Multipole expansion in quadrupolar devices comprised of planar electrode arrays

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ABSTRACT

Ion trap mass analyzers made using arrays of independent electrodes allow unprecedented control and variability of electric field shapes. We present a method to select and implement specific values for higher-order multipoles, which are known to affect ion trapping and mass analysis in quadrupole ion traps. Electrode arrays are amenable to microfabrication techniques, hence this method can be used to improve performance in miniaturized ion trap mass spectrometers. With ion traps made using two opposing electrode array plates, both even- and odd-order multipoles can be independently adjusted.

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1. Introduction

Quadrupole electric potentials find widespread use in containing, guiding, and analyzing charged particles. Most commercial mass spectrometers include quadrupole mass analyzers, mass filters, or ion guides. Quadrupoles are also used to trap ions for spectroscopic studies [1], for quantum computing applications [2], and for study of chemical reaction dynamics [3]. Despite the long history and wide application, purely quadrupolar potential distributions have never been created—all quadrupolar devices contain higher-order multipoles [4,5]. For some applications, including ion trap mass spectrometry, these higher-order multipoles can have a significant effect on performance [6,7]. A classic example is the original Finnigan ion trap mass spectrometer, which was manufactured with a "stretched" geometry to correct the negative octopole that interfered with mass analysis [4]. Other ion traps have also seen dramatic changes in mass resolution resulting from small changes in the magnitude and sign of higher-order multipoles [8].

Changing the shape and/or position of electrodes is commonly done to optimize the electric fields of quadrupole ion traps [5]. Additional "compensation" electrodes (positioned between the primary electrodes) have also been suggested as a method to modify the trapping fields [9,10]. However, these techniques have a simultaneous effect on all the components of the field. For example, increasing the separation between hyperbolic electrodes in an ion trap changes not only the quadrupole field, but also the other even, higher-order field components. Several studies have reported configurations that optimize a single component in linear ion traps and quadrupole mass filters (e.g., optimizing only the 12-pole), or that optimize a linear combination of higher-order fields [11,12], but individual field components cannot be independently optimized.

Higher-order multipoles are of particular concern for miniaturized and microfabricated quadrupole-based mass analyzers. Efforts to miniaturize ion trap mass analyzers have typically relied on simplified electrode geometries. For instance, the cylindrical ion trap—a geometrically simplified version of the quadrupole ion trap—has been the basis for many miniaturized and microfabricated mass analyzers [13–18]. Similarly, the rectilinear ion trap uses electrodes that are simpler, and therefore easier to miniaturize, than the linear ion trap from which it is conceptually derived [19,20]. However, with geometrical simplification comes a decrease in the quality of electric fields and the resulting mass resolution.

Our lab has previously introduced a new way of making radiofrequency ion traps, in which the trapping fields are created between two ceramic plates [21]. A series of independently-adjustable electrodes are lithographically deposited onto each plate. The first of these devices, the Halo ion trap, creates a toroidal trapping potential using a series of 15 concentric electrode rings per plate [22]. The planar Paul trap (a 3-D quadrupole) was later demonstrated using plates with 25 electrode rings [23]. Trapping fields in both devices were determined by the RF amplitude applied to each electrode ring. Although this approach allows significant control over the field shape, including control of multipole components, no method was

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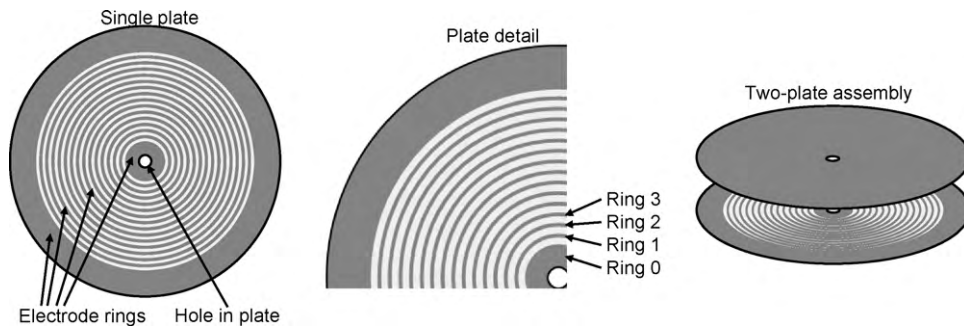*E-mail addresses:* austin@chem.byu.edu, dea@byu.edu (D.E. Austin).

