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On the Magnetic Moments of Tl^{203}, Tl^{205}, Sn^{115}, Sn^{117}, Sn^{119}, Cd^{111}, Cd^{113}, and Pb^{207}*,†

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A radiofrequency spectrometer for observing nuclear induction signals is described. The use of crossed radiofrequency coils allows, in a simple fashion, the determination of the signs of the magnetic moments. The spectrometer has been used to determine the magnetic moments and magnetic moment ratios given in Table II. These values are based on a proton moment, without diamagnetic correction, of (2.7935±0.0003) nuclear magnetons.

I. INTRODUCTION

THE phenomenon termed "nuclear magnetic resonance absorption"[1] or "nuclear induction"[2] by the two originating groups at Harvard and Stanford, respectively, has certain characteristic advantages for the observation of nuclear moments. One of them, its high accuracy, is of particular interest for the results obtained on the lightest nuclei,[3–8] since they can be readily interpreted in terms of nuclear forces. Another advantage lies in the fact that it can be applied to practically any element, and it is this feature which has been partially used in the work presented here, on the magnetic moments of Tl^{203}, Tl^{205}, Sn^{115}, Sn^{117}, Sn^{119}, Cd^{111}, Cd^{113}, and Pb^{207}.

In the design of apparatus used to measure a variety of nuclei two principal aspects must be kept in mind. The first is that, due to sample limitations such as low natural abundances, limitations in solubility, or unfavorable relaxation times, signals may occur with amplitude less than the noise contained in a convenient audio band-width within which nuclear induction signals are usually amplified. Through a radiofrequency amplification stage, preceding the rectifier and subsequent audio amplifier, it is possible to reduce the noise, due to shot effect, to a point at which the Johnson noise of the receiving circuit represents substantially the only limitation. A further reduction of the noise can then be obtained only by extending the observation over a longer time so that the noise fluctuations are largely canceled out; a practical method for achieving this aim is the use of a narrow band width. A second aspect is due to the circumstance that the magnetic moments and spins may only be approximately known or not known at all, so that the frequency of resonance cannot be anticipated. This calls for an automatic apparatus which can be made to examine a given range of radiofrequencies while making a record of any resonance that may occur. An instrument combining these two features was first devised and used successfully by Pound, and we have, in some respects, followed his design.[9]

Through mutual exchange of experience and parallel improvements, the originally rather different techniques employed at Harvard and Stanford have now developed to a point at which they are similar in many respects. We should like, however, to emphasize one of the main differences: While in the Harvard technique one observes the voltage induced by the precession of the nuclear moments in the same coil which provides the driving r-f field, a separate "receiver coil," wound at right angles both to the driving "transmitter coil" and to the d.c. magnetic field, is used in the Stanford technique for the observation of nuclear induction. It is primarily a matter of habit and experience which of the two arrangements one considers to be the simpler and easier to operate. The crossed coil arrangement has, however, the undeniable advantage that it gives directly the sign of the magnetic moment. This arises from the circumstance that the crossed coils intrinsically imply a sense of rotation around the d.c. field, a feature which the single coil arrangement does not possess. The fact that the sign of a nuclear moment is one of the relevant pieces of information to be obtained seemed to us to be a sufficient justification for our preference of crossed coils. The comparison of signs is one of the simple and regular routines which have been carried out in the performance of the measurements presented here.

Throughout this work we have used liquid samples with the advantage that narrow lines can usually be expected and that the addition of paramagnetic catalysts for the rapid establishment of thermal equilibrium is relatively simple. The chemical composition of the solute and that of the catalyst have been chosen in every case to obtain solutions of appropriate concentration.

To measure the gyromagnetic ratio of a nucleus, its resonance frequency is compared in the same field

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† Assisted by the joint program of the AEC and ONR.

[1] Purcell, Torrey, and Pound, Phys. Rev. **69**, 37 (1946).

