

Original article

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ANALYTICAL STUDY OF OPERATING MODES OF RF ION FUNNELS IN THE GAS DYNAMIC INTERFACES OF TANDEM TRIPLE-QUADRUPOLE MASS-SPECTROMETERS

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Abstract. The article considers analytical models of high-frequency electric fields which can be used effectively for fast, high-quality simulation of ion flow focusing and transport processes in the radio-frequency funnels. In particular, the use of such devices in the design of a tandem three-quadrupole mass spectrometer increases the amount of ions collected in the forevacuum region of the gas-dynamic interface of the electrospray ion source. The cases of funnels with two- and four-phase electrical voltages (options I and II), as well as with amplitude-modulated electrical voltages providing a pseudopotential mode with an Archimedean wave (III) have been analyzed. As a result, the most preferable design turned out to be option III. The use of such analytical models makes it possible to test effectively promising options and thereby significantly reduce costs for the preliminary selection of a principal scheme of a device with specified characteristics, including similar cases of other mass spectrometric designs.

Keywords: mass spectrometry, gas dynamic interface, radio-frequency trap, tandem mass spectrometer, triple-quadrupole mass-spectrometer

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АНАЛИТИЧЕСКОЕ ИССЛЕДОВАНИЕ РЕЖИМОВ РАБОТЫ РАДИОЧАСТОТНЫХ ВОРОНОК В ГАЗОДИНАМИЧЕСКИХ ИНТЕРФЕЙСАХ ТАНДЕМНЫХ ТРЕХКВАДРУПОЛЬНЫХ МАСС-СПЕКТРОМЕТРОВ

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Аннотация. В статье рассмотрены аналитические модели высокочастотных электрических полей, которые можно эффективно использовать для быстрого качественного моделирования процессов фокусировки и транспорта ионных потоков в радиочастотных воронках. В частности, применение таких устройств в конструкции тандемного трехквadrupольного масс-спектрометра увеличивает количество ионов, собираемых в форвакуумной области газодинамического интерфейса электроспрейного источника ионов. Проанализированы случаи функционирования воронок с двух- и четырехфазными электрическими напряжениями (варианты I и II), а также с амплитудно-модулированными электрическими напряжениями, обеспечивающими режим псевдопотенциала с архимедовой волной (III). В результате проведенного анализа наиболее предпочтительной конструкцией оказался III вариант. Использование подобных аналитических моделей позволяет эффективно проверять перспективные варианты и тем самым существенно снизить трудозатраты на предварительный выбор принципиальной схемы устройства с заданными характеристиками, в том числе и в других масс-спектрометрических разработках.

Ключевые слова: масс-спектрометрия, источник ионов, электрораспыление, газодинамический интерфейс, радиочастотная ловушка, тандемный трехквadrupольный масс-спектрометр

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Introduction

The paper considers one of the elements of a tandem triple-quadrupole mass spectrometer with electrospray ionization, namely, a radio frequency funnel placed in the forevacuum region of the gas dynamic interface of the ion source. Such a mass spectrometer is currently developed at the National Research Nuclear University MPhI as part of the Federal Project "Development of domestic instrumentation for civil purposes" supported by the Ministry of Science and Higher Education of the Russian Federation [1–10].

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Focusing RF funnels [11], designed to reduce ion losses in the gas-dynamic interface of the ion source, are considered in our study using analytical models. Unfortunately, direct simulation of such devices requires vast computing resources. On the other hand, while analytical models only provide qualitative rather than quantitative results for funnel operation, they allow to effectively select the most promising directions for optimizing the final designs. This way, the labor costs for selecting initial candidates are drastically reduced, so better characteristics of the final design can be achieved.

The paper considers analytical models of high-frequency electric fields of RF funnels, using them to preselect the design for such an element of the gas-dynamic interface of an electrospray ion source. Decisions are made for the potential of different designs. The main results briefly overviewed in this study will be presented in more detail in the future.

Model electric fields for transport channels with circular apertures

SRIG-type (Stacked Ring Ion Guide) RF electric traps, discussed in detail in [12, 13], are a chain of circular apertures (Fig. 1), to which high-frequency electric voltages with opposite phases at adjacent apertures are applied.

