Metastability transfer spectroscopy with two like ions in the same trap

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ABSTRACT High-resolution optical spectroscopy of an individual trapped ion is hampered by lack of sharp lasers. This suggests the use of a second metastable excited ion as an ultrasharp light source. To this end, laser-cool two barium ions to an equilibrium distance of ~8 μm on the z (symmetry) axis of the trap and, in this (earth)(Ba')2-molecule, visually or photoelectrically identify them as A and B by their location. Briefly turn on a 455-nm spectral lamp until one of the ions, say the A ion, is pumped into the metastable D_{5/2} level and turns invisible. Focus on the visible, spatially well-resolved B ion and turn off the blue and red illumination lasers for ~15 s. Then turn them back on again and check on whether the excitation by chance has been transferred to the B ion and is now in the D_{5/2} level and dark while the A ion is bright. The cross section for absorption of the \( \Delta (D_{5/2} \rightarrow S_{1/2}) = \lambda_0 = 1.76 \mu m \) radiation by a stationary ion can be \( > \lambda_0^2/2 \pi \). Thus, by pushing the two ions together to \( \lambda_0/4 \) by turning on a much stronger trapping field during the excitation exchange period, one might be able to detect excitation transfer in >10% of the attempts. The ions are tuned relative to each other by a 0- to 10-mV/cm variable dc field in the z direction, which displaces them axially and causes them to see different rf fields, which Stark-shifts their frequencies. In this way, a resonant transfer response as sharp as twice the natural width of the D_{5/2} level, 11 MHz or a \( Q \approx 0.4 \times 10^{12} \), might be demonstrated.

With the perfection of techniques used to trap and laser-cool individual atomic ions, considerable progress has been currently in ultimate-resolution atomic spectroscopy in the optical region. Single barium (1, 2) and mercury (3) ions have been trapped for relatively long periods of time and Doppler-free optical spectra (1, 3) have been taken. In addition, the technique of “shelving” (4) has been demonstrated (3, 5, 6), allowing one to achieve the theoretically maximum signal to noise in optical spectroscopy experiments on single ions. Currently the principal resolution-limiting element in such experiments is the finite breadth of the laser. We propose a series of experiments that bypass this problem by employing a second single ion of the same species or an emitter of extremely spectrally narrow radiation.

The experiment would make use of the essentially the same apparatus as used in the earlier “quantum jump” experiments (5). In this case, two ions would be simultaneously trapped. Very early compression of two barium ions to \( d = 4 \mu m \) in a (radial) well with \( \nu_e = 0.8 \) MHz was demonstrated (2). Thus they form essentially an (earth)(Ba')2-molecule in a low vibrational state. By applying a sufficiently large negative voltage to the ring electrode (with respect to the end caps), one would force the two ions to align themselves with the \( z \) axis (symmetry axis) of the trap; one thereby makes the “spring constant” in the radial direction greater than that in the axial direction (7). It can be shown (8) that in a rf trap whose axial frequency is \( \nu_z \), the equilibrium distance, \( d \), between two barium ions confined to the \( z \) axis will be \( d = (e^2/2 \pi \nu_z m)^{1/2} = 3.6 \nu_z^{-1/2} \) (with \( d \) in μm and \( \nu_z \) in MHz), where \( e \) is the electric charge and \( m \) is the ion’s mass.

