Collective oscillations of stored ions

K. Jungmann, J. Hoffnagle, R. G. DeVoe, and R. G. Brewer
IBM Research, Almaden Research Center, 650 Harry Road, San Jose, California 95120-6099
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When ions of slightly different mass and the same charge are stored in a radio-frequency electric quadrupole trap, a collective oscillation is observed rather than the independent motions usually assumed. A high-temperature model of ion clouds, each characterized by ions of one mass, shows that the ions of each cloud move as a unit and exhibit a correlated motion with the other clouds when a strong coupling condition is satisfied. This phenomenon is expected to arise whenever more than one ion is trapped and might be observable in neutral atom traps as well.

The current interest in trapping elementary particles or ions in Penning or electric quadrupole traps includes such studies as precision g-2 value measurements for a single electron or positron,[1] an improved determination of the electron to proton mass ratio,[2] the development of microwave and optical clocks,[3,4] sympathetic cooling of a two-species plasma,[5] and detecting optical quantum jumps in a single ion.[6] In this paper, we report an observation which appears to have gone unnoticed, namely, that ions stored in a quadrupole trap do not necessarily oscillate independently of one another, as previous analysis assumes, but instead collective oscillations can occur as if the ion clouds are giant particles with a large charge. These collective modes arise, as one might expect, from the ion-ion Coulomb potential, but rather remarkably this potential can be far weaker than the trapping potential. From the elementary theory of coupled oscillators given here, we conclude that the condition for collective motion depends simply on these two potentials and the relative mass of the ions. Even with a single-species ion cloud, collective motion is a certainty.[7] This phenomenon is unique to ion traps although there is an obvious resemblance to the plasma oscillations encountered in neutral plasmas.[8]

Consider the observations first. Our Paul quadrupole trap[9] consists of two end caps and a ring having hyperbolic surfaces where the dc potential is zero and the applied ac potential in the axial and radial positions, z and r, obeys $V=V_{\mathrm{ac}}[\cos(\Omega t)](z^2-r^2/2)/z_0^2$. The ions move in a time-averaged pseudopotential which in lowest order corresponds to a harmonic oscillator. The micromotion frequency $\Omega/2\pi=5.130$ MHz and the characteristic trap dimension z_0 satisfies $z_0^2=(\bar{z}_0^2+\bar{r}_0^2/2)$ where $\bar{r}_0/\bar{z}_0=\sqrt{2}$, $\bar{r}_0=0.250$ cm being the inner-ring radius and $\bar{z}_0$ one-half the end cap spacing. Ions such as Ho^+ and Er^+ were loaded into the trap, first by evaporating the neutral atom from a tungsten filament containing the metal, then by ionization with an electron gun, and finally by cooling the ion cloud with the addition of He gas at room temperature and at a pressure of 1×10^{-6} Torr. When two metal ion species were examined simultaneously, two independent ovens were used. The axial motion of the ions was detected[10] by sweeping the amplitude of the ac trapping field until the axial secular frequency ω_z matched the resonance of an external LC circuit tuned to 0.5939 MHz. A weak axial probe field also of frequency ω_z allowed synchronous detection. A Floquet solution of the appropriate Mathieu equation for this trap predicts a resonance at a voltage to mass ratio $V_{\mathrm{rms}}/m=3.814$ V/amu.

The mass spectra of Ho^+ and Er^+ in Fig. 1 provide one example of unexpected behavior which has forced us to conclude that collective motion is the proper interpretation of this phenomenon. As expected, $^{165}Ho^+$ yields only one mass peak since there is only one naturally occurring isotope, and it gives no hint of a collective oscillation. However, Er^+ shows only one peak also, and since the resolution is about 0.05 amu, we would have expected to see at least four isotopes in the range 166 to 170 and of comparable amplitude. As we shall see, our collective motion theory predicts one mass peak, the mass being the average of all species present. The third trace shows Ho^+ and Er^+ loaded simultaneously, the relative amounts being somewhat arbitrary and unknown. Again, only one mass peak appears, which always falls between the two peaks of pure Ho^+ and pure Er^+, and suggests an average mass value between the two extremes. Similarly, in the case of pure Sm^+ and pure Xe^+, which consist of several naturally occurring isotopes, only one mass peak was detected (see top trace of Fig. 2). A summary of these data appears in Table I where the measured mass is given relative to the single isotope ^{165}Ho in order to minimize systematic errors, particularly in the measurement of V_{ac}. We see that the accepted atomic masses of Xe, Er, and Sm are in excellent agreement with our measured values, supporting the premise that it is the average mass that is measured.

