

Experimental Study of the Diamagnetism of Gaseous Plasmas with Electron and Nuclear Spin Resonance Techniques*†

T. C. MARSHALL‡ AND L. GOLDSTEIN
University of Illinois, Urbana, Illinois

(Received December 15, 1960)

It has been possible to observe the occurrence of diamagnetism in active discharges by measuring shifts in spin absorption resonance frequencies of foreign substances located near the plasma. Narrow spin electronic resonances in DPPH have been used to show that the magnetization of a gaseous discharge in low-pressure mercury vapor increases linearly with applied magnetic field up to about 40 gauss, where maximization occurs. Observation of the phase change in the Larmor precession of protons in a strong homogeneous field showed that the diamagnetism in a modified Penning ionization gauge (P.I.G.) discharge increased linearly with power input and decreased approximately as $1/H$. Irradiation of the plasma with high-power microwaves with frequency near the free-electron gyrofrequency resulted in an increase of diamagnetism of the discharge. The discharge magnetic moment ranged from -0.001 to -0.32 erg/cm³-gauss, depending on experimental conditions. The sensitivity of the nuclear resonance technique is one part in 10^6 , and it permits observation of diamagnetism to be deferred until the discharge is over. The theory of plasma diamagnetism is summarized.

I. INTRODUCTION

WE report here the results of an experimental study of the diamagnetism of gaseous plasmas immersed in magnetic fields. This diamagnetism arises principally from the electronic component of the plasma, the ionic contribution being negligible except under special high-energy conditions. In a brief survey of the theories on plasma diamagnetism, we shall show that the energy density of the plasma may be inferred from experimental data. The purpose of this work is therefore to gain further understanding of the plasma-medium and to establish the potentialities of plasma diamagnetism experiments for diagnostic means as well.

Although the individual free electrons of either a metal or a gaseous plasma immersed in a magnetic field have orbital magnetic moments given by

$$M_z = -mV_{\perp}^2/2H \equiv -W_{\perp}/H, \quad (1)$$

where $V_{\perp}^2 = V_x^2 + V_y^2$, W_{\perp} is the transverse electronic kinetic energy, and H , the magnetic field, is directed along the z axis, it is incorrect to conclude that a macroscopic ensemble of n electrons per cm³ in a metal or a plasma must possess a moment n times as large as that in Eq. (1). Bohr¹ argued that the large magnetic moment of the free electron gas in the interior of a metal which was then thought to be classical $-nkT_e/H$, $\langle W_{\perp} \rangle = kT_e$ (T_e = electron temperature), was exactly canceled by a reversed surface electronic current formed by those electrons which make elastic impacts with the boundary potential of the metal. On this account, Bohr reasoned that the moment of such a system must be zero. Van Leeuwen² reached the same conclusion, by

noting that the energy of a system of charges is unchanged by a static magnetic field. Then, using the partition function Z , for a Maxwell-Boltzmann distribution

$$Z = \sum_i e^{-W_i/kT}, \quad (2)$$

where the energies of each of the i states of the system are W_i , one finds

$$M_z = nkT \frac{\partial}{\partial H} (\ln Z) = 0. \quad (3)$$

The assumption of thermal equilibrium allows one to disregard the detailed behavior of the electron trajectories near the boundary, since the walls must then act like more matter. The absence of a magnetic moment in such an "infinite" sample arises from the microscopic cancellation of all the random free electronic currents in the material.

Landau,³ who recognized that the magnetic field provides an axis of quantization for electronic motion, rejected the above classical approach. He found that the transverse electronic energy, W_{\perp} , depended on H and could assume only the "harmonic-oscillator" values

$$W_{\perp}(p) = \frac{e\hbar}{2mc} (2p+1)H, \quad p=0, 1, 2, \dots \quad (4)$$

Landau's equilibrium calculations predicted a small moment per electron in the metal compared with Eq. (1). One may regard the small "quantum-mechanical" moment which he calculated as a consequence of the uncertainty principle, which requires that the cancellation of the "volume" and "surface" currents of charge angular momentum can be exact only to $\approx \hbar$.

To treat plasma diamagnetism, we must further dispense with the restriction of thermal equilibrium; this complicates the theory in certain aspects which will be summarized below.

³ L. Landau, *Z. Physik* **64**, 629, (1930).

* Work supported by Air Force Cambridge Research Center.

† This article is based on a thesis submitted to the Graduate College of the University of Illinois by T. C. Marshall in partial fulfillment of the requirements for the degree of Doctor of Philosophy.

‡ National Science Foundation Predoctoral Fellow.

¹ N. Bohr, dissertation, Copenhagen, 1911.

² J. H. Van Leeuwen, dissertation, Leiden, 1919. See also J. phys. radium, **2**, 361 (1921).

II. BRIEF REVIEW OF THE THEORY OF THE MAGNETIC MOMENT OF A GASEOUS PLASMA

While several theories⁴⁻⁷ about the magnetic moment of plasmas have been proposed, experimental data have been too scarce to allow a satisfactory check to be made. The first successful experiments were conducted by Steenbeck, whose results agree with part of the data collected by the authors. However, Steenbeck's theory is questionable and would appear to contradict van Leeuwen's theorem.

Most theories agree that a plasma in a strong magnetic field must have a diamagnetic moment which approaches $-nkT_e/H$ as H grows large. A good derivation of this has been presented by Alfvén. Suppose the plasma is confined in a discharge tube in a magnetic field; if the walls of the tube are removed suddenly, the magnetostatic pressure inside the plasma is $(1+4\pi\chi)H^2/4\pi$ and $H^2/4\pi$ outside, where χ is the plasma susceptibility. Neglecting dynamical effects, which is valid if particle collisions are infrequent, the balance between the magnetostatic pressure and the gas kinetic pressure of the electrons and ions gives

$$-\chi H^2 = (n/3)(\langle mV^2 \rangle_i + \langle mv^2 \rangle_e) = nk(T_e + T_i), \quad (5)$$

which is the desired result. We have taken $n=n_e=n_i$.

