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Electric-field-dependent dynamic polarizability and state-insensitive conditions for optical trapping of diatomic polar molecules

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Selection of state-insensitive or "magic" trapping conditions with ultracold atoms or molecules, where pairs of internal states experience identical trapping potentials, brings substantial benefits to precision measurements and quantum computing schemes. Working at such conditions could ensure that the detrimental effects of inevitable inhomogeneities across an ultracold sample are significantly reduced. However, this aspect of confinement remains unexplored for ultracold polar molecules. Here, we present means to control the ac Stark shift of rotational states of ultracold diatomic polar molecules, when subjected to both trapping laser light and an external electric field. We show that both the strength and relative orientation of the two fields influence the trapping potential. In particular, we predict "magic electric field strengths" and a "magic angle," where the Stark shift is independent of the dc external field for certain rotational states of the molecule.

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The advantage of using state-independent light traps for precision frequency measurements with ultracold atoms has been demonstrated in several experiments [1,2]. Applications of this approach were analyzed in the context of optical atomic clocks and coherent control of atoms and photons within an optical cavity. For these applications the frequency of the laser beam that creates a far-off-resonant optical dipole trap is chosen such that the ac Stark shifts of the ground and one excited electronic atomic level are the same. In this way any optical atomic transition between these levels is unaffected by the trapping light. In Refs. [3–6] state-insensitive trapping conditions have also been found for microwave transitions in the atomic ground state by using a combination of the vector and tensor components of the ac Stark shift and an external magnetic field.

Ultracold molecular systems possess unique proporties that are considered to make them potentially useful as tools for precision measurements [7,8] and quantum computing [9,10]. Therefore it is desirable to extend the zero-differential ac Stark shift technique to these more complex systems. The idea of magic frequencies for vibrational Raman transitions in homonuclear Sr_2 molecules was first explored in Refs. [11,12]. This molecule is proposed for a search of possible time variation of the electron-to-proton mass ratio. The ac Stark shift of a molecule is determined by the dynamic molecular polarizability $\alpha(\nu)$, which is a function of radiation frequency ν and its polarization.

Polar molecules have a permanent dipole moment and their levels can be shifted and mixed with one another by applying an external electric field. This opens up a new way to create magic trapping conditions for two rotational levels of the molecule. In the presence of an external electric field, J is not a good quantum number and all states, even the "rotationless" ground state, have an anisotropic polarizability [13]. The anisotropy of the dynamic polarizability of these levels manifests itself as a dependence on the relative orientation of the polarization of the trapping laser and the dc electric field. The combined action of these two fields can be a powerful tool to manipulate and control ultracold molecules trapped in an optical potential.

The behavior described here has potential applications to several experiments that have been envisioned for diatomic polar molecules held in optical traps. The implications are particularly striking for the use of polar molecules in an optical lattice as quantum bits. As was described in Ref. [9], a pair of rotational states forms a suitable quantum bit. However, it was pointed out that this system is susceptible to decoherence due to intensity fluctuations in the optical trapping lasers, if the dynamic polarizabilities of these levels differ as is generally the case. This in turn leads to very stringent requirements on the laser intensity stability for such a system to be practical. As we will show, this limitation can be removed by adjusting the experimental parameters to guarantee that the dynamic polarizabilities of these states are equal. In particular, for this proposed system (where a spatial gradient of the electric field $\vec{\mathcal{E}}$ is required, so working at a magic electric field value is impossible) it should be possible to use light polarized at the "magic angle" relative to the static field in order to eliminate this potentially dangerous source of decoherence.

The existence of magic electric field values is also of possible use for envisioned applications where polar molecules in optical lattices are employed in novel types of many-body systems. This includes, for example, cases where the properties of long-range molecular interactions are tailored by a combination of a static electric field and resonant microwave fields coupling different rotational states [14–16]. Working at a magic electric field value in such systems could ensure that the inevitable inhomogeneities in the intensity of the trapping light across a large sample would not change the resonant condition for the microwave drive fields. Hence, working under such magic conditions might be necessary to implement proposals of this type.

Motivated by these ideas, we calculate the near-infrared dynamic polarizability of various rotational levels of the

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$v = 0$ vibrational state of the $X\,^1\Sigma^+$ potential of the KRb and RbCs molecules, under the simultaneous influence of trapping electromagnetic and static electric fields. We calculate dynamic polarizability using the computational techniques developed in previous publications [17–19]. Our calculations with an external electric field are predominantly performed at a laser frequency of 9174 cm^{-1} (or wavelength of 1090 nm), which corresponds to an often-used frequency to trap atoms and molecules in ultracold experiments. In the near infrared this laser frequency is sufficiently far away from molecular resonances of the excited electronic states that heating due to photon scattering is negligible. We focus on external electric field strengths up to 15 kV/cm, a value which should be experimentally accessible.

