Quantum state preparation and control of single molecular ions

D Leibfried

Time and Frequency Division, National Institute of Standards and Technology, 325 Broadway, Boulder, CO 80305, USA
E-mail: dil@boulder.nist.gov

Abstract. Preparing molecules at rest and in a very pure quantum state is a long-standing aspiration in chemistry and physics, so far achieved only for a select set of molecules in dedicated experimental setups. Here, a quantum-limited combination of mass spectrometry and Raman spectroscopy is proposed that should be applicable to a wide range of molecular ions. Excitation of electrons in the molecule followed by uncontrolled decay and branching into several lower-energy states is avoided. Instead, the molecule is always confined to rotational states within the electronic and vibrational ground-state manifold, while a co-trapped atomic ion provides efficient entropy removal and allows for extraction of information about the molecule. The outlined techniques might enable the preparation, manipulation and measurement of a multitude of molecular ion species with the same instrument, with applications including, but not limited to, precise determination of molecular properties and fundamental tests of physics.

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1. Introduction

Over the last 30 years, laser cooling [1, 2] and laser spectroscopy have led to spectacular developments in atomic physics. The electromagnetic vacuum of infrared to ultraviolet light provides a near-perfect entropy sink and allows us to optically pump and cool atoms if sufficiently closed cycles of electronic excitation and subsequent decay are realized. Entropy can be removed from the electronic state and the translational motion to where these degrees of freedom are in a very pure quantum state. Such a quantum state preparation is available for a sizeable number of neutral and charged atoms, but in most molecules the decay of excited electrons branches into a multitude of levels, and this prevents efficient entropy removal in all but a few cases. These exceptions include cooling molecules by buffer gas [3], exploiting fortuitous level structures that allow for closed cycles with a manageable number of laser sources [4–6], selective ionization and charge transfer [7] and assembling molecules from ultracold atoms [8–11]. In all these cases, the molecular states are destroyed or the molecules leave the interaction region during the process of their detection. Methods of preparing ensembles of specific molecules in their absolute ground state by pulsed adiabatic passage [12–14] or by collisions with a laser-cooled buffer gas [15] have also been envisioned. Some of the methods employed in the proposal here, namely preparation of the rotational ground state of a specific single molecular ion based on quantum logic spectroscopy [16], have been discussed in [17, 18].

2. Translational cooling of molecular ions

In the method proposed here, the molecular ion of interest is co-trapped with an atomic ion that can be manipulated precisely by laser and microwave fields [19]. Due to the Coulomb coupling between the co-trapped ions, laser cooling of the atomic ion will sympathetically cool the translational motion of the molecular ion as well. Typically, excited electronic and vibrational states of the molecular ion will decay to their ground states within nanoseconds

and milliseconds, respectively, and the rotational state distribution will equilibrate to the black-body temperature of the trap within a few seconds [5, 6]. After this initial equilibration, the molecule ideally never leaves the electronic and vibrational ground-state manifold. The mass of the molecular ion can be inferred quite precisely by determining the normal-mode frequencies of motion of the co-trapped ions [20, 21]. This can be accomplished by sweeping the frequency of an electronic drive signal that is applied to a trap electrode [20]. If the drive frequency coincides with a normal-mode frequency, the crystal gets excited and the laser-induced fluorescence changes. If the mass of the atomic ion m_a is known and has frequency ω_a in the potential well, the two-ion crystal with mass ratio $\mu = m_m/m_a \geqslant 1$ will exhibit two axial modes, with frequencies [22]

$$\omega_\pm = \omega_a \sqrt{\left(1 + \frac{1}{\mu} \pm \sqrt{1 + \frac{1}{\mu^2} - \frac{1}{\mu}}\right)} \tag{1}$$

and eigenvectors

$$v_\pm = \frac{1}{\sqrt{\left(1 - \mu \mp \sqrt{\mu^2 - \mu + 1}\right)^2 + 1}} \left\{1 - \mu \mp \sqrt{\mu^2 - \mu + 1}, 1\right\}, \tag{2}$$

where the first coordinate is of the the lighter mass atomic ion. If the molecular mass m_m is smaller than m_a, similar relations can be derived. From the measured frequencies ω_a and $\omega_\pm$, which can be determined with better than 10^{-4} relative accuracy [21], we can infer m_m:

