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Nonlinear Resonances and Phase Transitions of Two-Ion Coulomb Clusters in a Paul Trap: Calculations without Laser Cooling

A. W. Vogt

Institut für Theoretische Physik der Universität Göttingen, Bunsenstrasse 9, D-37073 Göttingen, Germany
(Fax: +49-551/39-9631, e-mail: (internet) vogt@hal9000.theo-phys.gwdg.de)

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Abstract. The mechanism responsible for transitions of laser-cooled trapped ions from an ordered "crystal" state to an irregular "cloud" state has been discussed controversially. A numeric and analytic study of the relative motion of two trapped ions without laser cooling is performed and compared with the results of previous simulations involving the laser. It turns out that the system without laser, in spite of its simplicity, already exhibits a non-monotonic dependence of crystal stability on trap parameters, which is linked to the presence of low-order nonlinear resonances.

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There is a persistent interest (see, e.g., [1]) in storing large numbers of laser-cooled ions in a Paul trap [2]. It has been known for several decades that under suitable conditions charged particles in such a trap are capable of forming clusters (referred to as Wigner crystals [3], Coulomb clusters, ordered states) in which trap forces and Coulomb repulsion are balanced in the average over one trap period [4]. Such crystals have been prepared by laser cooling of trapped ions [5, 6], and transitions (melting) back to an irregular (cloud) state were observed while trap parameters or laser power or detuning were being varied. The desire to identify the underlying mechanisms has spawned activities by experimenters and theorists alike [7, 8]. In particular, the question is still debated [9, 10], to which extent the melting of crystals is reproducible and what kind of forces have to be included into a model in order to explain the phenomenon.

Laplace's equation $\Delta V = 0$ for the electrostatic potential V excludes the possibility of trapping a point charge in a vacuum by static electric fields. The Paul trap achieves confinement of charged particles with the help of an oscillating field. The amplitude of the ac trap voltage is proportional to the trap parameter q_3 used below, see, e.g., [2, 4, 11] for details. A dc voltage may also be applied, which is proportional to the parameter a_3. Choosing a dimensionless time scale on which the ac voltage has period π, the equation of motion for a single ion and for the center-of-mass motion of several ions in a Paul trap has the Mathieu [12] form

$$\ddot{X}_j + (a_j - 2q_j \cos 2t) X_j = 0\,, \tag{1}$$

where

$$a_{1,2} = -a_3/2 \quad \text{and} \quad q_{1,2} = -q_3/2 \tag{2}$$

due to trap symmetry with respect to rotations about the z-axis and to the Laplace equation for the trap field. For certain parameter values the solutions of the Mathieu equation are stable [13]. Using (2) and intersecting the stability regions for j= 1, 2 on the one hand and $j = 3$ on the other hand, parameter values leading to three-dimensional confinement are obtained, see, e.g., [9]. In what follows, $q_{\mathrm{MI}}(a_3)$ denotes the upper bound of q_3 for fixed a_3 in the usual stability region, see Fig. 3.

In the case of several ions, Coulomb repulsion has to be included. Thus, the relative motion of two ions is governed by

$$\ddot{x}_j + (a_j - 2q_j \cos 2t) x_j - \frac{\alpha x_j}{|\mathbf{x}|^3} = 0\,, \tag{3}$$

where the constant α in the Coulomb force can be made equal to unity by an appropriate choice of the length scale, which is assumed in the following.

Interaction of the trapped ions with a laser beam gives rise to momentum changes, which may lead to damping and to diffusion (see, e.g., [9, 14, 15] for a discussion of laser cooling in a Paul trap).

Particular attention has been devoted to the melting of crystals of two (or a few) ions when the dc parameter a_3 is kept constant and the ac amplitude q_3 is increased.

