The Principle of Nuclear Induction[1]

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IT IS A TRIBUTE to the inherent harmony and the organic growth of our branch of science that every advance in physics is largely due to the developments that preceded it. The discovery for which Purcell and I have received the honor of the Nobel Prize award for the year 1952 is a typical example of this situation, and before describing the principle I shall therefore present an outline of its long and distinguished background.

Both the method and the object go back ultimately to spectroscopy, a field to which modern physics owes so much in other respects. Among the various aspects of this field there are two that are of particular importance here: the Zeeman effect for introducing magnetic fields as an essential element of spectroscopy, and the hyperfine structure of spectral lines for revealing the existence of nuclear moments. The correct interpretation of hyperfine structures was first given in 1924 by Pauli (*1*), who proposed that atomic nuclei may possess an intrinsic angular momentum (spin) and, parallel to its orientation, a magnetic moment. The energy of interaction of this magnetic moment with the magnetic field $H(0)$, produced by the atomic electrons at the position of the nucleus, depends upon the angle between them and leads thus to the observed small splitting of the energy levels. Conversely, it is possible under suitable conditions to determine from this splitting both the spin and the magnetic moment of the nucleus, and these two important quantities have indeed been determined in a great number of cases from the observation of hyperfine structures. The magnetic moments of the nuclei have been found, in all observed cases, to be of the order of the "nuclear magneton" that one obtains by substituting in the formula for the atomic Bohr magneton the mass of the proton in place of that of the electron. Nuclear moments are thus about a thousand times smaller than atomic moments, and this is plausible in view of the fact that one deals here with protons instead of electrons as elementary charged constituents. There are, however, distinct disadvantages in the optical determination of nuclear moments. In the first place, the accuracy is seriously limited, because the effect consists only in such a small splitting of spectral lines that one has to be concerned with their finite width. In the second place, it is necessary for the determination of the nuclear magnetic moment from the observed hyperfine structure to have a knowledge of the field $H(0)$ which is usually rather inaccurate since it involves complex electron configurations. In view of these limitations, one is led to values of nuclear magnetic moments with an accuracy of a few per cent at best. Finally, one is faced with the fact that hyperfine splittings tend to decrease with decreasing atomic number, with the result that it is not possible, by optical means, to observe them in the case of the greatest fundamental importance, that of hydrogen.

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Dr. Bloch, Professor of Physics at Stanford University, was co-winner with Dr. Edward M. Purcell of the Nobel Physics Prize for 1952. He was honored for his development of precision methods in the realm of nuclear magnetism and for the discoveries made by their use.

Dr. Bloch was born in Zurich, Switzerland, in 1905, and received his Ph.D. degree from the University of Leipzig in 1928. His work and study took him to Switzerland, Holland, Denmark, and Italy. In 1932 he returned to the University of Leipzig, but within a year, upon Hitler's ascent to power, decided to leave Germany. He held a Rockefeller Foundation fellowship in Rome, 1933–34 and in the spring of 1934, when he was offered a faculty appointment at Stanford, came to the United States. During the war years of 1942–45 he did research work at Stanford and Los Alamos and while associate group leader at the Radio Research Laboratory at Harvard discovered the theory of nuclear induction.

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A decisive step forward was made in 1933 by Stern (*2*), who applied his method of molecular beams to the determination of the magnetic moments of the proton and the deuteron in hydrogen molecules. Instead of the emitted light, it is the deflection of the molecule in an inhomogeneous magnetic field that is here affected by the nuclear moments. Although the observed effect was close to the limit of observability, it yielded the proton moment to within ten per cent, with the most important result that instead of having the expected value of one nuclear magneton it is about 2.5 times larger. Of similar importance was the result that the magnetic moment of the deuteron was between 0.5 and 1 nuclear magneton, since it indicated from the simplest plausible considerations of the structure of this nucleus that one should ascribe a moment of about 2 nuclear magnetons to the neutron. I shall come back later to this point; it represents the start from which my own experimental work has followed in an almost continuous line.

