

that power supply drift could also be responsible for a dispersion of results. Since the experimental electrodes act as a parallel plate capacitor, even a very slight voltage drift results in a displacement current in the circuit. This current easily could be of the order of magnitude of the current being measured. Consequently, it was necessary to insure a high degree of stability in the voltage source. The power supply used possessed sufficient stability so that the displacement current was always below 10^{-13} amp. This was less than one percent of the currents measured.

It has been suggested that in experiments of this kind the wall potential may influence the measured values of α/p . In this experiment the wall potential was varied in 5% steps from 45% to 100% of the anode potential and no effects on the values of α/p were found.

ACKNOWLEDGMENTS

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Moving Striations in Direct Current Glow Discharges*

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A theoretical examination of the ion balance equations for a plasma, such as the positive column of a direct current glow discharge might be, shows that a uniform plasma is not always possible. When ionization proceeds by a two-stage process involving accumulated metastable excited atoms, as may be the case in the noble gases, small perturbations of the ion concentrations from their equilibrium values may not remain small. Under such conditions, a spatially uniform steady plasma cannot exist. A general stability criterion is developed which may be used whenever accurate expressions are known for the ion and metastable production and loss rate terms. Application of the stability criterion is made using several approximations.

The instability of a spatially uniform plasma suggests that the ion concentrations may vary along the direction of current flow. The ion balance equations are re-examined for wave-like solutions.

Two approximate solutions showing some agreement with experimental observations on moving striations are obtained. The first solution, ignoring variations in the metastable concentration, yields ion and electron concentration waves traveling in the direction of the current flow with velocity probably considerably greater than the positive ion drift velocity. The amplitude of the positive ion wave is much greater than that associated with the electrons.

The second approximate solution ignores diffusion and represents a concentration wave dependent solely upon production and loss processes. It can travel in either direction, depending upon conditions in the plasma. General expressions for velocity, frequency, and wave number are given, but these cannot be evaluated numerically without better expressions for some of the quantities involved.

I. INTRODUCTION

MANY studies have been made of direct current glow discharges in the noble gases.¹ The characteristic appearance of such discharges is well known. Commonly displayed features include the cathode glow, Crookes dark space, negative glow, Faraday dark space, and a uniformly glowing positive column or plasma.

The behavior of the positive column, despite its uniform appearance, is far from simple. As Donahue and Dieke²⁻⁴ have demonstrated, moving striations are almost always present. When the positive column is observed by means of a phototube and cathode-ray oscilloscope, as was done by Donahue and Dieke, the light intensity from any small region of the positive column is seen to fluctuate rapidly. Often the fluctuation

is periodic, though not usually sinusoidal. Figure 1, taken from reference 3 or from Fig. 5 of reference 2, shows typical periodic fluctuations in the light intensity at several points in the positive column of an argon discharge at 12 mm pressure and 19.20 ma current. The tube diameter was about 13 mm, and the Faraday dark space extends to about 2.0 cm from the cathode. All the points in Fig. 1 are in the positive column, beyond the cathode region.

Points to be noted are (1) that the maximum amplitude varies little, and not in any systematic manner, and (2) that the light intensity actually becomes zero (or at least very small) during part of the cycle. The largest peak, labeled *P*, may be followed from one point in the tube to another. It is seen to appear at progressively later times as the phototube is moved toward the cathode. This peak is identified as a positive striation, since it moves in the direction of current flow (from anode to cathode). A second, smaller peak, labeled *N*, is observed to appear almost at the same time whenever it may be identified. It actually does appear slightly earlier as the cathode is approached,

* This work was supported by Office of Ordnance Research.

¹ The article by M. J. Druyvesteyn and F. M. Penning, *Revs. Modern Phys.* **12**, 87 (1940), contains many further references.

² T. M. Donahue and G. H. Dieke, *Phys. Rev.* **81**, 248 (1951).

³ T. M. Donahue and G. H. Dieke, Technical Report No. 1, The Johns Hopkins University, 1948 (unpublished).

⁴ T. M. Donahue and G. H. Dieke, Technical Report No. 3, The Johns Hopkins University, 1949 (unpublished).

indicating a rapid motion from the cathode toward the anode. Hence N is called a negative striation. The behavior at any one point is accurately periodic, with a period in this case of 4.1×10^{-4} sec. The average speed of the striation marked P is about 60 m/sec. Many more details are given in reference 2, which is generally available.

Evident conclusions are that the striations should not be regarded as small perturbations, slightly modifying a steady state, and that any theory of the positive column which ignores moving striations is certainly incomplete and probably incorrect. It is also evident that definite frequencies are generated in the discharge, essentially independent of external circuit parameters but dependent upon the actual discharge parameters themselves, such as current, pressure, and tube diameter. It is true that several modes of oscillation may exist with a given set of external circuit parameters. These modes correspond to different tube currents and voltages, so that the power supply voltage and the limiting series resistor are not without some influence. However, the behavior of any particular characteristic mode seems to depend only on internal parameters.

Probably the most serious and elaborate attempts to explain features of moving striations have been made in terms of plasma oscillations. Typically,⁵⁻¹² these attempts assume the existence of an ionized plasma (or a group of interacting ion streams, each satisfying a continuity equation) and proceed on the basis of small perturbations to arrive at a dispersion relationship for ion-density waves traveling in the medium. The actual techniques may involve finding solutions to Maxwell's equations, the Boltzmann transfer equation, the Fokker-Planck equation, or some consequence of these. In any case, the primary result is a dispersion relationship, valid for small perturbations around a steady state, but sometimes leading to the conclusion that for some frequencies a small perturbation will not necessarily remain small. Attempts to apply boundary conditions, obtain characteristic frequencies, and thus explain moving striations have not been noted for their success, although clear-cut failure is not often evident because the predictions are made in terms of electron temperature and other unobservable and perhaps meaningless parameters.

