

## The Intermediate State of a Hollow Superconducting Tin Cylinder\*†

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Small bismuth probes were used to measure the magnetic field inside and outside a long, hollow, thin-walled superconducting cylinder, both as a function of the applied field and as a function of the angular position of the probes. From the variation of the magnetic field the structure of the cylinder in the intermediate state was inferred. It would appear that with the onset of the intermediate state "windows" are formed at the poles of the cylinder. These "windows," through which the applied magnetic field can penetrate into the interior of the cylinder, probably consist of the usual rapidly alternating layers of normal and superconducting metal. The parts of the cylinder around the equator remain wholly superconducting until the applied field becomes equal to the critical field, at which time the entire cylinder becomes normal.

The intermediate state for the particular cylinder used in this experiment begins to appear at about  $0.3H_c$ , instead of at  $0.5H_c$ . It will be shown in a subsequent paper that this value is a function of the ratio of the inner and outside radii of the hollow cylinder.

### 1. INTRODUCTION

A LONG, solid metal cylinder in the superconducting state, when placed in a uniform magnetic field parallel to its axis, will remain in the superconducting state as long as the applied field is less than the critical value,  $H_c$ . When the applied field becomes equal to  $H_c$  the entire cylinder immediately goes into the normal state and remains in this state for all larger fields.<sup>1</sup> If, however, the cylinder is placed in a uniform magnetic field which is transverse to its axis, the transition from the superconducting state to the normal state becomes complicated. Instead of changing into the normal state at once, the cylinder first goes into the so-called intermediate state, since with the above geometry the magnetic field is not constant over the surface of the superconductor because of the magnetization of the cylinder.

A large amount of work has been done on the intermediate state of uniformly magnetized superconductors, particularly the solid sphere and the solid cylinder.<sup>2-11</sup> The results of this work may be summarized as follows: In the superconducting state, the magnetic field at the surface of the cylinder varies from zero at the two poles to a maximum at the two equators. (This statement

specifies what we mean by the poles and equators.) As the applied field is increased the field at the equators reaches the critical value before the field at any other point. When this occurs the whole body goes into an intermediate state, breaking up into a uniform distribution of small regions of superconducting metal and normal metal, in such a way that the average field inside the metal is everywhere equal to the critical value and is in the direction of the applied field. The superconductor remains in this state until the applied field equals  $H_c$ . For applied fields equal to or greater than  $H_c$  the entire superconductor is in the normal state.

A further step in the understanding of the intermediate state would be to investigate the field distribution around a superconductor whose geometry is such that it is not uniformly magnetized when placed in a uniform magnetic field. The simplest geometry of this type, experimentally, is the hollow circular cylinder.

The experiment to be described consists of measurements of the magnetic field distribution around the outer and inner surfaces of a hollow tin cylinder placed in a uniform magnetic field transverse to the axis of the cylinder. The experimental results show that the structure of the intermediate state, in the case of the hollow cylinder, is by no means uniform; that while the regions around the poles are in the usual intermediate state, the regions around the equator remain superconducting until the entire body goes into the normal state.

We shall show that the assumption of a homogeneous intermediate state throughout the body is in contradiction to the magnetostatic boundary conditions at the inner and outer surfaces of the cylinder. We shall also demonstrate that the observed results are in accord with these boundary conditions.

The magnetic field begins to penetrate into the bulk of our sample at applied fields of about  $0.3H_c$  instead of  $0.5H_c$  as would be expected for the cylindrical symmetry. Serin, Gittleman, and Lynton<sup>12</sup> have demon-

<sup>12</sup> Serin, Gittleman, and Lynton, following paper, *Phys. Rev.* **92**, 566 (1953).

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<sup>1</sup> D. Shoenberg, *Superconductivity* (Cambridge University Press, Cambridge, 1938), Chaps. I and II.

<sup>2</sup> W. J. de Haas and M. J. Casimir-Jonker, *Physica* **1**, 291 (1933).

<sup>3</sup> C. J. Gorter and H. G. B. Casimir, *Physica* **1**, 305 (1933).

<sup>4</sup> W. J. de Haas and O. A. Guinau, *Communs. Kamerlingh Onnes Lab. Univ. Leiden* **241a**; **247e**.

