State-Selective Detection of Near-Dissociation Ultracold KRb
$X^1\Sigma^+$ and a $3\Sigma^+$ Molecules

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We report on the state-selective detection of near-dissociation ultracold KRb molecules in the ground $X^1\Sigma^+$ state and the metastable $a^3\Sigma^+$ state. The molecules are produced by photoassociation of ultracold atoms followed by radiative decay into high vibrational levels of the $X$ and $a$ states. Detection utilizes resonance-enhanced one-color two-photon ionization, followed by time-of-flight mass spectroscopy. Scanning the detection laser frequency over the range 582-625 nm, we observe transitions from the $v''=86-92$ levels of the $X$ state, which are bound by up to 30 cm$^{-1}$, and the $v''=17-23$ levels of the $a$ state, which are also bound by up to 30 cm$^{-1}$. The measured vibrational spacings are in excellent agreement with those previously measured and those calculated from the relevant potential curves. Relative vibrational populations are also consistent with Franck-Condon factors for decay from the photoassociated levels.

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I. INTRODUCTION

Following in the footsteps of ultracold atoms, the area of ultracold molecules has developed rapidly in recent years [1, 2, 3]. Polar molecules, with their permanent electric dipole moments, have drawn particular attention [4]. The dipole moments of these heteronuclear molecules allow manipulation with applied electric fields. Because the dipole-dipole potential is both long-range and anisotropic, interactions between polar molecules are fundamentally different from those in homonuclear systems. Potential applications include novel quantum degenerate systems [5, 6, 7, 8, 9, 10, 11], quantum computation [12], and tests of fundamental symmetries [13]. Investigations of collisions and reactions at these extremely low temperatures also promise to open new areas of ultracold chemistry [14, 15, 16].

The technique of ultracold atom photoassociation (PA) [17] has been the primary means for producing molecules at translational temperatures below 1 mK [2]. In this process, laser light resonantly binds two colliding atoms into an excited molecule which subsequently decays by spontaneous emission. This method has recently been successfully adapted to heteronuclear systems [18, 19, 20, 21, 22, 23, 24, 25, 26]. Feshbach resonances have been used to make homonuclear molecules at even lower temperatures [27, 28, 29, 30, 31, 32, 33, 34, 35, 36], in some cases under quantum degenerate conditions. Based on recent observations of Feshbach resonances in homonuclear systems [37, 38], it can be expected that this route will also yield ultracold heteronuclear molecules in due course. Both PA and Feshbach resonances tend to produce molecules in high vibrational levels with outer turning points at long range. Although these high-$v''$ states are of interest for some studies, many applications require low-$v''$ states, because of their improved stability against inelastic processes and their larger dipole moments. Various schemes for populating low-$v''$ states have been proposed [3, 12, 21, 39, 40, 41], and some successful implementations have been reported [19, 42].

Direct detection of ultracold molecules has generally utilized photoionization combined with time-of-flight mass spectroscopy. State selectivity is essential to many applications and for diagnosing transfer of population. For example, measuring the $v''$ dependence of vibrational quenching rates due to collisions with ultracold atoms would be an important first step in studying ultracold molecule collisions. To date, the ability to unambiguously identify the vibrational state of ultracold molecules has been limited, particularly for levels near dissociation. Two-photon one-color ionization spectra have been reported in Rb$_2$ [43, 44, 45], but definitive assignments and initial state identifications were not made. We have recently demonstrated state-selective detection of Rb$_2$, details of which will be reported elsewhere [46]. In Cs$_2$, ultracold molecule detection spectra have been compared to absorption measurements, revealing the role of the diffuse bands in the detection process [47]. In K$_2$, two-photon two-color ionization spectra enabled some identification of the low-$v''$ states produced by both one-photon [48] and two-photon [42] PA. Dissociation with a separate cw laser helped to clarify the assignment [42]. In Na$_2$, high-resolution cw ionization spectroscopy yielded clear identification of the highest-lying bound and quasibound rovibrational-hyperfine states [49].