Subsequent to Stern's work, a number of far-reaching further developments have been achieved by Rabi in the application of atomic and molecular beams to the measurement of nuclear moments and hyperfine structures. Without attempting completeness, I want to mention some aspects of method in the brilliant series of investigations which he carried out with his collaborators. One of them is based upon a paper by Breit and Rabi (*3*), which treats the variation of the magnetic moment of an atom for the different Zeeman levels of hyperfine structure under the influence of an external magnetic field and which was applied to atomic beams where the deflection gives a direct measure of the magnetic moment. Another important aspect lies in the passage of the beam through two and later three separate field regions which can be adjusted to give zero deflection so that one deals with a null method. These innovations, besides giving many other interesting results, allowed the measurement of the hyperfine structure in the ground state of light and heavy hydrogen atoms; since the previously mentioned field $H(0)$, produced by the electron at the place of the nucleus, was given here, through a formula of Fermi (*4*), from the well-known theory of the hydrogen atom, this measurement resulted in the determination of the magnetic moments of the proton and the deuteron with an accuracy of a few per cent.

However, the most significant improvement in molecular and atomic beam techniques was the introduction of the magnetic resonance method. The beam here passes through a region where the magnetic field is homogeneous and constant with a weak alternating magnetic field superimposed at right angles to the strong constant field. Analogous to the resonance absorption of visible light, transitions occur here from one Zeeman level to another, if the alternating field satisfies Bohr's frequency condition for the energy difference between the two levels. However, instead of optical frequencies one deals here normally with frequencies in the radio range, so that this application of the magnetic resonance method, like our own, is properly labeled as belonging to the new field of radiofrequency spectroscopy. In the beam technique it has the great advantage of dispensing with a knowledge of the deflecting inhomogeneous fields, since the deflection is merely used now as an indicator for the occurrence of transitions in the homogeneous field region. A very much greater accuracy can thus be obtained; it led, for example, to the knowledge of the magnetic moments of the proton and the deuteron with an accuracy of about one part in a thousand and to the important discovery of a small but finite electrical quadrupole moment of the deuteron (*5*) in 1939.

The first use of the magnetic resonance method was suggested in 1936 by Gorter (*6*) in an attempt to detect the resonance absorption of radio quanta through the heating of a crystal. While the results of this experiment were negative, Rabi (*7*) in 1937 has treated the transitions in a rotating field and has pointed out their use in atomic and molecular beams.

Coming from quite a different direction, I was led at that time to similar ideas. They originated from my preceding work, which dealt with the magnetic moment of the neutron and which had been stimulated by Stern's previously mentioned measurement of the magnetic moment of the deuteron (*2*). The idea that a neutral elementary particle should possess an intrinsic magnetic moment had a particular fascination to me, since it was in such striking contrast to the then only existing theory of an intrinsic moment which had been given by Dirac (*8*) for the electron. Combining relativistic and quantum effects, he had shown that the magnetic moment of the electron was a direct consequence of its charge, and it was clear that the magnetic moment of the neutron would have to have an entirely different origin. It seemed important to furnish a direct experimental proof for the existence of a magnetic moment of the free neutron, and I pointed out in 1936 (*9*) that such a proof could be obtained by observing the scattering of slow neutrons in iron. The very strongly inhomogeneous magnetic field in the neighborhood of each iron atom was shown to affect a passing neutron through its magnetic moment and thus to lead to an appreciable magnetic scattering effect; it was shown at the same time that magnetic scattering would lead to the polarization of neutron beams. The existence of this effect was first clearly demonstrated in 1937 by a group of investigators at Columbia University (*10*), and it opened up the possibility of further work with polarized neutron beams.

The most desirable goal to be reached here was that of accurately measuring the magnetic moment of the neutron. It occurred to me that resonance depolarization could be achieved by passing a polarized neutron beam through a region where a weak oscillating field is superimposed on a strong constant

field, provided that the frequency of the former is equal to the frequency with which the neutron moment carries out a precessional motion around the direction of the constant field. A knowledge of this field and of the corresponding resonance frequency directly determines the magnetic moment under the safe assumption that the spin of the neutron is one-half, and the magnet scattering effect enters in this arrangement merely as an indicator for the occurrence of resonance depolarization. The application to polarized neutron beams was also noted by Rabi (*7*) in his previously mentioned original paper on the magnetic resonance method. It was first achieved in 1939 by Alvarez and myself (*11*) with the use of the Berkeley cyclotron, and it yielded a value for the magnetic moment of the neutron that was consistent with that of the deuteron if one assumed the latter to be additively composed of the moments of the proton and the neutron. The accuracy of this measurement amounted to about one per cent and was partly limited by that with which the strength of the constant field could be determined. Another limit of accuracy arose from the smallness of the observed polarization effect, but a subsequent systematic investigation of neutron polarization (*12*), carried out with the Stanford cyclotron, showed how this effect could be greatly increased.

