Precise spectroscopy for fundamental physics ☆

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We have applied experimental techniques that were developed for use in atomic frequency standards and clocks to investigations of local Lorentz invariance, the linearity of quantum mechanics, and anomalous long-range spin-dependent forces. These experiments used a hyperfine transition in $^{9}Be^{+}$ ions in a Penning trap. Recently, we have studied hyperfine transitions in $^{199}Hg^{+}$ ions in a linear rf trap. Hg^{+} ions might be used for similar investigations in the future.

1. Introduction

Ion traps are devices in which charged particles, such as ions or elementary particles, can be suspended by various combinations of electric and magnetic fields [1,2]. Since the ions are held in a vacuum, it is possible to measure internal resonance frequencies relatively unperturbed by collisions with gas molecules or with the walls of the enclosing chamber. An atomic beam has similar advantages in this regard, but the measurement time for an atom is limited to the flight time of the atoms through the apparatus, normally much less than one second. Ions can be held in traps for very long periods, even days. For these reasons, hyperfine and other internal resonances can be observed with very high resolution with trapped ions. Compared to neutral atoms, though, the numbers of ions that can be studied is low, because Coulomb repulsion places a limit on the densities that are possible. This limits the signal-to-noise ratio that can be achieved with trapped ions.

These techniques can be used to measure atomic and nuclear properties to great precision; they can be used to test fundamental physical principles or to search for unknown interactions. The general method is to devise a test theory that has an adjustable parameter. When the parameter is zero, the theory agrees with ordinary physics. A nonzero value, on the other hand, indicates a new physical effect. We set up experiments in which the existence of a nonzero parameter would lead to the

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shift of a resonance frequency. Such experiments have also been done by others with neutral atoms, often searching for the same effects as the trapped ion experiments.

2. Beryllium ion resonance

We have used a particular hyperfine transition in the ground state of the $^9Be^+$ ion for several tests of fundamental physical principles. The ground electronic state has the configuration $2s\,^2S_{1/2}$. The ^9Be nucleus has spin 3/2, so there are eight hyperfine-Zeeman states. In a high magnetic field, as is present in the experiments, the energy eigenstates are approximate eigenstates of I_z and J_z, the z-components of the nuclear and atomic angular momenta. Fig. 1 shows the hyperfine sublevels of $^9Be^+$ in a magnetic field. They are labeled by their main components in the $|m_I, m_J\rangle$ basis. The transition between the levels marked 1 and 2 at frequency ν_0 is of particular interest. At a particular value of the magnetic field B, near 0.8194 T, the first derivative of ν_0 with respect to B goes to zero. For small deviations δB from this field, the fractional shift of the transition frequency is only $\delta\nu/\nu_0 \approx -0.017(\delta B/B)^2$. For this reason, this transition, which has a frequency of 303 016 377.265 Hz, has been used in experimental atomic clocks and frequency standards [3,4].

3. Experimental methods

The experiments described here have taken place over several years, during which time the apparatus has undergone many technical changes. Details have

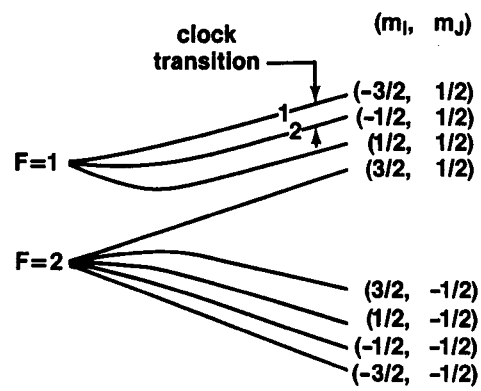

Fig. 1. Hyperfine structure of the $^9Be^+$ $2s\,^2S_{1/2}$ ground state as a function of the magnetic field B. At a particular value of B near 0.8194 T, the derivative with respect to B of the transition frequency between the levels labelled 1 and 2 is zero.

