

## Magnetic Moments of $\text{Si}^{29}$ , $\text{S}^{33}$ , $\text{Zn}^{67}$ , $\text{As}^{75}$ , $\text{Se}^{77}$ , $\text{Te}^{123}$ , and $\text{Te}^{125}$ \*†

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An improved nuclear induction spectrometer has been employed for the detection and measurement of the resonances of several stable nuclei. The sign and value of the magnetic moments are listed in Table I. No diamagnetic correction has been applied. The sensitivity of the apparatus is indicated by the photographs of the output signal of  $\text{H}^2$  and  $\text{O}^{17}$  which occur in water with a natural abundance of 0.02 and 0.04 percent, respectively.

### I. INTRODUCTION

A SPECTROMETER which employs the well-known principles of nuclear induction<sup>1,2</sup> has been used for the detection and measurement of unknown nuclear moments. Several of these measurements have been carried out with an apparatus described by Proctor<sup>3</sup> which has continued to be useful.

There are, however, instances wherein it is desirable to have greater sensitivity and stability than those available with the above-mentioned apparatus. Such instances occur particularly in the case of isotopes of relatively low abundance and large natural line widths. In order to have signal amplitudes sufficient for observation, it is not only necessary to use considerable rf amplification, but also to apply a relatively large driving rf field. Optimum conditions require, in fact, that the broadening of the lines, resulting from the rf field, is comparable to the natural line width. The combination of these two features introduces characteristic practical difficulties which must be minimized through careful design and construction of the nuclear induction head in particular and the transmitter, receiver, and power supplies in general. In the use of a spectrometer with crossed receiver and transmitter coils, it is desirable to decouple the two coils to such an extent that the voltage induced in the resonated receiver coil is of the order of a few millivolts; this voltage is referred to below as the "leakage voltage." Resulting from the thermal and mechanical instabilities, this decoupling is the more difficult to attain the larger the driving rf field, thus limiting in practice the useful range of operation of the apparatus.

A nuclear induction head has been constructed which achieves sufficient long-time mechanical stability in the presence of rf field amplitudes up to one gauss; at the same time it provides a simple and reproducible method of balancing to a minimum the induced leakage voltage.

The design details of the apparatus, as well as examples of nuclear induction signals which serve to test the behavior of the spectrometer, are given in the following section.

### II. APPARATUS

The essential principles of the present spectrometer have already been described by Proctor.<sup>3</sup> However, the design of new elements and certain other modifications merit discussion here. The new water-cooled electromagnet which supplies the electronically stabilized dc magnet field has pole faces of  $7\frac{1}{16}$  inches in diameter and a gap of  $1\frac{3}{4}$  inches and is of the double-yoke type. One particularly useful feature of this magnet lies in the fact that the poles may be tilted with respect to one another. It is thus possible to correct for any original lack of parallelism of the pole faces as well as for the distortion of the field produced by asymmetrical movement of the steel yoke under the application of the magnetic field in the gap. With this arrangement, it is quite simple to adjust the field homogeneity while observing the transients of the nuclear induction signal.<sup>4</sup> Thus fields with a homogeneity of the order of one part in hundred thousand over the sample volume of three cubic centimeters may be obtained by simple adjustment of the relative position of the pole pieces while the field is on. Since this uniformity of the dc field over the sample volume is sufficient for most purposes and relatively insensitive to field changes, no attempt has been made to shim the field.

The transmitter which supplies the rf driving field for the nuclear induction head, the head itself, and the rf receiver are illustrated schematically in the accompanying circuit diagram of Fig. 1. These units will be described in some detail in the following passages.

The variable frequency oscillator consists of a push-pull Hartley circuit employing a 6SN7 double triode which generates sufficient rf power to provide adequate grid driving current for the 832A amplifier over the entire frequency range of four to ten megacycles. The power amplifier serves both to isolate the master oscillator from load variations and to provide variable rf power to the head. The output inductance for the power amplifier is remotely located in the nuclear induction head. The air core twin-lead coaxial line serves not only

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<sup>1</sup> F. Bloch, *Phys. Rev.* **70**, 460 (1946), and Bloch, Hansen, and Packard, *Phys. Rev.* **70**, 474 (1946).

<sup>2</sup> Bloembergen, Purcell, and Pound, *Phys. Rev.* **73**, 679, 712 (1948).

<sup>3</sup> W. G. Proctor, *Phys. Rev.* **79**, 35 (1950).

<sup>4</sup> B. A. Jacobsohn and R. K. Wangsness, *Phys. Rev.* **73**, 942 (1948).