$$m_m = m_a \frac{2\omega_a^2}{\omega_+^2 + \omega_-^2 - 2\omega_a^2}. \tag{3}$$

For $\mu \gg 1$, ω_+ goes to $\sqrt{2}\omega_a$, while ω_- goes to zero. In the same limit the normal-mode amplitudes of the ions go to $\{-1, 0\}$ in the ω_+-mode and to $\{1/\sqrt{5}, 2/\sqrt{5}\}$ for the ω_--mode. Cooling of both modes will become more of a challenge with larger mass differences: for the ω_+-mode, the participation of the heavier ion tends to vanish, while the ω_--mode will go to low frequencies, where resolving sidebands becomes harder and ambient heating mechanisms are more detrimental [19].

For not too different masses, it should be possible to cool the normal modes of motion of both ions to the ground state, analogous to ground-state cooling demonstrated with different species of atomic ions [16, 23], which has been implemented for a range of masses $1/3 \leqslant m_a/m_{a'} \leqslant 2$. Outside this range, ground-state cooling becomes less effective, but might still be feasible. Suitable atomic ions range from 9 atomic mass units (^{9}Be$^+$) to 176 atomic mass units (^{176}Yb$^+$). Only one of the normal modes is required to be in the ground state for what follows. At this point the molecule is ideally in the electronic, vibrational and translational ground states, leaving only the rotational and, if the molecule has hyperfine structure, the hyperfine states undetermined.

3. Rotational spectroscopy and cooling

Manipulation of rotational states, ideally without altering the vibrational and the electronic state, might proceed by combining Raman transitions [24] driven by optical frequency combs [25] with a technique similar to what has been used for state preparation and readout in two-ion

quantum logic clocks [16]. In this way, entropy can be removed from the molecular rotational degree of freedom by indirectly coupling it to the near-perfect reservoir of a light field. This can be accomplished via closed-cycle electronic transitions of the co-trapped atomic ion. Raman transitions are a near-universal tool in molecular spectroscopy, because they can be driven with light sources that are not specific to the molecule under study. Direct electronic transitions in molecular ions typically require wavelengths shorter than 650 nm [26], below the central wavelengths of common femtosecond laser frequency combs (mostly 800 or 1064 nm) [25]. Two comb beams can be produced either by splitting the output of one laser and frequency shifting one part [27] or by using two separate combs with equal mode spacing and scanning the carrier-envelope phase of one or both. If the combs span about 30 THz, they are much broader than the transition frequencies between the highest-lying thermally occupied rotational states that need to be addressed (typically below 300 K $k_B/h \approx 6.25$ THz, where k_B and $\hbar$ are Boltzmann's constant and Planck's constant, respectively [26]). In that case, almost every comb-tooth in one laser beam can have a corresponding tooth in the other laser beam such that the pair is in Raman resonance with a certain rotational transition. All pairs are in resonance simultaneously and their effect adds up, leading to an interaction that is similar in strength to a pair of continuous-wave lasers with the same average power [27]. The two comb fields should counter-propagate along the ion crystal axis for maximal momentum transfer in transitions that change the state of one normal mode of the ion crystal while simultaneously driving a rotational state changing transition in the molecule (see figure 1).

Analogous processes have been utilized in experiments with a laser comb and two identical atomic ions, where an approximately 12.7 GHz hyperfine Raman transition was driven simultaneously and coherently with the ion crystal motion [27]. Raman transitions of the molecule require that the resonance condition

$$N\omega_m + \Delta\omega_o = \Omega_{J,J\pm1,2} + n\,\omega_\pm \tag{4}$$

is fulfilled. Here N and n are integers, ω_m is the mode spacing of both comb fields, set to be identical, and $\Delta\omega_o$ is the frequency shift between the two comb fields due to their individual carrier-envelope phases (or due to the frequency shift of one part of the output). The frequency difference between two rotational levels with quantum numbers J and $J\pm1,2$ is denoted by $\Omega_{J,J\pm1,2}$ (for linear and spherical-top molecules, $J \to J\pm1$ is forbidden by Raman selection rules) and $\omega_\pm$ is either of the normal mode frequencies of the two-ion crystal. Transitions with $n=0$ ('carrier') will have the largest transition rate, while the nth 'sideband' will be suppressed by the nth power of the Lamb–Dicke parameter. For Raman transitions induced by two counter-propagating beams with wave-vector modulus k_l, the Lamb–Dicke parameters of the molecular ion are given by [22]

$$\eta_m^{(\pm)} = 2|k_l|v_\pm^{(2)}\sqrt{\frac{\hbar}{2m_m\omega_\pm}}. \tag{5}$$

At the mode frequencies commonly used in resolved sideband cooling [19], the ground-state wavefunction extension of the molecule in either normal mode (typically a few nanometers) is much smaller than the light wavelength λ, so higher-order sidebands are largely suppressed. The first sidebands ($n = \pm1$) are the strongest motion-altering interaction and change the motional state by one quantum.

