Subharmonic Acceleration of Heavy Ions by the Cyclotron of Kyoto University

Fumio Fukuzawa, Nobutsugu Imanishi, Masakatsu Sakisaka and Koji Yoshida*

Department of Nuclear Engineering, Kyoto University

and

Yoshiaki UEMURA, Shigeru KAKIGI and Hirokazu FUJITA*

Institute for Chemical Research, Kyoto University

Received December 13, 1967

Characteristics of subharmonic acceleration of heavy ions by the cyclotron of Kyoto University were investigated experimentally. About 100 nA of third-subharmonically accelerated C^{2+} and N^{3+} ion beams were extracted out of the cyclotron. Several kinds of ion beams corresponding to the fifth-subharmonic acceleration were observed by a probe inserted in the cyclotron. Energies and vertical spreads of these ion beams were estimated carefully to examine the possibility of the fifth-subharmonic acceleration.

I. INTRODUCTION

The study of nuclear reactions induced by heavy ions has recently become more and more important. The heavy ion beam is also a very valuable tool for the investigations in the fields of atomic physics and solid state physics. In these fields, the energy of heavy ions is not necessarily to be so high as in the case of nuclear reactions.

We have accelerated C^{2+} and N^{3+} ions third-subharmonically by the cyclotron of Kyoto University,¹⁾ and extracted these ions successfully. Several experiments have been made using these ion beams. In this paper, we report on the characteristics of the acceleration of heavy ions, especially of the subharmonic acceleration.

II. CONDITION OF RESONANCE ACCELERATION

As is well known, the condition of resonance acceleration of an ion whose charge is e and mass is m is given by

$$B = \frac{m\omega}{e\nu} \quad (\nu = 1, 3, 5, \dots), \tag{1}$$

where B is the magnetic flux density and ω is the angular frequency of applied dee-to-dee voltage. The subharmonic number ν corresponds to such a mode of acceleration that the phase of dee-to-dee voltage changes by $\nu \pi$ during the time

^{*} 福澤 文雄, 今西 信嗣, 向坂 正勝, 吉田 紘二, 植村 吉明, 柿木 茂, 冨士田浩一

when an ion passes through one dee. The mode of acceleration with subharmonic number 1 is used usually in many cases, and is called "normal acceleration." Those with subharmonic numbers 3, 5, …… are called "the third-, the fifth-, …… subharmonic acceleration," respectively. We denote these mode of accelerations as I, III, V, ……. For example, C^{2+} III means the third-subharmonic acceleration of C^{2+} ion or the C^{2+} ion itself accelerated third-subharmonically.

The energy of ions accelerated by the ν th mode, E_{ν} , relates to that of accelerated by the normal mode, E_1 , as follows,

$$E_{\nu} = \frac{1}{\nu^2} E_{\rm I}.$$
 (2)

In order to obtain high energy ions, acceleration with smaller ν is favored. For the ions with large value of m/e, however, the resonance condition (1) cannot be fulfilled by the normal acceleration in some fixed-frequency cyclotrons. The subharmonic acceleration is inevitable in these cases.

The frequency of the dee-to-dee voltage and the maximum magnetic flux density of our cyclotron are 13 MHz and 1.8 Wb/m², respectively. Therefore, any ions whose mass-charge ratios M/z are less than 2.1 can be accelerated by the normal mode. Those ions which can be obtained directly from the ion source are H⁺, H₂⁺, D⁺, ³He²⁺, ⁴He⁺ and ⁴He²⁺. For heavier ions such as those of C, N and O, most ions extracted from the ion source are partially stripped ones which have the values of M/z far larger than 2, and cannot be accelerated by the normal mode.

