moment for the electronic transition [39,40], and $S_{J'J}$ is the Hön-London factor for the rotational levels [41]. The wave functions $\psi_{\nu e}(R)$ are calculated using the LEVEL code [37,42], which is based on the Cooley-Numberov method.

In the following sections, the described processes could be considered for atoms, ions, or molecules. Therefore, we will use the word “atom” for the sake of simplicity, unless otherwise stated when features specific to molecules will be invoked.

III. MOLECULE DETECTION WITH VRS

When multiple atoms are coupled to a cavity such that the atom-cavity system is in the collective strong coupling regime, the empty cavity single peak of transmission for a probe light beam through the cavity splits into two nondegenerate transmission peaks. This splitting is called vacuum Rabi splitting [29–33]. The frequency separation between the two peaks is equal to $2g_0 \sqrt{N}$, where $g_0$ is the coupling strength of a single atom with the cavity and $N_e$ is the effective number of atoms coupled to the cavity. The measured splitting between the two VRS peaks informs on the number of atoms coupled to the cavity. This splitting is clearly observed for a two-level atom, where the spontaneous radiative decay of the excited level $|g\rangle$ back to the ground-state level $|e\rangle$ does not uncouple the atom-cavity system. When the VRS is measured on an atom with an additional level $|g\rangle$, the atoms get rapidly pumped in the third state leading to a collapse of the VRS signal. In such a simple level scheme the VRS signal can be recovered by the addition of another laser to ensure the repumping of the $|g\rangle$ population into the $|g\rangle$ level [33]. However, this is not possible in molecules with a large number of loss channels. Having such a repumping laser is not practical for most molecular species due to the large number of rovibrational levels in the ground state of the molecules, which prevents the realization of a closed optical transition [43]. Therefore, the question is whether molecules can at all be detected using VRS, and if so under which conditions. A plausible strategy is to check if the VRS measurement is much faster than the photon absorption

\begin{table}[h]
\centering
\begin{tabular}{|l|c|}
\hline
Transition dipole moment with $v_e = 0, J_e = 0$ level of $X^1 \Sigma_g^+$ state & $4.8 \times 10^{-29} \text{ C m (14.2 D)}$ \\
Transition dipole moment with $v_e = 1, J_e = 0$ level of $X^1 \Sigma_g^+$ state & $5.1 \times 10^{-29} \text{ C m (15.3 D)}$ \\
Decay rate to $v_e = 0, J_e = 0$ level of $X^1 \Sigma_g^+$ state & 401.5 kHz \\
Decay rate to $v_e = 1, J_e = 0$ level of $X^1 \Sigma_g^+$ state & 456.6 kHz \\
Total decay rate to all levels of $X^1 \Sigma_g^+$ state & 6.44 MHz \\
\hline
\end{tabular}
\caption{Parameters for molecular transitions considered in this article, with $v_e = 1$ and $J_e = 1$ level of the state $B^1 \Pi_u$ as the excited state [38–40].}
\end{table}
For the atom-cavity system defined above, the evolution of the atomic states and the cavity field, after including spontaneous emission rates, and assuming the cavity field to be classically denoted by a coherent state |α⟩, result in the set of coupled differential equations [44–46],

\[ \frac{d\rho_{ge}(t)}{dt} = -\frac{\Gamma_1}{2} \rho_{ge}(t) + i g_0 \alpha(t) \rho_{ge}(t), \]

(3a)

\[ \frac{d\rho_{eg}(t)}{dt} = -\frac{\Gamma_1}{2} \rho_{eg}(t) + i g_0 \alpha(t) \rho_{ge}(t) - \eta, \]

(3b)

\[ \frac{d\rho_{ec}(t)}{dt} = -\frac{\Gamma_1}{2} \rho_{ec}(t) + i g_0 \alpha(t) \rho_{eg}(t) - \eta, \]

(3c)

Here, ρ’s with single letter as subscript are the populations of the levels and the ones with two-letter subscripts are coherences between levels, ∆pc = ωp − ωcv. Γ1 and Γ2 are the decay rates of the excited state (e) to the ground states |g⟩ and |g’⟩ respectively, η is the rate at which classical light is injected into the cavity from the incident probe light, κ1 is the decay rate of the cavity field, and Γ1 = Γ1 + Γ2 is the total decay rate of the excited state. |g⟩ represents all the dark ground states of molecules as can be seen from Fig. 1. Its major role is to bleach the atoms from the transition interacting with the cavity so it is irrelevant whether it is just one dark ground state or many, for calculations in this section.

