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The influence of a non-uniform RF field on the ion trajectories in an omegatron II: harmonic peaks

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Abstract The quadrupole field component of a nonuniform RF field can cause harmonic mass peaks. This effect is mathematically analysed. It is shown theoretically and experimentally that these spurious peaks can be suppressed by a DC drift field.

Nomenclature

This list is additional to the nomenclature given in our previous article (Bijma and Hoenders 1971).

C_1, C_2	coefficients defined in (20) and (21) (m)
$f_1(t), f_2(t)$	functions defined in (12) and (28) (m)
$g_1(U, V, t), g_2(U, V, t)$	functions defined in (18) and (19) (m s ⁻¹)
$h_1(U, V, t), h_2(U, V, t)$	functions defined in (22) and (23) (m)
O''	origin coordinate system defined in (29)
U, V	coefficients defined in (10) (m)
U_u, V_u	zero order approximation of U and V , defined in (22) and (23) (m)

1 Introduction

This second publication is a continuation of a previous paper (Bijma and Hoenders 1971, to be referred to as I) describing one of the effects of a nonuniform RF field – the so-called RF drift-off. In that paper it was shown that the RF drift-off increases the resolving power without reducing the ion detection sensitivity. This paper describes the effect of the harmonic peaks. A harmonic peak is a spurious peak at the double cyclotron frequency, caused by the nonuniform RF field. In certain circumstances these harmonic peaks can be observed in all omegatrons with a nonuniform RF field, so we have studied this effect to some extent.

Experimental data are given among others by Reich (1961), Averina (1966) and Bijma *et al.* (1968). We give a review of the equations of motion as derived in the previous paper from the Newton-Lorentz equations, with the assumptions concerning the electromagnetic field of the so-called long omegatron. An approximate calculation shows an exponential growth of the ion trajectories at the double resonance frequency. For a

Figure 1 Influence of the quadrupole field component (a) $E_x = bx \sin 2\Omega t$ and (b) $E_x = bx \sin (2\Omega t + \pi)$ on an ion with initial velocity $\dot{x}(0)$. (c) Harmonic ion path for $E_x = bx \sin 2\Omega t$ and initial velocity $\dot{x}(0)$

cubical omegatron the analysis will be more complicated but the results will be similar. The trapping field for a cubical omegatron, however, is inferior to the combined drift and trapping field used in a long omegatron. Fortunately the harmonic peaks can be easily suppressed by a DC drift field.

2 Principle of operation

As a consequence of its initial velocity and the homogeneous magnetic field the ion will describe a circular path if no electric field is applied. The quadrupole component $bx \sin (2\Omega t + \phi)$ of the applied RF field will perturb this circular path in such a way that it is transformed into an exponential spiral. This can be explained as follows. Consider figure 1(a) where we have drawn the force acting on the ion caused by the quadrupole field term

$$E_x = bx \sin 2\Omega t \quad (\text{cf. I, equation (1)}).$$

The tangential part of this force in this case will accelerate the ion in the four quadrants. Therefore the radius of the path will increase with time. As can be seen from the quadrupole field term this force will be proportional to the x coordinate of the radius, so that an exponential increase of the radius can be expected in this case. We have drawn another typical case in figure 1(b) from which we deduce the tangential force to decelerate the ion in all four quadrants if

$$E_x = bx \sin (2\Omega t + \pi).$$

In either case, the initial velocity of the ion determines the initial path radius which has a strong influence on the path radius as a function of time. In figure 1(c) a calculated path is given.

3 Mathematical treatment

In figure 2 the RF and DC voltages applied to the omegatron are drawn. The electric field in the omegatron is generated by the RF voltage $V_{rf} \sin (\omega t + \phi)$, the DC drift voltage V_{dr} , and the DC trapping voltage V_{tr} .

In figure 1 of I it has been shown that V_{rf} causes a nonuniform RF field which can be approximated by

$$\begin{aligned} E_x &= (a + bx) \sin (\omega t + \phi) \\ E_y &= 0 \\ E_z &= -bz \sin (\omega t + \phi). \end{aligned} \quad (1)$$

The DC potential distributions caused by the drift potential and the trapping potential are drawn in figure 3. The DC field can be approximated by

$$\begin{aligned} E_x &= a' + b'x \\ E_y &= 0 \\ E_z &= -b'z. \end{aligned} \quad (2)$$

Figure 2 Sketch of the omegatron. The mass peaks of figures 5 and 6 are measured on collector C₂. The ion path is indicated by a solid spiral, the electron path by a dotted line

The magnetic field is assumed to be uniform and directed along the z axis; thus $\mathbf{B} = (0, 0, B)$.