[2] Bloch, Hansen, and Packard, Phys. Rev. **69**, 127 (1946).

[3] H. L. Anderson and A. Novick, Phys. Rev. **71**, 372 (1947).

[4] Bloch, Graves, Packard, and Spence, Phys. Rev. **71**, 373, 551 (1947).

[5] Bloch, Levinthal, and Packard, Phys. Rev. **72**, 1125 (1947).

[6] Bloch, Nicodemus, and Staub, Phys. Rev. **74**, 1025 (1948).

[7] H. L. Anderson and A. Novick, Phys. Rev. **73**, 919 (1948).

[8] H. L. Anderson, Phys. Rev. **76**, 1460 (1949).

[9] R. V. Pound, Phys. Rev. **72**, 527, 1273 (1947); **73**, 523, 1112 (1948). The essential feature of Pound's apparatus, the use of a self-excited oscillating receiver of special design, is not incorporated in our spectrometer.

with that of another nucleus, the gyromagnetic ratio of which is known. The two frequencies would be in proportion to their gyromagnetic ratios, if it were not for the diamagnetic field corrections, due to atomic electrons, which are only known with limited accuracy. A summary of results, presented in Table II, has not included any diamagnetic correction in the computation for magnetic moments; instead, the magnitude of this correction has been given separately, in order to keep the experimental and the theoretical values distinct.

The uncertainty of the diamagnetic field corrections, however, does not enter into the calculation for the ratio of the gyromagnetic ratios of isotopes, since the same percentage corrections apply here for each. The elements whose nuclei have been chosen for measurement (except Pb^{207}) have been those which have isotopes of the same spin, so that the relatively small effect on the magnetic moment by adding two neutrons to a nucleus may be measured with good accuracy. Tin is the only element which has three stable isotopes of substantial abundance, Sn^{115}, Sn^{117}, and Sn^{119} each having a spin $\frac{1}{2}$ and differing in turn by two neutrons. For this reason, this sequence was considered particularly interesting; together with the cadmium isotopes Cd^{111} and Cd^{113} one obtains a sequence of five nuclei differing in turn by two nucleons.

II. APPARATUS AND METHOD

A. Outline of Spectrometer Design

The operation of the spectrometer is best understood with the aid of the block diagram, given in Fig. 1. The rectifier supplies current for the magnet; this current is regulated by hard tubes in the regulator. The d.c. magnetic field produced is modulated at a rate of 92.5 c.p.s. by the driver unit, which supplies current at this frequency to coils mounted on the nuclear induction head, located between the pole faces of the magnet. The head, described in greater detail below, contains a transmitter coil which is part of the plate impedance of a radiofrequency oscillator. The head also contains a tuned receiver coil, perpendicular to the transmitter coil and to the magnetic field, in which the nuclear signals are induced. The induced voltages are first amplified by a radiofrequency amplifier; after rectification by a diode, the nuclear induction signals are further amplified at audiofrequencies by an amplifier, built in the oscilloscope, and presented as vertical deflections on the screen. The horizontal deflection of the oscilloscope trace is provided by a voltage derived from the driver and therefore of the same frequency as the modulating field. If, for a given radiofrequency, the range of the modulated magnetic field includes the resonance value, there will thus be traced upon the screen two resonances during each cycle of the field modulation. Except for the introduction of the radiofrequency amplification stage preceding the rectifying diode, the method described so far is largely the same as that explained in detail in the original paper of Bloch, Hansen, and Packard.[10]

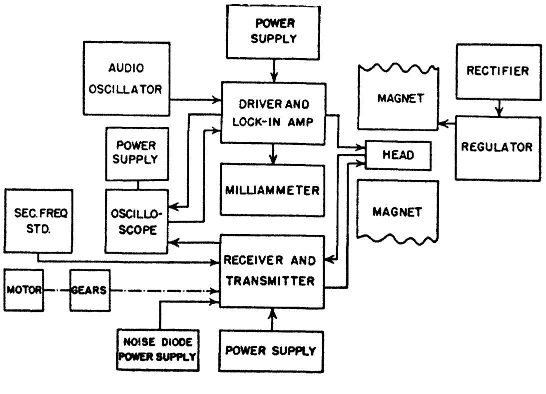

FIG. 1. Block diagram of the spectrometer.