Masses are resolved, however, when the mass difference is sufficiently large. For example, when Xe^+ and Ho^+ are loaded together (Fig. 2), two peaks appear corresponding to the pure species but shifted slightly to higher mass.

How are these results to be explained? The simplest model, which we consider first, is that of two singly charged ions of mass m_1 and m_2 that are confined in a quadrupole trap with force constants k_1 and k_2, the particles repelling one another with a force constant a. The reduced equations of motion are

$$m_1\frac{d^2x}{dt^2}=-k_1x+a(x-y)\ ,$$
$$m_2\frac{d^2y}{dt^2}=-k_2y-a(x-y)\ . \tag{1}$$

Here $k_i=m_iq_i^2\Omega^2/8$ with $i=1,2$ and $q_i=4(e/m_i)V_{\rm ac}/(z_0\Omega)^2$, which implies a time average over the micromotion, and we have ignored higher-order contributions that arise in the Mathieu equation.[9,10] The Coulomb force constant is defined by $a\equiv 2e^2/r_0^3<0$ which is the result of an expansion of e^2/r^2 about an average particle separation $r_0\equiv(x_0-y_0)<0$, determined by initial conditions. The determinantal solution of (1) yields two secular frequencies

$$\omega_{1,2}^2=\frac{1}{2m_1m_2}(m_1(k_2+a)+m_2(k_1+a)$$
$$\pm\{[m_1(k_2+a)-m_2(k_1+a)]^2+4m_1m_2a^2\}^{1/2})\ . \tag{2}$$

Rearranging terms in the square root and with the reasonable assumption that $a\ll k_{1,2}$, we define a strong coupling condition

$$\left|\frac{a}{k_1}\right|\gg\left|\frac{m_1-m_2}{m_2}\right|\ , \tag{3}$$

where the modes of oscillation are

$$\omega_1^2=\frac{1}{2}\left[\frac{k_1}{m_1}+\frac{k_2}{m_2}\right]\ , \tag{4}$$
$$\omega_2^2=\frac{1}{2}\left[\frac{k_1+2a}{m_1}+\frac{k_2+2a}{m_2}\right]\ . \tag{5}$$

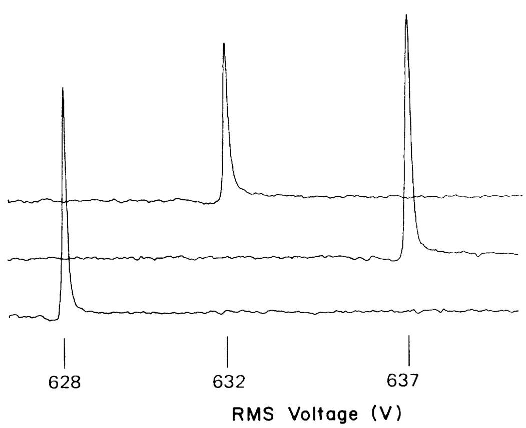

FIG. 1. The strong-coupling condition is satisfied in detecting axial resonances of an electric quadrupole trap for pure Ho^+ (lowest trace), pure Er^+ (middle trace), and a mixture of Ho^+ and Er^+ (uppermost trace). The single mass peak in the top two traces is a signature of strong coupling and corresponds to the average mass.