been published previously [3–7]. However, the basic principles have remained the same. The $^9Be^+$ ions, in numbers ranging from a few up to tens of thousands, are confined in a Penning trap. A Penning trap consists of electrodes which create an electrostatic quadrupole potential, combined with a uniform magnetic field. This combination of fields traps ions in all three dimensions. In the form used in recent experiments [4,6,7], the Penning trap consists of hollow cylindrical electrodes, to which static electric potentials are applied. It is inserted into the bore of a superconducting solenoid magnet, which generates a uniform magnetic field of approximately 0.82 T. In the earlier experiments [3,5], the trap electrodes had approximately hyperboloidal surfaces, and a conventional electromagnet generated the magnetic field. The pressure in the trap was approximately 10^{-8} Pa. The ions were created by electron-impact ionization of neutral atoms.

Laser cooling [8,9] of the ions reduces the Doppler shifts of the resonance frequencies and is necessary in order to achieve the present levels of accuracy. The 313 nm transition from the 2s $^2S_{1/2}$ to the 2p $^2P_{3/2}$ electronic state is used for state selection and detection, as well as for laser cooling. It is necessary to turn off the 313 nm light for some periods in order to avoid perturbations of the energy levels. In the earlier experiments, the need to turn off the 313 nm light limited the frequency resolution, since the $^9Be^+$ ions heated and increased their spatial extent when the light was not available for laser cooling. In the more recent experiments, $^{26}Mg^+$ ions were trapped and laser cooled at the same time. They cooled the $^9Be^+$ ions by long-range Coulomb collisions [10]. The 280 nm transition between 3s $^2S_{1/2}$ and 3p $^2P_{3/2}$ was used for laser cooling the $^{26}Mg^+$.

The 313 and 280 nm beams required for state selection and detection of the $^9Be^+$ ions and for laser cooling of the $^{26}Mg^+$ were generated by frequency doubling the outputs of cw dye lasers in nonlinear crystals. Fluorescence from the atoms was focused by a lens system onto the photocathode of either an imaging photon-counting tube or a simple photomultiplier tube.

Multiple resonance was used to drive the rf resonance between states 1 and 2 to measure the number of ions that underwent a transition (see fig. 2). In the more recent experiments, the measurement sequence was as follows: The ions were subjected for about 10–20 s to 313 nm radiation, polarized perpendicular to the magnetic field. The frequency of the 313 nm radiation was slightly below the 2s $^2S_{1/2}$ ($m_I = +3/2$, $m_J = +1/2$) to 2p $^2P_{3/2}$ ($m_I = +3/2$, $m_J = +3/2$) transition frequency. This is a cycling transition, since electric dipole selection rules require that the ion return to the ground-state sublevel from which it starts. Spontaneous Raman transitions, induced by the 313 nm radiation, established a steady state in which approximately 16/17 of the ions were in state 4 and the remaining 1/17 were in state 5. This optical pumping has been discussed previously [11,12] and studied experimentally [13]. The 313 nm beam was then turned off to stop the optical pumping and to prevent perturbations to the $^9Be^+$ energy levels. Next, the ions in state 4 were transferred to state 3 and then to state 2 by on-resonance rf pulses. Ramsey's method of separated oscillatory fields [14] was then used to drive the transition

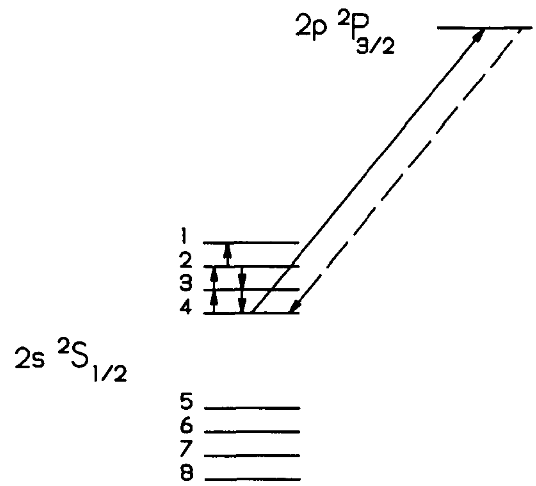

Fig. 2. Level diagram showing the transitions involved in the state preparation and detection for the $^9Be^+$ $2s\,^2S_{1/2}$ $(m_I = -3/2, m_J = 1/2)$ to $(m_I = -1/2, m_J = 1/2)$ (state 1 to 2) transition in the electronic ground state.

