PHYSICAL REVIEW A

Fractional frequency collective parametric resonances of an ion cloud in a Paul trap

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(Received 23 January 1998)

We report the observation of the first ten collective parametric resonances of an uncooled N_2^+ ion cloud in a Paul trap driven at simple fractions of twice the secular frequency of the trap by an additionally applied quadrupole field. The fractional resonances are observable only if the excitation field surpasses a critical strength. Odd-even staggering of the thresholds is observed. [S1050-2947(98)51307-8]

PACS number(s): 32.80.Pj, 07.75.+h

The Paul trap [1] has a dual nature. On the one hand, it serves as an auxiliary device in high-precision spectroscopy and applied physics; on the other hand, it is an excellent system for the investigation of dynamical effects. The central point of this paper is to demonstrate theoretically and experimentally the existence of high-order collective fractional parametric resonances in an externally driven cloud of ions stored in a Paul trap.

The equations of motion of a single particle in an ideal Paul trap are derived in many publications (see, e.g., [1]). In essence, the particle's trajectory is the solution of three uncoupled Mathieu equations [2] in the three spatial directions. The same holds for the center-of-mass of a cloud of charged particles in an ideal Paul trap, since the two-body Coulomb interaction cancels in the derivation of the equations of motion for the center of mass [3]. In this paper we consider oscillations in the *z* direction only and obtain for the center-of-mass coordinate *Z* of a cloud of charged particles the following equation of motion [1,3]:

$$\ddot{Z} + [\hat{a} - 2\hat{q}\cos(2\hat{\tau})]Z = 0.$$
 (1)

The equation of motion (1) is written in scaled time $\hat{\tau} = \Omega t/2$, where Ω is the trap frequency. The two parameters \hat{a} and \hat{q} in Eq. (1) are the standard Paul trap control parameters given by

$$\hat{a} = -\frac{8QeU_0}{m\Omega^2 r_0^2}, \quad \hat{q} = \frac{4QeV_0}{m\Omega^2 r_0^2},$$
 (2)

where Qe and m are the charge and mass of the particles in the cloud, U_0 and V_0 are the dc voltage and the zero-to-peak ac voltage applied to the trap, and r_0 is the equatorial radius of the ring electrode of the trap. In a pseudopotential approximation [4] Eq. (1) becomes

$$\frac{d^2Z}{dt^2} + \omega_z^2 Z = 0, \qquad (3)$$

where we have reverted back to natural time t and ω_z is the secular frequency of the cloud in the z direction. We now couple an external parametric drive to Eq. (3), which in our Paul trap [3] is realized by adding an additional driving voltage V_{exc} (peak-to-peak) to the end-cap electrodes of the trap. Adding also a damping term we obtain

$$\frac{d^2 Z}{dt^2} + \Gamma \frac{d Z}{dt} + \omega_z^2 Z = F \cos(\omega t) Z, \qquad (4)$$

where Γ is the damping constant, $F = eQV_{\text{exc}}/(mr_0^2)$ is the strength of the additional drive, and ω is its frequency. Introducing $\tau = \omega t/2$ turns Eq. (4) into the damped Mathieu equation

$$\ddot{Z} + \lambda \dot{Z} + [a - 2q\cos(2\tau)]Z = 0, \tag{5}$$

where $\lambda = 2\Gamma/\omega$, $a = (2\omega_z/\omega)^2$, and $q = 2F/\omega^2$. Depending on the values of the control parameters λ , a, and q, Eq. (5) exhibits stable and unstable behavior. For $\lambda = 0$ the stability diagram of Eq. (5) is shown in Fig. 1. The white regions in Fig. 1 correspond to the stable solutions of Eq. (5); in the shaded regions of Fig. 1 the solutions of Eq. (5) are exponentially unstable. As indicated in Fig. 1 the instability regions have the shape of narrow tongues touching the *a* axis at $a = n^2$, n = 1, 2, 3, ... [2]. Therefore, for small values of λ , we expect instabilities in Eq. (5) if ω is in the vicinity of



FIG. 1. Stability diagram of the Mathieu equation. The shaded regions correspond to exponentially unstable solutions. The white regions correspond to stable solutions.

R34

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$$\omega_n = 2 \omega_z / n, \quad n = 1, 2, 3, \dots$$
 (6)