We begin by studying the dynamic polarizability or ac Stark shift for the $J = 0$ and $J = 1$ rotational levels of the $v = 0$ $X\,^1\Sigma^+$ state of KRb and RbCs in the near infrared, without an external electric field. The procedure used to determine the complex molecular polarizability has already been described in our previous paper [17]. Figure 1 illustrates the results of this study in the absence of an electric field by showing the dynamic polarizability α as a function of laser frequency $h\nu$. The range of laser frequencies spans a technologically relevant near-infrared optical domain and includes the lasing frequency near 9174 cm^{-1} used in Ref. [13]. The figure also shows that the dynamic polarizability of the molecular state depends on the rotational quantum number J and its projection M. The curves for the different states do not cross as a function of the laser frequency. In other words, at zero electric field we can not find a magic frequency for $J = 0$ and $J = 1$ states using trapping light in the near infrared.

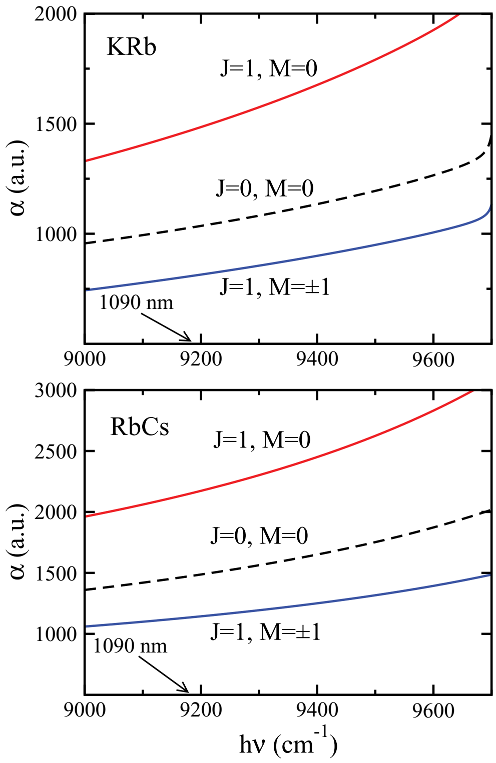

FIG. 1. (Color online) Dynamic polarizability in the absence of an electric field and in atomic units for the $v = 0$, $J = 0$ and 1 levels of the $X\,^1\Sigma^+$ ground state of KRb and RbCs as a function of trapping frequency in the near infrared domain. The dashed lines show the polarizability for the $J = 0$ and $M = 0$ level, which is independent of the light polarization. The solid lines correspond to the $J = 1$ state with magnetic sublevels $M = 0$ and ± 1 illuminated by linear polarized light along the $\hat z$ direction. One atomic unit of polarizability corresponds to $4.686\,45 \times 10^{-8}$ MHz/(W/cm^2).

I. FIELD-DEPENDENT DYNAMIC POLARIZABILITY

Here we extend the idea of the ac Stark shift for the rotational levels of the $X\,^1\Sigma^+$ ground state to the mixing of these levels in a static electric field $\vec{\mathcal{E}} = \mathcal{E}\hat z$ along the space-fixed $\hat z$ direction. Our analyses show that the experimentally accessible electric fields mix only a few low-lying rotational states. The ultimate goal is to introduce the dynamic polarizability of the mixed rotational levels as a function of an external dc electric field and the polarization of the ac trapping field. In a heteronuclear molecule this Stark mixing is primarily due to the permanent electronic dipole moment $\vec d$; we neglect the much smaller effects due to Stark mixing with other electronic states. For the Hund's case (a) $X\,^1\Sigma^+$ state of an alkali-metal dimer, the total electron spin and orbital angular momentum are not coupled to the molecular rotation. The molecular wave function in the laboratory frame, $|vJM\rangle_z$, is then given by

$$|vJM\rangle_z \equiv |vJ\rangle Y_{JM}(\hat R) = \left\{ \frac{\psi_{vJ}(R)}{R} |X\,^1\Sigma^+\rangle \right\} Y_{JM}(\hat R), \qquad (1)$$