With $a_3 = 0$, an order–chaos transition in the relative motion of two ions was reported at some critical value $q_c < q_{\mathrm{MI}}(0)$ for both experiments and computer simulations, the latter including trap forces, Coulomb repulsion, a cooling term and random displacements in coordinate space [8]. In contrast to this, other authors [9] have not found any such transition in numerical calculations, and fluctuations arising from spontaneous emissions were deemed to weak to cause order–chaos transitions in the case $a_3 = 0$ [10]. Further-

more, in experiments with short q_3-scan times, crystals were observed to remain stable in several – but not in all – runs of the experiment until $q_3 = q_{\mathrm{MI}}(a_3)$ was reached [9].

Collisions with background molecules were found to be capable of inducing transient chaos for $q_3 < q_c$ and stationary chaos for $q_3 > q_c$, but such collisions were calculated to occur on a time scale of roughly one minute [10], which left unexplained the relatively high number of melting events in the experiments of [9], which lasted only seconds.

Recently, melting at some realistic trap parameters was reproduced in computer simulations [16] mimicking two-ion experiments reported in [9]. The model used in these simulations featured trap forces, Coulomb repulsion of the ions and interaction of each ion with the laser beam, including fluctuating forces from spontaneous emissions. The model did not include collisions with background gas molecules. The trap parameters at which melting occurred in those simulations were found to be correlated with nonlinear resonances in the relative motion of two ions without laser cooling. This link between the behavior of laser-cooled ions (a non-Hamiltonian system) on the one hand and the Hamiltonian system of interacting trapped ions *without a laser* on the other hand was established for four different areas in the a_3–q_3 plane. A number of questions remained open, though, in particular:

- there was no proof that the system *without laser* itself was less stable at the trap parameters in question than elsewhere, and
- it was unexplained why the presence of a low-order nonlinear resonance did not always lead to melting in simulations including laser cooling.

The purpose of this paper is to clarify these issues by studying the system of two interacting ions in a Paul trap *without laser cooling* (mainly). The change of coordinates necessary to make apparent the nonlinear resonances mentioned in [16] will be given.

1 Numerical Calculations without a Laser

This section is devoted to a numerical study of the solutions of (3), the equations of motion of a time-dependent Hamiltonian system. It is not quite obvious that insight into the behavior of the non-Hamiltonian system with laser cooling can be gained from studying the Hamiltonian one. An attempt to do this is based on the observation that in the presence of a laser beam, even in those cases where melting does not occur, deviations from the periodic crystal solution exist, the size of which is the result of a balance between the net cooling effect of the laser light pressure and heating caused by fluctuations. The properties of the Hamiltonian system are studied in an area of phase space whose size is chosen somewhat larger than these deviations in a no-melting run of the simulation program with laser cooling. For constant trap parameters and 400 different initial values close to the crystal solution, (3) is integrated numerically over 10^4 trap cycles, using a fourth-order symplectic integrator [17]. Two types of trajectories can be distinguished: some stay close to the crystal solution, while others deviate from it by amounts