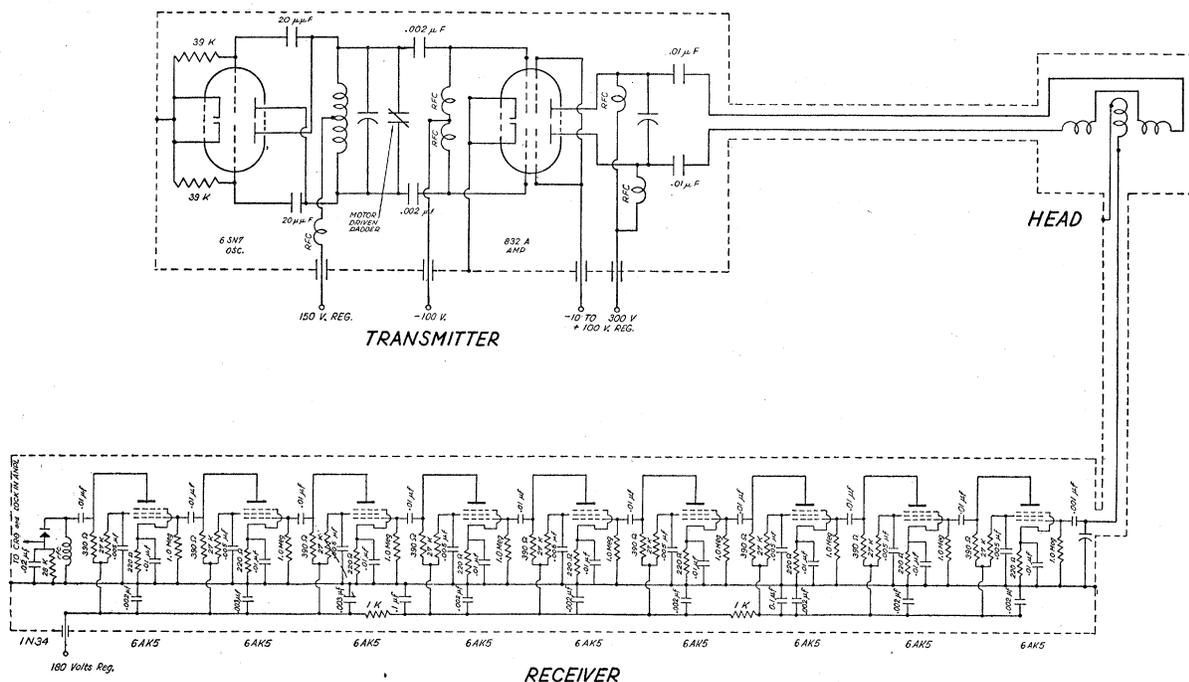


FIG. 1. Schematic diagram illustrating the transmitter, the nuclear induction head, and the rf receiver.

for the transmission of rf power, but also for the mechanical support of the head.

The intensity of the rf driving field is controlled by means of the screen voltage of the 832A amplifier tube. The rf half-amplitude  $H_1$  increases in such a way that, with approximately 300 and 150 volts on the plates of the 832A and 6SN7, respectively, and with two milliamperes driving current on the control grid of the 832A, intensities from 0.1 to 1.0 gauss are included in a range of zero to one hundred volts on the screen grid of the amplifier tube. Somewhat lower rf fields may be obtained by using negative bias on the screen grid. Owing to the fact that the receiver amplifier and the transmitter are located next to each other, care must be exercised to provide shielding such that unwanted direct

coupling between these two components does not cause an excessive leakage voltage on the final diode detector. For this reason the entire transmitter is mounted in a brass cylinder about  $5\frac{1}{4}$  inches in diameter and 12 inches long. The two end plates which support the nuclear induction head and the power feed-through condensers, respectively, are machined to fit the cylinder snugly in order to minimize the leakage (see Fig. 2). In order to reduce undesirable coupling of the rf units through the regulated power supplies, all the power for the transmitter is brought in through standard  $0.01 \mu\text{f}$  "Hypass" condensers.

The construction of the nuclear induction head is shown in the external view of Fig. 3 and in the drawing of Fig. 4. The head is of the crossed coil type as mentioned by previous authors<sup>5,6</sup> with certain alterations in design which will be mentioned below.

For an rf field intensity  $H_1$  of the order of  $\frac{1}{3}$  gauss, there exist voltages in excess of 100 volts across the transmitter coil. However, the maximum allowable induced voltage across the resonated receiver coil at the input of the amplifier is of the order of 10 millivolts, and is in fact generally less than 2 millivolts. Voltages larger than two millivolts cause, after amplification, unduly large leakage voltages at the final diode and are undesirable from the standpoint of optimum over-all signal-to-noise ratio of the apparatus. The reduction is obtained by the balancing schemes inherent in the "crossed-coil" technique. Gross decoupling between the

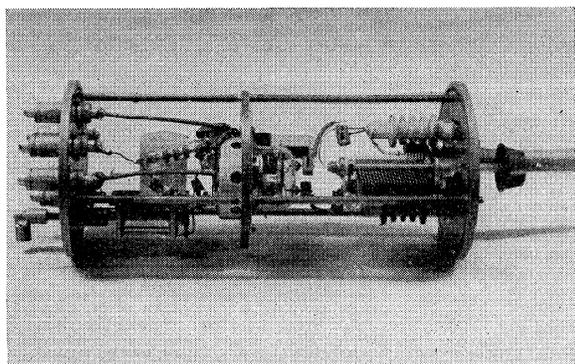


FIG. 2. The photograph illustrates the arrangement of the components of the transmitter. The external shield has been removed.

<sup>5</sup> M. E. Packard, Rev. Sci. Instr. 19, 435 (1948).

<sup>6</sup> F. Bloch and D. H. Garber, Phys. Rev. 76, 585 (1949).

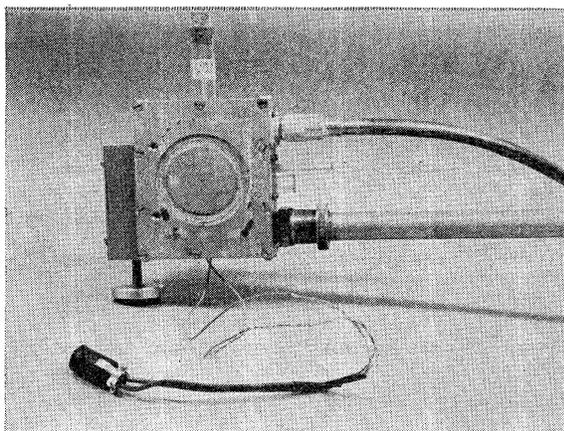


FIG. 3. External view of the nuclear induction head showing the relative position of the various leakage voltage controls. A sample test tube is shown in place.

transmitter and receiver coils is accomplished by having the axes of the coils perpendicular. Additional compensation is achieved by "tilting" the rf field with respect to the axis of the receiver coil. This is accomplished with a high degree of facility and stability by means of a differential screw drive moving a Lucite form on which half of the transmitter coil is wound. The relative movement of the two halves of the coil tilts the rf field and thereby produces complete balancing of the quadrature voltage of the rf leakage induced in the resonated receiver coil. This particular phase of the driving rf field is associated with the absorption mode, or  $v$  mode,<sup>7</sup> of the nuclear induction signals. As may be noted from Fig. 4, the transmitter and receiver coaxial lines are located at the lower and upper extremities of the head. Between these leads as well as between the receiver and transmitter coils proper are internal ground planes which further minimize any coupling to the receiver coil. With this arrangement at an rf field of about  $\frac{1}{3}$  gauss half-amplitude, the residual leakage after complete " $v$  mode" balance amounts to somewhat less

