Towards a Molecular Ion Qubit

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Abstract We discuss the application of a novel spectroscopy protocol to study the Zeeman structure of single molecular ions. From molecular structure calculations for $^{14}N_2^+$ and $^{16}O_2^+$ we deduce their Zeeman spectra, and discuss their potential to be used as a resource to encode quantum information.

1 Introduction

Information is central to our society, but for it to become a major commodity, a century of theoretical and technological developments were needed. Central to our understanding, processing and communication of information is the notion of bit, the smallest quantum of information, that can usually take on the values zero or one. As Shannon showed [33], bits also adapt beautifully to the type of logic, machines and electrical circuits that conform our technology. In other words, all classical technology, from the earliest machines to the latest transistor-based circuits accommodate perfectly to the paradigm of Boolean logic and our classical understanding of information as a collection of symbols.

The developments in the last decade have seriously challenged the *statu quo* in information processing. To start with, we are rapidly approaching the limits of what is currently achievable by silicon integrated technologies. As we reduce the size of circuits and strive for faster and less energy-consuming information processing, or as we increase the storage density of information, we get closer to the nanoscale, a physical domain governed by the rules of quantum mechanics. At these scales the states of electrons, atoms and molecules are ill defined and, while the theory is deterministic, the outcomes of measurements are not. More precisely, we no longer

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have 0's and 1's, but *quantum bits* or *qubits*, which are superpositions of these possibilities with different (complex) amplitudes. Surprisingly, this apparent complexity turns out to be an advantage. A novel information theory can be built using the concepts and tools from quantum mechanics [26], centered on the notions of qubit and quantum operations. This new theory accommodates new types of algorithms for standard procedures, such as factorization, database search or unbreakable cryptography, with a performance that sometimes exceeds exponentially that of their classical counterparts. The double promise of a technology at the nanoscale which is more efficient both energetically and computationally, has spun an extraordinary development of quantum technologies at all levels: from superconducting circuits, quantum dots and nano-mechanical resonators down to atoms and molecules.

In general terms a qubit can be made of any two-level quantum system. Among the many possible ways or possible platforms for quantum information, trapped ions have many advantages. The ions can be trapped by electric fields and/or cooled down and manipulated with laser sources. So far the practical materialization has been mainly carried out using *atomic* ions. In this regard there are typically three types of qubits using the energy levels of an ion:

a) *Zeeman qubits.* Here the energy splitting between the qubit levels is linearly dependent on the magnetic field B. They usually have lifetimes on the order of milliseconds or larger. One obvious drawback of the Zeeman qubit is that since the energy difference between the two qubit states depends linearly on the magnetic field, any relevant magnetic field fluctuations in the environment of the ion will cause dephasing, which in practice limits their applicability, unless a suitable pair of states can be found where this effect is not present, at least to first order.

b) *Hyperfine qubits.* In isotopes with non-zero nuclear spin, we can have hyperfine splittings. At low magnetic field there is also a linear dependence that can remove the degeneracy of different Zeeman levels. A clear advantage of the hyperfine qubit choice is that pairs of levels can be found such that their energy separation does not depend, to first order, on the magnetic field (we present below a molecular example, where this is the case too). Hyperfine qubits are extremely long-lived (with lifetimes on the order of thousands to even millions of years!) and phase/frequency stable, which has led to their application for defining time and frequency standards.

c) *Optical qubits.* In atomic ions with a low-lying D level, such as $^{40}\text{Ca}^+$, there exists the possibility of using the $S \rightarrow D$ optical transition. This typically gives a lifetime of the order of seconds, not large but long enough to perform logic gate operations (on the order of microseconds). There is therefore a rather wide range of possible applications and laboratory implications. In the case of molecules this is a less popular choice and we will not consider it here.
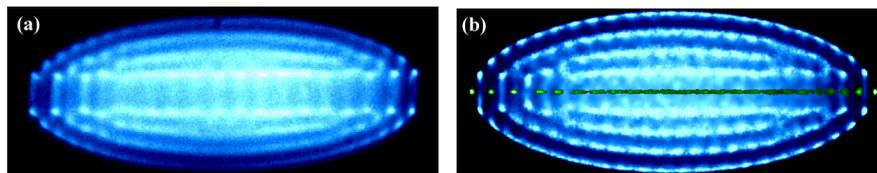
Fig. 1 (a) False-color fluorescence image of 24 quantum-state selected, sympathetically-cooled N_2^+ ions embedded into an ensemble of localized, laser-cooled Ca^+ ions in an ion trap. Because the molecular ions are not laser cooled and therefore do not fluoresce, their location is only visible as a non-fluorescing region in the center of the image. (b) Simulation of the experimental image. The distribution of the molecular ions has been made visible in green. Adapted from Ref. [39].