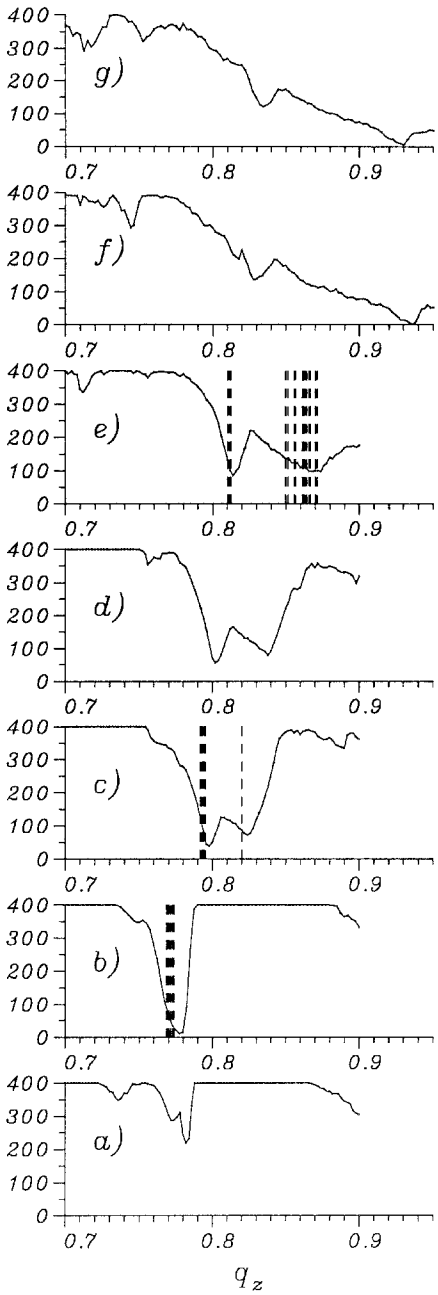

Fig. 1a–g. Number of crystals stable over 10^4 trap periods in simulation without laser and with fixed trap parameters. **a** $a_3 = 0$; **b** $a_3 = -0.003$; **c** $a_3 = -0.018$; **d** $a_3 = -0.023\ldots$; **e** $a_3 = -0.036$; **f** $a_3 = -0.050$; **b** $a_3 = -0.055\ldots$. Step width is $\Delta q_3 = 0.002$ (0.0025 in **b** and **g**). *Dashed lines* in **b**, **c** and **e** mark parameter values where melting occurred in simulations with laser cooling

several orders of magnitude larger than initially. In the latter case, the crystal is considered "molten". Figure 1 shows the number of cases in which no melting occurs as a function of the trap parameter q_3, the step width being $\Delta q_3 = 0.002$ or 0.0025. This number is used as a crude measure of stability of the two-ion crystal. The results show that crystal stability depends *non-monotonically* on the ac trap parameter q_3. This suggests that the melting of crystals might not be just a question of increasing q_3 beyond a critical value.

In order to see whether melting of laser-cooled two-ion crystals can be understood on the basis of properties of

the system without laser, in the cases $a_3 = -0.018$ and $a_3 = -0.036$, ac amplitudes q_3 where melting occurred in simulations including the laser [16] have been marked by dashed lines in Fig. 1. They coincide with local minima in the curve obtained from simulations without laser in a way which strongly suggests that properties of the Hamiltonian system under consideration provide a key to understanding the more complex system which includes laser cooling. In simulations with laser cooling, in the case $a_3 = -0.036$ melting took place in a narrow area of the q_3-axis at about $q_3 = 0.81$ and in a broader one at $q_3 = 0.85 \ldots 0.87$. This feature corresponds to a narrow dip and a broader one, respectively, in the curves indicating stability of the system without laser cooling.

In [16], simulations of laser cooling were performed for $a_3 = 0$, too, but no melting was found. In the corresponding Fig. 1a, at about $q_3 = 0.78$, there is a pronounced dip in the number of crystals which survived 10^4 trap periods in calculations without a laser. This enhanced sensitivity apparently was not strong enough to give rise to melting in simulations with laser cooling though.

Upon changing the dc parameter to $a_3 = -0.003$, the dip in question becomes deeper (see Fig. 1b), and simulations including the laser now result in melting (dashed lines). In contrast to [16], these simulations have no experimental counterpart yet. To save computer time, they are limited to a small q_3 range: calculations start at $q_3 = 0.76$, and upon increasing this parameter, melting takes place in all ten simulations (performed with different random number sequences for the modelling of spontaneous emissions) in the range $0.769 \leq q_3 \leq 0.774$.

A certain amount of arbitrariness is involved in producing Fig. 1. Clearly, the choice of initial values has an influence on the outcome. The above results have been produced with initial values obtained by adding to the crystal solution at $t = 0$ velocities on a two-dimensional grid with nonuniform spacing, as shown in Fig. 2. It corresponds to a regular spacing in variables $I_{1,2}$ to be introduced in the next section.