The equations of motion for the ion are derived with the aid of the Newton-Lorentz relations

$$\frac{d}{dt}(m\mathbf{v}) = e(\mathbf{E} + \mathbf{v} \wedge \mathbf{B}) \quad (3)$$

thus

$$\ddot{x} + (\Omega^2 - \beta')x = (\alpha + \beta x) \sin(\omega t + \phi) + \Omega \dot{y}(0) + \alpha' \quad (4)$$

$$\dot{y} = -\Omega x + \dot{y}(0) \quad (5)$$

$$\ddot{z} = -\beta z \sin(\omega t + \phi) - \beta' z \quad (6)$$

with

$$\alpha' = \frac{ea'}{m}, \quad \beta' = \frac{eb'}{m}, \quad \alpha = \frac{ea}{m}, \quad \beta = \frac{eb}{m} \quad \text{and} \quad \Omega = \frac{eB}{m}. \quad (7)$$

The homogeneous part of (4):

$$\ddot{x} + \Omega^2 x = \beta x \sin(\omega t + \phi) \quad (8)$$

is known as the Mathieu equation. The z component of the ion motion, being a periodic movement, will not be considered.

We shall now try to find an approximate solution of equation

Figure 3 DC potential distribution in a long omegatron (a) of the drift potential V_{dr} , and (b) of the trapping potential V_{tr}

(4) for the case $\omega = 2\Omega$. For reasons of mathematical convenience, from now on we will assume $\beta' = 0$, because in this case the frequency shift will be small, thus

$$\ddot{x} + \Omega^2 x = (\alpha + \beta x) \sin(2\Omega t + \phi) + \Omega \dot{y}(0) + \alpha'. \quad (9)$$

This equation can be transformed, using a modified form of the method of variation of constants; consequently we assume the solution to be of the following form:

$$x(t) = U(t) \sin \Omega t + V(t) \cos \Omega t + f_1(t) + \frac{\dot{y}(0)}{\Omega}. \quad (10)$$

In this equation, the function $f_1(t)$ is the solution of equation (9) in the case where $\beta = 0$ and the initial conditions are given by:

$$f_1(0) = 0 \quad \text{and} \quad f_1'(0) = 0 \quad (11)$$

thus

$$f_1(t) = \frac{-\alpha}{3\Omega^2} \{ \sin(2\Omega t + \phi) - \sin \phi \cos \Omega t - 2 \sin \Omega t \cos \phi \} + \frac{\alpha'}{\Omega^2} (1 - \cos \Omega t). \quad (12)$$

The initial conditions of $U(t)$ and $V(t)$ are given by

$$U(0) = \frac{\dot{x}(0)}{\Omega} \quad \text{and} \quad V(0) = -\frac{\dot{y}(0)}{\Omega}. \quad (13)$$

The functions $x(t)$ being described with the aid of two independent functions $U(t)$ and $V(t)$, we may assume a suitably chosen relation between $U(t)$ and $V(t)$, thus

$$\dot{U} \sin \Omega t + \dot{V} \cos \Omega t = 0. \quad (14)$$

Substituting (10) in (9), condition (14) results in an equation containing first order derivatives only, thus

$$\Omega \dot{U} \cos \Omega t - \Omega \dot{V} \sin \Omega t = \beta x \sin(2\Omega t + \phi). \quad (15)$$

With the aid of Cramer's rule we can solve $\dot{U}$ and $\dot{V}$ from equations (14) and (15), thus

$$\dot{U} = \frac{\beta x}{\Omega} \sin(2\Omega t + \phi) \cos \Omega t \quad (16)$$

$$\dot{V} = -\frac{\beta x}{\Omega} \sin(2\Omega t + \phi) \sin \Omega t. \quad (17)$$

Substituting $x(t)$ from (10) in (16) and (17) gives

$$\dot{U} = \frac{\beta}{4\Omega} (U \cos \phi + V \sin \phi - C_1) + g_1(U, V, t) \quad (18)$$

$$\dot{V} = \frac{\beta}{4\Omega} (U \sin \phi - V \cos \phi + C_2) + g_2(U, V, t) \quad (19)$$

with

$$C_1 = \frac{\alpha}{3\Omega^2} (1 + \cos^2 \phi) + \frac{\alpha'}{\Omega^2} \sin \phi \quad (20)$$

$$C_2 = \frac{-\alpha}{3\Omega^2} \sin \phi \cos \phi + \frac{\alpha'}{\Omega^2} \cos \phi. \quad (21)$$

The functions $g_1(U, V, t)$, $g_2(U, V, t)$ are given in Appendix 1.

