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Laser-Fluorescence Mass Spectroscopy

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Measurements of ion cyclotron-resonance frequencies in a Penning trap, by a laser fluorescence technique, are described. This technique has been applied to indirect measurements of the proton-to-electron mass ratio and the $^9Be^+$ electron g_J factor. It is found that $m_p/m_e = 1836.152\,38(62)$ (0.34 ppm) and $g_J(^9Be^+) = 2.002\,262\,06(42)$ (0.21 ppm). Ultimately, ion cyclotron-resonance accuracies near 1 part in 10^{13} should be possible.

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The possibility of performing high-resolution mass spectroscopy in a Penning electromagnetic trap has been realized for some time.[1] Recently, experiments[2-4] have determined the proton-electron mass ratio m_p/m_e by alternately storing protons and electrons in the same Penning trap apparatus and comparing their cyclotron frequencies. In Ref. 2, cyclotron resonance is detected by measuring ion loss from the trap after resonant excitation. In Ref. 3, resonant excitation of cyclotron motion is detected by the increase of the ion's orbital magnetic moment; this appears as a change in the time-of-flight spectrum when ions are ejected from the trap into an axially symmetric inhomogeneous magnetic field. In Ref. 4, ion cyclotron resonance is detected by synchronously observing induced currents in the ring electrode, which is split into quadrants. The respective accuracies of m_p/m_e in these experiments are ± 2.9 ppm, ± 0.60 ppm, and ± 0.14 ppm. Here we demonstrate an alternative technique for precision mass spectroscopy in a Penning trap and apply it to an indirect measurement of m_p/m_e.

In our experiment, we measure the axial (ν_z), magnetron (ν_m), and electric-field-shifted cyclotron (ν_c') frequencies of a small cloud of atomic ions stored in a Penning trap by observing the changes in ion fluorescence scattering from a laser beam which is focused onto the ion cloud as shown schematically in Fig. 1. That is, when the ion motional frequencies are excited by an externally applied oscillating electric field, the ion orbits increase in size causing a decrease in laser fluorescence. To a good approximation, the electric field excites only the collective center-of-mass modes, whose frequencies are equal to those of a single isolated ion in the trap.[5] The three measured frequencies can then be combined to yield the free-space cyclotron frequency (ν_c) from the expression[6]

$$qB_0/2\pi m = \nu_c = [(\nu_c')^2 + \nu_z^2 + \nu_m^2]^{1/2} \text{(SI units)}, \quad (1)$$

where B_0 is the (uniform) applied magnetic field, q is the ion charge, and m is the ion mass. Therefore, if ν_c is measured for different ions, mass comparisons can be made.

As a demonstration of the technique, we have compared the cyclotron frequency to magnetic-field–dependent nuclear-spin-flip hyperfine

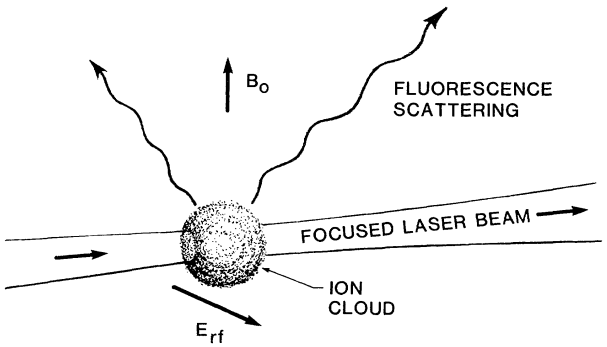

FIG. 1. Basic technique. A small sample of ions is confined by static magnetic and electric fields (not shown). A laser beam is focused onto the sample and the fluorescence scattering is observed. When the cyclotron motion is excited by an externally applied electric field ($\vec{E}_{rf}$) the ion orbits increase in size which results in a decrease in fluorescence.