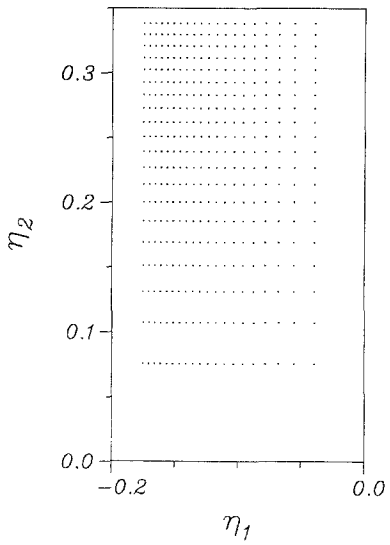

Fig. 2. 400 velocities added to the periodic crystal solution to obtain initial values for Fig. 1. Variables $\eta_{1,2}$ defined in Sect. 2

It has been checked that the main features can be obtained with other sets of initial values as well. Integration time is another interesting parameter: in examples it was found that no melting occurred over the first 100 cycles whereas the situation after 10^5 cycles did not differ from that after only 10^4 cycles. Therefore integration over 10^4 cycles was used throughout Fig. 1. The outcome was not sensitive to the integration step width in an example where this was checked, comparing step widths $\pi/20$ and $\pi/200$.

The main result of this section is that the stability of the crystal solution of the Hamiltonian system of two interacting trapped ions in a Paul trap *without laser cooling* depends in a non-monotonic fashion on trap parameters and that the melting of *laser-cooled* ion crystals takes place at trap parameters where this stability is particularly low. The remainder of this paper will try to identify a pattern behind that dependence of stability on trap parameters.

2 Nonlinear Resonances

The analysis in this section is based on a change of variables, cf. [18, 19]. Trap symmetry and the use of cylindrical coordinates allow to restrict the problem to two coordinates $\mathbf{u} \equiv (u_1, u_2) \equiv (\varrho, z)$ with momenta $\mathbf{v}$, whose equations of motion are obtained from the Hamiltonian

$$H_1 = \frac{1}{2}\mathbf{v}^2 + U(\mathbf{u}, t)\,, \tag{4}$$

where the potential is

$$U = \frac{1}{2}\frac{p_\varphi^2}{u_1^2} + \frac{1}{|\mathbf{u}|} + \frac{1}{2}(a_3 - 2q_3\cos 2t)\left(u_2^2 - \frac{1}{2}u_1^2\right) \tag{5}$$

and the angular momentum p_φ is a constant of motion. Let $\mathbf{u}^0$ denote the crystal solution in the x–y-plane $\mathbf{u}^0 = [\varrho^0(t), 0] = [\varrho^0(t + \pi), 0]$ for some fixed trap parameters and angular momentum p_φ under consideration. A new coordinate $\boldsymbol{\varepsilon}$ with momentum $\boldsymbol{\eta}$ is defined as the deviation from the crystal solution: $\boldsymbol{\varepsilon}(t) \equiv \mathbf{u}(t) - \mathbf{u}^0(t)$. The time evolution of $\boldsymbol{\eta}$ and $\boldsymbol{\varepsilon}$ is governed by the Hamiltonian

$$H_2(\boldsymbol{\eta}, \boldsymbol{\varepsilon}, t) = \frac{1}{2}\boldsymbol{\eta}^2 + U[\boldsymbol{\varepsilon} + \mathbf{u}^0(t), t] - \left.\frac{\partial U}{\partial \mathbf{u}}\right|_{\mathbf{u}^0} \cdot \boldsymbol{\varepsilon}\,. \tag{6}$$