To date, the only heteronuclear system in which state selectivity has been achieved is RbCs. Using two-photon two-color ionization, ultracold molecules were detected in specific vibrational levels of the $a^3\Sigma^+$ state [18, 20]. In addition, these molecules have been transferred by stimulated emission pumping to the $X^1\Sigma^+$ state ($v''=0,1$) and detected in these states with vibrational selectivity [19].

In the present work, we report on vibrationally state-selective detection of ultracold KRb molecules in high-$v''$ levels of both the ground $X^1\Sigma^+$ state and the metastable $a^3\Sigma^+$ state. These molecules are formed by cold-atom photoassociation followed by radiative decay, as shown in Fig. 1. Two-photon one-color ionization proceeds...
through resonant intermediate levels of the $4^1\Sigma^+$, $5^1\Sigma^+$, $4^3\Sigma^+$, and $3^3\Pi$ states, allowing vibrational state identification and determination of the relative populations. Using wavelengths in the range of 582-625 nm, we have observed vibrational levels $v''=86-92$ for the $X$ state and $v''=17-23$ for the $a$ state. We have also analyzed spectroscopy of the excited states, details of which will be reported elsewhere.

II. EXPERIMENT

Details of the experimental setup have been described previously. Here we recount it briefly, focusing on the ionization detection. The KRb PA takes place in overlapping clouds of ultracold $^{39}$K and $^{85}$Rb. High atomic densities, estimated at $3 \times 10^{10}$ cm$^{-3}$ for K and $1 \times 10^{13}$ cm$^{-3}$ for Rb, are achieved using “dark-SPOT” MOTs for each species. Temperatures for K and Rb of 300 $\mu$K and 100 $\mu$K, respectively, are expected.

The PA process is driven with a cw tunable titanium-sapphire laser (Coherent 899-29). Its output, typically $>400$ mW, is focused into the overlapping MOT clouds. PA spectra, as described previously, are obtained by scanning this laser and measuring the ionization signal from molecules which have radiatively decayed into the $X^1\Sigma^+$ ground state and the metastable $a^3\Sigma^+$ state.

In our earlier work, the ionization detection used a pulsed laser with a broader linewidth and significant amplified spontaneous emission (ASE). This prevented state-selective detection but had the benefit of yielding at least some ion signal at most wavelengths, thereby facilitating the location of PA resonances.

In the present work, ionization detection is achieved with a pulsed dye laser (Continuum ND6000) pumped by a frequency-doubled Nd:YAG laser at a 10 Hz repetition rate. The use of two dyes, Rh610 and DCM, provides spectral coverage over the range 582 nm to 625 nm. The 0.05 cm$^{-1}$ linewidth of the dye laser is sufficient to resolve the vibrational structure, but not the rotational structure, of transitions from levels of the $X^1\Sigma^+$ and $a^3\Sigma^+$ states. This detection laser, with a pulse width of 7 ns and a typical output power of 3 mJ, is focused to a diameter of 1 mm. This is significantly larger than the 0.3 mm diameter of the MOT clouds in order to illuminate a larger fraction of the ballistically expanding cloud of cold molecules. Ions from the laser pulse are accelerated to a channeltron ion detector. KRb$^+$ is discriminated from other species ($K^+$, Rb$^+$, Rb$_2^+$) by its time of flight (TOF).

III. DETECTION SPECTRA FOR SINGLET MOLECULES

Detection spectra are obtained by fixing the PA frequency on a resonance and recording the KRb$^+$ ion signal while scanning the pulsed laser. A 400 cm$^{-1}$ scan for $X^1\Sigma^+$ state molecules is shown in Fig. 2. The spectra display structure on two scales. On a gross scale ($\sim 20$ cm$^{-1}$), nearly periodic spacings correspond to vibrational levels of the upper state of the detection transition, specifically the $4^3\Sigma^+$ state for the spectrum shown in Fig. 2. The spectroscopy of this upper state will be described separately. Measured level spacings and the range of levels observed are both in very good agreement with calculations based on ab initio potentials. We note that the increased complexity of the spectrum at higher frequencies is likely due to the appearance of transitions to the $5^3\Sigma^+$ state, which are not yet assigned.