These instabilities are of the parametric type, and are well known in the standard literature [2,5]. The properties of the n=1 resonance were recently studied in detail [3]. An important result is the existence of a critical voltage below which the collective resonance is not observed. In order to obtain a theoretical prediction for the critical voltages of the higher-order parametric resonances we scale Eq. (4), defining $\tilde{\tau} = \omega_z t$. We obtain the equation of motion

$$\frac{d^2 Z}{d\tilde{\tau}^2} + \gamma \frac{dZ}{d\tilde{\tau}} + Z = f\cos(\nu \tilde{\tau})Z,$$
(7)

where $\gamma = \Gamma/\omega_z$, $f = F/\omega_z^2$, and $\nu = \omega/\omega_z$. The parametric resonances of Eq. (7) are expected in the vicinity of $\nu_n = 2/n, n = 1, 2, 3, \dots$ We investigated Eq. (7) numerically using a fourth-order Runge-Kutta method. For γ values ranging from 10^{-2} to 10^{-4} we determined numerically the critical $f_c^{(n)}$ necessary for the onset of parametric instability of resonance number *n*. Our numerical results are conveniently summarized by the formula

$$f_c^{(n)}(\gamma) = a_n(\gamma) \gamma^{1/n}, \qquad (8)$$

where for $\gamma \ll 1$ the coefficient $a_n(\gamma)$ is only weakly dependent on *n* and γ . For example, to within an error of 5% $a_n(\gamma = 10^{-3})$ is given by 2.0, 2.03, 1.83, 1.73, 1.73 for $n = 1, \ldots, 5$, respectively. This result is consistent with a formula stated without derivation in Ref. [5]. Thus, especially for rough estimates, $a_n(\gamma)$ may be replaced by $a_n(\gamma) \approx 2$. Relating Eq. (8) to the physical parameters of our experiment we obtain a direct prediction for the critical voltage (peakto-peak) necessary to excite the parametric resonance number *n*:

$$V_{c}^{(n)} = \alpha_{n} \gamma^{1/n} = a_{n} \frac{\omega_{z}^{2} m r_{0}^{2}}{e Q} (\Gamma/\omega_{z})^{1/n}.$$
 (9)

Note that the associated critical q of Eq. (5) given by $q_c^{(n)} = 2F_c^{(n)}/\omega_n^2 \approx n^2 \gamma^{1/n}$ (for $a_n \approx 2$) is not necessarily small and in fact diverges for $n \to \infty$ for any value of γ . This is to be expected, since for large n the unstable tongues in Fig. 1 remain exponentially thin for $q \ge 1$ (see, e.g., Fig. 20.1 of Ref. [2]). Even for such large values of q our theory remains valid since it is based on Eq. (8), which was established nonperturbatively (numerically).

We turn now to an experimental verification of the existence of high-order fractional parametric resonances in the Paul trap. We used the experimental setup described in [3] to create a cloud of N₂⁺ ions stored in a Paul trap. It has the standard geometry ($r_0^2 = 2z_0^2$) and hyperbolic electrodes, with a minimum radius of the ring electrode $r_0 = 2$ cm. The ions are confined by an rf field of 750 kHz with an ac amplitude $V_0 = 190$ V (zero-to-peak) and a dc voltage $U_0 = +10$ V applied at the ring electrode. The additional excitation field is applied at the end-cap electrodes during the interaction time. About 4000 ions are stored in the trap for these experiments. After a "creation time" [3] of about 1 s, in which the N₂⁺ ions were created by electron bombardment from the rest gas in the trap (rest gas pressure ~10⁻⁹ mbar) we exposed the



FIG. 2. Experimental signatures of the first ten collective fractional parametric resonances for N_2^+ .

cloud to the excitation field generated by applying an additional ac voltage to the trap. The effectiveness of the coupling of the field to the ion cloud depends on the excitation voltage and the excitation frequency. After an "interaction time" [3] of about 1 s, the ions remaining in the trap are counted by extracting them electrically from the trap by a voltage pulse through a hole in one of the end caps of the trap. The ion loss due to interaction is a measure of the effect of the excitation voltage.

Figure 2 shows the number of surviving ions as a function of the excitation frequency. The macromotion spectrum is recorded at an excitation field amplitude $V_{\text{exc}} = 3.0 \text{ V}$ (peakto-peak) and a frequency step of 50 Hz between the data points. Every data point in Fig. 2 corresponds to a complete creation-interaction-extraction cycle. Thus there are no memory effects in the data of Fig. 2. The working point in Fig. 2 is chosen such that the secular trap potential is shallow (9 eV) in the z direction and steep (41 eV) in the x, y directions. Hence it is easier to excite higher-order resonances in the z direction. This way, as shown in Fig. 2, we were able to observe a sequence of fractional parametric resonances for n=1 to n=10. It is to be noted that the observed resonances are not produced due to direct excitation of the $2\omega_{z}$ resonance by the higher harmonics of the function generator used for excitation (SRS model DS345). This was experimentally verified by using a spectrum analyzer. The amplitude of the strongest harmonic is less than -55 dB and is much smaller than the critical amplitude required to excite the $2\omega_z$ resonance directly.

In Fig. 3 we present the n=3 resonance in more detail. The inset in Fig. 3 shows the shape of the n=3 resonance. The shapes of all the parametric resonances are qualitatively the same as the one of the n=1 resonance [3]. The inset shows a relatively weak and broad resonance at $\sim 2\omega_z/3$ due to excitation of individual ions, and a strong and sharp resonance corresponding to collective oscillations of the ion cloud. The full width at the base of the collective resonance is plotted as a function of the excitation amplitude V_{exc} (peak-to-peak) in Fig. 3. It shows a threshold behavior reminiscent of the n=1 resonance [3].

