ac Stark effect in ultracold polar ^{87}Rb^{133}Cs molecules

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We investigate the effect of far-off-resonant trapping light on ultracold bosonic ^{87}Rb^{133}Cs molecules. We use kHz-precision microwave spectroscopy to measure the differential ac Stark shifts between the ground and first excited rotational levels of the molecule with hyperfine-state resolution. We demonstrate through both experiment and theory that coupling between neighboring hyperfine states manifests in rich structure with many avoided crossings. This coupling may be tuned by rotating the polarization of the linearly polarized trapping light. A combination of spectroscopic and parametric heating measurements allows complete characterization of the molecular polarizability at a wavelength of 1550 nm in both the ground and first excited rotational states.

Ultracold molecules in optical traps and optical lattices have many potential applications, ranging from quantum-state-controlled chemistry [1–4] to quantum simulation [5,6] and quantum information [7,8]. Many of these applications rely on coherent microwave transfer between rotational states of the molecules. However, all the molecules that have been prepared at ultracold temperatures so far [9–13] have nuclei with nonzero spins, resulting in complex hyperfine and Zeeman structures [14–19]. In such cases, the laser fields used to confine the molecules have important effects through the ac Stark effect, particularly for molecules in rotationally excited states. A thorough understanding of these effects is essential in order to eliminate differential Stark shifts detrimental to internal state transfer and thus to develop ultracold polar molecules into a controllable resource for use in quantum science.

The key quantity that determines the ac Stark effect is the molecular polarizability. Following a theoretical proposal by Kotochigova and DeMille [20], Neyenhuis *et al.* [21] carried out parametric heating experiments at fixed laser intensity to determine the polarizabilities of different molecular states. They showed that there exists a *magic* angle for the linear polarization of optical trapping light. In analogy with magic-wavelength traps in atomic systems, at the magic angle, the ac Stark shift of hyperfine levels of different rotational states are the same. This allowed Ramsey interferometry between two rotational levels of the molecule with a reasonably long coherence time and led to a pivotal study of the dipolar spin-coherence time in a three-dimensional (3D) optical lattice

[22]. More recently, Deiß *et al.* [23,24] carried out parametric heating experiments on aligned triplet Rb$_2$ molecules and extracted both isotropic and anisotropic polarizabilities.

In this Rapid Communication, we explore the dependence of the ac Stark effect on laser intensity both experimentally and theoretically, using microwave spectroscopy of the chemically stable and bosonic ^{87}Rb^{133}Cs molecule. We show that there is a subtle interplay between the ac Stark effect and the hyperfine structure. The trapping light couples neighboring hyperfine states, giving rich and complex structure with many avoided crossings as a function of laser intensity. We use our measurements to extract a precise value for the anisotropic component of the molecular polarizability. We complete the characterization of the polarizability tensor by performing parametric heating and spectroscopic measurements to extract the isotropic component of the polarizability. Our findings will allow us to engineer trapping potentials suitable for internal state transfer with long coherence times and have implications for experiments with other molecular species.

The ac Stark effect arises from the interaction of the electric field due to a laser of intensity I with the polarizability α of an atom or molecule and results in a perturbation in energy of $-\alpha I$. By contrast with the atomic case, the molecular polarizability is anisotropic; in the case of a linear diatomic molecule like ^{87}Rb^{133}Cs, the highest polarizability is along the internuclear axis. We apply a magnetic field B_z in the vertical z direction, and the orientation of the molecule is defined with respect to this magnetic field. The polarizability of the molecule at an angle θ to the internuclear axis is

$$\alpha(\theta) = \alpha_\parallel \cos^2\theta + \alpha_\perp \sin^2\theta = \alpha^{(0)} + \alpha^{(2)} P_2(\cos\theta), \quad (1)$$

where $\alpha_\parallel$ and $\alpha_\perp$ are the polarizability parallel and perpendicular to the internuclear axis respectively, $\alpha^{(0)} = \frac{1}{3}(\alpha_\parallel + 2\alpha_\perp)$ and $\alpha^{(2)} = \frac{2}{3}(\alpha_\parallel - \alpha_\perp)$. The trapping light is linearly polarized in the xz plane at an angle β to the magnetic field. We therefore

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rotate the polarizability tensor through an angle β and find matrix elements

$$
\begin{aligned}
&\langle N',M_N'|I\alpha|N,M_N\rangle \\
&= I\alpha^{(0)}\delta_{NN'}\delta_{M_N M_N'} \\
&\quad + I\alpha^{(2)}\sum_M d_{M0}^2(\beta)(-1)^{M_N'}\sqrt{(2N+1)(2N'+1)} \\
&\quad \times \begin{pmatrix} N' & 2 & N \\ 0 & 0 & 0 \end{pmatrix}\begin{pmatrix} N' & 2 & N \\ -M_N' & M & M_N \end{pmatrix},
\end{aligned} \tag{2}
$$

where N is the rotational angular momentum of the molecule, with projection M_N along the magnetic field axis, and $d^2(\beta)$ is a reduced rotation matrix.