Despite lack of even qualitative success, the plasma oscillation theories have clearly shown that a highly ionized gas is capable of response to perturbations over a wide range of frequencies. By proper application of boundary conditions, inclusion of omitted considera-

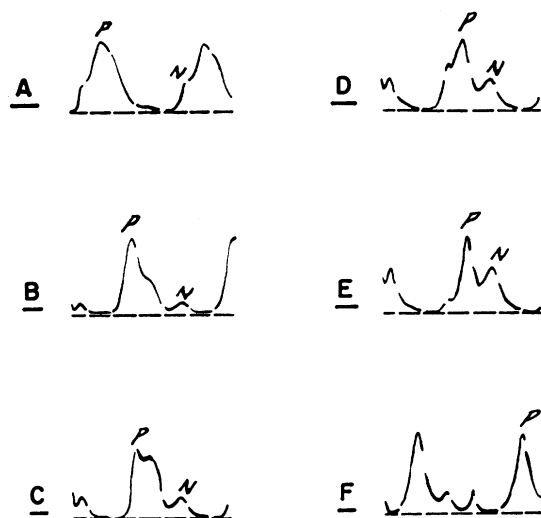


Fig. 1. Oscillograms of moving striations in argon at 12 mm pressure. The sweep markers are $100 \mu\text{sec}$ apart and the zero line is provided by an electronic switch. The primary positive striation is marked P , and a small negative striation is marked N . The Faraday dark space extends to about 2.0 cm from the cathode, and these oscillograms are taken at the following distances from the cathode: A , 4.83 cm; B , 4.05 cm; C , 3.80 cm; D , 3.67 cm; E , 3.45 cm; F , 2.53 cm. Taken from reference 3.

tions such as inelastic collisions, ion production, and ion loss, modification of the distribution functions, and a careful handling of both physical and mathematical difficulties, including those associated with a nonlinear system of equations, it should in principle be possible to understand moving striations from the point of view of plasma oscillation theory.

Because of the mathematical complexity associated with even very poor physical approximations (see, for example, references 9, 10, and 11), a different approach may be mathematically less formidable and physically closer to actual conditions in the positive column.

The usual theory of the positive column, ignoring striations, assumes that ion loss exists, either by diffusion to the walls or by volume recombination, and that the rate of production is just sufficient to offset this loss.¹³⁻¹⁶ The positive column is considered usually to be longitudinally uniform and axially symmetric. It is at this point that the present development begins.

If longitudinally uniform positive columns can exist, why should they oscillate any more than an organ pipe full of air should oscillate? Obviously some external oscillatory forcing could cause waves to propagate in the plasma, but the problem is then to identify the external source of oscillation. Loeb¹⁷ has attempted to

⁵ L. Tonks and I. Langmuir, *Phys. Rev.* **33**, 195 (1929).

⁶ D. Bohm and E. P. Gross, *Phys. Rev.* **75**, 1851 (1949).

⁷ D. Pines and D. Bohm, *Phys. Rev.* **85**, 338 (1951).

⁸ R. W. Twiss, *Phys. Rev.* **88**, 1392 (1952).

⁹ N. G. van Kampen, *Physica* **21**, 949 (1955).

¹⁰ A. A. Luchina, *Soviet Phys.* **1**, 12 (1955).

¹¹ G. Ia. Miakishev and A. A. Luchina, *Soviet Phys.* **1**, 21 (1955).

¹² H. W. Batten and H. C. Early, Technical Report No. 2, University of Michigan, 1954 (unpublished).

¹³ A. von Engel and M. Steenback, *Elektrische Gasentladungen* (Verlag Julius Springer, Berlin, 1934), Vol. 2, p. 83.

¹⁴ Reference 1, p. 158.

¹⁵ M. Hoyaux and P. Gans, Report—EOARDC-TN-54-1 Ateliers de Constructions Electriques de Charleroi (unpublished).

¹⁶ K. S. W. Champion, *Proc. Phys. Soc. (London)* **B65**, 329, 345 (1952).

¹⁷ L. B. Loeb, *Phys. Rev.* **76**, 255 (1949).

attribute to the cathode the function of originating the waves. The process he invokes seems to require a long positive column of positive resistance characteristic, which seldom exists,¹⁸ and an autocatalytic action at the cathode which results in overproduction of secondary electrons by positive ion bombardment. Since moving striations seem to be independent of cathode material and, in particular, may be easily observed in tubes with thermionic cathodes,¹⁹ Loeb's hypothesis does not seem applicable.

Another attempt to answer the question of origin was recently made by Emeleus and Daly.²⁰ They assume that ions may oscillate in a potential minimum near the cathode with a frequency proportional to the square root of the current. This dependence of frequency on current is not characteristic of moving striations, as shown by reference 2, Fig. 3, or, more clearly, by reference 4, Figs. 2 and 7. In almost every case the frequency decreases as the current increases.

The remainder of this paper will be devoted to a consideration of the ion balance equations and their consequences. It will be shown that when ion and metastable atom production and loss rates are related in a plausible way, a stable, homogeneous positive column may not exist at all, and the instability which results in moving striations may originate in the positive column itself.

II. STABILITY OF SPATIALLY UNIFORM PLASMAS

1. The Problem

The equations satisfied by ion and metastable concentrations in the positive column are modified continuity equations containing production and loss terms. If the positive column is uniform, Z dependence is eliminated. If the radial dependences are eliminated by well-known

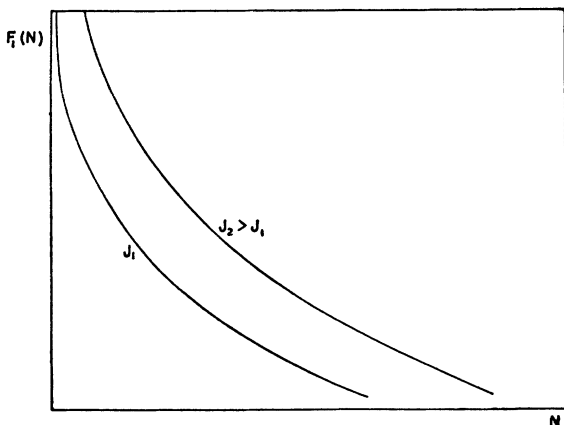


FIG. 2. The direct ionization rate as a function of ion concentration at two current densities.

¹⁸ Reference 1, p. 157.

¹⁹ Reference 2, p. 256.

²⁰ K. G. Emeleus and N. R. Daly, Proc. Phys. Soc. (London) B433, 114 (1956).

methods, the equations to be satisfied are of the form

$$\dot{Y} = G - L, \quad (1)$$

where Y is the appropriately averaged concentration of the component of interest, \dot{Y} is its time-derivative, G is the production rate of the component Y , and L is its loss rate, usually containing a term representing a radial current of Y to the tube walls. At equilibrium, $\dot{Y} = 0$.