The following quantity \tilde{B} is very useful to represent the kind of ions and the mode of acceleration simultaneously,

$$\widetilde{B} = 9 \frac{M}{z} \cdot \frac{1}{\nu} . \tag{3}$$

Ion						\widetilde{B}				
M/z	$^{1}_{1}\mathrm{H}$	$^{2}_{1}\mathrm{D}$	³ ₂ He	⁴ ₂ He	¹² ₆ C	$^{14}_{7}{ m N}$	¹⁶ ₈ O	ν=1	$\nu = 3$	ν=t
1.0	H+							9.0	3.0	1.8
1.5			³ He ²⁺					13.5	4.5	2.7
2.0	H_2^+	D^+		$^{4}\mathrm{He^{2+}}$				18.0	6.0	3.6
2.4					C^{5+}				7.2	4.3
2.7							O6+		8.0	4.8
2.8						N^{5+}		1	8.4	5.0
3.0	Н	D+	³ He ⁺		C4+				9.0	5.4
3.2							O^{5+}		9.6	5.8
3.5						N^{4+}			10.5	6.3
4.0		D_2 +		⁴ He ⁺	C ³⁺		O4+		12.0	7.2
4.7						N^{3+}			14.0	8.4
5.3							O^{3+}		16.0	9.6
6.0					C^{2+}				18.0	10.8
7.0						N^{2+}				12.6
8.0							O^{2+}			14.4

Table 1. Values of \widetilde{B} for various kinds of ions.

The numerical factor 9 is added for convenience. The resonance magnetic field B for the acceleration of the ions specified by a given value of \tilde{B} is evidently proportional to this value. Values of \tilde{B} for some ions are given in Table 1. From the curve of B vs. magnet current I_B , we can determine the necessary magnet current for the acceleration of the ion with a given \tilde{B} . Fig. 1 shows the \tilde{B} vs. I_B curve which is calibrated with the experimental values of H⁺I, N³⁺III, C²⁺III and H₂⁺I.

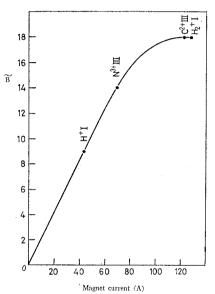


Fig. 1. \overline{B} vs. I_B curve which is calibrated with the experimental values of H⁺I, N³⁺III, C²⁺III and H₂⁺I.

III. INNER ION BEAM

1. Observation of Resonances²⁾

Resonance accelerations were observed by a probe electrode which was inserted between two dees to measure the inner ion beam. As shown in Fig. 2, the probe electrode was covered with a copper tube and only the tip of width 5 mm was effective in the measurement of the ion current.

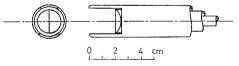


Fig. 2. Schematic view of the probe electrode used to measure internal beam intensity. Material is water-cooled copper.

The ion source used is a modified arc type, the construction of which is described in our previous report.¹⁾ However, materials of some constitutions are different from the reported ones. First, cavity material was changed from Cu-W to Mo, and the copper cap for shielding the quartz insulator was also replaced

by that of Mo. Second, the canal part of the arc chamber was made of wolfram instead of graphite. Due to these improvements, the present ion source has excellent properties of the smallness of out-gas and stability at high temperature.

The area of ion outlet is 0.15 cm^2 . Optimum quantity of gas flow was in the range of $0.1 \sim 0.3$ atm cc/min for any gases used, and the pressure in the cavity of the ion source was estimated to be about 1×10^{-3} torr. Usually, the power of about 750 W is used for gas discharge, and the lifetime of a filament of $2 \text{ mm}\phi$ wolfram wire was 10 hours for He gas and $2 \sim 4$ hours for N₂ and CO₂ gases.

In order to observe resonances for several kinds of gases, the probe tip was placed at the radial distance of 40 cm from the center of the magnet. Some of

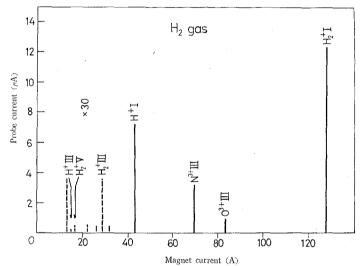


Fig. 3. Probe current at the radius of 40 cm vs. magnet current. H_2 gas is supplied for the ion source.

