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The Electric Mass Filter

W. PAUL M. RAETHER



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TRANSLATION OF

THE ELECTRIC MASS FILTER

("Das elektrische Massenfilter")

by

W. Paul and M. Raether

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THE ELECTRIC MASS FILTER

by W. Paul and M. Raether

In a previous treatise, Paul and Steinwedel [1] proposed a new, entirely electrically operated mass filter promising to have advantages over the present methods for determining the specific charge e/m of ions. In the meantime, the new method has been tested experimentally and the assumptions have been confirmed.

A. Theoretical Section

The mass spectrographs or electromagnetic isotope separators utilized in the past operate on the following principle:

The e/m of the ions is determined by measuring two of the three quantities impulse, energy and velocity of ions, viz., by eliminating velocity. Usually a spatial or temporal separation of ions of different mass takes place. The impulse is measured by deflection of the ions in a magnetic field; the energy is measured by deflection or acceleration of the ions in a static electric field. The velocity is derived from the elapsed time the ions require to cover a prescribed straight or curved path. Highfrequency equipment is used for measurement. However, in this process only the ions which start during a certain brief interval (impulse operation) or which satisfy certain phase relationships are measured.

In the present method, none of the aforementioned quantities is used to measure e/m; instead, the acceleration of an ion (dependent on time) in a high-frequency electric field is measured and used for segregating the masses. The spatial distribution of the electric field is characterized by the need for its potential V (x, y, z, t) to be a purely quadratic function of the coordinates x, y, z.

Disregarding revolutions and displacements of the coordinate system, the most general potential of this kind is represented by :

Here f(t) is any periodic function of time; on account of the Laplace equation $\Delta V = 0$, the constants a, β and γ of the relationship $a + \beta = \gamma$ have to suffice.

If ions are introduced into a field of this kind, their equations of motion lead to differential equations with periodic coefficients (periodic elasticity), which are distinguished by ranges of stable and unstable solutions. Hence, there are two different types of ion paths; either the ions execute, around the center of symmetry of the field, oscillations whose amplitudes do not exceed a certain maximum value (stable paths), or the amplitudes increase unusually rapidly so that the particles migrate in a very short time to the field-generating electrodes (unstable paths), and hence are eliminated. With a given field and a time dependence f(t), the specific charge of the ion alone will determine whether it will describe a stable or unstable path. In particular, the stability or instability of the paths is quite independent of the starting point and the direction or magnitude of the initial velocity of the ions, resulting in a high degree of non-dependence on the properties of the ion source. If e/m of an ion is in a stable range, any possible path is stable; if it is in an unstable range, any possible path is unstable. The theory is largely in keeping with that of the focusing process with alternating field gradients in the case of the synchrotron, in which, however, instabilities of the path of the very kind exploited here must be avoided.

Figure 1. Quadrupolar field symmetrical to the x-axis with hyperbolic electrodes.

(1)

A Specific Case

For the following investigation, a layout was chosen in which a = 0 in eq. (1); i.e., the potential of the field used has the form

$$V(y, z, t) = c f(t) (y^2 - z^2).$$

It is implemented by a quadrupolar field symmetrical to the x-axis with hyperbolic electrodes (fig. 1). If the distance from the vertex of the hyperbola to the x-axis is called r_0 , and if the voltage U is located at the electrodes alternately,

$$V = \frac{U(t)}{r_0^2} (y^2 - z^2)$$
 (2)

and the field strength

$$E_v = \pm \frac{2U}{r_0^2} y_t^{u} = E_s = -\frac{2U}{r_0^2} z_s$$

If U is ac voltage of the form $U = U_0 \cos \omega t$, the equations of motion of an ion are

$$m\ddot{x} = 0$$

$$m\ddot{y} - \frac{2eU_0}{r_0^3} \cdot y \cdot \cos\omega t = 0$$

$$m\ddot{z} + \frac{2eU_0}{r_0^2} \cdot z \cdot \cos\omega t = 0.$$
(3)

The first equation yields $\dot{x} = \text{const}$; i.e., the ion migrates in the x-direction at constant velocity. The other two equations are special cases of Mathieu's differential equation, which in its general form reads [2]:

$$\frac{d^{2}x}{d\zeta^{2}} + (a - 2q\cos 2\zeta)x = 0.$$
 (4)



Figure 2. Stability diagram of the Mathieu functions. Ions whose a and q values are within the shaded range describe stable paths.