G and L are in general functions of the ion and excited atom concentrations, the pressure, and the electric field intensity. If appropriate expressions can be written for G and L , it is then possible to reduce the number of independent variables in the positive column by imposing the equilibrium condition. It is not evident, however, that equilibrium is either stable or even a good approximation to average conditions in the plasma. It will be shown, in fact, that the solutions so obtained are not stable when metastable atoms are present in sufficient quantities.

In the remainder of this section, expressions for G and L will be examined, and a stability criterion will be applied to plasmas in which ionization occurs only by direct electron impact and to plasmas in which ionization occurs by a two-step process involving metastables. No loss of generality occurs in a *uniform* plasma if photon processes are ignored, since they could only modify the radial ion distributions and thereby not alter anything more than a numerical detail in the theory. The present state of knowledge of the G functions is not good enough to require accurate numerical details at present.

2. Electric Field Intensity

The electric field intensity in the G functions may be replaced in terms of its dependence upon discharge parameters and ion concentrations. Since the voltage drop across the positive column in typical discharge tube circuits is much less than that across the current limiting resistor plus the cathode fall voltage, the tube current may be regarded as approximately constant, independent of positive column conditions. The electric field intensity E may be written

$$E = J / [e(\mu_- N + \mu_+ P)], \quad (2)$$

where J is the current density (regarded as constant), μ_- and μ_+ are the negative and positive ion mobilities, and N and P are the negative- and positive-ion concentrations. Equation (2) is a relationship which will serve to eliminate E from the G and L functions. Its essential feature is the inverse dependence of E upon ion concentrations. As a further simplification, justified at present because the dependence of G and L is only approximately known, it is assumed that μ_- is independent of E/p_0 . This assumption, while not realistic, is probably accurate within 20% for the usual range of E/p_0 found in noble gas positive columns. Other

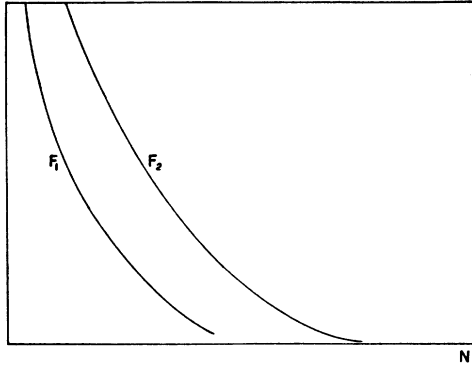


FIG. 3. Qualitative comparison of the direct ionization function $F_1(N)$ with the excitation function $F_2(N)$ at fixed pressure and current density.

approximations, such as neglect of the radial component of field strength, the assumption of constant current density, and the use of certain explicit electron energy distribution functions, are at least as unrealistic. These approximations are justifiable only as simplifications which allow preliminary calculations to be made to illustrate the concepts.

For a uniform plasma, it will be shown that $N=P$. In any case, P will probably not differ from N by more than a factor of 10, and the mobilities are weighted far more heavily than that in favor of N . Thus, for a uniform plasma, Eq. (2) becomes

$$E = p_0\theta/N, \quad (3)$$

where $p_0\theta = J/[e(\mu_- + \mu_+)]$, with θ to be considered a parameter independent of E , N , or P .

3. Balance Equations

Equation (1) for each of the components of the positive column may be written. In this analysis, in addition to N and P , a concentration of metastable excited atoms, denoted by M , is considered. This could be further subdivided, as could P , but since the present analysis is intended to illustrate method, rather than numerical detail, every reasonable simplification was used. The balance equations for N , P , and M are

$$\dot{N} = F_1(N) + MF_3(N) + \frac{1}{2}\alpha_1 M^2 - \gamma N - \alpha NP, \quad (4)$$

$$\dot{P} = F_1(N) + MF_3(N) + \frac{1}{2}\alpha_1 M^2 - \gamma P - \alpha NP, \quad (5)$$

$$\dot{M} = F_2(N) - MF_4(N) - \alpha_1 M^2 - \nu M + \beta\alpha NP. \quad (6)$$

$F_1(N)$ is a function representing the rate at which ion pairs are produced by direct electron impact. It is a function of N alone because the E dependence has been eliminated by means of Eq. (3). The use of various approximate distribution functions which may be approximately valid in the high-energy tail of the electron distribution yield the result that F_1 is a monotone decreasing function of N , as shown in Fig. 2. Also F_1 should increase with the parameter J . A

prototype function based on a method outlined by Druyvesteyn and Penning¹ is given by

$$F_1(N) = (A/N)e^{-HN}, \quad (7)$$

where A is a constant proportional to $\theta^2 p_0$ and H is a constant proportional to θ^{-1} .

$F_2(N)$ is the rate at which metastables are produced by direct electron-atom interaction. Much of the discussion of $F_1(N)$ applies to F_2 . Figure 3 shows a comparison of F_1 and F_2 at the same current density as functions of N . F_2 may also be represented by a function similar to that of Eq. (7), with a larger A and smaller H . The term $MF_3(N)$ represents the rate of ionization of metastables by electron interaction. It is proportional to M , and $F_3(N)$ is indicated by Fig. 4. $F_3(N)$ is easier to calculate accurately than are F_1 and F_2 , since the electron energies involved in ionization of metastables are not so far out in the tail of the distribution. The calculation is therefore much less sensitive to the form of the electron distribution function. A prototype function based on the Druyvesteyn distribution and the assumption of constant cross section σ gives

$$F_3 = 0.9N\sigma(\bar{\epsilon}/2m)^{\frac{1}{2}} \exp(-0.55\epsilon_1^2/\bar{\epsilon}^2), \quad (8)$$

where $\bar{\epsilon}$ is the mean electron energy and ϵ_1 is the energy of excitation. Graham and Ruhlig²¹ have calculated $\bar{\epsilon}$ for noble gases as a function of E/p_0 . For argon, this curve may be approximated by taking

$$\bar{\epsilon} = 5E/p_0, \quad (9)$$

where $\bar{\epsilon}$ is in ev and E/p_0 is in v/cm mm Hg. The use of Eqs. (3) and (9), together with the excitation energy of argon, allows Eq. (8) to be written

$$F_3(N) = BN^{\frac{1}{2}} \exp(-LN^2), \quad (10)$$

where $B = 0.9\sigma(5\theta/2m)^{\frac{1}{2}}$ and $L = 0.35/\theta^2$.