It was of considerable importance to improve the accuracy of the determination of the neutron moment to at least one part in a thousand in order to test small deviations from the additivity of the moments of the proton and the neutron, which could be expected in connection with the finite electric quadrupole moment of the deuteron, according to the theoretical work of Rarita and Schwinger (*13*). The fact that higher accuracy hinged essentially upon that of a field calibration and the search for a suitable and convenient standard led me to new ideas when, toward the end of the last world war, my thoughts turned back to the continuation of my previous work.

The essential fact of the magnetic resonance consists in the change of orientation of nuclear moments, and the methods to be employed in molecular and atomic beams as well as in neutron beams are primarily indicated by different ways to detect this change. The acquaintance with radio techniques during the war suggested to me still another and much simpler way, that of detecting the reorientation of nuclear moments through the normal methods of radio reception. The signals to be detected would be due to the electromagnetic induction caused by nuclear reorientation and should appear as a voltage difference between the terminals of an external electric circuit. I believe that this is the most general and distinctive feature of our discovery, and it is for this reason that I chose for it the name of "nuclear induction." Purcell, whose independent approach was largely based on considerations of energy relations, has chosen to call it "nuclear magnetic resonance absorption," but soon after our respective initial work and despite its apparent difference, it became clear that it was based upon the same principle.

In order to understand this principle, one can start from macroscopic quantities and describe the underlying phenomenon in classical terms. Consider for this purpose, as a typical example, about one cubic centimeter of water with the protons contained in it as the nuclei under investigation. Their magnetic moments are oriented in a completely random manner in the absence of an external magnetic field; after the sample has been brought into such a field, however, there will be established a new thermal equilibrium in which the magnetic moments are distributed with a slight surplus parallel to the field. Even in relatively strong fields of the order of 10,000 gauss this surplus will, at room temperature, amount to no more than about one part in a million. While its direct observation would be difficult, there thus exists a "nuclear paramagnetism" in the sense that one deals with a finite macroscopic nuclear polarization which is both parallel and proportional to the applied external field. The establishment of thermal equilibrium demands the transfer of the energy released by the partial orientation of the nuclear moments into heat, and it can take place only through interaction of these moments with their molecular surroundings. The strength of this interaction determines the time interval required for the nuclear moments to adjust themselves to the equilibrium conditions; it is measured by the "relaxation time," as in the analogous case of atomic paramagnetism. The role of the relaxation time is of basic significance for our experiments, and I shall soon come back to its discussion.

For the moment, we shall return to the equilibrium state, once it is established, and to the description of the nuclear polarization under the conditions of magnetic resonance. A simple mechanical consideration of the gyroscope shows that an alternating field at right angles to the constant field has the effect of tilting the direction of the polarization with respect to the constant field, and that the polarization will thereupon continue to perform a precessional rotation around this field. The angular frequency of precession is proportional to the field with a constant of proportionality which is called the "gyromagnetic ratio" of the nuclei and which is equal to the ratio of their magnetic moment and their intrinsic angular momentum. From a perfectly macroscopic point of view, one thus deals with a situation in which the protons in our cubic centimeter of water have the effect of an invisible compass needle rotating in its interior. The "invisibility" refers actually only to observation of optical frequencies; the rotation occurs in the range of radiofrequencies, and it can very well be observed by using Faraday's law of induction. Indeed, the rotation of our compass needle is accompanied by that of a magnetic field which possesses an alternating component perpendicular to the axis of rotation, and hence by an electromotive force,

induced in a suitably wound coil of wire around the sample. From here on it is merely a matter of the standard techniques of radio reception to rectify and amplify this electromotive force so that it can be recorded on a voltmeter, displayed on a cathode-ray oscillograph, or made audible in a loudspeaker.

What amazed me most in my first calculations on this effect was the magnitude of the signals which one could expect from nuclear induction. In our example of a cubic centimeter of water in a normal field of a few thousand gauss they turned out to amount to the order of a millivolt. This magnitude is well above the noise which accompanies any radio receiver and which sets the ultimate limit of signal detection. It should be observed here that, being a phenomenon of fluctuations, the noise can always be reduced by averaging over sufficiently long times. This procedure was used later to increase very greatly the sensitivity of the method; it is characteristic of the present possibilities that my collaborators have succeeded in the last few years in detecting in natural water signals arising from deuterium and from the isotope of oxygen with atomic mass 17, despite their low abundances of 0.02 and 0.04 per cent, respectively.