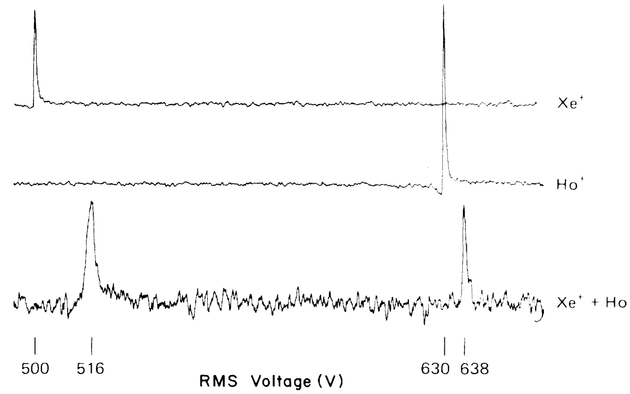

FIG. 2. The weak-coupling condition is satisfied for the mixture $Xe^+ + Ho^+$ (lowest trace) which shows two resolved peaks corresponding to the Xe^+ and Ho^+ ion clouds moving independently of one another. The mass peaks of the two are shifted relative to the pure elements (top two traces).

From the phase relations of the solutions of (1), we see that the ω_1 mode is a translational motion of the two masses with a fixed spacing r_0, the ion-ion Coulomb repulsion being unchanged as indicated by the absence of an a term in Eq. (4). It is this translational mode that results in an axial current that is detectable. For the ω_2 mode, the ion-ion distance is stretched and compressed about an unchanging center-of-mass position, and the current and hence the signal vanish. While this mode cannot be detected electronically, we predict that optical detection is possible. We rewrite Eq. (4) as

$$\omega_1=\left[(\sqrt{2}e/z_0^2\Omega)V_{\rm ac}\left[\left\langle\frac{1}{m^2}\right\rangle\right]\right]^{1/2} \tag{6}$$

to emphasize that the average quantity $\langle 1/m^2\rangle\equiv\frac{1}{2}(1/m_1^2+1/m_2^2)$ is of importance, and for small mass differences $(\langle 1/m^2\rangle)^{1/2}\cong\langle 1/m\rangle\cong 1/\langle m\rangle$. Thus, for these collective modes, it is predicted that only one mass peak will be observed corresponding to the average mass.

For the case of weak coupling, the inequality sign of Eq. (3) is reversed and we obtain from (2)

$$\omega_{1,2}^2=(k_{1,2}+a)/m_{1,2}\ , \tag{7}$$

i.e., each mode corresponds to a single mass, the reso-

TABLE I. Atomic and measured masses in amu. Naturally occurring Xe has nine isotopes in the range 124 to 136, Ho has one isotope, Er has six isotopes in the range 162 to 170, and Sm has seven isotopes in the range 144 to 154. Masses are measured relative to Ho.

Element	Atomic mass	Measured mass
Xe	131.30	130.6
Ho	164.93	164.93
Er	167.26	167.3
Sm	150.35	149.6

nance being shifted by the a term. Furthermore, in the limit $m_1=m_2$, the strong coupling condition (3) is always satisfied, *no matter how weak the Coulomb coupling is,* making the weak-coupling condition inapplicable.

As a second model, we consider the many-particle case of one or more clouds of ions, each composed of ions of one mass. In a high-temperature regime, which is appropriate to the experiments described here, we find that each cloud moves as a unit retaining its spheroidal shape. Thus collective oscillations can exist on two levels, first in the motion of a single cloud of ions and second in the correlated motion of several clouds.