The term $MF_4(N)$ in Eq. (6) represents the loss of metastables by electron impact. F_4 should resemble F_3

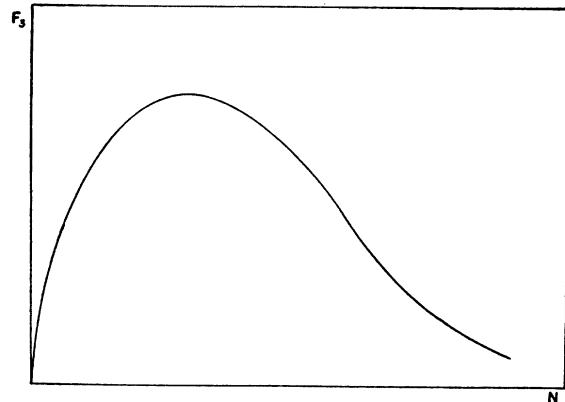


FIG. 4. The form of $F_3(N)$ as indicated by the prototype function $F_3 = BN^{\frac{1}{2}} \exp(-LN^2)$.

²¹W. J. Graham and A. J. Ruhlig, Phys. Rev. **94**, 25 (1954).

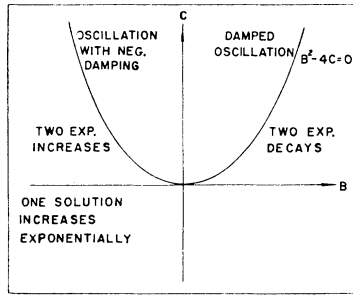


FIG. 5. Diagram showing the nature of the solutions of Eqs. (11) and (12) for Γ satisfying Eq. (12) as determined by b and c .

but always exceed in magnitude, since not all electron processes which destroy metastables will yield ions.

The $\alpha_1 M^2$ terms represent ion pair formation and consequent metastable loss as a result of the interaction of two metastable atoms with sufficient total energy to produce one ion pair and one normal atom. Biondi²² has shown that these reactions are of significance in noble gas discharges.

The terms γN , γP , and νM represent losses to the walls and, in the metastable case, deexcitation by collisions with gas atoms. The recombination terms αNP lead to a fraction β of recombined ions which become metastable.

4. Stability of Plasma Equilibrium

When $\dot{N} = \dot{P} = \dot{M} = 0$, Eqs. (4), (5), and (6) give relationships which can in principle be solved for the equilibrium concentrations M_0 , N_0 , and P_0 . One obvious result is that $N_0 = P_0$. The equilibrium solution may be such that small fluctuations around equilibrium will never increase, in which case the solution is said to be stable. If small fluctuations may lead to large departures from equilibrium, the solution is not stable.

In order that the equilibrium stability may be readily examined, assume that $N = P$, and let $N = N_0 + \delta N$, $M = M_0 + \delta M$. Expansion of the terms of Eqs. (4) and (6) about the singular point and elimination of the equilibrium conditions gives a pair of equations of the form

$$\delta \dot{N} = (\dot{N}, N)_0 \delta N + (\dot{N}, M)_0 \delta M, \quad (11)$$

$$\delta \dot{M} = (\dot{M}, N)_0 \delta N + (\dot{M}, M)_0 \delta M, \quad (12)$$

where (\dot{N}, N) has been written for $\partial \dot{N} / \partial N$, etc., and the subscript indicates that these derivatives are to be evaluated at the singular point. Solutions of the form $\delta N = (\delta N)_0 e^{\Gamma t}$ and $\delta M = (\delta M)_0 e^{\Gamma t}$ lead to the familiar consistency condition

$$\Gamma^2 + b\Gamma + c = 0, \quad (13)$$

where

$$b = -(\dot{N}, N) - (\dot{M}, M),$$

$$c = (\dot{N}, N)(\dot{M}, M) - (\dot{N}, M)(\dot{M}, N).$$

²² M. A. Biondi, Phys. Rev. **82**, 453 (1951); **88**, 660 (1952). L. B. Loeb, *Basic Processes of Gaseous Electronics* (University of California Press, Berkeley, 1955), Chap. VI.

The subscripts have been omitted since these derivatives are always to be evaluated at equilibrium. If either of the roots of Eq. (13) has a positive real part, then one of the solutions to Eqs. (11) and (12) will diverge from the singular point and equilibrium will be unstable. Figure 5 is a diagram showing the nature of the solutions to Eqs. (11) and (12) as determined by b and c in Eq. (13). Stable solutions will occur only if $b > 0$ and $c > 0$.

Substitution of the indicated derivatives in the expression for b gives

$$b = F_4 + \nu + 2\alpha_1 M + \gamma + \alpha N - F_1' - MF_3', \quad (14)$$

where $F_1' = dF_1/dN$, etc. Each term in Eq. (14) is clearly positive except the last. If equilibrium occurs for values of N such that $-MF_3'$ is sufficiently negative, b can be negative, and the plasma will be unstable. As shown by Fig. 4, the slope of F_3 is sometimes positive. That the metastable concentration may be high enough to have great influence on the plasma characteristics in noble gases has been demonstrated clearly by Meissner and Miller.²³

An interesting qualitative observation on the range of currents for which instability (and therefore perhaps striations) may exist follows from the prototype functions given by Eq. (10). Its maximum occurs for $LN^2 = 0.25$. Obviously b cannot be negative for N larger than that for which F_3 is maximum. But since for argon $LN^2 = 0.35 p_0^2 / E^2$, a limit for instability is given by $E/p_0 > 1.2$ v/cm mm Hg.

Comparison of this last result with experimental data²⁴ gives the curve of Fig. 6 for argon. This is in qualitative agreement with Pupp's curve showing maximum current for moving striations as a function of pressure.²⁵ Quantitatively, however, the lower limit on E/p_0 is too great, since moving striations occur at much lower values. Similar results may be obtained from the expression for c . Since a plasma may show instability as a whole, however, it seems plausible that localized fluctuations could cause longitudinal, or axial, instabilities even more easily than the type already examined, since axial flow of ions could take place. Numerical analysis based on the results of this section is therefore expected to indicate greater stability than actually exists.

A similar analysis for a plasma in which the metastable concentration is zero, but N and P fluctuations are allowed independently, yields the result that such a plasma is always stable.

Analysis of the three-component system produces a cubic equation corresponding to Eq. (13) in Γ . If it is written $\Gamma^3 + a\Gamma^2 + b\Gamma + c = 0$, stable solutions will exist only if a , b , and c are all non-negative and $ab - c > 0$. Thus the condition for stable equilibrium of a three-

²³ K. W. Meissner and W. F. Miller, Phys. Rev. **92**, 896 (1953).