The existence and detection of a precessing nuclear polarization in a sample represents to my mind the basis of nuclear induction. It is, however, necessary to consider also the features which produce and counteract the tilt of the polarization with respect to the constant field. Magnetic resonance enters here as the most important means of producing the tilt, since it allows its achievement under the application of relatively weak oscillating fields. In fact, it is a common feature of every resonance phenomenon that relatively weak external forces can produce large effects if their frequency is equal to the natural frequency of the system to which they are applied. The natural frequency in question is, in our case, that with which the nuclear polarization precesses by itself around the constant field, and the practical way to determine this frequency is to vary either that of the applied alternating field or the magnitude of the constant field until resonance conditions are established and detected by a maximum of the observed nuclear induction signal. The simultaneous knowledge of resonance field and frequency then directly yields, as in the use of magnetic resonance in molecular beams, the gyromagnetic ratio and, with a knowledge of the spin, the magnetic moment of the nucleus. Actually, it is also possible to determine the spin separately by using the additional piece of information contained in the intensity of the observed signal.

To follow the analog of mechanical resonance we must now come back to relaxation, which can be seen to act like a friction and which counteracts the tilt produced by the alternating field. If the friction is large, i.e., if the relaxation time is short, it will either reduce the effect for a given amplitude or require a correspondingly larger amplitude of the alternating field. It will, in either case, result in a relatively broad resonance line, thus diminishing the accuracy of the measurement. While from this point of view it is undesirable to have too short a relaxation time, it is equally undesirable to have it too long, since the very circumstance of producing its tilt diminishes the magnitude of the polarization so that it requires the refreshing influence of the relaxation mechanism to bring it back to its equilibrium value.

There was not much known about the magnitude of nuclear relaxation times when Purcell and I started our first experiments on nuclear induction, and the main doubt about their success arose from the possibility of insufficient relaxation. In fact, it seems, in retrospect, that the failure of Gorter's first attempt (*6*), as well as of a second one, undertaken in 1942 (*14*), was primarily due to this circumstance. While E. M. Purcell, H. C. Torrey, and R. V. Pound (*15*), toward the end of 1945, obtained their first positive results from protons in paraffin, the late W. W. Hansen, M. E. Packard, and I (*16*) found ours a few weeks later in water without either group knowing anything about the work of the other. The relaxation time of paraffin has the convenient value of about 1/100 second, whereas pure water has a somewhat unfavorably long relaxation time of about 2 seconds. Neither of these two values had been foreseen, and I was fully prepared to find the relaxation time of pure water considerably longer and, in fact, too long for our method of observation. It was known, however, that the conversion of ortho- and parahydrogen was accelerated by the presence of paramagnetic atoms and molecules; this mechanism has the common feature, with the attainment of the equilibrium polarization of protons, that it requires a random process of nuclear reorientation, and it had been understood to take place through the magnetic field of the paramagnetic catalyst acting upon the magnetic moment of the proton. An estimate showed that, depending upon the concentration of a paramagnetic salt dissolved in water, a wide range of relaxation times, going down to values of the order of 10^{-5} second, could be obtained. Before starting our observations we therefore had prepared solutions of the paramagnetic iron nitrate in water, and although the first weak signals were received from pure water we found, shortly afterward, considerably stronger signals from a solution with about one-half molar concentration of iron nitrate. The signals appeared as rather broad lines on the cathode-ray oscillograph because of insufficient homogeneity of the constant magnetic field.

Since the width of the resonance line determines the accuracy with which magnetic moments can be determined, we shall briefly consider the conditions necessary for obtaining sharp lines. In the first place, it is necessary that the constant field have the same value in all parts of the sample; in the second place, one must not choose an excessive amplitude of the alternating field, since this too would cause an excessive broadening. The ultimate limit is given by the

natural width of the line, and it is closely related to the relaxation time; it can be seen, in fact, that the relative accuracy of a measurement by nuclear induction, due to the natural line width, is limited to the order of the number of cycles which the nuclear polarization carries out in its precession around the constant field during the relaxation time. As an example, we shall again consider protons in pure water and in a field of 10,000 gauss; the frequency of precession is here 42.5 megacycles per second, so that about 10^8 cycles are performed during the relaxation time of approximately 2 seconds. This means that an accuracy of about 1 part in 100 millions could be, in principle, achieved here, provided that one had a sufficiently homogeneous field available. Although this limit has not yet been reached, it is noteworthy that in water, alcohol, and other liquids, resolutions of one part in 10 millions have actually been achieved. It is indeed the possibility of coherent observation over a large number of cycles which allows the use of nuclear induction as a method of high precision measurements. In fulfillment of my original plans, it was applied by H. H. Staub, D. B. Nicodemus, and myself to the magnetic moments of the proton, the neutron and the deuteron (*17*), and it resulted not only in the verification of the previously mentioned deviation from additivity in the deuteron (*13*), but in its measurement with an accuracy that is beyond the present scope of the theory of nuclear forces. It was particularly gratifying to me to obtain these results from experiments combining the polarization and magnetic resonance depolarization of neutrons with nuclear induction.