Another approach to the "diffusionless" plasma may be made using the quantum mechanical treatment outlined by Van Vleck⁸ and Teller⁹ for metals. If diffusion is absent in a plasma, then the effective radial electric potential $\varphi(r)$ arising from the density gradients and space charge fields caused by diffusion does not enter into the particle Hamiltonian, and Eq. (4) correctly gives the energy of the electrons. Thus the moment per particle is

$$M(p) = -\frac{\partial W_1(p)}{\partial H} = -\frac{e\hbar}{2mc}(2p+1) = -\frac{W_1(p)}{H}. \quad (6)$$

Van Vleck showed that it was possible to break up the average moment for the electronic system into separate volume and surface contributions. Summation over p and averaging in the equilibrium case gives results identical with Landau's treatment in the calculation for metals. However, collisions of electrons with walls in a gaseous plasma are absorbing rather than reflecting, hence the charge concentration is in general very low near the plasma surface; furthermore, unlike a metal, both electrons and ions can move tangentially

in whatever radial static electric field exists in the boundary sheath, and thus the net magnetic moment of such motion is zero to the extent that collisions are infrequent [that is, we require $(\nu/\omega_H)_e \ll 1$, where ν is the momentum-transfer collision frequency and ω_H is the cyclotron frequency of the electrons (e) and ions (i), respectively]. Thus the surface current is suppressed, and

$$\langle M_z \rangle = - \int_0^\infty \frac{n}{H} W_{ei}(p) f_e(p) dp - \int_0^\infty \frac{n}{H} W_{ii}(p) f_i(p) dp, \quad (7)$$

where f is the normalized distribution function for either ions or electrons, and integrals have been employed because of the close spacing of the Landau energy levels. In our experiments, $\langle W_{ei} \rangle \gg \langle W_{ii} \rangle$. The advantage of the quantum treatment lies in the fact that the energy of an orbiting charge does in fact depend on H ; this clarifies some of the misconceptions of "classical" arguments.

Alternatively, it is easy to show that owing to a radial gradient in electronic concentration and/or temperature throughout the plasma, the microscopic azimuthal currents of the particles no longer exactly cancel at every point in space (as with Bohr's model), but instead gradually accumulate over the radius to yield a net diamagnetic susceptibility given by (5).

A calculation of the moment where radial ambipolar diffusion predominates has been made by Tonks, Alfvén, and Allis and Gordon. Allis' result, which holds even for weak H ,

$$M \approx -\frac{nk(T_e + T_i)}{H} \frac{1}{1 + \nu_e \nu_i / \omega_H \omega_{Hi}}, \quad (8)$$

where we have assumed $H \gg 4\pi M$. The electrons now move in trochoidal paths around H under the influence of the potential $\varphi(r)$, rather than the former elliptical gyration. However, the field $-\partial\varphi/\partial r$ itself exerts no azimuthal torque on the charges, hence the angular momentum and magnetic moment of the plasma remain unchanged for large H , whether $\varphi(r)=0$ or not. After maximizing in magnitude near $\nu_e \nu_i \approx \omega_H \omega_{Hi}$, the moment from Eq. (8) decreases linearly with H to zero for $H=0$. The latter effects arise from the different mobilities of ions and electrons, and hence Eq. (8) is probably sensitive to the assumptions on diffusion for weak fields. In an active mercury-vapor plasma at 5- μ pressure, the maximum of M occurs at ≈ 50 gauss.

As Simon¹⁰ has pointed out, under certain conditions electrons in gaseous discharges may cross the magnetic field by diffusing along the magnetic lines of force to a conducting electrode. This mechanism "short-circuits"

⁴ M. Steenbeck, *Wiss. Veröffentl. Siemens-Werken* **15**, 2, 1 (1936).

⁵ H. Alfvén, *Cosmical Electrodynamics* (Oxford University Press, New York, 1950) 1st ed.

⁶ L. Tonks, *Phys. Rev.* **56**, 360 (1939).

⁷ E. I. Gordon, Research Laboratory of Electronics Quart. Progr. Rept., July 15, 1957 (unpublished), p. 13.

⁸ J. H. Van Vleck, *The Theory of Electric and Magnetic Susceptibilities* (Oxford University Press, New York, 1932).

⁹ E. Teller, *Z. fur Phys.* **67**, 311 (1931).

¹⁰ A. Simon, *Phys. Rev.* **98**, 317 (1955).

the conventional transverse ambipolar diffusion process; the usual restriction that the diffusive flow of charges must equalize separately in each coordinate direction is replaced by the more general statement that the sum of charged particle fluxes over all directions be zero. Therefore, by suitable choice of gas pressure, magnetic field, and discharge dimensions, it is possible to cause $\varphi(r)=0$ in the plasma. The transition between the strong magnetic field condition ($\varphi(r)=0$), and the weak field region where ambipolar diffusion predominates can be determined approximately by setting the time required for an electron to diffuse along the magnetic field or transverse to it by the ambipolar process equal:

$$H_{\text{transition}} \approx \frac{mc}{e} (1836A)^{\frac{1}{2}} \left(\frac{L^2}{R^2} - 1 \right)^{\frac{1}{2}} (\nu_e \nu_i)^{\frac{1}{2}}, \quad (9)$$

where A is the mass number of the ion, and L and R are the length and radius of the discharge plasma, respectively.¹¹ The collision frequency ν_e will be determined by the momentum transfer impacts with neutral atoms, because the electronic kinetic energy will be high and the degree of ionization low. While our work has been done primarily where $H > H_{\text{transition}}$, the most significant results for magnetic conditions of plasmas in "low" magnetic field ($H < H_{\text{transition}}$) will be reported presently.

It should not be surprising that for large H the magnetic moment calculated from the undisturbed orbits of the charges should reduce to that predicted by diffusion arguments, since the effective force field for electronic diffusion is radial and the creation of a purely radial force field cannot alter the azimuthal angular momentum of a system of charges.

III. INTRODUCTION TO THE EXPERIMENTAL SECTIONS

Plasma diamagnetism has been observed by detecting the disturbance in the magnetic field near a discharge tube and on its axis with a small sensitive spin-resonance "gaussmeter" probe. In the presence of magnetized matter, the magnetic field measured by such a probe is given by

$$\mathbf{B} = \mathbf{H}_{\text{applied}} + 4\pi\mathbf{M} + \int_s \frac{(\mathbf{M} \cdot \mathbf{n}) \hat{r}_0}{r^2} dS - \int \frac{(\text{div} \mathbf{M}) \hat{r}_0}{r^2} dV, \quad (10)$$

where \mathbf{n} is the unit normal to the magnetized surface and \hat{r}_0 is the usual unit radius vector. Since the probe was located *outside* the plasma, the term $4\pi\mathbf{M}$ is zero. The spin probe will therefore observe only a fraction of the maximum change in local field, $4\pi\mathbf{M}$, owing to magnetic depolarization effects. The data for M re-

ported here has had a correction applied (deduced from the experimental geometry and/or a separate experiment) to compensate for this depolarization.