The locking effect of the RF electric field increases exponentially for these devices away from the axis and approaching the boundaries of the electrodes, while the locking effect of such an electric field for standard multipole RF traps with long cylindrical electrodes polynomially increases away from the axis and approaching the boundaries of the electrodes. Thus, SRIG traps provide more reliable retention of charged particles (in our case, ions) than classical multipole RF traps.

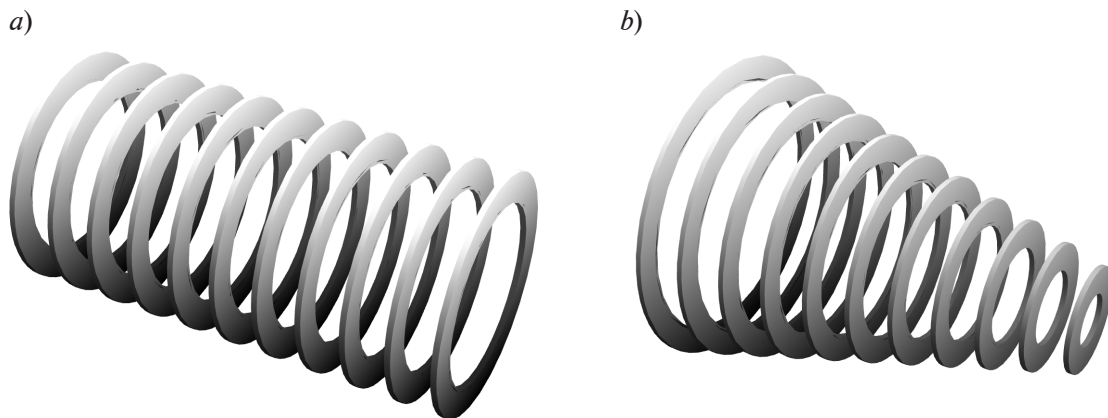


Fig. 1. Electrode configurations of SRIG-type RF traps with cylindrical (a) and conical (b) channels for ion retention and transport

The electric potential $U(z, r, t)$ near the axis in a classical SRIG-type RF electric trap with apertures of the same radius (see Fig. 1, a), where the phase difference of RF voltages applied to adjacent apertures is equal to π , is described with good accuracy by either of the two formulas:

$$U(z, r, t) = U_C(z, r) \cos(\omega t + \varphi), \quad U(z, r, t) = U_S(z, r) \cos(\omega t + \varphi), \quad (1)$$

where

$$U_C(z, r) = \frac{U_R}{I_0(\pi R/L)} \cos\left(\frac{\pi z}{L}\right) I_0\left(\frac{\pi r}{L}\right),$$

$$U_S(z, r) = \frac{U_R}{I_0(\pi R/L)} \sin\left(\frac{\pi z}{L}\right) I_0\left(\frac{\pi r}{L}\right). \quad (2)$$



U_R , ω , φ in Eqs. (1), (2) are the amplitude, circular frequency and phase of RF voltages applied to the electrodes; R is the inner radius of circular apertures; L is the distance between adjacent apertures; $r = \sqrt{x^2 + y^2}$ is the distance in the radial direction to the axis from a point with

Cartesian spatial coordinates (x, y, z) ; t is time; I_0 is the modified zero-order Bessel function [14].

The electric potential near the axis for a radio frequency trap with a conically narrowing transport channel (see Fig. 1,b), in which the phase difference of RF voltages applied to adjacent apertures is equal to π , is described with good accuracy by either of the two formulas:

$$V(z, r, t) = V_C(z, r) \cos(\omega t + \varphi), \quad V(z, r) = V_S(z, r, t) \cos(\omega t + \varphi), \quad (3)$$

where

$$\begin{aligned} V_C(z, r) &= \frac{U_R}{I_0(\pi R/L)} \left[z \cos\left(\frac{\pi z}{L}\right) I_0\left(\frac{\pi r}{L}\right) + r \sin\left(\frac{\pi z}{L}\right) I_1\left(\frac{\pi r}{L}\right) \right], \\ V_S(z, r) &= \frac{U_R}{I_0(\pi R/L)} \left[z \sin\left(\frac{\pi z}{L}\right) I_0\left(\frac{\pi r}{L}\right) - r \cos\left(\frac{\pi z}{L}\right) I_1\left(\frac{\pi r}{L}\right) \right]. \end{aligned} \quad (4)$$

In these formulas, I_1 , a modified first-order Bessel function, is added to the previous notation [14].