For a very general rough estimate of achievable Raman rates, we can assume that the intermediate level in the Raman transition has a dipole matrix element of
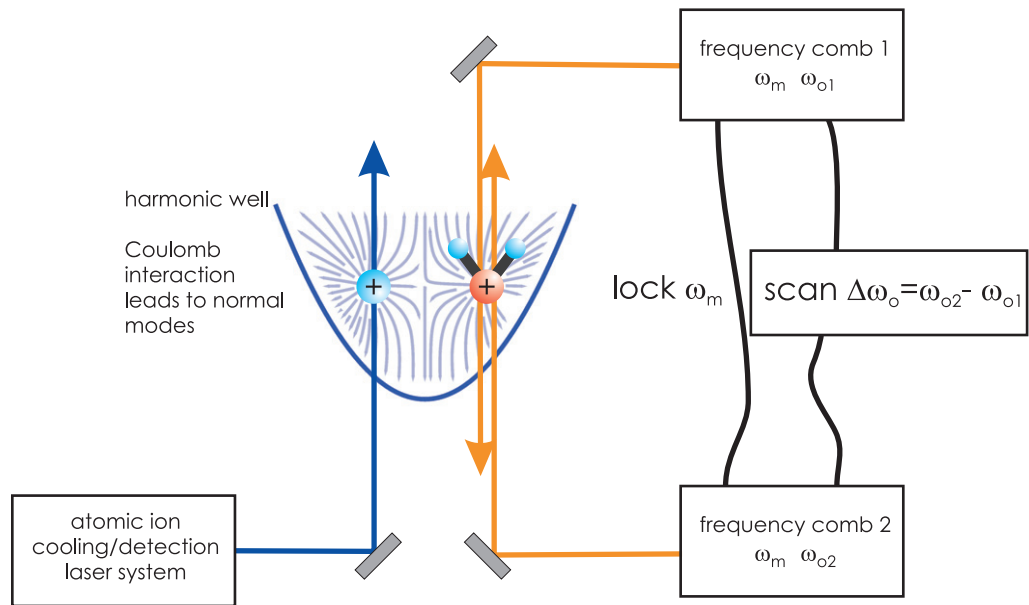
Figure 1. Schematic setup of a quantum-limited molecular spectrometer. A known atomic ion (blue) and a possibly unknown molecular ion (red and blue stick model) are confined in the same harmonic trapping well. The atomic ion is initialized to a known internal state. Due to the Coulomb interaction between atomic and molecular ions the normal modes of vibration of the entire crystal can be sympathetically cooled to their ground state by coupling exclusively to the atomic ion with laser interactions (blue arrow). The rotational states of the molecule are then manipulated by inducing Raman transitions due to the light fields of two counter-propagating frequency combs (orange arrows) of equal repetition rate $\omega_m/(2\pi)$ and variable relative carrier-envelope phase $\Delta\omega_o$. The photon recoils produced by Raman transitions of the molecule can excite the shared crystal motion, and this excitation can subsequently be detected on the atomic ion (see text). Detection of crystal excitation thus establishes that the molecule has undergone a transition.

$e \cdot a_0 \approx 8.48 \times 10^{-30}\,\mathrm{C\,m}$, where e is the electron charge and a_0 the Bohr radius. The transition rate on the $n = 0$ carrier is then approximately [28]

$$|\Omega_0| = \frac{(e\,a_0 E_{\mathrm{av}})^2}{4\hbar\Delta_l}, \tag{6}$$

with Δ_l being the detuning of the comb central frequency to the resonance frequency from the molecular ground state to the intermediate level and E_{av} the average electric field per comb, set to be identical for both combs for simplicity. If each comb emits 1 W on average into a beamwaist of $20\,\mu\mathrm{m}$ around 800 nm and the dipole transition resonance is at 400 nm, the $n = 0$ carrier Raman transition rate is $|\Omega_0| \approx 2\pi \times 824\,\mathrm{kHz}$. The rate $|\Omega_1|$ on $n = \pm 1$ sideband transitions is given by $|\Omega_1| = \eta|\Omega_0|$, with the Lamb–Dicke parameter η typically ranging between 0.03 and 0.3, depending on the normal mode structure and the comb wavelength (see equation (5)).