Fig. 1. Quadrupole ion trap made using two plates, each with lithographically-defined electrode rings, and overlaid with a resistive layer (not shown).

reported for determining what potentials should be applied to each ring in order to produce a given field.

This article presents a solution to this problem using standard matrix algebra. The multipole expansion of each independent electrode and the desired overall trapping field form a set of linear equations which can be solved to determine the needed potentials for each electrode. This method can be used to create fields in which the higher-order terms are independently variable. For instance, a quadrupole ion trap in which the octopole, 12-pole, and 16-pole terms are all exactly zero, can be experimentally realized. Using this approach, miniaturized mass analyzers can be produced in which the electric fields are not compromised by simplification of electrode geometry. This solution applies to all quadrupolar devices consisting of arrays of electrodes on one or two plates [24]. This paper discusses the basis for and implementation of this solution.

2. Theory

Consider a quadrupole ion trap made using two rigid, insulating plates, as shown in Fig. 1. Each plate contains a 1-mm hole in the center, and includes electrode rings (with width 100 μm) centered at radii 2, 2.5, 3, 3.5, . . . , 12 mm. An additional electrode spans the distance between the 1-mm hole and a radius of 1.8 mm. Electrodes are numbered as shown. The plates are separated by 5 mm. These dimensions are similar to those of the planar Paul ion trap reported by Zhang et al. [23]. The potential on each ring is independently adjustable. A thin layer of germanium, providing a uniform surface resistance, is deposited on top of the array of electrode rings. The potential of the germanium layer directly above an electrode ring is equal to the potential applied to that ring, while the potential of the germanium between electrode rings can be solved using Ohm's Law. The same solution would be obtained if the substrate was made of a resistive material.

The potential distribution in the region between the plates is constrained by the Laplace equation:

$$\nabla^2_{x,y,z}\Phi = 0 \qquad (1)$$

The system being considered has cylindrical symmetry, so this equation is satisfied by the following potential distribution in spherical polar coordinates:

$$\Phi(\rho, \theta, \phi, t) = \Phi_0(t)\sum_{n=0}^{\infty} A_n \frac{\rho^n}{\rho_0^n} P_n(\cos\theta) \qquad (2)$$

where P_n is the Legendre polynomial of order n, and ρ_0 is the characteristic dimension (for ion traps this is typically the inner radius of the ring electrode). The set of terms with $n = 0$ (monopole), $n = 1$ (dipole), $n = 2$ (quadrupole), $n = 3$ (hexapole), etc., are referred to as the multipole expansion of a given potential distribution. The A_n-terms in this equation represent the magnitude of each pole. The A_0-term does not affect the behavior of charged particles within the potential distribution because it produces no electric field. For primarily quadrupolar devices, the A_2-term dominates. Odd-order terms (A_1, A_3, A_5, etc.) are zero for systems with symmetry about a plane perpendicular to the rotational axis, including conventional quadrupole ion traps. Even, higher-order multipoles (i.e., A_4, A_6, A_8, etc.) sometimes have significant effect on the behavior of trapped ions, and are therefore of particular concern for quadrupole-based mass analyzers. Eq. (2) can be expanded for terms $n = 2$, 4, and 6 in cylindrical coordinates as follows:

$$\Phi(r, z, \phi, t) = \Phi_0(t)\left[A_2\frac{r^2 - 2z^2}{2r_0^2} + A_4\frac{3r^4 - 24r^2z^2 + 8z^4}{8r_0^4} + A_6\frac{5r^6 - 90r^4z^2 + 120r^2z^4 - 16z^6}{16r_0^6}\right] \qquad (3)$$