If it is necessary to achieve an additional effect, the electric field of the funnel should be modified in a non-linear way, using additive correction term, quadratic with respect to the z coordinate. When such field distortions are introduced, the design of the funnel, i.e., the positions and diameters of individual circular apertures, must be changed accordingly. The problem of calculating the position and shape of the electrodes from a given electric field is elementary, unlike the opposite problem, which once again shows the advantages of an analytical approach using model distributions of the electric field.

Any of these formulas can be used for analytical description of the correcting potential $W(z, r, t)$ with a quadratic dependence on the z coordinate (along the axis of the device):

$$W(z, r, t) = W_C(z, r) \cos(\omega t + \varphi), \quad W(z, r, t) = W_S(z, r) \cos(\omega t + \varphi), \quad (5)$$

where

$$\begin{aligned} W_C(z, r) &= \frac{U_R}{I_0(\pi R/L)} \left\{ z^2 \cos\left(\frac{\pi z}{L}\right) I_0\left(\frac{\pi r}{L}\right) + 2zr \sin\left(\frac{\pi z}{L}\right) I_1\left(\frac{\pi r}{L}\right) - \right. \\ &\quad \left. - \frac{1}{2} r^2 \cos\left(\frac{\pi z}{L}\right) \left[I_0\left(\frac{\pi r}{L}\right) + I_2\left(\frac{\pi r}{L}\right) \right] \right\}, \\ W_S(z, r) &= \frac{U_R}{I_0(\pi R/L)} \left\{ z^2 \sin\left(\frac{\pi z}{L}\right) I_0\left(\frac{\pi r}{L}\right) - 2zr \cos\left(\frac{\pi z}{L}\right) I_1\left(\frac{\pi r}{L}\right) - \right. \\ &\quad \left. - \frac{1}{2} r^2 \sin\left(\frac{\pi z}{L}\right) \left[I_0\left(\frac{\pi r}{L}\right) + I_2\left(\frac{\pi r}{L}\right) \right] \right\}, \end{aligned} \quad (6)$$

I_2 is a modified second-order Bessel function [14].

To check if the functions (2), (4), (6) satisfy the three-dimensional Laplace equation and, therefore, whether they can be considered as electric potentials of some electrostatic field, the Wolfram Mathematica program [15] can be used, which provides an effective tool for symbolic calculations.

Note. The assumption of quasi-static high-frequency electric field is used for formulas of model electric potentials (1), (3), (5). It is valid when the time of the characteristic change in electrical voltages at the electrodes significantly exceeds the time of propagation of an electromagnetic perturbation within the device. Typical sizes of electrode configurations used in mass spectrometer designs are tens of centimeters. Taking into account the equality of the speed of light and the propagation velocity of electromagnetic perturbation, this assumption is obviously fulfilled for the frequencies of electrical voltages used in mass spectrometers (they amount to several gigahertz). In this case, the high-frequency electric potential, which varies in time and space, can be expressed as a product of a time function (it describes the temporal change in electrical voltages) by an electrostatic potential (corresponds to DC voltages at the electrodes). Such a step is, in fact, neglecting the electrodynamic effects, i.e., the accompanying electromagnetic wave.

Pseudopotential model of ion motion in the presence of viscous friction effect

A qualitative description of the motion of charged particles in high-frequency electric fields can be obtained with a pseudo-potential model of motion, according to which motion is divided into the sum of two terms: a «slow» component in some effective quasi-stationary force field and motion as high-frequency oscillations with a small amplitude.