This Hamiltonian is written as a sum

$$H_2 = H_{20} + H_{21}\,, \tag{7}$$

where

$$H_{20} \equiv \frac{1}{2}\boldsymbol{\eta}^2 + \frac{1}{2}\sum_{i,j}\varepsilon_i\varepsilon_j \left.\frac{\partial^2 U}{\partial u_i \partial u_j}\right|_{\mathbf{u}^0} \tag{8}$$

$$H_{21} \equiv U[\boldsymbol{\varepsilon} + \mathbf{u}^0(t), t] - \left.\frac{\partial U}{\partial \mathbf{u}}\right|_{\mathbf{u}^0} \cdot \boldsymbol{\varepsilon} - \frac{1}{2}\sum_{i,j}\varepsilon_i\varepsilon_j \left.\frac{\partial^2 U}{\partial u_i \partial u_j}\right|_{\mathbf{u}^0}. \tag{9}$$

H_{20} alone yields decoupled equations of motion which are linear and whose coefficients have period π (Hill's equation, see, e.g., [12, 13]. The stability of this linearized problem is checked by calculating the Floquet exponent [12, 13]. It turns

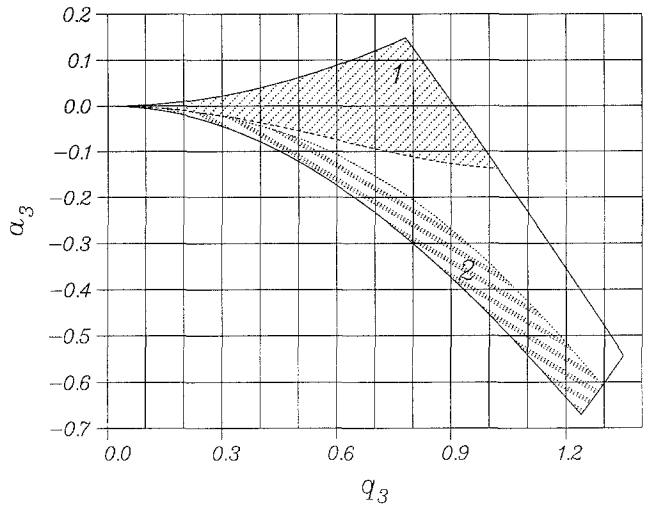

Fig. 3. The usual stability region of a Paul trap (*solid borders*). In regions 1 and 2, two-ion crystals in the x–y-plane and on the z-axis, respectively, are stable according to linearized equations of motion

out that its solutions are bounded and stable, indeed, in most of the parameter region of interest in the present context, see region 1 in Fig. 3. ($p_\varphi = 0$ was assumed here, the influence of realistic p_φ values on the location of the stability region's boundary being small.) A similar analysis can be done for the relative motion of two ions along the z-axis (rather than in the x–y-plane as treated so far); the linearized problem obtained in that case is stable in region 2 in Fig. 3. All calculations presented in this paper have trap parameters in region 1. In experiments [9] and simulations [16] with dc parameter $a_3 = -0.036$, region 2 was intersected at low q_3 values; the change of orientation of the crystal from the z-axis to the x–y-plane upon increasing q_3 was visible in the simulations.

Having established that all solutions of the linearized problem are bounded and stable in the parameter region of interest here, they will be used to define new variables for the investigation of the original nonlinear problem. The general motion of the j component in the linearized problem is a linear combination of two basic solutions

$$h^j_{1,2} = w_j(t) \begin{matrix}\cos\\ \sin\end{matrix} [\omega_j t + \beta_j(t)], \tag{10}$$

where $w_j(t)$ and $\beta_j(t)$ are real and periodic with period π. The index j refers to $u_1 \equiv \varrho$ for $j = 1$ and to $u_2 \equiv z$ for $j = 2$. It is assumed that the (constant) Wronskian

$$\det \begin{pmatrix} h^j_1(t) & h^j_2(t) \\ \dot{h}^j_1(t) & \dot{h}^j_2(t) \end{pmatrix} \tag{11}$$

equals unity for $j = 1, 2$. To achieve this, in addition to multiplying w_j by a constant, it may be necessary to interchange cos and sin in (10).