In deriving (18) and (19) we have expressed the goniometric product terms as sums of goniometric functions. The functions g_1 and g_2 consist of those terms containing any of the following factors $\sin \Omega t$, $\cos \Omega t$, $\sin 2\Omega t$, ... With the aid of the Laplace transformation (see Appendix 2) we obtain a set of so-called coupled Volterra integral equations:

$$U(t) = U_u(t) + h_1(U, V, t) \quad (22)$$

$$V(t) = V_u(t) + h_2(U, V, t) \quad (23)$$

with

$$U_u(t) = \frac{1}{2} \{ [U(0) + C_1] (1 + \cos \phi) + [V(0) - C_2] \sin \phi \} \\ \times \exp(\beta t/4\Omega) + \frac{1}{2} \{ [U(0) - C_1] (1 - \cos \phi) \\ + [-V(0) - C_2] \sin \phi \} \exp(-\beta t/4\Omega) \\ + C_2 \sin \phi - C_1 \cos \phi \quad (24)$$

$$V_u(t) = \frac{1}{2} \{ [V(0) - C_2] (1 - \cos \phi) + [U(0) + C_1] \sin \phi \} \\ \times \exp(\beta t/4\Omega) + \frac{1}{2} \{ [V(0) + C_2] (1 + \cos \phi) \\ + [-U(0) + C_1] \sin \phi \} \exp(-\beta t/4\Omega) \\ - C_2 \cos \phi - C_1 \sin \phi. \quad (25)$$

$U_u(t)$ and $V_u(t)$ are the solutions of equations (18) and (19) in cases where $g_1(U, V, t)$ and $g_2(U, V, t)$ both equal zero. $h_1(U, V, t)$ and $h_2(U, V, t)$ are defined in Appendix 2 and are convolution integrals containing the functions g_1 and g_2 . As is shown in Appendix 2 these terms cause only slight perturbations with respect to the functions $U_u(t)$ and $V_u(t)$ and will therefore be neglected.

Substituting the approximate solutions $U_u(t)$ and $V_u(t)$ in (10) we get

$$x(t) - \frac{\dot{x}(0)}{\Omega} - f_1(t) = U_u(t) \sin \Omega t + V_u(t) \cos \Omega t. \quad (26)$$

Equation (26) gives the approximate solution for the movement of the ion in the x direction. We also have to consider the movement in the y direction which can be obtained from (26) and (5); thus

$$y(t) + \frac{\dot{x}(0)}{\Omega} - f_2(t) = U_u(t) \cos \Omega t - V_u(t) \sin \Omega t \quad (27)$$

with

$$f_2(t) = \frac{\alpha}{3\Omega^2} \{ \cos \phi + \frac{1}{2} \cos(2\Omega t + \phi) + \sin \Omega t \sin \phi \\ - 2 \cos \Omega t \cos \phi \} - \frac{\alpha'}{\Omega} t + \frac{\alpha'}{\Omega^2} \sin \Omega t \quad (28)$$

Integrating (26) it is assumed that the functions $U_u(t)$ and $V_u(t)$ can be placed before the integral sign (see Appendix 3).

We now calculate the radius of the ion path from (26) and (27) with respect to a new moving origin given by:

$$O'' \left(\frac{\dot{x}(0)}{\Omega} + f_1(t), -\frac{\dot{x}(0)}{\Omega} + f_2(t) \right). \quad (29)$$

If we consider the asymptotic behaviour of the origin O'' , and neglect the oscillating terms which are small with respect to the dimensions of the omegatron, we get the following approximation for the origin:

$$O''(0, -\alpha' t/\Omega). \quad (30)$$

The radius of the ion path with respect to this origin is thus given by

$$r^2 = x^2 + \left(y + \frac{\alpha' t}{\Omega} \right)^2 = U_u^2 + V_u^2. \quad (31)$$