from state 2 to state 1. In Ramsey's method, two coherent rf pulses are applied, with a delay between them. The main advantage over applying a single rf pulse of the same total duration is that the resonance linewidth is about a factor of two smaller. Typically, the rf pulses were 1 s long. The delay between the two pulses was varied up to as long as 550 s (see fig. 3). The frequency resolution is inversely proportional to the time between the rf pulses, if the durations of the rf pulses are short compared to the time between them. For 550 s between the pulses, the reso-

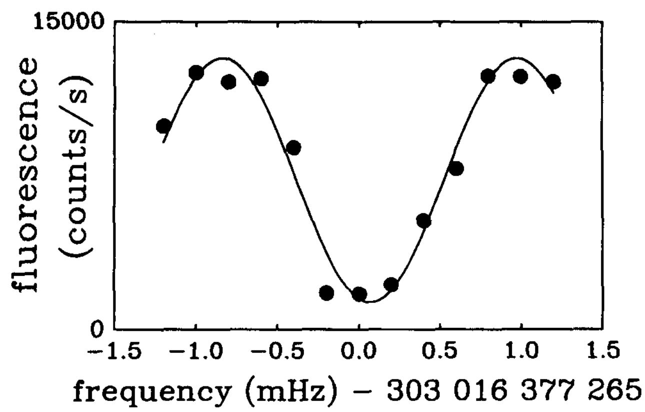

Fig. 3. Example of a 303 MHz $^9Be^+$ hyperfine resonance obtained by Ramsey's method of separated oscillatory fields. The time between the two rf pulses was 550 s. The width of the resonance is 0.9 mHz (0.0009 Hz). (From ref. [4].)

nance linewidth is 900 μHz. After applying the rf pulses, the number of ions in state 2 was measured. First, the ions in state 2 were transferred to state 3 and then to state 4 by applying the rf pulses in the opposite order from that which was used to prepare the ions in state 2. Then the 313 nm beam was turned back on and the fluorescence photons were counted.

4. Local Lorentz invariance

The Einstein equivalence principle is the basis of all metric theories of gravitation, including general relativity [15]. According to the Einstein equivalence principle, (i) all bodies fall in a given gravitational field with the same acceleration (weak equivalence principle), (ii) the outcome of any local nongravitational experiment is independent of the velocity and orientation of the freely falling apparatus (local Lorentz invariance (LLI)), and (iii) the outcome of any local nongravitational experiment is independent of where and when it is performed. It follows from LLI that the relative frequencies of two atomic frequency standards whose positions and orientations are fixed with respect to the same freely falling laboratory frame do not depend on the velocity and orientation of that frame.

There are many possibilities for the form which a breakdown of LLI might take. One which has been extensively studied is the $TH\epsilon\mu$ formalism [15–17]. This formalism allows different values for the limiting speed for material particles c_0 and the speed of light c_L. A specific prediction of this formalism is that the frequency of the $^9Be^+$ transition discussed in the previous section depends on the angle between the magnetic field and the velocity of the laboratory with respect to some preferred frame. The frequency of a hydrogen maser frequency standard is not predicted to vary in this way. The ratio of the $^9Be^+$ resonance frequency to that of the hydrogen maser was measured for different orientations of the magnetic field with respect to the fixed stars [18]. For practical reasons, it was convenient to let the direction of the magnetic field remain fixed in the laboratory frame and let the rotation of the Earth change the orientation. Fig. 4 is a plot of the $^9Be^+$ frequency as a function of the sidereal time. No variation in the frequency was found, within the limits set by the experimental uncertainty of about 100 μHz. If we take the frame in which the cosmic blackbody radiation is isotropic to be the preferred frame, this result implies that the $TH\epsilon\mu$ preferred frame parameter $|1 - c_0^2/c_L^2|$ is less than or equal to 10^{-18}. This limit was about 300 times more stringent than earlier results obtained by conventional nuclear magnetic resonance methods [19,20]. Interpretations of the $^9Be^+$ results in terms of other possible preferred frames or other forms of violations of LLI are given in ref. [18]. Our experiments were followed by experiments in which the nuclear magnetic resonance frequencies of gas-phase neutral atoms were observed by optical pumping techniques [21,22]. These experiments improved the limits on $|1 - c_0^2/c_L^2|$ by about three orders of magnitude.