From Eq. (7) or (8), taking $n \approx 10^{13} \text{ cm}^{-3}$, $kT \approx 6 \text{ eV}$, and $H \approx 10^3$ gauss, we find $4\pi M \approx 1$ gauss, a field change which is easily detected by spin resonance methods. Field shifts $\delta B \approx 1/10$ to 1 gauss may easily be observed by *electron* spin resonance gaussmeters, where the rf spin absorption resonance linewidth may be as low as ≈ 1 gauss; with these methods, an rf frequency of 100 Mc/sec corresponds to a magnetic field of 36 gauss. Owing to the heavier mass of nuclei, magnetic fields ≈ 1000 – $10\,000$ gauss may be easily explored by *nuclear* resonance methods which, because of the very narrow absorption resonance, allow field shifts as low as 10^{-3} gauss to be detected.

Unlike a pickup coil, the spin-resonance probe may be easily shielded electromagnetically from the plasma, which is often a troublesome noise source in these measurements. The spin-resonance equipment can be operated in this manner because the changes in local magnetic field from the plasma have been made static or quasi-static.

IV. OBSERVATION OF PLASMA DIAMAGNETISM IN WEAK MAGNETIC FIELDS

A few simple experiments were performed under the weak magnetic field and $\varphi \neq 0$ conditions with a long hot-cathode mercury-vapor discharge plasma. One such experiment has been discussed in a previous note.¹² An electron spin resonance was produced in a small coil containing roughly 0.1 g of α, α , diphenyl β picryl hydrazyl (DPPH)¹³ placed on the axis and between two series-connected discharge tubes located within large Helmholtz coils. The magnetic field from the latter, up to 60 gauss, was sawtooth modulated repetitively to allow oscilloscopic display of the spin absorption resonance. The DPPH coil formed one arm of a balanced rf bridge such as described by Thomas and Huntoon.¹⁴ As the spin system was passing through resonance, a pulsed dc plasma was produced in the discharge tubes by a specially designed pulser.¹⁵ By pulsing the plasma at half the field-modulation frequency, a displacement of the spin resonance peak could be observed (Fig. 1). The pulsing sequence was synchronized to the 60-cps line frequency. The displacement of the spin resonance resulted from *diamagnetism* in the nearby plasma and was not from currents passing through the leads of the discharge tube. This conclusion may be easily verified by reversing the direction of the magnetic field. The field shift from diamagnetism reverses in sense if the applied magnetic field be reversed, whereas the spurious field shift does not.

¹² T. C. Marshall, R. A. Kawcyn, and L. Goldstein, *Nature* **187**, 584 (1960).

¹³ C. H. Townes and K. J. Turkevich, *Phys. Rev.* **77**, 148 (1950).

¹⁴ H. A. Thomas and R. C. Huntoon, *Rev. Sci. Instr.* **20**, 516 (1949).

¹⁵ R. A. Kawcyn, *Rev. Sci. Instr.* (to be published).

¹¹ This derivation is only approximate, and Eq. (9) is not expected to be valid for $L/R \lesssim 1$.

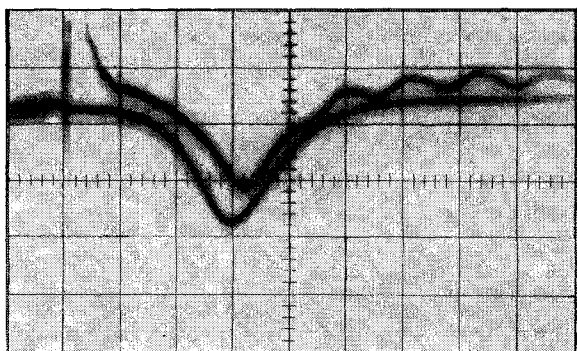


FIG. 1. Shift of the spin resonance field in DPPH from diamagnetism of a mercury vapor argon plasma. Horizontal scale, 1 gauss/cm.

Field changes of 1/10 to 1/2 gauss were observed for the hot cathode discharge; measurement was made photographically by comparing the shift of the DPPH absorption maximum with its linewidth. A linear increase in diamagnetism was observed up to a field ≈ 55 gauss, where a "maximum" occurred. The value of M measured at this point set $nkT_e = 2.8$ erg/cm³ whereas 2.6 erg/cm³ was measured using a pulsed Langmuir probe technique.¹⁶ Theoretical values of ionic and electronic cross sections gave $\nu_e \nu_i / \omega_H \omega_{H_i} \approx 3/4$ at this field and vapor pressure $\approx 5\mu$.

The precision of the above experiment was improved by subjecting the spin system to magnetic field modulation with amplitude considerably less than the spin absorption line width. Figure 2 shows a clearly defined "maximum" of the plasma magnetic moment for a cold-cathode mercury vapor discharge at $\approx 3\mu$ pressure. The "maximum" has shifted to a weaker field than above because of the reduced mercury vapor pressure. The magnetization is of course zero at zero field because of the vanishing of the electronic orbital angular momentum.

V. OBSERVATION OF PLASMA DIAMAGNETISM IN STRONG MAGNETIC FIELDS

Measurement of diamagnetism of plasmas in strong fields is of greatest interest because the plasma energy density may be directly inferred uncomplicated by collision effects. For these observations, a nuclear magnetic resonance (NMR) technique, which can detect field changes as small as one part per million, has been found convenient. This technique permits the observer to measure the plasma magnetization after the plasma, the chief noise source in such investigations, has become inactive.

The NMR apparatus is based on a variant of the pulsed rf spin flip and nuclear induction method such as discussed by Hahn¹⁷ and Spokas and Slichter.¹⁸

¹⁶ J. F. Waymouth, J. Appl. Phys. **30**, 1404 (1959).

¹⁷ E. L. Hahn, Phys. Rev. **80**, 580 (1950).

¹⁸ J. J. Spokas and C. P. Slichter, Phys. Rev. **113**, 1462 (1959).