<sup>5</sup> R. Peierls, *Proc. Roy. Soc. (London)* **A155**, 613 (1936).

<sup>6</sup> F. London, *Physica* **3**, 450 (1936).

<sup>7</sup> D. Shoenberg, *Proc. Roy. Soc. (London)* **A155**, 712 (1936).

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<sup>9</sup> E. R. Andrew, *Proc. Roy. Soc. (London)* **A194**, 98 (1948).

<sup>10</sup> A. Shalnikov, *J. Phys. U.S.S.R.* **9**, 202 (1945).

<sup>11</sup> A. Meshkovski and A. Shalnikov, *J. Phys. U.S.S.R.* **11**, 1 (1947).

strated that this effect is a real one. They have shown that the value of magnetic field at which the penetration into the hollow cylinder takes place depends on the ratio of the inner diameter to the outer diameter.

## 2. EXPERIMENTAL DETAILS

The cylinder used in these experiments was made from spectroscopically pure tin (99.998 percent) supplied by Johnson-Matthey and Company. The sample was cast in a brass cylinder with a steel drill rod fitted through the center to form the inner surface. It was necessary to machine the inner and outer surfaces to obtain the proper dimensions. The cylinder was 4.79 inches long. The inside diameter was 0.580 inch and the outside diameter was 0.674 inch. No attempt was made to grow the sample into a single crystal.

A schematic representation of the arrangement inside the cryostat is shown in Fig. 1. *B* and *C* represent two sets of clamps which held the field measuring probes. The nature of these probes will be described later. A pair of leads was connected to each probe and brought up out of the cryostat through the shaft *G* to the detecting apparatus. This shaft also provided a means of

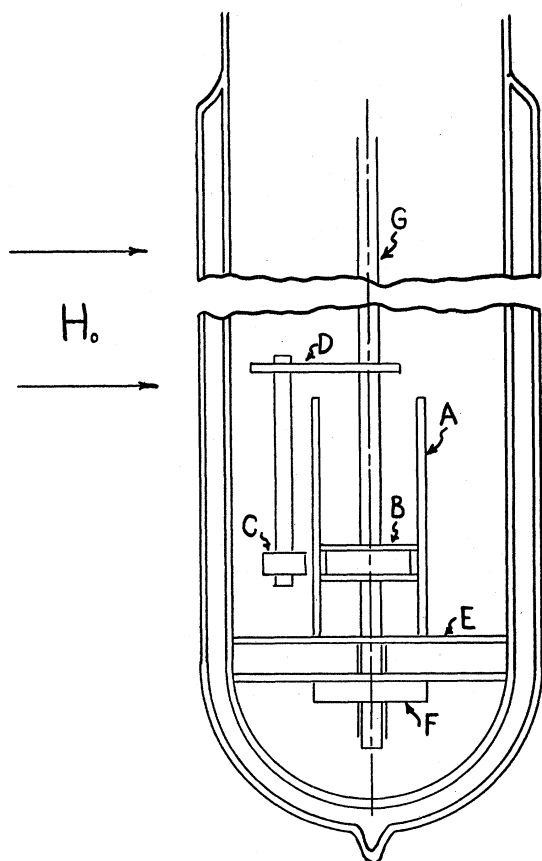


FIG. 1. The arrangement inside the cryostat. *A*=sample; *B*=inner probe; *C*=outer probe; *D*=clamp; *E*=sample mount; *F*=heater; *G*=driveshaft;  $H_0$ =applied field direction.

positioning the probes or driving them at constant speed around the sample.

The field measuring apparatus consisted of two bismuth probes, one for the inside surface of the cylinder and one for the outside surface. The effective dimensions of the probes were about 60 microns in length and 60 microns in diameter. The proper diameter was obtained by drawing the bismuth into wire while in soft glass. The glass was then etched off with hydrofluoric acid. The length was obtained in the following way. The clamps, which had the dual purpose of holding the bismuth in place and of providing electrical connections to the probes, were insulated by means of a single layer of Cellophane tape which also provided the proper spacing.