²⁴ Reference 13, p. 110.

²⁵ Reference 1, p. 167.

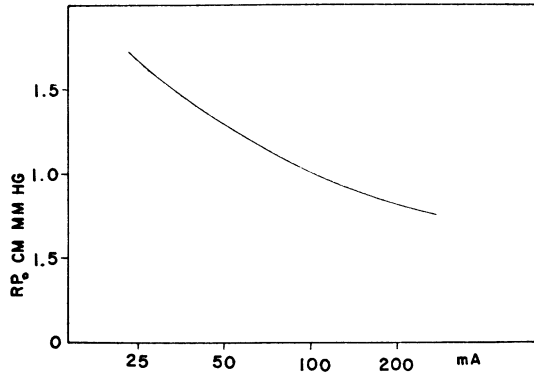


FIG. 6. Maximum current for instability of the plasma as a whole for argon as predicted by $E/p_0 > 1.2$.

component system is even more restrictive and less likely than for the two-component system.

5. Summary

In this section, analysis of the stability of a uniform plasma has been carried out for conditions of constant current density. It is shown that metastable-dependent plasmas can be unstable if the singular point in the ion and metastable rate equations occurs at sufficiently high metastable concentration and in the proper ion concentration range.

The possibility of such instability suggests that, in addition, longitudinal instability may arise from localized departures from equilibrium, and that those instabilities might be the cause of moving striations.

III. MOVING STRIATIONS

1. Method of Approach

In order to obtain wave motion, space- and time-dependent variables must be used. In this section, the basic Eq. (1) is written in much the same way as in Sec. II, except that the Z dependence is no longer ignored in the divergence of the particle currents. The radial particle currents are still considered to be included in the loss terms, but the particle currents in the Z

direction are given by

$$Nv_N = -D_- \partial N / \partial z - \mu_- NE, \quad (15)$$

$$Pv_P = -D_+ \partial P / \partial z + \mu_+ PE, \quad (16)$$

$$Mv_M = -D^* \partial M / \partial z, \quad (17)$$

where the D 's are the appropriate diffusion coefficients, the V 's are the velocities in the Z direction, and E is the effective field strength in the Z direction. Inclusion of the divergence terms in the rate equations, and the substitution $E = J e(\mu_+ P + \mu_- N)$, with J considered constant, gives

$$\dot{N} = D_- \frac{\partial^2 N}{\partial Z^2} + \frac{\mu_+ \mu_- J \left(P \frac{\partial N}{\partial Z} - N \frac{\partial P}{\partial Z} \right)}{e(\mu_+ P + \mu_- N)^2} + F_1 + MF_3 + \frac{1}{2} \alpha_1 M^2 - \gamma N - \alpha NP, \quad (18)$$

$$\dot{P} = D_+ \frac{\partial^2 P}{\partial Z^2} + \frac{\mu_+ \mu_- J \left(P \frac{\partial N}{\partial Z} - N \frac{\partial P}{\partial Z} \right)}{e(\mu_+ P + \mu_- N)^2} + F_1 + MF_3 + \alpha_1 M^2 - \gamma P - \alpha NP, \quad (19)$$

$$\dot{M} = D^* \frac{\partial^2 M}{\partial Z^2} + F_2 + \beta \alpha NP - \alpha_1 M^2 - MF_4 - \nu M. \quad (20)$$

Let $N = N_0 + \delta N$; $P = P_0 + \delta P$; $M = M_0 + \delta M$, where the equilibrium values, considered to be independent of time and space, are denoted by the subscript 0 and the departures from equilibrium are considered small. Since

$$\mu_+ \mu_- J / e N_0 (\mu_+ + \mu_-)^2 = \mu_+ \mu_- E_0 / (\mu_+ + \mu_-),$$

which is approximately equal to $\mu_+ E_0$, the average positive ion drift velocity, it is reasonable to let

$$\mu_+ \mu_- J / e N_0 (\mu_+ + \mu_-)^2 = V_+. \quad (21)$$

Also at equilibrium, $N_0 = P_0$. Substitution of these values into Eqs. (18), (19), and (20), and subsequent substitutions of the type $\delta N = \delta N_0 \exp(\Gamma t + Kz)$, leads in a routine manner to the consistency determinant

$$\begin{vmatrix} K^2 D_- - \Gamma + K V_+ + (\dot{N}, N) & (\dot{N}, P) - K V_+ & (\dot{N}, M) \\ (\dot{P}, N) + K V_+ & K^2 D_+ - \Gamma - K V_+ + (\dot{P}, P) & (\dot{P}, M) \\ (\dot{M}, N) & (\dot{M}, P) & (\dot{M}, M) + K^2 D^* - \Gamma \end{vmatrix} = 0. \quad (22)$$

The derivatives in parentheses are identical to those given in Sec. II, and are not modified by consideration of space dependence since the equilibrium condition is taken to be independent of Z . These derivatives should be evaluated at equilibrium, and therefore should be designated with the 0 subscript. However, since they always appear in this development evaluated at the equilibrium point, the subscript is redundant and will be omitted.

Equation (22) is cubic in Γ and of sixth degree in K . No attempt in this paper will be made to solve Eq. (22) as such. Approximate and simplified solutions will be obtained, however, and these will be compared to experimental data.

2. Simplification of the Basic Equation

The parameters Γ and K in Eq. (22) may be complex. The observations on moving striations indicate, as

pointed out in the introduction, that no exponential growth or decay of the striation in either space or time is experienced. There is an increase in amplitude of positive striations as they enter the cathodic region of the discharge, but this can be explained in terms of significant departures from plasma conditions.

Probably Eq. (22) is satisfied by some waves which increase exponentially, but physical reasoning requires that the solutions be bounded. Hence the physically meaningful approximation will be made that Γ and K must be pure imaginaries. It is then expected that the consequences of solutions to Eq. (22) will resemble reality in much the same way as the solutions to the ordinary acoustic wave equation resemble conditions in a high-amplitude shock wave. Where sine waves are predicted, saw-toothed waves may exist, and where a constant velocity is predicted, an amplitude-dependent one may be found. The assumption that K and Γ are pure imaginaries so greatly enhances the availability of the contents of Eq. (22) that the resultant loss of generality is overbalanced in the present state of the theory.