$|\Delta M_I| = 1$ transition frequencies in the $^9Be^+$ $2s^2S_{1/2}$ ground state. Approximately twenty $^9Be^+$ ions (spherical cloud of diam $\cong 100$ μm) were stored in a Penning trap[4] with $1.64 z_0 = r_0 = 0.417$ cm. Typical conditions were $B_0 = 1.134$ T and applied trap voltage $V_0 = 1$ V, giving $\nu_c' = 1.922$ MHz, $\nu_z = 198$ kHz, and $\nu_m = 10.2$ kHz. The trap was made of gold mesh end caps and a molybdenum-mesh ring electrode. The center of the trap was at one focus of an ellipsoidal mirror; the second focus was outside the vacuum system. A lens was used to collimate the fluorescence light into a photomultiplier tube. The ions were laser cooled, compressed, and pumped into the $(M_I, M_J) = (-\frac{3}{2}, -\frac{1}{2})$ ground state by a laser tuned to the $2s\text{-}^2S_{1/2}(-\frac{3}{2}, -\frac{1}{2}) \rightarrow 2p^2P_{3/2}(-\frac{3}{2}, -\frac{3}{2})$ ($\lambda \cong 313$ nm) transition.[7] The size of the cloud was determined by using a second probe laser. The density was determined by measuring the space-charge-shifted $\vec{E}\times\vec{B}$ cloud-rotation frequency via the change in Doppler shift across the cloud by means of the probe laser.[8] We note that this cloud-rotation frequency is $> \nu_m$; only when space charge is negligible are these frequencies equal. From these measurements of cloud size and density, ion number was determined. The magnetic-field–dependent $(-\frac{3}{2}, -\frac{1}{2}) \rightarrow (-\frac{1}{2}, -\frac{1}{2})$ ground-state spin flip hyperfine transition frequency (ν_s) was measured by an rf-optical double-resonance technique described elsewhere.[7] In some cases, the $(\frac{3}{2}, \frac{1}{2}) \rightarrow (\frac{1}{2}, \frac{1}{2})$ transition was measured. The spin-flip frequency and cyclotron frequency were measured nearly simultaneously on the same ion cloud by stepping rf oscillators in frequency across ν_s and ν_c'. That is, the cyclotron resonance was probed at one frequency near ν_c' and then the spin resonance probed at a frequency near ν_s. Then the cyclotron and spin oscillators were incremented (by typically 0.25 and 2 Hz, respectively) and the cycle repeated. For each step of the spin and cyclotron resonances, the laser was shut off while the resonance was driven to avoid light shifts. Resonance curves taken in this fashion are shown in Fig. 2. This simultaneous sweeping reduced the effect of slow fluctuations in B_0 (ca. $\pm$ 1.5 ppm/min). Axial and magnetron resonances could be taken separately by using the same frequency-stepping technique; however, much better axial resonance curves were observed by leaving the laser on continuously. Potentially this causes a light shift as discussed below. From the measured values of ν_s and the values for the $^9Be^+$ hyperfine constant (A) and nuclear-to-electron g-factor ratio (g_I/g_J), the Breit-Rabi formula was used to determine

$$\nu_e(^9Be^+) \equiv g_J(^9Be^+) e B_0/(4\pi m_e), \quad (2)$$

where $g_J(^9Be^+)$ is the electron g factor in $^9Be^+$ and e is the electron charge. Since $\nu_c(^9Be^+)$ and $\nu_e(^9Be^+)$ are measured under essentially the same conditions we have

$$R \equiv 2\nu_e(^9Be^+)/\nu_c(^9Be^+)$$
$$= g_J(^9Be^+) m(^9Be^+)/m_e, \quad (3)$$

where $m(^9Be^+)$ is the $^9Be^+$ mass. To determine A and g_I/g_J, we have measured the $(-\frac{3}{2}, +\frac{1}{2}) \rightarrow (-\frac{1}{2}, +\frac{1}{2})$ and $(\frac{3}{2}, -\frac{1}{2}) \rightarrow (\frac{1}{2}, -\frac{1}{2})$ ground-state transition frequencies at field-independent points.[7] We obtain the preliminary values $A = -625\,008\text{-}837.048(10)$ Hz and $g_I/g_J = 2.134\,779\,853(2) \times 10^{-4}$. The only significant deviation from the Breit-Rabi formula is a shift in the effective value of A, proportional to B_0^2 and approximately equal to -0.017 Hz at $B_0 = 1$ T.[9] This has a negligible effect on the present experiment.