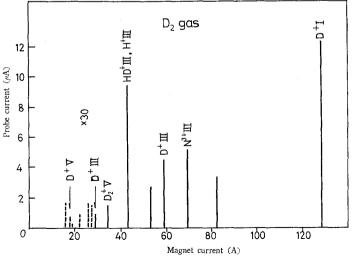


Fig. 4. Probe current at the radius of 40 cm vs. magnet current. D_2 gas is supplied for the ion source.

the results obtained are shown in Figs. $3\sim7$. Kinds of ions suggested from Fig. 1 and Table 1 are designated to resonance peaks in these figures. Some of them do not originated from gas used, and may be due to residual gas, leaked air and decompositions of diffusion pump oil. Intensities of these peaks varied depending on the process of evacuation.

Because of the rather small angle of dee sector of our cyclotron (143°) , it is very doubtful for the fifth-subharmonic acceleration to be realized. It appeared, however, that there were many resonance accelerations corresponding to III- and V-mode. The possibility of V-mode acceleration will be examined in a later section of this paper.

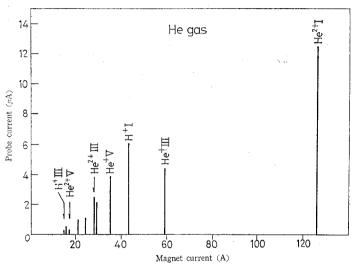
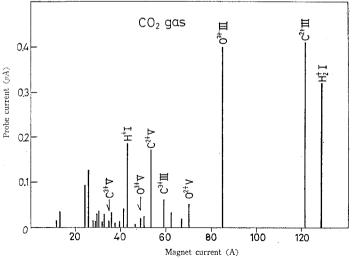
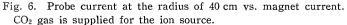
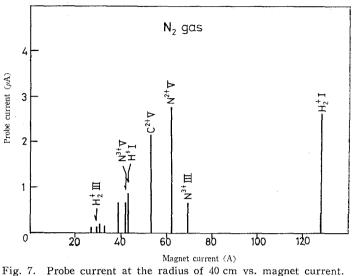


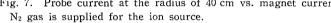
Fig. 5. Probe current at the radius of 40 cm vs. magnet current. He gas is supplied for the ion source.





Fumio FUKUZAWA, et al.





2. Radial Extension of the Resonant Ion Beam

The feature of the radial extension of inner beam under a given value of B can be seen by the measurement of the probe current at varying position of the probe tip.

For some values of *B* near the resonance of N³⁺III, the results obtained are shown in Fig. 8. In this figure, we can see clearly that the inner beam extends to large radial distance as the value of *B* comes near to the optimum one, and this corresponds to the improvement of the phase relation. Sudden increase of the probe current just before fall is probably due to the radial bunching of inner beam near the value of the phase shift of $\pi/2$. Small fluctuations of the probe current associated with the probe position is caused by the discreteness of the radius of ion circulation. This can be confirmed as follows: in our cyclotron, the relation of orbit radius r(cm) and energy E(MeV) is given by

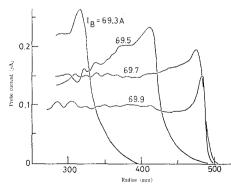


Fig. 8. Probe current vs. radius for different magnet current I_B .

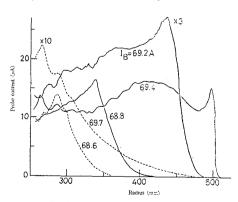


Fig. 9. Probe current vs. radius for different magnet current I_B ; two shims of radius 42 cm and thickness 1 mm were installed.

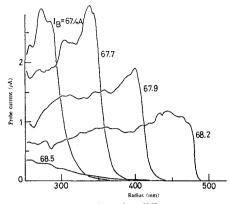


Fig. 10. Probe current vs. radius for different magnet current I_B ; four shims of radiur 45 cm and thickness 1 mm were installed.

$$E = 7.25 \left(\frac{r}{47}\right)^2 \frac{M}{\nu^2}.$$
 (4)

The figure 47 in this formula implies that the distance between the septum and the center of the magnet is 47 cm. Accelerating voltage of 100 kV in the dee gap produces the increase of radius of orbit per turn $\Delta r(\text{cm})$ at r(cm) as given in Table 2.* These values agree very well with the observed intervals between successive maxima of probe current.