The potential along the z axis (the axis of rotation) can be found by evaluating the above equation at $r = 0$:

$$\Phi(z, \phi, t)_{r=0} = \Phi_0(t)\left[A_2\frac{-z^2}{r_0^2} + A_4\frac{z^4}{r_0^4} + A_6\frac{-z^6}{r_0^6}\right] \qquad (4)$$

The system under consideration does not possess a characteristic radial dimension, so it is more convenient to define the A-terms based on z_0, which is half the plate spacing:

$$\Phi(z, \phi, t)_{r=0} = \Phi_0(t)\left[A_2\left(\frac{z}{z_0}\right)^2 + A_4\left(\frac{z}{z_0}\right)^4 + A_6\left(\frac{z}{z_0}\right)^6 + \ldots\right] \qquad (5)$$

For convenience, the signs of some of the terms in Eq. (5) have been switched, such that the dependences of all terms on z have the same sign. In this case, all A-terms of even-order multipoles will have the same sign when the poles themselves have the same sign with respect to z. A superlinear field under this convention is one in which the higher-order multipoles have the same sign as the primary quadrupole. This latter equation is the definition that will be used throughout this paper.

Because of the superposition principle, the multipole expansion of an ion trap made with an array of electrodes is equal to the sum of the multipole expansions created by each individual electrode in such an array. For instance, the quadrupole content, A_2, of an ion trap made using an array of m rings is

$$A_2 = A_{2,1}\Phi_{01} + A_{2,2}\Phi_{02} + A_{2,3}\Phi_{03} + \ldots + A_{2,m}\Phi_{0m} \qquad (6)$$

where Φ_{0m} is the applied RF amplitude (Φ_0) to ring m, and $A_{2,m}$ is the quadrupole ($n = 2$) term created by ring m alone. Similar equations can be written for each of the higher-order multipoles. This process leads to n equations in m variables, the $A_{n,m}$-terms of which

can be expressed as a matrix:

$$
\begin{pmatrix}
A_{2,1} & A_{2,2} & A_{2,3} & \cdots & A_{2,m} \\
A_{4,1} & A_{4,2} & A_{4,3} & \cdots & A_{4,m} \\
\vdots & \vdots & \vdots & & \vdots \\
A_{n,1} & A_{n,2} & A_{n,3} & \cdots & A_{n,m}
\end{pmatrix}
\tag{7}
$$

The above matrix multiplied by the set of RF amplitudes for each ring, Φ_{0m}, yields the set of A-terms for the whole device:

$$
\begin{pmatrix}
A_{2,1} & A_{2,2} & A_{2,3} & \cdots & A_{2,m} \\
A_{4,1} & A_{4,2} & A_{4,3} & \cdots & A_{4,m} \\
\vdots & \vdots & \vdots & & \vdots \\
A_{n,1} & A_{n,2} & A_{n,3} & \cdots & A_{n,m}
\end{pmatrix}
\times
\begin{pmatrix}
\Phi_{0^1} \\
\Phi_{2^2} \\
\Phi_{0^3} \\
\vdots \\
\Phi_{0^m}
\end{pmatrix}
=
\begin{pmatrix}
A_2 \\
A_4 \\
\vdots \\
A_n
\end{pmatrix}
\tag{8}
$$

The above equation can be solved using standard matrix methods. When $n > m$, and the equations are all linearly independent, the matrix is overdetermined, and no solution is possible. For $n < m$, the system is underdetermined, and a family of solutions will always exist. In theory, for a device consisting of m electrode rings, up to m separate even-ordered A-terms can be constrained. In practice, however, the RF amplitudes applied to each ring must lie within a reasonable range. It may also be desirable to maximize the well depth for a given RF power supply, or to have only one phase of RF used. These factors add additional constraints to the system of equations and reduce the number of A-terms that can be selected for a given number of independent electrodes. For underdetermined systems (i.e., there are more electrode rings than there are multipole terms we wish to constrain) the matrix can be simplified to echelon form using Gaussian elimination, and then additional constraints (i.e., voltage ranges) can be included. Mathematical software, including Excel, can also be used to solve the above set of equations.

