Magic Conditions for Multiple Rotational States of Bialkali Molecules in Optical Lattices

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We investigate magic-wavelength trapping of ultracold bialkali molecules in the vicinity of weak optical transitions from the vibrational ground state of the \(X^1\Sigma^+\) potential to low-lying rovibrational states of the \(b^3\Pi_0\) potential, focusing our discussion on the \(^{87}\)Rb\(^{133}\)Cs molecule in a magnetic field of \(B = 181\) G. We show that a frequency window exists between two nearest neighbor vibrational poles in the dynamic polarizability where the trapping potential is “near magic” for multiple rotational states simultaneously. We show that the addition of a modest DC electric field of \(E = 0.13\) kV/cm leads to an exact magic-wavelength trap for the lowest three rotational states at a angular-frequency detuning of \(\Delta \nu = 2\pi \times 218.22\) GHz from the \(X^1\Sigma^+(v = 0, J = 0) \rightarrow b^3\Pi_0(v' = 0, J = 1)\) transition. We derive a set of analytical criteria that must be fulfilled to ensure the existence of such magic frequency windows and present an analytic expression for the position of the frequency window in terms of a set of experimentally measurable parameters. These results should inform future experiments requiring long coherence times on multiple rotational transitions in ultracold polar molecules.

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I. INTRODUCTION

Ultracold polar molecules present a wealth of opportunities in quantum science and technology [1]. Proposed applications span the fields of precision measurement and metrology [2–5], quantum-state resolved chemistry [9–13], dipolar quantum matter [14–18], quantum simulation [20–25] and quantum information processing [26–32]. Recent experimental progress on the production of ultracold molecules by association [33–40] and direct laser cooling [41–49] has brought many of these applications within reach.

In the realm of quantum simulation and computation, the rotational structure of ultracold molecules provides a rich basis of long-lived states in which to encode pseudo-spins or quantum information. Owing to the permanent molecular-frame electric dipole moment, the rotational states can be conveniently manipulated with microwave fields, as already demonstrated in a number of settings [50–54]. Moreover, laboratory-frame dipole moments can be engineered using applied electric fields or superpositions of rotational states. The resulting long-range interaction between molecules can be exploited to realise model Hamiltonians in quantum magnetism [20–22, 24–26, 63–65] and two-qubit gates for quantum information processing [26–32]. To generate useful interaction strengths necessitates inter-molecular distances below a micrometre. This is most readily achieved using optical potentials, either in the form of an optical lattice [59–60] or an array of optical tweezers [61–62].

For diatomic molecules, such as ground-state bialkali molecules [33–35], the dynamic polarizability along the molecular axis (\(\alpha_\parallel\)) is, in general, different from that perpendicular to it (\(\alpha_\perp\)). For light polarized at an angle \(\theta\) to the molecular axis, this leads to a dynamic polarizability in the body-fixed frame given by

\[
\alpha(\theta) = \alpha^{(0)} + \alpha^{(2)} P_2(\cos(\theta)),
\]

where \(\alpha^{(0)} = \frac{1}{3}(\alpha_\parallel + 2\alpha_\perp)\) and \(\alpha^{(2)} = \frac{2}{3}(\alpha_\parallel - \alpha_\perp)\) are the isotropic and anisotropic components of the polarizability tensor, respectively. \(\alpha_\parallel\) and \(\alpha_\perp\) result from a sum over all allowed molecular transitions for the component of the dipole operator parallel or perpendicular to the molecular axis, respectively, and are smooth functions of wavelength in the regime where the frequency of the trapping laser is far-detuned from any rovibrionic transitions [66–69]. In the lab frame, the dynamic polarizability can be thought of as the spatial average of \(\alpha(\theta)\). Although \(\alpha^{(0)}\) is the same for all rotational states, \(\alpha^{(2)}\) strongly mixes states with different rotational projections in excited rotational states. It follows that for molecules confined in an optical potential, the anisotropic polarizability leads to rotational transition frequencies that are strongly dependent on the intensity and polarization of the trapping light. The concomitant state-dependent light shifts make it highly challenging to achieve rotational coherence times that are sufficiently long to be sensitive to the \(\sim\)kHz interaction strengths [51–55] typical of most molecules. Nevertheless, several approaches have been developed to match the polarizabilities of two specific states within a molecule. These include judicious choice of the intensity and polarisation of the trapping light [67–69] and the addition of applied electric fields to simplify the couplings within the molecule [55–56, 70].

Inspired by the magic-wavelength traps used in atomic clocks [71, 72], it is natural to investigate magic-wavelength trapping for molecules. Intuitively, magic trapping independent of the molecular rotational state

\[
E_{\parallel} = \frac{1}{2} m (v + 1)^2 \left(1 + \frac{1}{3} + \frac{1}{3} + \frac{1}{3} \right) = 218.22 \text{ GHz}
\]
can be realized under the condition of \( \alpha^{(2)} = 0 \). To search for this condition, one needs to tune the trapping laser wavelength into a regime where there is significant interplay between several ro-vibrational poles in \( \alpha_0 \) and \( \alpha_1 \). Indeed, following this approach, very recent work has demonstrated state-insensitive trapping for two vibrational [73] or rotational [74] levels. These magic-frequency traps show reduced sensitivity to experimental parameters, enabling longer coherence times to be achieved. However, numerous proposed applications make greater use of the rich internal structure of molecules by simultaneously addressing multiple rotational states simultaneously.