2 Quantum Technology with Molecular Ions

With all these possibilities at hand with an *atomic ion*, it is immediate to enquire about the use of *molecular ions* instead. A molecule presents the advantage of much closer energy levels (rotation and vibration vs only electronic in atoms) that gives, in principle, longer lifetimes for excited states since the Einstein's coefficients are proportional to the third power of the radiation frequency. As in most situations these features have advantages and inconveniences. The most obvious advantage is the large number of energy levels and frequency scales available in molecules that envisages many possible future applications, e.g., as novel types of qubits or as interfaces between other quantum systems which operate on different frequency scales. Moreover, the intricate couplings between the different angular momenta in molecular systems often lead to a breakdown of the linear dependence of the level energies with applied magnetic fields at smaller values than for atomic ions, which opens up new opportunities for the design of qubits with minimal decoherence. These in turn could serve, for instance, as long-lived quantum memories as a part of a hybrid quantum system. In this context, maybe the biggest advantage of molecules lies in their enormous chemical diversity: it is practically always possible to find (or indeed "engineer") a molecule with the desired properties for the relevant application.

On the other hand, the large density of quantum states in molecules complicates their preparation in *single* states as well as their coherent manipulation, which are fundamental requirements for quantum information processing, novel approaches to precision spectroscopy [3, 30, 31] and quantum-controlled chemistry [28, 39, 40]. Moreover, the large density of states implies the presence of a variety of decay and dephasing mechanisms which render the implementation of high-fidelity quantum operations in molecules very challenging.

However, over the past years there has been impressive progress in the localization of single molecular ions in traps by sympathetic cooling of their translational motion with laser-cooled atomic ions [23]. Very recently, also the initialization of sympathetically-cooled molecular ions in well-defined quantum states has been accomplished, either by optical pumping in molecules like MgH^+ [34] and HD^+ [3, 32] or by the state-selective generation of the ions prior to sympathetic cooling in N_2^+ [35, 36] (see Fig. 1). These developments now pave the way for the

first realization of coherent experiments with single, localized molecular ions in traps.

With atomic ions a number of very accurate preparation, manipulation and measurement techniques have been developed [13], among which we remark the detection by electron shelving. Unfortunately, no such scheme is generally available for molecular ions due to the general absence of cycling transitions. This has led to using alternative detection methods, such as laser-induced charge transfer [35, 36] or state-selective photodissociation [3, 32, 34], which, however, entail the destruction of the molecular ion and are hence inapplicable if repeated measurements are desired, as in quantum metrology [31] or quantum information [13].

In a recent work [24] we have proposed a fast, accurate and *non-destructive* spectroscopy protocol suitable to address the different internal degrees of freedom – rotational, vibrational, hyperfine– of molecular ions. Here, we briefly review the main ideas underlying that work and focus on addressing hyperfine states of a molecular ion, and go forward to analyse the prospects of two particular molecular ions, $^{14}N_2^+$ and $^{16}O_2^+$, for quantum information processing tasks.

3 Phase-sensitive Quantum Logic Spectroscopy

One can define spectroscopy as the study of the effect of a periodic force or driving on a physical system of interest. Typically, one observes the back-action of the system on the driving field, e.g., by measuring absorption spectra, or how the intensity or phase of the driving field is changed by the interaction with the system. In Ref. [31] the idea of quantum logic spectroscopy (QLS) was introduced, which advocates a careful study of the changes on the driven system itself (in that case, an Al^+ ion), by using quantum gates to enhance the precision of the measurements performed. The protocol utilized in [31] had stringent requirements on the temperature of the ions studied, and the overall speed of the process was limited by using the collective motional modes of the system as a quantum bus. By appealing to ideas from coherent control, it is possible to remove these limitations [11, 12]. We have resorted on these ideas to design a novel protocol that extends QLS to molecular ions. In the following, we present a brief summary of the main concepts involved, and then we apply them to analyze two molecular ions in the light of their potential use as qubits.