For computational ease, in deriving Eq. (3) we assumed Nc atoms couple to the cavity with equal strength g0. Nc can be obtained by computing the overlap of the cavity mode function f(x,y,z) and the atomic density profile [33,46–48]. Hence we remove the subscript j while going from Eq. (1) to Eq. (3) (see [46,48] for more details). This simplification enables solving the time-dependent differential Eq. (3) by numerical integration to obtain the power of light transmitted by the atom-cavity system [46,48]. Scanning across the atom-cavity resonance, we obtain a VRS signal, as shown in Fig. 2. In the simulation, the detuning of the probe laser is adiabatically increased and the corresponding change in cavity output power is monitored. Here the change is adiabatic with respect to the atomic and cavity rates. The detuning of the probe laser is scanned over 200 steps of 0.005 ms (0.1 ms in total), resulting in a scan rate of 10 kHz. The probe light power (Ppw = 0.23 nW) is such that the maximum output of the cavity during the detection stage is Pout = 10 pW, thus corresponding to 4 × 10⁷ photons per second. With these parameters, the maximum number of photons available for detection at each step of the scan in Fig. 2(a) is 10. If the photon detection efficiency is 50%, this gives 50-ns delay time between two photons on average. This time duration is equal to the typical dead time of single-photon avalanche photodetectors (APD) [49] making such a detection feasible. The maximum photon occupancy in the cavity mode for these parameters is three. The cavity mirror separation is taken to be 11.8 mm and the radius of curvature of the mirrors is 10 mm yielding a waist size of 30 μm for the cavity mode at the center of the cavity. These typical cavity parameters are taken...
atom-cavity resonances. At the start of the scan, all atoms are assumed showing VRS for various atom numbers. (b) Probability for the atoms function of the detuning of the probe laser from the cavity frequency from the experimental work of Albert et al. [47]. Here |e⟩ is the νe = 1, J_e = 0 level of the B1Π_u electronically excited state, |g⟩ is the νg = 0, J_g = 0 level of the X1Σ_g^+ electronic ground state as explained in Sec. II. Throughout this article, we assume that only the above mentioned transition couples to the cavity giving a maximum coupling constant of g_0/2π = 219.2 kHz. |g^⟩ represents all other levels of X1Σ_g^+. Other parameters for Fig. 2 are, κ_g/2π = 2.5 MHz, the loss rate from input mirror of cavity κ_{itin} = 0.1κ_0, and loss rate from output mirror of cavity κ_{oout} = 0.8κ_0.

From Fig. 2(b) we see that the larger the number of atoms the smaller the leakage into the dark ground state (|g^⟩). This results from the higher atom number shifting the resonance frequencies of the VRS peaks away from the atomic resonance, and hence the probability of photon absorption for an atom reduces. For N_c = 5 × 10^4, 0.54% of the atoms will be lost to the dark ground state per scan and for N_c = 1 × 10^6, 0.003 atoms will be lost. These numbers suggest that such a detection scheme is feasible.

In order to keep the absorption and spontaneous emission of a photon by single atoms low, the photon occupancy of the cavity needs to be minimal. This results in a lower flux of photons out of the cavity, consequently reducing the detection probability. To ensure an optimal detection, the loss from cavity mirrors other than the loss due to leakage from output mirror should be minimized. This is achieved using an ultrahigh reflectivity input mirror and a moderately reflective output mirror such that most of the losses from the mirrors are due to transmission rather than absorption and scattering losses. This results in a high ratio κ_{oout}/κ_0, where κ_{oout} is the loss rate from the output mirror of the cavity and κ_0 the total loss rate from the cavity mirrors. For instance, keeping the output photon flux and all other parameters identical to those of the simulation of Fig. 2, a low ratio of κ_{oout}/κ_0 will result in a 64% loss of atoms in the case of N_c = 5 × 10^4 and a 0.3% loss of atoms in the case of N_c = 1 × 10^6, per scan. Hence it is important to keep κ_{oout}/κ_0 as high as possible. In addition, having a larger value of κ_0 is advantageous in this case as this means a smaller lifetime of the photon inside the cavity. This reduces its probability of being absorbed, and the steady state inside the cavity is achieved faster, enabling a faster scan. For example, if we keep the ratio mentioned above, output photon flux, and other parameters same as for Fig. 2, κ_0 = 20 MHz will result in a 0.3% loss of atoms in the case of N_c = 5 × 10^4 and a 0.0015% loss of atoms per scan in the case of N_c = 1 × 10^6, and κ_0 = 0.5 MHz will result in 2% loss of atoms in the case of N_c = 5 × 10^4 and a 0.012% loss of atoms per scan in the case of N_c = 1 × 10^6. However, we cannot keep on increasing κ_0 because this will result in a very broad cavity linewidth which will engulf the VRS. This suggests that a careful choice of cavity parameters is critical for the success of the scheme.