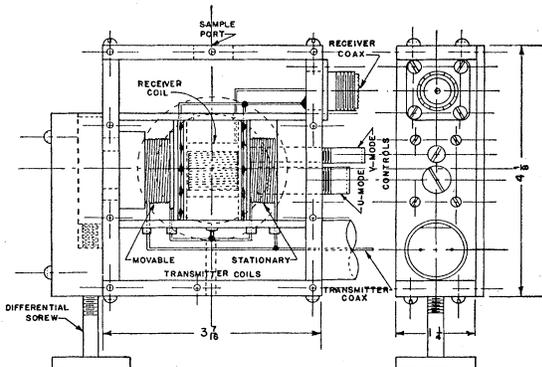


FIG. 4. View of nuclear induction head with the left cover plate removed showing the arrangement of the component parts.

<sup>7</sup> F. Bloch, *Phys. Rev.* **70**, 460 (1946).

than one-tenth of a volt. This residual leakage is due to the in-phase voltage induced in the receiver coil and is associated with the dispersion mode, or  $u$  mode,<sup>7</sup> of the nuclear induction signals. Although this remaining voltage is quite small in comparison with the voltage across the transmitter coil, it is still too large to permit realization of the maximum sensitivity of the rf amplifier. The final cancelling of the remaining leakage is accomplished by a  $u$  mode control<sup>8</sup> consisting of an inductance loop and a series resistance of such a magnitude that the total series impedance is predominantly resistive. The design of the control is simple and is schematically indicated in Fig. 5. Since the field resulting from the transmitter coil is predominantly in the  $y$  direction, the voltage induced in the circular loop, located in the  $xz$  plane, does not change upon rotation around the  $y$  axis. With the axis of the receiver coil fixed and perpendicular to the  $y$  axis, its coupling with the rectangular loop and, thereby, with the transmitter coil changes upon rotation around the  $y$  axis. The control is mounted in a threaded Lucite rod which is machined to fit very accurately the corresponding threaded hole in the main body of the nuclear induction head. An additional control provides a fine adjustment of the  $v$  mode leakage and is located just above the  $u$  mode control as shown in Figs. 3 and 4. This control consists of a small copper disk about 2 mm thick and approximately 5 mm in diameter mounted on the end of a threaded Lucite rod. The combined effect of these two controls is to affect the magnitude and phase of the "homodyning leakage" voltage applied to the input of the rf receiver. This leakage voltage is important for providing sufficient voltage for the proper operation of the crystal detector and for determining the mode of the observed resonance line.

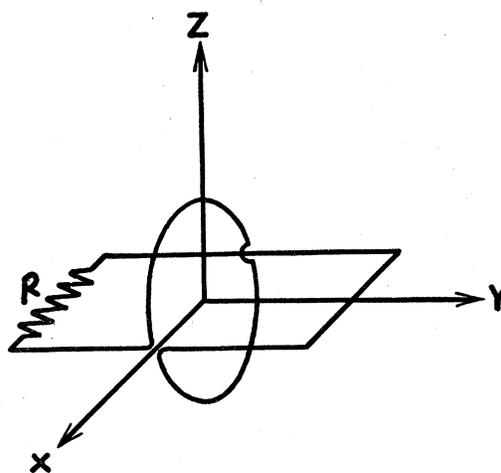


FIG. 5. Schematic diagram of the  $u$  mode control.

<sup>8</sup> This method of balance is that of F. Western at Varian Associates. The author wishes to thank Mr. Western for pointing it out to him.

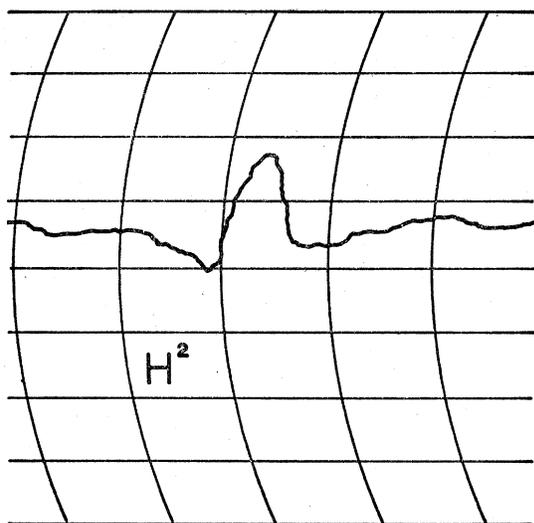


FIG. 6. Deuterium signal,  $u$  mode, in natural water with 8 molar  $\text{MnCl}_2$  added. Rf half amplitude of  $\frac{1}{3}$  gauss and sweep amplitude of  $\frac{1}{2}$  gauss were used.

It is the stability of the balancing which limits the magnitude of the rf intensity  $H_1$ , for thermal heating and sensitivity to microphonics increase rapidly with the intensity of the rf field. In the case of overnight searching when the apparatus runs unattended for many hours, the rf field,  $H_1$ , is generally held below one-half gauss. At fields of the order of one-third gauss or less, and after having warmed up, the apparatus is sufficiently stable to run without rebalance for as much as twenty hours.