3.1. High field effects and accidental resonances

The laser parameters used in the previous section call for relatively short pulses at high peak intensity. In the Fourier limit, a 30 THz wide comb requires a pulse length of about 15 fs (computed for a Gaussian pulse envelope). For the waist size and average power assumed above and 1 GHz repetition rate, this pulse length results in peak intensities of approximately 10^{10} W cm^{-2}. Multi-photon above-threshold ionization and above-threshold dissociation require the electric field of the lasers to become comparable to the Coulomb field of the atoms. Typical intensities for the onset of these effects are estimated to be of the order of 10^{12} W cm^{-2}, about two orders of magnitude higher than anticipated here.

Even if off-resonant multi-photon effects are unlikely, one might have to deal with accidental resonances driven by the pulsed lasers. No rigorous general statement can be made regarding how problematic such resonances might be. In most cases, single-photon transitions would correspond to high-order vibrational overtones and should be correspondingly weak. Two-photon transitions would most likely be of the Raman type. Spontaneous Raman transitions, where one photon comes from the pulsed laser while the other photon is spontaneously emitted, should be suppressed relative to the desired stimulated Raman transitions that connect rotational levels with both photons provided by the lasers. A rough estimate based on a typical spontaneous Raman scattering cross-section of 10^{-28} cm^2 steradian^{-1} [29, 30] and the intensities and duty cycles discussed in section 4 yield a spontaneous Raman scattering rate of approximately 8×10^{-6} s^{-1}. If accidental resonances still pose a problem in a particular case, the intensity of the Raman beams can be reduced at the expense of a proportionally lower transition rate on the desired rotational Raman transitions. Besides the longer time spans required for scanning a rotational spectrum, lower transition rates pose a problem if they become comparable to either the black-body rates between rotational levels or the heating rate of the motional state of the ion crystal in the ion trap potential. In large ion traps at room temperature, both these time scales should be a good fraction of a second, so the transition rates could be reduced by several orders of magnitude before being limited by such decoherence effects.

3.2. Rotational cooling for known molecular constants

For known rotational constants and transition rates of the molecule, we can set the relative detuning of the combs such that two rotational levels with high thermal occupation probability are connected by an $n = -1$ resonance condition in equation (4). As long as the motion of the two-ion crystal is in the ground state, this transition can proceed only one way, from the J to the $J - 1$, 2 states, while one quantum of motion is added to the mode at ω_+ (see figure 2(a)), the reverse direction cannot conserve energy.