The general solution of this equation can be represented by:

 $z = A \cdot e^{\mu} : \sum_{n=1}^{+\infty} c_n e^{inc} + B e^{-\mu} : \sum_{n=\infty}^{+\infty} c_n e^{-inc}.$

It will be perceived from this that there are two types of solutions. If the characteristic exponent μ , which can be calculated from a and q, is imaginary, the solution for all ζ remains limited. However, if μ is real or complex, the amplitude grows exponentially and the path is unstable. The stability or instability of a solution depends only on the size of the parameters a and q. It is independent of the initial values z_0 and \dot{z}_0 , as already mentioned in the introduction. If the stable ranges in the a, q plane are plotted, the result is fig. 2. The stable ranges are shown by shading. A comparison of eqs. (3) and (4) reveals that in our example

$$a = 0$$
 and $q = \frac{4e U_0}{r_0^* m \omega^*}$, (5)

 $\zeta = \frac{\omega t}{2}$. Accordingly, the ions may be located at any point on the q-axis of fig. 2. In fig. 2a, the vicinity of the q-axis is diagrammed. For the sake of clarity, the upper ranges are represented with exaggerated width. From $\mathbf{q} = 0$ to q = 0.92, the ion is located in a stable range, from q = 0.92 to q = 7.50 in an unstable one, from q = 7.50 to q = 7.52 in a stable one again, etc.

According to eq. (5), the range in which the ion is located depends only on e/m when the ac voltage and the frequency are given. For an alternating current of 1000 v, the first two stable ranges in an M, ν diagram are shown by the shaded area in fig. 3 (M = mass number defined by $\frac{e}{m} = \frac{e}{m_0 M}$, ν = frequency). From this, the following is seen: If an ion beam contains different

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Figure 2. Points of intersection of the stable ranges with the q-axis.

masses, in the first range, with given ac voltage and frequency, only masses greater than m = $\frac{4eU_0}{0.92 \times r_0^2 \omega^2}$ are allowed to bass

through the field. In the second range, only a finite mass interval

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 $\Delta m = \frac{\Delta q_m}{q}$ is passed. Since $\Delta q/q$ for the second range is $\approx 1\%$, it should be expected that isotopes can still be separated when the



Figure 3. Transmissivity ranges of the mass filter with an alternating current $U_0 = 1000v$, $r_0 = 0.5$ cm. Ions whose mass number is located in the shaded area can pass through the filter. Massenzahl = mass number; Bereich = range; Frequenz = frequency; MHz = megacycles.

masses are smaller. A disturbing effect in this conjunction is that a mass continuum in the first range is superposed on the second range. From the third range on, $\Delta q/q$ becomes very small, so that a very good resolution should be expected, but superposition occurs even in these higher ranges. However, these higher ranges can hardly be used owing to their intensity. As the eqs. (12) show, the initial energies of the ions in the y and z-directions, respectively, in the third range are not likely to exceed the sum of 10^{-6} ev.

If a dc voltage is superimposed on the ac voltage, hence selecting a $U = U_g + U_0 \cos \omega t$, a small mass interval ΔM can be transmitted in the first range as well. The equations of motion now read:

Comparing eq. (6) with eq. (4), we get:

$$a_y = -\frac{8e\,U_R}{r_R^2 m\,\omega^2}; \quad a_z = +\frac{8e\,U_R}{r_R^2 m\,\omega^2}$$
 (7)

while $q = \frac{4eU_0}{r_0^2 m\omega^2}$ obtains in both directions of motion. The relationship $\frac{a}{q} = \pm 2\frac{U}{U_0^g}$ depends only on dc and ac voltage, not on frequency and ion mass. In the a, q diagram, the curves $u = \frac{u}{U_0^g} = \frac{a}{2q} = \text{const are straight lines which start at the origin}$ and are symmetrical to the q-axis.



Figure 4. The first range of stability. Enlarged section taken from fig. 2. z-Richtung = z-direction; stabiler Bereich = stable range; y-Richtung = y-direction; q_{grenz} = q_{boundary}.

A section taken from the first range of stability and a straight-line couple u = const are plotted in fig. 4. The half-plane a < 0 corresponds to the y-direction and the half-plane a > 0, to the z-direction [cf. (7)].