In this work, we investigate magic-wavelength trapping of ultracold bialkali molecules in the vicinity of weak optical transitions from the vibrational ground state of the \( X^1 \Sigma^+ \) potential to low-lying rovibrational states of the \( b^3 \Pi_0 \) potential, focussing our discussion on the \( ^{87} \text{Rb}^{133}\text{Cs} \) molecule. We show that a magic trapping frequency window for multiple rotational states of the \( X^1 \Sigma^+ \) potential exists between two nearest neighbor vibrational poles of the \( b^3 \Pi_0 \) potential, far away from any rotational poles. Within this window, the laser trapping is “near magic” for multiple rotational states simultaneously and is exactly magic for pairs of neighboring rotational states at specific laser frequencies. Moreover, the “near magic” frequency window can be tuned to a true magic frequency for the lowest three rotational states by applying an experimentally accessible DC electric field. This true triple magic condition is expected to be useful for future studies of synthetic spin-1 systems using ultracold molecules. The existence of such a magic frequency window relies on a set of strict criteria which we derive analytically. We show that these criteria can be satisfied near the narrow \( X^1 \Sigma^+ \to b^3 \Pi_0 \) transitions for heavy molecules, including \( ^{87} \text{Rb}^{133}\text{Cs} \) and \( ^{23} \text{Na}^{87}\text{Rb} \). We also derive an analytic expression for the position of the frequency window in terms of a set of experimentally measurable parameters, such as transition widths and transition wavelengths. This will provide a straightforward, self-consistent approach to search for the magic trapping frequency window in future experiments.

This paper is organized as follows. Section II presents the general theoretical framework describing the molecular rotational states in the lowest vibrational state of the ground electronic potential in the presence of applied magnetic, electric and optical fields. In section III, we discuss the hyperfine structure of the \( ^{87} \text{Rb}^{133}\text{Cs} \) molecule in the presence of applied magnetic and electric fields with a view to identifying the best target states in each rotational level for magic trapping. In section IV, we consider the AC-Stark shift and dynamic polarizability of \( ^{87} \text{Rb}^{133}\text{Cs} \) molecules in the vicinity of the weakly allowed \( X^1 \Sigma^+ \to b^3 \Pi_0 \) transitions. In section V, we identify magic trapping frequencies by searching for crossings among the frequency-dependent dynamic polarizability curves of different rotational states. We present a simple analytic treatment that shows excellent agreement with our numerical results, both near-resonance and in the magic frequency window between two vibrational poles. Imaginary polarizabilities for rotational states in the magic frequency window are also calculated. In section VI, we discuss the wider significance of our work, before concluding in section VII.

II. THEORETICAL FRAMEWORK

We focus on the molecular rotational states \( J \) associated with the \( v = 0 \) vibrational state of the ground electronic state of \( \text{RbCs} \). The effective Hamiltonian that describes the system in the presence of a static magnetic field \( \vec{B} \), a static electric field \( \vec{E} \), and an optical laser field of intensity \( I \) [52, 65, 76] is given by:

\[
H = H_{\text{rot}} + H_Z + H_{\text{hf}} + H_{\text{DC}} + H_{\text{AC}},
\]

where the rotational Hamiltonian is

\[
H_{\text{rot}} = B_\alpha \vec{J}^2,
\]

the Zeeman Hamiltonian is

\[
H_Z = -g_\mu_N \vec{J} \cdot \vec{B} - \sum_{k=1}^2 g_k \mu_N \vec{I}_k \cdot \vec{B}(1 - \sigma_k),
\]

the nuclear quadrupole interaction is

\[
H_{\text{hf}} = \sum_{k=1}^2 \frac{(eqQ)_k}{I_k(I_k-1)} C_2(\alpha, \beta) T_2(\vec{I}_k, \vec{I}_k),
\]

and the DC-Stark shift is

\[
H_{\text{DC}} = -\vec{d} \cdot \vec{E}.
\]

In Eqs [3]-[6], \( \vec{J}, \vec{I}_k \), and \( \vec{d} \) denote the molecule orbital angular momentum operator, the nuclear spin operators for the \( k \)-th atom, and the permanent molecular electric dipole moment operator, respectively. The nuclear quadrupole interaction \( H_{\text{hf}} \) couples the nuclear spin to rotational states and depends on the quadrupole coupling constants \( (eqQ)_k \) for \( \text{Rb} \) and \( \text{Cs} \) obtained from Refs [52]. The operator \( T_2(\vec{I}_k, \vec{I}_k) \) is a rank-2 tensor and \( C_2(\alpha, \beta) = \sqrt{4\pi/5}Y_{20}(\alpha, \beta) \) is the modified spherical harmonic function, where the angles \( \alpha, \beta \) describe the orientation of the diatomic molecule in the space-fixed coordinate frame. In these equations, \( B_\alpha \) is the rotational constant, \( \mu_N \) is the nuclear magneton, and \( g_\nu \) is the molecule rotational \( g \)-factor. Moreover, \( g_k \) and \( \sigma_k \) with \( k = 1, 2 \) are nuclear-spin \( g \)-factors and isotropic molecular nuclear shielding factors, respectively.
Here, the direction of the external magnetic field is our quantization axis along which we define projection quantum numbers of angular momenta. The matrix elements of the Hamiltonian are determined in low-energy set of basis functions $|J, M; m_1, m_2\rangle$, where $J$ and $M$ are the orbital angular momentum and its associated projection, respectively. Quantum numbers $m_k$ are nuclear spin projections of the $k$-th atom.

