with experiment. A more accurate treatment should also include the effect of something resembling surface tension of the (probably diffuse) cavity wall.

Extending the simplified particle-in-a-spherical-box treatment to a system of *two* 1s electrons trapped in a cavity, application of first-order perturbation theory yields

$$W=\frac{h^2}{4mr_0^2}-\frac{2e^2}{r_0}+\int\frac{e^2}{r_{12}}\psi_0^2dT.$$

The third term (involving the unperturbed wave functions and the separation r_{12} of the electrons) corresponds to electrostatic repulsion between the electrons, and is evaluated by adaptation of the analogous treatment for the normal helium atom.[4] By numerical methods, its value approximates to $+e^2/r_0$. The corresponding approximate values of r_0 and W would be $h^2/2me^2$ and $-me^4/h^2$. From physical analogy with the helium atom like problem, it appears that higher approximations would yield a value of W numerically *greater* than me^4/h^2. (For example, first order perturbation treatment gives for hydride ion[4] $W=-0.75\ W_H$, while variation methods yield $W=-1.05\ W_H$.) The above theoretical trapping energy for a single electron is $-me^4/2h^2$. That is, the trapped pair should be stable with respect to dissociation into two electrons trapped in separate cavities. This at first sight paradoxical result arises from the greatly increased polarization energy of the pair (*vide* the factor Z^2 in the Born formula) which more than compensates for electrostatic repulsion. It appears highly probable that this energetic stability of the pair would be retained by a more accurate treatment (i.e., one considering the penetration of the wave functions outside the cavity). The Pauli exclusion principle requires the trapped pair to be in a singlet state—i.e., to be diamagnetic.

The hitherto neglected inter-ionic forces become increasingly important with greater solute concentration. In the limit, the electrostatic attractive potential on a cluster of Z electrons at average distance ρ from singly charged positive ions approaches $-AZ(e^2/\rho)$. The Madelung constant A for random configuration should have a value of the order of magnitude of 1.5, and hence this potential can become (for $Z=1$) much larger than the polarization potential $-Z^2e^2/2r_0$. One result is a greatly decreased value of the radius of the electron cavity—the molar volume in concentrated solutions of sodium is about 40 cm^3, as compared with some 700 cm^3 for highly dilute solutions. Further, the linear dependence on Z gives no appreciable energetic advantage to the pairing of electrons, and their mutual repulsion would render the pairs unstable. This unpairing of electrons in concentrated solutions is indicated by the reappearance of appreciable paramagnetic susceptibility, as contrasted with diamagnetism of the solute at intermediate concentrations.

Extension of the "particle in a box" treatment indicates that the kinetic energy of a trapped electron excited to a p state exceeds the potential energy of the s ground state. By the Franck-Condon principle a continuous absorption spectrum should result, leading to photo-ejection of the electron from the cavity. The infra-red absorption maximum of very dilute metal-ammonia solutions corresponds to transition to the first p state. Current experimental studies reveal a far ultraviolet absorption maximum, corresponding to transition to the second p state. The theory of the continuous absorption spectrum of the trapped pair is developed in like fashion.

The theory as outlined applies in essential detail to any solvent of *high dielectric constant* which is chemically indifferent to free electrons. The known examples would appear to be ammonia, organic amines, and possibly fused alkali metal amides.

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[2] C. A. Kraus, *The Properties of Electrically Conducting Systems* (Chemical Catalogue Company, 1922).
[3] For fuller mathematical details, see G. Gamow, *Atomic Nuclei and Radioactivity* (Oxford University Press, New York, 1931), pp. 42–49.
[4] L. Pauling and E. B. Wilson, Jr., *Introduction to Quantum Mechanics* (McGraw-Hill Book Company, Inc., New York, 1935), Appendix V.

Frequency Modulated Cyclotron

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THE application of the synchrotron phase stability principle[1,2] to the frequency modulated cyclotron has been investigated experimentally on the 37-inch Berkeley cyclotron. A radial decrease in magnetic field has been used such that it requires a frequency change simulating the change which would be required by the relativistic increase of mass with velocity as the ions are accelerated to high speeds.

The frequency of revolution of an ion of kinetic energy W (units Mc^2) in a magnetic field H is

$$f=\frac{f_c}{1+W}\frac{H}{H_c}$$

where f_c is the frequency the ion would have with very low energy in the field H_c. Thus the change in frequency produced by an increase in W to a large value can be simulated by the proper radial decrease in magnetic field even though ions of a comparatively low energy are used. The purpose of the experiments described here was to simulate the acceleration of deuterons to 200 Mev in the giant 184-inch cyclotron. Here $W=0.107$ so that a total frequency change of 13 percent would leave 2.3 percent for radial decrease of magnetic field in the 184-inch case.

We have succeeded in accelerating a time average deuteron current of 0.2 microampere through a radial fall in magnetic field of 13 percent. This was accomplished by a radio frequency peak potential of 3 kv on the dee which accelerated the deuterons to 7 Mev. Without frequency modulation the maximum energy obtainable is 0.5 Mev with this voltage on the dee (in accord with theory). So far our maximum modulation frequency has been 600 cycles per second which corresponds to an acceleration time of 500 μs or about 5000 turns. The threshold appears at 5 milliseconds or 5×10^4 turns.