where v is a vibrational quantum number, J and M are the molecular rotational angular momentum and its projection along the z axis, $\psi_{vJ}(R)$ is the radial rovibrational wave function, $Y_{JM}(\hat R)$ is a spherical harmonic, $\hat R$ is the orientation of the molecule relative to the electric field direction z, and $|X\,^1\Sigma^+\rangle$ is the electronic wave function with projections defined along the internuclear axis. For the $X\,^1\Sigma^+$ state, in even modest electric fields the nuclear spins are well decoupled from the other spins and angular momenta; hence we ignore the nuclear spins here. Then the Hamiltonian for such a system becomes

$$H = \sum_{vJM} E_{vJ} |vJM\rangle_z \times {}_z\langle vJM| - \vec d \cdot \vec{\mathcal{E}}, \qquad (2)$$

where $E_{vJ} = G_v + B_v J(J+1)$ and G_v and B_v are the vibrational energy and rotational constant of vibrational level v, respectively. Higher order rotational corrections are negligible. For the alkali-metal dimers KRb and RbCs we have that $\Delta G_v = G_{v+1} - G_v$ is on the order of 50–100 cm^{-1} for small v, while B_v is on the order of 0.017–0.037 cm^{-1}.

We evaluate the matrix elements of the operator $-\vec d \cdot \vec{\mathcal{E}}$ by noting that after averaging over the electronic wave function $|X\,^1\Sigma^+\rangle$ it reduces to $-d(R)C_{10}(\hat R)\mathcal{E}_z$, where $d(R) = \langle X\,^1\Sigma^+|d|X\,^1\Sigma^+\rangle$ is the R-dependent permanent electric dipole moment, $C_{lm}(\hat R) = \sqrt{4\pi/(2l+1)}Y_{lm}(\hat R)$ are tensors of rank l, and $\mathcal{E}_z$ is the electric field strength. Consequently, this operator conserves the projection quantum number M. The matrix element between two rovibrational states is

$$
\begin{aligned}
&{}_z\langle vJM| - \vec{d}\cdot\vec{\mathcal{E}}|v'J'M'\rangle_z \\
&= -\delta_{MM'} d_{vJ,v'J'} \mathcal{E}_z \int d\hat{R}\, Y^*_{JM}(\hat{R}) C_{10}(\hat{R}) Y_{J'M'}(\hat{R}) \\
&= -\delta_{MM'} d_{vJ,v'J'} \mathcal{E}_z (-1)^M \sqrt{(2J+1)(2J'+1)} \\
&\quad \times \begin{pmatrix} J & 1 & J' \\ -M & 0 & M' \end{pmatrix} \begin{pmatrix} J & 1 & J' \\ 0 & 0 & 0 \end{pmatrix}, \qquad (3)
\end{aligned}
$$

where $\delta_{MM'}$ is the Kronecker δ function, $d_{vJ,v'J'} = \int_0^\infty dR\, \psi_{vJ}(R) d(R) \psi_{v'J'}(R)$, and $(\cdots)$ are 3-j symbols (see, e.g., [20]). This matrix element is nonzero when $J+1+J'$ is even, according to the parity selection rules, and is independent of the sign of M and M'. For the small J values of interest here we can assume that the J dependence of $\psi_{vJ}(R)$ is negligible. Moreover, coupling between vibrational levels can also be ignored, as the dipole moment $d(R)$ is a slowly varying function with R and the spacing between vibrational levels is large compared to the rotational splitting. For tensor operators $C_{lm}(\hat{R})$ of rank 0, 1, and 2 the relationship between Cartesian x, y, and z components and spherical $m = -1$, 0, and $+1$ components can be found in Refs. [21,22].

For each projection M and vibrational level v, eigenvalues and eigenvectors of the Hamiltonian, Eq. (2), are obtained by the direct diagonalization of the Hamiltonian matrix including rotational states J from $|M|$ up to some value $J_{\max}$. For the external electric field strength and J values of interest, $J_{\max} = 10$ is sufficient for convergence. We label the eigenenergies by $E_{v\tilde{J}M}$ with corresponding eigenvectors $|v\tilde{J}M\rangle = \sum_J U^{vM}_{\tilde{J},J} |vJM\rangle_z$. Here $\tilde{J}$ is an integer index with values $\tilde{J} = |M|, |M|+1, \ldots$, such that the eigenstate $|v\tilde{J}M\rangle$ adiabatically connects to the electric field–free eigenstate $|vJM\rangle_z$ with $J = \tilde{J}$. For $|M| > 0$ the levels with projection quantum number $-M$ and M remain degenerate. The dipole matrix element between states $|v\tilde{J}M\rangle$ and $|v'\tilde{J}'M'\rangle$ of the X potential is given by $\langle v\tilde{J}M|d_\sigma|v'\tilde{J}'M'\rangle = \sum_{J,J'} U^{vM}_{\tilde{J},J} U^{v'M'}_{\tilde{J}',J'} \times {}_z\langle vJM|d_\sigma|v'J'M'\rangle_z$, where σ is a spatial index that can be expressed in either Cartesian coordinates x,y,z, or spherical coodinates $q = 0,\pm 1$.