The strength of the magnetic field was obtained by measuring the electrical resistance of the bismuth during calibration of the probes in known fields, making use of the well-known fact that at low temperatures the magnetoresistance in bismuth is very large.<sup>2,4</sup>

The detecting apparatus consisted of a Wheatstone bridge which was excited at 213 cps. The output of the bridge was amplified and detected, and the detected signal was measured by a Brown recording potentiometer. Figure 2 shows a block diagram of the apparatus. With this arrangement it was possible not only to measure the magnetic field at various points as a function of the applied field; but also, for a given applied field, to make continuous measurements of field as a function of the angular position of the probes around the cylinder.

The probes were calibrated at various temperatures. The results are plotted in Fig. 3. As can be seen from the figure, the sensitivity of the probes was independent of temperature in the helium range; thus it was possible to calibrate the probes at temperatures above the critical temperature of tin.

The magnetic field was provided by a large pair of Helmholtz coils, the field at the center of the coils being 27.6 oersted/amp.

The runs can be classified into two categories, static runs and dynamic runs. The static runs were made by placing a probe at the desired location and plotting the change in the probe resistance as a function of the applied magnetic field. The applied field was varied in convenient steps from zero up to a value greater than

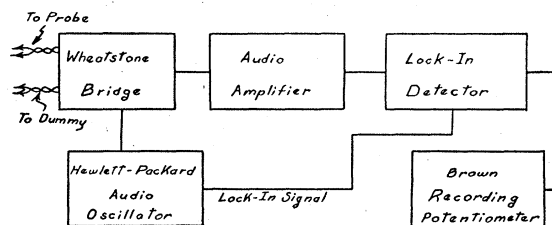


FIG. 2. The detecting apparatus.

the critical field value,  $H_c$ . Repetition of a run could not be made without first warming the sample to a temperature greater than the critical temperature because when the applied field was reduced to zero the cylinder retained a large magnetic moment. This "freezing in" of flux in multiply-connected bodies is well known.<sup>1</sup>

The procedure for the dynamic runs was the same as for the static runs with one exception. Instead of the probe being fixed at a single position, it was driven, by means of a synchronous motor, around the surface of the sample.

Since the chart of the Brown recording potentiometer is also run by a synchronous motor, we obtained in this way a plot of the magnetic field at the inner and outer

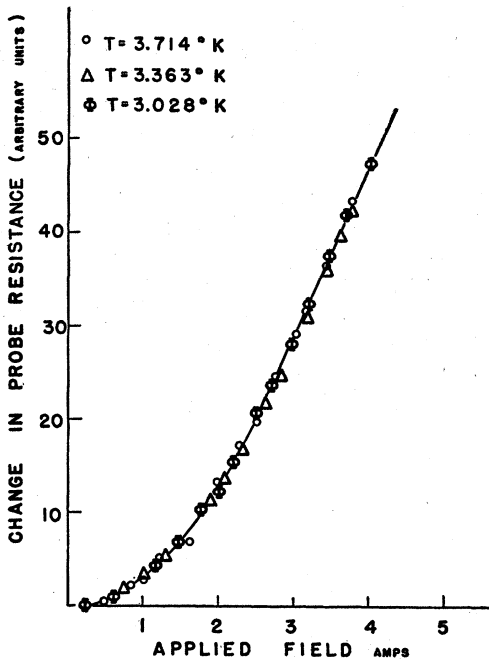


FIG. 3. Calibration of the bismuth probes at various temperatures.

surfaces as a function of the angular position of the probes.

3. EXPERIMENTAL RESULTS

Figure 4 shows typical results obtained from the dynamic runs. The superconducting state is characterized by an absence of magnetic field in the interior and a field at the outer surface whose magnitude varies as the sine of the polar angle.

When the cylinder goes into the intermediate state, the curves show that there is a broad region around the poles within which the magnitude of the magnetic field is constant and approximately equal to the applied field. We call this region the "window." For magnetic fields greater than about 1.8 amp<sup>18</sup> the angular width of this region was roughly constant and equal to about

<sup>18</sup> The critical field corresponded to 3.62 amp.