If

$$\Gamma = i\omega \quad \text{and} \quad K = -ik, \quad (23)$$

$$\begin{vmatrix} -k^2 D_- - i\omega - ikV_+ + (\dot{N}, N) & (\dot{N}, P) + ikV_+ \\ (\dot{P}, N) - ikV_+ & -k^2 D_+ - i\omega + ikV_+ + (\dot{P}, P) \end{vmatrix} = 0. \quad (24)$$

This is equivalent to the assumption that the metastables and normal atoms form a two-component gas, each of which is so populous that no rate equation need be written for either the metastable gas or the normal gas. Ions and electrons can be made from both gases, and rate equations are then written for only ions and electrons.

Separation of Eq. (24) into real and imaginary parts gives

$$\omega^2 = k^4(D_- D_+) - k^2[D_+(\dot{N}, N) + D_-(\dot{P}, P)] + (\dot{N}, N)(\dot{P}, P) - (\dot{N}, P)(\dot{P}, N), \quad (25)$$

and

$$\omega = \frac{k^3 V_+(D_- - D_+) + kV_+[(\dot{P}, P) - (\dot{N}, N) + (\dot{P}, N) - (\dot{N}, P)]}{k^2(D_- + D_+) - (\dot{N}, N) - (\dot{P}, P)}. \quad (26)$$

The coefficient of kV_+ in Eq. (26) is zero.

Before these equations are combined to yield a sixth degree equation for k , an elementary solution may be examined.

3. Simplest Striation

If all production and loss derivatives are ignored, Eqs. (25) and (26) become

$$\omega^2 = (D_- D_+) k^4, \quad (27)$$

and

$$\omega = kV_+(D_- - D_+)/ (D_- + D_+), \quad (28)$$

where ω and k are assumed to be real, Eq. (22) may be separated into real and imaginary parts. Each part is a relationship between ω and k , and these must be simultaneously valid. Elimination of ω between them gives rise to an equation of 18th degree in k . For each of the eighteen roots of this equation, a unique ω exists, so it is possible for Eq. (22) to lead to eighteen frequencies and wave numbers which may be present. Probably not all of the roots of the k equation are real, and those that are not are meaningless, since the separation of Eq. (22) into real and imaginary parts was made under the assumption that k and ω are real. Analysis of the coefficients in the k equation could be made to determine the number of real roots as a function of the discharge parameters. The presence on occasion of six or eight distinct waves in a complete striation, the observation of several modes of oscillation, and the transition from one mode to another²⁻⁴ are at this point not surprising.

Since the 18th degree equation in k is still beyond the reach of practicality, a further simplification will be made. Assume that $\delta M = 0$, so that the equivalent of Eq. (22) becomes 2×2 determinant,

The phase velocity is $\omega/k = V_+$, approximately, since usually $D_- \gg D_+$. To this same approximation,

$$k = V_+(D_- D_+)^{-\frac{1}{2}} = \mu_+ E_0 (D_- D_+)^{-\frac{1}{2}}, \quad (29)$$

and

$$\omega = V_+^2 (D_- D_+)^{-\frac{1}{2}} = (\mu_+ E_0)^2 (D_- D_+)^{-\frac{1}{2}}. \quad (30)$$

The phase velocity is thus seen to be approximately the positive-ion drift velocity, and in the direction of the current flow (i.e., toward the cathode).²⁶

This is a poor approximation for many obvious reasons. It is therefore not surprising that the striation velocity as given by this approximation is much too low. Comparison of the results of Donahue and Dieke⁴ with the published data on the average field conditions²⁴ and mobilities²⁷ indicates that the actual striation velocities are on the order of thirty times the values obtained here. The velocities and frequencies behave qualitatively in the manner suggested by this approximation, remaining nearly constant at a given current for a wide range of pressures which leaves E/p_0 also nearly constant. At sufficiently low pressure E/p_0 rises rapidly as the pressure is reduced, and so do the velocity and frequency of the striations. At higher currents, E/p_0 is reduced and so are frequency and velocity. The wavelength, however, as obtained from

²⁶ This result is similar to that reported by S. Watanabe and N. L. Oleson, Phys. Rev. **99**, 1701 (1955). Their approach to the entire problem differs in many ways from this one, particularly in the treatment of ion production and loss.

²⁷ J. A. Hornbeck, Phys. Rev. **84**, 615 (1951).

Eq. (29), does not agree qualitatively with observation, but rather k itself behaves in the same manner as Donahue and Dieke find λ to behave.

Two obvious criticisms and one comment should be made at this point. First, if ion production and loss play an important part in the striation mechanism, it is certainly not correct to ignore all pertinent terms in Eqs. (25) and (26). Second, as pointed out in the introduction and elsewhere, no one should expect to explain moving striations in terms of a small perturbation theory. The large amplitudes observed experimentally, as indicated in this section, suggest by analogy to shock waves that departures from a simplified theory might well be in the direction of higher velocities. Donahue and Dieke² have mentioned that the striation velocities are greatest where the striation is brightest. It should be brightest where excitation, and field strength, are greatest. At high field strengths, the drift velocity of the positive ions will be greater than the value computed for the average field strength, and this effect alone will account for much of the discrepancy between experiment and theory. For example, Donahue and Dieke report voltage fluctuations of the order of ten volts, associated with striation motion on the order of one centimeter. This would suggest field strengths associated with the striations of 10 v/cm, which is on the order of ten times the average field strength.

The final comment before continuation of the calculation is that the results so far yield definite frequencies and wave numbers, rather than merely dispersion relationships. These characteristic frequencies and wave numbers are properties of the plasma itself and are not dependent upon boundary conditions. This is in agreement with the experimental result that striation properties are not dependent upon tube length, provided the tube is long enough, or upon external circuit parameters or electrode materials.

Equation (28) may be put back in one of the relationships between δN_0 and δP_0 to give, if $\delta P = \delta P_0 \exp[i(\omega t - kz)]$,

$$\delta N_0 = i\delta P_0 / (D_-^{1/2} D_+^{-1/2} + 2i) \quad (31)$$

or approximately

$$\delta N_0 = i(D_+/D_-)^{1/2} \delta P_0, \quad (32)$$

since $D_- \gg D_+$. This indicates that the amplitude of the negative-ion fluctuation is considerably less than the positive-ion fluctuation, and that the negative-ion maximum leads the positive-ion maximum by approximately a quarter-wavelength as they travel down the tube. This is typical of the striation behavior described by Donahue and Dieke, where the positive striation seems to draw electrons toward it as it moves toward the cathode.