The description of nuclear induction which I have presented follows closely my own original thoughts on the subject, but it can equally well be approached from other angles. The simplest one is probably that of Gorter (*6*) in his first attempt to detect nuclear magnetic resonance. We have seen before that the alternating field tilts the nuclear polarization against the constant field. This process requires a certain amount of work which, through relaxation, will reappear in the form of heat produced in the sample. The effect in fact does not involve induction but represents pure nuclear resonance absorption; however, it would be very slight, and it has not yet been established. A second attempt of Gorter (*14*), carried out later, is based upon the fact that the nuclear paramagnetic susceptibility has a maximum for radiofrequencies, corresponding to magnetic resonance conditions; it would manifest itself in the frequency of an electric oscillator of which a coil, surrounding the sample, forms the self-inductance. This scheme is actually one of the many others that can be devised for the observation of nuclear induction and, if successful, would have represented the first demonstration of the effect. Purcell's first successful experiment involved the electrodynamical aspect of absorption in so far as its occurrence under resonance conditions was manifested through the increased loss of a

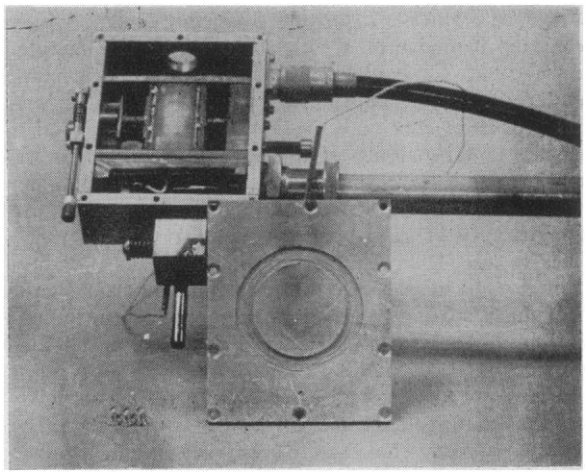

FIG. 1. The head of the crossed coil arrangement with the cover plate removed. The bottom tube to the right contains the leads to the transmitter coil which is wound in two sections visible in black in the head. The black cable leads from the receiver coil to the amplifier; the receiver coil is wound with a vertical axis inside the hollow lucite piece between the two sections of the transmitter coil. The sample test tubes are placed in its interior through the circular hole at the top of the supporting frame.

cavity resonator; the cavity was replaced in his succeeding arrangements by more conventional circuit elements. A particularly suitable and convenient ar-

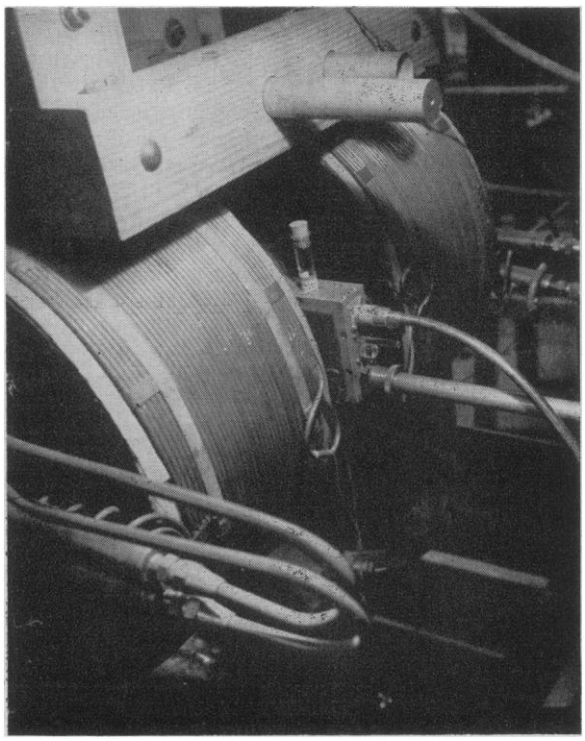

FIG. 2. The same head as in Fig. 1, about to be inserted in the gap of an electromagnet and containing a sample test tube. The two protruding lucite rods between the leads reach into the interior of the head and carry small copper disks; a fine adjustment of the coupling is achieved by rotation of these "paddles."