Using formulae (13), (20), (21), (22), (23), (24), (25) in (31) we get:

$$r^2 = \left[\left\{ \frac{\dot{x}(0)}{\Omega} \cos \frac{1}{2}\phi - \frac{\dot{y}(0)}{\Omega} \sin \frac{1}{2}\phi + \frac{2\alpha}{3\Omega^2} \cos^3 \frac{1}{2}\phi + \frac{\alpha'}{\Omega^2} \sin \frac{1}{2}\phi \right\} \right. \\ \times \exp(\beta t/4\Omega) - \frac{2\alpha}{3\Omega^2} \cos^3 \frac{1}{2}\phi - \frac{\alpha'}{\Omega^2} \sin \frac{1}{2}\phi \left. \right]^2 \\ + \left[\left\{ \frac{\dot{x}(0)}{\Omega} \sin \frac{1}{2}\phi + \frac{\dot{y}(0)}{\Omega} \cos \frac{1}{2}\phi - \frac{2\alpha}{3\Omega^2} \sin^3 \frac{1}{2}\phi - \frac{\alpha'}{\Omega^2} \cos \frac{1}{2}\phi \right\} \right. \\ \times \exp(-\beta t/4\Omega) + \frac{2\alpha}{3\Omega^2} \sin^3 \frac{1}{2}\phi + \frac{\alpha'}{\Omega^2} \cos \frac{1}{2}\phi \left. \right]^2. \quad (32)$$

For a further analysis we consider the asymptotic behaviour of the path radius r , because we are only interested in those ions which reach the collector, thus

$$r \approx \left| \frac{\dot{x}(0)}{\Omega} \cos \frac{1}{2}\phi - \frac{\dot{y}(0)}{\Omega} \sin \frac{1}{2}\phi + \frac{2\alpha}{3\Omega^2} \cos^3 \frac{1}{2}\phi + \frac{\alpha'}{\Omega^2} \sin \frac{1}{2}\phi \right| \\ \times \exp(\beta t/4\Omega). \quad (33)$$

It is assumed that the velocity distribution is rotational symmetric, therefore we introduce cylindrical coordinates:

$$\dot{x}(0) = \dot{r}(0) \cos \zeta \quad (34)$$

$$\dot{y}(0) = \dot{r}(0) \sin \zeta$$

$$r \approx \left| \frac{\dot{r}(0)}{\Omega} \cos(\zeta + \frac{1}{2}\phi) + \frac{2\alpha}{3\Omega^2} \cos^3 \frac{1}{2}\phi + \frac{\alpha'}{\Omega^2} \sin \frac{1}{2}\phi \right| \exp(\beta t/4\Omega) \quad (35)$$

$$r \approx \left| \frac{\dot{r}(0)}{\Omega} \cos(\zeta + \frac{1}{2}\phi) + \frac{2a}{3B\Omega} \cos^3 \frac{1}{2}\phi + \frac{a'}{B\Omega} \sin \frac{1}{2}\phi \right| \exp(bt/4B). \quad (36)$$

4 Suppression of the harmonic peaks

A resonant ion will reach the collector in a detection time t_2 when the path radius r equals the collector distance d for $t = t_2$, thus

$$r = \frac{a}{2B} t_2 = d \quad (\text{cf. I, equation (9) with } \Delta\omega \rightarrow 0). \quad (37)$$

We will now investigate whether harmonic ions could reach the collector within this time t_2 . This is of interest because we have chosen our drift-off field so that ions with a detection time $t > t_2$ will drift out of the collection range, i.e. not reach the collector. This point is further discussed below. With (36) we find that these ions must satisfy the following inequality:

$$r \approx \left| \frac{\dot{r}(0)}{\Omega} \cos(\zeta + \frac{1}{2}\phi) + \frac{2a}{3B\Omega} \cos^3 \frac{1}{2}\phi + \frac{a'}{B\Omega} \sin \frac{1}{2}\phi \right| \\ \times \exp(bt_2/4B) > d \quad (38)$$

or with the aid of (37)

$$\left| \frac{\dot{r}(0)}{\Omega} \cos(\zeta + \frac{1}{2}\phi) + \frac{2a}{3B} \cos^3 \frac{1}{2}\phi + \frac{a'}{B} \sin \frac{1}{2}\phi \right| \\ > d\Omega \exp(-bd/2a). \quad (39)$$