As it was shown in [11, 12, 19], it is possible to increase the versatility and robustness of the original QLS protocol by using geometric gates based on state-dependent forces. Geometric gates are quantum gates whose performance does not depend on the motional state of system and are thus particularly appealing to address sympathetically-cooled ions, such as molecular ions trapped in Ca^+ or Be^+ Coulomb crystals. The physical principle at the root of these quantum operations is that a chain of ions shaken by an external driving will have its quantum state modified by the acquisition of a phase depending on the driving forces. Now, for the particular case of the system being two trapped ions, if the forces applied on them
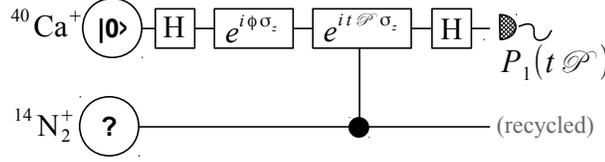
Fig. 2 *State detection protocol.* The control ion is subject to a series of single-qubit gates together with a two-qubit gate in which the observable property $\mathscr{P}$ of the spectroscopy ion induces a state-dependent phase on the two-ion quantum state. Measurement of the control ion then allows access to information on $\mathscr{P}$.

are such that their effect depends on the internal state of the ion (as can be realized, e.g., by applying a laser of a given polarization that can only affect one internal state of the ion), then the phase accumulated by the two-ion system will also depend on their internal states. As a consequence, if one is able to measure this accumulated phase (e.g., via an interferometric process), one will gain information on the state of the trapped ions.

Let us consider a system composed of two trapped ions, a control or logic ion (that in our case will be an atomic ion such as $^{40}\text{Ca}^+$ or $^9\text{Be}^+$) and a spectroscopy ion (such as a molecular ion). The first ion is actively laser-cooled, while the spectroscopy ion is sympathetically cooled by the control ion thanks to the Coulomb interaction. Let us assume that the control ion can be described as a qubit and that we apply some force on it which depends on its internal state, $f_C(t)\sigma_C^z$, where σ_C^z is the Pauli z-matrix for the internal-state space of the control ion. On the other hand, the spectroscopy ion can have in principle any number of internal levels, and applying a force to it will depend on some internal property, $\mathscr{P}$, such as a magnetic moment of a dipole moment, $f_S(t)\mathscr{P}$. As explained in Ref. [24], under the influence of these forces, the quantum state of the two-ion system will acquire a total phase due to both forces $f_{C,T}$ given by

$$\Phi_{\mathscr{P}} = \sigma_C^z \mathscr{P} \phi_{CS},\tag{1}$$

where ϕ_{CS} is a function of the collective modes of the two-ion complex, ω_{com} and ω_{str}, and of the forces applied on the ions. Let us emphasize at this point that the forces on the control and spectroscopy ions are independent, and do not need to have a common origin or even be based on the same effect. In particular, below we consider applying an AC Stark shift to the atomic ion, and a magnetic field gradient (Zeeman shift) on the molecular ion.

The phase 1 does not depend on the motional state of the ions at the start of the operation, which makes the gate temperature-independent and robust. This allows us to design the following spectroscopy protocol (cf. Fig. 2a) to accurately measure the phase $\Phi_{\mathscr{P}}$ and, hence, obtain a measurement of the physical observable $\mathscr{P}$.

1. Prepare the control ion in the initial state $|0\rangle$;
2. apply a Hadamard gate on this ion, $H = \exp(-i\sigma_C^y\pi/2)$; in practice, this amounts to starting a Ramsey sequence;

3. optionally, apply a reference phase on the control ion, $\exp(i\phi\sigma_C^z)$;
4. apply the state dependent forces $f_{C,S}(t)$ on the control and spectroscopy ions, ensuring that the initial motional state of the ions is restored (see Sect. 4);
5. close the Ramsey sequence by applying again a Hadamard gate, H; and
6. measure whether the control ion is in the excited state $|1\rangle$.