On a finer scale ($\sim 5$ cm$^{-1}$), the structure corresponds to the spacings between high-lying vibrational levels of the $X^1\Sigma^+$ ground state. Specific X-state levels are assigned by matching the measured spacings with those calculated from the potential. Examples are shown in Fig. 3 for PA to two different vibrational levels of the $3(0^+)$ state. As can be seen, the agreement between measured and predicted vibrational spacings is excellent, allowing unambiguous identification of the ground-state levels. A previously measured spacing between $v''=86$
Each group of lines is labeled according to the vibration of the molecules. The KRb molecule is aligned and excited to a 3(0) state at 12569.94 cm$^{-1}$, which corresponds to a detuning of $-246.66$ cm$^{-1}$ below the K(4S) + Rb(5P$_{3/2}$) asymptote.

The blue end of the spectrum becomes congested and is not yet assigned.

FIG. 3: (Color online) An expanded view of the $v'' = 45$ group of lines in the singlet detection spectrum for two different 3(0) PA detunings: (a) $-246.66$ cm$^{-1}$ and (b) $-307.69$ cm$^{-1}$. The calculated X-state level spacings (dashed vertical lines) are superimposed. Most unlabelled lines belong to adjacent $v'$ groups. Solid circles indicate the calculated FCF's for decay from 3(0) to $X^1\Sigma^+$ for these two cases. The maximum FCF value in (a) is 0.11 and the maximum in (b) is 0.12. Note the shift of the FCF peak to lower $v''$ (smaller outer turning point) with larger PA detuning.

IV. DETECTION SPECTRA FOR TRIPLET MOLECULES

State-selective detection of triplet $a^3\Sigma^+$ molecules is carried out in a similar manner to that described above for singlet $X^1\Sigma^+$ molecules. The only difference is that the PA laser is tuned to a level which decays to the $a$ state. For this work, we use the 3(0) state from the K(4S) + Rb(5P$_{3/2}$) asymptote, which correlates to the $1^3\Pi$ state at short range. Higher detection laser intensities are required for comparable signal levels from
triplet molecules. The triplet detection spectra are obtained over the same wavelength range, but of course involve different upper states: $4\Sigma^+$ and probably $3\Pi$. The spectroscopy of the $4\Sigma^+$ state, along with that of the $4\Pi$ state (used for singlet detection), will be described in a separate publication [50]. A ~400 cm$^{-1}$ scan is shown in Fig. 4. The structure repeating at ~50 cm$^{-1}$ corresponds to the vibrational spacing of the $4\Sigma^+$ state. Assigned vibrational levels are indicated in the figure. The congested region of larger signal to the blue may involve the $3\Pi$ state, but specific assignments have not yet been made.

Two-photon transitions to atomic Rb Rydberg states sometimes can be seen as well in the triplet spectra, due to leakage from the space-charge-broadened Rb$^+$ TOF peak into the KRb$^+$ TOF peak. We also see ions at the Rb atomic 5P$_{3/2}$→5f one-photon transition. The 5P$_{3/2}$ level is populated by the MOT lasers and this dipole-forbidden transition is enabled by the ~160 V/cm electric field used for extraction. These atomic lines serve as useful frequency markers and verify that the laser frequency measurements are accurate to 0.16 cm$^{-1}$. These lines are not readily observable in the spectra of singlet molecules (Fig. 2) because lower laser intensities are used.