Instead of equation (39) which contains the initial velocity, we can calculate a relation which contains the initial energy expressed in electron volts:

$$\frac{m}{2e} \left\{ \frac{\dot{r}(0)}{\Omega} \cos(\zeta + \frac{1}{2}\phi) + \frac{2a}{3B} \cos^3 \frac{1}{2}\phi + \frac{a'}{B} \sin \frac{1}{2}\phi \right\}^2 \\ > \frac{m}{2e} d^2 \Omega^2 \exp(-bd/a) \text{ [eV]} \quad (40)$$

As an example we substitute for $\dot{r}(0)$ in (40) the average initial velocity in the $(\dot{r}, \zeta)$ plane, equation (A13) of I, where we assumed that the energy distribution is a Maxwell distribution. We further substitute the other numerical values from Appendix 4 of I. This leads to:

$$5 \times 10^{-9} M \left(\frac{1950}{M^{1/2}} \cos(\zeta + \frac{1}{2}\phi) + 120 \cos^3 \frac{1}{2}\phi + 80 \sin \frac{1}{2}\phi \right)^2 \\ > \frac{300}{M} \text{ [eV]}. \quad (41)$$

The energy expressed in the left-hand side is of the order of the thermal energy, i.e. $\approx 3 \times 10^{-2}$ eV. So no thermal ion will reach the collector within the detection time $t = t_2$.

Another group of ions to be considered are the ions which are created by dissociative ionization of molecules by electron impact (Rapp *et al.* 1965). The energy of these ions is up to about 10 eV and has an anisotropic distribution depending both on the kind of molecules and the electron energy. To reach the collector within a time t_2 the ions must have an initial energy greater than $300/M$ eV in the (r, ζ) plane. But at the same time the energy component in the z direction must be less than or approximately equal to 0.2 eV because otherwise the trapping field is too small and these ions will be neutralized on the z side electrodes.

The chance that both conditions are fulfilled is very small, so hardly any dissociated ion will reach the collector within the detection time $t = t_2$.

5 Time of flight distribution for thermal harmonic ions

Since in many omegatrons a smaller DC drift field is used than assumed in §4 it is illustrative to calculate which of the ions will reach the collector within a given time. If we assume that the initial velocity distribution is a Maxwellian one, the calculation can be performed, but the terms $(2a/3B\Omega) \cos^3 \frac{1}{2}\phi$ and $(a'/B\Omega) \sin \frac{1}{2}\phi$ give rise to a very complex mathematical calculation.

As can be seen from (41) the average value of the term $(\dot{r}(0)/\Omega) \cos(\zeta + \frac{1}{2}\phi)$ is dominant. Therefore, neglecting the terms $(2a/3B\Omega) \cos^3 \frac{1}{2}\phi$ and $(a'/B\Omega) \sin \frac{1}{2}\phi$ we get the following approximate expression for the ion path radius:

$$r \approx \left| \frac{\dot{r}(0)}{\Omega} \cos(\zeta + \frac{1}{2}\phi) \right| \exp(bt/4B). \quad (42)$$

We now calculate the fraction $\Delta N/N$ of the thermionic ions created at $t=0$ reaching the collector within an arbitrary time t_3 . For this fraction the following inequality has to be valid:

$$\left| \frac{\dot{r}(0)}{\Omega} \cos(\zeta + \frac{1}{2}\phi) \right| \exp(bt_3/4B) > d. \quad (43)$$

The Maxwell velocity distribution in the $(\dot{r}, \zeta)$ plane is given by

$$f(\dot{r}) d\dot{r} d\zeta = \frac{m}{2\pi kT} \exp\left(\frac{-m\dot{r}^2}{2kT}\right) \dot{r} d\dot{r} d\zeta$$

(cf. I, equation (A12))

$$\dot{r} = \dot{r}(0).$$

Integration of (I, A12) between the boundaries of integration determined by the inequality (43) yields the fraction $\Delta N/N$ reaching the collector within the time t_3 . This leads to the following integral:

$$\frac{\Delta N}{N} = \frac{m}{2\pi kT} \int_{\Omega d \exp(bt_3/4B)}^{\infty} \exp\left(\frac{-m\dot{r}^2}{2kT}\right) \dot{r} d\dot{r} \\ + \arccos\left(\frac{d\Omega/\dot{r}}{\exp(-bt_3/4B) - \phi/2}\right) \times 2 \int d\zeta \\ - \arccos\left(\frac{d\Omega/\dot{r}}{\exp(-bt_3/4B) - \phi/2}\right) \quad (44)$$