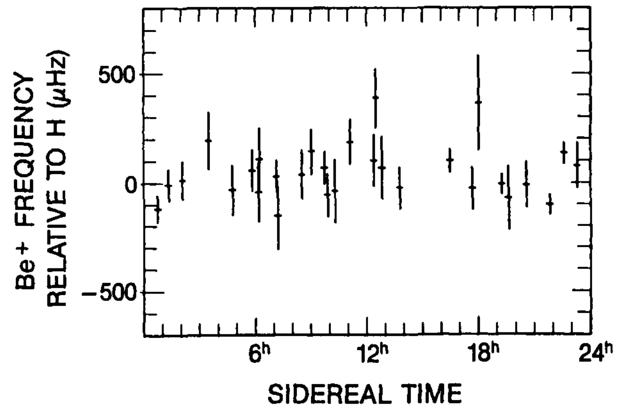

Fig. 4. The $^9Be^+$ transition frequency with respect to sidereal time. The orientation of the trap magnetic field (which defines the quantization axis for the $^9Be^+$ ions) with respect to the fixed stars varies due to the rotation of the Earth. A daily variation of the frequency would indicate a violation of local Lorentz invariance. (From ref. [18].)

5. Nonlinear quantum mechanics

One of the most fundamental properties of quantum mechanics is linearity. The Schrödinger equation is linear, so any linear combination of solutions is itself a solution. Weinberg has formulated a nonlinear generalization of quantum mechanics [23]. It reduces to ordinary quantum mechanics when a parameter in the theory goes to zero.

In Weinberg's nonlinear quantum mechanics, physical systems are still represented by state vectors. However, the time development of the state vectors is governed by a nonlinear equation, rather than by the Schrödinger equation. An energy eigenstate is defined as a state whose time dependence is a simple exponential phase factor $\exp(-iEt/\hbar)$, where E is defined as the energy eigenvalue. The method of determining the eigenstates and eigenvalues differs from that in ordinary, linear quantum mechanics [23]. One of the observable consequences of Weinberg's theory is a different kind of time development for a superposition state. Consider a two-level system having energy eigenstates $|1\rangle$ and $|2\rangle$ and corresponding energy eigenvalues $\hbar\omega_1$ and $\hbar\omega_2$. At time $t = 0$, let the state be the superposition

$$|\psi(0)\rangle = (1-a)^{1/2}|1\rangle + a^{1/2}|2\rangle . \tag{1}$$

In linear quantum mechanics, the state at a later time t is

$$|\psi(t)\rangle = \exp(-i\omega_1 t)\{(1-a)^{1/2}|1\rangle + a^{1/2}\exp[i(\omega_1-\omega_2)t]|2\rangle\} . \tag{2}$$

However, in nonlinear quantum mechanics, the state at a later time is

$$|\psi(t)\rangle = \exp(-i\omega_1(a)t)\{(1-a)^{1/2}|1\rangle + a^{1/2}\exp[i(\omega_1(a)-\omega_2(a))t]2\rangle\}\,. \quad (3)$$

The difference is that, in eq. (3), ω_1 and ω_2 are functions of the relative amplitudes of the two components of the state. Although the overall phase factor $\exp(-i\omega_1(a)t)$ in eq. (3) is not observable, the relative phase factor $\exp[i(\omega_1(a)-\omega_2(a))t]$ is observable.