The AC-Stark Hamiltonian $H_{AC}$ in Eq. (2) is constructed up to second order in the electric field strength of the driving laser in the regime where the AC-Stark shift is much smaller than the rotational constant. In this regime, the AC-Stark Hamiltonian $H_{AC}$ is

$$H_{AC} = -\frac{I}{\epsilon_0 c} \sum_{J, M, m_1, m_2} |J, M; m_1, m_2\rangle \langle J, M; m_1, m_2|$$

$$\times \sum_{J} \frac{(J, M'|\vec{d}_{\text{tr}} \cdot \vec{\epsilon}|f)(f|\vec{d}_{\text{tr}} \cdot \vec{\epsilon}|J, M)}{E_{J} - (E_{J} + \hbar \omega)} , \tag{7}$$

where energies $E_J$ are the eigenvalues of $H_{\text{rot}}$, $\vec{d}_{\text{tr}}$, $\vec{\epsilon}$, and $\omega$ are the molecular transition electric dipole moment operator, the laser polarization, and the laser angular frequency, respectively. The summations over $J$, $M$, $M'$, and $m_k$ only contain basis functions in the low-energy space. The summation $f$ in Eq. (7) is over all ro-vibrational states and continua of excited electronic states with energies $E_f$ excluding their Zeeman, hyperfine, and DC-Stark shifts. We have included previously studied excited electronic states that dissociate to limits where only one of Rb or Cs is excited to its energetically-lowest excited $nP$ state. In this work, we are interested in the regime where the AC-Stark shift is much smaller than the rotational constant. Thus, in writing Eq. (7), couplings between the states with different orbital angular momenta $J$ are neglected. Finally, $\epsilon_0$, $c$, and $\hbar$ are the vacuum permittivity, the speed of light in vacuum, and the reduced Planck’s constant, respectively.

We diagonalize Eq. (2) in the basis $|J, M; m_1, m_2\rangle$, including $J \leq 20$ to find eigenenergies $E_i$ and corresponding eigenstates $|i\rangle$ of the molecular system. The dynamic polarizability of an eigenstate is $-\partial E_i/\partial I$. By mapping out the intensity-dependence of the eigenenergies of the effective low-energy Hamiltonian, we obtain the dynamic polarizabilities for various rotational states. The electric field, magnetic field, and laser frequency serve as our tuning parameters which can be manipulated, as shown in the following discussions, to realize various magic trapping conditions. Although, in this work we focus our discussion on the $^{87}$Rb,$^{133}$Cs molecule, the extension to other diatomic alkali molecules is implied.

III. ZEEMAN SPLITTINGS AND DC-STARK SHIFTS IN RBCS MOLECULES

The nuclear spins of $^{87}$Rb and $^{133}$Cs atoms are $I_1 = 3/2$ and $I_2 = 7/2$, respectively. Because of the multiple combinations of the atomic nuclear spin projections and the molecular orbital angular momentum projections, there exist $(2J + 1)(2I_1 + 1)(2I_2 + 1)$ energy levels that are associated with the rotational state with orbital angular momentum $J$. In the presence of the magnetic field, the static electric field, and the hyperfine interactions, these “near” degenerate energy levels split. Before we discuss the magic trapping conditions, it is necessary to select the best target states to be trapped among these levels for each rotational state.

The left column of Fig. 1 shows the magnetic field strength dependence of the rotational energy manifold $E_J$ ($J = 0, 1, 2$) with vanishing static electric field. In the weak magnetic field regime, the splitting between the levels of the same energy manifold are dominated by the hyperfine interactions. In this regime, the total angular momentum $F^2 = (\vec{J} + \vec{I}_1 + \vec{I}_2)^2$ and the total

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**FIG. 1:** The hyperfine energy levels for the $J = 0$, 1, and 2 manifolds as functions of the magnetic field strength $B$ (the left column (a), (c), and (e) panels, respectively) and the static electric field strength $E$ applied parallel to a magnetic field of $B = 181$ G (the right column (b), (d), and (f) panels, respectively). The red dashed lines in (a), (c), and (e) mark the target trapping state (see text). Panel (b) consists of a band with 32 energy levels. Panel (d) consists of two bands; the upper one contains 32 energy levels with $M = 0$ and the lower one 64 energy levels with $M = \pm 1$. Panel (f) consists of three bands; the upper one contains 32 energy levels with $M = 0$, the middle one 64 energy levels with $M = \pm 1$, and the lower one 64 energy levels with $M = \pm 2$. Then, we discuss the magic trapping conditions, it is necessary to select the best target states to be trapped among these levels for each rotational state.
projection $M_F = M + m_1 + m_2$ are approximately good quantum numbers which means that the eigenstates consist of strong admixture of states with different nuclear spin projections. The level repulsion is strong in this regime, leading to quadratic Zeeman shifts dominating over linear Zeeman shifts for $B < 50$ G in the left column of Fig. 1. With increasing magnetic field strength, the linear Zeeman shift dominates. Due to the differences in the various $g$-factors, $g_r = 0.0062$, $g_1 = 1.836(3)$, and $g_2 = 0.738(1)$ in Eq. (4) for $^{87}$Rb$^{133}$Cs [72, 77], the projections $M$, $m_1$, and $m_2$ are all approximately good quantum numbers in the high-field regime. Thus, the eigenstates have significantly reduced admixture. The levels marked by the red dashed lines in the left column of Fig. 1 correspond to the states containing more than 50% occupation in the $|J, M = 0; m_1 = 3/2, m_2 = 7/2\rangle$ component. Note this corresponds to the spin-stretched state in $J = 0$ and is the initial state created in experiments [36, 37]. We select these states as our target trapping states. For $B > 150$ G, the admixture of the other components into the target trapping states is less than 20% for $J = 0, 1$, and 2. The red dashed lines in Fig. 1 (c) and Fig. 1 (e) terminate around $B = 25$ G and $B = 100$ G, respectively, because no target state can be identified in the small $B$ regime due to strong admixture.

In this paper, we focus on a magnetic field strength of $B = 181$ G according to the experimental work [37]. To further suppress the admixture of other components into our target trapping states, we take advantage of an applied static electric field. Due to the small magnitude of the rotational $g$-factor $g_r$, the states with different orbital angular momentum projections $M$ and the same nuclear spin projections are close in the spectrum. For example, the energy splitting between states with a unit difference in the orbital angular momentum projection $M$ is $\sim 10$ kHz for $B = 181$ G. A static electric field along the magnetic field direction separates the levels with different absolute values of $M$ in the spectrum.

