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J. Phys. B: At. Mol. Opt. Phys. 39 (2006) S1259-S1265

doi:10.1088/0953-4075/39/19/S31

# LETTER TO THE EDITOR

# Probabilistic state preparation of a single molecular ion by projection measurement

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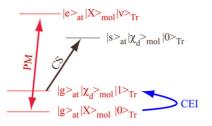
Received 27 March 2006 Published 25 September 2006 Online at stacks.iop.org/JPhysB/39/S1259

#### Abstract

We propose how to prepare a single molecular ion in a specific internal quantum state in a situation where the molecule is trapped and sympathetically cooled by an atomic ion and where its internal degrees of freedom are initially in thermal equilibrium with the surroundings. The scheme is based on the conditional creation of correlation between the internal state of the molecule and the translational state of the collective motion of the two ions, followed by a projection measurement of this collective mode by atomic ion shelving techniques. We estimate that state preparation in a large number of internal states is possible.

Investigations of production and trapping of cold neutral (van de Meerakker et al 2005, Doyle et al 2004, Egorov et al 2002) and ionic molecules (Mølhave and Drewsen 2000, Drewsen et al 2004, Bertelsen et al 2006) point to a wealth of possible applications, including studies of molecular Bose-Einstein condensates (Zwierlein et al 2004, Ohashi and Griffin 2002, Bourdel et al 2004, Szymanska et al 2005, Greiner et al 2005), investigations of collision and reaction dynamics at low temperature (Smith 1994, Kreckel et al 2002), high-resolution spectroscopy (Krems et al 2004, Roth et al 2005), coherent control experiments (Rice and Zaho 2000) and state-specific reaction studies (Harich et al 2002, Liu et al 2000). For much of this research, long-term localized and state-specific targets are highly desirable. One way to obtain such targets is to work with trapped molecular ions sympathetically cooled by atomic ions where previous investigations show that molecular ions can be translationally cooled to temperatures of a few mK, at which stage they become immobile and localize spatially in Coulomb crystal structures (Mølhave and Drewsen 2000, Drewsen et al 2004). Though these molecules are translationally cold, studies indicate that the internal degrees of freedom of at least smaller hetero-nuclear molecules, due to their interaction with the black-body radiation, are close to being in equilibrium with the temperature of the surroundings (Bertelsen et al 2006). This is not unexpected since the many orders of magnitude difference between the internal transitions frequencies in the molecule  $(\gtrsim 10^{11} \text{ Hz})$  and the frequency of the collective vibrational modes

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**Figure 1.** Idealized sketch of the state-selection sequence. The first step is cooling to the ground state of the collective translational mode,  $|0\rangle_{Tr}$  with the molecule in the electronic ground state and with the internal rovibrational states characterized by a statistical ensemble  $|X\rangle_{mol}$ . The second step involves the correlation of external motion of the two-ion system and the internal state of the molecule (CEI) by exciting the collective mode to  $|1\rangle_{Tr}$  if the molecule is in a specific internal state,  $|\chi_d\rangle_{mol}$ . Next, if the collective mode is excited, conditional shelving (CS) transfers the atom to the long-lived metastable state,  $|s\rangle_{at}$ . Finally, the projection measurement (PM) proceeds by exposing the atomic ion to light which is resonant with the transition between the atomic ground state,  $|g\rangle_{at}$ , and an excited state,  $|e\rangle_{at}$ . Rescattered light will then be absent only if the atom is in the shelved state and hence the molecule is in the desired internal state  $|\chi_d\rangle_{mol}$ .

in the Coulomb crystals ( $\leq 10^7$  Hz) leads to very inefficient coupling between these degrees of freedom. Several schemes were recently proposed to decrease the rotational temperature of translationally cold hetero-nuclear molecular ions (Vogelius *et al* 2002, 2004).