After steps $1-5$, the control ion and the spectroscopy ions will be entangled, and the excited state population of the control ion will oscillate as

$$P_1 = \sin^2(t\mathscr{P}\phi_{CS} + \xi). \tag{2}$$

The measurement step 6 projects the entangled state and, after reinitializing the control ion to $|0\rangle$, a repeated application of the protocol allows to determine the value of $\mathscr{P}$ with high precision [15].

4 QLS protocol for molecular Zeeman states

In this section we present a particular application of the protocol described in Sec. 3 when one is interested in discriminating the Zeeman levels within a rovibrational level of the molecular ion of interest. The different magnetic-field dependence of the Zeeman shifts for different levels result in general in different magnetic moments for each state. Hence, we envision to use the magnetic moment as our observable of interest, $\mathscr{P} = \mu$, and to use of magnetic field gradients to couple to it [16, 22]: $f_S(t) = \mu\partial B(t)/\partial x$. Without loss of generality, we take the forces to be characterized by a driving frequency ν and have a Gaussian envelope of duration T:

$$f_{C,S}(t) = f_{C,S}^0 e^{-(2t/T)^2} \cos(\nu t). \tag{3}$$

With this time dependence, and for a sufficiently long pulse, $T \geq 5\pi/\tilde{\omega}$ ($\tilde{\omega} = \min\{|\nu - \omega_{com}|, |\nu - \omega_{str}|\}$), one can ensure that *all* motional degrees of freedom of the system are restored to their initial state, and the two ions accumulate a quantum phase given by Eq. (1) with (cf. Ref. [24])

$$\phi_{CS} = \frac{1}{4}\sqrt{\frac{\pi}{2}} \frac{f_C^0 f_S^0 a^2 T}{\omega} \left[\frac{\omega^2}{\omega_{com}^2(\tilde{m}) - \nu^2} - \frac{\omega^2}{\omega_{str}^2(\tilde{m}) - \nu^2} \right], \tag{4}$$

where ω is the frequency of the control ion trap, $\tilde{m} = m_T/m_C$ is the mass ratio of the two ions, and $a^2 = \hbar/[(m_C + m_T)\omega]$. We note that the driving frequency ν is a free parameter. This can be useful for the case of ions with small magnetic moments, where one can set ν close to one of the collective mode frequencies of the system to enhance the effect (at the expense of a longer gate time, see [24] for details).

Using this protocol it would be possible to study the Zeeman structure of, for example, $^{14}N_2^+$ molecular ions trapped with $^{40}Ca^+$ as in [35,36]. We have performed structure calculations to estimate the magnetic moments of the corresponding states
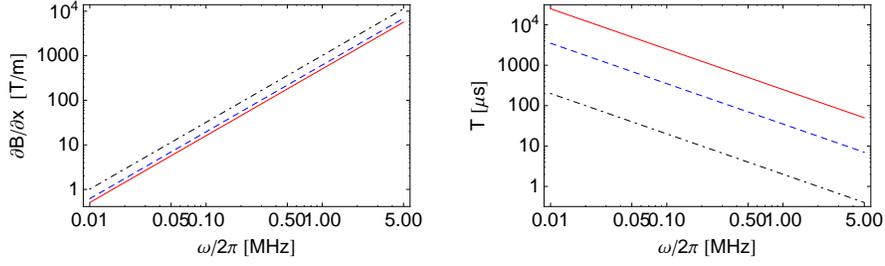
Fig. 3 *Experimental requirements.* Gradient (left) and gate time (right) required to perform the spectroscopy protocol with $^{40}\text{Ca}^+$ and $^{14}\text{N}_2^+$ ions, utilizing a magnetic field gradient $\partial B/\partial x = \exp[-(2t/T)^2]B' \cos(\nu t)$ in a harmonic trap of frequency ω, for driven at frequencies $\nu/\omega = 0$, 1.1 and 1.01 (dash-dotted, blue dashed, red solid).

and found them to be on the order of the Bohr magneton, see the left panels in Fig. 4. With this, it is straightforward to calculate the required field gradients to realize the spectroscopy protocol, assuming a laser is driving AC Stark forces on the $^{40}\text{Ca}^+$ ion. It turns out that for typical trap frequencies $\omega/2\pi = 10^2 - -10^3$ Hz, field gradients on the order of 10 T/m applied during a few hundred microseconds will suffice, see Fig. 3. Moreover, even lower gradients and shorter times can be used in case of smaller trap frequencies, making this approach very attractive.