Let us consider the motion of ions in a high-frequency electric field, whose potential has the form [16]:

$$U_{rf}(x, y, z, t) = \sum_k \left(p_k(t) \cos(\omega_k t + \varphi_k) U^{(k)}(x, y, z) + q_k(t) \sin(\omega_k t + \varphi_k) V^{(k)}(x, y, z) \right), \quad (7)$$

where $p_k(t)$, $q_k(t)$ are «slow» functions of time; ω_k are "fast" frequencies far apart from each other on the frequency scale; $U^{(k)}(x, y, z)$, $V^{(k)}(x, y, z)$ are electrostatic fields corresponding to DC voltages at the electrodes of the device.

It is worth noting here that such concepts as "slow", "fast" and "far away" correspond to the characteristic times T_0 of the motion of ions in the transport channel:

$$dp_k(t)/dt \sim 1/T_0, dq_k(t)/dt \sim 1/T_0, \omega_k \gg 1/T_0, \forall i \neq j: |\omega_i - \omega_j| \gg 1/T_0,$$

in accordance with the pseudopotential model of ion motion (see [16, 18] and references therein).

In the presence of neutral gas, its action can be replaced by the presence of effective viscous friction, whose strength is set by Stokes' law [17]. Then, e , m are the charge and mass of the ion; $\Omega = \gamma/m$ is the effective frequency of collisions of ions with neutral gas molecules ($\gamma = \gamma(x, y, z, t)$) is the Stokes coefficient for the effective viscosity due to collisions of ions with neutral gas molecules in the vicinity of the considered point in space at the given time; this coefficient varies slowly over time and does not depend (in the first approximation) on the relative velocity of ions.

In further calculations, the subscripts for the functions $U^{(k)}(x, y, z)$ and $V^{(k)}(x, y, z)$ denote partial derivatives of potentials with respect to the corresponding spatial variables, the arguments of potentials are omitted for brevity.

After careful averaging of the equations of motion in a high-frequency electric field, it turns out that the above slow motion of the ion is carried out in a pseudopotential electric field with the pseudopotential $\bar{U}(x, y, z, t)$, which is due to the spatial gradient of the amplitude of the high-frequency electric field. This pseudopotential is expressed as [18]:

$$\bar{U}(x, y, z, t) = \sum_k \frac{e}{4m(\Omega^2 + \omega_k^2)} \left(p_k^2(t) \left[\left(U_x^{(k)} \right)^2 + \left(U_y^{(k)} \right)^2 + \left(U_z^{(k)} \right)^2 \right] + q_k^2(t) \left[\left(V_x^{(k)} \right)^2 + \left(V_y^{(k)} \right)^2 + \left(V_z^{(k)} \right)^2 \right] \right), \quad (8)$$



In addition, there is a non-potential pseudo-electric field in the equations of slow motion, with components $(\bar{E}_x, \bar{E}_y, \bar{E}_z)$ associated with the presence of viscous friction and with the spatial phase gradient of the high-frequency electric field:

$$\begin{aligned} \bar{E}_x(x, y, z, t) = \sum_k \frac{e(\Omega/\omega_k)}{2m(\Omega^2 + \omega_k^2)} p_k(t) q_k(t) & \left(U_x^{(k)} V_{xx}^{(k)} - U_{xx}^{(k)} V_x^{(k)} + \right. \\ & \left. + U_y^{(k)} V_{xy}^{(k)} - U_{xy}^{(k)} V_y^{(k)} + U_z^{(k)} V_{xz}^{(k)} - U_{xz}^{(k)} V_z^{(k)} \right), \end{aligned} \quad (9)$$

$$\begin{aligned} \bar{E}_y(x, y, z, t) = \sum_k \frac{e(\Omega/\omega_k)}{2m(\Omega^2 + \omega_k^2)} p_k(t) q_k(t) & \left(U_x^{(k)} V_{xy}^{(k)} - U_{xy}^{(k)} V_x^{(k)} + \right. \\ & \left. + U_y^{(k)} V_{yy}^{(k)} - U_{yy}^{(k)} V_y^{(k)} + U_z^{(k)} V_{yz}^{(k)} - U_{yz}^{(k)} V_z^{(k)} \right), \end{aligned} \quad (10)$$