To calculate the hyperfine levels in the presence of an ac Stark effect, we construct the Hamiltonian matrix in a decoupled basis $|N M_N\rangle |I_{Rb}m_I^{Rb}\rangle |I_{Cs}m_I^{Cs}\rangle$, where $I_{Rb}=3/2$, $I_{Cs}=7/2$, and m_I^{Rb}, m_I^{Cs} are the corresponding projections. We supplement the Zeeman and hyperfine matrix elements

of Ref. [14] with the ac Stark terms of Eq. (2), which are diagonal in and independent of m_I^{Rb} and m_I^{Cs}. We include all basis functions with $N \leqslant 3$ in the calculation. Diagonalizing the resulting Hamiltonian gives us both energy levels and wave functions in the presence of off-resonant trapping light. We then use the wave functions to calculate spectroscopic transition strengths for the required polarization of microwaves.

The representation of $\alpha\theta$ in terms of $\alpha^{(0)}$ and $\alpha^{(2)}$ conveniently separates the effects of the two components of the polarizability. The isotropic component $\alpha^{(0)}$ shifts all diagonal matrix elements by the same amount and has no effect on transition frequencies, though it does contribute to optical trapping. The anisotropic component $\alpha^{(2)}$, on the other hand, mixes different hyperfine states through matrix elements diagonal and off-diagonal in M_N and dependent on β. For $N=0$, the matrix elements of $\alpha^{(2)}$ are zero, so the polarizability is simply $\alpha^{(0)}$ for all hyperfine states. For $N=1$, however, $\alpha^{(2)}$ has important effects; if we neglect terms off-diagonal in N, the matrix of the polarizability tensor between basis functions with $N=1$, $M_N=0$, $+1$ and -1 is

$$
\langle 1,M_N'|I\alpha|1,M_N\rangle = I\alpha^{(0)} + \frac{I\alpha^{(2)}}{5}\begin{pmatrix} 2P_2(\cos\beta) & -\frac{3}{\sqrt2}\sin\beta\cos\beta & +\frac{3}{\sqrt2}\sin\beta\cos\beta \\ -\frac{3}{\sqrt2}\sin\beta\cos\beta & -P_2(\cos\beta) & -\frac{3}{2}\sin^2\beta \\ +\frac{3}{\sqrt2}\sin\beta\cos\beta & -\frac{3}{2}\sin^2\beta & -P_2(\cos\beta) \end{pmatrix}. \tag{3}
$$

In the absence of the trapping laser, $M_F = M_N + m_I^{Rb} + m_I^{Cs}$ is a good quantum number, but M_N, m_I^{Rb}, and m_I^{Cs} are not individually conserved. When the trap laser is polarized along B_z, corresponding to $\beta=0$, M_F is still conserved. However, when $\beta \neq 0$, the ac Stark effect mixes levels with different values of M_F and there are no good quantum numbers except reflection symmetry in the xz plane.

Our experimental apparatus and method for creating ultracold ^{87}Rb^{133}Cs molecules have been discussed in previous publications [11,25–31]. In this work we create a sample of up to $\sim$3000 ^{87}Rb^{133}Cs molecules in their absolute ground state at a temperature of $\sim$1 μK by magnetoassociation on a Feshbach resonance [29] followed by transfer to the hyperfine and rovibronic ground state by stimulated Raman adiabatic passage (STIRAP) [11]. The transfer of the molecules to the ground state is performed in free space [31]. We recently reported the coherent control of the rotational and hyperfine state of the molecules using external microwave fields, also in free space [19].