Each attempt to excite a Raman transition between rotational states of the molecule with the comb-lasers would be followed by an attempt to drive an $n \to (n - 1)$ resolved sideband on the cooling and detection ion (see figure 2(b)). As long as the selected normal mode of crystal motion is in the ground state ($n = 0$), this transition is energetically forbidden, but if the preceding transition of the molecule has deposited a quantum of motion ($n = 1$), the electronic state of the atomic ion can be changed while this quantum is removed [16]. The electronic state change of the atom can be detected by state-selective fluorescence on the cooling and detection transition, in analogy to quantum-logic ion clocks [16, 19] (see figure 2(c)). Crucially, if a state change of the atomic ion is detected, this near-ideal measurement projects the rotational state of the molecule into a pure and known quantum state, reducing the entropy of the molecular state
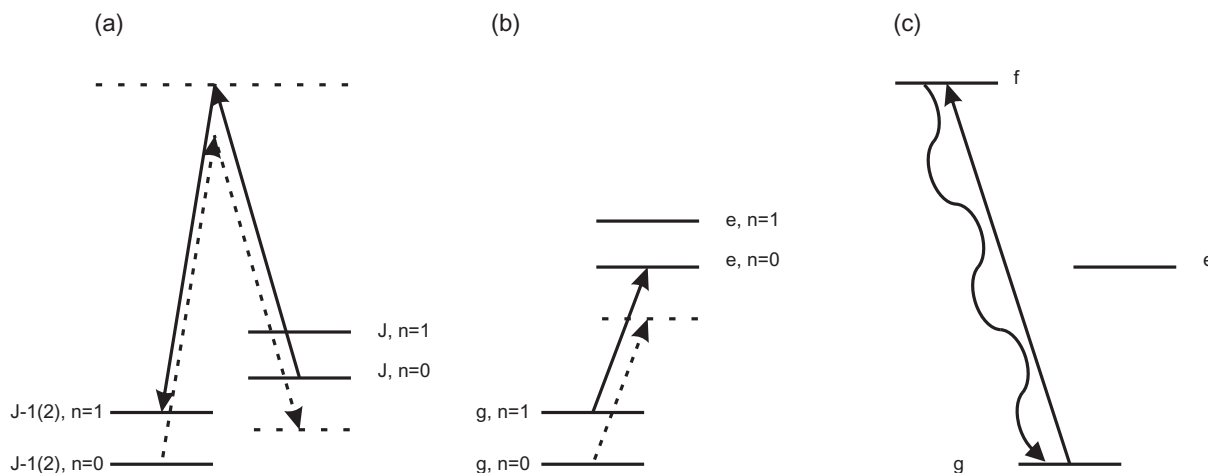
Figure 2. Molecular rotational Raman transition and subsequent detection (energy differences not to scale). (a) The molecule undergoes a Raman transition on the $n = -1$ sideband of the selected normal mode of the two-ion crystal in equation (4). Since this mode is in the ground state, only the rotational transition $J \to J - 1$ is energetically allowed (solid lines), while the opposite direction is not resonant with an existing state (dashed lines). (b) The conditionally added quantum of motion is transferred into an excitation of the co-trapped atom by the use of a $(g, n = 1) \to (e, n = 0)$ resolved sideband transition, where g and e refer to two long-lived states in the atomic ion. (c) The electronic state of the atomic ion is detected by laser-induced fluorescence on a cycling transition connecting g with the fast-decaying level f [16, 19]. If no fluorescence is detected, the atom is found to be in the excited state e. This near-ideal measurement projects the state of the molecule into $J - 1$. The atom can then be reset to g by optical pumping and the crystal can be re-cooled to the motional ground state. This resets atom and two-ion-crystal motion to the initial condition before step (a) without affecting the rovibrational or electronic state of the molecule. The reset removes entropy from the system and prepares it for further manipulations.

considerably in one successful attempt [17]. In case the excitation attempt is not successful, the addressed rotational state is almost certainly not occupied, so we can proceed to probing another highly occupied rotational state in an updated distribution that excludes the unsuccessfully probed level(s). Once the initial distribution of rotational states is projected into a pure state in this manner, we can drive the system straight into the rotational ground state with carrier-type transitions ($n = 0$ in equation (4)) if the Raman transition rates are known. It might also be possible to use techniques similar to stimulated rapid adiabatic passage (STIRAP) [13, 14, 31] to provide more robust carrier and sideband transitions without exact knowledge of the transition rates.

3.3. Determination of transition rates

If the Raman transition rates of the molecule are not known, they can be determined as follows: once the molecular ion has been projected into the rotational state J, we can probe the molecule

on the $(J, n = 0) \to (J - 1, 2, n = 1)$ transition for several different excitation periods and transfer any motional excitation of the two-ion crystal into an electronic state change of the atom by inducing a red sideband transition of the atom, as in the preceding step (section 3.2) that narrowed the thermal J-distribution to just one state. If the atomic ion changes state in such an excitation attempt, the transition to $(J - 1, 2, n = 1)$ has most likely been made; therefore, the molecule is now in the state $J - 1, 2$ with high probability, and can be returned to J by re-cooling the two-ion crystal to the $n = 0$ ground state, followed by repeated attempts to do a $(J - 1, 2, n = 0) \to (J, n = 1)$ sideband transition (detuned by $2\omega_{\pm}$ to the blue from the transition previously attempted). By recording transition probabilities as a function of length of excitation in many repetitions of this cycle, the transition rate and subsequently the magnitude of transition matrix elements can be inferred.