Finally, variables $(\mathbf{I}, \boldsymbol{\phi})$ are defined to satisfy

$$\varepsilon_j = \sqrt{2I_j}[\cos(\omega_j t - \phi_j) h^j_1(t) + \sin(\omega_j t - \phi_j) h^j_2(t)], \tag{12}$$

$$\eta_j = \sqrt{2I_j}[\cos(\omega_j t - \phi_j) \dot{h}^j_1(t) + \sin(\omega_j t - \phi_j) \dot{h}^j_2(t)]. \tag{13}$$

Their time evolution is given by the Hamiltonian

$$H_3 = \mathbf{I} \cdot \boldsymbol{\omega} + H_{21}[\boldsymbol{\varepsilon}(\mathbf{I}, \boldsymbol{\phi}, t), t] \equiv \mathbf{I} \cdot \boldsymbol{\omega} + H_{31}(\mathbf{I}, \boldsymbol{\phi}, t) \tag{14}$$

according to $\dot{\mathbf{I}} = -\partial H_3/\partial \boldsymbol{\phi}$, $\dot{\boldsymbol{\phi}} = \partial H_3/\partial \mathbf{I}$. This is the result given without proof in [16].

As for the interpretation of the new variables, $\mathbf{I} = 0$ corresponds to the periodic crystal solution. The frequencies $\omega_{1,2}$ depend on trap parameters a_3, q_3 and, in principle, on angular momentum p_φ. They are obtained from the Floquet exponent of the linearized problem. The influence of p_ϕ again turns out to be negligible for the present purpose, and $p_\varphi = 0$ is assumed. The physical meaning of the frequencies $\omega_{1,2}$ is similar to that of frequencies of small oscillations about the stable equilibrium position in a model which replaces the trap with a time-independent harmonic potential [11, 20, 21]. For low q_3, even the size of the frequencies agrees well with those obtained in the time-independent model, but this approximation would not be good enough for use in Fig. 4.

Since $\varepsilon_j = \sqrt{2I_j} w(t) \cos[\phi_j + \beta_j(t)]$ is periodic, H_{31} has period 2π as a function of the ϕ_j and period π as a function of t, whence a Fourier expansion

$$H_{31} = \sum c_{k_0 k_1 k_2}(\mathbf{I}) \exp[\mathrm{i}(2k_0 t + k_1\phi_1 + k_2\phi_2)], \tag{15}$$

where the sum is over integer k_j's. For small $|\mathbf{I}|$ one has [22] $d\phi_j/dt \approx \omega_j$, which implies that the arguments in the exponentials are slowly varying whenever a resonance relation

$$2k_0 + \omega_1 k_1 + \omega_2 k_2 \approx 0 \tag{16}$$

holds. The significance of such resonances for the long-time behavior of trajectories is well known (e.g., [18, 23- 25]). In particular, those resonances for which k_1 and k_2 have equal sign (sum resonances) may lead to instability of solutions [18, 23, 26]. In Fig. 4, resonance lines for $|k_1| + |k_2| \leq 8$, k_2 even [27], $k_1 k_2 \geq 0$ (sum resonances) have been drawn in the ω_1–ω_2-plane. Dotted curves (closely resembling

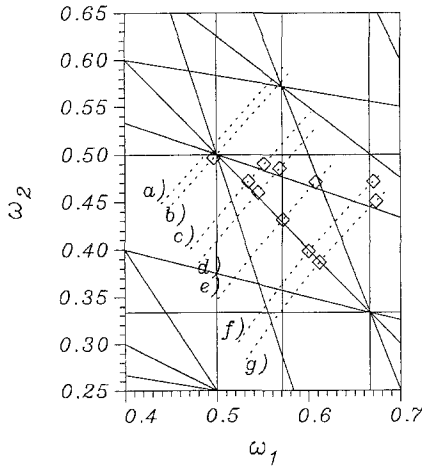

Fig. 4. On *solid lines* a relation $2k_0 + k_1\omega_1 + k_2\omega_2 = 0$ holds. Lines have been drawn for sum resonances with $|k_1| + |k_2| \leq 8$, k_2 even. *Dotted curves* show (ω_1, ω_2) values belonging to parameters a_3, q_3 as in Fig. 1, with lowest q_3 at lower left end in each curve. Symbols ($\diamond$) mark points where enhanced sensitivity of crystals was found in simulations without laser