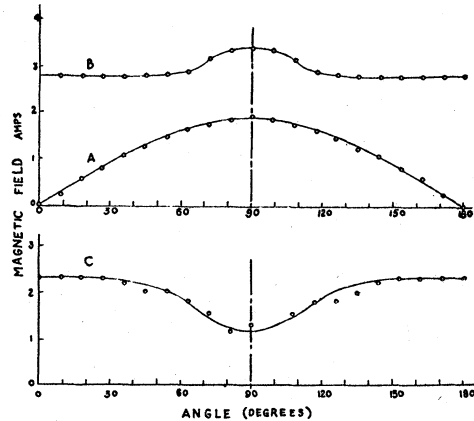


FIG. 4. Typical results of measurements of the magnetic field as a function of the angular position of the probe. A=outside probe; sample in the superconducting state. B=outside probe; sample in the intermediate state. C=inside probe; sample in the intermediate state.

110°. This corresponded quite closely to the angle subtended at the outer surface by a pair of parallel tangents to the inner surface of the cylinder.

At the outer equators the magnetic field is at all times greater than the applied field whereas, at the inner equators, the field is considerably less than the applied field. At high fields (approaching the critical value) the field at the inner equator becomes slightly greater than the applied magnetic field (see Fig. 5).

Figure 5 shows the magnetic field at the outer and inner equators as a function of the applied field. Figure 6 shows similar curves for the outer and inner poles.

The magnetic field distribution, before penetration, is clearly the same as that for a perfect diamagnet. The field at the inner and the outer poles is zero, whereas the field at the outer equator increases linearly with the applied magnetic field. The slope of this part of the curve is 1.94 rather than the expected 2.00 because the probe was located not at the surface of the cylinder but about 0.01 inch from it.

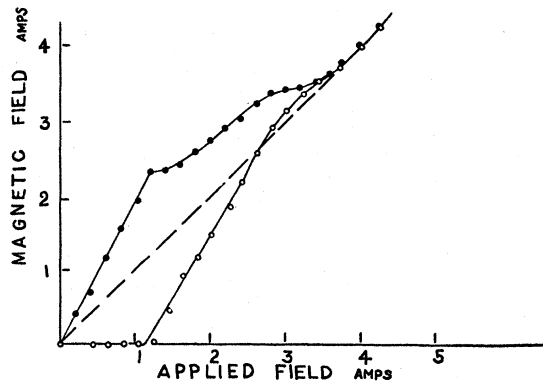


FIG. 5. The magnetic field at the equator as a function of the applied field. Open circles=inside cylinder. Solid circles=outside cylinder.

Time effects were observed throughout the intermediate state. That is to say, measurable times were required for the magnetic fields at the surfaces of the sample to stop changing when the applied field was changed. The longest observed times were of the order of ten minutes; these occurred when the applied field was in the neighborhood of those values for which penetration first begins to take place.

When the applied field was reduced in steps from values greater than  $H_c$  to zero, pronounced hysteresis was observed at all positions. In fact, hysteresis curves were obtained by cycling the applied field from  $H_c$  to  $-H_c$  and back. This state of affairs is due to the setting up of persistent currents around isolated regions of normal conductor as the applied field is reduced from  $H_c$ , thus "freezing in" flux and giving rise to a residual magnetic moment. Experimental studies of time effects and hysteresis were made by Babiskin<sup>14</sup> for the hollow sphere. In this paper we make no attempt to interpret the time effects or hysteresis but concern ourselves only with the initial transition from the virgin superconducting phase to the normal phase in increasing magnetic field.

#### 4. THE STRUCTURE OF THE INTERMEDIATE STATE

We shall now attempt to infer the structure of the intermediate state of the sample by studying the field distribution after penetration takes place. We note that when the field penetrates into the cylinder, a window is formed around the poles. The magnetic fields adjacent to the inner and outer surfaces of the window are approximately equal to the applied field. A hasty guess as to the structure of the tin composing the window would be that it is in the normal state. This is both unsatisfactory and unnecessary. It is unsatisfactory because it requires that a large amount of tin be in the normal state in the presence of a magnetic field which is much less than the critical value. That it is unnecessary can be seen in the following way. Since the wall thickness of the cylinder is small compared with the diameter,<sup>15</sup> the windows will appear to the field as a wall which is long compared to its thickness. In the case of such a wall, one obtains for the magnetic field distribution in the intermediate state:  $B = H = H_0$  outside the wall, and  $H = H_c$ ,  $B = H_0$  within the walls for  $H_0$  less than  $H_c$ ,  $H_0$  being the applied field. Thus we conclude that the structure in the window is that of the ordinary intermediate state.