If the amplitude and frequency of the ac voltage is regarded as constant, all masses are located on the straight line u = const. The value q_1 is referred to mass m_1 , $q_2 \triangleq m_2$ and $q_3 \triangleq m_3$. Only the masses m_1 to m_3 are included in a stable range in the y-direction, and only the masses greater than m_2 in the z-direction. Since an ion can pass through the field only if it is stable both in the y-direction and in the z-direction, in all a mass interval of only $\Delta m = m_1 - m_2$ will be transmitted. The width of the transmissivity range as a function of the dc voltage is obtained from the

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intersections of the straight line u = const with the boundary curves of the first range of stability. When the dc voltage is increased, the angle between the straight lines and the q-axis becomes greater and the transmissivity range narrower. With a relationship $U_g/U_0 = 0.166 = u_{max}$, the result in the boundary case is an infinitely narrow range. Above u_{max} , no ion can pass through the field. In this case, q assumes the value $q_{boundary} = 0.706$. The boundary curves are represented by the equations

$$a = -\frac{q^{4}}{2} + \frac{7}{128}q^{4} + 0(q^{4})$$
(8)

for the lower boundary curve, and

$$a = 1 - q - \frac{q^2}{8} + \frac{q^3}{64} - \frac{1}{1536} q^4 + O(q^8)$$

for the upper boundary curve. In the vicinity of the point $q = q_{boundary}$, the result is

$$\Delta q = (0,236 - 1,410 \,\mu) \left(\frac{1}{1,176 + 2 \,\mu} + \frac{1}{0,629 - 2 \,\mu} \right)$$

or, by way of approximation for a very narrow range ($\Delta q/q < 5\%$),

$$\Delta q \approx 4 (0,236 - 1,410 \mu)$$

With constant field values, this corresponds to a mass range

$$\Delta m = 4 \, \frac{m}{a} \, (0,236 - 1,410 \, u) \, .$$

The theoretical mass resolving power can be calculated from this as a function of the quantity u; it is

$$\frac{m}{1m} = \frac{1}{2} \frac{\nu}{A\nu} = \frac{0.75}{1 - u/u_{\text{max}}}.$$
 (9)

The frequency band width for a mass evolves as

$$21v = \frac{v}{v_{s,5}} \left(1 - \frac{u}{u_{m}}\right)$$
(10)

In calculating the transmissivity range, it is understood that the field is infinitely long in the x-direction, since the amplitudes of the unstable ion paths tend toward ∞ when $t \rightarrow \infty$. In practice, it is sufficient to require that the ions undergo so many changes of field that the amplitudes of the unstable paths grow at an adequate rate to make the ions impinge on the electrodes. Accordingly, the length of the field, L, must be made just large enough so that the ions' period of existence in the field is great compared with the duration of a high-frequency period. The latter is $\tau = \frac{L}{V}$

when $v = \sqrt{\frac{2eU_B}{m}}$ is the velocity of the ions and U_B is the acceleration voltage. This expression should be great compared with 1/v; hence $L \ge r_0 \pi \frac{1}{2U_B} q$

$$2 r_0 \pi \sqrt{\frac{2 U_H}{U_0}} q$$

(11) For L = 50 cm, $r_0 = 0.5$ cm and $U_0 = 1000$ v, the number of changes of field is represented by $n = \frac{850}{\sqrt{U_B}}$. Hence, 100-v ions undergo 85 changes of field, and 1000-v ions only 25, a value which entails doubt as to whether the amplitudes of the unwanted masses have already grown to the extent that they no longer impinge on the collector.

If the desired masses are actually to pass through the field, their amplitudes in the y-direction and in the z-direction must not exceed the figure r_0 . This requirement signifies a limitation of the initial coordinates. Calculation of the amplitudes from the Mathieu differential equation for the possible a, q values within the stable range as a function of the initial conditions y_0 , z_0 and the penetration angle, is complicated and will be left to a later study. Paul and Steinwedel gave a rough estimate of the maximum penetration angle and the energy of the motion in the radial direction, respectively. Their estimate reads:

$$e U_y < e(U_0 + U_k) e^{-\pi \sqrt{2}q + \tilde{a}}$$

$$e U_k < e(U_0 - U_k) e^{-\pi \sqrt{2}q - \tilde{a}} \uparrow.$$

$$(12)$$

The work of Paul and Steinwedel is based on Mathieu's differential equation in the form:

$$\frac{1}{d\zeta^2} \stackrel{\text{def}}{\to} (\lambda + q \cdot \cos 2\zeta) = 0$$

V in that case is twice the vertex voltage.