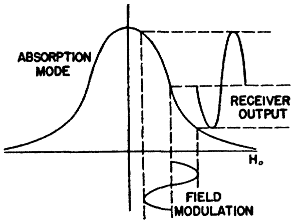

FIG. 2. Absorption mode for slow passage, illustrating receiver output for a sinusoidal field modulation.

The nuclear induction signals, in order to be observable on the oscilloscope screen, must be larger than the noise also presented there. If, for reasons such as those given in the introduction, this is not the case, they can nevertheless be made visible by taking a photograph of the oscilloscope screen, making a long exposure so that the random noise fluctuations are smoothed out.[11] Another alternative is based upon reduction of the band width; it is this method, first outlined by Pound[9] and by Bloembergen, Purcell, and Pound,[12] which we have followed and which is described below.

The radiofrequency flux intensities from the nuclei in the receiver coil can be resolved into two characteristic phase components, one in phase with the impressed radiofrequency field and one 90° behind it; the latter corresponds to dispersion and is called the u-mode, while the former is the v-mode and corresponds to absorption.[13] Either mode can be observed by homodyning the nuclear induction voltage with a radiofrequency voltage which is large compared to the maximum amplitude of the former and of the same phase as the desired mode.[12] Described as a function of the radiofrequency deviation from resonance $\Delta\omega$, the amplitude of each mode has its characteristic shape and magnitude. The work described in this paper has been done using the v-mode signal under slow passage conditions described by Bloch who derived the formula

$$v=M_0\frac{\gamma H_1T_2}{1+(\Delta\omega T_2)^2+\gamma^2H_1{}^2T_1T_2}, \tag{1}$$

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[10] Bloch, Hansen, and Packard, Phys. Rev. **70**, 474 (1946).

[11] F. Bloch and D. H. Garber, Phys. Rev. **76**, 585 (1949).

[12] Bloembergen, Purcell, and Pound, Phys. Rev. **73**, 679 (1948).

[13] F. Bloch, Phys. Rev. **70**, 460 (1946).

where M_0 is the equilibrium polarization per unit volume of the sample, H_1 the half-amplitude of the exciting radiofrequency field, T_1 and T_2 the longitudinal and transverse relaxation times, respectively, and γ the gyromagnetic ratio of the nucleus in question.

In Fig. 2 we have plotted the *v*-mode as it would appear upon slow, monotonic variation of the d.c. field H_0 in the neighborhood of resonance. If, instead, the field is modulated sinusoidally over a relatively small region as indicated, one will obtain from the audio output of the receiver a signal which contains principally the first harmonic of the modulating frequency. It appears with the largest amplitude and opposite phase if the center of the modulation is located near one or the other of the two points of inflection of the *v*-mode, and its first harmonic content is here comparable to the maximum of this mode, if the sweep amplitude is chosen to be of the order of the line width. The observation of the first harmonic alone gives under these conditions practically the full magnitude of the available signal, while sweep amplitudes which are either small or large compared to the line width result in an appreciable loss of signal magnitude.