There are several facts which lead us to believe that the regions around the equators, not included in the windows, remain substantially in the superconducting state until the applied field attains the value  $H_c$ . First, the field at the outside equator remains greater than the applied field for all values less than  $H_c$ . Second, in contrast to the field at the inside pole, the field at the

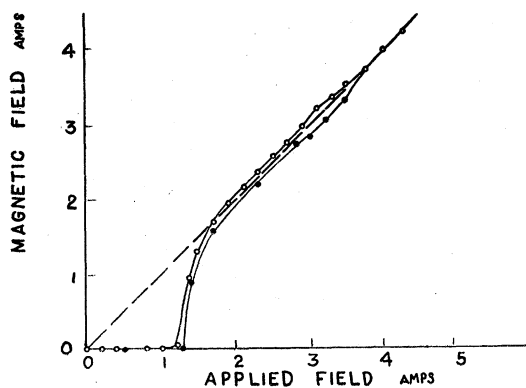


Fig. 6. The magnetic field at the pole as a function of the applied field. Open circles=inside cylinder. Solid circles=outside cylinder.

inside equators remains appreciably less than the applied field for a large fraction of the transition. This indicates that this point is being shielded by a large concentration of superconducting metal near the equator. The fact that the field eventually exceeds the applied field indicates that the superconducting region is shrinking about the equator with increasing applied field. The third indication is that the magnetic field at the outside pole is slightly less than the applied field whereas the magnetic field at the inside pole is slightly greater than the applied field. If the regions at the equators are truly superconducting, then their induced magnetic moments would contribute a field which is superimposed on the applied field. This field, equivalent to the field of two line dipoles oriented antiparallel to the applied field and situated at the equators, would add to the applied field in such a way as to account for the slight deviations at the poles.

The structure of the intermediate state of a hollow superconducting cylinder, as described above, is thus quite inhomogeneous. This result is in sharp contrast to the structures observed in uniformly magnetized solid bodies. For example,  $H$  equals  $H_c$  throughout the body of a solid superconductor in the intermediate state, and it is supposed to consist of a homogeneous mixture of superconducting and normal regions. We shall now show that such a structure is not possible for a hollow cylinder.

We shall use a two-dimensional polar coordinate system,  $r$  and  $\theta$  being the coordinates. Let  $a$  and  $b$  be the outer and inner radii of the cylinder; and let the applied field  $H_0$  be in the  $x$  direction. Then the field outside the cylinder will be equal to the applied field plus a dipole term due to the magnetization of the cylinder. Thus,

$$\begin{aligned}
 B_r = H_r &= [H_0 - M/r^2] \cos\theta, \\
 B_\theta = H_\theta &= -[H_0 + M/r^2] \sin\theta, \quad \text{for } r \geq a; \\
 B_r = H_r &= H_1 \cos\theta, \\
 B_\theta = H_\theta &= -H_1 \sin\theta, \quad \text{for } r \leq b;
 \end{aligned}$$

<sup>14</sup> J. Babiskin, Phys. Rev. 85, 104 (1952).

<sup>15</sup> The ratio of the wall thickness to the outer diameter is 0.07.

and

$$\begin{aligned} H_r &= H_c \cos\theta, \\ H_\theta &= -H_c \sin\theta, \quad \text{for } a \gg r \gg b. \end{aligned}$$

$B$  in the walls of the cylinder must be determined so that  $\text{div}B$  vanishes. Also  $H_1$  and  $M$  must be determined from the usual boundary conditions that the tangential component of  $H$  and the normal component of  $B$  be continuous on all boundaries. Applying the boundary condition for the tangential component of  $H$  on the inner boundary, we obtain  $H_1 = H_c$ . Similarly, on the outer boundary,  $-[H_0 + M/a^2] = -H_c$ , or

$$M = [H_c - H_0]a^2.$$

Applying the boundary conditions to  $B$ , we have at  $r = b$ ,  $B_r = H_c$  and at  $r = a$ ,  $B_r = 2H_0 - H_c$ .