About 1/10 cm³ of glycerine is enclosed in a small coil placed between the polefaces of a large Varian electromagnet (Fig. 3). In the static magnetic field, the protons in the glycerine precess with frequency

$$\omega_{\text{spins}} = \gamma H, \quad \gamma = 2.68 \times 10^4 \text{ (gauss sec)}^{-1}; \quad (11)$$

if, however, a strong pulse of rf with frequency near ω_{spins} be applied to the coil, the proton spins will be nutated by H_1 , the rf magnetic field, $\pi/2$ into the plane perpendicular to H in a time

$$t' \approx \pi/2\gamma H_1. \quad (12)$$

The rf field, H_1 , lies perpendicular to H ; in practice, the pulse width $t' \approx 5\text{--}10$ μ sec for an rf pulse of a few hundred watts power. This technique allows the largest component of the net nuclear magnetization to be observed.

After this high-level rf signal has been removed, the receiver detects the nuclear induction signal from the freely precessing spins by a phase detection method in which the precession frequency of the spins is compared with the frequency of the coherent rf source which flipped them. A sizeable decaying audio beat signal, with frequency dependent on how far the rf source is detuned from ω_{spins} , can be displayed on the oscilloscope. Should the magnetic field change by an amount δB while the spins are precessing, the shift in phase of the beat signal is

$$\theta = \int_0^\tau \gamma \delta B(t) dt, \quad (13)$$

where δB persists for an interval τ . These concepts are illustrated in Fig. 4, which shows the progressive shift in phase of the nuclear induction signal when the spin probe is located in an appropriate position near an active plasma. The nuclear signal persists in measurable amplitude for several milliseconds, and the active discharge, from which δB is derived, is over before the NMR signal decays. The plasma was pulsed at half the pulsed NMR frequency (20 pps) to permit

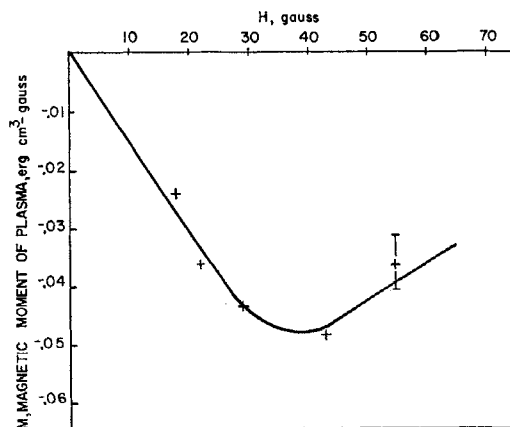


FIG. 2. Diamagnetism of a cold cathode mercury vapor plasma.

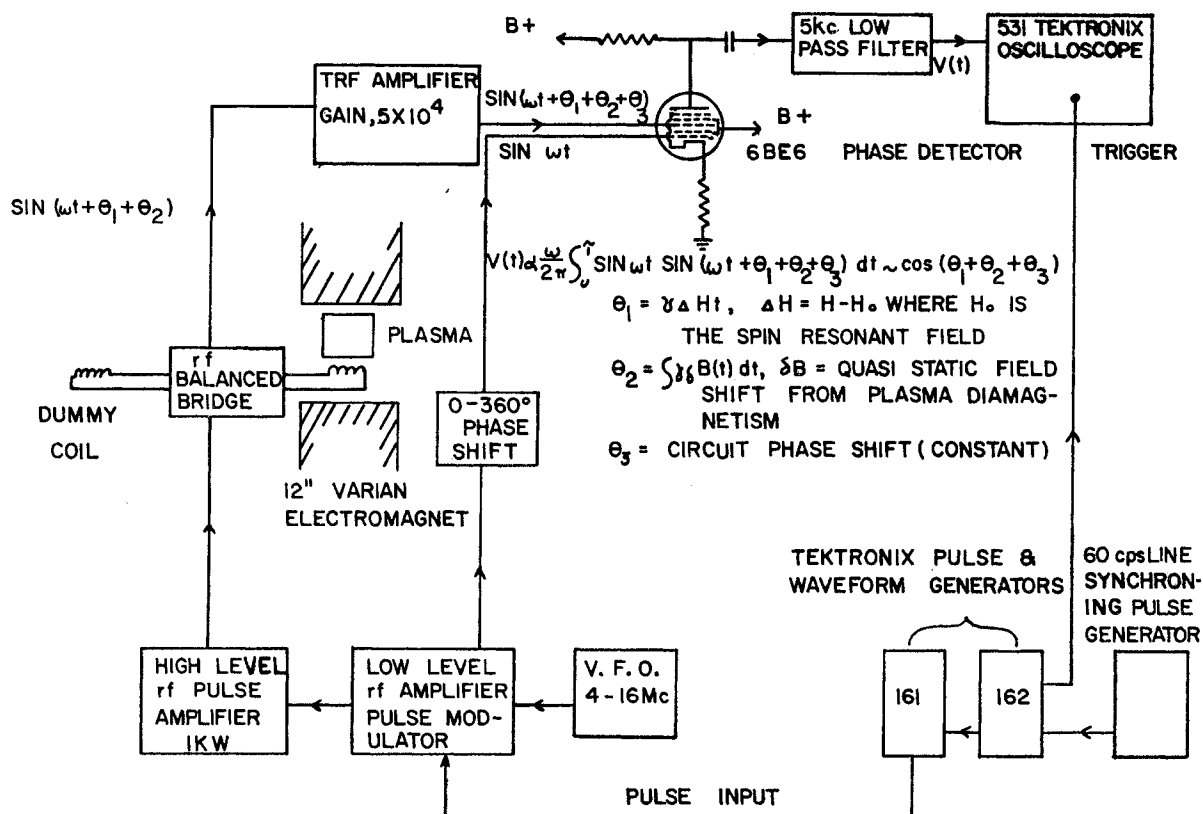


FIG. 3. Block diagram of pulse NMR equipment.

immediate display of the phase shift on an oscilloscope. In the instance illustrated by Fig. 4, the field is 1650 gauss, and the frequency of the rf source, about 7 Mc/sec, is within 4 kc/sec of resonance.¹⁹ A 5-kc/sec low-pass filter improved the NMR signal/noise.