The rf receiver, the circuit of which is also shown in Fig. 1, has a pass-band frequency range of four to ten megacycles. When used in conjunction with the rest of the spectrometer, it has a noise figure of  $3.0 \pm 0.6$  at an rf level in the head of 0.3 gauss. With the exception of the input coil, the receiver is untuned and the gain-bandwidth design parameters were determined according to the principles outlined in the book *Electronics* by Elmore and Sands.<sup>9</sup> There are nine stages of amplification with a gain per stage of 1.67 resulting in a total gain of 100 over a usable frequency range of 4.0 to 9.4 Mc. The maximum frequency is determined by the minimum capacitance of the tuning condenser, the capacitance of the copper coaxial line, and the distributed capacitance of the receiver coil in the head. The detected output from the 1N34 is then fed into the "lock-in" amplifier in exactly the same manner as has been described earlier by Proctor.<sup>3</sup>

There are two examples of nuclei which are readily available and whose induction signals serve as excellent tests of the sensitivity and performance of the spectrometer. They are  $\text{H}^2$  and  $\text{O}^{17}$ , both occurring naturally in water. These two nuclei have been chosen because

<sup>9</sup> W. C. Elmore and M. Sands, *Electronics* (McGraw-Hill Book Company, Inc., New York, 1949), first edition.

they are not only of low abundance, but also because they require little preparation. Both  $\text{H}^2$  and  $\text{O}^{17}$  possess small quadrupole moments which, when interacting with the molecular fields, provide a means for attaining thermal equilibrium. However, in the case of  $\text{H}^2$  it is necessary in general to add paramagnetic catalyts in order to obtain natural line widths in excess of field inhomogeneities over the sample volumes. These signals are reproduced in Figs. 6 and 7.

### III. APPLICATION OF THE SPECTROMETER

For many nuclei under investigation information is available from spectroscopic data concerning the spin and the approximate value of the magnetic moment. In order to search for the resonance of a nucleus, the frequency is fixed at some convenient value and the magnetic field is slowly varied so as to cover a range of values on both sides of the expected gyromagnetic ratio. For a given frequency setting, the total search time as determined by the clock drive of the magnet current control amounts to about thirty hours. Once the resonance has been detected on the recording tape, the field is held fixed at the point of resonance and the signal is again traced out by slowly varying the frequency on either side of the resonance. This slowly changing frequency is constantly monitored by a General Radio Model LR-1 Heterodyne Frequency Meter and in this way the frequency at which the resonance occurs can be measured to within fifty cycles. The resonance of the unknown nucleus is then compared, at the same field,

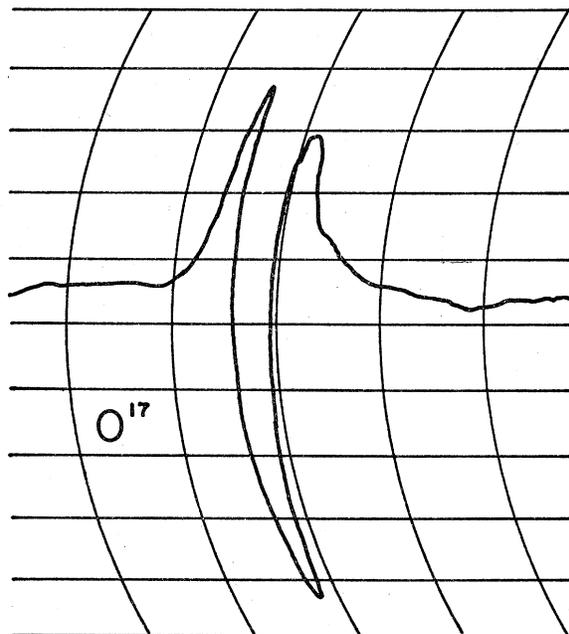


FIG. 7.  $\text{O}^{17}$  signal,  $u$  mode, in natural water without any catalyst added. Rf half-amplitude of  $\frac{2}{10}$  gauss and sweep amplitude of  $\frac{3}{10}$  gauss were used. The audio gain for this trace was reduced to  $\frac{7}{10}$  of that used in the trace of Fig. 6.

with that of a known nucleus. Typical examples of standard nuclei are  $H^2$  in heavy water,  $Na^{23}$  in NaCl, and  $N^{14}$  in  $HNO_3$ .

In order to insure that a systematic error arising from the drift of the dc field does not occur while measurements are being made, the measured values of the frequency are plotted as a function of time, and by interpolation these values can be obtained as if they were measured at the same time and hence at the same field.

In this manner the magnetic moments of seven new isotopes have been determined with high precision and are listed in Table I.<sup>10</sup> Only the frequency ratios, known spins, and the known magnetic moments of standard nuclei have been used in the computation of these values and no diamagnetic corrections have been applied.

Due to the fact that the resonance frequency is sometimes dependent upon the compound used in a sample,<sup>11</sup> the compounds employed for the actual frequency measurement have been included in the tabulation. The compounds used for the standard nuclei have been mentioned above. For the purpose of computation the value of the magnetic moment of  $Na^{23}$  was determined by taking  $\mu(H^1) = 2.79268 \pm 0.00006$  nm<sup>12</sup> and  $\nu(Na^{23})/\nu(H^1) = 0.26450 \pm 0.00003$ .<sup>13</sup> The value of the magnetic moment of deuterium used in these comparisons is  $\mu(H^2) = 0.857606$ .<sup>14</sup>

The sign<sup>3</sup> of the magnetic moment was ascertained in all cases, while the spin determinations were not carried out except as noted. In the instances where the spin of the nucleus has been measured, the phenomenological equations<sup>7</sup> for the description of nuclear induction have been applied under suitable conditions.<sup>7,15</sup>

### A. Silicon

A tentative assignment of the value of the spin<sup>16</sup> and the magnetic moment<sup>17</sup> of the  $Si^{29}$  nucleus has been reported to be  $\frac{1}{2}$  and 0.55 nm, respectively. These experimental results are consistent with the model of the nucleus proposed by Mayer<sup>18</sup> and with the value of the magnetic moment of the  $Si^{29}$  nucleus predicted by the scheme of Schawlow and Townes.<sup>19</sup>

The first nuclear induction signals of silicon were reported by Hatton *et al.*<sup>17</sup> as having been observed in beryl and also in two types of glass. Accordingly, a

<sup>10</sup> As the measurements mentioned in this section progressed, the results were published in a preliminary form: S. S. Dharmatti and H. E. Weaver, Phys. Rev. **83**, 845 (1951); **84**, 367 (1951); **84**, 843 (1951); **85**, 927 (1952); **86**, 259 (1952).