5 Towards a molecular ion qubit: comparison of $^{14}\text{N}_2^+$ and $^{16}\text{O}_2^+$

As an example of the different features that molecules may exhibit in this context, we have considered the effect of an external magnetic field in the lowest energy levels of $^{14}\text{N}_2^+$ and $^{16}\text{O}_2^+$. We write a general Hamiltonian as

$$H = H_0 + H_Z, \tag{5}$$

where H_0 is the field-free molecular Hamiltonian and H_Z represents the interaction with the magnetic field. The ground electronic state of $^{14}\text{N}_2^+$ is $^2\Sigma_g^+$ and, in this way, H_0 involves rotational, spin-rotation and, in addition, hyperfine interactions (the nuclear spin of ^{14}N is one). The $^{16}\text{O}_2^+$ cation, on the other hand, has a $^2\Pi_g$ ground state and the zero-field Hamiltonian includes rotational, spin-orbit, spin-rotation and Λ doubling terms [8]. There is no hyperfine structure involved since the ^{16}O nuclei have zero spin. Another consequence of the oxygen nuclear spin statistics is that one of the two Λ-doublet components is always missing for each value of the total angular momentum J [8]. All this will make the $^{16}\text{O}_2^+$ spectrum much less congested than that of $^{14}\text{N}_2^+$. Differences also arise regarding the coupling with the applied magnetic field. First, the leading term of the Zeeman Hamiltonian for a $^2\Sigma$ molecule such as $^{14}\text{N}_2^+$ reads

$$H_Z(\mathrm{N}_2^+) = \mu_B B g_S S_Z, \tag{6}$$

where g_S is the electron spin gyromagnetic factor (taken as $g_S = 2.002$), B is the magnetic field, μ_B is the Bohr magneton, and S_Z is the component of the electronic spin along the direction of the external field. This interaction differs in $^{16}\mathrm{O}_2^+$ since the orbital motion of the unpaired electron also couples to the field so, in this case,

$$H_Z(\mathrm{O}_2^+) = \mu_B B \left(g_L L_Z + g_S S_Z \right), \tag{7}$$

where g_L is the electron orbital g-factor (taken as $g_L = 1$) and L_Z is the projection of the electronic orbital angular momentum onto the external field.
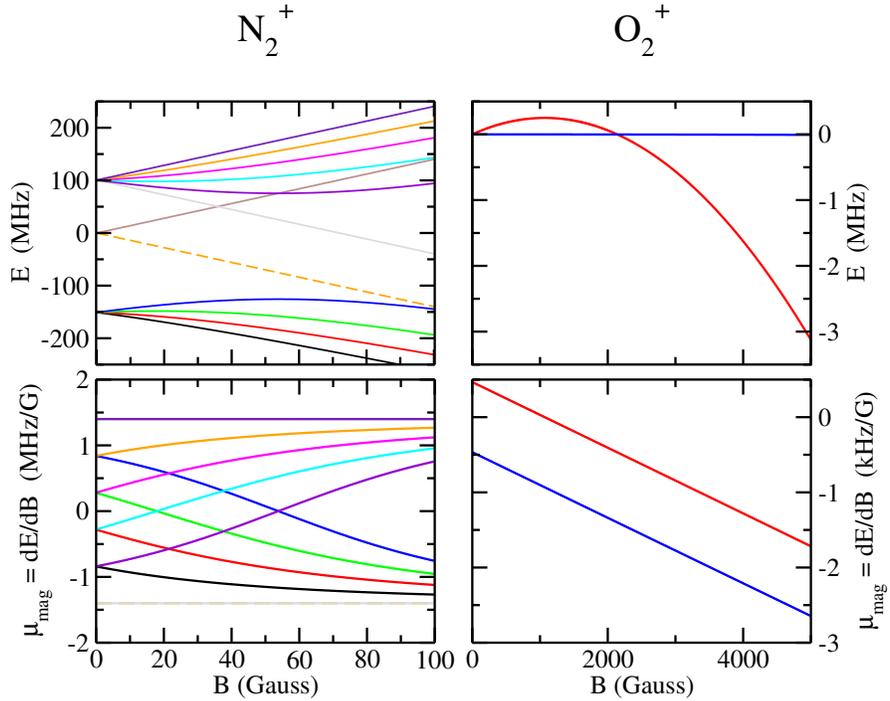