The right column of Fig. 1 shows the dependence of the rotational energy manifold $E_J (J = 1, 2, 3)$ on the electric field strength in the presence of a parallel magnetic field of $B = 181$ G. With increasing $E$, the energies of the $J = 0$ manifold decrease quadratically [see Fig. 1 (b)] due to the second-order level repulsion with the states $|J = 1, M = 0; m_1, m_2\rangle$. It turns out that the energies of the states $|J = 1, M = 0; m_1, m_2\rangle$ are pushed up. Due to the level repulsion between the states $|J = 1, M = 0; m_1, m_2\rangle$ and $|J = 2, M = 0; m_1, m_2\rangle$, the states with $M = 0$ in the $J = 1$ manifold are pushed down with increasing $E$. Thus, the static electric field separates the $J = 1$ rotational energy manifold into two bands, the upper one with $M = 0$ and the lower one with $M = \pm 1$ [see Fig. 1 (d)]. Similarly, for the $J = 2$ manifold, a three-band structure is seen with the upper, the middle, and the lower one corresponding to $M = 0$, $M = \pm 1$, and $M = \pm 2$, respectively. For $B = 181$ G, a static electric field of strength $E = 0.1$ kV/cm already makes the admixture of the states with finite $M$ into the state with $M = 0$ negligible.

IV. AC-STARK SHIFTS NEAR THE NARROW $X^1\Sigma^+ \rightarrow b^3\Pi_0$ TRANSITIONS

To study the AC-Stark shift of the $^{87}$Rb$^{133}$Cs molecule, we consider the application of a driving laser field with the angular frequency $\omega$ to induce coupling between the target trapping states and electronically excited states. Figure 2 shows the selected relativistic adiabatic potential $\Omega = 0^+$ curves of the $^{87}$Rb$^{133}$Cs molecule, where $\Omega$ is the total projection quantum number of the electronic angular momentum and nuclear spins along the diatomic molecule axis. The $b^3\Pi_0$ potential and the $A^1\Sigma^+$ potential are coupled by the spin-orbit coupling terms which lead to an avoided crossing near $R_c = 10a_0$. Here, the potentials and the spin-orbit coupling functions are generated based on the data in Refs. [64, 78, 80]. Due to the spin-orbit coupling, the few lowest bound states lying near the bottom of the $b^3\Pi_0$ potential have some
FIG. 3: Microwave transition frequencies from the $|J = 0, M = 0; m_1 = 3/2, m_2 = 7/2\rangle$ ground state to the $|J = 1\rangle$ manifold as a function of the laser intensity for a laser frequency near the resonance transition to the $\Sigma^+$ states. The panels (a) and (c) correspond to vanishing static electric field. Panels (b) and (d) correspond to a static electric field of $E = 2$ kV/cm applied in the $z$-direction. The red circles in all panels mark the energy level of the target trapping state.

We start the discussion with the dynamic polarizabilities near these narrow transitions, indicated by the blue dashed line in Fig. 2. We denote $\omega_{\nu'}$ the resonance transition frequency from the $(v = 0, J = 0)$ state of the $X^1\Sigma^+$ potential to the $(\nu' = 1, J = 1)$ state of the $b^3\Pi_0$ potential. For $\nu' = 0$, the resonance frequency reads $\omega_0 = 2\pi \times 261.533$ THz which corresponds to a wavelength of $1146.287$ nm. When the driving laser frequency $\omega$ is close to the resonance frequency $\omega_{\nu'}$, we reference $\omega$ to $\omega_{\nu'}$ through the detuning $\Delta_{\nu'} = \omega - \omega_{\nu'}$.

Figure 3 shows the impact of the static electric field on the AC-Stark shifts of the microwave transition frequencies from the $|J = 0, M = 0; m_1 = 3/2, m_2 = 7/2\rangle$ ground state to the $|J = 1\rangle$ rotational state of the $b^3\Pi_0$ potential in the small and large detuning regimes. The driving laser is linearly polarized with a polarization parallel to the magnetic field. The red circles correspond to the target trapping state as discussed in Sec. III. For the case with the detuning of $\Delta_{\nu'} = 2\pi \times 3$ GHz and vanishing static electric field [Fig. 3(a)], the AC-Stark shifts can be characterized into two bands; one going up with increasing laser intensity while the other staying almost independent of the laser intensity. The former corresponds to states with $M = 0$ while the latter to states with $M = \pm 1$. As shown by the red circles in Fig. 3(a), the energy level of the target trapping state in the $|J = 1\rangle$ manifold crosses those of the other levels with increasing laser intensity. These crossings lead to strong level interactions [see the gap in the red circles near $I = 0.1$ kW/cm$^2$ in Fig. 3(a)], hence to large hyper-polarizabilities which makes the system unstable with respect to fluctuations of the trapping laser intensity.

The level-crossing behavior in the AC-Stark shift can be avoided by separating the $M = 0$ band and the $M = \pm 1$ band using a static electric field as discussed in Sec. III. Figure 3(c) shows the AC-Stark shifts in the presence of a static electric field of $E = 0.2$ kV/cm. Compared to Fig. 3(a), the $M = 0$ band lies roughly $5$ MHz above the $M = \pm 1$ band for $I = 0$. With increasing laser intensity, the energy gap between the $M = 0$ band and the $M = \pm 1$ band keeps increasing. The energy of the target trapping state does not cross any of the $M = \pm 1$ states any more.

The level crossings seen in Fig. 3(a) result from the fact that the AC-Stark shift of the target trapping state is larger than the energy splitting between the nearest neighbor hyperfine levels. With larger laser detuning, the differential AC-Stark shift is greatly reduced. For example, for a detuning of $\Delta_{\nu'} = 2\pi \times 200$ GHz as shown in Fig. 3(b), the level crossings between the target trapping state and the other states in the $|J = 1\rangle$ manifold disappear for the laser intensity regime shown here. A finite static electric field still separates the $M = 0$ band from the $M = \pm 1$ band as shown in Fig. 3(d), which does make the system more robust, but is not necessary in this case.