<sup>11</sup> W. G. Proctor and F. C. Yu, Phys. Rev. **77**, 717 (1950); W. C. Dickenson, Phys. Rev. **77**, 736 (1950).

<sup>12</sup> Hipple, Sommer, and Thomas, Phys. Rev. **80**, 487 (1950).

<sup>13</sup> F. Bitter, Phys. Rev. **75**, 1326 (1949).

<sup>14</sup> Supplement 1 to *Nuclear Data*, National Bureau of Standards Circular No. 499 (1951), p. 1.

<sup>15</sup> F. Alder and F. C. Yu, Phys. Rev. **81**, 1067 (1951).

<sup>16</sup> Townes, Mays, and Dailey, Phys. Rev. **76**, 700 (1949).

<sup>17</sup> Hatton, Rollin, and Seymour, Phys. Rev. **83**, 672 (1951).

<sup>18</sup> M. G. Mayer, Phys. Rev. **78**, 16 (1950); Haxel, Jensen, and Suess, Phys. Rev. **75**, 1766 (1949).

<sup>19</sup> A. L. Schawlow and C. H. Townes, Phys. Rev. **82**, 268 (1951).

TABLE I. Magnetic moments and magnetic moment ratios.

Nuclei	Spin	Compound used	Magnetic moment in nuclear magnetons
$Si^{29}$	(1/2?)	Cobalt glass	$(-0.55492 \pm 0.00004)$
$S^{33}$	3/2	$CS_2$	$+0.64292 \pm 0.00014$
$Zn^{67}$	5/2	$ZnSO_4$	$+0.87378 \pm 0.00013$
$As^{75}$	3/2	$Na_3AsO_4$	$+1.4347 \pm 0.0003$
$Se^{77}$	1/2	$H_2SeO_3$	$+0.53326 \pm 0.00005$
$Te^{123}$	1/2	$TeCl_2$	$-0.73188 \pm 0.00004$
$Te^{125}$	1/2	$TeCl_2$	$-0.88235 \pm 0.00004$
$\mu(Te^{125})/\mu(Te^{123}) = 1.20560 \pm 0.00007$			

Pyrex rod was inserted into the receiver coil of the induction head and the spectrometer set to search over a small region on either side of the reported value of 0.55 nm. Resonances were subsequently detected not only in samples containing Pyrex glass, but also in cobalt-, lead-, uranium-, vicor-, and soft glass.

A comparison of the resonant frequency of the  $Si^{29}$  signal in cobalt glass with that of deuterium in  $D_2O$  gave

$$\nu(Si^{29})/\nu(H^2) = 1.29410 \pm 0.00007.$$

Assuming the spin of  $Si^{29}$  to be  $\frac{1}{2}$ , the value of the magnetic moment is

$$\mu(Si^{29}) = -0.55492 \pm 0.00004.$$

The sign of the magnetic moment was observed to be negative. It should be pointed out that according to the shell model this observation further supports the assumption of a spin of  $\frac{1}{2}$  for  $Si^{29}$ . In fact this isotope belongs to the third shell of odd-neutron nuclei, so that only  $S_{\frac{1}{2}}$ ,  $D_{\frac{3}{2}}$ , and  $D_{\frac{5}{2}}$  states are available for the odd nucleon. Since a  $D_{\frac{3}{2}}$  state would lead to a positive moment, the spin value of  $\frac{3}{2}$  can safely be excluded. The only other possibility for a negative moment would be the  $D_{\frac{5}{2}}$  state which, however, would lead to the implausibly small value of  $-0.111$  for the magnetic moment. Although no conclusive proof has been given yet that the spin of  $Si^{29}$  is  $\frac{1}{2}$ , this value must be considered by far the most probable one.

Nuclear induction resonances with a signal-to-noise ratio of about ten have also been observed in a three molar solution of  $SiO_2$  in NaOH and in a three molar solution of silicon metal in KOH, without the addition of paramagnetic catalyst. The aspects of these signals as registered by the recording dc milliammeter indicate that in these samples the experimental conditions are not those of slow passage. Measurement of the transverse relaxation times of the protons in these same solutions<sup>20</sup> yielded values of the order of thirty milliseconds, while the normal relaxation times of the protons in the cp reagents, NaOH and HOH, are considerably longer. This suggests that the commercially prepared samples of  $SiO_2$  and silicon metal contain small amounts of paramagnetic catalysts as con-

<sup>20</sup> The author wishes to thank D. E. Maxwell of the Physics Laboratory of Stanford University for the measurement of the proton relaxation times by means of the spin-echo method.

taminants and that it is these contaminants which cause relaxation of the protons and, to a much smaller degree, also of the  $\text{Si}^{29}$  nuclei.

Attempts have been made to achieve the proper slow passage conditions which would allow a spin determination from the magnitude of the signal by adding several paramagnetic catalysts to the solutions of  $\text{SiO}_2$ . With none of the catalysts exhibiting the desired effects, these attempts of a conclusive spin determination for  $\text{Si}^{29}$  have been temporarily abandoned.

Resonances have also been observed in samples of powdered silicon metal, and these signals exhibited a slight chemical shift of about 1/5500 toward lower fields. By far the best signals with a signal-to-noise ratio of about one hundred are obtained in cobalt and lead glass. However, these samples of glass are not suitable for spin determination because of the uncertainty in the number of nuclei which actually contribute to the nuclear induction signal and because of the fact that the measured longitudinal and transverse relaxation times were considerably different.