Here, we focus on an alternative route to the production of molecular ions in specific states. The physical system used for this purpose consists of one trapped molecular ion sympathetically cooled by a simultaneously trapped atomic ion. Such a situation was previously realized and it was shown to be possible to determine the molecular ion species non-destructively by a classical resonant excitation of one of the two axial collective modes of the two-ion system (Drewsen *et al* 2004). With this setup, we now propose to exploit the quantum aspect of the same collective modes to create correlations between the internal state of the molecular ion and the collective motional state in the trap potential. Previously, correlations in two-ion systems were essential, e.g., in demonstrations of quantum logical gates (Schmidt-Kaler *et al* 2003) and in high-resolution spectroscopy (Wineland *et al* 2003).

As depicted in figure 1, the state preparation of the molecular ion ideally involves the following steps. First, the two-ion system is cooled to its collective motional ground state (Rohde et al 2001, Barrett et al 2003) with the molecule in the electronic ground state and with a Maxwell-Boltzmann distribution over rovibrational states. We consider only one of the two independent axial modes of the two-ion system and refer to it as the collective mode. Second, laser fields are applied to induce transitions between the ground and the first excited motional states conditioned on the specific rovibrational state of the molecular ion. This procedure creates correlations between the motional state of the two-ion system and the internal state of the molecular ion. Next, conditioned on an excitation of the collective mode, an atomic shelving transition to a metastable state is driven by another laser field. Finally, laser fields are applied to project the atomic ion on the shelved (non-fluorescing) or non-shelved (fluorescing) state. If no fluorescence is observed, we conclude that the molecular ion is in the internal state of interest. In contrast, if fluorescence is present, the ion is not in the desired state. In the latter case, after a duration of time sufficiently long to bring the molecule back into thermal equilibrium (typically through interaction with black-body radiation), the procedure is repeated. Eventually, no fluorescence is detected in the final step, and the molecular ion is known to be in the desired quantum state. A state-to-state analysis of the procedure is presented schematically in table 1. The same type of state-projection measurement was

Table 1. The evolution of the system through the state-selection sequence depicted in figure 1.

| IE  | $ g\rangle_{\rm at} X\rangle_{\rm mol} 0\rangle_{\rm Tr}$      |  |
|-----|--|--|
| CEI | $ g\rangle_{\rm at} \chi_d\rangle_{\rm mol} 1\rangle_{\rm Tr}$ | $ g\rangle_{\rm at} X'\rangle_{\rm mol} 0\rangle_{\rm Tr}$ |
| CS  | $ s\rangle_{\rm at} \chi_d\rangle_{\rm mol} 0\rangle_{\rm Tr}$ | $ g\rangle_{\rm at} X'\rangle_{\rm mol} 0\rangle_{\rm Tr}$ |
| PM  | Dark   | Fluorescent  |

IE: initialization of the external state of the two-ion system by cooling the collective mode to the ground state. CEI: correlation of collective external modes and internal molecular state. CS: conditional shelving. PM: projection measurement. In the table,  $|X'\rangle_{mol}$  denotes the modified statistical ensemble of internal molecular states when the desired state,  $|\chi_d\rangle_{mol}$ , has been removed from  $|X\rangle_{mol}$ . The portion of the ensemble of rovibrational states that was initially in the desired molecular state  $|\chi_d\rangle_{mol}$  is conditionally shelved and unaffected by the projection measurement. The remaining rovibrational states, on the other hand, are unaffected by the conditional shelving and therefore lead to fluorescence upon projection measurement.

previously proposed in connection with stochastic cooling of a single atomic ion (Eschner *et al* 1995).

Before a discussion of a realistic implementation of the proposed scheme, we evaluate the effect of imperfections in the various steps of the procedure.

(1) Initialization of the external state of the two-ion system (IE). Cooling of a two-ion system completely to the motional ground state,  $|0\rangle_{Tr}$ , is unrealistic, but several experiments show that it is feasible to achieve  $W_0 = 95\%$  population in  $|0\rangle_{Tr}$  in the case of two atomic ion species (Barrett *et al* 2003, Rohde *et al* 2001). The same degree of cooling is expected for an atomic–molecular ion system. We therefore use a Boltzmann distribution with  $W_0 = 95\%$ , and this results in  $W_1 \simeq 4.7\%$  and  $W_2 \simeq 0.3\%$  for the populations in  $|1\rangle_{Tr}$  and  $|2\rangle_{Tr}$ , respectively.