4. Effect of Production and Loss Terms

Figure 7(A) shows a plot of Eq. (25), labeled R , and Eq. (26), labeled I , in the first quadrant, with the rate

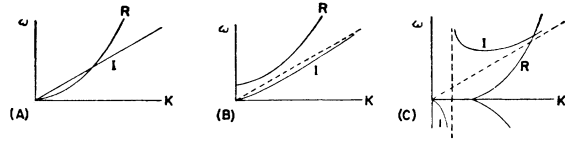


FIG. 7. Comparison of Eqs. (25) and (26) (A) when rate terms are ignored, (B) when metastables are unimportant, and (C) when the metastable term MF_3' is sufficiently positive to cause instability.

terms set equal to zero. The intersection of these curves corresponds to the simple solution in Sec. III3. The inclusion of the rate terms can result in two possible types of change. First, if $(\dot{N}, N) + (\dot{P}, P)$ is negative and $(\dot{N}, N)(\dot{P}, P) - (\dot{N}, P)(\dot{P}, N)$ is positive, as will be the case when the MF_3' term is either unimportant or negative, curve R will be shifted upwards and curve I shifted downward, as indicated in Fig. 7(B). [For small contributions from the rate terms, the curves may intersect and a solution may therefore be possible, but certainly if these terms contribute heavily, the curves in Fig. 7(B) are representative of the change.] A point of intersection is not possible in the curves as they are drawn, and therefore no real values of k and ω will simultaneously satisfy Eqs. (25) and (26).

If the metastable contribution MF_3' is sufficiently positive, however, significant changes in the other direction will result. Figure 7(C) shows how curve I is greatly modified and curve R is shifted downward so that the intersection will always exist, and furthermore it will occur at higher values of ω and k than in the case of Fig. 7(A). The phase velocity ω/k will also be increased in this case, since for a given k , ω is obviously greater than its value on the line I in Fig. 7(A). Slight modification of the drawing will show that considerably increased values of the phase velocity are possible, particularly if the intersection occurs for values of k^2 only slightly larger than $[(\dot{P}, P) + (\dot{N}, N)] / (D_- + D_+)$.

The increase in k and ω/k caused by the metastable production rate is seen to reduce the ratio

$$\delta N_0 / \delta P_0 = iV_+ / [kD_- + i(V_+ + \omega/k)]. \quad (33)$$

The net effect, then, of metastable participation of the type which leads to the instabilities of Sec. II is to increase frequency and phase velocity, while reducing the amplitude of the negative-ion fluctuation.

5. Striations Without Diffusion

Considerable simplification can be made in Eq. (22) if all diffusion coefficients are set equal to zero. There is no real justification for choosing one and not another, either to be zero or to differ from zero, even though D_- is certainly much greater than D_+ or D^* . The reason is that Sec. III3 has already indicated that the amplitude of δP is probably considerably greater than that of δN and no estimate of δM_0 has been made. The amplitude differences are largely ascribable to diffusion or lack of it, and therefore the selection of a single diffusion term

to remain probably contributes error as well as complication.

The substitutions $K = -ik$, $\Gamma = i\omega$, $D_- = D_+ = D^* = 0$ in Eq. (22), with consequent separation into real and imaginary parts, give

$$\omega^2 = \Delta / \text{Tr}\Delta, \quad (34)$$

and

$$\begin{aligned} \omega^3 - \omega [& (\dot{M}, M)(\dot{P}, P) - (\dot{P}, M)(\dot{M}, P) + (\dot{N}, N)(\dot{P}, P) \\ & - (\dot{N}, P)(\dot{P}, N) + (\dot{N}, N)(\dot{M}, M) + (\dot{N}, M)(\dot{M}, N)] \\ & + kV_+ [(\dot{N}, N)(\dot{M}, M) - (\dot{N}, M)(\dot{M}, N) \\ & - (\dot{P}, P)(\dot{M}, M) + (\dot{P}, M)(\dot{M}, P) \\ & + (\dot{M}, M)(\dot{N}, P) - (\dot{M}, M)(\dot{P}, N)] = 0, \end{aligned} \quad (35)$$

where

$$\Delta = \begin{vmatrix} (\dot{N}, N) & (\dot{N}, P) & (\dot{N}, M) \\ (\dot{P}, N) & (\dot{P}, P) & (\dot{P}, M) \\ (\dot{M}, N) & (\dot{M}, P) & (\dot{M}, M) \end{vmatrix} \quad (36)$$

and $\text{Tr}\Delta$ is the trace of Δ , or

$$\text{Tr}\Delta = (\dot{N}, N) + (\dot{P}, P) + (\dot{M}, M). \quad (37)$$

Equation (34) gives ω directly, and this combined with Eq. (35) gives the phase velocity. The frequency here is determined entirely by rate terms, and is given by

$$\omega^2 = \frac{\gamma [(F_4 + 2\alpha_1 M + \nu)(F_1' + MF_3' - \gamma - \alpha N) + (F_3 + \alpha_1 M)(F_2' - MF_2' - \alpha \beta N)]}{F_1' + MF_3' - 2\gamma - \alpha N - \nu - 2\alpha_1 M - F_4}. \quad (38)$$

The frequency so obtained is clearly different from any of the plasma frequencies⁵⁻¹² derived in the literature. Since it is dependent upon poorly defined functions, numerical predictions are not possible with any accuracy. For a given geometry, gas and pressure, it should be not difficult to obtain values for γ , α , ν , and α_1 . But it is still necessary to put in specific functions in order to obtain a numerical answer.

The sign of neither numerator nor denominator in Eq. (38) is clearly determinate, so without further analysis the reality of ω is in doubt.

As an approximation, suppose that $\alpha = \nu = \alpha_1 = F_1 = 0$ and

$$F_2 = (Q/N) \exp(-HN), \quad F_3 = BN^{\frac{1}{2}} \exp(-LN^2)$$

as suggested by Eqs. (7) and (10). The rate equations at equilibrium yield

$$MF_3 = \gamma N = F_2. \quad (39)$$

Substitution of these relationships and their consequences in Eqs. (38) and (35) yield

$$\omega^2 \gamma^{-2} = 2NM^{-1}(HN + 2)(1 + 4LN^2 + 2NM^{-1})^{-2}, \quad (40)$$

$$\omega^2 \gamma^{-2} = NM^{-1}(HN + 3) + \frac{1}{2} + 2LN^2 + kV_+ \omega^{-1} NM^{-1}(HN + 2LN^2 + \frac{1}{2}). \quad (41)$$

Thus Eq. (40) shows that ω is real, since every letter is chosen to be intrinsically non-negative. Equation (41) alone allows all positive values of kV_+/ω ; i.e., it

forbids no wave propagating in the direction of the positive ion drift velocity. It allows waves to propagate in the direction opposite to V_+ only if $|kV_+/\omega|$ is small enough to keep the right hand side of Eq. (41) from becoming negative. This acts as a lower limit to the phase velocity of waves traveling toward the anode, and may well be related to the fast "negative striations" observed by Donahue and Dieke.