The plasma was produced in a modified Penning ionization gauge (P.I.G.) discharge tube, a section of which is shown in Fig. 5. In addition to the electrical configuration, the principal features are: (1) the transverse cylindrical cavity which allows probing and/or

disturbing 4500-Mc/sec microwaves to propagate across the magnetic field through the plasma in a geometry which permits the angle between the rf electric field and the dc magnetic field to be varied continuously, and (2) the demountable metal construction, which permits ready repair and alteration. Currents as high as 5 amp could be passed through helium gas at a pressure as low as 10 μ . Operation of

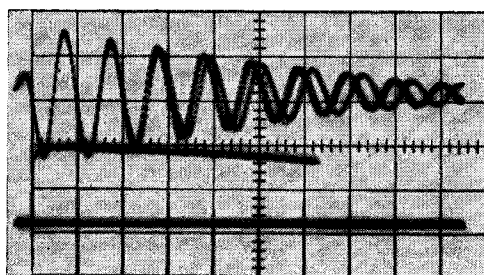


FIG. 4. Phase displacement of proton spin induction beat signal from a diamagnetic plasma. Discharge current pulse lasts about 2 msec.

¹⁹ For further information, the reader is referred to Tech. Rept. No. 8, AF2152, University of Illinois, by T. C. Marshall, R. A. Kawcyn, and L. Goldstein (unpublished).

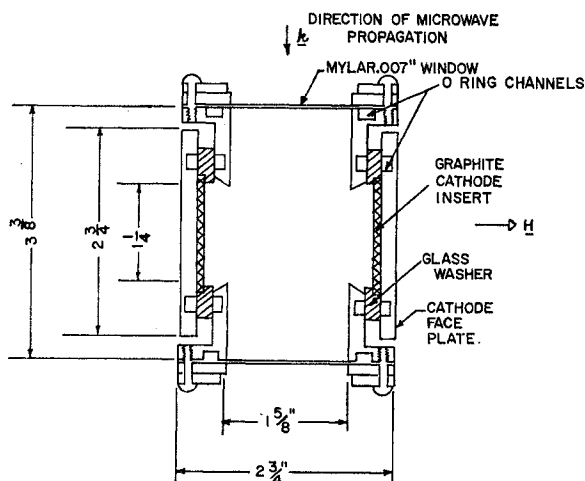


FIG. 5. Section of the modified P.I.G. discharge tube.

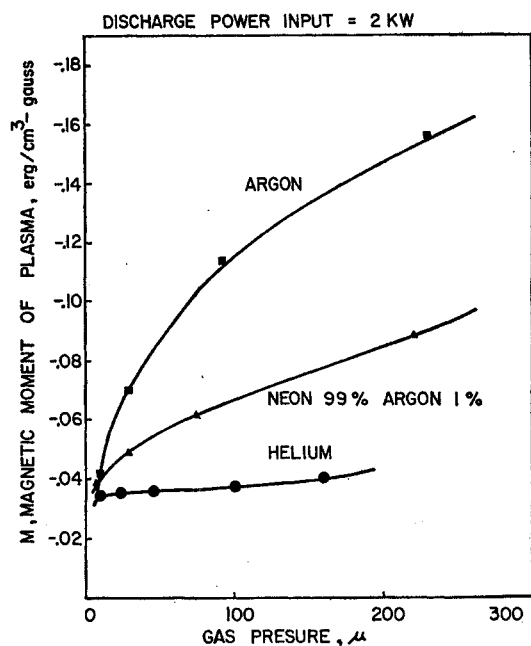


FIG. 8. The dependence of plasma diamagnetism upon gas pressure and power input.

plasma diamagnetism upon magnetic field. The theoretical $1/H$ behavior of the diamagnetism fits the data reasonably well; however, only if the operation of the discharge tube is relatively insensitive to magnetic field changes can the above be valid. In view of our preceding remarks, we believe this comparison is justified. It was also possible to observe the change in local field while the plasma was present using a variation of the simple marginal oscillator NMR gaussmeter described by Hopkins.²⁰ This circuit may be easily tuned to the NMR absorption for a wide range of magnetic fields. The magnetization can be inferred by the phase change of the "wiggles" following a nuclear resonance in water, displayed on the oscilloscope by weakly modulating the static magnetic field at low frequency. The size of this probe is, however, larger than the glycerine probe and the circuit is more sensitive to interference than the one discussed previously.

By inserting an insulating glass sleeve into the discharge cavity, it was established that the metallic lateral walls of the discharge tube had little effect on the plasma diamagnetism.

VI. PLASMA DIAMAGNETISM UNDER THE INFLUENCE OF MICROWAVE ENERGY OF FREQUENCY NEAR THE ELECTRON CYCLOTRON FREQUENCY

If the frequency of an intense microwave beam is tuned at or near the gyrofrequency of the electrons in a plasma $\omega \approx \omega_H = eH/mc$, a considerable resonance increase of the plasma electron kinetic energy density

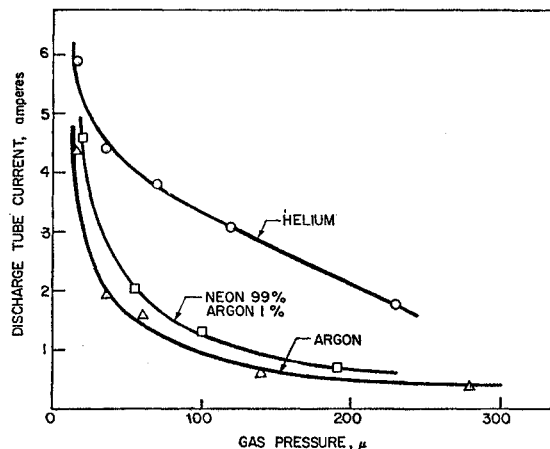


FIG. 9. The discharge current required to produce an electron concentration of about $9 \times 10^{12} \text{ cm}^{-3}$ in the P.I.G. tube at 1600 gauss field.

is to be expected. In principle, an observation of the enhanced plasma diamagnetism resulting from this source then may be used, in conjunction with other independent measurements, to yield data on the energy relaxation processes in the plasma. We shall be interested in studying this enhancement, which for the following treatment will be defined by the second term of

$$M_{\text{total}} = -[(nkT_e/H)_{\text{dc plasma}} + (\Delta p_W/H)_{\text{rf}}], \quad (14)$$

where Δp_W is the enhanced kinetic energy density in the plane perpendicular to H .

The absorption of rf energy may be estimated from an expression for the transverse rf conductivity σ_{\perp} of the plasma near the electron cyclotron resonance:

$$\sigma_{\perp} = \frac{ne^2}{mv} \left[\frac{v^2/\omega^2(1 + \omega_H^2/\omega^2 + v^2/\omega^2)}{(\omega_H^2/\omega^2 - 1 + v^2/\omega^2)^2 + 4v^2/\omega^2} \right]. \quad (15)$$

Equation (15) was obtained as a solution of the simple Langevin equation, and has the typical resonance form. In Fig. 11, a very narrow cyclotron resonance absorption profile is shown, which follows Eq. (15) in form.