### B. Sulfur

Eshbach, Hillger, and Jen,<sup>21,22</sup> by observing the Zeeman effect on the microwave rotational spectrum of OCS enriched in  $\text{S}^{33}$ , determined the magnetic moment of the stable odd sulfur isotope to be  $\mu(\text{S}^{33}) = 0.632 \pm 0.010$  nm. The nuclear induction resonance of  $\text{S}^{33}$  in a sample of  $\text{CS}_2$  was first observed near 3.05 Mc and in the neighborhood of 9.4 kilogauss. A comparison of the resonant frequency of  $\text{N}^{14}$  in 3.2 normal  $\text{HNO}_3$  with that of  $\text{S}^{33}$  in  $\text{CS}_2$  yielded the result

$$\nu(\text{S}^{33})/\nu(\text{N}^{14}) = 1.06174 \pm 0.00013.$$

Taking for the spin the value  $\frac{3}{2}$ ,<sup>23</sup> the magnetic moment was calculated to be

$$\mu(\text{S}^{33}) = +0.64292 \pm 0.00014.$$

The positive sign of the moment was verified by a comparison of the sign of the  $\text{S}^{33}$  signal with that of  $\text{N}^{14}$  at the same frequency and different magnetic fields.

### C. Zinc

The isotope  $\text{Zn}^{67}$  with an abundance of 4.11 percent is the only odd nucleon nucleus of the five naturally occurring isotopes of zinc. Lyshede and Rasmussen<sup>24</sup> by means of spectroscopic analysis tentatively assigned a value of 5/2 for the spin of the nucleus and also reported a value of +0.9 nm for the magnetic moment. Later Arroe<sup>25</sup> from a study of the hfs of an enriched sample confirmed the above value of the nuclear spin.

Nuclear induction signals in  $\text{Zn}(\text{NH}_3)^{++}$  were ob-

served at about 2.35 Mc and 8.84 kilogauss, with a signal-to-noise ratio of about six. A comparison of the resonant frequency of  $\text{N}^{14}$  with that of  $\text{Zn}^{67}$  and  $\text{ZnSO}_4$  yielded the result

$$\nu(\text{Zn}^{67})/\nu(\text{N}^{14}) = 0.86580 \pm 0.00001.$$

Taking for the spin the value of 5/2, the following magnetic moment results:

$$\mu(\text{Zn}^{67}) = +0.87378 \pm 0.00013.$$

The sign of the magnetic moment was determined to be positive and the nuclear spin of 5/2 was verified by careful comparison of the signal amplitudes and line widths of  $\text{Zn}^{67}$  in  $\text{ZnSO}_4$  and of  $\text{H}^2$  in 0.5 percent solution of  $\text{D}_2\text{O}$  in 8 molar  $\text{MnCl}_2$ .

### D. Arsenic

Arsenic has only one naturally occurring isotope and a spin of  $\frac{3}{2}$ .<sup>26</sup> Spectroscopically, Schuler and Marketu<sup>27</sup> measured the magnetic moment as  $1.5 \pm 0.3$  nm and also reported a nuclear quadrupole moment of  $+0.3 \times 10^{-24}$  cm<sup>2</sup>. This value of the magnetic moment would seem to be an exception to the average distribution of nuclear moments as represented by Bloch.<sup>28</sup> It was therefore advisable to attempt a more precise determination of the magnetic moment by means of nuclear induction. Owing to the rather large quadrupole moment, it was necessary to choose a compound which when dissociated would place the arsenic nucleus in a symmetrical configuration. The solution of 2 molar  $\text{Na}_2\text{HAsO}_4$  in 3 molar  $\text{NaOH}$  was employed on the assumption that  $\text{AsO}_4^{-3}$  ions of tetrahedral symmetry are formed. As a result, the gradient of the electric fields to which the arsenic nucleus is subjected would be minimized for the reason of symmetry and an excess quadrupole broadening of the line would be avoided. The nuclear resonance obtained in the above solution was compared to the sodium resonance resulting from a solution of  $\text{NaCl}$  to which 0.5 molar  $\text{MnCl}_2$  had been added as a catalyst. This comparison yielded the result

$$\nu(\text{As}^{75})/\nu(\text{Na}^{23}) = 0.64745 \pm 0.00015. \quad (1)$$

Taking the spin of  $\frac{3}{2}$ , the sign and value of the magnetic moment were found to be

$$\mu(\text{As}^{75}) = +1.4347 \pm 0.0003. \quad (2)$$

Since the publication of the above result, Staub and collaborators<sup>29</sup> have also independently measured the magnetic moment of arsenic in both  $\text{AsO}_4^{-3}$  and  $\text{AsS}_4^{-3}$  ions. The value reported by Staub *et al.* agrees within the experimental error with the value given in Eq. (2).

<sup>21</sup> Eshbach, Hillger, and Jen, *Phys. Rev.* **80**, 1106 (1950).

<sup>22</sup> Eshbach, Hillger, and Strandberg, *Phys. Rev.* **85**, 532 (1952).

<sup>23</sup> C. H. Townes and S. Geschwind, *Phys. Rev.* **74**, 626 (1948).

<sup>24</sup> J. M. Lyshede and E. Rasmussen, *Z. Physik* **104**, 375 (1937).

<sup>25</sup> O. H. Arroe, *Phys. Rev.* **74**, 1263 (1948).

<sup>26</sup> Dailey, Rusinow, Shulman, and Townes, *Phys. Rev.* **74**, 1245 (1948).

<sup>27</sup> H. Schuler and M. Marketu, *Z. Physik* **102**, 703 (1936).

<sup>28</sup> F. Bloch, *Phys. Rev.* **83**, 839 (1951).

<sup>29</sup> Staub, Jeffries, and Loeliger, *Helv. Phys. Acta* **24**, 643 (1951) and *Phys. Rev.* **85**, 478 (1952).