In our determination of R we have considered the following systematic effects:

(1) Deviations of the electric potential from a pure quadratic ($r^2 - 2z^2$). The only clearly observable systematic effect was an axial anharmonicity of approximately $(\partial\nu_z/\partial E_z)\nu_z^{-1} \cong -0.1/\text{eV}$ ($V_0 = 1$ V). This is about 10 times larger than that observed with a trap of solid uncompensated electrodes.[10] However, because of the small excitation required to observe the axial resonance, typical linewidths were < 4 Hz. We have also ob-

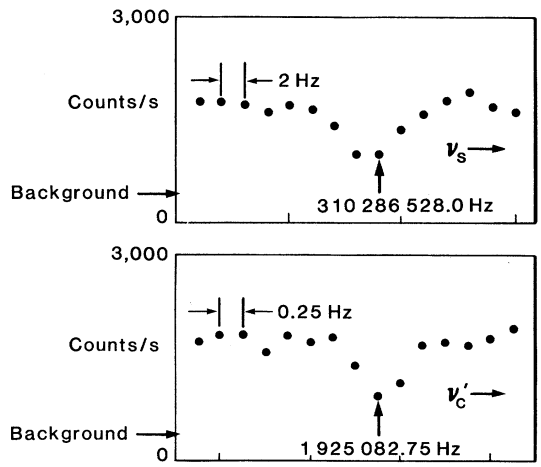

FIG. 2. Example cyclotron (ν_c') and spin (ν_s) resonances. These curves were taken nearly simultaneously by the technique described in the text. $B_0 \cong 1.134$ T, $\nu_z \cong 215$ kHz.

served a slight (< 0.1 ppm) negative cyclotron anharmonicity which agrees in sign and approximate magnitude with the axial anharmonicity. For fixed V_0, an error in ν_c' or ν_z due to a fourth-order anharmonic term in the electric potential causes an error in R proportional to B_0^{-2}. An error in our measurement of ν_c' which is independent of B_0 causes an error in $R \propto B_0^{-1}$.

(2) Induced-charge frequency shifts.[5] From the measured numbers of ions this shift should be < 0.1 ppm and causes an error in $R \propto B_0^{-2}$ for constant ion number.

(3) Optical-well shift. Since a focused laser beam creates an optical potential well,[11] we might expect a shift in ν_z. For the conditions realized here, we estimate $\Delta\nu_z/\nu_z \cong 10^{-7}$ and measure $\Delta\nu_z/\nu_z < 10^{-7}$. For fixed ν_z this shift causes an error in $R \propto B_0^{-2}$.

(4) Magnetic bottle shift.[4] The trap could contribute to the magnetic field inhomogeneity through its susceptibility. Because the amount of material constituting the electrodes is small, we estimate this shift to be small. It causes an error in $R \propto B_0^{-1}$.

(5) Light shift on spin transitions. Although the laser is off while the ν_s power is applied we typically repeated the ν_s cycle two or four times on each point to increase the signal-to-noise ratio. This might cause a spin coherence between spin pulses which could then be light shifted. Measurements indicated that such a shift was $< 0.5 \times 10^{-7}$. This shift is independent of B_0.

(6) Space charge. Other kinds of ions were driven from the trap with strong cyclotron excitation; therefore no space-charge shifts should occur.[5]

Values of R vs B_0^{-2} and B_0^{-1} were extrapolated to the limit $B_0 \to \infty$. Axial frequencies of 292 and 215 kHz at magnetic fields of 0.673 T $[(\frac{3}{2},\frac{1}{2}) \to (\frac{1}{2},\frac{1}{2})$ transition], 0.764 T, and 1.134 T were used. The total spread of unextrapolated values was 0.4 ppm and the total spread of extrapolated values was approximately 0.15 ppm. From these extrapolations and our estimates of systematic effects we find

$$g_J(^9\mathrm{Be}^+)m(^9\mathrm{Be}^+)/m_e = 32891.5710(49)\ (0.15\ \mathrm{ppm}). \quad (4)$$

The numbers in parentheses represent estimates of one standard deviation. We note that we have checked this result to 2 ppm accuracy by comparing $\nu_e(^9\mathrm{Be}^+)$ to the cyclotron frequency of electrons which are alternately stored in the trap.