The actual phase velocity predicted from Eqs. (40) and (41) may be either positive or negative, depending upon what is required of the kV_+/ω term to satisfy equality of the ω^2 values in the two equations. Numerical analyses have not been made because the theory appears not to be in a state to yield satisfactory numbers.

6. Discussion

Two approximations to the solution of Eq. (22) were made. Both were based on the assumption that $k = iK$ and $\omega = -i\Gamma$ are real. It is believed that this assumption is not at variance physically with the assumption of small perturbations. Further, the inclusion of a real part in K (or imaginary in k), leading to attenuation or amplification of the wave as it travels along the tube, results in either large perturbations or no perturbations. Therefore a correct theory, in which the actual nonlinear equations are solved, would probably show alternate amplification and attenuation, together with variations in velocity, as the point representing the concentrations moves along its trajectory in N, P, M space. It is obvious that the definite frequencies, wave numbers, and phase velocities predictable from this development are a consequence of the conversion of Eq. (22) into two separate equations in two unknowns. That there is a consequent loss of generality not already inherent in the small perturbation approximation is not obvious. A careful study of this point should be made.

The first approximation considered the metastable atoms to be so numerous that they may be treated as an admixed gas without the need to account for production or loss. The electron and ion concentrations then exhibit wave-like behavior with the concentration waves moving in the direction of the current flow. The positive ion amplitude is much greater than the electron concentration, and the electron wave leads the ion wave by approximately a quarter wavelength. The field intensity should therefore be maximum only slightly less than a quarter wavelength ahead of the positive-ion maximum, and excitation and metastable production should be greatest in this high-field region. This result is in agreement with the observation of Donahue and Dieke,²⁸ that the metastable excitation maximum leads the main striation as they travel toward the cathode. A calculation by Farris²⁹ also shows a region of high electric field strength (he represents this as electron temperature) traveling so as to represent an ionization wave.

²⁸ Reference 2, Sec. XI.

²⁹ V. D. Farris, Proc. Phys. Soc. (London) **B68**, 383 (1955).

In this first approximation, diffusion is important and variations in metastable concentration were ignored. The rate terms were found to be important in just the way suggested by Sec. II. The basic diffusion-type wave is slowed down, the frequency is reduced, and the wavelength is increased by rate terms which would cause the uniform plasma to be stable. If the rate terms of this type are of sufficient importance, no periodic behavior or wave motion of this type is possible. On the other hand, rate terms of the type which lead to instability of a uniform plasma also lead to higher phase velocity, higher frequency, and shorter wavelength, predictions which are in qualitative agreement with experiment.

The second approximate solution considered variations in all three concentrations but ignored diffusion. The wavelength, frequency, and phase velocity so obtained are dependent entirely on the rate terms. It was shown that use of the prototype rate terms leads to a real frequency. The wave number and phase velocity may be either positive or negative, depending upon the rate terms. A lower limit exists to the speeds

allowable by striations moving in the direction opposite to the current flow.

Inclusion of all possibilities results in equations which have not yet been solved. Preliminary successes, however, are sufficiently promising to warrant further study based on the mechanism suggested.

The most important weaknesses in this theory of striations appears to be (1) the use of a small-perturbation theory and (2) lack of reliable expressions for the ionization and excitation rates. Less important deficiencies include (3) the approximation of constant current density, (4) the failure to include a P dependence (which affects the field strength) in the ionization and excitation rate terms, (5) the use of first-order diffusion theory, (6) the use of ambipolar type diffusion to describe ion and electron losses even when the concentrations differ, (7) the use of the loss term γN even when the concentration profile is probably not the J_0 Bessel function, and (8) neglect of the many possible components that may be present with sufficiently long lifetimes to be of significance, such as molecular ions and several species of metastables.

Ferromagnetic Resonance in Thin Films of Permalloy*

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Microwave resonance measurements have been made in evaporated (82% Ni, 18% Fe) films of thickness 760 Å to 1600 Å. The longest relaxation, as measured by the line width, gives $T_2 = 3 \times 10^{-8}$ sec. The sample thickness was equal to or less than the skin depth, resulting in a Lorentzian-type line. Multiple resonances were obtained according to shape anisotropy theory.

THE original discovery of ferromagnetic resonance¹ was made in thin films of iron, nickel, and cobalt. Subsequent resonance experiments were mostly confined to bulk metals and the ferrites. Renewed interest has recently been shown in the magnetic properties of ferromagnetic films because of their likely single-domain structure and their consequent single-domain rotation magnetization process.²⁻⁴ Evidence has been presented that the spin reversal time by domain rotation may be faster than 10^{-9} sec.⁵ An ultimate limit must be set by damping of the motion. We have started an investigation of the damping as well as other features of the resonance at microwave frequencies.

The films, (82% Ni, 18% Fe) evaporated onto microscope slides, varied in thickness from 760 Å to

1600 Å. The microwave experiments were carried out using standard, well-known techniques.

Damping, as measured by the line width, turns out to be much smaller than previously observed in ferromagnetic metals or alloys. The longest relaxation time we observed occurred at 2800 Mc/sec with $T_2 = 3 \times 10^{-8}$ sec, where the magnetic field H was applied perpendicular to the plane of the film.

With H making other angles with the plane of the film at 9000 Mc/sec, and at temperatures down to 4.2°K, the full line width ΔH varies in no regular fashion. Some selected resonance curves are shown in Fig. 1. In all cases the lines had more of a Lorentzian shape than $\sqrt{\mu_R}$ shape, evidence of the fact that the sample thickness was approximately equal to or less than the skin depth on resonance. The appropriate electromagnetic calculations show that in this range line broadening with respect to the μ'' line is in the vicinity of 1.5. For a thick sample $(\Delta H)_{\sqrt{\mu_R}}/(\Delta H)_{\mu''} = 2.5$, and the line would have a characteristic $\sqrt{\mu_R}$ shape.

From parallel and perpendicular orientations with respect to H , the magnetization $4\pi M$ and the g factor

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