(2) Correlation of external motion and the internal molecular state (CEI). This part of the procedure can, e.g., be accomplished by inducing transitions between rotational sub-states of the molecule using Raman  $\pi$ -pulses (Schmidt-Kaler *et al* 2003, Monroe *et al* 1995) in a way similar to that demonstrated for sub-states in atomic ions (Monroe et al 1995). Such a transition can be made state-selectively by a suitable choice of laser frequencies, i.e., by assuring a very large detuning for states other than the desired one. Alternatively, if there are no rotational sub-states, as in the case of J = 0, or if for some reason it is desired to stay in a specific sub-state, a sequence of two laser pulses can be applied like in stimulated Raman adiabatic passage (STIRAP) processes (Bergmann et al 1998). First, a pulse couples the final state,  $|\chi_d\rangle_{mol}|1\rangle_{Tr}$ , and intermediate states,  $|\chi'\rangle_{mol}|\nu\rangle_{Tr}$ , with a coupling strength characterized by the free molecule Rabi frequency,  $\Omega_s(t)$ , while a delayed pulse couples the initial state,  $|\chi_d\rangle_{mol}|0\rangle_{Tr}$ , to  $|\chi'\rangle_{mol}|\nu\rangle_{Tr}$  with free molecule Rabi frequency  $\Omega_p(t)$ . Here,  $|\chi_d\rangle_{mol}$  and  $|\chi'\rangle_{mol}$  denote the desired and intermediate molecular state, respectively. Though the pulse sequence resembles a STIRAP process, there is an important difference since the two laser pulses, which are only shifted in frequency by the collective mode frequency ( $\simeq 10$  MHz), do interact with the same internal transitions of the molecule. To model the effect of such a two-pulse process, we expand the state of the two-ion system as  $|\Psi(t)\rangle = \sum_{\nu_{\text{Tr}}=0}^{\nu_{\text{Tr},\text{max}}} c_{\nu_{\text{Tr}}}(t) |\chi_d\rangle_{\text{mol}} |\nu\rangle_{\text{Tr}} + b_{\nu_{\text{Tr}}}(t) |\chi'\rangle_{\text{mol}} |\nu\rangle_{\text{Tr}}$  with  $\nu_{\text{Tr},\text{max}} = 5$  for convergence, initial condition  $c_0$  (t = 0) = 1 and desired final state  $|\chi_d\rangle_{mol}|1\rangle_{Tr}$ . In the simulations, both laser pulses are assumed to be Gaussian in time with a width  $\tau = 50 \,\mu s$  (FWHM) and separated by  $1.3\tau$ . Furthermore, we assume the intermediate molecular state to be a vibrational excited state with a spontaneous decay rate of  $100 \text{ s}^{-1}$ . With a realistic molecular transition wavelength of 6  $\mu$ m (e.g., the vibrational transition between the ground and the first excited state in the  $X^{1}\Sigma$  state of MgH<sup>+</sup> (Vogelius *et al* 2004)), a maximum free molecule Rabi frequency of 7 MHz for both pulses, and a detuning from the intermediate state of  $\delta \simeq 10$  MHz, we find that more than 80% of the population can be transferred to motional excited states. Of experimental importance, the transfer efficiency was found to be stable when varying the detuning a few MHz.<sup>3</sup> In the following, we use the more conservative estimate of the transfer efficiency,  $\wp_{CEI} = 0.7$ .