$$\begin{aligned} \bar{E}_z(x, y, z, t) = \sum_k \frac{e(\Omega/\omega_k)}{2m(\Omega^2 + \omega_k^2)} p_k(t) q_k(t) & \left(U_x^{(k)} V_{xz}^{(k)} - U_{xz}^{(k)} V_x^{(k)} + \right. \\ & \left. + U_y^{(k)} V_{yz}^{(k)} - U_{yz}^{(k)} V_y^{(k)} + U_z^{(k)} V_{zz}^{(k)} - U_{zz}^{(k)} V_z^{(k)} \right). \end{aligned} \quad (11)$$

In addition to these pseudo-forces, the equations of slow motion also include the viscous friction force (originally present in them) with components (F_x, F_y, F_z) :

$$\begin{aligned} \bar{F}_x(x, y, z, t) &= -\gamma(x, y, z, t) (\dot{x}(t) - u_x(x, y, z, t)), \\ \bar{F}_y(x, y, z, t) &= -\gamma(x, y, z, t) (\dot{y}(t) - u_y(x, y, z, t)), \\ \bar{F}_z(x, y, z, t) &= -\gamma(x, y, z, t) (\dot{z}(t) - u_z(x, y, z, t)), \end{aligned} \quad (12)$$

where (u_x, u_y, u_z) are the components of the gas flow velocity (it changes slowly over time) in the vicinity of the considered point in space at the considered time; $(\dot{x}, \dot{y}, \dot{z})$ are the velocity components of the slow (averaged over fast oscillations) motion of the ion.

Analysis of the properties of a conical funnel with two-phase power supply

We use a pseudopotential model to describe the motion of ions in a high-frequency electric field (3). The pseudopotential is calculated using the general Eq. (8). In this case, there is no spatial phase gradient of the high-frequency electric field, so we are dealing with a non-potential pseudo-electric force. The three-dimensional graph of the pseudopotential has the form of a kind of gutter with the edges that grow sharply with distance away from the axis and approaching the electrodes, and the slope of the edges of the gutter increases as it approaches the outlet from the funnel. This means that such a high-frequency electric field effectively «presses» the ions to the axis of the device, and this pressing force increases significantly along the direction of motion, making the ion beam progressively narrower.

The presence of pseudopotential corrugation on the axis of the system can be considered an unpleasant effect, since it can create parasitic local traps for ions. In addition, in such a design, there is no relying on cooling of the ions (discharging their excess kinetic energy), since the high-frequency electric field on the axis is not zero and therefore the ions are constantly oscillated by this field.

In addition to these disadvantages, the distribution of the pseudopotential function along the funnel axis slowly increases towards its outlet, which slows down the motion of ions and makes it difficult for them to escape through the outlet. For such a funnel, it turns out necessary to apply an additional pulling electric field. Such a measure can be implemented if additional static potentials are applied to the apertures and a constant increase in static electric potential is ensured between adjacent apertures.

Analysis of the properties of a conical funnel with four-phase power supply

For a radio frequency trap with conical electrodes (see Fig. 1,*b*), in which the phase difference of RF voltages applied to adjacent apertures is equal to $\pi/2$, the electric potential near the axis is described with good accuracy by the expression

$$V(z, r, t) = V_C^*(z, r) \cos(\omega t + \varphi) + V_S^*(z, r) \sin(\omega t + \varphi), \quad (13)$$

where

$$\begin{aligned} V_C^*(z, r) &= \frac{U_R}{I_0(\pi R/2L)} \left[z \cos\left(\frac{\pi z}{2L}\right) I_0\left(\frac{\pi r}{2L}\right) + r \sin\left(\frac{\pi z}{2L}\right) I_1\left(\frac{\pi r}{2L}\right) \right], \\ V_S^*(z, r) &= \frac{U_R}{I_0(\pi R/2L)} \left[z \sin\left(\frac{\pi z}{2L}\right) I_0\left(\frac{\pi r}{L}\right) - r \cos\left(\frac{\pi z}{2L}\right) I_1\left(\frac{\pi r}{2L}\right) \right]. \end{aligned} \quad (14)$$