The potential energy of a single cloud of N ions of one mass and unit charge e is

$$U(\mathbf{R}_i)=e\int n(\mathbf{r},\mathbf{R}_i)\Psi(\mathbf{r})d^3r + e^2\int\int \frac{n(\mathbf{r},\mathbf{R}_i)n(\mathbf{r}',\mathbf{R}_i)}{2|\mathbf{r}-\mathbf{r}'|}d^3r\,d^3r' \ . \tag{8}$$

The first term arises from the trap pseudopotential $\Psi=(\frac{1}{2}e)kr^2$ and the second from the self-energy or Coulomb interaction, R_i being the distance from the center of mass (c.m.) of the ith cloud to the trap minimum and r the corresponding distance of an ion. The high-temperature limit[11] implies a Gaussian charge density $n(r,R_i)=N(\alpha/\pi)^{3/2}e^{-\alpha(\mathbf{r}-\mathbf{R}_i)^2}$ with $\alpha\equiv m\omega^2/(2k_BT)$. Integration of the first term of Eq. (8) yields

$$U(R_i)/N=\tfrac{1}{2}kR_i^2+\frac{3}{4}\frac{k}{\alpha} \ , \tag{9}$$

while the self-energy integral is a constant. These constant terms do not affect the dynamics of the problem. Thus the average potential energy per ion, $\frac{1}{2}kR_i^2$, is exactly the same as a single ion located at the position R_i, and the ion cloud behaves as a single giant particle. In this way, the ions of an individual cloud are strongly coupled and oscillate collectively in the trap as in the two-particle problem when $m_1=m_2$.

If we introduce a second cloud of another species, the potential of the first cloud is $U_1(\mathbf{R}_1,\mathbf{R}_2)=U_1(\mathbf{R}_1)+U_{12}(\mathbf{R}_1-\mathbf{R}_2)$ where the trap potential U_1 is given by Eq. (9) and the intercloud repulsion is

$$U_{12}(\mathbf{R}_1-\mathbf{R}_2)=e^2\int\int\frac{n_1(\mathbf{r}_1,\mathbf{R}_1)n_2(\mathbf{r}_2,\mathbf{R}_2)}{|\mathbf{r}_1-\mathbf{r}_2|}d^3r_1d^3r_2 \ . \tag{10}$$

For small displacements $R\equiv R_1-R_2$, numerical and analytic solutions of Eq. (10) as well as symmetry arguments reveal that

$$U_{12}(R)=U_{12}(0)-\tfrac{1}{2}AR^2\ldots \tag{11}$$

where A is a positive constant. The potentials (9) and (11) result in the same equations of motion as for the two-particle problem, Eq. (1), but with one difference. The sign of A in (11) is positive whereas for two particles the coupling constant a is negative. Indeed, Fig. 2 shows that the mass peaks are shifted to higher values in agreement with $A>0$ while for two particles we predict a shift to smaller masses. The physical reason for the difference is that the two clouds can overlap with a repulsive force that vanishes at $R=0$ but initially increases when $|R|>0$ and then decreases as $1/R$ for large R. However, two charged particles can never overlap and their repulsive force always decreases with increasing R.

We note that a determination of the minimum mass difference that can be resolved allows an independent determination of the ion density. For this purpose, we define the transition between the strong- and weak-coupling regimes by $|a/k_1|=|(m_1-m_2)/m_2|_{\min}$ where $|m_1-m_2|_{\min}$ is the minimum resolvable mass difference. Assuming a Gaussian distribution, we derive in (11) the Coulomb force constant per ion of two clouds as $a\equiv A/N=\frac{1}{3}\sqrt{2/\pi}N\alpha^{3/2}e^2$, N being the number of ions in each cloud. Experiments on a mixture of $^{165}\mathrm{Ho}^+$ and $^{150}\mathrm{Sm}^+$ suggest that a mass difference of 15 divides the two regimes. This result implies that $|a/k_1|=0.01$, and since $k_1=3.7\times10^{-9}$ dyne/cm for Ho^+, we conclude that the density at the peak of the distribution $n_p=N(\alpha/\pi)^{3/2}\cong1.1\times10^8$ cm^{-3}, which is significantly less than the maximum value[10,11] or Brillouin density $n_{\max}=(3/4\pi)m\omega_z^2/e^2=3.8\times10^9$ cm^{-3}. From the above expression for a and using n_p and a measured $N=10^5$ ions, we estimate a plasma temperature $T\cong4\times10^4$ K.