3.4. Quantum-non-demolition measurement of the rotational state

For known transition rates, one should be able to repeat the cycle of successive $n = \pm 1$ transitions and detection of the molecular state changes outlined in section 3.3 several times. This ideally constitutes a quantum-non-demolition (QND) readout as we should leave a motional quantum that can be subsequently read out through the state of the atom in each cycle step. Every time this happens, we gain more certainty about the state the molecule is in even if single measurements have only modest fidelity. An analogous state-detection scheme with atomic ions has reached a detection fidelity of 99.94%, with a fidelity of about 80% for a single measurement [32]. Several cycles of such QND readouts also prepare the molecule rotation into a pure quantum state with higher confidence than that for a single detection of a transition.

3.5. Determination of molecular constants

If the rotational levels of the molecular ion are unknown, we can slowly scan the frequency difference $\Delta\omega_{\mathrm{o}}$ of the two combs over the comb spacing ω_{m}. At some point, an $n = \pm 1$ resonance will lead to a molecular Raman transition that will leave a quantum in the selected mode of motion of the crystal. Detecting a state change in the atom after converting the motion into atomic excitation implies that the molecule is in either of two J-levels that the previous Raman interaction has resonantly connected, so the distribution of rotational states is narrowed to two levels. We can reset the atom state by optical pumping, and recool the crystal motion to the ground state and then attempt to drive a molecular transition at a relative detuning that is $2\omega_{\pm}$ red-detuned from the previous pulse. Again, the transition is possible only if a quantum of motion is added that can be subsequently detected with the atom. Detection of the atomic state change would establish that the molecule has undergone a transition from the higher energy J to the $J - 1$ or $J - 2$ state (for linear and spherical-top molecules, the $J - 2$ state has been prepared with certainty). If several attempts to drive at this red-detuned frequency fail, the molecule is most likely already in the lower-energy state and the failed attempts were actually on an $n = -3$ sideband transition that is also energetically forbidden if the selected mode of crystal motion is in the ground state. In this case, attempts to drive transitions $2\omega_{\pm}$ blue detuned from the pulse that produced the original transition should excite the molecule to the higher-energy J state, while a motional quantum is deposited ($n = 1$) that can subsequently be converted into a state change of the atom and detected. In either case, the unknown frequency $\Omega_{J,J\pm1,2}$ can be found from the mean of the two resonant frequencies in the preceding steps.

It is then possible to find the rate of these transitions and gain information about the related transition matrix elements, as described in section 3.3. Further rotational levels may be identified by completing the scan of $\Delta\omega_o$ over the full comb mode spacing ω_m. After several distinct transitions are found, it should be possible to assign J and determine all rotational constants and centrifugal distortion constants. This can be done unambiguously if the absolute frequency of every transition is known. This is possible even if N in equation (4) is initially unknown. From the range of rotational transitions and assuming that ω_m is approximately $2\pi \times 1\,\mathrm{GHz}$, N can take values between 1 and 10^4. After finding a resonance, we can change ω_m by a small amount of (say) the order of $10^{-3}\omega_m$, easily observable by monitoring the repetition rate of the comb(s) on a fast photodiode. Then $\Delta\omega_o$ can be scanned until the resonance condition is re-established. The closest integer to the fraction of the changes in ω_m and $\Delta\omega_o$ is N. We can eventually infer the full rotational spectrum of the molecule by systematically scanning the complete range ω_m and then determining N for each resonance found. With this knowledge, the spectrum, originally compressed into one range ω_m, can be deconvoluted to yield rotational state information with a resolution limited only by the lifetimes of the involved states.

For simplicity, the preceding discussion has neglected any hyperfine structure that the molecule might possess. Hyperfine structure complicates the spectrum-finding procedure by adding more resonances and one more degree of freedom that needs to be determined. Still, it seems straightforward to incorporate hyperfine structure with slight modifications into the procedures outlined.