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B. Experimental Section

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For practical implementation, the hyperbola field was approximated by four round brass rods 10 ± 0.01 mm in diameter, 50 cm long and 10 mm apart; i.e., the hyperbolas were replaced by their circles of curvature at the vertex (fig. 5). Measurements in the electrolytic trough and an estimate by calculation showed that this arrangement within a circle with a radius of $2/3r_0$ around the zero point embodies the exact field with sufficient precision. The rods were bedded in brass disks with Ergan as an insulating material, after which their position with respect to each other could not be adjusted. The rod system was screened on both sides from the ion source and the collector by grounded circular diaphragms 8 mm in diameter. Rubidium ions were shot into this field from an incandescent anode. Rubidium was chosen because its two isotopes, 85 and 87, are two mass units apart and have a favorable mixture ratio of 2.67:1.

From the field quantities, it can be calculated that with a high-frequency amplitude of 1000 v and ideal resolving power, mass 85 is located at a frequency of 2.55 mc. Masses 4 to 250 embrace a frequency range of 12 to 1.5 mc.



Figure 5. Cross-section of the electrode arrangement.

The high-frequency voltage was generated with a selfexcited push-pull transmitter. In this connection, the high-frequency voltage had to be kept constant over a rather large frequency range. This was achieved by indirect control of the anode voltage. The constancy thus attained was 0.01% over a frequency range of 200 kc. The controlled amplitude of the high frequency could be varied from 700 to 1100 v peak voltage, and the frequency from 2 to 3 mc at a frequency constant of 1×10^{-4} after a half-hour starting period. The high-frequency output required was about 5 w.



Figure 6. Cross-section of the setup. Ionenquelle = ion source; Justiervorrichtung = adjustment device; Zuführung der Hochfrequenz = high-frequency lead; Auffänger = collector; Kühlung = cooling; Glaszyl. = glass cylinder; Eintrittsblende = inlet diaphragm; Messingrohr= brass pipe; Austrittsblende = outlet diaphragm; Schnitt A-B = cross-section A-B.

The rod system had a capacity of about 60 picofarads; it was coupled in such a way that it was in the oscillating circuit as a booster capacity. The dc voltage was supplied symmetrically to the system via 2 high-frequency chokes and grounded. It could be taken either from a special line device or from the high-frequency generator by means of rectification, as desired. This had the following advantage: If the high-frequency amplitude should vary, the ratio U_g/U_0 would automatically remain constant. Figure 6 shows a cross-section of the assembly.

Measurement Results

The cathode influx was measured in a collector located at the end of the field as a function of the frequency or amplitude of the ac voltage and the amount of superimposed dc voltage. In the measurements shown in fig. 7, the amplitude was kept constant at 925 v and the frequency was varied, with increasing dc voltage as a parameter. The penetration voltage of the ions was 110 v.

At the resulting speed, the ions undergo about 75 high-frequency periods in the field. In fig. 7, the frequency is plotted as the abscissa, and the cathode influx as the ordinate. The cathode influx of isotope 85 was made equal to 100 in each instance. The fraction of the intensity of rubidium 87 which remains standing between the lines is stated in % as a measurement of the resolving power. When u = 0.164, the background is less than 1%. The half-width of the lines is 5 kc. Thus, the resolving power is $M/\Delta M = 250$. If the measured half-width Δv is plotted as a function of u, the result is a straight line of the kind to be expected from eq. (10) (fig. 8). The dependence on u expected from eq. (9) likewise applies to the resolving power $\nu / \Delta \nu$. This shows the sensitivity with which the resolving power reacts to changes of the superimposed dc voltage. It is noteworthy that the theoretical line width is almost twice as large as the measured half-width. The diagrammed theoretical width is based on the assumption that each ion located in the stable range also enters the collector. i.e., that the amplitudes of all particles of small \mathbf{r}_0 persist. However, this is not the case. Ions located in the vicinity of the stability boundary, as a thorough observation shows, execute very great amplitude surges in the field according to their individual initial phases, so that they too are eliminated. This leads to an apparent name wing of the stable range and thus to an amplification of the resolving power.

The mixture ratio of isotopes 85 and 87 (2.67:1) was reproduced with a tolerance of 3% without elaborate measurement devices.

Theory reveals that the stability properties of solutions do not depend on the initial conditions. Accordingly, the initial velocity must not have any influence on the location of a mass line. The fact that this is not actually the case is shown in an appropriate way by fig. 9. In the measurement reproduced here, the ions were accelerated with a 220-v alternating current, and thus they had energies between 0 and 310 ev. Notwithstanding, a good resolving power could be achieved.