Table 2. Values of the increase of orbit radius and periods of fluctuation in the probe current.

Orbit radius r (cm)	Increase of orbit radius Δr (cm)	Period of fluctuation (cm)
30	2.0	2.0
35	1.6	1.6
45	1.2	1.1

Similar measurements with some shims installed were made. The results are shown in Figs. 9 and 10. The former corresponds to the case with two shims of radius 42 cm and thickness 1 mm, the latter to the case with four shims of radius 45 cm and thickness 1 mm. Indices of magnetic field at the distance of 35 cm from the center of the magnet were 1.3×10^{-2} and 7.0×10^{-2} , respectively, while that without shim was zero. These figures show, for *B* larger than the optimum one, the exponential decrease of probe current with increase of radial distance. The feature of the radial extension of the inner ion beam changes drastically when the value of *B* exceeds the optimum one. This may be expected from the phase relation, and is very important in the practical technique of ion-acceleration. Besides the small fluctations, there are large bumps of width 5~10 cm. These imply the orbit bunching caused by the radial

^{*} Because of the wide space of dee gap of our cyclotron, the accelerating field varies considerably while an ion is in the field. The approximation that an ion is accelerated by the constant field causes about 20% error in the estimation of Δr .

oscillation of the ion path.

It should be noted that the probe current given in Figs. $8 \sim 10$ should be considered as relative values. Because the intensity of the inner ion beam depends very sensitively on the conditions of the ion source and the dee-to-dee voltage, the values of the probe current obtained under the different conditions cannot be compared on the same level.

IV. EXTRACTION OF ION BEAM³⁾

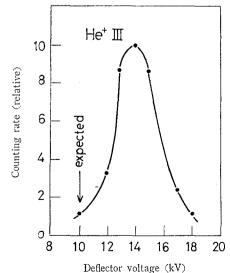
In order to decide whether the assignments of ions for the peaks shown in Figs. $3\sim7$ are correct or not, it is desirable to extract the corresponding ions out of the cyclotron. The ions of He⁺III, C²⁺III and N³⁺III were extracted successfully through the deflector and confirmed in the following manner.

These ions should have energies of 3.2, 9.7 and 11.3 MeV, respectively, and be detectable by a solid state semiconductor detector. A p-n junction type semiconductor detector with the window of 0.75 mg/cm^2 thick was used to detect ion beams.

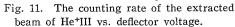
The deflector voltage V_{def} necessary to extract the ion of charge ze_0 (e_0 is electronic charge) and energy E is given approximately by the relation

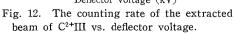
$$V_{def} \propto \frac{E}{ze_0},\tag{5}$$

and it is known that the value of V_{def} for the extraction of 29 MeV He²⁺I ions is 45 kV. For the above three ions they are estimated to be about 10, 15 and 12 kV, respectively. In Figs. 11 and 12, the counting rates of the extracted beams of the He⁺III and C²⁺III are shown as a function of deflector voltage. For N³⁺III, the probe current at the exit of deflector was measured and corrected for the background current, which was remained even when the deflector voltage was



C²⁺ Ш 10 Counting rate (relative) 8 6 4 2 10 14 16 18 20 12 22 Deflector voltage (kV)





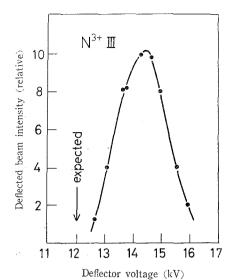


Fig. 13. The intensity of the deflected beam of N³⁺III vs. deflector voltage.

turned off. The result obtained is shown in Fig. 13. These figures show the expected resonance peaks about estimated values of V_{def} .

The energies of ions can be determined by the pulse heights from the detector. After correction of the energy loss in the window of the detector by use of Papineau's semi-empirical values of effective charge,⁴⁾ we obtained energies of He⁺III, C²⁺III and N³⁺III as 3.5 ± 0.1 , 9.8 ± 0.1 and 12.0 ± 0.1 MeV, respectively. These values agree very well with the estimated values for suggested ions.