Now we are able to calculate the dynamic polarizability of the mixed rotational eigenstates at laser frequency ν. It is determined by the properties of the operator $\alpha_{\sigma\sigma'}(\nu)$ defined by [21,23]

$$
\begin{aligned}
\alpha_{\sigma\sigma'}(\nu) = \sum_\gamma \left\{ \frac{1}{E_\gamma - E_{v\tilde{J}M} + h\nu} + \frac{1}{E_\gamma - E_{v\tilde{J}M} - h\nu} \right\} \\
\times d_\sigma |\gamma\rangle\langle\gamma| d_{\sigma'} \qquad (4)
\end{aligned}
$$

with γ enumerating eigenstates of the ground as well as excited electronic potentials in the presence of the electric field and $\sigma,\sigma' = x$, y, or z. We will focus on the $J = 0$ and $J = 1$ levels and without loss of generality assume that the x axis of our coordinate system lies in the plane spanned by the electric-field direction and the orientation of the linearly polarized laser light. Moreover, we are interested in the situation where the level shifts due to the laser are small compared to those induced by the electric field. For the isolated levels with $M = 0$, the polarizability is determined by the diagonal matrix element $\alpha^{v\tilde{J}M=0}_{\sigma\sigma'}(\nu) = \langle v\tilde{J}M = 0|\alpha_{\sigma\sigma'}(\nu)|v\tilde{J}M = 0\rangle$. The $|M| > 0$ dynamic polarizability needs to be treated by degenerate perturbation theory within the two-dimensional subspace $|v\tilde{J}M\rangle$ and $|v\tilde{J} -M\rangle$. In fact, our choice of the x direction and the symmetry properties of the dynamic polarizability ensure that the linear combinations $|v\tilde{J}M,\pm\rangle = \{|v\tilde{J}M\rangle \pm |v\tilde{J} -M\rangle\}/\sqrt{2}$ with $M > 0$ are the correct eigenstates. For these states the dynamic polarizability is $\alpha^{v\tilde{J}M,\pm}_{\sigma\sigma'}(\nu) = \langle v\tilde{J}M,\pm|\alpha_{\sigma\sigma'}(\nu)|v\tilde{J}M,\pm\rangle$. For diatomic species, only the diagonal elements $\alpha^{v\tilde{J}M,\pm}_{xx}$, $\alpha^{v\tilde{J}M,\pm}_{yy}$, and $\alpha^{v\tilde{J}M,\pm}_{zz}$ are nonzero. Hence in an oscillating electric field $\vec{\mathcal{E}}_o(t) = \mathcal{E}_o(0)\mathrm{Re}\{\vec{\epsilon} e^{i2\pi\nu t}\}$ (where $\vec{\epsilon}$ is the complex unit vector indicating the polarization), the state $|v\tilde{J}M,\pm\rangle$ shifts in energy by an amount ΔE given by $\Delta E = -\sum_{\sigma,\sigma'} |\mathcal{E}_o(0)|^2 \alpha^{v\tilde{J}M,\pm}_{\sigma\sigma'}(\nu)\epsilon_\sigma\epsilon^*_{\sigma'}/4$. The sum is over the spatial indices $\sigma,\sigma' = x,y,z$.

We are interested in near-infrared laser frequencies, which are detuned away from resonances with rovibrational levels of the electronically excited potentials. In particular, we focus on wavelengths between 1000 and 1100 nm. Starting from the $v = 0$ vibrational level of the $X\,^1\Sigma^+$ states of KRb and RbCs, photons of this wavelength do not possess sufficient energy to reach rovibrational levels of the electronically excited singlet $^1\Lambda$ potentials. The photons do have enough energy to reach vibrational level of the triplet $b^3\Pi$ potential considering only single-photon excitations. However, such transitions require relativistic spin-orbit coupling to the $A\,^1\Sigma^+$ potential to acquire a nonzero dipole matrix element; such spin-orbit induced couplings are small enough to neglect under the conditions of interest here.

This allows us to make several approximations. First, we can use nonrelativistic potentials and transition dipole moments and in the calculation of the dynamic polarizability consider only singlet $^1\Sigma^+$ and $^1\Pi$ potentials. Second, assuming a large detuning (such that $|E_\gamma - E_{v\tilde{J}M} - h\nu| \gg E_{vJM}$ for all states of interest) we can neglect the electric field and rotational dependence of the energy denominators in Eq. (4). Finally, we find that in the near infrared the contribution to the polarizability from intermediate states γ in the ground $X\,^1\Sigma^+$ state is small.