(3) *External state-conditional shelving (CS)*. Shelving conditioned on the excitation of the collective mode can, e.g., be achieved by driving red sideband transitions between motional states by  $\pi$ -pulses (Monroe *et al* 1995). As an alternative, we consider the STIRAP type process from the atomic ground state,  $|g\rangle_{at}$ , to the shelved atomic state,  $|s\rangle_{at}$ , via an intermediate state,  $|i\rangle_{at}$ . We expand the state of the combined system as

$$|\Psi(t)\rangle = \sum_{\nu_{\mathrm{Tr}}=0}^{\nu_{\mathrm{Tr},\mathrm{max}}} (c_{\nu(t)_{\mathrm{Tr}}}|g\rangle_{\mathrm{at}}|\chi_d\rangle_{\mathrm{mol}}|\nu\rangle_{\mathrm{Tr}} + b_{\nu(t)_{\mathrm{Tr}}}|i\rangle_{\mathrm{at}}|\chi_d\rangle_{\mathrm{mol}}|\nu\rangle_{\mathrm{Tr}} + a_{\nu(t)_{\mathrm{Tr}}}|s\rangle_{\mathrm{at}}|\chi_d\rangle_{\mathrm{mol}}|\nu\rangle_{\mathrm{Tr}}).$$

As initial conditions we use the final amplitudes from the step setting up the correlation between the external motion and the internal molecular state (CEI). These amplitudes showed significant population in the first few excited motional states. Our simulations show that a transfer efficiency  $\wp_{CS}$  of more than 95% is obtained even with population in several excited motional states. We use the more conservative estimate  $\wp_{CS} = 0.7$  in the following.

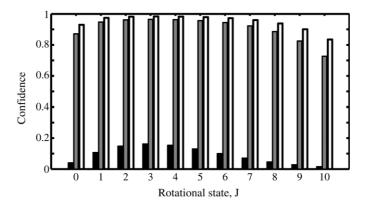
(4) Projection measurement (PM). The final step, projection measurement on the atomic ion, can be made very efficient. With a typical exposure time T = 5 ms, one should be able to determine the projected atomic state with more than a 95% confidence (Rowe *et al* 2001).

Over-all confidence of steps (1)–(4). The probability of being in the ideal initial state  $|g\rangle_{\text{at}}|\chi_d\rangle_{\text{mol}}|0\rangle_{\text{Tr}}$  is  $P_{\chi_d}W_0$ , where  $P_{\chi_d}$  denotes the initial population in  $|\chi_d\rangle_{\text{mol}}$  and  $W_v$  denotes the initial population in the collective motional state  $|v\rangle_{\text{Tr}}$ . This latter state has to go through the step setting up the correlation between the external motion and the internal molecular state (CEI) as well as the conditional shelving (CS) step for the external state to reach the shelved state. Therefore, the probability of finding the molecule in the shelved state after the selection sequence is  $S_{\text{prep}} = P_{\chi_d} W_0 \wp_{\text{CEI}} \wp_{\text{CS}}$ . Unfortunately, a false positive result can occur if the system is initially in a state  $|v\rangle_{\text{Tr}}$  with  $v \ge 1$  since the system may proceed through CS without exciting the motional state during CEI. The probability of a false positive measurement is then  $E = (1 - P_{\chi_d})(1 - W_0) \wp_{\text{CS}}$ , where the first two factors account for the initial population in the excited states of the collective motion, but not in  $|\chi_d\rangle_{\text{mol}}$ , and the last factor accounts for the necessary application of conditional shelving. We define the confidence of a measurement as  $F = S_{\text{prep}}/(S_{\text{prep}} + E)$ .

As a test case we assume  $P_{\chi_d} = 5\%$  and use a thermal distribution over external vibrational states determined by letting  $W_0 = 95\%$ . Non-fluorescence in the final stage of the state preparation then gives a confidence of the molecule being in  $|\chi_d\rangle_{mol}$  of about  $F \simeq 0.4$  which is too marginal for the procedure to be useful.