In the following discussion of dynamic polarizabilities, we describe the detuning as near-resonance when $\Delta_{\nu'} < 2\pi \times 10$ GHz and as medium-detuned otherwise. According to the above discussion, the static electric field is always turned on for the near-resonance cases and not mandatory for the far-detuned cases. This setup makes our results independent of the laser intensity in a broad intensity regime for both cases.

V. MAGIC CONDITIONS FOR MULTIPLE ROTATIONAL STATES

We may identify magic trapping frequencies by searching for crossings among the frequency-dependent dynamic polarizability curves of different rotational states. We start the discussion with the dynamic polarizabilities $\alpha_{\perp,\parallel}$ near the resonance from which we extract the parallel and perpendicular background polarizabilities $\alpha_{b,\parallel}$ and $\alpha_{b,\perp}$ and the transition width $\Gamma_{0,\nu'}$. Given the values of $\alpha_{b,\parallel}$, $\alpha_{b,\perp}$, and $\Gamma_{0,\nu'}$, it is proved analytically and verified by our numerical calculations that there exists a "near" magic frequency window for multiple rotational
FIG. 4: The dynamic polarizabilities near the resonance transition to the $v = 0$ vibrational state of the $b^3Π_0$ potential. A magnetic field of strength $B = 181$ G and a static electric field of strength $E = 0.2$ kV/cm are applied in the $z$-direction. The driving laser polarization is (a) parallel and (b) perpendicular to the external static fields. The black circles and red squares correspond to the numerical results of the dynamic polarizabilities of the $J = 0$ and $J = 1$ target trapping state. The black solid lines and the red solid lines correspond to the analytical results generated using the Eqs. (8) and (9). The green upper triangle in Panel (b) marks the crossing between the black circles and red squares.

states in the medium-detuned regime between vibrational poles. By tuning the static electric field, a true triple magic frequency is found for the $J = 0, J = 1$, and $J = 2$ target trapping states for the $87$Rb$^{133}$Cs molecule.

A. Near-Resonance Dynamic Polarizabilities

In the near-resonance regime, we fix the strength of the static electric field to be $E = 0.2$ kV/cm. The angle between the laser polarization and the magnetic field is denoted $\theta$. In this case, the dynamic polarizabilities $\alpha_{J=0}$ of the $J = 0, M = 0$ target trapping state and $\alpha_{J=1}$ of the $J = 1, M = 0$ target trapping state can be approximated using (67) by

$$\alpha_{J=0} = -\frac{3\pi c^2}{2\omega_{v'}} \frac{\Gamma_{0,v'}}{3\Delta_{v'}} + 1 3 \alpha_{bg,\|} + \frac{2}{3} \alpha_{bg,\perp}, \tag{8}$$

and

$$\alpha_{J=1} = -\frac{3\pi c^2}{2\omega_{v'}^2} \left[ \cos^2(\theta) \frac{\Gamma_{0,v'}}{3 \Delta_{v'} + 2B_v + 2B_{v'}} + \frac{3 + \cos^2(\theta)}{15} \frac{\Gamma_{0,v'}}{\Delta_{v'} + 2B_v - 4B_{v'}} \right] + \frac{2\cos^2(\theta) + 1}{5} \alpha_{bg,\|} + \frac{4 - 2\cos^2(\theta)}{5} \alpha_{bg,\perp}, \tag{9}$$

respectively. Here, the parameters $B_v$ and $B_{v'}$ correspond to the rotational constants for the $v = 0$ vibrational state of the $X^1Σ^+$ potential and the $v' = 0$ vibrational state of the $b^3Π_0$ potential. The transition width $\Gamma_{0,v'}$ can be calculated via

$$\Gamma_{0,v'} = \frac{\omega_{v'}^3}{3\pi\epsilon_0\epsilon_r} |\mu_{0,v'}|^2 \tag{10}$$

where the $\mu_{0,v'}$ is the transition dipole momentum between the $v = 0$ vibrational state of the $X^1Σ^+$ potential and the $v'$ vibrational state of the $b^3Π_0$ potential. The parallel and perpendicular background polarizabilities $\alpha_{bg,\|}$ and $\alpha_{bg,\perp}$ contain the contributions from all the far-detuned rovibronic states with $\Omega = 0$ and $\Omega = 1$, respectively [63–65]. For $87$Rb$^{133}$Cs, we find $B_v = 2\pi \times 0.490$ GHz, $B_{v'} = 2\pi \times 0.510$ GHz, $\Gamma_{0,v'=0} = 2\pi \times 15.5$ kHz, $\alpha_{bg,\|} = h \times 0.127$ kHz/(W/cm$^2$), and $\alpha_{bg,\perp} = h \times 0.0340$ kHz/(W/cm$^2$). Experimentally, these values can be extracted by fitting the measured dynamic polarizability curves near the poles.

Figure 3 shows the dynamic polarizabilities for laser polarizations parallel and perpendicular to the magnetic field direction in the near-resonance regime. The symbols correspond to the numerical results and the lines show the analytical results generated using Eqs. (8) and (9). The agreement in both cases is excellent. As can be seen, there is no crossing between the $\alpha_{J=0}$ curve and the $\alpha_{J=1}$ curve in the near-resonance regime for $\theta = 0°$. According to Eq. (8), the dynamic polarizability $\alpha_{J=1}$ can be tuned by varying the polarization direction of the driving laser. For example, for $\theta = 90°$, the term in the first row of Eq. (8) inside the square bracket vanishes and the pole structure at $\Delta_{v'} = -2\pi \times 2.00$ GHz is missing, as shown by the red squares in Fig. 3 (b). In addition, the pole at $\Delta_{v'} = 2\pi \times 1.06$ GHz is slightly narrower compared to the $\theta = 0°$ case. In this case, the $\alpha_{J=1}$ curve crosses the $\alpha_{J=0}$ curve at the magic detuning of $2\pi \times 6.28$ GHz, as shown by the green upper triangle in Fig. 3 (b). The value of the polarizability at the magic detuning is $-h \times 2.71$ kHz/(W/cm$^2$). The negative polarizability indicates that the molecules can be trapped at the nodal point of an optical lattice where the laser intensity is the local minimum. This trapping condition is beneficial for also minimizing heating and loss from incoherent photon scattering.