straight lines) mark values $\omega_{1,2}$ calculated from trap voltages appearing in Fig. 1. Symbols ($\Diamond$) mark those frequencies which correspond to local minima (as a function of q_3, see Fig. 1) in the number of stable crystals in simulations without laser. Only those with less than 175 stable trajectories have been chosen in order to concentrate on the main features and because the location of these low minima is determined by the behavior of trajectories with initially small $|I_{1,2}|$, for which any frequency shifts caused by H_{31} are small, cf. [22]. It turns out that the frequencies corresponding to minima, i.e., relatively unstable crystals, lie close to resonance lines $(k_0, k_1, k_2) = (-1, 3, 0)$, $(-1, 2, 2)$, and $(-1, 1, 3)$ [27], i.e., it appears that the presence of low-order resonances is reflected in the pattern of minima in Fig. 1, thus explaining why the sensitivity of crystals to displacements from the periodic solution depends non-monotonically on trap parameters. At this point it should be noted that the explicit time dependence of the trap field is essential here since in a time-independent two-dimensional model no sum resonances can occur. Besides, there is a well-known topological argument (see, e.g., [28]) according to which two-dimensional time-dependent systems are eligible for a universal instability – Arnold diffusion [25] – while two-dimensional time-independent ones are not.

3 Conclusion

Coming back to the questions raised initially, the main results of this work can be summarized as follows.

- The melting of crystals of two laser-cooled ions in a Paul trap can indeed be understood from an enhanced sensitivity of the crystals to displacements from the ideal crystal solution which is visible already in the Hamiltonian system without laser cooling.
- This enhanced sensitivity in turn is correlated to the presence of low-order nonlinear resonances, as has been demonstrated for 11 of the local minima of Fig. 1, whose frequencies are marked in Fig. 4. But already for the system without laser the size of the effect varies strongly for different trap parameters, even for the same resonance (different heights of minima in Fig. 1a, b, e.g.), which makes it plausible that under laser cooling, melting need not always take place in the presence of such a resonance.

In the present context it is not possible to *predict* the degree of destabilization in the presence of a given resonance – this cannot be done without taking into account the dependence of coefficients $c_{k_0 k_1 k_2}(\mathbf{I})$ in (15) on action variables $\mathbf{I}$. The simple argument given above for the relevance of resonances (namely, the occurrence of slowly varying arguments in the exponentials), clearly does not contain everything known today on the complex behavior of nonlinear systems. Further work might in particular address the question of overlapping of resonances in the sense of Chirikov's overlap criterion [25].

This work dealt with the case of a two-dimensional time-dependent system. Higher dimensional ones arise if more than two ions are considered. Another way to generalize the problem would be to include higher order contributions of the trap field [29], or to consider an asymmetric trap. From what has been said above it is clear that the mere presence of low-order nonlinear resonances does not allow to predict melting of a general laser-cooled Coulomb cluster in a Paul trap, but the above results provide a warning that trap parameters with increased sensitivity of crystals to perturbations might exist. Ultimately, the occurrence of melting will also depend on laser parameters [5].

In spite of all difficulties in comparing theoretical and experimental results, arising from trap imperfections such as asymmetries and contact potentials [5], e.g., the significant non-monotonic dependence of crystal stability on trap parameters found in this paper should be amenable to experimental tests beyond the experiments already performed in [9]. In particular it would be interesting to confirm the prediction of destabilization at $a_3 \approx -0.003$, $q_3 \approx 0.78$ experimentally.

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