The output of the receiver is analyzed, as indicated in Fig. 1, by a "lock-in amplifier"[14] for the presence of voltages of the sweep frequency, resulting in a direct current whose magnitude is proportional to the amplitude and whose sign depends on the phase of the input voltage. This current is smoothed by an *RC*-filter, and it is registered on a chart by a recording, d.c. milliammeter. If the frequency response of the recording milliammeter is sufficiently high, one has an output band width of the spectrometer given by $1/\pi RC$, where *RC* is the product of the resistance *R* and capacitance *C* determining the final output time constant. With a noise voltage proportional to the square root of the pass-band width, one thus obtains upon the milliammeter record a signal-to-noise ratio equal to (signal-to-noise ratio present at the receiver output) $(\pi RC \times \text{receiver-band width in c.p.s.})^{\frac{1}{2}}$ (α), where α is a number less than unity which has been introduced to account for the loss of signal described above. By utilizing this method within practical limits, a large improvement in the signal-to-noise ratio can be realized.

If the transmitter and receiver are tuned together while the frequency is varied slowly and the magnetic field remains fixed, the recording milliammeter will record the presence of a resonance. In this way the apparatus may be used to search for nuclear resonances. It is only necessary that the rate at which the radiofrequency is changed is such that the time which it takes to vary the frequency through a line width is long compared to *RC*. With a sweep amplitude comparable to the line width one records, in this way, essentially the derivative of the resonance line as a function of the frequency.

It was pointed out in the introduction that the use of crossed transmitter and receiver coils enables the spectrometer to distinguish the sign of nuclear moments. This is due to the fact that the phase relation between the nuclear signal induced in the receiver coil and the radiofrequency voltage across the transmitter coil is inverted for nuclear moments with opposite sign. Superimposed in a linear detector upon the homodyning "local oscillator" voltage derived from the driving radiofrequency field, the radiofrequency nuclear induction signal manifests this phase relation through the sign of the audiofrequency signal, which, according to Fig. 2, will determine the sign of the recorded trace. Hence, if two nuclei are observed, leaving all radiofrequency and sweep-frequency conditions unchanged and achieving their respective resonances through a

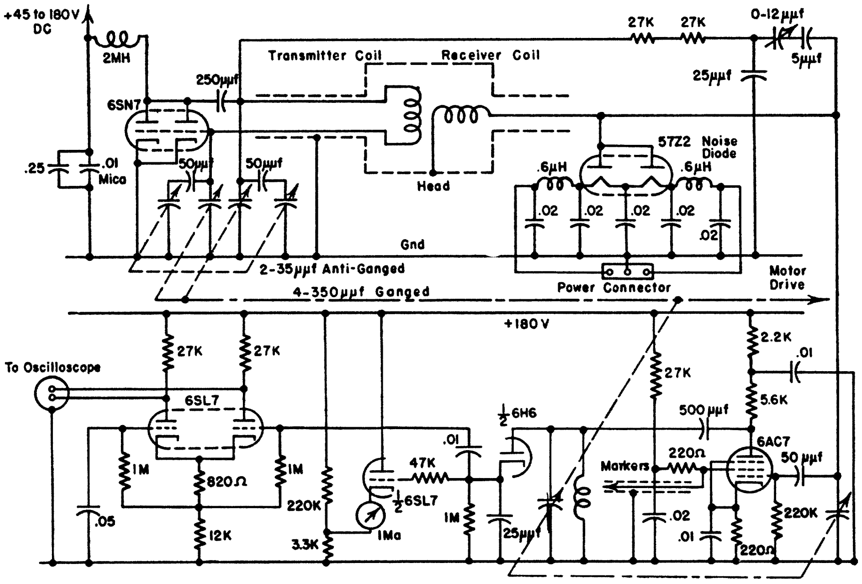

FIG. 3. Circuit diagram of the transmitter and the receiver.

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[14] R. Dicke, Rev. Sci. Inst. 17, 268 (1946).

mere change of the d.c. field, the relative sign of their recorded signals will tell directly whether the signs of their magnetic moments are the same or opposite.