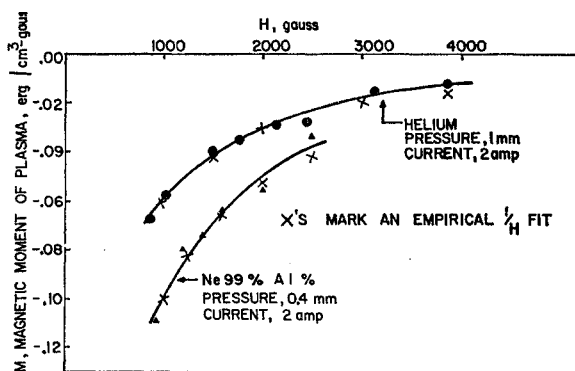


FIG. 10. The dependence of plasma diamagnetism upon the applied magnetic field.

²⁰ N. J. Hopkins, Rev. Sci. Instr. 20, 401 (1949).



FIG. 11. Simultaneous display of free electron ($g=2$) orbital cyclotron resonance absorption of microwave energy and an electron spin resonance in DPPH ($g=2.0043$), the DPPH located in a capsule inside the P.I.G. tube. Horizontal scale, 2.3 gauss/cm.

This resonance was observed in low pressure air ($<10\mu$) with an exceedingly low-energy density plasma in the discharge tube; there was no shift in the resonance frequency eH/mc as the electron concentration was increased. Broadening of the absorption resonance was, however, much greater than that predicted by Eq. (15) on the basis of collisions alone as the dc plasma energy density was increased to $\approx 10\text{--}50$ erg/cm³. This is possibly due to the Doppler effect of the random electronic motions or the interaction of the electrons with the thermal radiation field near the cyclotron frequency. The latter effect arises from the broadening of the electronic Landau levels from random induced transitions, and amounts to

$$\Delta f/f \approx \frac{32\pi^3}{3\hbar^2 c^3} f k T |er|_{nn'}^2, \quad (16)$$

where $|er|_{nn'}$ is the electric dipole matrix element between two Landau states and f is the radiation frequency. Equation (16) is approximate because it assumes first order perturbation theory and radiative equilibrium (which requires an opaque plasma). The resonance width from this source is ≈ 100 gauss for the higher electronic temperatures.

An increase of the diamagnetism of an existing dc plasma was observed with the NMR probe when the plasma was disturbed by absorption of microwave energy (4500 Mc/sec) radiated by a cw magnetron (Raytheon QK 273). Figure 12 shows that the additional NMR beat signal phase displacement, corresponding to a plasma energy density increase, from the absorption of ≈ 1 watt/cm² (50 w) of rf is comparable with the phase displacement from the existing plasma energy density of 200 w of dc power. In this example, the NMR pulsing rate was 20 pps, the dc discharge, 10 pps, and the magnetron, 5 pps. Changes in the dc current through the discharge tube when the magnetron

was switched on were negligibly small because of a large resistance in series with the discharge tube. The pulse duration of the magnetron was the same as that for the dc current discharge, 2 msec.

Langmuir probe and optical studies of the plasma showed that the electronic kinetic energy was on the average little influenced by the rf. This is plausible, because in a hot, weakly ionized active plasma a small increase in electronic kinetic energy yields a comparatively large increase in the number (n_i) of electrons capable of ionization, and therefore a large increase in n . This number, n_i , is roughly

$$n_i/n \approx \frac{2}{\sqrt{\pi}} \left[\frac{W_i}{kT} \right]^{-\frac{1}{2}} e^{-W_i/kT}, \quad (17)$$

assuming a purely Maxwellian energy distribution, and taking W_i as the ionization energy for the atom. The Maxwell distribution results from the rapid thermalization of the kinetic energy gained from the rf field.

In the steady state, since electron loss under these conditions is due mainly to diffusion along H , Eq. (17) must be combined with the diffusion equation

$$\partial n / \partial t = 0 = -D_{11}^{(\text{amb})} \nabla^2 n + n_i \langle V_i \rangle / \lambda_i, \quad (18)$$

where $D_{11}^{(\text{amb})}$ is the ambipolar diffusion constant for motion parallel to H , and the energy conservation equation

$$\begin{aligned} n \frac{2m}{M_a} \frac{\langle V_e \rangle}{\lambda_e} W + n_x \frac{\langle V_x \rangle}{\lambda_x} W_x + n_i \frac{\langle V_i \rangle}{\lambda_i} (W_i + W) \\ = P_{\text{rf}} + P_{\text{dc}}, \end{aligned} \quad (19)$$

$$kT \approx \frac{2}{3} W.$$

Here, P_{rf} and P_{dc} are the absorbed rf and dc input power densities, respectively, $\lambda_{e,i,x}$ and $\langle V_{e,i,x} \rangle$ are the mean free path and average velocity for collision, ionization, and excitation; and n_x is the number of electrons which may inelastically excite the neutral atom of mass M_a . If we take as typical experimental

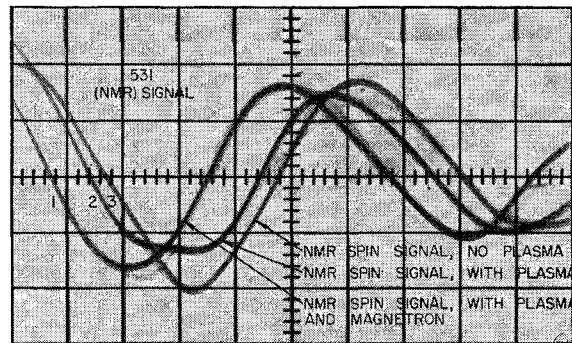


FIG. 12. Enhanced phase displacement of NMR beat signal from an active plasma irradiated by high power gyro-resonant microwaves. Helium gas, pressure 0.2 mm; discharge current, 0.3 amp.

data for helium a gas pressure of 100μ , $n \approx 5 \times 10^{11} \text{ cm}^{-3}$, $P_{\text{de}}^{\text{effective}} \approx 1 \text{ watt/cm}^3$, $P_{\text{rf}} = 0$, $D_{11}(\text{amb}) \approx 5 \times 10^6 \text{ cm}^2/\text{sec}$, then Eqs. (18) and (19) give W , the average electronic energy, $\approx 10 \text{ ev}$. Further rf heating with $P_{\text{rf}} \approx 1 \text{ watt/cm}^3$ power density will cause an increase of only 2 ev in W , but this increase nearly doubles n_i/n .