To measure the differential ac Stark shift between $N = 0$ and $N = 1$, we perform microwave spectroscopy in the presence of the dipole trapping light. A single beam ($\lambda = 1550$ nm, waist $= 95$ μm) is switched on for 500 μs before the microwave pulse to allow the intensity of the trapping light to stabilize. The laser polarization is tunable to a precision of $\pm 1°$ by a $\lambda/2$ wave plate. The molecules experience a reasonably homogeneous laser intensity within 2% of the peak intensity. With the molecules initially in the spin-stretched rotational and hyperfine ground state ($N=0, M_N=0, m_I^{Rb}=3/2, m_I^{Cs}=7/2$), we pulse on the microwave field for a time (t_{pulse}) that is less than the duration of a π pulse for the relevant transition. We measure the number of molecules remaining in the ground

state by reversing the STIRAP sequence to dissociate the molecules and using absorption imaging to detect the resulting atoms. We observe the microwave transition as an apparent loss of ground-state molecules. All the features we measure are Fourier-transform limited, with widths proportional to $1/t_{pulse}$. The microwave power is tuned to allow pulse times of 100–180 μs, yielding Lorentzian lines with 5–10 kHz full width at half maximum. We observe transitions due to microwaves polarized along the z and y axes.

We begin by setting the laser polarization perpendicular to the magnetic field, i.e. $\beta = 90°$. Figure 1 shows the measured transition frequencies for $B_z = 181.5$ G as a function of laser intensity. These are superimposed on calculations of the 96 hyperfine levels of $N = 1$, using molecular constants fitted to the experiments as described below. Calculated transition strengths are indicated with blue and red shading for microwaves polarized along z and y, respectively. Many avoided crossings appear in the region where $I\alpha^{(2)}$ is comparable to the hyperfine couplings and Zeeman splittings. The basis functions that carry the spectroscopic intensity cut through the manifold of states, resulting in a complicated variation in transition strengths as each state brightens and fades. At sufficiently high laser intensities, the ac Stark effect dominates the Zeeman splittings; N eventually requantizes along the laser polarization axis, and the pattern of transition strengths and frequencies simplifies.

Figure 2 shows analogous results for the laser polarization parallel to the applied magnetic field, $\beta = 0°$. A single beam is used at low intensities, as in Fig. 1. At the highest intensities shown, however, two beams are used to form a crossed optical dipole trap. The beams propagate in the xy plane and cross at an angle of $\sim 27°$. In this case M_F is a good quantum number
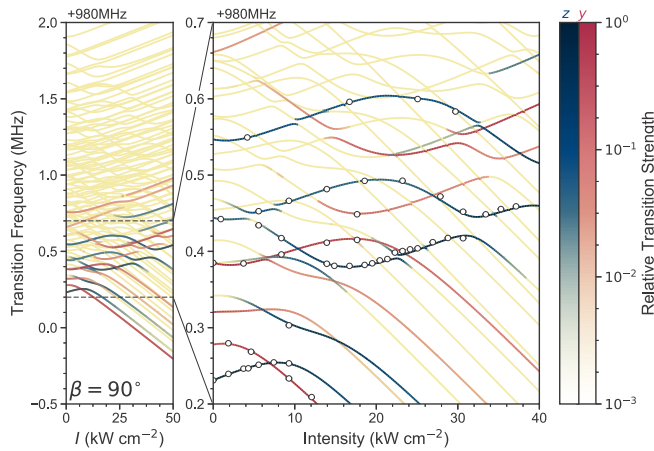
FIG. 1. Transition frequencies from the lowest-energy hyperfine state of the rovibronic ground state $N = 0, M_F = 5$ to the 96 hyperfine states for $N = 1$ as a function of laser intensity, for laser polarization perpendicular to the magnetic field, $\beta = 90°$. The relative transition strengths for microwaves polarized along z and y and are shown as blue and red color maps respectively. The data points show experimental microwave frequencies.

even in the presence of the trapping laser. The three $M_F = 5$ hyperfine states for $N = 1$ (blue) diverge as a function of laser intensity, and there are no avoided crossings between them. At high intensity M_N becomes an increasingly good quantum number, and the two states with $M_N = \pm 1$ lose intensity for microwaves polarized along z. Nevertheless, strong avoided crossings still exist where states with the same M_F cross.