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Figure 7. The cathode influx measured in the collector as a function of the frequency of the alternating current. Parameter $u = U_G/U_0$. Ionenstrom = cathode influx; Frequenz = frequency; MHz = megacycles.



Figure 8. The resolving power and the line width, respectively, as functions of the superimposed direct current. u = 100%. The dotted curves are calculated. kHZ = kilocycles.

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 ~ 2.5 X



Figure 9. Transmissivity curve of the mass filter with ions accelerated by 220-v alternating current. Ionenstrom = cathode influx.

Influence of Finite Field Length

As demonstrated above, as the velocity of the ions increases, they experience fewer high-frequency periods, so that the amplitudes of the unwanted masses cannot increase sufficiently by the end of the field. Consequently, the resolving power diminishes above a certain velocity. Figure 10 shows a transmissivity curve for 100 and 1000-v acceleration current with equal range width. It is seen that as few as 25 oscillations, corresponding to the initial energy of 1000 ev, yield a poor resolving power. However, even in the case of high penetration voltages the resolving power could be improved by further narrowing of the range.

During the measurements, the high-frequency voltage was usually kept constant and the frequency was varied. As a matter of principle, the reverse measurement process can also be used. An experimental test revealed this assumption to be correct.

Cathode Influx and Gain

Theoretical predictions cannot be made out of hand concerning the anticipated efficiency (usable cathode influx/cathode influx introduced) as a function of the penetration voltage. Experiments showed the following: The current in the collector decreases exponentially with increasing dc voltage, and hence with diminishing range width. At $\nu / \Delta \nu = 500$, the efficiency was 10^{-4} .



Figure 10a and b. Influence of the ion velocity on the line width. Acceleration voltage a $U_B =$ 100 v; b $U_B = 1000$ v. In both cases, u = 0.161.

On the other hand, by settling for a resolving power $\nu/\Delta \nu = 110$, which is perhaps adequate for an isotope separation, we could attain an efficiency of 10%. Measurement was usually made with cathode influxes of 10^{-6} amp. The efficiency depends substantially on the transversal energy of the ions and on the field adjustment. Certainly it can be improved.

Error Influences

Essentially, there are two effects which occur and interfere with the action of the mass filter, viz., field errors and charging of the electrodes. If the field does not have precisely the potential $V \sim (y^2 - z^2)$, non-linear terms appear in Mathieu's differential equation and influence the stability boundaries. Field errors of this kind occurred in conjunction with poor adjustment of the rods and the use of a field with hyperbolically milled electrodes which did not have a fine enough tolerance from the mechanical point of view. Consequently, the mass lines became double and triple, and hence unstable areas occurred within the stable range. These interesting phenomena will be studied further.

If the high-frequency voltage is not purely sinusoidal (i.e., if harmonics are present), Mathieu's equation cos 2 ζ must be replaced by the Fourier factorization of the voltage. However, this has only very slight influence. For example, if a rectangular voltage is used instead of the sinusoidal voltage, the only result is a somewhat different position of the stable ranges. The intersection of the boundary curve of the first stability range with the q-axis, for example, is displaced by about 20% in this case. Naturally, this is completely negligible in terms of the operating principle of the mass filter. On the other hand, subharmonics behave differently. They give rise to instabilities within the stable ranges. There is a definite possibility of using these "stop bands," as they are called in the theory of the high-focus synchrotron, for mass separation.

Even with an ideal field, the theoretically possible resolving power [eq. (9)] is limited in actual practice by the errors in the amplitude and frequency of the high-frequency voltage. The applicable equation is:

$$\frac{du}{du} = \frac{1}{\frac{dq}{q} + 2\frac{dv}{v} + \frac{dU_0}{U_0}},$$

It is an intrinsic feature of the filter described here that a relatively large cathode influx impinges on the electrodes constantly. If insulating layers form on the electrodes -- which is the case to a greater or lesser extent according to the material used -- the impinging charge cannot flow off rapidly enough. In the less serious cases, this has the effect of an additional dc voltage which blocks the range to a greater or lesser extent according to the magnitude of the cathode influx. This phenomenon falsifies the frequency ratio of the isotopes. It was observed especially on brass electrodes. This effect can be suppressed in large measure by using nickel-plated electrodes. High charges lead to considerable field distortions, which give rise to double and triple lines, as in the case of mechanical field errors. Nonconducting layers of this kind were removed by cleaning the system occasionally.

Space charge influences were not observed in conjunction with the low cathode influxes used.

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