With this in mind the polarizability becomes

$$
\begin{aligned}
\alpha^{v\tilde{J}M,\pm}_{\sigma\sigma'}(\nu) \cong & \sum_{J,J'} U^{v|M|}_{\tilde{J},J} U^{v|M|}_{\tilde{J},J'} \\
& \times \sum_{ev_e\Lambda} \left\{ \frac{1}{E_{ev_e\Lambda} - E_v + h\nu} + \frac{1}{E_{ev_e\Lambda} - E_v - h\nu} \right\} \\
& \times \sum_{J_eM_e} {}_z\langle vJM,\pm|d_\sigma|ev_eJ_eM_e\Lambda\rangle_z \\
& \times {}_z\langle ev_eJ_eM_e\Lambda|d_{\sigma'}|vJ'M,\pm\rangle_z, \qquad (5)
\end{aligned}
$$

where the energy E_v is a typical vibrational energy in the ground state potential, and the rovibrational wave functions $|ev_eJ_eM_e\Lambda\rangle$ of the electronically excited states with approximate energy $E_{ev_e\Lambda}$ are given by

$$
\begin{aligned}
|ev_eJ_eM_e\Lambda\rangle_z &\equiv |ev_e\Lambda\rangle \times |J_eM_e\Lambda\rangle_z \\
&\equiv \left\{ \frac{\phi_{v_e}(R)}{R} |e\,^1\Lambda\rangle \right\} \times \left\{ \sqrt{\frac{2J+1}{4\pi}} D^{J_e*}_{M_e\Lambda}(\hat{R}) \right\},
\end{aligned}
$$

where the vibrational and electronic dependence has been isolated in the ket $|ev_e\Lambda\rangle$ and the rotational dependence in $|J_eM_e\Lambda\rangle_z$, respectively. The wave function $\phi_{v_e}(R)$ is the radial rovibrational wave function, the ket $|e^1\Lambda\rangle$ is the electronic state with projection quantum number Λ defined along the internuclear axis, and $D^J_{MM'}(\hat{R})$ is a Wigner rotation matrix that describes a symmetric top rotational wave function. The sum over Λ includes both positive and negative values, where $\Lambda = 0$ corresponds to excited ${}^1\Sigma^+$ electronic states and $\Lambda = \pm 1$ to ${}^1\Pi$ states.

As stated before, the energy of the excited state and $\phi_{v_e}(R)$ depend on the electronic state $|e^1\Lambda\rangle$ and vibrational level v_e, but not J_e and M_e. Consequently, the transition dipole moments separate into ${}_z\langle vJM,\pm|d_\sigma|ev_eJ_eM_e\Lambda\rangle_z = \langle vJ|d(R)|ev_e\Lambda\rangle F^{J_eM_e\Lambda}_{JM\pm,\sigma}$, where $F^{J_eM_e\Lambda}_{JM\pm,\sigma}$ is an integral of the product of three Wigner rotation matrices $D^J_{MM'}(\hat{R})$ over the orientation of the molecule $\hat{R}$ that can be evaluated using angular momentum algebra [20].

Moreover, we have verified that $\langle vJ|d(R)|ev_e\Lambda\rangle$ is nearly independent of J. The sums over J_e and M_e in Eq. (5) can now be performed and we finally find

$$\langle v\tilde{J}M,\pm|\alpha_{\sigma\sigma'}(\nu)|v\tilde{J}M,\pm\rangle = \sum_{\Lambda}\alpha^{\Lambda}(\nu)\sum_{J,J'}U^{v|M|}_{\tilde{J},J}U^{v|M|}_{\tilde{J},J'} \times \sum_{J_eM_e}F^{J_eM_e\Lambda}_{JM\pm,\sigma}F^{J_eM_e\Lambda}_{J'M\pm,\sigma'} \qquad (6)$$

with the Λ-dependent

$$\alpha^{\Lambda}(\nu) = \sum_{ev_e}\langle vJ|d|ev_e\Lambda\rangle\langle ev_e\Lambda|d|vJ\rangle \times \left\{\frac{1}{E_{ev_e\Lambda}-E_v+h\nu}+\frac{1}{E_{ev_e\Lambda}-E_v-h\nu}\right\}. \qquad (7)$$

The parallel $\alpha^0(\nu)$ and perpendicular $\alpha^1(\nu)$ contributions to the polarizability are due to transitions to the Σ and Π states, respectively.