Fig. 5 is an expanded view of the spectrum, encompassing only one vibrational level of the $4\Sigma^+$ upper state. The structure here is due to the near-dissociation levels of the $a^3\Sigma^+$ state. The vibrational levels show spacings of 2.4 to 5.6 cm$^{-1}$. Such spacings correspond to levels $\nu'$=20 to 26 in the ab initio potential of Kotochigova et al. [55] and $\nu'$=18 to 24 in the ab initio potentials of Park et al. [56] and of Rousseau et al. [51]. These three potentials have been compared by Zemke et al. [57]. However, a definitive assignment has very recently become available, based on new Fourier Transform Spectra in Hannover [58]. This clearly indicates we have observed levels $\nu''$=17 to 23, as shown in Table 1. We also plan to calculate improved Franck-Condon factors for the Hannover $a^3\Sigma^+$ potential once it is available. The FCFs in Fig. 5 are based on the Kotochigova et al. [55] and $\nu''$=18 to 24 in the ab initio potential once it is available. The FCFs in Fig. 5 are based on the Kotochigova et al. potential [55] for levels 20 to 26. Finally, we plan direct measurement of the binding energies by scanning a separate cw laser to deplete the ground-state levels. We have recently observed this “ion dip” spectroscopy for $X^1\Sigma^+$ $\nu''=89$.

Table I. Level spacings ($\Delta G_{\nu+1/2}$, in cm$^{-1}$) in the $39^K^8$Rb $a^3\Sigma^+$ state

| $\nu$ | Fourier Transform Spectra | PA-REMPI(This work) |
|-------|-----------------------------|----------------------|
| 17    | 5.49                        | 5.60                 |
| 18    | 4.84                        | 4.90                 |
| 19    | 4.23                        | 4.23                 |
| 20    | 3.60                        | 3.56                 |
| 21    | 2.99                        | 2.96                 |
| 22    | 2.41                        | 2.38                 |

FIG. 4: (Color online) Detection spectrum for triplet molecules. The PA laser is tuned to a 3(0$^-$) $(J=1)$ level at 12493.91 cm$^{-1}$, which corresponds to a detuning of -322.69 cm$^{-1}$ below the K(4S)+Rb(5P$_{3/2}$) asymptote. Each group of lines is labeled according to the $v'$ level of the $4\Sigma^+$ state which is excited. The blue end of the spectrum becomes congested due in part to the emergence of transitions to the $3\Pi$ state, which have not yet been assigned. The triangle indicates a one-photon atomic transition from Rb(5P$_{3/2}$)→(5f). Asterisks indicate two-photon transitions from Rb(5s) to atomic Rydberg states: 13d, 14d, 15d, 16d from left to right.

FIG. 5: (Color online) An expanded view of the $v'=9$ group of lines in the triplet detection spectrum for two different 3(0$^-$) PA detunings: (a) −296.21 cm$^{-1}$ and (b) −322.69 cm$^{-1}$. The calculated $a$ state level spacings (dashed vertical lines) are superimposed. Solid circles indicate the calculated FCF’s for decay from the 3(0$^-$) state to the $a^3\Sigma^+$ state. The maximum FCF value in (a) is 0.10 ($\nu''=20$) and the maximum in (b) is 0.10 ($\nu''=19$). Asterisks indicate the two-photon atomic transition: Rb(5s)→(13d).
As for the singlet molecules, we can use the peak heights as a measure of the lower level ($a^3\Sigma^+$) populations. There is a small, but noticeable shift in the distribution to lower $v''$ for larger (negative) PA detunings. Also shown in these figures are the calculated FCF’s for decay from each $3(0^-)$ PA level to various levels of the $a^3\Sigma^+$ state. Calculations using the LEVEL program reproduce the overall locations of the distributions, including the shift with PA detuning. However, compared to the singlet spectra (Fig. 3), individual peak heights are not as accurately predicted. We hope future calculations based on the Hannover $a^3\Sigma^+$ potential, once it is available, can give more accurate results. Linewidths in the triplet spectra are somewhat broader than those in the singlet spectra. Power broadening, spin-spin, second-order spin-orbit, rotational and hyperfine structure should all contribute to the line shapes observed.