rangement consists of a radiofrequency bridge, which contains in one arm a coil, surrounding the sample. As a consequence of nuclear induction there occurs, under resonance conditions, a change of the impedance of this coil and thereby a readily detectable change in the balance of the bridge. It should be remarked that the change of impedance is complex, with its real part corresponding to absorption, its imaginary part to dispersion. This fact can be traced back to the phase relation between the nuclear induction signal and the applied radiofrequency field, and the phase sensitivity of the bridge allows the observation of the effect either as absorption or as dispersion or as a combination of both.

Finally, I shall give a brief description of our own original arrangement, which we still use in its principal features. The essential balance which Purcell has obtained by a bridge method is here to a large extent achieved geometrically by using two radiofrequency coils with their axes oriented at right angles to each other and to the constant field. One of them, the "transmitter coil," produces the alternating field, while the other, the "receiver coil," serves for detection of the nuclear induction signal (Figs. 1 and 2). A small amount of coupling between the two coils is admitted to produce a voltage across the receiver coil, and its phase with respect to the superimposed voltage induced by the nuclei can be adjusted for the observation of either absorption or dispersion (Figs. 3 and 4) in similarity to the bridge method.

A considerable variety of other circuits has been introduced by different investigators. Except for the greater or lesser ease of avoiding instrumental difficulties, they lead to the same ultimate sensitivity and accuracy of the method, since they all observe the same basic phenomenon.

There is, however, one distinctive feature in the crossed coil arrangement, which automatically yields another significant piece of information. The two coils imply a sense of rotation around the constant field; depending upon whether the nuclear polarization precesses in the same or the opposite sense of

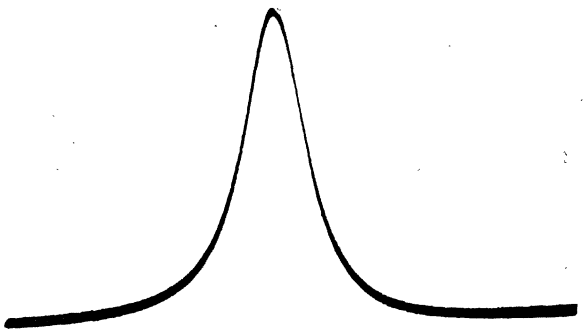

FIG. 3. A resonance line of protons in water, containing $MnSO_4$ as a paramagnetic catalyst and obtained from the phase component of the nuclear induction signal which corresponds to absorption. The photograph is that of the trace on a cathode-ray oscillograph with the vertical deflection arising from the rectified and amplified signal and the horizontal deflection corresponding to different values of the constant field.

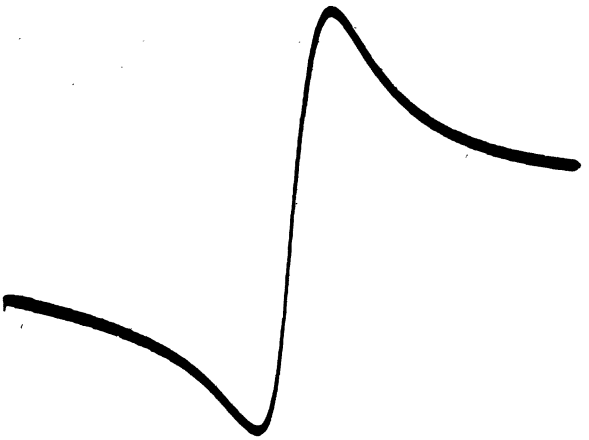

FIG. 4. The only difference between this line and that of Fig. 3 lies in the adjustment of the observed phase, which here is that corresponding to dispersion.

rotation, there results a phase difference of 180 degrees between the voltage in the receiver coil due to coupling with the transmitter coil and the superimposed voltage due to nuclear induction. The action of the rectifier translates this phase difference into an inversion of the signal, which is directly displayed on the oscillograph or on the recording instrument. One obtains in this sample manner information about the sign of the magnetic moment of the nucleus, defined by its relative orientation to the angular momentum, since it is this sign which determines the sense of rotation of the nuclear polarization in a given field. The sign of nuclear moments represents an important clue to their interpretation in terms of nuclear structures; usually it is referred to the sign of the proton moment, which has been known for a considerable time to be positive. It has been determined in this manner for a number of nuclei where it was not previously known.

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