II. MAGIC DC ELECTRIC FIELD

The polarizability depends on the strength of an external electric field through the use of the unitary matrices $U^{vM}_{\tilde{J},J}$ of Eq. (6). Figure 2 shows the dynamic polarizability of the ground states of the KRb and RbCs molecules as a function of the external electric field strength. The left panels of these figures correspond to the parallel ($\vec{\epsilon} = \hat{z}$) and the right panels to the perpendicular ($\vec{\epsilon} = \hat{x}$) polarization of the trapping light relative to the electic field direction $\vec{\mathcal{E}}$. In all cases the polarizability depends on both the state index $\tilde{J}$ and the angular momentum projection M. For KRb (Fig. 2, top row) a "magic" electric field strength exists at $\mathcal{E} = 10$ kV/cm, where the polarizability of $\tilde{J}=0$, $M = 0$ and $\tilde{J}=1$, $M = 0$ states coincide. This is possible due to the polar character of the molecules. For RbCs (Fig. 2, bottom row) two magic electric field strengths exist. For the pair of states $\tilde{J}=0$, $M = 0$ and $\tilde{J}=1$, $M = 0$ a crossing occurs near 2 kV/cm. Another crossing appears for the states $\tilde{J}=1$, $M = 0$ and $\tilde{J}=1$, $M = \pm 1$ at 4.7 kV/cm. They both are at a much smaller field strength than for KRb, since RbCs has a larger permanent dipole moment and smaller rotational splittings in the ground state. Note that the $M = 0$ magic electric field occurs at the same field strength for both parallel and perpendicular polarization of the trapping light. In fact, they are the same for any polarization.

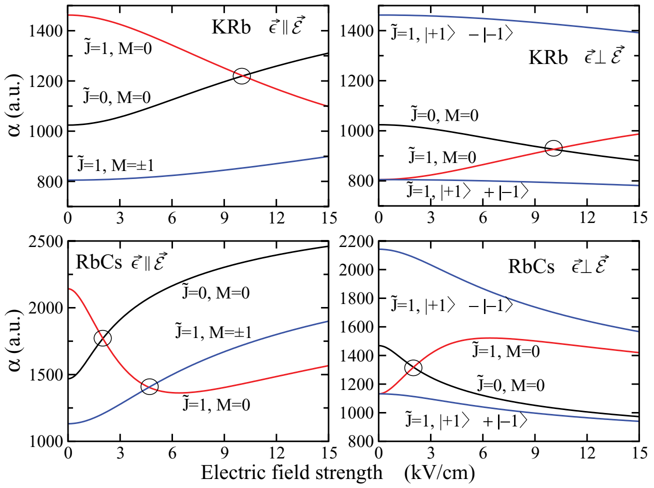

FIG. 2. (Color online) Dynamic polarizability at a wave number of 9174 cm^{-1} of the $v = 0$, $\tilde{J}=0$ and 1 levels of the $X\,{}^1\Sigma^+$ ground state of KRb (top row) and RbCs (bottom row) as a function of the field strength of an external electric field. The polarization of the trapping field is parallel (left panel) and perpendicular (right panel) to the direction of the electric field. The circles indicate a crossing point between $\tilde{J}=0$ and 1 polarizabilities. The degeneracy of the states $\tilde{J}=1$ and $M = \pm 1$ is lifted by perpendicularly polarized laser light. The new states are labeled $|+1\rangle \pm |-1\rangle$, corresponding to $|\tilde{J}=1, M\pm\rangle$ respectively. The $M = \pm 1$ states remain degenerate for parallel polarization.

III. MAGIC ANGLE

In future experiments one would expect to be able to change the angle between the static and dynamic electric fields. Figure 3 shows a surface plot of the dynamic polarizability as a function of $\mathcal{E}$ and the angle θ between $\vec{\mathcal{E}}$ and polarization of the

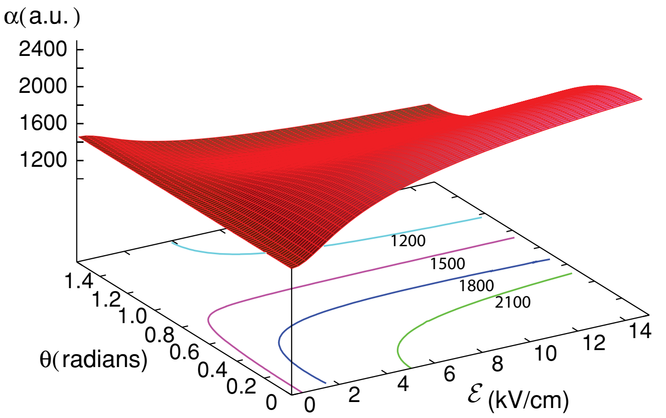

FIG. 3. (Color online) Dynamic polarizability of the $v = 0$, $\tilde{J}=0$, $M = 0$ level of the $X\,{}^1\Sigma^+$ ground state of RbCs as a function of external electric field strength and the angle θ between the direction of the electric field $\vec{\mathcal{E}}$ and the polarization $\vec{\epsilon}$ of the trapping light at a wave number of 9174 cm^{-1}. The contour lines are marked by the polarizability values in atomic units.