The experimental uncertainties are not visible on the scale presented in Figs. 1 and 2. The statistical uncertainties in the transition frequencies are typically ± 0.5 kHz. The dominant uncertainty in the laser intensity on the other hand is systematic and due to the uncertainty in the beam waist at the position of the molecules. We estimate this uncertainty to be $\pm 3\%$ of the peak intensity.
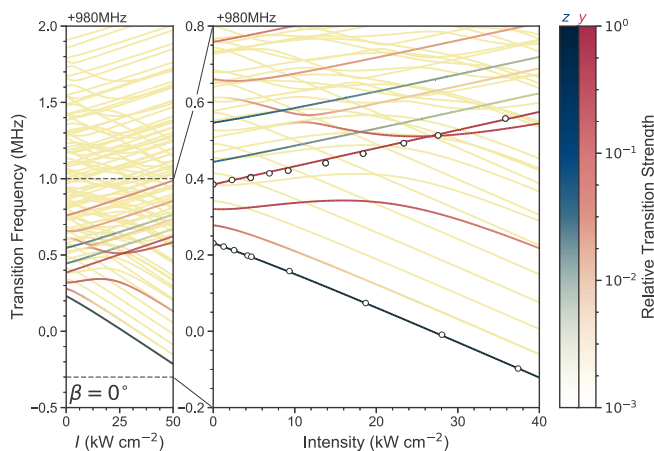

FIG. 2. Transition frequencies from $N = 0, M_F = 5$ to $N = 1$ as a function of laser intensity, for laser polarization parallel to the magnetic field, $\beta = 0°$. The relative transition strengths are coded as in Fig. 1. The data points show experimental microwave frequencies.
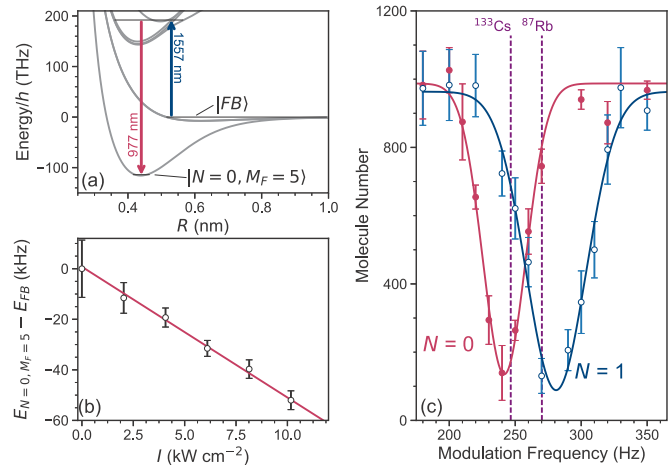

FIG. 3. Measurement of the isotropic component $\alpha^{(0)}$. (a) Transitions used in STIRAP of molecules to the lowest-energy hyperfine state of the rovibronic ground state. (b) ac Stark shift of the STIRAP dark-state resonance. (c) Parametric heating measurements of trap frequency for molecules in the lowest-energy hyperfine states of $N = 0$ (red closed circles) and $N = 1$ (blue open circles) in traps with the same peak laser intensity. The dotted lines show center frequencies for ^{87}Rb and ^{133}Cs atoms.

We have fitted the experimental results of Figs. 1 and 2 independently to obtain $\alpha^{(2)}$, while holding the hyperfine constants fixed at the values of Ref. [19]. For laser polarizations $\beta = 0°$ and $90°$, we obtain values of $\alpha^{(2)}_{\beta=0°}/4\pi\epsilon_0 = 507(1)\, a_0^3$ and $\alpha^{(2)}_{\beta=90°}/4\pi\epsilon_0 = 602(2)\, a_0^3$ respectively. The uncertainties given are the statistical uncertainties found during fitting, but both results are subject to the same systematic uncertainty in intensity described above. Despite the difference between the two fitted values, the theory in each individual case describes the observed ac Stark shift well.

To characterize the polarizability of the molecule fully, we must also measure the *isotropic* component $\alpha^{(0)}$. To do this, we perform several complementary measurements. First, we observe the energy shift of $N = 0, M_F = 5$ with respect to an initial weakly bound Feshbach state. This is given by the ac Stark shift of the two-photon transition used in STIRAP [32], shown in Figs. 3(a) and 3(b). This energy shift gives the difference in polarizability between the two molecular states, and the polarizability of the Feshbach state is simply the sum of the polarizabilities of the constituent atoms, which are well known [33].