Inside the cylinder walls  $B$  is parallel to  $H$  and thus is everywhere in the  $x$  direction. Thus, the condition  $\text{div}B = 0$  reduces to  $\partial B_x / \partial x = 0$ , and we see that  $B$  must be independent of  $x$ . This is in obvious contradiction to the boundary conditions. Along the  $x$  axis, where it is wholly normal to the boundaries,  $B$  varies from  $(2H_0 - H_c)$  on the outer boundary to  $H_c$  on the inner boundary. Thus it cannot be independent of  $x$ . The converse must also be true, that if  $\text{div}B$  vanishes, the boundary conditions cannot be met. Thus we cannot expect the intermediate state of a hollow cylinder to be the same as for a solid body.

In order to determine the proper model for the intermediate state, one must calculate the Gibbs free energy for every possible magnetostatically sound model. That model which yields the least free energy is, then, the correct one. It is, however, impossible to carry out this calculation unless the precise way in which the normal regions grow is determined. Therefore, we shall not attempt to show that the suggested model of the intermediate state of a hollow cylinder is unique, but rather that it is a reasonable one.

Let us assume that the field has penetrated into the cylinder so that the cylinder is in the intermediate state. Further, to simplify the problem, we shall put the effective permeability of the sample equal to a constant  $\mu$ . Then the solution of the field inside this cylinder in a uniform magnetic field  $H_0$  for the case in which the ratio of the inner radius to the outer radius,  $b/a$ , is of the order of unity, is

$$H \simeq H_0/\mu, \quad B \simeq H_0, \quad \text{at the poles;}$$

and

$$H \simeq H_0, \quad B \simeq \mu H_0, \quad \text{at the equator.}$$

Peierls<sup>5</sup> has determined the conditions for the existence of the intermediate state of uniformly magnetized bodies. For these bodies  $B$ ,  $H$ , and  $H_0$  are related by  $H(1-n) + Bn = H_0$ , where  $4\pi n$  is the demagnetizing coefficient. For the long cylinder in a longitudinal field,  $n=0$  and  $H=H_0$ . Peierls shows that in

this case the body remains in the superconducting state until  $H_0 = H_c$  and then becomes normal without an intermediate state. Comparing this with the observations at the equators, we see that it is reasonable that the equators remain in the superconducting state until the applied field reaches the critical value.

For a flat wall transverse to the field,  $n=1$  and  $B=H_0$ . Peierls has shown that in this case the body goes into the intermediate state for any  $H_0$ , however small, and that  $H=H_c$  inside. Here, again, comparing this with the results at the poles, we see that the model based on our observations is a reasonable one.

Thus Fig. 7 gives a rough sketch of the inferred structure. This sketch should not, of course, be taken too literally since the determination of the normal-superconducting boundaries was, of course, impossible. We must also remark that the above structure probably only appears in hollow cylinders for which the wall

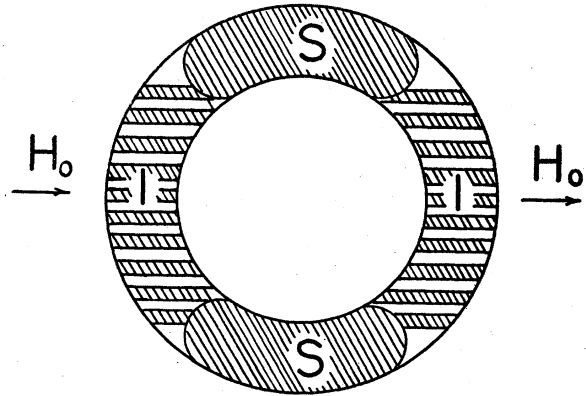


FIG. 7. The schematic representation of the inferred structure of the intermediate state of a hollow cylinder.

thickness is small compared with the diameter. As the wall gets thicker, the structure undoubtedly approaches the usual structure for a solid cylinder.

An interesting, but unexpected, result of the experiment was that the transition from the superconducting state to the intermediate state took place not when the applied field was one-half of the critical value but rather when it was approximately one-third of the critical value. This effect is discussed in the paper<sup>12</sup> following this one.

I would like to take this opportunity to thank those members of the Rutgers University Physics Department who have given me invaluable aid in this work. I would like to thank Dr. B. Serin, who suggested this problem and who advised me throughout this work, and Dr. P. R. Weiss, Dr. C. A. Reynolds, and Mr. S. Brooks for their interesting and inspiring discussions. I would also like to thank Mr. R. Campbell, Mr. J. Teza, and Mr. A. Siemons for their aid and advice in the design and construction of the apparatus.