With the sign of the moment manifested in the manner described and the gyromagnetic ratio given by the value of the resonance frequency, there remains one more important piece of information, furnished by the magnitude of the signal. Indeed, the voltage induced in the receiver coil by the nuclei is, among other factors, strongly dependent upon their spin. Its value could be ascertained from the magnitude of the recorded signal by calibrating the total gain of the radiofrequency, audio, and d.c. circuits. A simpler procedure is, however, to compare the signal magnitudes from the nucleus of type *a* under observation and from a nucleus of type *b* with known spin by merely changing the value of the field H_0, leaving all other conditions of observation unchanged, as in the determination of the relative signs mentioned. Indeed, if H_1 is made small, so that $\gamma^2 H_1^2 T_1 T_2 \ll 1$, and if the signals are modulated with a small amplitude of magnetic field, it follows from Eq. (1) and the Curie expression for M_0 that

$$r = [n_a I_a(I_a+1)\gamma_a^3 T_{2a}^2]/[n_b I_b(I_b+1)\gamma_b^3 T_{2b}^2], \quad (2)$$

is the ratio of the maximum derivative of the signal from n_a nuclei of spin I_a to that of n_b nuclei of spin I_b, contained in the sample where $\gamma_{a,b}$ and $T_{2a,b}$ are their respective gyromagnetic ratios and transverse relaxation times. With I_b being known and the other quantities readily measureable, I_a can thus be determined from (2). Throughout this work we have made use of this relationship to check spin values, although in the instances observed so far this has merely served as a verification of results known previously from hyperfine structure measurements.

B. Detailed Description of Principal Components

The magnet used with the spectrometer has pole faces $7\frac{13}{16}$ in. in diameter and a gap of $1\frac{3}{4}$ in. The pole faces have been shimmed to obtain a more homogeneous field in the central region, but this procedure has not been carried out to the highest perfection. For a typical cylindrical sample contained in a test tube, 1.3 cm in diameter and 2.5 cm in length, the variation of the magnetic field over the region occupied by the sample was about 3×10^{-3} percent.

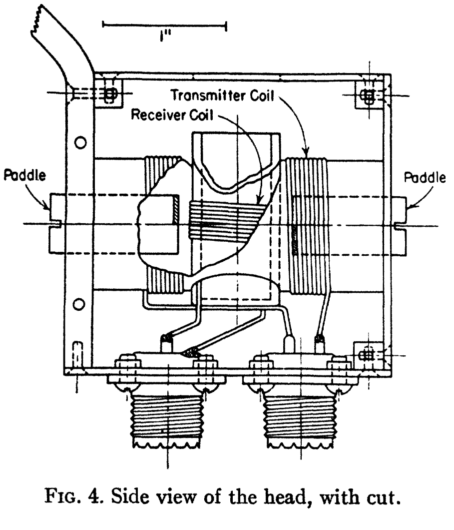

FIG. 4. Side view of the head, with cut.

There are two water-cooled windings, one around each pole piece. Each winding consists of about 17,000 turns of B and S No. 18 copper wire, so that the magnetization arises from a large number of turns rather than from a large current. This feature makes it possible to regulate the current by hard tubes, specifically eight 304*TL*'s in parallel. In the unsaturated region the magnetic field changes by 10 gauss/ma; a current of 2 amp. produces slightly less than 12 kgauss. The voltage for 2 amp. is about 3000 v, which is supplied, after transformer step-up, by full-wave rectification from the 220-v, 3-phase lines. To obtain current regulation the magnet current is passed through a variable resistor of high wattage. The difference of the voltage across this resistor and that of a 28.5-v battery is amplified in a d.c. amplifier of two stages with a total gain of 1600. The output is applied to the grids of the hard tubes, thus providing a feedback capable of reducing line voltage variations of five percent to current variations of 0.0005 percent approximately, depending on the magnitude of the variable resistor. The warming up of the magnet windings causes current drifts which, however, subside about one hour after turning on.