The multipole expansion created from a single electrode can be calculated, as explained below. Once the expansions for each elec-trode have been calculated, these values combined with the desired multipole expansion for the whole device determine the operating voltages for each electrode. A related method was used to optimize the rectilinear ion trap [25] by dividing each electrode into thin strips and then superimposing the multipole expansions of each strip, although the strips did not employ variable or independent potentials.

3. Computational methods

The potential distributions created by individual electrode rings were calculated using SIMION 8 software. For each electrode, the potential of that electrode was set to 1000 V while the potentials on all other electrodes were set to zero. Spacing between electrode plates was set to 1000 grid units to provide enough calculated points for an accurate multipole expansion. The potentials along the resistive germanium layer were calculated in SIMION using a method presented previously [21]. Isopotential contour profiles created by five representative electrodes are shown in Fig. 2. Note that all show significant contributions from higher-order fields—all show strong deviations from the hyperbolic isopotentials of an ideal quadrupole.

Once the potential distribution was solved for an individual electrode, a neutral was flown along the z axis between the two plates, and the potential recorded at every grid unit (using the "Ion's Every Time Step" function with the default value (0) for trajectory computational quality). For this calculation a neutral particle gives more consistent point spacing than a charged particle, the latter being accelerated by the electric field of the device. Values of z were normalized with respect to the plate spacing. The multipole expansion of the whole trapping volume is characterized by the potential function along the z axis, as seen from Eq. (5).

The multipole expansion created by each electrode was solved by fitting the potentials along the z axis to a 25th order polynomial using the *polyfit* function in Matlab. Calculated multipole expansions did not converge well when less than 20 fit variables were used, although more variables increased computation time. A previous report utilizing a similar approach to calculate multi-
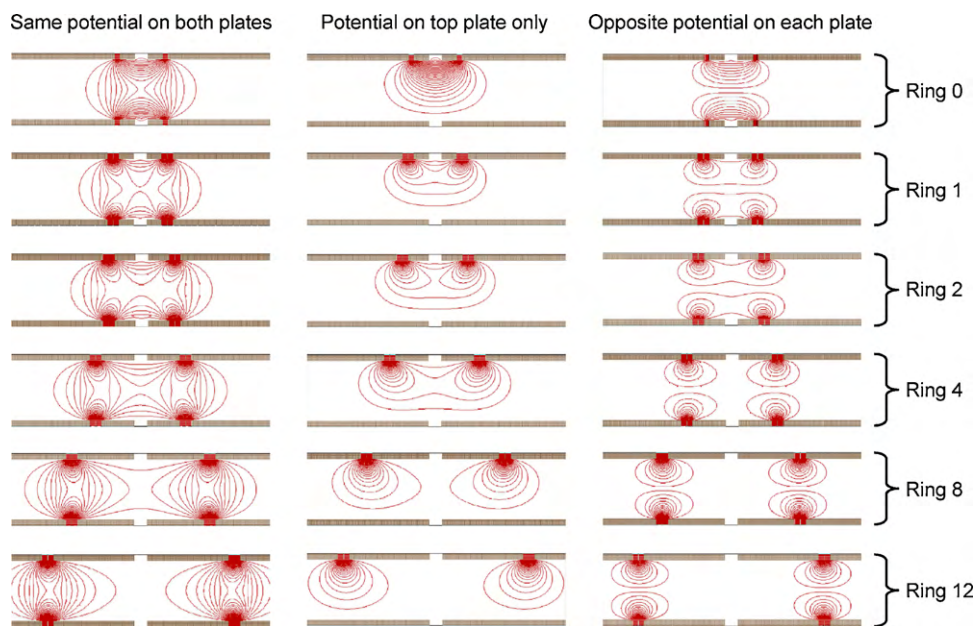
Fig. 2. Isopotential contours created by applying potentials to individual electrode rings (0, 1, 2, 4, 8, and 12). Left column: potential of the same polarity applied to given ring on both plates. This mode creates the primary quadrupolar field. Center column: potential applied to given ring on one plate only. Right column: opposite polarity applied to given ring of both plates. This is the mode that would be used for resonant ejection and ion excitation. The number of contour lines per volt applied is not constant throughout the diagrams, and was chosen in each case for visual clarity.