### E. Selenium

Of the six naturally occurring isotopes, selenium has only one odd isotope  $\text{Se}^{77}$ , with an abundance of about 7.5 percent. The spin of this nucleus has recently been determined to be  $\frac{1}{2}$ ,<sup>30</sup> which is the value predicted by the shell model of the nucleus. According to this model, the odd nucleon would be in the  $P_{1/2}$  level and hence would exhibit a positive magnetic moment. Taking the Schmidt value of 0.64 nm as the upper limit for the magnetic moment of  $\text{Se}^{77}$ , the spectrometer was set to search over a range of values extending as far below this limit as possible. Since the spin of the selenium isotope is known to be  $\frac{1}{2}$ , the question arises concerning the addition of a paramagnetic catalyst. Normally for nuclei with spin  $\frac{1}{2}$ , it is necessary to add a catalyst in order to enhance the establishment of thermal equilibrium. However, in this case, the catalyst was not only unnecessary but actually deleterious in the sense that the effect of the catalyst was to cause a less favorable ratio of the relaxation times  $T_1$  and  $T_2$ .

The fact that relatively narrow lines were observed in a solution of  $\text{H}_2\text{SeO}_3$  in water, not containing any catalyst, can possibly be attributed to the slight action of the paramagnetic monatomic selenium in acid solutions. The presence of a paramagnetic catalyst was supported by the observed fact that it not only affected the relaxation times of the selenium nuclei, but also shortened significantly the relaxation times of the sodium nuclei which were added to the selenous acid for the purpose of comparing the resonant frequencies.

A comparison of the resonant frequency of  $\text{Se}^{77}$  with that of  $\text{Na}^{23}$  gave the result:

$$\nu(\text{Se}^{77})/\nu(\text{Na}^{23}) = 0.72193 \pm 0.00002.$$

With the known magnetic moment of  $\text{Na}^{23}$  and the fact that the spin of  $\text{Se}^{77}$  is  $\frac{1}{2}$ , the sign and value of the magnetic moment were found to be

$$\mu(\text{Se}^{77}) = +0.53326 \pm 0.00005.$$

### F. Tellurium

Tellurium has two odd isotopes  $\text{Te}^{123}$  and  $\text{Te}^{125}$ , which occur naturally with an abundance of 0.85 percent and 7.0 percent, respectively. The spins of these isotopes have been measured as  $\frac{1}{2}$ .<sup>31,32</sup> Mack and Arroe<sup>32</sup> from their studies of hfs reported that the ratio of the magnetic moments of the isotopes is 1.208 with a maximum deviation of 5 percent. Recently Ross and Murakawa<sup>33</sup> have reported the magnetic moments to be

$$\mu(\text{Te}^{125}) = -0.7 \pm 0.2 \text{ nm}$$

and

$$\mu(\text{Te}^{123}) = -0.6 \pm 0.2 \text{ nm}.$$

Using a 3.1 molar solution of  $\text{TeO}_2$  in HCl and also one of cp tellurium metal in aqua regia, signals of  $\text{Te}^{125}$  and  $\text{Te}^{123}$  were observed in both the above solutions without the addition of paramagnetic catalyst.

The resonant frequencies of  $\text{Te}^{125,123}$  were compared with that of  $\text{Na}^{23}$  in NaCl, with the result

$$\nu(\text{Te}^{125})/\nu(\text{Na}^{23}) = 1.19457 \pm 0.00004$$

and

$$\nu(\text{Te}^{123})/\nu(\text{Na}^{23}) = 0.99085 \pm 0.00003.$$

Taking the value of the spin of the two isotopes to be  $\frac{1}{2}$ , the signs and values of the magnetic moments of  $\text{Te}^{125,123}$  are, respectively,

$$\mu(\text{Te}^{125}) = -0.88235 \pm 0.00004$$

and

$$\mu(\text{Te}^{123}) = -0.73188 \pm 0.00004.$$

The ratio of the magnetic moments of the two isotopes is therefore calculated to be

$$\mu(\text{Te}^{125})/\mu(\text{Te}^{123}) = 1.20560 \pm 0.00007,$$

which is in good agreement with the original value obtained by Mack and Arroe.<sup>32</sup>

If the leakage voltage of the spectrometer is adjusted for the observation of the absorption mode of the nuclear resonance, the tellurium signals in solutions of  $\text{TeCl}_2$  are found to saturate at a half-amplitude of the rf field of about 0.06 gauss. The application of the phenomenological equations to this case and the measurement of the line widths (for the most part determined by the field inhomogeneity over the sample) yielded for the ratio of  $T_1/T_2$  the approximate value of 12. The fact that slow passage conditions were achieved in these solutions with nuclei of spin  $\frac{1}{2}$  suggests that there are paramagnetic contaminants in the chemical reagents. It is known that there are small traces of iron in every stock solution of cp HCl which might possibly account for the relatively short relaxation time. However, it is also known that there exists monatomic tellurium<sup>34</sup> in acid solutions and further that these atoms are likely to be paramagnetic in analogy with oxygen. This situation is similar to that encountered in the case of  $\text{Se}^{77}$  (see Part E of this section), which belongs to the same group of the periodic table. The nuclei of both elements had sufficiently short relaxation times to exhibit good signals in spite of the fact that different solvents were used in the two cases. This result indicated, therefore, that the relaxation in this case is probably due to the action of paramagnetic atoms rather than of possible contaminants. Another similarity of tellurium with selenium lies in the observed fact that the addition of manganese ions with a few tenths molar concentration was sufficient to diminish the signal amplitudes to a point where they could no longer be detected.

The first tellurium signals were detected in a sample

<sup>30</sup> S. P. Davis and F. A. Jenkins, *Phys. Rev.* **83**, 1269 (1951).

<sup>31</sup> F. Bitter, *Phys. Rev.* **75**, 1326 (1949).

<sup>32</sup> C. R. Fowles, *Phys. Rev.* **76**, 571 (1949); **78**, 744 (1950); J. E. Mack and O. H. Arroe, *Phys. Rev.* **76**, 1002 (1949).

<sup>33</sup> J. S. Ross and K. Murakawa, *Phys. Rev.* **85**, 559 (1952).