IV. DETECTION THROUGH CAVITY EIT

In this section, we explore the detection of molecules interacting with a cavity using electromagnetically induced transparency [34–36]. In this phenomenon a frequency window of transparency is opened for a probe laser which would have been absorbed by an ensemble of atoms (or ions, or molecules) resonant with the probe light. This transparency is induced by another strong light beam interacting with the same three-level ensemble. There is a steep dispersive effect, and little or no absorption for a probe light near the EIT peak [35,50].

To start with, we first analyze the ideal case of three-level atoms coupled to the cavity and later extend the analysis to molecules which are equivalent to leaky four-level atoms. The three levels with two ground states (|g⟩ and |g^⟩) along with an excited state (|e⟩) form a lambda system [35] as shown in Fig. 1(b). In contrast to VRS, a second light field which couples the ground state (|g⟩) to the excited state is required for EIT. We call it control beam which has a frequency ω_c. In a frame rotating at the probe frequency (ω_p), the Hamiltonian for stationary atoms for such a case is

\[
\hat{H} = \hbar \sum_{j=1}^{N} \left[ -\Delta_{pa}\hat{a}_{ej}^\dagger + (\Delta_{pa} + \Delta_{ra})\hat{a}_{gj}^\dagger \right.
\]

\[
+ g_j \left( \hat{a}_{gj}^\dagger \hat{a}_{ej} + \hat{a}_{ej}^\dagger \hat{a}_{gj} \right) + \Omega \left( \hat{a}_{gj}^\dagger + \Omega \hat{a}_{ej} \right). \]

(4)

Here, \(2\Omega = -\mu g_e E/\hbar\) is the Rabi frequency for the control beam, where \(\mu g_e\) is the transition dipole moment for the transition |g⟩ → |e⟩, and E is the electric field amplitude of the control beam. \(\Delta_{ra} = \omega_r - (\omega_c - \omega_p)\) is the control...
laser detuning from the $|g′⟩ \leftrightarrow |e⟩$ transition. Using Eq. (2) the system evolution is described by the coupled differential equations

$$\frac{dα}{dt} = -\eta - \alpha(κ_i - iΔ_p) - i \sum_{j=1}^{N} g_{j}ρ_{ge}^j,$$

(5a)

$$\frac{dρ_{ge}^j}{dt} = \left( -\frac{Γ_i}{2} + iΔ_p \right) ρ_{ge}^j - iαg_{j}(ρ_{ge}^j - ρ_{eg}^j) - iρ_{ge}^jΩ,$$

(5b)

$$\frac{dρ_{ge}^j}{dt} = (-γ_{ge}^j + iΔ_p - iΔ_n)ρ_{ge}^j + iαg_{j}ρ_{eg}^j - iρ_{ge}^jΩ^*,$$

(5c)

$$\frac{dρ_{eg}^j}{dt} = \left( -\frac{Γ_i}{2} + iΔ_n \right) ρ_{eg}^j - iαg_{j}ρ_{ge}^j + Ω(ρ_{eg}^j - ρ_{ge}^j),$$

(5d)

$$\frac{dρ_{eg}^j}{dt} = Γ_1ρ_{eg}^j - iα^*g_{j}ρ_{ge}^j + iαg_{j}ρ_{eg}^j,$$

(5e)

$$\frac{dρ_{ge}^j}{dt} = Γ_2ρ_{ge}^j + iρ_{ge}^jΩ^* - iρ_{ge}^jΩ^*,$$

(5f)

$$\frac{dρ_{eg}^j}{dt} = -Γ_1ρ_{eg}^j + iα^*g_{j}ρ_{ge}^j - iαg_{j}ρ_{eg}^j - ρ_{ge}^jΩ + iρ_{ge}^jΩ^*,$$

(5g)