The integral is calculated in Appendix 4, yielding

$$\frac{\Delta N}{N} = 1 - \phi \left\{ \Omega d \left(\frac{m}{2kT} \right)^{1/2} \exp\left(-\frac{bt_3}{4B}\right) \right\}. \quad (45)$$

Figure 4 Fraction $\Delta N/N$ of the resonant ions, indicated by a broken line, and of the harmonic ions, indicated by a solid line, that reach the collector within a time $t_3 = 4Bt_3^*/b$, as a function of t_3^* . $\Delta N/N = 1 - \phi\{\Omega d(M/2RT)^{1/2} \exp(-bt_3/4B)\}$; $T = 300$ K; $d = 10^{-2}$ m; $b/a = 10^2$ m $^{-1}$

In this expression $\phi(x)$ is the well known error function defined by

$$\phi(x) = \frac{2}{\pi^{1/2}} \int_0^x \exp(-t^2) dt. \quad (46)$$

In figure 4 this function is plotted for different masses. Introducing the argument of the exponential factor in equation (45) as a new variable, we get

$$t_3^* = \frac{bt_3}{4B}. \quad (47)$$

This is the abscissa in figure 4.

According to equation (37) a resonant ion will reach the collector after a time

$$t_2 = \frac{2Bd}{a} \quad (48)$$

thus

$$t_3^* = \frac{bd}{2a}. \quad (49)$$

With the numerical values of I, Appendix 4, valid for the omega-tron drawn in figure 2, we calculate that

$$t_3^* = 0.5 \quad (50)$$

for a resonant ion.

As can be seen from figure 4 a harmonic ion will be dependent on its mass, and reach the collector after a time $t_3^* \geq 2$. Hence the time of flight of a harmonic ion is more than four times as long as that for a resonant one.

6 Experimental verification and conclusions

The theoretical analysis of the RF drift-off and the harmonic peaks given in these two papers is not restricted to the shape of the long omegatron considered, but can be applied to any other omegatron for which the linear approximation of the electric field holds. Starting from the deduced theory of the nonuniform RF field, one can try to design an omegatron that approximates the theoretically assumed field, for instance by taking hyperbolic electrodes. A detailed experimental verification of both the RF drift-off and the harmonic effects on some of these omegatrons will be given in a future paper.

However, for the sake of clarity, some of the measurements obtained with the omegatron in figure 2 will be given below.

Figure 5 Mass peaks of N_2^+ and N_2^{++} measured at different values of the drift potential V_{dr} . The peak height of N_2^{++} without harmonic peak is about 7% of the peak height of N_2^+ . This value corresponds with that of Klopfer and Schmidt (1960). For $V_{dr} \approx 0$ a harmonic peak of N_2^+ is superimposed on the N_2^{++} peak. See figure 6. $V_{rf} = 3$ V, $V_{tr} = 0.4$ V, $P = 50$ nTorr, $i = 5$ μ A

Using N_2 as a test gas, figure 5 gives the mass peaks of N_2^+ and N_2^{++} as a function of both the frequency ν and the DC drift potential V_{dr} . The harmonic peak of N_2^+ is superimposed on the N_2^{++} peak because $\Omega(N_2^{++}) = 2\Omega(N_2^+)$. As we have seen in the deduced theory, the time of flight is much greater for a harmonic ion than for a resonant one. Hence an increasing drift field will gradually suppress the harmonic peaks. For this reason the harmonic ions will be collected only if V_{dr} is about zero. This can also be seen from figure 6 where the peak heights from N_2^+ and N_2^{++} are recorded as functions

Figure 6 Peak heights of the N_2^+ and N_2^{++} peaks as function of the drift potential V_{dr} . The superimposed harmonic peak of N_2^+ is indicated at N_2^{++}

of the DC drift potential V_{dr} . It is possible to choose the working conditions in such a way that no harmonic peaks are detected. Harmonics caused by quadratic and higher order field terms in the RF field are almost impossible because of the very high initial ion velocity needed. In contradiction to the conclusions of Schuchhardt (1960) it is impossible that the higher order DC field terms can cause harmonics.