The essential radiofrequency components of a nuclear induction apparatus are the transmitter, providing the driving radiofrequency field, the "head" in which the sample is located, and the receiver circuit. They are diagrammatically shown in Fig. 3. The transmitting circuit is a Colpitts oscillator, with a capacitance feedback ratio almost exactly equal to two. A series of commercial *B*-batteries serves as a source of constant plate voltage variable between 45 and 180 v. The half-amplitude H_1 of the driving radiofrequency field increases with increasing voltage and can be varied between 0.13 and 0.82 gauss. The same set of batteries also supplies 180 v for the plate voltage in the receiver circuit. The inductance of the transmitter coil in the head is resonated by variable condensers ganged with the tuning condensers of the receiver. This allows a continuous frequency variation between 4.35 and 9.00 Mc and thereby in the range of d.c. field between 12,000 and 1020 gauss the observation of nuclei with gyromagnetic ratios ranging between 2.28×10^3 and 5.52×10^4 gauss^{-1} sec.$^{-1}$. As standards of comparison in this range we have used the nuclei H^1, Na^{23}, and H^2, for which the spins as well as the signs and, with high accuracy, the magnetic moments are known.

The construction of the head is shown in its side view, Fig. 4, with one shielding plate removed. It is of the type first used by Bloch and Garber[11] with the advantage over previously described forms[15] that the

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[15] M. Packard, Rev. Sci. Inst. 19, 435 (1948).

sample can be simply inserted through an opening without interruption of the operation of the apparatus. This is achieved by splitting the transmitter coil into two halves of approximate Helmholtz proportions, leaving the center free for the hollow receiver coil form, shown in the cut with an inner diameter sufficient to accommodate 15-mm test tubes. Receiver and transmitter coils are held rigidly on Lucite forms and are shielded by a silver-plated brass shell. Two Lucite "paddles,"[10] carrying semicircular copper disks at their inner ends, are used to control by their rotation the inductive coupling of the receiver coil to the transmitter coil. The head has a depth of $1\frac{1}{4}$ in.; its other dimensions are indicated by the scale given in Fig. 4. The coils which are used to modulate the field H_0 are cemented to the outside of the shielding plates, and they are likewise of Helmholtz proportions.

The receiver coil is resonated in order to enhance the induced voltage by the resonance rise factor Q. This enhanced voltage appears across a tuning condenser in the receiver, from which point it is amplified as a radiofrequency voltage, detected, and amplified again as an audio signal, the audio band width of the detector time constant and amplifier band pass being 5 kc. The radiofrequency amplifier, detector, and audio amplifier are conventional, although there are some unusual features which merit a short discussion.

There are two controls used to adjust the magnitude and phase of the radiofrequency "leakage" voltage applied to the grid of the radiofrequency amplifier for the purpose of serving as a homodyning voltage for the nuclear induction signals. The amplitude and phase of this leakage voltage is important for detector sensitivity and for determining which mode of the resonance is to be observed. The purpose of the 35-$\mu\mu$f antiganged condensers shown in Fig. 3 across two of the principal tuning condensers is to change the ratio of the capacitances of these two tuning condensers without changing their total capacitance. By this change the center point of the radiofrequency voltage across the transmitter coil can be varied without changing the frequency. Since the receiver coil is grounded at one end, its effective capacitive coupling to the transmitter coil is sensitively dependent upon the center voltage of the latter. The above-mentioned change of the 35-$\mu\mu$f condensers provides thus a variable, capacitively induced voltage in the receiver circuit. This voltage has the same phase as that across the transmitter coil and therefore controls the v-mode leakage. The RC network indicated in the upper right of Fig. 3 provides principally a potential across the capacitance which lags in phase 90° behind the potential across the transmitter coil and thus contributes to the homodyning phase component of the u-mode. A small voltage of this phase is coupled to the radiofrequency amplifier grid by means of the 0- to 12-$\mu\mu$f variable capacitance shown. By means of these two controls it is possible to observe either the pure v-mode or the pure u-mode or any mixture of them. To obtain, for example, the pure v-mode, the paddles are first adjusted to eliminate the principal part of the in-phase voltage, which is induced in the receiver coil. Proper setting of the 0- to 12-$\mu\mu$f condenser provides cancellation of the remaining u-mode leakage and the antiganged 35-$\mu\mu$f condensers, serving as a fine control for the in-phase leakage, are adjusted to provide, after amplification, a homodyning voltage of approximately 0.2 v r.m.s., sufficient for linear detection. Conversely, by using the u-mode control to obtain a sufficient voltage of the other phase component and canceling the remaining in-phase leakage by means of the v-mode control, one can observe the pure u-mode.