Next we turn to an experimental check of the analytical prediction (9). According to Eq. (9) a plot of the logarithms of the critical voltages observed in the experiments versus 1/n should result in a straight line. Thus for every single one of the ten parametric resonances shown in Fig. 2 we deter-

R36



FIG. 3. Width of the n=3 parametric resonance as a function of the excitation voltage (peak-to-peak). Inset: shape of the n=3 resonance.

mined the threshold excitation strength and plotted it logarithmically versus 1/n. The result is shown in Fig. 4. Surprisingly the even and the odd resonances have a vastly different threshold behavior not anticipated by the theory. Odd and even resonances clearly fall into two classes, each of which forms approximately a straight line in Fig. 4. For fixed Γ the staggering behavior cannot be explained on the basis of the theory developed above, since the a_n (α_n) coefficients are only slowly varying with n and do not exhibit any odd-even staggering. While the straight-line behavior of the data was anticipated and is understood on the basis of Eq. (9), the separation of the data into two classes (odd-even staggering) is currently not understood.

We compare now the details of Fig. 4 with the theory of parametric resonances developed above. Using $\omega_z = 2\pi$ imes40 kHz extracted from Fig. 2 and the approximate value of 2 for a_n in Eq. (9) we obtain $V_c^{(n)} = \alpha \gamma^{1/n}$, where $\alpha = 14.8$ V. This theoretical value for α agrees astonishingly well with the α values of 10.3(3) V and 9.6(8) V extracted by means of a linear fit from Fig. 4. Thus, within the experimental errors, both curves in Fig. 4 converge to the same critical voltage close to the theoretically expected value for $n \rightarrow \infty$. The slopes of the two curves, however, are vastly different, corresponding to $\gamma_{\text{even}} \approx 1.0(3) \times 10^{-6}$ and $\gamma_{\text{odd}} = 2.3(2) \times 10^{-3}$ for the even and odd resonances, respectively. These γ values correspond to $\Gamma_{\text{even}} \approx 0.25 \text{ s}^{-1}$ and $\Gamma_{\text{odd}} \approx 580 \text{ s}^{-1}$, respectively. The time constant for the even resonances $T_{\rm even} = 1/\Gamma_{\rm even} \approx 4$ s is of the order of the storage time of N₂⁺ ions in our experiment. Thus in the case of the even resonances the damping mechanism for the parametric resonances and the ion loss may have a similar origin. At present, however, we have no theory that would be able to explain the large difference in the Γ_{even} and Γ_{odd} values.

In this paper we presented experimental evidence for the observation of fractional parametric resonances of order n = 1, ..., 10 in the collective excitation of the center-of-mass motion of N₂⁺ ion clouds at frequencies $\omega_n = 2\omega_z/n$. We repeated the experiments leading to the results shown in Fig.



FIG. 4. The logarithms of the measured critical (peak-to-peak) voltages V_c versus 1/n for fractional resonances of order n = 1, 2, ..., 10. The solid line shows a least-squares fit of the experimental data points by the function $V_c = \alpha \gamma^{1/n}$. The data point for n = 9 is missing due to problems with an insufficient signal-to-noise ratio and a multitude of other resonances in the immediate vicinity of the n = 9 parametric resonance.

2 with clouds of H_2^+ ions and obtained qualitatively identical results. The parametric resonances are observable only if the excitation voltage exceeds a critical voltage that depends on n. The theoretical prediction Eq. (9) is only partially confirmed. A surprising odd-even staggering of the excitation thresholds is observed. The shape of the n > 1 resonances is found to be qualitatively the same as the shape of the n=1resonance. At present the existence of the observed excitation thresholds is most naturally explained by the presence of a damping mechanism in our experiments. A possible candidate for this mechanism is proposed in [3]: damping by the ambient rest gas. We found recently that in our experiments other species of charged molecules are trapped simultaneously with the ion species under investigation. Because of the mass difference of the additionally stored charged particles, these particles do not participate in the resonant excitation of the species under investigation. Thus one may think of the additionally present charged particles as a charged inert background gas. This results in a considerable enhancement of the relevant scattering cross sections. Thus this observation lends additional support to the collisional damping mechanism proposed in [3]. Further investigation of the damping mechanism together with investigation of the oddeven staggering of the excitation thresholds for high-order parametric resonances are important directions for future research.

We benefited from fruitful discussions with Harel Primack. The experiment was supported by the Deutsche Forschungsgemeinschaft. R. B. is grateful for financial support from the Deutsche Forschungsgemeinschaft (SFB 276). X. Z. C. acknowledges a grant from the Deutscher Akademischer Austauschdienst. M. A. N. R. is grateful for financial support from the DLR International Buro's Indo-German bilateral scientific exchange program.

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