Appendix 1 The functions $g_1(U, V, t)$ and $g_2(U, V, t)$

The expressions for the functions $g_1(U, V, t)$ and $g_2(U, V, t)$ appearing in (18), (19) can be given explicitly, thus:

$$\begin{aligned} g_1(U, V, t) = & \frac{\alpha\beta}{6\Omega^3} \cos \Omega t + \frac{\beta\dot{y}(0)}{2\Omega^2} \sin(\Omega t + \phi) + \frac{\beta V}{2\Omega} \sin(2\Omega t + \phi) \\ & - \frac{\alpha\beta}{6\Omega^2} \sin \phi \sin(2\Omega t + \phi) - \frac{\alpha\beta}{12\Omega^3} \cos(3\Omega t + 2\phi) \\ & + \frac{\beta\dot{y}(0)}{2\Omega^2} \sin(3\Omega t + \phi) - \frac{\beta U}{4\Omega} \cos(4\Omega t + \phi) \\ & + \frac{\beta V}{4\Omega} \sin(4\Omega t + \phi) - \frac{\alpha\beta}{12\Omega^3} \sin \phi \sin(4\Omega t + \phi) \\ & + \frac{\alpha\beta}{6\Omega^3} \cos \phi \cos(4\Omega t + \phi) - \frac{\alpha\beta}{12\Omega^3} \\ & \times \cos(5\Omega t + 2\phi) + \frac{\beta}{\Omega} \left[+ \frac{\alpha'}{2\Omega^2} \sin(\Omega t + \phi) \right. \\ & - \frac{\alpha'}{2\Omega^2} \sin(2\Omega t + \phi) + \frac{\alpha'}{2\Omega^2} \sin(3\Omega t + \phi) \\ & \left. - \frac{\alpha'}{4\Omega^2} \sin(4\Omega t + \phi) \right] \end{aligned} \quad (A1)$$

$$\begin{aligned} g_2(U, V, t) = & -\frac{\alpha\beta}{6\Omega^3} \sin \Omega t - \frac{\beta\dot{y}(0)}{2\Omega^2} \cos(\Omega t + \phi) - \frac{\beta U}{2\Omega} \\ & \times \sin(2\Omega t + \phi) + \frac{\alpha\beta}{3\Omega^3} \cos \phi \sin(2\Omega t + \phi) \\ & - \frac{\alpha\beta}{12\Omega^3} \sin(3\Omega t + 2\phi) + \frac{\beta\dot{y}(0)}{2\Omega^2} \cos(3\Omega t + \phi) \\ & + \frac{\beta U}{4\Omega} \sin(4\Omega t + \phi) + \frac{\beta V}{4\Omega} \cos(4\Omega t + \phi) \\ & - \frac{\alpha\beta}{12\Omega^3} \sin \phi \sin(4\Omega t + \phi) - \frac{\alpha\beta}{6\Omega^3} \\ & \times \cos \phi \sin(4\Omega t + \phi) + \frac{\alpha\beta}{12\Omega^3} \sin(5\Omega t + 2\phi) \\ & + \frac{\beta}{\Omega} \left[-\frac{\alpha'}{2\Omega^2} \cos(\Omega t + \phi) + \frac{\alpha'}{2\Omega^2} \cos(3\Omega t + \phi) \right. \\ & \left. - \frac{\alpha'}{4\Omega^2} \cos(4\Omega t + \phi) \right]. \end{aligned} \quad (A2)$$

Appendix 2 Derivation of the equations (22) and (23)

The set of differential equations (18), (19)

$$\dot{U} = \eta U \cos \phi + \eta V \sin \phi - \eta C_1 + g_1 \quad (18)$$

$$\dot{V} = \eta U \sin \phi - \eta V \cos \phi + \eta C_2 + g_2 \quad (19)$$

with

$$\eta = \frac{\beta}{4\Omega} \quad (A3)$$

can be transformed with the aid of the Laplace transformation, yielding

$$U(s) (\eta \sin \phi) + V(s) (-\eta \cos \phi - s) = -V(0) - \frac{\eta C_2}{s} - g_2(s) \quad (A4)$$

$$U(s) (\eta \cos \phi - s) + V(s) (\eta \sin \phi) = -U(0) + \frac{\eta C_1}{s} - g_1(s). \quad (A5)$$