(5) *State purification (SP)*. The principal source of error is false positive detections stemming from initial population in  $|1\rangle_{Tr}$ . These errors are excluded by using the state-purification procedure presented in table 2. If no fluorescence is detected after the state-preparation process, the step creating correlation between the external motion and the internal molecular state (CEI) is reapplied. The second step is another conditional shelving process transferring

<sup>&</sup>lt;sup>3</sup> Two laser pulses with a difference frequency which is insensitive to fluctuations in the laser frequency may be generated from a single CW laser source by application of acousto-optical modulators. A Rabi frequency of 7 MHz can be achieved by focusing CW laser beams with a modest power of  $\sim 10$  mW to spot sizes of  $\sim 1 \text{ mm}^2$ .



**Figure 2.** Confidence of finding a MgH<sup>+</sup> ion in rotational states. The black columns correspond to a 300 K Boltzmann distribution over rotational states. The grey columns present the achievable confidence, F', when state purification is applied. Finally, the white columns show the confidence, F'', obtainable when selections using the second sideband are applied without state purification. The confidence levels were derived using the efficiencies of CEI, CS and PM, given in the text.

**Table 2.** State purification (SP). Start: the states of the system ending in the shelved atomic state after the state-preparation process (1)–(4) when assuming finite temperature of the collective mode. SP proceeds by a second correlation of the external mode and internal molecular state (CEI), exciting the collective mode if the molecule is in  $|\chi_d\rangle_{mol}$ . Then conditional shelving (CS) transfers the atom back to the ground state  $|g\rangle_{at}$  if the collective mode is excited. Finally, a second projection measurement (PM) detects the atom in  $|g\rangle_{at}$  if the molecule is in  $|\chi_d\rangle_{mol}$ .

| Start | $ s\rangle_{\rm at}  \chi_d\rangle_{\rm mol}  0\rangle_{\rm Tr}$ | $ s\rangle_{\rm at} X'\rangle_{\rm mol} 0\rangle_{\rm Tr}$ |
|-------|--|--|
| CEI   | $ s\rangle_{\rm at}  \chi_d\rangle_{\rm mol}  1\rangle_{\rm Tr}$ | $ s\rangle_{\rm at} X'\rangle_{\rm mol} 0\rangle_{\rm Tr}$ |
| CS    | $ g\rangle_{\rm at} \chi_d\rangle_{\rm mol} 0\rangle_{\rm Tr}$   | $ s\rangle_{ m at} X'\rangle_{ m mol} 0 angle_{ m Tr}$     |
| PM    | Fluorescent  | Dark   |

 $|s\rangle_{at}$  to  $|g\rangle_{at}$  on the red sideband of the collective motion. Finally the projection measurement is repeated. The desired final state is now fluorescent while other states are dark.

The probability of a successful detection after the state-purification procedure is estimated by  $S_{pur} = S_{prep} \wp_{CEI} \wp_{CS}$ . This result follows from the fact that the population has to go through both the correlation step (CEI) and the conditional shelving step after the state preparation. The probability of a false positive detection caused by population initially in excited motional states is now given by  $E1 = W_{l \ge 2}(1 - P_{\chi_d})\wp_{CS}^2$ , since the system must start in the second excited state or higher to pass the conditional shelving step twice without being excited during the correlation step (CEI). Another source of false positive measurements appears due to stochastic heating during the relatively slow projection measurement. Assuming a heating rate of  $\Gamma = 10$  vibrational quanta per second (Rohde *et al* 2001), the error induced by stochastic heating is  $E2 = T\Gamma(1 - P_{\chi_d})W_1\wp_{CS}^2$ , since only the population initially in  $|1\rangle_{Tr}$  will introduce the error which is not accounted for in E1.

The confidence of state preparation after state purification is defined as  $F' = S_{pur}/(S_{pur} + E1 + E2)$  and the state-purification step improves the confidence of state preparation to more than 80% (see figure 2). Note that if  $W_0 = 100\%$ , the measurement cycle leads to 100% certain state preparation even without the state-preparation step. Increasing the efficiency of the CEI correlation step,  $\wp_{CEI}$ , to 100% leads to only 10% increase in the confidence of the

final state preparation. Finally, we note that the preparation efficiency is independent of the conditional shelving efficiency,  $\wp_{CS}$ .