In our experimental arrangement we use a single dee whose diametral edge is perpendicular to the axis of the

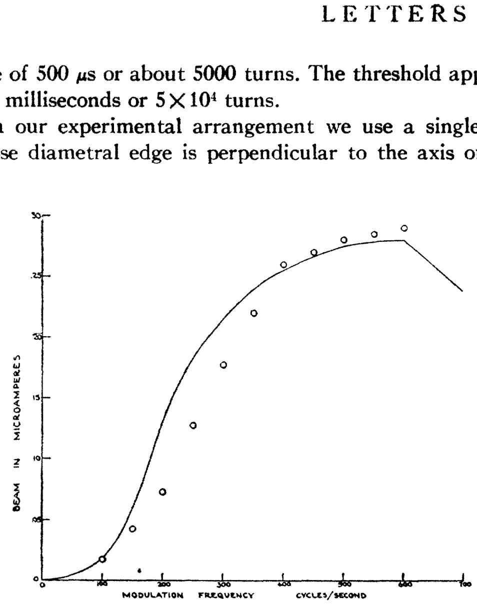

FIG. 1. The beam current collected on a probe at a radius of 17 inches where the field is 11.5 percent smaller than H_c, the field at the center. This current is plotted as a function of the modulator frequency.

resonant line, with the mechanical rotating condenser[3] on the other end of the line. The line resonates with a voltage node near the center moving back and forth as the resonant frequency is changed by the rotating capacitor. A 20 percent change in frequency is produced in this way. The condensor rotor consists of 36 teeth on a circle two feet in diameter. Thus a rotation of 1000 r.p.m. corresponds to 600 cycles per second for the modulating frequency. The grounded grid r-f oscillator required 600 watts of plate power.

Under the usual operating conditions (2800 volts on the dee) the dee and resonant line system float at a potential of about +1500 volts d.c. The output under these conditions is a factor of ten greater than when the dee system is grounded. We believe that this unusual situation is concerned with the ion starting conditions under these low single dee voltages and is not concerned with the frequency modulation characteristics of the instrument.

Figure 1 shows the probe current as a function of the angular velocity of the mechanical condenser expressed in cycles per second of the modulating frequency. Readings on a "protection meter" ionization chamber placed six feet outside the cyclotron indicated strict proportionality between the radiation from the cyclotron and the probe current. The smooth curve shows the theoretical prediction[4] of the yield as a function of modulator frequency. With no modulation and the probe at 2.5-inch radius, 22μa of current was picked up, indicating an efficiency of 1 percent for the f-m system. This is in close agreement with theory, if one takes into account the fact that only about one-sixth of the modulation cycle is used in this experiment.

The results of an investigation of the beam *vs.* radius are shown in Fig. 2. The probe current indicates a very slow fall off with radius, while the onset and rapid increase of ionization chamber reading with radius is consistent with the deuterons being accelerated to about 7 Mev as predicted from the values of magnetic field and radius. The order of magnitude of this energy was verified by excitation of the 12.8-hour radioactivity in copper.

The increase in beam with increasing r.f. voltage on the dee is very marked—a factor of ten in going from 2 to 3 kv. This must be due to an increase in source efficiency since it can easily be shown that the increase in f-m efficiency will only go as the square root of the dee voltage if the shape of the modulation cycle is unchanged. An evacuated rotating condenser is being constructed so that we can raise the voltage to 10 kv on the dee and increase the rate of rotation.

In view of the success of these experiments it has been decided to complete the 184-inch instrument as an f-m cyclotron. From Fig. 2 we can say that appreciable values

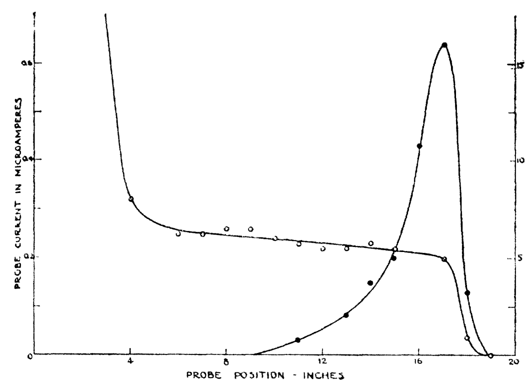

FIG. 2. Beam current as a function of radius. The modulator frequency was 590 cycles per second, the peak r-f dee voltage was 2800 volts, and the dee bias +1500 volts.

for the time average current of 200-Mev deuterons would be expected from the giant cyclotron even with 3 kv on the dee. At ordinary dee voltages of 50 kv, say, the output would be larger both because of the increase in efficiency and the increase in source efficiency. A reasonably conservative expectation would appear to be a time average current of one microampere of 200-Mev deuterons with 50 kv on the dee.

It is clear, also, that the f-m system will have advantages for smaller cyclotrons where it is desired to obtain high energy protons with reasonable dee voltages. Bringing the beam out of the f-m cyclotron is an important problem, but there are several solutions which look feasible and which we intend to try.

We wish to express our thanks to Drs. E. M. McMillan and R. L. Thornton for many helpful suggestions and criticisms and our gratitude to Professor Ernest Lawrence whose inspiration and encouragement made this development possible. This work was done under the auspices of the Manhattan District.

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