Further confirmation was made by nuclear emulsions which were exposed to suggested ion beams of $C^{2+}III$ and $N^{3+}III$. Many tracks with same length (10 micron) and same incident angle were observed under a microscope at the portion of irradiation.⁵⁾ Length of these tracks agrees with the ones of 10 MeV Cion and 12 MeV N-ion which were estimated by the extrapolation of the published data.⁴⁾

From the above facts, assignments of He⁺III, C^{2+} III and N³⁺III are correct. For other kinds of ions corresponding to low resonance magnetic fields, it is impossible to extract ion beams out of cyclotron because indices of magnetic fields at the position of the deflector become negative due to the effect of the Roseshim. Measurement of energies of ions by the inner probe electrode were tried as described in the next section.

So far, the extracted beam intensities of $C^{2+}III$ and $N^{3+}III$ amount to at best 100 nA at the exit of the deflector. The experiments on charge-changing collisions^{6,7)} and on Coulomb excitation have been made by use of these ions.^{8,9)} In order to make stable acceleration, it is necessary to stabilize the frequency of dee-to-dee voltage and the magnetic flux density to the order of 1×10^{-4} .

V. DETERMINATION OF ENERGY OF INNER ION BEAM

In order to determine the energy of ions which collide with the inner probe

electrode, the rise of temperature of the probe was measured by a thermocouple attached to it. So as to be able to measure simultaneously the vertical spread of the ion beam, a three-finger probe was used. Construction of the tip of this probe is shown in Fig. 14. Three separated pieces B, A and C are independent probe electrodes. A thermocouple was fixed to the middle probe A.

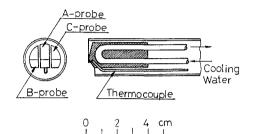


Fig. 14. Schematic view of the three-finger probe used to measure the vertical spread of the ion beam. Material is water-cooled copper.

When ions of charge ze_0 and energy E collide with A-probe, the heat quantity Q produced per unit time is proportional to $I_A E/z$, where I_A is the A-probe current. On the other hand, A-probe losses its thermal energy by the conduction of heat of probe-support, the heat transfer in cooling water and the heat radiation. So long as the rise of temperature is not large, the last effect can be ignored. Heat loss q by the former two causes may be proportional to the difference of temperature of the A-probe (θ) and of cooling water (θ_0) . In the equilibrium, q is equal to Q. Therefore, the equilibrium temperature of the probe is given by

$$\theta = c \frac{I_A E}{z} + \theta_0, \tag{6}$$

where c is a constant depending on the structure of probe and the flow rate of the cooling water. As thermo-electromotive force of the thermocouple is proportional to the temperature θ , we can use the magnitude of thermo-electromotive force itself for θ , so far as the value of E/z is concerned.

Placing A-probe at the distance of 35 cm from the center of the magnet, we

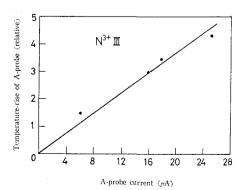


Fig. 15. Temperature-rise of A-probe vs. A-probe current for N³⁺III beam.

measured temperature θ for some values of I_A . In Fig. 15, the temperature rise $(\theta - \theta_0)$ obtained for N³⁺III is shown. For other kinds of ions, similar linear relation of θ vs. I_A were obtained. Linear relation warrants Eq. (6), at least within the error of these measurements. To determine values of E/z of various kinds of ions, the mean value of c deduced from the data on N³⁺III and C²⁺III was used, because these ions were confirmed as described in the preceding section. Experimental values of E/z, obtained in such a manner, are shown in column 5 of Table 3. Predicted values of E/z for supposed ions, shown in column 4, were calculated in consideration of the energy relation at r=35 cm, that is,