The Ramsey resonance method is a very sensitive way to search for a dependence of the relative phase factor on the state amplitudes. By varying the product of the amplitude and duration of the first rf pulse, different initial superposition states can be generated. During the time between the pulses, the phase factor oscillates at the frequency $[\omega_1(a)-\omega_2(a)]$.The signal observed after applying the second rf pulse depends on the accumulated phase. If there is a nonlinear contribution to the Hamiltonian, the center of a resonance like that of fig. 3 will depend on the initial amplitudes in the superposition state. In the experiment [7], the initial amplitudes of the two states labeled 1 and 2 in fig. 1 were varied between two superpositions (called A and B), chosen to maximize the predicted effect. The superposition A was $0.4886|1\rangle + 0.8725|2\rangle$ (corresponding to $a = 0.75$). For B, the coefficients were reversed. Fig. 5 is a plot of the center frequency of the resonance as a function of time. The sections labeled A and B indicate the different superposition states. The average difference on frequency between the A and B conditions was $3.8 \pm 8.3\,\mu$Hz, which is not measurably different from zero. This sets a limit of 4×10^{-27} on the fraction of the binding energy per nucleon of the ^{9}Be nucleus that could be due to nonlinear contributions to the Hamiltonian.

Other experiments have searched for nonlinearities of the same general form. Some of them used nuclear magnetic resonance [24,25]. These experiments set

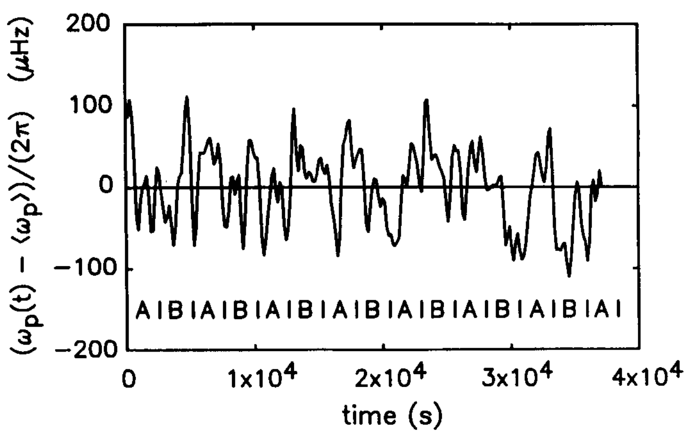

Fig. 5. The oscillation frequency of the coherence between the two $^9Be^+$ states as a function of time. The state amplitudes in the superposition state are varied between the two conditions called A and B, which are described in the text. A systematic variation of the frequency with the state amplitudes would indicate a violation of the linearity of quantum mechanics. (From ref. [7].)

roughly the same limit on the nonlinearity parameter as the $^9Be^+$ experiment. Another used a hydrogen maser, which was operated at different values of the state population inversion [26]. The limit set on the contribution to the energy arising from the nonlinear Hamiltonian was approximately the same as for the nuclear magnetic resonance experiments in absolute energy units. However, it is probably more meaningful to consider the ratio of the nonlinear contribution to the binding energy of the system. In this case, the hydrogen maser experiment is not as stringent a test, because the binding energy of an atomic system is much less than that of a nuclear system. The reason that the hydrogen nucleus cannot exhibit nonlinear behavior of the type postulated by Weinberg is that it has spin 1/2. It is impossible to construct a nontrivial nonlinear Hamiltonian for a spin 1/2 particle which satisfies rotational invariance [23]. However, it is possible for the composite system of a bound proton and electron to exhibit a nonlinear effect. These experiments and the status of the theory are reviewed in ref. [27].

6. Anomalous spin-dependent interactions

The $^9Be^+$ resonance has also been used in a search for previously unobserved spin-dependent forces of macroscopic range [28]. Forces of a monopole–dipole or a dipole–dipole nature might potentially be observed. Such forces are predicted to arise from scalar or pseudo-scalar couplings of weakly interacting bosons, such as axions.