Here, $γ_{ge}^j$ is the decoherence rate for the coherence between the two ground states (|$g⟩, |g′⟩$). In steady state, $\frac{dα}{dt} = 0$ and $\frac{d\rho_{mn}}{dt} = 0$, ∀($m,n$), and Eqs. (5) become a set of linear equations which can be solved algebraically. Eliminating the atomic variables we get

$$-\eta - \alpha(κ_i - iΔ_p) = i \sum_{j=1}^{N} g_{j}ρ_{ge}^j$$

$$= iα \sum_{j=1}^{N} χ_j$$

$$= iα \frac{2g^2_{j}(Δ_n - Δ_p)}{2|Ω|^2 + (2Δ_p + iΓ_1)(Δ_n - Δ_p)},$$

(6)

where

$$χ_j = \frac{2g^2_{j}(Δ_n - Δ_p)}{2|Ω|^2 + (2Δ_p + iΓ_1)(Δ_n - Δ_p)}$$

(7)

is the linear susceptibility of the $j^{th}$ atom, $χ$ is the total linear susceptibility, and we make use of $\sum_{j=1}^{N} g^2_{j} = g^2_{0}N_c$ as we are interested in the average effect. In deriving the above equation, we have assumed that the intracavity light amplitude is very small compared to other relevant parameters, i.e., $g_{0}|α| \ll Ω, Γ_1$ and hence the susceptibility $χ$ shows linear dependence with the cavity field amplitude $α$ after neglecting the small nonlinear terms. We also assume that there is no decoherence between the two ground states, i.e., $γ_{ge} = 0$, which is valid for a dilute gas. The average photon number inside the cavity can then be written as

$$\bar{n} = |α|^2 = \frac{η^2}{(Δ_p - χ)^2 + (κ_i - χ)^2},$$

(8)

where

$$\chi_1 = \frac{4g^2_{0}N_c(Δ_n - Δ_p)(|Ω|^2 - Δ_p(Δ_n - Δ_p))}{Γ_i^2(Δ_n - Δ_p)^2 + 4(|Ω|^2 - Δ_p(Δ_n - Δ_p))^2},$$

and

$$\chi_2 = -\frac{2Γ_1g^2_{0}N_c(Δ_n - Δ_p)^2}{Γ_i^2Δ^2 + 4(|Ω|^2 + Δ^2)^2}.$$ 

In the limit of $Δ^2 \ll |Ω|^2, 4Γ^2/|Ω|^2$, i.e., small detuning of probe laser near the EIT peak, the intracavity photon number ($\bar{n}$) reduces to

$$\bar{n} = |α|^2 = \frac{η^2}{κ_i^2} \frac{d^2}{Δ^2 + d^2}.$$ 

This is a Lorentzian function with full width at half maxima (FWHM),

$$2d = \frac{2|Ω|^2κ_i}{\sqrt{Γ_i^2g^2_{0}N_c + (g^2_{0}N_c + |Ω|^2)^2}}.$$ 

For $|Ω|^2 \gg Γ_iκ_i$, the FWHM reduces to

$$2d = \frac{2κ_i}{(g^2_{0}N_c + 1)}.$$ 

(11)

This expression yields a simple dependence of the FWHM on the atom number. The greater the atom number, the lower the linewidth of the Lorentzian. Importantly, FWHM is linear in $N_c$ in contrast with the square-root dependence of VRS, thus making the EIT method of atom number detection more sensitive. Such a linewidth narrowing for cavity transmission was predicted earlier by Lukin et al. [51]. It was observed for thermal atoms coupled to a ring cavity [52], laser-cooled atoms coupled to a Fabry-Pérot cavity [53], and for laser-cooled ions coupled to a Fabry-Pérot cavity [54]. However, for a cavity-based EIT, the dependence of EIT linewidth on the number of atoms derived in this article has not been explored and requires experimental verification.

In the above analysis, the lambda system for the atoms was a closed system, and there was no leakage to any other dark state. However, this will not be the case for molecules due to the presence of a large number of ground-state levels. To check if Eq. (11) still holds for such a leaky system we solve the time-dependent partial differential Eq. (5) similar to Sec. III after
removing the $j$ subscript, and assuming the average number of atoms $N_c$ couple equally to the cavity. Here, in addition to the decays in Eq. (5), an additional decay $\Gamma_3$ to the dark ground state $|g'\rangle$ is also included. The total decay rate from the excited state becomes $\Gamma_t = \Gamma_1 + \Gamma_2 + \Gamma_3$ [see Fig. 3(b)] and all other parameters remain the same. An equivalence diagram between molecular levels and atomic levels is shown in Fig. 3.