As an alternative to the state-purification step, we may perform the correlation step (CEI) and the conditional shelving on the second motional sideband, i.e., consider  $|g\rangle_{at}|\chi_d\rangle_{mol}|0\rangle_{Tr} - |g\rangle_{at}|\chi_d\rangle_{mol}|2\rangle_{Tr}$  in the CEI step and  $|g\rangle_{at}|\chi_d\rangle_{mol}|2\rangle_{Tr} - |s\rangle_{at}|\chi_d\rangle_{mol}|0\rangle_{Tr}$  in the conditional shelving step. Since the initial probability  $W_2$  for being in  $|2\rangle_{Tr}$  is much smaller than  $W_1$ , the confidence of the preparation is improved considerably without applying state purification, and is simply given by  $F'' = S_{prep}/(S_{prep} + E'')$  with  $E'' = E \times (1 - W_0 - W_1)/(1 - W_0) \sim E \times$ 0.06. The required laser intensity would, however, increase significantly due to the weaker coupling between  $|0\rangle_{Tr}$  and  $|2\rangle_{Tr}$  compared with the  $|0\rangle_{Tr} - |1\rangle_{Tr}$  coupling.

The average time needed for the performance of a successful detection depends partly on the probability of finding the molecule in  $|\chi_d\rangle_{mol}$  at thermal equilibrium, partly on the time needed for the projection measurement cycle including the purification step, and partly on the time scale for re-thermalization. Even for polar diatomic molecules with a relatively large rotational constant, the time scale for re-thermalization, due to the interaction with black-body radiation ( $\tau_{re} \sim 5$  s), is much longer than the projection measurement cycle. Hence, the average time is estimated by  $\tau_{re}/S_{prep}$  which for a state with a thermal population of ~10% is ~100 s.

As a specific test case for an implementation of the state-preparation scheme, we now focus on a MgH<sup>+</sup> ion internally in equilibrium with black-body radiation at 300 K and trapped together with an atomic ion amenable for conditional shelving, e.g., a Ca<sup>+</sup> ion. We neglect the vibrational quantum number and take into account only the rotational states of the MgH<sup>+</sup> ion since at room temperature it is in the vibrational ground state with more than 99% certainty. The rotational levels are populated according to the Boltzman distribution presented in figure 2. We calculate the confidence F' for the molecular ion to be found in a specific rotational state after application of the state preparation- and state purification-scheme. The results presented in figure 2 show that the ion can be prepared in all 11 represented rotational states with high confidence. Hence, if the aim is to study the dependence of a process on the internal state of a molecule, starting out with an internally hot molecular ion may turn out to be advantageous.

The above-presented scheme is not restricted to diatomic molecular ions. For more complex molecules at thermal equilibrium, the number of populated internal states increases, and hence the population of the individual states will eventually be too low to prepare a molecule in a fully specified quantum state. In cases where the density of rovibrational levels is not too high, however, the scheme could be used to specify one specific quantum number by a suitable choice of the intermediate state in the Raman process used to correlate the internal state of the molecule with the external motional mode.

At a more refined level of manipulation and of interest for quantum information processing, the two-ion system discussed here may be used to create and study entanglement between atomic and molecular species.

In conclusion, we presented a method to prepare a single trapped molecular ion in specific states with high confidence. Of great prospect for state-specific investigations, the probabilistic nature of the preparation process makes it possible to access a large number of states within a thermal distribution in a relatively simple experimental way.

### Acknowledgments

We thank K Mølmer for discussions. LBM was supported by the Danish Natural Science Research Council (grant no 21-03-0163) and the Danish Research Agency (grant no

2117-05-0081). MD was supported by the Danish Natural Science Research Council, the Danish National Research Foundation through the Quantum Optics Center QUANTOP and by the Carlsberg Foundation.

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