Magnet current	\widetilde{B}		E/z			
(A)	В	Supposed ion	Predicted	Experimental	$ u_{exp}$	
27.57	5.7	He ²⁺ III	0.89	0.05	50.7	
34.10	7.0	He ⁺ V	0.64	0.19	16.4	
37.00	7.7	?		<0.30	>11.4	
42.14	8.8	N ³⁺ V	0.75	0.85	4.6	
42.93	9.0	H+I	4.0	5.6	0.73	
49.77	10.3	$\begin{cases} C^{2+}V\\ O^{3+}V \end{cases}$	0.96 0.85	0.72	6.4	
60.58	12.3	$N^{2+}V$	1.1	0.44	12.4	
69.70	14.0	N ³⁺ III	2.1	1.9	3.3	
122.19	18.0	C ²⁺ III	2.7	3.0	2.7	

Table 3, Predicted and experimental values of E/z and experimental subharmonic numbers.

$$E = 4.01 \frac{M}{\nu^2} \quad \text{(MeV).} \tag{7}$$

There are agreement between the predicted values of E/z and experimental ones in some cases, and not in other cases. In order to see the fitness more distinctly, we treat this problem from the different point of view.

Combining Eqs. (3) and (7), we obtain

$$\frac{\tilde{B}}{E/z} = \frac{9}{4}\nu. \tag{8}$$

The value of \tilde{B} for any magnet current can be read from Fig. 1. Putting this value of \tilde{B} and experimental value of E/z in Eq. (8), we can determine the sub-harmonic number ν without specification of the kind of ions. Values of ν determined in this manner, ν_{exp} , are shown in column 6 of Table 3.

It seems likely to assume the supposed ion is correct one in the case that the value of ν_{exp} is near to the value of subharmonic number of supposed ion. However, even when the value of ν_{exp} becomes very large, we can retain the supposed ion by interpreting as follows. For ion beams with larger ν_{exp} values than predicted ones, there should be a lot of scattered ions in them, which have small energies and cannot contribute to the temperature-rise of the probe but to the probe current. Consequently, the value of E/z estimated from this probe current I_A should be smaller than the expected one, and lead to larger value of

 ν_{exp} . From values of ν_{exp} , we estimated the amount of the scattered ions as $2\sim$ 20 times as large as that of regular ions. If this interpretation is right, there should be corresponding effect in the vertical spread of beam. This is considered in the next section.

VI. VERTICAL SPREAD OF INNER ION BEAM

It is very interesting to know the extent of spread of the inner ion beam in the vertical direction (direction of the magnetic field). This can be estimated from three probe currents measured with the three-finger probe following the method of calculation described in the Appendix.

Assuming the vertical distribution of beam intensity to be Gaussian, we have the following expressions for the width of beam spread Γ and the vertical shift z_A of the middle A-probe axis from the beam center,

$$\Gamma^2 = \frac{8\Delta^2}{2i_A - (i_B + i_O)} \tag{9}$$

$$z_A = \frac{(i_0 - i_B)\varDelta}{2\{2i_A - (i_B + i_0)\}} \tag{10}$$

where i_A , i_B and i_0 are the logarithmic values of A-, B- and C-probe currents, respectively. The Δ is the distance of separation between centers of nearby probes, and is 7 mm in the present case.

The values of Γ and z_A obtained for some supposed ions are shown in Table 4. The positive (negative) value of z_A corresponds to the axis of A-probe to be lowered (raised) than the center of beam. The black mark (\times) in the column

Magnet current (A)	Supposed ion	Γ (mm)	$z_A \ (mm)$	
27.57	He ²⁺ III	×		
34.10	He ⁺ V	×		
11	//	23.7	-3.3	
11	11	27.7	+3.5	
37.00	$\widetilde{B} = 7.7$	×		
42.14	N ³⁺ V	12.9	+0.43	
//		×		
//	//	15.5	+1.2	
42.93	H+I	16.7	-0.45	
49.77	$C^{2+}V, O^{3+}V$	×		
60.58	N ²⁺ V	14.5	-1.7	
//	//	\times \cdot		
	"	17.9	-2.0	
//	//	24.0	-1.8	
69.70	N ³⁺ III	15.6	-0.76	
//	//	16.3	-1.5	
122.19	C ²⁺ III	13.2	+0.11	
128.23	H_2 ⁺ I	14.2	-1.1	

Table 4. Values of the width of beam spread I' and shift z_A of a probe axis from the beam center for various kinds of ions. Different values were obtained in different runs.

of Γ means that the value of Γ calculated by Eq. (9) became imaginary and Gaussian distribution could not be applied. From values for H₂+I, C²⁺III and N³⁺III, it seems that reasonable values of Γ and z_A are 13~17 mm and 0~-1.5 mm, respectively.