For such a scenario, the EIT peaks obtained by performing a numerical integration for different atom numbers are shown in Fig. 4. The parameters differing from the numerical calculations of VRS (Fig. 2) are, the power of light input to the cavity $P_m = 40 \text{ pW}$, $\kappa_t/2\pi = 0.5$ MHz, and $\Omega = 10$ MHz. $|g'\rangle$ is the $\nu_c = 1, J_e = 0$ level of the $X^{1}\Sigma_g^+$ state, and $|g''\rangle$ represents all other $X^{1}\Sigma_g^+$ levels. Here, for $N_c = 1 \times 10^3$, 10% of the atoms will be lost to the dark ground state per scan, and for $N_c = 5 \times 10^4$, 0.06% of the atoms will be lost. For the analysis here, we have kept the scan duration at 1 ms because it takes more time to reach steady state for detection using EIT. Additionally, at the end of the scan, very few atoms go to the state $|g'\rangle$ as can be seen from Fig. 4(c).

This EIT detection is a significant improvement in terms of loss of molecules compared to the VRS detection. For the same $N_c$, the loss rate per scan is two orders of magnitude smaller, and hence detection of a smaller number of molecules is possible. For such an EIT-based detection scheme, having smaller $\kappa_t$ is better because the EIT window is usually small. For example, if we keep the output photon flux and other parameters the same, $\kappa_t = 2.5$ MHz will result in a loss of 80% in the case of $N_c = 1 \times 10^3$ and a 0.3% loss of atoms per scan in the case of $N_c = 5 \times 10^4$. Although small $\kappa_t$ is desirable in this case, we cannot lower it arbitrarily because the linewidth of the probe laser will also require narrowing. As in the case of detection using VRS, here too, the ratio $\kappa_{12}/\kappa_1$ should be on the higher side to avoid high intracavity photon number occupation.

EIT is accompanied by the phenomenon of slowing of group velocity of the probe light [35,55–57]. Hence, we expect an increase in trapping time of the photon inside the cavity as observed in previous experiments [53,58]. This can be exploited to detect the molecules placed inside the cavity. Below we explore a simple detection scheme, now in the time domain. The scheme is to set $\Delta_{pa} = \Delta_{pc} = \Delta_{ra} = 0$, to switch on the probe laser, to allow the system to reach the steady state [59], and obtain a constant light intensity output of the cavity. Following this the probe laser is suddenly switched off and the decay of the cavity output light is observed. The results of the numerical simulations are shown in Fig. 5.
MHz and $\kappa$ of cavity light decay for various atom numbers. (a) is indeed what is seen from the full numerical simulations be exponential with the decay rates, $2\pi/\kappa$ decay in the time domain; we expect the decay curves to 
event in the case of $\kappa$. However, very few photons will be collected during a single decay due to a finite dead time of an APD. However, multiple detection cycles can be added up. For example, the observation of one decay event in case of $\kappa$ MHz gives a maximum of 1 decay event in case of $\kappa$. In Fig. 5(a) for $\kappa = 0.05$ MHz, we get a maximum of 10 detections per observation interval if the total intervals are 20 for 10 detection cycles in the case of $\kappa$. In the presence of such fluctuations, higher value of $\kappa$ will be more advantageous due to large linewidths. However, higher value of $\kappa$ will require multiple interrogation of the ensemble, which may present a problem for some experiments. Experimentally the fluctuations can be minimized by using stabilized probe and control lasers which are locked to a high-finesse cavity [61]. Availability of such a locking scheme makes such a detection process feasible.

A Lorentzian in frequency space implies an exponential decay in the time domain; we expect the decay curves to be exponential with the decay rates, $2d$ of Eq. (11). This is indeed what is seen from the full numerical simulations in Fig. 5. For this detection scheme, a smaller value of $\kappa$ is very advantageous because the decay time and thus the observation time is longer, as seen in Fig. 5(b) for $\kappa = 0.05$ MHz. In Fig. 5(a) for $\kappa = 0.5$ MHz, the observation time is very short. Hence, very few photons will be collected during a single decay due to a finite dead time of an APD. However, multiple detection cycles can be performed, results of which can be added up. For example, the observation of one decay event in the case of $\kappa = 0.5$ MHz gives a maximum of 1 photon detection per 50 ns, so for 10 detection cycles, we get a maximum of 10 detections per observation interval if the total number of interval is 20 for 1-\(\mu\)s decay. Similarly, in the case of $\kappa = 0.05$ MHz, we get a maximum of 10 detections per observation interval if the total intervals are 20 for 10-\(\mu\)s decay [60] if a maximum of 1 photon is detected per 50 ns. Comparing the two cases with respect to the total measurement time and the total photon flux, 10 decay events in case of $\kappa = 0.5$ MHz and 1 decay event in case of $\kappa = 0.05$ MHz give the same statistics for detection.