In one experiment, the frequency ν_0 of the 303 MHz $^9Be^+$ transition was measured for directions of the magnetic field parallel and antiparallel to the local acceleration of gravity. A monopole–dipole force between the polarized ^9Be nuclei and the unpolarized particles of the Earth would lead to a difference of the frequency for the two different orientations of the magnetic field. The magnitude of the shift of the frequency for a vertical magnetic field was found to be less than 13.4 μHz (less than 5.5×10^{-20} eV).

In a second experiment, the frequency ν_0 was measured in a trap in which the magnetic field was generated with a superconducting solenoid and in a trap in which it was generated with an electromagnet having an iron yoke. The difference between the two cases is that the iron yoke contains polarized electrons. Hence, the $^9Be^+$ transition frequency might be shifted by a dipole–dipole interaction between the electrons and the ^9Be nucleus (aside from the ordinary magnetic interaction). The magnitude of the frequency shift was found to be less than 186 μHz (less than 7.7×10^{-19} eV). This places limits on possible nonmagnetic dipole–dipole interactions.

Other sensitive searches for spin-dependent forces have been made and are reviewed in ref. [28]. Recently, the relative nuclear magnetic resonance frequencies of ^{199}Hg and ^{201}Hg were measured for two different orientations of the magnetic field with respect to the Earth's gravitational field [29]. This result places somewhat more stringent limits on a monopole–dipole interaction than does the $^9Be^+$ experiment.

7. New experiments

Recently, a linear rf trap has been developed for observing hyperfine resonances in Hg^+ ions[30]. The main advantage of this kind of trap is that many ions can be trapped along the central axis of the trap, where the oscillating electric fields approach zero. All of the ions trapped along this line can be laser-cooled to very low velocities and hence can have low Doppler shifts. In most of the rf or Paul traps which have been used previously the fields approach zero only at one point, so only one ion can have very low Doppler shifts [1]. There is a similar problem in a Penning trap, since the crossed electric and magnetic fields cause the ions to rotate about the central axis.

In preliminary results with a linear rf trap, the 40.5 GHz ground-state hyperfine transition in $^{199}Hg^+$ has been observed with a linewidth of 0.25 Hz [30]. The ^{199}Hg nucleus has spin 1/2. The ^{201}Hg nucleus has spin 3/2, which should make it more suitable than ^{199}Hg for tests of local Lorentz invariance and the linearity of quantum mechanics. It may be possible to repeat many of the tests of fundamental physics which were previously performed with $^9Be^+$ with $^{199}Hg^+$ or $^{201}Hg^+$ ions with comparable accuracy. If any of the anomalous effects were to increase with the mass, charge, or quadrupole moment of the nucleus, they might be enhanced relative to $^9Be^+$.

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Presentation: W.M. Itano

B.I. Deutch: Why haven't you used a baked vacuum apparatus which results in pressures about 10^{-12} Torr?

W.M. Itano: We plan to reduce the pressure in future apparatuses by cooling them to liquid helium temperatures. This will have other beneficial effects, such as the possibility of using superconducting magnetic shielding.

Th. Kühl: As to the relativity test mentioned in your talk, is there any positive result reported, where a change of the signal is seen depending on the sideral times?

W.M. Itano: As far as I know, there are no positive results.

T.P. Das: Is it possible for you to measure accurately hyperfine constants in highly charged ions such as Bi^{80+}, that is, lithium-like bismuth ion? The reason for the interest in this system is that the radiative effects on hyperfine properties here are comparable to and perhaps stronger in magnitude than the many-body correlation effects. We have made estimates of this by relativistic many-body perturbation theoretic methods (Phys. Rev. A) two years back and it would be interesting to attempt to verify these predictions on radiative effects in many-electron systems.

W.M. Itano: No, with current technology, we cannot study such highly charged ions, because lasers suitable for state preparation and detection are not available.