B. Multiple Magic Frequency Window

For arbitrary $J$, we derive the general formula for the dynamic polarizability near the resonance transition to one of the states of the $b^3Π_0$ potential,

$$\alpha_J = \frac{3\pi c^2}{2\omega_{v'}^2} \left[ A_J(\theta) \frac{\Gamma_{0,v'}}{\Delta_{v'} + L_J} + B_J(\theta) \frac{\Gamma_{0,v'}}{\Delta_{v'} + R_J} \right] + \left[ A_J(\theta) + B_J(\theta) \right] \alpha_{bg,\|} + \left[ 1 - A_J(\theta) - B_J(\theta) \right] \alpha_{bg,\perp}, \tag{11}$$
where the pole positions $L_J$ of the left branch and $R_J$ of the right branch read

$$L_J = J(J+1)B_v - [J(J-1) - 2]B_{v'},$$  \hspace{1cm} (12)$$

and

$$R_J = J(J+1)B_v - [(J+1)(J+2) - 2]B_{v'},$$  \hspace{1cm} (13)$$

respectively. The angular factors $A_J(\theta)$ and $B_J(\theta)$ in Eq. (11) are,

$$A_J(\theta) = \begin{cases} 
\frac{(J+1)(J-1)}{2(2J+1)(2J-1)} + \frac{J^2 + 1}{2(2J+1)(2J-1)} \cos^2(\theta), & J > 0 \\
0, & J = 0,
\end{cases}$$  \hspace{1cm} (14)$$

and

$$B_J(\theta) = \frac{(J+2)(J+1)}{2(2J+3)(2J+1)} + \frac{J(J+1)}{2(2J+3)(2J+1)} \cos^2(\theta).$$  \hspace{1cm} (15)$$

By Taylor-expanding the right hand side of Eq. (11) with respect to $L_J$ and $R_J$, we obtain,

$$\alpha_J = [A_J(\theta) + B_J(\theta)] \left( -\frac{3\pi c^2}{2\omega^2} \Gamma_{0,v'} \Delta_{v'} + \alpha_{bg,\|} - \alpha_{bg,\perp} \right) + \alpha_{bg,\perp} + T_J(\Delta_{v',\theta}),$$  \hspace{1cm} (16)$$

where the remaining term $T_J(\Delta_{v',\theta})$ reads,

$$T_J(\Delta_{v',\theta}) = \frac{3\pi c^2}{2\omega^2} \Gamma_{0,v'} \left[ A_J(\theta)L_J + B_J(\theta)R_J \right] + \mathcal{O} \left( \frac{\Gamma_{0,v'}^2 L_J^2}{\Delta_{v'}^3} \right) + \mathcal{O} \left( \frac{\Gamma_{0,v'}^2 R_J^2}{\Delta_{v'}^3} \right).$$  \hspace{1cm} (17)$$

Based on Eq. (16), we can always find a detuning $\Delta_{v',cr}$ such that,

$$\alpha_J = \alpha_{bg,\perp} + T_J(\Delta_{v',cr},\theta),$$  \hspace{1cm} (18)$$

where,

$$\Delta_{v',cr} = \frac{3\pi c^2}{2\omega^2} \frac{\Gamma_{0,v'}}{\alpha_{bg,\|} - \alpha_{bg,\perp}}.$$  \hspace{1cm} (19)$$

For the transitions with $\Delta_{v',cr}$ lying in the medium-detuned regime, i.e., $|\Delta_{v',cr}| \gg |L_J|$, $|\Delta_{v',cr}| \gg |R_J|$, and $|\Delta_{v',cr}| \gg \Gamma_{0,v'}$, the remaining term $T_J(\Delta_{v',cr},\theta)$ can be neglected. In this case, both the $\theta$-dependence and the $J$-dependence of $\alpha_J$ in Eq. (18) disappear, indicating that the frequency-dependent dynamic polarizabilities of all rotational states pass through the same fixed point; the trap is magic for all rotational states at this laser detuning. The multiple magic frequency is approximately given by Eq. (19) and the value of the dynamic polarizability is approximately equal to the background perpendicular dynamic polarizability $\alpha_{bg,\perp}$.

Figure 5(a) shows the triple crossing magic frequency for $\alpha_J$ with $J = 0, 1,$ and 2 near the resonance transition to the $v' = 0$ state of the $b^3Π_0$ potential. A magnetic field $B = 181$G is applied in the $z$-direction. The laser field is parallel to the magnetic field. The circles mark the crossings between different curves in (b) and (c). The static electric field is vanishing in (a) and (b). A finite static electric field of $E = 0.13$ kV/cm is applied along the $z$-direction in (c). A near triple magic condition exists in (b) and a true triple magic condition exists in (c).
this detuning is \( \alpha_J = h \times 0.03392 \text{kHz/(W/cm}^2) \).