Figure 13(a) shows the influence of the relatively high-level rf energy on the plasma diamagnetism near cyclotron resonance, which is thus seen to be enhanced, and 13(b) shows the effects for rf frequencies considerably below resonance. Complete absorption was obtained in the first case, but only about 50% absorption in the second instance. The "saturation" effect shown in 13(a) has been attributed to a local breakdown of the gas near the plasma surface; the region between the microwave port and the plasma was stuffed with glass wool. In the limit of zero power, i.e., $E_{\text{rf}} \rightarrow 0$, the ratio of the slopes of 13(a) and 13(b) is given by Eq. (15):

$$\left[\frac{\text{slope of (a)}}{\text{slope of (b)}} \right]_{E_{\text{rf}} \rightarrow 0} \approx \delta^2 \omega^2 / (\nu^2)^{\text{effective}}, \quad (20)$$

where $\omega_H \equiv \omega(1 + \delta)$, and we take $2\nu_{\text{eff}}/\omega$ as the observed resonance breadth from all causes. Here $\delta \approx 0.2$ and $2\nu/\omega = \Delta H/H \approx 1/10$ from the radiation effect which dominates the purely collisional resonance broadening for these pressures and electron temperatures ($kT \approx 10 \text{ ev}$) in helium. Thus $\delta^2 \omega^2 / \nu^2 \text{ eff} \approx 16$, whereas the experimental data yield ≈ 14 .

Assuming the energy density in the plasma responds exponentially in time when the rf power is altered, the steady state power balance between rf power input P_{rf} and the losses is

$$P_{\text{rf}} = \frac{3}{2} \Delta \rho W / \tau_W, \quad (21)$$

which defines the kinetic energy relaxation time, τ_W [see Eq. (14)]. Photomultiplier studies of the initial decay of excitation light from the plasma as all power was removed suggested $\tau_W \approx 10^{-5} \text{ sec}$. This relaxation time is consistent with $D_{11}(\text{amb})$. At 1650 gauss field, a maximum augmented magnetization of $-0.047 \text{ erg/cm}^3\text{-gauss}$ could result from complete absorption ($\approx 1 \text{ watt/cm}^3$) of microwave energy, which corresponds to a plasma energy density increase of 75 erg/cm^3 (reflection of the microwaves at the vacuum-plasma interface has been accounted for). A linear extrapolation of the graph in Fig. 13(a) yields $M \approx -0.035$ at 70 watts input, which appears to be fair agreement.

VII. CONCLUSION

On the basis of these experiments, the authors believe that substantial agreement exists between theory and

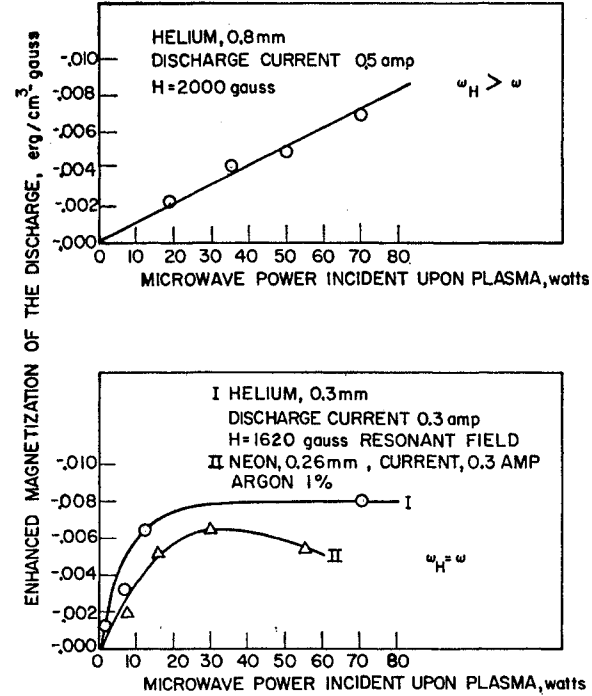


Fig. 13. Enhanced magnetization of the discharge as a function of microwave power incident upon the plasma (a) near cyclotron resonance; (b) considerably below resonance (upper figure).

experiment for gaseous plasma diamagnetism. At low fields, the behavior of the magnetic moment agrees with the diffusion theory of Allis and Gordon, and is in reasonable agreement with the tabulated values of electron mobility in mercury vapor for this example. When the magnetic field is strong, the gas pressure sufficiently low, and the plasma short compared to its diameter, ambipolar diffusion of electrons directly across the field in the plasma is unfavored; nevertheless, a strong diamagnetism is observed, in agreement with the predicted behavior nkT_e/H . This must arise indirectly from the electronic orbits and the lack of a "reverse" plasma boundary current.

The measurement of diamagnetism in a plasma in a strong magnetic field affords an excellent method for determining the total energy density of the plasma charges. The observations can be made with a small NMR probe which need not be inserted directly into the plasma, nor need it be an integral part of the particular plasma experiment being performed. Since the nuclear resonance absorption linewidth of protons in liquids is very narrow, it is little affected by the electromagnetic interactions among plasma constituents. Figure 11 implies that the narrow electron cyclotron absorption resonance of a weak low-pressure discharge may also be used as a sensitive gaussmeter to detect the diamagnetic field shift from a more energetic nearby plasma.

Although this need have no connection with diamag-

netism measurements, we point out that the NMR technique may be used for probing the transient, quasi-static magnetic fields in a plasma. The NMR apparatus yields $\int \delta B_z(t) dt$ directly; the integration is done by the spin "memory" of the protons, and not by an external circuit. Of course, δB_z should not contain important Fourier components above ω_{spins} . A considerable advantage of this method resides in the

fact that the observations may be postponed until the noisy transient plasma has been deactivated.

ACKNOWLEDGMENTS

The authors wish to thank Professor C. P. Slichter for his suggestion on the nuclear resonance aspect of this experiment and R. A. Kawcyn for his experimental assistance and contribution to the electronic design.

PHYSICAL REVIEW

VOLUME 122, NUMBER 2

APRIL 15, 1961

Electrical Breakdown in Hydrogen at Low Pressures*

A. L. WARD

Diamond Ordnance Fuse Laboratories, Washington, D. C.