<sup>34</sup> *Textbook of Inorganic Chemistry*, edited by J. Newton Friend (Charles Griffin and Company, Ltd., London, 1931), Vol. VII, Part 2, p. 365.

of powdered tellurium metal. Under the conditions of slow passage and with sweep amplitudes small compared to the natural line width of the nuclear induction signals, the output voltage of the spectrometer as registered by the dc milliammeter is the derivative of the slow passage signal. Under these conditions the area under the trace is zero. The aspect of the signals in metallic tellurium indicated that the experimental conditions of the metal were not those of slow passage; in addition, it was not possible to detect an absorption mode with the lowest available half-amplitude of the rf field of about 0.01 gauss. Both facts indicate that the longitudinal relaxation time in the metal is compar-

tively long. No observable chemical shift was detected between the resonant frequency of  $\text{Te}^{125}$  in the pulverized metal and in the solutions.

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## Atomic Excitation and Ionization Accompanying Orbital Electron Capture by Nuclei\*

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The process of atomic excitation and ionization accompanying orbital electron capture by nuclei is treated on the basis of the general theory of  $\beta$ -decay using a configuration space representation for both nucleons and leptons. Quantitative expressions are obtained (a) for the total number of double holes produced in the  $K$ -shell due to  $K$ -capture accompanied by excitation or ejection of the other  $K$ -electron [Eq. (15)], and (b) for the total probability of electron ejection in orbital electron capture together with the ejected electron momentum spectrum [Eqs. (17)–(18)]. A discussion is given of the possibilities of experimental verification of the theory developed.

### I. INTRODUCTION

IN the customary physical description of orbital electron capture, one views the process as involving the transformation of one of the atomic orbital electrons (most probably from the  $K$ -shell) into an emitted neutrino with the simultaneous transformation of one of the nuclear protons into a neutron. In this description, no other particles are considered to be emitted, and the other electrons in the atom are presumed to remain in their original orbits, adjusted from the charge of the parent nucleus  $Z_i$  to that of the daughter  $Z_f = Z_i - 1$ . Actually, however, a certain nonvanishing probability exists (a) for photon emission as a result of the charge acceleration involved in the orbital electron capture<sup>1</sup> and (b) for the excitation of one (or more) of the noncaptured orbital electrons into unoccupied atomic bound<sup>2</sup> states and into unbound states as a consequence of the nuclear charge alteration as conditioned by the electron-electron Coulomb interaction. In a certain proportion of the orbital electron captures then, the emitted neutrino is accompanied by another particle, *viz.*, a photon or a previously bound electron

with which the neutrino must share the available energy. The energy release in orbital electron capture, which is otherwise not directly obtainable, may then be found by measuring the maximum energy carried off by the photon or by the ejected electron. Such a measurement has indeed been carried out recently for the case of photons associated with orbital electron capture in  ${}_{26}\text{Fe}^{55}$ ,<sup>3,4</sup> and the energy release in the transition accurately determined.

In the present paper we shall calculate (a) the total number of double holes produced in the  $K$ -shell due to the transformation of one of the  $K$ -electrons into the emitted neutrino and the excitation of the other  $K$ -electron into an unoccupied bound<sup>2</sup> or into an unbound (ejected) state, and (b) the total probability of (other) electron ejection in orbital electron capture together with the ejected electron momentum distribution. Our work is closely related to the work of Migdal<sup>5</sup> and of Feinberg<sup>6</sup> on the ejection of atomic orbital electrons during nuclear negatron decay.<sup>7</sup>

<sup>3</sup> Bell, Jauch, and Cassidy, *Science* **115**, 12 (1952).

<sup>4</sup> D. Maeder and P. Preiswerk, *Phys. Rev.* **84**, 595 (1951); see also the work on  ${}_{18}\text{A}^{37}$  by Anderson, Wheeler, and Watson, *Phys. Rev.* **87**, 668 (1952).

<sup>5</sup> A. Migdal, *J. Phys. (U.S.S.R.)* **IV**, 449 (1941).

<sup>6</sup> E. L. Feinberg, *J. Phys. (U.S.S.R.)* **IV**, 423 (1941).

<sup>7</sup> See also the careful discussion of atomic electron excitation and ejection in the negatron transition  ${}_{2}\text{He}^6 \rightarrow {}_{3}\text{Li}^6$ , by A. Winther,

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<sup>1</sup> P. Morrison and L. Schiff [*Phys. Rev.* **58**, 24 (1940)] give the theory.

<sup>2</sup> The binding is virtual in all cases of interest (see reference 11).

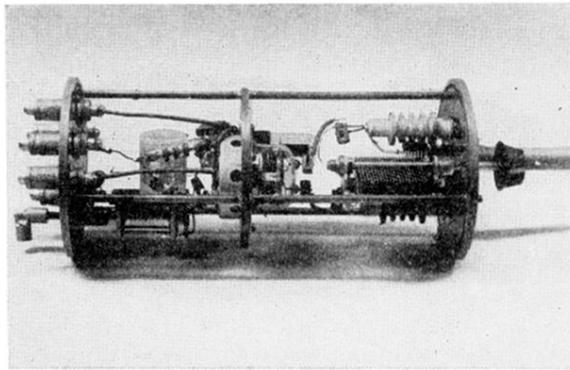


FIG. 2. The photograph illustrates the arrangement of the components of the transmitter. The external shield has been removed.

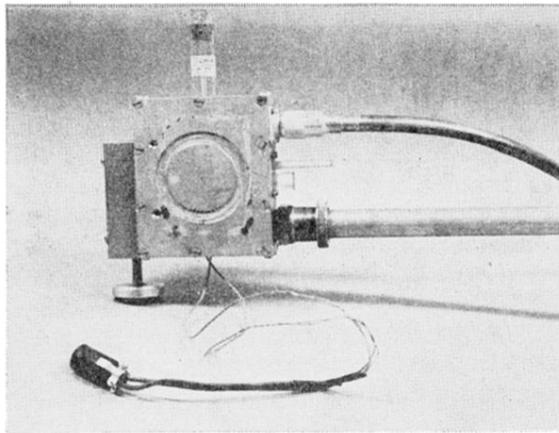


FIG. 3. External view of the nuclear induction head showing the relative position of the various leakage voltage controls. A sample test tube is shown in place.