4. MgH⁺ co-trapped with Mg⁺

Estimated parameters for the concrete case of a MgH⁺ molecule, co-trapped with a Mg⁺ atom, establish the feasibility of the proposed scheme. The rotational state of MgH⁺ has been previously prepared close to its ground state in an ensemble of molecular ions with dedicated laser systems and a destructive detection method [5]; therefore all relevant molecular parameters are reasonably well known. As a linear molecule, MgH⁺ admits only $J \to J \pm 2$ rotational Raman transitions. The low mass and small size produce a rotational constant of approximately 190 GHz and a centrifugal correction of the order of 10 MHz. In thermal equilibrium with a room-temperature environment only the first 11 rotational states have occupation probabilities above 1%. The highest occupation probability is approximately 14% for $J = 4$. The $J \to J \pm 2$ selection rule dictates that we should first try to prepare a state with even J to be able to connect to the ground state. The dipole transition matrix element between the ground $X^1\Sigma^+$ and the lowest electronically excited $A^1\Sigma^+$ state with an energy difference corresponding to 280 nm is about $1.57e\,a_0$ [33]. For the same laser parameters as in section 3.2, the carrier Raman transition rate, equation (6), is approximately $|\Omega_0| \approx 2\pi \times 1.1\,\mathrm{MHz}$. For a crystal consisting of MgH⁺ and Mg⁺ at a single Mg⁺ trap frequency of $\omega_a \approx 2\pi \times 1\,\mathrm{MHz}$ the two normal-mode frequencies, equation (1), are $\omega_+ \approx 2\pi \times 0.99\,\mathrm{MHz}$, and $\omega_- \approx 2\pi \times 1.72\,\mathrm{MHz}$, with the Lamb–Dicke factors, equation (5), of $\eta_+ \approx 0.16$ and $\eta_- \approx 0.13$, respectively. These numbers imply $n = \pm 1$ sideband transition rates of $|\Omega_1| \approx 2\pi \times 174\,\mathrm{kHz}$ (complete rotation between states achieved in $1.43\,\mu\mathrm{s}$) and $|\Omega_1| \approx 2\pi \times 137\,\mathrm{kHz}$ (complete rotation in $1.82\,\mu\mathrm{s}$), respectively. Together with initial ground-state cooling and subsequent transfer of motional excitation and state readout of the Mg⁺ ion, one experimental cycle should take not more than 1 ms [19]. An initial scan could

involve 100 attempts per comb detuning $\Delta\omega_o$, yielding a scan speed of ten frequency points per second. A 10 μs window with about 10 000 Raman pulses applied at 1 GHz repetition rate to the molecule would correspond to a Fourier limited width of about 100 kHz for each resonance; so, to collect a reasonably densely sampled spectrum over 1 GHz, we would need 10^5 points taken in 10^4 s or about 3 h. This duration represents a worst-case estimate, where nothing about the rotational constants is known, and is at the same time compatible with achievable lifetimes in ion traps. If we assume that ten transitions with initial state population larger than 1% are present, it should take less than 1 h to identify the first resonance. With each newly identified resonance, finding the remaining resonances should take fewer attempts. In this manner, it should be possible to determine a complete rotational spectrum in a few hours. Once the most populated sub-levels are determined, these could be directly prepared within a time span comparable to the rotational re-thermalization period of the molecule by repeating excitation attempts until a transition is successfully made. An experiment in a low-temperature environment would reduce the initial spread of J-levels considerably, while suppressing background gas collisions with the crystal and thus further increasing the ion lifetimes in the trap.

5. Conclusions

In the recent past, the quantum control of neutral and ionized atoms has proven to be enormously fruitful. Controlling the quantum state of a multitude of molecules using the methods proposed here and other methods that have recently emerged promises to take us to the next level of complexity. We can hope to keep and study the same molecule with a resolution limited only by natural linewidths under full control of its quantum states. It should also be possible to prepare certain rotational states efficiently, including the rotational ground state and coherent superpositions of these states. This may be further aided by a higher-fidelity QND way of measurement/preparation that is described in section 3.4. After preparation, the molecular quantum state can be coherently manipulated with the same or other laser sources within the limits imposed by decay and re-thermalization of the involved states. For rotational states within the electronic and vibrational ground state manifold, re-thermalization is often on a time scale of many seconds.

Aided by these tools, we might test quantum mechanics and its application to coupled systems to unprecedented levels and search for energy shifts due to parity violation, a finite value of the electron electric dipole moment or a variation of fundamental constants, such as the proton-to-electron mass ratio. It might also become feasible to induce highly controlled molecular collisions. Because reaction cross-sections can strongly depend on the vibrational and rotational quantum states, we might glean better control and understanding of chemical reactions in this way.

On completion of preparing the manuscript, I became aware of similar work by the group of D N Matsukevich at the National University of Singapore [34].

Acknowledgments

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