Our theory also predicts that the triple magic frequency window also holds for higher rotational states. Figure 6 shows the \( \alpha_J \) curves up to \( J = 4 \) for the parallel driving case in the presence of the static electric field of strength \( E = 0.13 \text{ kV/cm} \). It can be seen that all the values of \( \alpha_J \) are very close to \( \alpha_{bg,\perp} \) in the same magic frequency window as discussed before. A further zoom-in of the magic frequency window, shown in the inset of Fig. 6, indicates that \( \alpha_{J = 3} \) and \( \alpha_{J = 4} \) almost run parallel to \( \alpha_{J = 2} \) and, consequently, do not pass through the triple magic frequency point for the \( \alpha_{J = 0,1,2} \) curves. The higher rotational states make the contribution from the remaining term \( T_J(\Delta v', \theta) \) more important due to larger values of \( |L_J| \) and \( |R_J| \). Thus, no crossings among the polarizability curves of higher \( J \) values are expected within the magic frequency window.

The similarity of the \( \alpha_J \) curves in the medium-detuned regime with increasing \( J \) values is explained by the asymptotic behavior of the angular factors \( A_J(\theta) \) and \( B_J(\theta) \) in Eqs. (14) and (15) in the large \( J \) limit. Expanding \( A_J(\theta) \) and \( B_J(\theta) \) in terms of \( 1/J \), we obtain,

\[
A_J(\theta) = \frac{1 + \cos^2(\theta)}{8} + O\left(\frac{1}{J^2}\right),
\]

and,

\[
B_J(\theta) = \frac{1 + \cos^2(\theta)}{8} + \frac{\sin^2(\theta)}{8J} + O\left(\frac{1}{J^2}\right).
\]

With increasing \( J \), the leading order terms of both \( A_J(\theta) \) and \( B_J(\theta) \) are independent of the value of \( J \); hence the expression for \( \alpha_J \) in Eq. (16) becomes the same for all \( J \), neglecting the remaining \( T_J(\Delta v', \theta) \) term. Thus, for large \( J \), the various \( \alpha_J \) curves are close and almost parallel to each other in the medium-detuned regime. Combining the true triple magic condition for the lower \( J \) values and the similarity between \( \alpha_J \) for higher \( J \) values, leads to a “near magic” trapping window for multiple rotational states that should be possible to realize experimentally.

The \( \theta \)-independence of \( \alpha_J \) within the multiple magic frequency window is also verified by our numerical results. Figure 7 shows the dynamic polarizability \( \alpha_{J = 1} \) for angles between 0° and 90°. All the curves nearly cross the same point around the detuning of \( 2\pi \times 218 \text{ GHz} \).

Based on all the results and observations discussed above, we conclude that the existence of the multiple magic frequency window presents a frequency region of a few gigahertz within which the system is super robust with respect to the fluctuations of the trapping laser frequency and the polarisation direction for arbitrary rotational states. Within this window long-rotational coherences should be possible on multiple rotational transitions in the \(^{87}\text{Rb}^{133}\text{Cs} \) molecule.

C. Criteria for the Multiple Magic Frequency Window

The existence of the multiple magic frequency window relies on the condition that the remaining \( T_J(\Delta v', \theta) \) term in Eq. (18) is much smaller than the \( \alpha_{bg,\perp} \) and thus can be neglected. Taking the leading order term of \( T_J(\Delta v', \theta) \) in Eq. (17), the condition \( |T_J(\Delta v', \theta)| \ll |\alpha_{bg,\perp}| \) yields a lower bound on the transition width \( \Gamma_{0,\perp} \) in terms of
FIG. 8: The dynamic polarizabilities of the $J = 0$, 1, and 2 rotational states near the resonance transitions to the (a) $v' = 1$, (b) $v' = 2$, and (c) $v' = 3$ vibrational states of the $b^3\Pi_0$ potential. A magnetic field of $B = 181$ G is applied in the $z$-direction. No static electric field is applied. The black solid, red dashed, and blue dotted lines correspond to the dynamic polarizabilities of $J = 0$, 1, and 2 rotational states, respectively. A near triple magic condition exists in (a) and (b) but not in (c).

For $^{87}$Rb$^{133}$Cs molecules near the narrow transitions to the bottom of the $b^3\Pi_0$ potential, the right hand side of Eq. [22] is equal to $2\pi \times 0.125$ kHz. As the transition linewidth $\Gamma_{0,v'}$ decreases with increasing $v'$, this condition puts a constraint on the number of vibrational poles around which the multiple magic frequency window exists.

Figure 8 shows $\alpha_J$ for $J = 1, 2,$ and $3$ near the $v' = 1, 2,$ and $3$ vibrational poles at the bottom of the $b^3\Pi_0$ potential. With increasing $v'$, the transition is narrower and the triple crossing moves towards the pole of $\alpha_J$. The transition widths are $\Gamma_{0,v'=1} = 2\pi \times 6.84$ kHz for the $v' = 1$ pole and $\Gamma_{0,v'=2} = 2\pi \times 1.44$ kHz for the $v' = 2$ pole. Triple crossings can be seen around $\Delta v'=1 = 2\pi \times 120$ GHz for the $v' = 1$ vibrational pole (Fig. 8(a)) and around $\Delta v'=2 = 2\pi \times 22$ GHz near the $v' = 2$ vibrational pole (Fig. 8(b)). For $v' = 3$, the transition width $\Gamma_{0,v'=3}$ is $2\pi \times 0.206$ kHz which is already close to the lower bound. Thus, no triple crossings can be seen in Fig. 8(c).

D. Imaginary Polarizability in the Magic Trapping Window

Light-induced decoherence of rovibrational levels of a polar molecule is often characterized by the imaginary part of the polarizability \[81\], which accounts for losses due to spontaneous emission and other decay mechanism of intermediate electronically excited states. Here, we evaluate the imaginary part of the complex molecular dynamic polarizability $\alpha(h\omega, \epsilon)$ as

\[
\alpha(h\omega, \epsilon) = \frac{1}{\epsilon_0 c} \sum_f \frac{(E_f - i h \gamma_f / 2 - E_i)^2 - (\hbar \omega)^2}{(E_f - i h \gamma_f / 2 - E_i)^2 - (\hbar \omega)^2} \times |\langle \vec{d}_{tr} \cdot \epsilon | \rangle|^2,
\]

assuming that each of these intermediate $E_f$ state has a line width $\gamma_f$ equal to 6 MHz, the atomic line width of Rb $5p(2P)$ state. This assumption is justified by previous calculations of the imaginary polarizability of rovibrational levels of ground state KrB molecules \[76\] and a comparison of $\alpha_{imag}$ with an experimentally measured value \[81\]. The sum over $f$ in Eq. [23] is limited to transi-
tions to relativistic electronic excited potentials that dissociate to either a singly excited Rb or a singly excited Cs atom.