Table 1
Even A_n-terms for the first 12 electrode rings of ion trap from Fig. 1. The same potential is applied to each plate, so all odd A_n-terms are zero.

| | Ring # | | | | | | | | | | | |
	0	1	2	3	4	5	6	7	8	9	10	11
A_2	532.4	−31.1	−70.9	−76.4	−73.3	−61.8	−49.9	−39.2	−30.5	−23.4	−17.9	−13.6
A_4	172.0	−123.4	−65.9	−24.6	−3.4	4.8	7.2	6.9	6.0	4.8	3.8	2.8
A_6	−68.4	−2.7	27.2	25.3	11.5	6.9	1.7	2.0	0.2	0.3	−0.2	0.5
A_8	−75.2	25.0	34.6	−14.3	11.7	−7.9	6.3	−7.3	1.2	−1.6	1.4	−3.7
A_{10}	67.2	109.9	−108.5	92.7	−76.5	32.5	−35.9	33.3	−6.5	6.5	−8.3	17.4

pole expansions found that 22 variables provided a sufficient level of accuracy for low-order multipoles [26]. In all cases, more fitting parameters are needed for accurate values of higher-order terms in the multipole expansion. After the potentials were fit to 25th order polynomial functions, the coefficients were normalized with respect to the value of the potential applied to the electrode (1000 V is used in SIMION to reduce rounding errors in subsequent steps). The coefficients determined by Matlab were used to populate the matrix of Eq. (8). The multipole expansion of the whole device calculated this way can later be normalized with respect to the overall quadrupole. In this work, higher-order multipoles of the whole device are expressed as ratios of A_n to A_2, as is commonly done elsewhere.

Variables such as plate spacing, electrode location and width, the size of the central hole, and the electrode density are easily studied using this approach. In addition, odd-order terms (dipole, hexapole) can be explored—both theoretically and experimentally—by applying potentials to electrodes of only one of the two plates. Odd-order terms may be useful for ion excitation, dissociation, and ejection.

4. Results and discussion

Table 1 shows the even A_n-terms ($2 \leq n \leq 10$) for the first 12 electrode rings of the ion trap described in Fig. 1. The potential is applied to the same ring on both plates. These terms depend only on physical structure, and are not dependent on the applied RF, or on the voltages selected for operation. These values were calculated assuming 1 V applied to the electrode of interest, and zero volts applied to all other electrodes. Fig. 3 shows the A_n/A_2 ratios ($n = 4, 6, 8$) for the first twelve electrode rings of the same trap. Even-order multipoles (the top plot) are created by applying the identical potential to a given electrode ring on both plates. Odd-order multipoles (the lower plot) are created by applying potentials of opposite sign to the given electrode ring on the two plates (i.e., +V on one plate, −V on the other plate). This figure illustrates the independent nature of the equations represented in Eq. (8), and shows that the higher-order terms of the whole device can be controlled by selection of the potentials applied to each electrode ring.

Fig. 4 shows examples of four ion traps, each with different values of several higher-order multipoles. Above are plots of the isopotential lines within each device. Below is a plot of the higher-order components of the axial electric field (at $r = 0$) in each trap. The centers of each trap appear similar in this figure. However, differences appear as small distortions near the trapping plates in each design. These correspond to field components of higher order than those specified during optimization. For ion trap operation, field components beyond the 16-pole have less effect on ions at the trapping center, but may have effects on ion trapping and ejection. Nevertheless, these designs illustrate that higher-order components can be independently varied to optimize a particular ion trap experiment. For each trap in Fig. 4 the same maximum voltage is applied to each plate, although the voltages applied to individual rings vary between each design. Because relative field components are independent of the magnitude of the applied voltage, the same field shape can be made with any operating parameters (voltage or

frequency). Thus the fields shown in Fig. 4 apply to typical operating conditions for this ion trap.