Still a third approach that might be appropriate for low temperatures is to model the plasma as a three-dimensional lattice where masses m_1 and m_2 lay in alternating planes that vibrate as a unit. The motion then reduces to a one-dimensional problem. With the assumption that only nearest neighbors interact, the resulting modes are found to be virtually identical to the two-particle results when $Kl\ll1$ (strong coupling) or $Kl=\pi$ (weak coupling) where $\hat{K}$ is the propagation vector of the traveling wave and l the lattice spacing. However, at low temperatures the ion density distribution is expected to be flat prior to an abrupt falloff,[11] and the assumption that the shape of the charge clouds is unaffected by a neighboring cloud will not be obeyed as demonstrated recently for Hg^+ and Be^+.[5]

It would seen, therefore, that the two-particle and the high-temperature ion cloud models are a reasonable first approximation to understanding collective oscillations of stored ions. The theory could be generalized further to allow for more than two mass numbers. However, as the number of different masses increases, so will the number of modes, but with axial detection, only the ω_1 translational mode will be observed. We hope to detect the remaining modes optically. The predictions for two trapped ions should be tested also, and with optical cooling, the present observations are expected to change.[12] Moreover, the influence of collective motions on precision measurements should be considered. Finally, collective oscillations are predicted to occur in neutral atom traps where the Coulomb coupling is replaced by a van der Waals interaction.

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[1]P. B. Schwinberg, R. S. Van Dyck, Jr., and H. G. Dehmelt, Phys. Rev. Lett. **47**, 1679 (1981).

[2]R. S. Van Dyck, F. L. Moore, D. L. Farnham, and P. B. Schwinberg, Int. J. Mass Spectrom. Ion Phys. **66**, 327 (1985).

[3]J. J. Bollinger, J. D. Prestage, W. M. Itano, and D. J. Wineland, Phys. Rev. Lett. **54**, 1000 (1985).

[4]J. C. Bergquist, D. J. Wineland, W. M. Itano, H. Hemmati, H.-U. Daniel, and G. Leuchs, Phys. Rev. Lett. **55**, 1567 (1986).

[5]D. J. Larson, J. C. Bergquist, J. J. Bollinger, W. M. Itano, and D. J. Wineland, Phys. Rev. Lett. **57**, 70 (1986).

[6]W. Nagourney, J. Sandberg, and H. Dehmelt, Phys. Rev. Lett. **56**, 2727 (1986); J. C. Bergquist, R. G. Hulet, W. M. Itano, and D. J. Wineland, *ibid.* **57**, 1699 (1986); Th. Sauter, W. Neuhauser, R. Blatt and P. E. Toschek, *ibid.* **57**, 1696 (1986); see also A. Schenzle and R. G. Brewer, Phys. Rev. A **34**, 3127 (1986).

[7]D. J. Wineland and H. G. Dehmelt [J. Appl. Phys. **46**, 919 (1974)] discuss some aspects of collective motion of an electron cloud.

[8]J. P. Schermann and F. G. Major [Appl. Phys. **16**, 225 (1978)] consider the case of a trapped neutral plasma, but collective motions were not examined.

[9]R. F. Wuerker, H. Shelton, and R. V. Langmuir, J. Appl. Phys. **30**, 342 (1959).

[10]H. G. Dehmelt, Adv. At. Mol. Phys. **3**, 53 (1967); *ibid.* **5**, 109 (1969).

[11]L. S. Cutler, R. P. Giffard, and M. D. McGuire, Appl. Phys. B **36**, 137 (1985); L. S. Cutler, C. A. Flory, R. P. Giffard and M. D. McGuire, *ibid.* **39**, 251 (1986).

[12]A phase transition is predicted to occur when the coupling parameter $\Gamma \equiv e^2(4\pi n/3)^{1/3}/k_B T = 168$; see, for example, L. R. Brewer, J. D. Prestage, J. J. Bollinger, and D. J. Wineland, in *Strongly Coupled Plasma Physics,* edited by F. J. Rogers and H. E. Dewitt (Plenum, New York, 1987). In our experiments, $\Gamma \ll 1$.