Figure 9 shows the calculated imaginary part of the polarizability of the $v = 0, J = 0, 1, 2$ $X^1\Sigma_+^+$ states as functions of laser frequency. By construction the imaginary part is negative. It is several orders of magnitude smaller than the real part. The resonances in the graph correspond to poles due to the lowest vibrational $\nu'$ of the $\Omega = 0$ relativistic component of the $b^3\Pi_0$ potential. For a detuning of $\Delta\nu' = 0 = 2\pi \times 218$ GHz close to the triple magic frequency shown in Fig. 3 the value of the imaginary part of the polarizability is $1.0 \times 10^{-9}$ kHz/(W/cm$^2$). For comparison, the polarizability at this detuning is $\alpha_J = h \times 0.03392$ kHz/(W/cm$^2$), as stated earlier.

VI. DISCUSSION

Although all the results above are derived by considering transitions to the $b^3\Pi_0$ potential, similar results to Eqs. (15) and (19) are found for $\Omega = 1$ potentials with $\alpha_{bg,1}$ replaced by $\alpha_{bg,\parallel}$ and vice versa. These observations indicate that any rovibrational pole that is associated with a resonance transition to the state with quantum number $\Omega$ can be used to cancel the contributions to the rank-2 dynamic polarizability tensor from all the other far-detuned states with the same quantum number $\Omega$. What remains is the contribution to the dynamic polarizability from the states with different $\Omega$. This cancellation happens at a frequency that is independent of the rotational quantum number $J$ and the polarization direction of the laser.

Even though the derivation of the equations in Sec. VI.B is “universal”, i.e. independent of the molecule species, the existence of the magic frequency window does require certain conditions to be fulfilled. For example, Eq. (22) gives us a lower bound on the transition width. For heavier molecules, such as $^{87}$Rb,$^{133}$Cs, this condition can be satisfied near the narrow transitions to the bottom of $b^3\Pi_0$ potential, since the spin-orbit coupling effect is stronger and the rotational constants, $B_v$ and $B_{\nu'}$, are smaller. For $^{23}$Na$^{87}$Rb, we also find that the multiple magic frequency window exists near the narrow transitions to the $b^3\Pi_0$ potential. However, compared to $^{87}$Rb,$^{133}$Cs, the window only exists near the $\nu' = 0$ and $\nu' = 1$ vibrational poles and missing near the $\nu' = 2$ pole.

Here, we emphasise that the condition on the lower bound of the transition width given by Eq. (22) is not the only criteria for the existence of the multiple magic frequency window. Eq. (22) allows the multiple magic frequency window to also be found near to broad transitions. However, in this case, the predicted magic frequency position in Eq. (19) cannot be larger than the energy spacing between two nearest neighbor vibrational poles (i.e., $|\Delta\nu',\nu'| \ll |\omega_{\nu'}|\pm 1$ – $\omega_{\nu'}$). This condition puts an upper bound for the transition width,

$$\Gamma_{0,\nu'} \ll \frac{2\omega_{\nu'}}{3\pi c^2} |\alpha_{bg,\parallel} - \alpha_{bg,\perp}| \times |\omega_{\nu'}|$$

where the “$+/−$” should be used for the positive/negative value of $\alpha_{bg,\parallel} - \alpha_{bg,\perp}$. This condition is very easily satisfied near the narrow transitions, however, it needs to be examined near to the broad ones. This condition implies that we need to be in the “medium-detuned” regime to find the multiple magic frequency window.

Although the existence of the multiple magic frequency windows needs to be checked case-by-case, the results derived in this work will greatly benefit the search for them. In experiments, the background values of the polarizabilities and the transition widths can both be straightforwardly measured. According to Eq. (19), the magic detuning can then be predicted based entirely upon these measured values.

VII. CONCLUSION

We have investigated magic-wavelength trapping of ultracold bialkali molecules in the vicinity of weak optical transitions from the vibrational ground state of the $X^1\Sigma_+^+$ potential to low-lying rovibrational states of the $b^3\Pi_0$ potential, focussing our discussion on the $^{87}$Rb,$^{133}$Cs molecule. We have shown that a magic trapping frequency window for multiple rotational states exists between two nearest neighbor vibrational poles, far away from any rotational poles. Within this window, the laser trapping is “near magic” for multiple rotational states simultaneously and is exactly magic for pairs of neighboring rotational states at specific laser frequencies. Moreover, the “near magic” frequency window can be tuned to a true magic frequency for the lowest three rotational states by applying an experimentally accessible DC electric field. This true triple magic condition is expected to be useful for future studies of synthetic spin-1 systems using ultracold molecules.

We have derived a set of criteria that must be fulfilled to ensure the existence of such magic frequency windows and have also presented an analytic expression for the position of the frequency window in terms of a set of experimentally measurable parameters. These will provide a straightforward, self-consistent approach to search for the magic trapping frequency window in future experiments. We expect the realization of optical traps which are simultaneously magic for multiple rotational states will enable the implementation of highly tunable models in quantum magnetism [57] and the mapping of many rotational levels onto a synthetic dimension [26]. More broadly, our work is relevant in settings where there is a need to control the relative polarizabilities of different molecular rotational states, facilitating, for example, the study of Hopf insulators in dipolar systems [82].
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