As is noted in the preceding section, if the large value of ν_{exp} estimated from the measurement of the temperature of the probe is caused by the scattered ions, the vertical distribution of beam intensity should be very large in Γ or could not be the Gaussian distribution. Comparing the results shown in Tables 3 and 4, we find actually that ions with large values of ν_{exp} have large values of Γ or imaginary Γ . The shifts z_A in these cases are also large.

It is very regret that we have few data until now on the variation of the vertical distribution of beam as the probe distance from the center of magnet varied. This will be studied in near future.

VII. CONCLUSION

We have been able to accelerate ions of C^{2+} and N^{3+} by the third-subharmonic mode, and extract these ion beams of about 100 nA. Many resonances corresponding to the fifth-subharmonic acceleration were observed by the inner probe. The result of the measurements of their energies and vertical spread contains some ambiguities, and we cannot decide, at present, whether the fifth-subharmonic acceleration takes place or not. If these resonances are not due to the fifth mode, what on earth are these? The reproducibility of the phenomena is too good to attribute to the ions accelerated initially at the place other than the center of the cyclotron. It is also difficult to seek the ions with appropriate values of M/z to be accelerated by the normal or the third mode. On the other hand, affirmation of the fifth mode should cause serious difficulty in the problem of ion acceleration by the dees with so much small sector-angle of 143°. If this is true, the way of thinking of the ion-acceleration should be changed drastically. Before proposing definite solution on this problem, we must proceed further measurements on the spread of beam and calculate precisely the ion path (phase relation, beam focusing etc.).

ACKNOWLEDGEMENTS

Authors would like to express sincere thanks to Profs. T. Yanabu, S. Yamashita and I. Kumabe for thir interest in this problem and many valuable discussions. They are also grateful to Messrs. T. Takeuchi, K. Sano, T. Maruyama and S. Kakui for their cordial help in the operation of the cyclotron.

APPENDIX

Vertical Distribution of Beam Intensity

Using a three-finger probe (see text), the width of the beam spread and the vertical shift of the probe axis from the beam center are estimated in the following way.

The vertical distribution of the beam intensity, I(z), is assumed to be Gaussian,

$$I(z) = I(0) \exp\left(-\frac{4z^2}{\Gamma^2}\right) \tag{A-1}$$

where Γ is the width parameter. Then, the beam current measured by the probe of width b placed at a distance of z from the center of beam is expressed as

$$I(z, b) = \int_{z-b/2}^{z+b/2} I(z) dz$$
 (A-2)

This is approximated as

$$I(z, b) \approx I(z)b \tag{A-3}$$

When three probes B, A and C are placed side by side as shown in Fig. A-1, each probe currents I_B , I_A and I_C are expressed, in this approximation, as

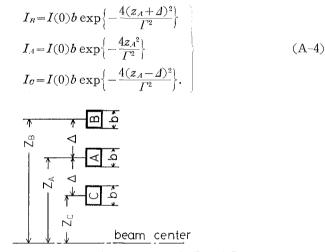


Fig. A-1. Arrangement of three probes A, B and C.

These equations can be solved easily, provided Γ to be real,

where

 $i_B = \ln I_B$, $i_A = \ln I_A$, $i_C = \ln I_C$ and $i_0 = \ln \{I(0)b\}$.