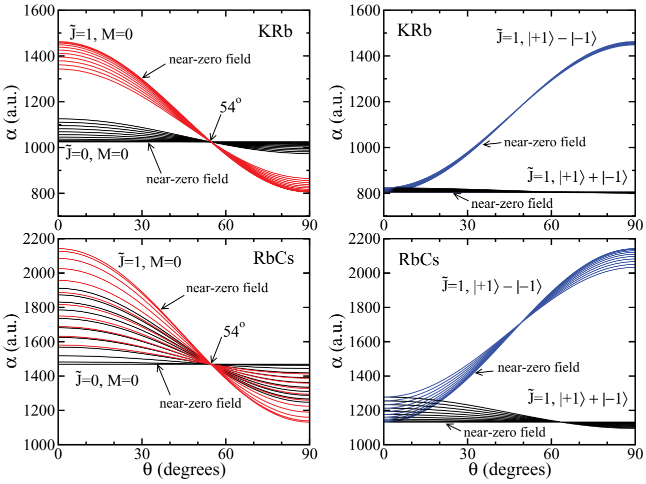

FIG. 4. (Color online) Dependence of the dynamic polarizability on the angle between the polarization $\vec{\epsilon}$ of the trapping light at a wave number of 9174 cm^{-1} and the direction of the external electric field $\vec{\mathcal{E}}$, for different values of $\mathcal{E}$. The polarizability of states with projections $M = 0$ and $M = \pm 1$ of the $X\,^1\Sigma^+$ ground state of KRb and RbCs is shown in the left and right panels, respectively. For clarity $\mathcal{E}$ ranges from 0.6 kV/cm to 6 kV/cm in steps of 0.6 kV/cm for KRb and from 0.3 kV/cm to 3 kV/cm in steps of 0.3 kV/cm for RbCs. At the magic angle $\theta = \theta_0 = 54^\circ$, the polarizabilities of the states $\tilde{J} = 0$, $M = 0$, and $\tilde{J} = 1$, $M = 0$ are identical and independent of the strength of the static electric field. For the $\tilde{J} = 1$, $M = \pm 1$ states no magic angle exists.

trapping light $\vec{\epsilon}$, for linearly polarized light. The polarizability depends smoothly on both θ and $\mathcal{E}$.

Figure 4 compares the dynamic polarizability of the states $|\tilde{J} = 0\rangle$ and $|\tilde{J} = 1, M = 0, \pm 1\rangle$ as a function of the angle θ of the linear polarization of the optical field relative to the static field (such that $\hat{\epsilon} = \cos\theta\hat{z} + \sin\theta\hat{x}$), for several static electric field strengths within the range from 0 to 6 kV/cm for KRb and 0 to 3 kV/cm for RbCs. All curves with $M = 0$ cross at the angle $\theta = \theta_0$ such that $\cos^2\theta_0 = 1/3$, or $\theta_0 \approx 54^\circ$. We refer to this as the "magic angle" since here the ac Stark shift is independent of the internal state of the molecule. This behavior occurs in many contexts and is a simple consequence of the rank-2 tensor structure of the polarizability [24]. The angular dependence of the $\tilde{J} = 0$ level is smaller than that of the $\tilde{J} = 1$ levels as expected. It inherited the zero-electric field properties of the scalar $J = 0$ state. As seen in Fig. 2, the RbCs polarizabilities of the $\tilde{J} = 0$, $M = 0$ and $\tilde{J} = 1$, $M = 0$ levels cross at a magic electric field strength of 2 kV/cm. For Fig. 4 this implies that the curves start to "overlap" when $\mathcal{E}$ is equal or larger than this magic field value.