PA detunings for the $3(0^-)$ state from 244 cm$^{-1}$ to 323 cm$^{-1}$ (measured relative to its K(4P) + Rb(5P$_{3/2}$) asymptote) have been used to observe $a$-state levels from $v''=17$-23. These have binding energies from 29.02 cm$^{-1}$ to 5.31 cm$^{-1}$, respectively. This numbering, based on the Fourier Transform Spectra in Hannover, is definitive because vibrational assignment is based on two different isopotes. None of the three sets of $ab~initio$ potentials from 54, 55, 56 can give this exact numbering, although a good vibrational spacing match can be found if we adjust their numbering by one or three 57.

An important difference between singlet and triplet molecules is that the triplets have a non-zero magnetic moment and can therefore be magnetically trapped. We have previously demonstrated this trapping in the quadrupole magnetic field of the MOT 25. 26 by delaying the molecule detection with respect to the turn-off of the PA laser. This difference in magnetic properties could be utilized as a singlet/triplet “filter” to distinguish the two types of molecules. We do see cases where both $X^1\Sigma^+$ and $a^3\Sigma^+$ molecules appear in the same region of the detection spectrum. An example, using PA to the $3(0^+)$ state, is shown in Fig. 6. At long range, the $3(0^-)$ state should decay to both $X^1\Sigma^+$ and $a^3\Sigma^+$ states 27, so we expect to detect both. However, the FCF’s for the first step of the detection process play an important role. For high-$v''$ levels of $X^1\Sigma^+$, the overlap with $a^3\Sigma^+$ levels comes primarily from the outer turning point. On the other hand, overlap of high-$v''$ levels of $a^3\Sigma^+$ with $a^3\Sigma^+$ levels comes primarily from the inner turning point and becomes more favorable in the lower energy region of the spectrum. The overall detection efficiencies (including the ionization step) for $X^1\Sigma^+$ and $a^3\Sigma^+$ become comparable in the region shown in Fig. 6. Although we do see triplet features in a primarily singlet detection spectrum (using PA to $3(0^+)$), we do not see singlet features in the triplet detection spectra (using PA to $3(0^-)$). This is consistent with the fact that at long range, $3(0^+)$ can decay to both to both $X^1\Sigma^+$ and $a^3\Sigma^+$ states, while $3(0^-)$ can decay only to $a^3\Sigma^+$ 21.

V. CONCLUSIONS

In summary, we have realized vibrationally state-selective detection of near-dissociation levels of ultracold KRB molecules in the $X^1\Sigma^+$ ground state and the $a^3\Sigma^+$ metastable state. This state-selectivity will be crucial to future experiments in ultracold molecular collisions and reactions where specific initial and final states must be measured. This capability is equally important in diagnosing population transfer, e.g., from high-$v''$ to low-$v''$ 14. In fact, the first step in our singlet detection ($4^3\Sigma^+ \leftarrow X^1\Sigma^+$) is a good candidate for realizing this type of transfer. As an example, if we start in $X^1\Sigma^+$ ($v''=89$), the FCF for excitation to $4^3\Sigma^+$ ($v''=40$) is quite large ($0.02$ due to overlap at the outer turning point. On the other hand, overlap at the inner turning points gives a favorable FCF of $\sim0.01$ for decay (or stimulated emission) of $4^3\Sigma^+$ ($v''=40$) to the absolute ground state, $X^1\Sigma^+$ ($v''=0$). These large FCF’s indicate that coherent two-photon transfer, such as STIRAP 55, should be feasible with narrow-linewidth quasi-cw lasers.

The two-photon, one-color detection we have employed is particularly convenient because only one tunable laser is required. However, the second (ionizing) step generally requires high intensity, resulting in power broadening of the first (bound-bound) step. Two-photon, two-color detection (e.g., through states from the K(4s)+Rb(5p) asymptotes) may offer some benefits. The ionizing step can be driven with high intensity from a fixed-frequency...
pulsed laser, such as a frequency-doubled YAG laser. If the first step is driven with a narrow-linewidth cw laser at low intensity, rotational resolution should be achievable.

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