From these equations we solve for $U(s)$ and $V(s)$ with the aid of Cramer's rule:

$$U(s) = \{[-V(0) - \eta C_2/s - g_2(s)] \eta \sin \phi - (\eta \cos \phi + s) \{U(0) - \eta C_1/s + g_1(s)\} / (\eta^2 - s^2)\} \quad (\text{A6})$$

$$V(s) = \{[-U(0) + \eta C_1/s - g_1(s)] \eta \sin \phi + \{V(0) + \eta C_2/s + g_2(s)\} (\eta \cos \phi - s) / (\eta^2 - s^2)\} \quad (\text{A7})$$

The inverse transformation of (A6) and (A7) leads to (22) and (23) with $h_1(U, V, t)$ and $h_2(U, V, t)$ given by (A8) and (A9), thus

$$h_1(U, V, t) = \int_0^t \sinh \eta(t-\tau) \{g_1(\tau) \sin \phi + g_2(\tau) \cos \phi\} d\tau + \int_0^t \cosh \eta(t-\tau) g_2(\tau) d\tau \quad (\text{A8})$$

$$h_2(U, V, t) = \int_0^t \sinh \eta(t-\tau) \{g_2(\tau) \sin \phi - g_1(\tau) \cos \phi\} d\tau + \int_0^t \cosh \eta(t-\tau) g_1(\tau) d\tau \quad (\text{A9})$$

The unique solution of the set of coupled Volterra integral equations can be given with the aid of the Neumann series. These series are obtained by repeated iteration of the equations (22) and (23). The first term $h_1(U_u, V_u, t)$, $h_2(U_u, V_u, t)$ consists of oscillating terms with an amplitude of which the magnitude differs by an order $\beta/4\Omega^2$ from U_u and V_u .

Appendix 3 On the approximate integration of equation (26)
Integrating (26) we have to calculate integrals of the form

$$\int \exp(\beta t/4\Omega) \sin(\Omega t + \phi) dt \quad (\text{A10})$$

If $\beta/4\Omega^2 \ll 1$ the following approximation is valid

$$\begin{aligned} \int \exp(\beta t/4\Omega) \sin(\Omega t + \phi) dt \\ = \frac{\exp(\beta t/4\Omega)}{(\beta/4\Omega)^2 + \Omega^2} \left\{ \frac{\beta}{4\Omega} \sin(\Omega t + \phi) - \Omega \cos(\Omega t + \phi) \right\} \\ \simeq \exp(\beta t/4\Omega) \int \sin(\Omega t + \phi) dt. \end{aligned} \quad (\text{A11})$$

Appendix 4 Calculation of the integral (44)

With the aid of the substitutions

$$C = \frac{m}{2\pi kT}, \quad a = \frac{m}{2kT} \quad \text{and} \quad u^{1/2} = \Omega d \exp\left(-\frac{bt_3}{4B}\right) \quad (\text{A12})$$

the integral (44) can be written as

$$2C \int_{u^{1/2}}^{\infty} \exp(-a\rho^2) \rho d\rho \int_{-\arccos u^{1/2}/\rho - \phi/2}^{+\arccos u^{1/2}/\rho - \phi/2} d\zeta \quad (\text{A13})$$

Performing the integration over ζ and partial integration leads to

$$\frac{-2C}{a} \exp(-a\rho^2) \arccos \frac{u^{1/2}}{\rho} \Big|_{u^{1/2}}^{\infty} + \frac{2C}{a} \int_{u^{1/2}}^{\infty} \frac{\exp(-a\rho^2) u^{1/2} d\rho}{\rho^2 (1 - u/\rho^2)^{1/2}} \quad (\text{A14})$$

In this expression the first two terms are zero. Substituting $x = \rho^2$ in equation (A14) yields

$$\frac{Cu^{1/2}}{a} \int_u^{\infty} \frac{\exp(-ax) dx}{x(x-u)^{1/2}} \quad (\text{A15})$$

According to Gradshteyn and Ryzhik (1965, 3.363.2)

$$\int_u^{\infty} \frac{\exp(-ax) dx}{x(x-u)^{1/2}} = \frac{\pi}{u^{1/2}} [1 - \phi\{(u/a)^{1/2}\}] \quad (\text{A16})$$

where $\phi(x)$ is the well known error function

$$\phi(x) = \frac{2}{\pi^{1/2}} \int_0^x \exp(-t^2) dt \quad (\text{A17})$$

(This function is, for instance, tabulated in: Jahnke *et al.* 1966, p 31.)

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