This result, with a theoretical value[12,13] of $g_J(^9\mathrm{Be}^+)$ and the value[14] of $m(^9\mathrm{Be}^+)/m_p$, can be used to give an indirect determination of m_p/m_e. Using Veseth's value[12] for $g_J(^9\mathrm{Be}^+)$, we obtain

$$m_p/m_e = 1836.152\,38(62)\ (0.34\ \mathrm{ppm}). \quad (5)$$

This value agrees with but is (0.34 ± 0.37) ppm lower than the most precise direct determination.[4] If we assume the value of m_p/m_e from Ref. 4, we obtain

$$g_J(^9\mathrm{Be}^+) = 2.002\,262\,06(42)\ (0.21\ \mathrm{ppm}). \quad (6)$$

The potential accuracy for direct mass comparisons with use of this method is extremely high. Assume that cyclotron resonance is performed on a single Be^+ ion in a trap with $z_0 = r_0/\sqrt{2} = 1$ cm, $B_0 = 6$ T, $\nu_z \cong 150$ kHz. If maximum laser cooling[15] is achieved, then $z_{\max} \cong 1\ \mu\mathrm{m} \gg r_c(\max), r_m(\max)$; this implies that the effects of anharmonicities are greatly suppressed. The third- and fourth-order axial corrections to the electric potential can be nulled by proper biasing of the end cap and guard electrodes on a compensated trap.[4] The largest uncompensated terms in the electric potential appear to be due to residual third- and fourth-order terms which violate axial symmetry. Using perturbation theory,[16] we estimate these terms to be at the level of a few parts in 10^{13} (and proportional to B_0^{-2}). The induced-charge frequency shift would be at the level of a few parts in 10^{13} (proportional to B_0^{-2}). Magnetic bottle effects could be nulled[17] and relativistic shifts would be negligible.

Eventually, precision mass comparisons could be made between isotopes of the same species or, more interestingly, between different nuclear isomers. In the case of nuclear isomers, if the energy difference could also be determined in terms of γ-ray wavelengths, a conversion factor from wavelength to atomic mass unit would be obtained.[18] Such techniques could also be employed to study radiation pressure forces.[11]

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Uncertainty in Quantum Measurements

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The object of this Letter is to show that except in the case of canonically conjugate observables, the generalized Heisenberg inequality does not properly express the quantum uncertainty principle. It is, in general, too weak. An inequality is obtained which does express the principle.

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In quantum theory, any single observable or commuting set of observables can in principle be measured with arbitrary accuracy.[1-3] But there is in general an irreducible lower bound on the uncertainty in the result of a simultaneous measurement of noncommuting observables. Equivalently, there is an upper bound on the accuracy with which the values of noncommuting observables can be simultaneously prepared. These are qualitative statements of the uncertainty principle in quantum theory. My purpose here is to obtain a quantitative expression of the principle. We shall see that the customary generalization

$$V_{\hat{A}}(|\psi\rangle)V_{\hat{B}}(|\psi\rangle) \geq \tfrac{1}{4}|\langle\psi|[\hat{A},\hat{B}]|\psi\rangle|^2 \qquad (1)$$

of Heisenberg's inequality[4]

$$V_{\hat{x}}(|\psi\rangle)V_{\hat{p}}(|\psi\rangle) \geq \tfrac{1}{4} \quad ([\hat{x},\hat{p}]=i), \qquad (2)$$

though it is of course true, will not fit the bill. The quantity

$$V_{\hat{A}}(|\psi\rangle) \equiv \langle\psi|\hat{A}^2|\psi\rangle - \langle\psi|\hat{A}|\psi\rangle^2 \qquad (3)$$

is the variance of $\hat{A}$ in the state $|\psi\rangle$ and the units are chosen so that $\hbar = 1$.

In order to express the principle

$$\left[\begin{array}{c}\text{Uncertainty in the result of a}\\ \text{measurement of } \hat{A} \text{ and } \hat{B}\end{array}\right] \geq \left[\begin{array}{c}\text{An irreducible lower}\\ \text{bound}\end{array}\right] \qquad (4)$$

quantitatively, I shall seek a theorem of linear algebra in the form

$$\mathfrak{U}(\hat{A},\hat{B},|\psi\rangle) \geq \mathfrak{B}(\hat{A},\hat{B}). \qquad (5)$$

Here $\hat{A}$ and $\hat{B}$ are the observables which are simultaneously measured or prepared and $|\psi\rangle$ is the relative state representing the outcome of the measurement or preparation.

It is logically possible that the bound $\mathfrak{B}$ could also depend on the initial state of the system, but this could not be the case in quantum theory where there always exists a dynamical evolution which transforms any initial state into any other.