Although low-order field components can be controlled, the distortions in Fig. 4 illustrate that there will always be higher-order components that are not negligible. In a sense, this represents a conservation of edge effects: one can push the edge effects out of the low-order field components, but only at the expense of the higher components.

The two-plate approach can be used as the basis for experiments on the effects of individual multipole components. For instance, the effects of octopole and dodecapole, which are often treated together in the ion trap literature, can be separately explored and optimized. Undesirable non-linear resonances can be reduced or eliminated during dipole resonance ejection in regions of the stability diagram where such non-linear resonances are a problem. The effects of odd-order poles can be studied. The speed with which ions accumulate energy during resonant ejection depends on the order of the field in which the resonance takes place, hence it may be possible to improve mass resolution by taking advantage of novel combinations of fields. Non-linear resonances with higher-order multipoles have been used by Moxom et al. to improve mass resolution in miniaturized cylindrical ion traps [27]. The current procedure may be used in combination with this
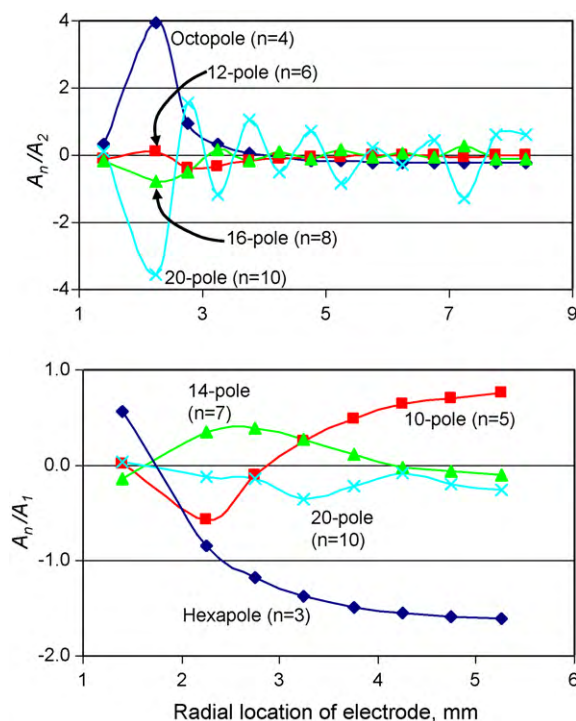