AND

EIFIONYDD JONES†

Department of Physics, University of Wales, University College of Swansea, Swansea, Wales

(Received December 13, 1960)

Experimental and theoretical determinations of the static voltage-current characteristics, extending from the region of the Townsend (self-maintained) discharge to the normal glow discharge, have been carried out in hydrogen at low pressures, (7–25 mm Hg). The calculations, made on an electronic computer, were based on the distortion of the electric field by space charge, and used the experimentally determined variation of both the primary and secondary Townsend ionization coefficients on the ratio of the field to pressure. Good agreement is obtained between the measured and calculated breakdown and glow voltages, and both the experimental and theoretical curves of the characteristic are of similar shape.

I. INTRODUCTION

THE breakdown potential of a gas between parallel plate electrodes was derived by Townsend, who gave a criterion which expressed the condition that internal ionization processes, in the gas and at the cathode, just replace the externally initiated current.¹ However, the actual breakdown transition, i.e., the lowering of the potential with the increase of current, is less well understood.

The voltage-current characteristic of a discharge in this region of transition from breakdown to glow voltage is difficult to obtain experimentally. Mathematical calculations for this region are complex because of the behavior of the space-charge field and its subsequent effect on the discharge parameters. However, with the aid of an electronic computer, Ward² has calculated static characteristics which extend well into the glow region at low pressures. The present paper introduces certain improvements into these calculations and compares such theoretical characteristics with those determined experimentally in hydrogen at low pressures. Measurements and calculations extend from breakdown to the normal glow discharge.

* Supported in part by the Office of Ordnance Research.

† Now at CERN, European Organization for Nuclear Research, Geneva, Switzerland.

¹ See, for example, F. Llewellyn-Jones, *Ionization and Breakdown in Gases* (Methuen and Company, London, 1957).

² A. L. Ward, *Phys. Rev.* **112**, 1852 (1958).

A report on a similar comparison for hydrogen at high pressure, but restricted to the breakdown region and below, has been published recently.³

II. DESCRIPTION OF APPARATUS

(a) Discharge Tube and Vacuum System

The experiments were carried out in a simple two-electrode parallel plate discharge tube. The electrodes were made of copper in the shape of disks 3 cm in diam, rounded at the edges to prevent local field intensification, mounted parallel and 0.195 cm apart. The electrodes were first cleaned by washing in grease solvents and then carefully polished. The final polishing was carried out with a Selvyt cloth with micro-alumina as the polishing agent. After further washing in distilled water and drying, the electrodes were sealed into a borosilicate glass envelope which was quickly sealed into a vacuum system and pumped down to a pressure of the order of 10^{-6} mm Hg with the aid of an oil diffusion pump and a conventional backing pump. Liquid air traps were always used to prevent back diffusion of oil into the discharge tube.

The gas used in the experiments was hydrogen produced by electrolysis of pure barium hydroxide dissolved in distilled water. The gas was dried by many hours

³ D. J. De Bitetto, L. H. Fisher, and A. L. Ward, *Phys. Rev.* **118**, 920 (1960).

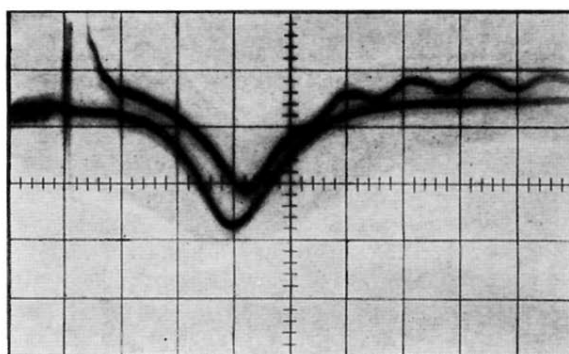


FIG. 1. Shift of the spin resonance field in DPPH from diamagnetism of a mercury vapor argon plasma. Horizontal scale, 1 gauss/cm.

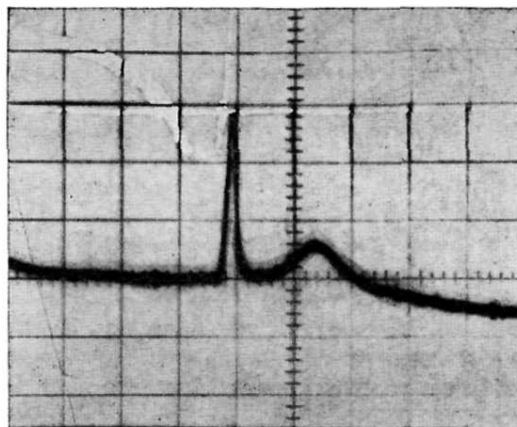


FIG. 11. Simultaneous display of free electron ($g=2$) orbital cyclotron resonance absorption of microwave energy and an electron spin resonance in DPPH ($g=2.0043$), the DPPH located in a capsule inside the P.I.G. tube. Horizontal scale, 2.3 gauss/cm.

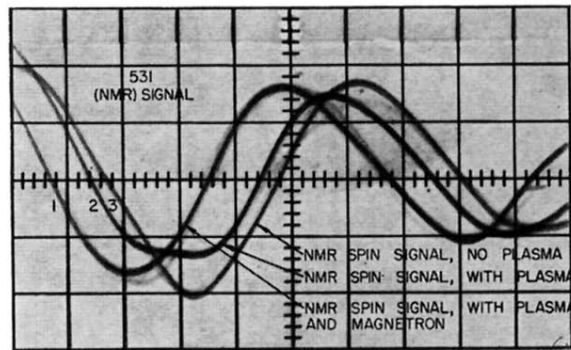


FIG. 12. Enhanced phase displacement of NMR beat signal from an active plasma irradiated by high power gyro-resonant microwaves. Helium gas, pressure 0.2 mm; discharge current, 0.3 amp.

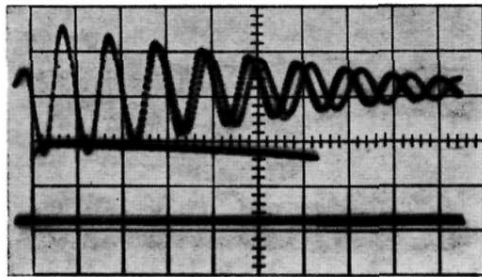


FIG. 4. Phase displacement of proton spin induction beat signal from a diamagnetic plasma. Discharge current pulse lasts about 2 msec.