The error caused from the approximation of (A-3) is estimated as follows. Putting $y=2\sqrt{2}z/\Gamma$ and $W=2\sqrt{2}b/\Gamma$, Eq. (A-2) can be rewritten as

$$I(z,b) = \frac{\sqrt{\pi}}{2} I(0) \Gamma\left\{ \phi\left(y + \frac{W}{2}\right) - \phi\left(y - \frac{W}{2}\right) \right\},\tag{A-6}$$

where

$$\Phi(y) = \int_0^y \varphi(t) dt \text{ and } \varphi(t) = \frac{1}{\sqrt{2\pi}} \exp\left(-\frac{t^2}{2}\right).$$

While, the approximated probe current (A-3) is rewritten as

$$I(z)b = \frac{\sqrt{\pi}}{2}I(0)\Gamma\varphi(y)W.$$
 (A-7)

The ratio of the exact to the approximated probe current is given as

$$\frac{I(z)b}{I(z,b)} = \frac{\varphi(y)W}{\varphi\left(y + \frac{W}{2}\right) - \varphi\left(y - \frac{W}{2}\right)},\tag{A-8}$$

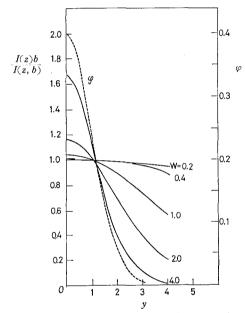


Fig. A-2. The ratio of the exact to the approximated probe current for some values of W.

and is shown in Fig. A-2 as a function of y for some values of W. Using Eq. (A-5), we can calculate values of Γ , z_A , z_B and z_σ from experimental values of probe currents and estimate above ratios from Fig. A-2. If they are nearly equal to unity, we can regard it as appropriate to use the approximation (A-3). In the present case, these ratios corresponding to A-, B- and C-probe currents for C²⁺III, for example, are 1.07, 0.93 and 0.93, respectively. For other kinds of ions, we have also values of nearly equal to unity. The relative error caused from this approximation is estimated to be less than 5% in all cases.

REFERENCES

- K. Kimura, Y. Uemura, M. Sonoda, S. Shimizu, T. Yanabu, R. Ishiwari, J. Kokame, A. Katase, I. Kumabe, S. Yamashita, H. Takekoshi, K. Miyake, H. Ikegami and H. Fujita, Bull. Inst. Chem. Res., Kyoto Univ. 39, 368 (1961).
- (2) Y. Uemura, H. Fujita, S. Kakui, M. Sakisaka, F. Fukuzawa, M. Tomita, K. Yoshida, S.

Okumura, S. Imanishi, K. Kobayashi, T. Takeuchi and T. Nakata, Genshikaku Kenkyu (circular in Japanese) 8, 617 (1964).

- (3) Y. Uemura, H. Fujita, S. Kakui, M. Sakisaka, F. Fukuzawa, M. Tomita, K. Yoshida, S. Okumura, S. Imanishi, K. Kobayashi, T. Takeuchi and T. Nakata, *Genshikaku Kenkyu* (circular in Japanese) 9, 363 (1964).
- (4) A. Papineau, J. Phys. radium 17, 556 (1956), Compt. rend. 242, 2933 (1956).
- (5) K. Kimura, Decennial Report 1955–1965, Bull. Inst. Chem. Res., Kyoto Univ., 43, 499 (1965).
- (6) Y. Uemura, H. Fujita, T. Maruyama, M. Sakisaka, F. Fukuzawa, M. Tomita, K. Ogino, K. Yoshida, S. Okumura, N. Imanishi and T. Nakata, *Genshikaku Kenkyu* (circular in Japanese) 10, 303 (1965).
- (7) N. Imanishi, M. Sakisaka, F. Fukuzawa and Y. Uemura, J. Phys. Soc. Japan 20, 2100 (1965).
- (8) M. Sakisaka, F. Fukuzawa, N. Imanishi, K. Shima, K. Tsuji, T. Yamazaki and Y. Uemura, Genshikaku Kenkyu (circular in Japanese) 11, 597 (1967).
- (9) N. Imanishi, F. Fukuzawa, M. Sakisaka and Y. Uemura, Nuclear Physics, A101, 654 (1967).