Fig. 3. Above: relative even-order multipoles (A_n/A_2 for $n = 4, 6, 8$, and 10) as a function of electrode ring location. Potentials of the same sign are applied to the given electrode on each plate. Below: relative odd-order multipoles (A_n/A_1 for $n = 3, 5, 7$, and 9) as a function of electrode ring location. Potentials of opposite sign are applied to the given electrode on each plate.
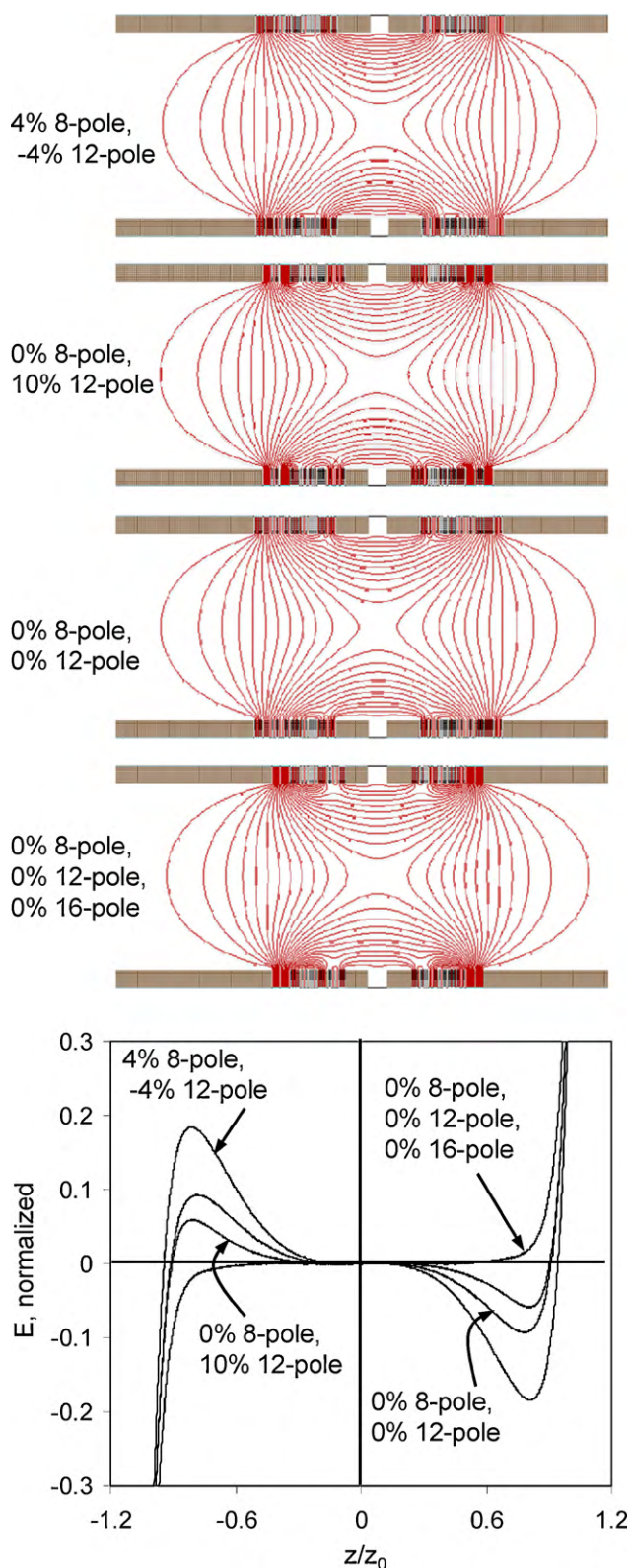
technique to provide higher-order fields optimized for resonant ejection.

The above method can be extended to other types of ion traps, including a toroidal ion trap and a linear ion trap, or to a quadrupole mass filter. It is important to note that no multipole expansion (similar to Eq. (2)) has yet been identified that exactly satisfies the Laplace equation for a toroidal trapping geometry. Although it may still be useful to think about ion behavior in a toroidal trap using spherical or cylindrical harmonics, it is not strictly correct to do so.

An advantage of the above approach is that the A-terms for each electrode need to be calculated only once for a given physical configuration. Both in calculations and in practice, changing the multipole expansion of the whole device requires changing only the RF amplitudes applied to each electrode.

5. Conclusions

Multipoles higher than quadrupole can be specified and controlled in quadrupolar devices made using two lithographically-patterned plates. The superposition principle allows calculation of the multipole expansion of the device using the expansions of each electrode elements patterned on the plates. Sample calculations demonstrate the ability to create ion traps in which the octopole, dodecapole, and 16-pole are independently adjustable. Edge effects, including the effect of an ion ejection hole, still create higher-order field components, but these can be pushed out to higher orders. This method represents a new tool to study the effects of higher-order fields on ion trapping, mass analysis, and ion activation in quadrupole ion traps. The approach can also be used to compensate for the field deterioration present in miniaturized ion traps for portable mass spectrometers.

Acknowledgement

Fig. 4. Isopotential contours for four ion traps with specified multipole profiles: 4% octopole and −4% dodecapole; 0% octopole and +10% dodecapole; 0% octopole and 0% dodecapole; 0% octopole, dodecapole, and 16-pole. The number of contour lines per volt is constant throughout this figure. For 1000 V applied, isopotential lines are separated by 42 V. Bottom: higher-order fields (trapping field minus quadrupolar field component) for each of the above devices.

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