The polarizability operator $\alpha_{\sigma\sigma'}(\nu)$, defined by Eq. (4), is a reducible rank-2 tensor operator. Hence it can be expressed as a sum of irreducible tensor operators $\alpha^{(k)}(\nu)$ of rank $k = 0,1,2$. In terms of these irreducible tensor operators, the ac Stark shift ΔE is proportional to the diagonal matrix element of the operator $\sum_{\sigma\sigma'} \alpha_{\sigma\sigma'}(\nu)\epsilon_\sigma\epsilon^*_{\sigma'}$, which can be written in the general form

$$\sum_{k=0}^{2}\sum_{q=-k}^{k}(-1)^q\alpha^k_q(\nu)\epsilon\epsilon^k_{-q}, \tag{8}$$

where q is a spherical tensor projection index and the explicit forms of the irreducible spherical tensors $\alpha^k_q(\nu)$ and $\epsilon\epsilon^k_q$ are given in Refs. [21,22].

In order to derive the magic angle condition for $M = 0$ states, we consider the effect of each term in the expansion (8). The term with $k = 0$ corresponds to the scalar polarizability; the operator $\alpha^0_0(\nu) = \sum_{\sigma\sigma'} \alpha_{\sigma\sigma'}(\nu)\delta_{\sigma\sigma'}$ has, under our approximations, diagonal matrix elements that are independent of J (or $\tilde{J}$) and M for all states of interest, and similarly the quantity $\epsilon\epsilon^0_0 \propto \vec{\epsilon}\cdot\vec{\epsilon}^* = 1$ is independent of the polarization of the optical field. The term with $k = 1$, corresponding to the vector polarizability, in general is significant. However, for the special case of linearly polarized light where $\vec{\epsilon}$ is real, the quantities $\epsilon\epsilon^1_q = (\vec{\epsilon}\times\vec{\epsilon}^*)_q$ vanish and hence the effect of the vector polarizability is zero. For the tensor polarizability terms (with $k = 2$), from the Wigner-Eckhart theorem only the operator component with $q = 0$ gives rise to a nonzero diagonal matrix element for $M = 0$ states. Hence the contribution of this term is proportional to $\epsilon\epsilon^2_0$. Without loss of generality we can define the linear polarization as $\vec{\epsilon} = \epsilon_x\hat{x} + \epsilon_z\hat{z} = \cos\theta\hat{z} + \sin\theta\hat{x}$. In this case $\epsilon\epsilon^2_0 \propto \epsilon_z\epsilon^*_z - \vec{\epsilon}\cdot\vec{\epsilon}^*/3 = \cos^2\theta - 1/3$. Hence the contribution to ΔE due to the tensor polarizability also vanishes for all states with $M = 0$, when the optical field is linearly polarized at the "magic angle" $\theta = \theta_0$. Under this condition the only contribution to the dynamic polarizability is from the $k = 0$ scalar term, which is the same for all states of interest.

The property of the magic electric field discussed in Fig. 2 can also be understood in terms of the tensor structure of the polarizability. At certain values of the applied dc electric field $\mathcal{E}$, the rank-2 components of the polarizability, α^2_0, of the $\tilde{J} = 0$, $M = 0$ and $\tilde{J} = 1$, $M = 0$ levels become the same. When this condition is met, the ac Stark shift becomes independent of the direction of the trapping light's linear polarization.

Here we finish with an example of a specific implementation of a magic angle 3D lattice. Let the lattice be formed by three orthogonal retroreflected laser beams a, b, and c, with initial propagation directions $\hat{k}$ given by $\hat{k}_a = \hat{y}$, $\hat{k}_b = (\hat{x} + \hat{z})/\sqrt{2}$, and $\hat{k}_c = (\hat{x} - \hat{z})/\sqrt{2}$. These three beams each have a different frequency ν, such that $\nu_a = \nu_b - \delta_b = \nu_c - \delta_c$; here the offset frequencies δ_b and δ_c must satisfy $\nu_a,\nu_b,\nu_c \gg \delta_b,\delta_c \gg f_{\mathrm{mot}}$, where f_{mot} is the motional frequency of the molecules in the optical trapping potential. The use of different frequencies (which can be generated from a single laser by using, e.g., acousto-optic modulators) in this manner eliminates the effect of interference terms between the different laser beams: such terms average to zero rapidly over the time of motion of the atom, and hence can be neglected. The resulting average trap potential is then simply the sum of the potentials due to each individual laser beam. Finally, the polarizations of the three beams can be chosen as $\hat{\epsilon}_a = \sqrt{2/3}\hat{x} + \sqrt{1/3}\hat{z}$; $\hat{\epsilon}_b = \sqrt{2/3}\hat{k}_c + \sqrt{1/3}\hat{y}$; and $\hat{\epsilon}_c = \sqrt{2/3}\hat{k}_b + \sqrt{1/3}\hat{y}$. In each case, $|\hat{\epsilon}\cdot\hat{z}| = \cos\theta_0$.

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