In this case, a gutter also appears on the pseudopotential graph, which effectively «presses» the ions to the axis of the device, and the force of such an impact increases quadratically as it approaches the outlet from the funnel. However, there is no pseudopotential corrugation along the axis, which guarantees the absence of local parasitic traps for ions along the axis of their motion. Nevertheless, the value of the pseudopotential on the axis is still not zero, which means that a high-frequency electric field is formed, which oscillates the ions. Therefore, in such a design, there is also no relying on cooling of the ions (discharging their excess kinetic energy). In addition, the distribution of the pseudopotential function along the funnel axis slowly increases with increasing distance from zero in the z coordinate, which somewhat slows down the motion of ions and prevents their passage through the outlet, similar to the previous case.

In addition to the described features of the model, in the case of a high-frequency electric field of the form (13), there is a spatial gradient of its phase. But it plays a positive role, since the non-potential electric pseudoforce that arises in this case only additionally presses the ions against the axis of the device and provides a constant pulling force directed towards the outlet from the funnel. This factor could make it possible to do without an additional pulling static electric field, however, the pulling electric pseudoforce depends on the mass and, therefore, a new obstacle is possible, additional ion mass discrimination. A situation may arise for too large masses when the pulling of the pseudo-force is unable to overcome the braking of moving ions and an obstacle will prevent them from escaping the funnel.

Analysis of the properties of a conical funnel in pseudopotential mode with an Archimedean wave

Systems with a traveling pseudopotential wave are considered in [19–21]. In this case, the properties of an RF trap with conical electrodes are investigated, for which a high-frequency electric field forms a slowly traveling pseudopotential wave along the axis of the device. At the minima of the pseudopotential wave, the high-frequency electric field is zero, and it is at these points that local ion clusters are formed, which then move along the axis of the device to escape the funnel, simultaneously with the shift of the local minima of the pseudopotential wave. Importantly, the transport of ions is ensured by this, independent of their mass, since the speed with which the minima of the pseudopotential wave move is determined by the parameters of high-frequency voltages applied to the electrodes of the device (and nothing else).



For such an RF trap, the electric potential near the axis is described with good accuracy by the following formula:

$$V(z, r, t) = [V_C^*(z, r) \cos(2\pi t/T) + V_S^*(z, r) \sin(2\pi t/T)] \cos(\omega t + \varphi), \quad (14)$$

where T is the period of «slow» time that determines the speed of transport; the potentials $V_C^*(z, r)$ and $V_S^*(z, r)$ are given by Eqs. (14).

In this case, the graph also shows a corrugated gutter of the pseudopotential, and as the process progresses over time, the corrugation slowly moves along with the transporting pseudopotential wave, effectively pressing the ions against the axis of the device. In addition, a slow-moving pseudopotential wave appears on the axis of the system, forcibly transporting ions from the inlet to the outlet.

The traveling pseudopotential wave on the axis of the RF funnel is characterized by a variable maximum amplitude, which increases quadratically as it approaches the outlet from the funnel, but at the points of the minimum, the pseudopotential value is zero. As noted above, ions are trapped and their clusters are formed at the points of the minimum, and with the slow motion of the pseudopotential wave along the axis of the device, synchronized transport of ions is carried out, regardless of their mass. Since the high-frequency electric field in the centers of ion clusters is exactly zero, and the high-frequency electric field turns out to be very small for minor deviations of ions within the volume of the cluster, it is quite possible to count on at least partial cooling of ions during transport through the forevacuum region of the gas-dynamic interface.

The spatial phase gradient of the high-frequency electric field for the electric potential (15) is zero, therefore, there are no additional effects associated with the presence of a non-potential pseudoelectric force (see Eqs. (9)–(11)) in such a system.

Conclusion

Analytical models of the high-frequency electric field were constructed for radio-frequency ion funnels with circular apertures. They were used for qualitative analysis of RF ion funnels in different operating modes. We can conclude that the most promising configuration considered is the RF funnel with a conical transport channel and electric power supply, allowing to generate a pseudopotential Archimedean wave on the axis of the device.

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