One of the ions is now excited to the \( 5^2D_{5/2} \) level by turning on a filtered barium hollow cathode lamp, which causes \( 6^1S_{1/2} \rightarrow 6^3P_{3/2} \) transitions only (via radiation at 455 nm). After a few such transitions, the ion will end up in the metastable \( 5^2D_{5/2} \) level. The state of each ion is monitored by focusing the strong \( 6^3P_{1/2} \rightarrow 6^3S_{1/2} \) fluorescence from each ion onto a small pinhole, which allows only light from that ion to reach a photomultiplier tube (PMT). Two PMTs or pinholes would be used: one for each ion. The photomultipliers and associated optics for each ion would be conveniently placed on opposite sides of the trap. (It might in practice be simpler to use a moveable pinhole and a single PMT to sample at will the light from each ion.) Excitation rates to the metastable level may be of ~1 s^{-1} and one ion will obviously be excited first. When this occurs (as determined by the PMTs), the lamp is immediately turned off and the excited ion is allowed to decay via electric quadrupole radiation; the average decay time is one lifetime (5) of the metastable level (32 s). To prevent broadening of the transition, the red and blue ("cooling and interrogating") lasers will be turned off for 15–30 s. At the end of this period, the interrogating lasers would be turned on to determine whether an excitation transfer took place. When the excited ion decays, the probability of transferring its excitation to the second ion is \( P = \lambda/Q/4\pi \) (\( \lambda_0/d^2 \)), where \( \lambda_0 = 1.76 \mu m \) is the wavelength of the emitted photon and \( Q \) is the absorption cross section. For \( \lambda_0/d = 4 \), this gives \( P > 0.2 \). Realizing \( d = 0.5 \mu m \) or \( \nu_z = 18 \) MHz, though not easy, should nevertheless be feasible. Any difficulty in optically resolving the ions could be reduced by reducing the well depth during the interrogation phase of the experiment. For example, the ion separation could be increased to \( 8 \mu m \) by reducing the frequency to 0.3 MHz.

The ions could be tuned relative to one another by placing them in a magnetic field with a small controllable gradient. Assuming a magnetic field sensitivity of ~1.4 MHz/G and a linewidth of 0.005 Hz, a field gradient that varied from 0 to \( 1 \times 10^{-4} \) G/cm would suffice to tune the ion over one linewidth (1 G = 0.1 mT). Suitable magnetic shielding is obviously required.

Another tuning technique would exploit the Stark shifts due to the trapping rf fields (9). We estimate this shift as \( \delta_S = -24.6^2 \text{MHz}/(V/cm)^2 \), the value that has been measured (10) for the sodium D lines. This is justified because the S-P level separations and the P-D level separations in Na and Ba are about the same in absolute value. Therefore, one expects that the down-shift of the S level is about the same in Na and Ba. Likewise, the shifts of the Ba'' D level and of the Na P level cannot be very different either. The shift \( \delta_S \) may of course be calculated quite accurately from the known Ba'' energy levels.

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Thus, the ions can be tuned over one linewidth by displacing them until there is about a 10-mHz difference in their Stark shifts, $\delta_S$. For $d = 0.5 \mu m$, the dc field generated by one ion at the site of the other is $60 V/cm$, which implies a rf field (1) that is larger by a factor (8) of $\sqrt{2}\Omega/\omega_h > 2\sqrt{2}$, or $E_{rf} \approx 200 V/cm$, where $\Omega$ is the frequency of the trapping field and $\omega_h$ is the secular frequency. This produces a Stark shift, $\delta_S = 100 Hz$. To generate a differential of $10^{-2} Hz$ or 1 part in $10^4$, the dc field $= 60 V/cm$ at the ions must be changed by $\pm 0.25$ part in $10^4$ or 1.5 mV/cm, which is done by applying such a dc field—namely, $\pm 0.1 mV$ across the end caps of the trap.

Metastability exchange may initially be greatly speeded up by Stark-quenching by means of a manageable auxiliary $\approx 100-\text{MHz}$ field. As a warm-up exercise here we will study with a rf field applied between the cap electrodes the Stark-quenching of the $D_{5/2}$ level in a single ion.

Thus, we see that this experiment using two laser-cooled ions in a rf trap can obtain extremely narrow resonances. Such a system could study the intrinsically interesting problem of resonant excitation transfer between two nearby atoms essentially at rest; it would also set a record in spectral narrowness that would not be improved upon until a breakthrough in the stability of laser local oscillators occurs.

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