Fig. 4 *Zeeman diagrams.* Upper panels: Zeeman energy shifts of the ground rovibronic states of $^{14}\mathrm{N}_2^+$ (left) and $^{16}\mathrm{O}_2^+$ (right). Lower panels: $^{14}\mathrm{N}_2^+$ (left) and $^{16}\mathrm{O}_2^+$ (right) magnetic moments, $\mu = (\partial E/\partial B)$, for the levels above.

The Zeeman splittings and corresponding magnetic moments, $\mu = \partial E/\partial B$, of the rovibrational ground states of $^{14}\mathrm{N}_2^+ \left(^2\Sigma_g^+\right)$ and $^{16}\mathrm{O}_2^+ \left(^2\Pi_g\right)$ as functions of B are shown in Fig. 4. For nitrogen, the hyperfine structure at zero field gives three distinct sublevels corresponding to total angular momenta $F =$, 3/2, 1/2 and 5/2 in order of ascending energy (for total nuclear spin $I = 2$, 0 and 2, respectively). Details of the calculations and the molecular constants used can be found in Ref. [24]. For $^{16}\mathrm{O}_2^+$,

there is only one level at zero field corresponding to a total angular momentum, $F = J = 1/2$. The calculations at various values of B were performed by diagonalizing the Hamiltonian matrix using a Hund's case (a) representation [4, 5] and taking the molecular constants for the ground vibrational state from the work of Coxon and Haley [8] (a more complete account will be given elsewhere [25]). We note that, for both ionic species, the levels plotted in Fig. 4 are well separated from the next upper rotational levels of both ionic species studied (energy splitting of about $\hbar \times 10^5$ MHz $\sim k_B \times 4.8$ K) .

It is quite noticeable the very different behavior of both ions in the presence of a magnetic field. The $^{14}N_2^+$ levels have a much stronger dependence with B leading, in most cases, to rather high values of the state-dependent magnetic moments: for the highest fields they tend to μ_B in absolute value, and for lower fields ($B < 80$ G) almost all states are distinguishable by the values of their magnetic moments, this feature serving for efficient non-destructive measurements as proposed in Ref. [24]. For oxygen, on the contrary, the magnetic sublevels depend extremely weakly on B: notice the very different scales in the axes of the graphs with respect to nitrogen. This is due to the fact that for this level, the Zeeman effect is governed by $g_L - g_S/2 \approx 0.001$ (see Ref. [20] for a discussion). Hence, the $^{16}O_2^+$ magnetic moments are about 1000 smaller than the $^{14}N_2^+$ ones. As a consequence, the two lowest Zeeman levels in the ground rovibrational state of $^{16}O_2^+$ will feature very small coupling to ambient magnetic field fluctuations. In other words, they may constitute a convenient two-level (qubit) system for storage of quantum information.

We finally note that the couplings with higher molecular states (off-diagonal matrix elements) play an important role in the dependence of the energy levels with the field, since they are the cause for the departure of a linear dependence with B [17]. Indeed, it can be seen that the magnetic moments are not exactly constant with B, and that the one corresponding to the upper level ($M = -1/2$, in red in the Fig. 4) changes sign at about 1000 Gauss.

6 Conclusions and outlook

In conclusion, we have presented a non-destructive molecular spectroscopy protocol and discussed in detail its application to study the Zeeman spectrum of molecular ions. From an analysis of the calculated spectra of $^{14}N_2^+$ and $^{16}O_2^+$, we deduce that the lowest Zeeman levels of the latter constitute an promising candidate to realize a quantum memory.

In the future, we will study the application of this protocol to other molecular ions of interest, and analyse in further detail the susceptibility of each one to ambient fields, so as to determine the best candidates for several quantum information tasks.

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