In addition to this spectroscopic method, we perform parametric heating on the molecular sample. Here, we retrap $N = 0$ or $N = 1$ molecules in their lowest hyperfine state with two beams with $\beta = 0°$ and total intensity $I = 36$ kW cm^{-2}. The intensity of one of the beams is then modulated sinusoidally by $\pm 20\%$ for 1 s. When the modulation frequency is twice the trapping frequency, we resonantly heat the molecules and observe evaporative loss from the trap as shown in Fig. 3(c). If the ac Stark shift is linear, the trap frequency ω is proportional to $\sqrt{\alpha/m}$, where m is the mass. We compare the trap frequency for molecules with those for ^{87}Rb and ^{133}Cs atoms in a dipole trap of the same intensity [dashed lines in Fig. 3(c)] to find the absolute polarizabilities
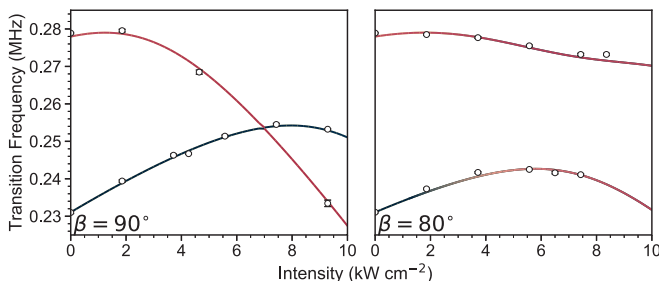
FIG. 4. Transition frequencies from $N = 0, M_F = 5$ to the two lowest energy hyperfine states of $N = 1$ for $\beta = 90°$ and $\sim 80°$. Transition strengths are coded as in Figs. 1 and 2.

of the molecules in both states. The value for $N = 1$ is corrected for the contribution of $\alpha^{(2)}$ obtained above for $\beta = 0°$. The two parametric heating results agree with one another and with the spectroscopic result within experimental uncertainty. We find an uncertainty-weighted average value $\alpha^{(0)}/4\pi\epsilon_0 = 8.8(1) \times 10^2 a_0^3$, in reasonable agreement with theoretical predictions [20,34,35]. Note that the parametric heating approach does not require knowledge of the absolute intensity of the trapping beams and thus gives a smaller uncertainty in $\alpha^{(0)}$.

The avoided crossings between laser-dressed levels can cause trap loss if the molecules undergo Landau-Zener transitions to different hyperfine states as they move around the trap or if the intensity of the dipole trap is changed dynamically. This is particularly important when retrapping molecules that have been manipulated in free space. Since in our experiment trapping requires a minimum laser intensity around 15 kW cm^{-2}, such losses occur for example when attempting to retrap the spin-stretched $N = 1, M_F = 6$ state with $\beta = 90°$. Avoided crossings will also produce anharmonic and anisotropic trapping potentials, which may result in complicated density profiles for molecular clouds and cause coupling between translational and rotational degrees of freedom in optical lattices. The strengths of avoided crossings may be tuned by varying β; for example, Fig. 4 shows how the avoided crossing between the two lowest-energy hyperfine levels of $N = 1$ varies due to a change in laser polarization of approximately 10°. Understanding the avoided crossings will allow us to identify optimum laser intensities and polarizations for optical trapping. Furthermore, given sufficient broadening of the avoided crossing and precise control of the laser

intensity, it may be possible to traverse the avoided crossings in a controlled manner during the retrapping of molecules. This may allow access to hyperfine states that are not easily produced with microwave transfer.

A further consequence of the ac Stark effect is that the frequencies of microwave transitions depend on the position within an optical trap. This has important ramifications for the design of experiments to achieve coherent control of trapped molecules. Neyenhuis *et al.* demonstrated coherence times in Ramsey interferometry up to 1.5 ms in ^{40}K^{87}Rb by optimizing a "magic angle" between the magnetic field and the laser polarization [21]. To achieve longer coherence times, it is desirable to find excited states that are parallel to the ground state as a function of laser intensity. This condition is met at the turning point of an avoided crossing. This will make it possible to achieve longer coherence times by controlling laser intensity as well as polarization.

In summary, we have completely characterized the anisotropic polarizability of ^{87}Rb^{133}Cs for $\lambda = 1550$ nm. We have measured microwave spectra of several hyperfine components of the the $N = 0 \rightarrow 1$ microwave transition as a function of laser intensity and used them to extract precise values of the anisotropic component $\alpha^{(2)}$ of the molecular polarizability. We have supplemented this with parametric heating and spectroscopic measurements to determine the isotropic component $\alpha^{(0)}$. We have discovered a subtle interplay between the ac Stark effect and the hyperfine structure, which produces a rich and complex pattern of avoided crossings between levels as a function of laser intensity and polarization. Understanding this pattern has allowed us to trap molecules in well-defined hyperfine states and control their polarizability. This lays the foundations for enhanced coherent microwave control of the internal state of polar molecules confined in optical traps and lattices, which will underpin many exciting proposed applications of ultracold molecules.

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