For this detection scheme, the condition $\Delta_{pa} = \Delta_{rs}$ is always satisfied. Hence, we expect complete transparency and very little absorption of photons. For 10 detection cycles in the case of $\kappa = 0.5$ MHz, 0.09% of the atoms are lost for $N_c = 1 \times 10^3$, and 0.006% of the atoms are lost for $N_c = 5 \times 10^4$. In contrast, for single detection cycles in case of $\kappa = 0.05$ MHz, 0.008% atoms are lost for $N_c = 1 \times 10^3$ and 0.0006% of the atoms are lost for $N_c = 5 \times 10^4$. From the above numbers, we can say that the detection involving lower $\kappa$ is more efficient. However, less than one atom is lost during each detection cycle in each case so the detection scheme with $\kappa = 0.5$ MHz results in very tiny loss of molecules from the ground state relevant for detection and it is not required to make the detection scheme less lossy. Less loss for this detection scheme implies more flexibility for the ratio $\kappa_2/\kappa_1$. For example, if $\kappa = 0.5$ MHz, and if all other parameters are same as the ones for the case of VRS in Sec. III, a low ratio of $\kappa_2/\kappa_1 = 0.01$ will result in 6.5% loss of atoms in the case of $N_c = 1 \times 10^3$ and a 0.4% loss of atoms in the case of $N_c = 5 \times 10^4$ MHz atoms for the 10 decay events as opposed to 65% in the case of detection through VRS.

V. DISCUSSION

If we consider loss of molecules from the level used for detection as the measure for the efficiency, the detection of molecules through delay in decay times mentioned above is seen to be better than other detection schemes explored in this article. However, the condition $\Delta_{pa} = \Delta_{rs} = 0$ should always be satisfied, and any fluctuations around this condition will hinder the detection as the EIT effect is very sensitive near this condition, as it can be seen from narrow linewidths of Fig. 4. In the presence of such fluctuations, higher value of $\kappa$ will be more advantageous due to large linewidths. However, higher value of $\kappa$ will require multiple interrogation of the ensemble, which may present a problem for some experiments. Experimentally the fluctuations can be minimized by using stabilized probe and control lasers which are locked to a high-finesse cavity [61]. Availability of such a locking scheme makes such a detection process feasible.

In the above analysis, we have ignored the hyperfine structure of the molecules. For some molecular states the hyperfine splitting will make the above calculations more complicated. But for the singlet state the hyperfine splitting at zero magnetic field is typically of the order of a few tens of kHz for alkali-metal diatomics [62–66]. This is smaller than the cavity and atomic linewidths explored in this article and therefore should not create complications. This is supported experimentally by the observation of the phenomena explored here, with atoms [52]. For atoms the magnetic sublevels of the hyperfine levels are nearly degenerate and behave as a single level for the observation of VRS and EIT phenomenon. The other extreme where the hyperfine splitting is resolvable is also not a problem as this will just add more loss channels without affecting the detection process. In addition, for singlet molecular states the hyperfine splitting decreases when a high electric field is applied [62,65]. This can be exploited for species where the hyperfine structure creates a problem.

VI. CONCLUSION

In this article, we explored dispersion based nondestructive techniques with the help of numerical simulations and theoretical analysis for detecting molecules using a cavity. The advantage of the method results from the collective strong coupling of molecules to the cavity, which allows the molecules to
be probed with a small laser intensity and under off-resonance conditions, thus keeping losses low. It is clear from the analysis that the large number of decay channels for molecules does not preclude cavity detection of molecules. Both VRS and EIT-based arrangements were analyzed and the parameters for molecule detection using each of these phenomena identified. The detection of molecules using the EIT feature is not just feasible, but also very efficient. With care the technique can detect a very small number of molecules. These techniques will be useful for not just detection of molecules but can be used to detect atoms/ions with multiple levels. A consequence of this is that the need for a repumping laser is mitigated. Hence, an extended to molecules. The analysis done here enables highly efficient exploration of cold, dilute molecular gases.

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[60] There are few recent works which do use such a repumping scheme for the purpose of laser cooling of specific molecules [15,16]. However, after scattering of a few thousand photons the molecules are lost for further laser cooling as levels not accessible any more by the laser are populated.