TABLE I. Data from the measurements on Sn^{115}.

Nucleus	Time of resonance in minutes	Resonance frequency in kilocycles
Sn^{115}	9	6236.2
	60	6236.7
	106	6236.7
	328	6236.3
	378	6236.0
Na^{23}	27	5044.9
	75	5044.7
	161	5044.6
	343	5044.5
	396	5044.5

The noise diode, shown connected directly to the receiver coil, serves the purpose of calibrating the observed noise against the known shot noise in the diode. It is inoperative when its filaments are cold, and is turned on by applying heater power through the indicated connector. For low levels of oscillation, a noise figure of four is obtained; this means that the noise power per unit band width in the spectrometer output is four times as great as that which one would expect, were there no noise other than the amplified Johnson noise from the resonated receiver coil.

The harmonics of a 25-kc multivibrator from a commercial secondary frequency standard are coupled to the screen grid of the radiofrequency amplifier. Tube currents proportional to the product of the control grid and screen grid potentials give rise to audio beats between the radiofrequency leakage from the transmitter and these harmonics. When the beat frequency is near the sweep frequency, the beats are detected by the lock-in amplifier, and a characteristic mark is made on the tape of the recording milliammeter.

The meter shown carries a current proportional to the average cathode potential of the detecting diode; it serves to register the amplified amount of leakage, and its use is important during adjustment.

The ganged receiver and transmitter capacitors are turned by a clock motor. The rate is variable, and it is chosen according to the line width of the resonance being observed and the final band width of the spectrometer. As an example, a final band width frequently used is 0.069 c.p.s.; for nuclei with a gyromagnetic ratio of 10^4 gauss^{-1} sec.$^{-1}$, providing a line of one gauss

TABLE II. Magnetic moments and diamagnetic corrections.

Nucleus	Magnetic moment in nuclear magnetons	Diamagnetic correction in percent
Cd^{111}	-0.5923 ± 0.0001	0.47
Cd^{113}	-0.6196 ± 0.0001	0.47
Sn^{115}	-0.9134 ± 0.0002	0.50
Sn^{117}	-0.9951 ± 0.0002	0.50
Sn^{119}	-1.0411 ± 0.0002	0.50
Tl^{203}	1.5962 ± 0.0003	0.98
Tl^{205}	1.6118 ± 0.0003	0.98
Pb^{207}	0.5837 ± 0.0001	1.00

Ratios of magnetic moments

$\mu(Cd^{113})/\mu(Cd^{111}) = 1.0461 \pm 0.0001$
$\mu(Sn^{117})/\mu(Sn^{115}) = 1.0894 \pm 0.0001$
$\mu(Sn^{119})/\mu(Sn^{117}) = 1.0465 \pm 0.0001$
$\mu(Tl^{205})/\mu(Tl^{203}) = 1.0098 \pm 0.0002$

between points of maximum slope, a sufficiently slow rate is 2 kc/min. Searching the complete frequency range at this rate would require 39 hours.

In order to have constant leakage voltage over a wide frequency range it would be necessary that the capacities of the two tuning condensers of the transmitter remain equal to each other while the frequency changes. In practice, this cannot be achieved with sufficient accuracy since the leakage is very sensitive to any difference between these capacities. This imperfection is not serious, however, over a frequency interval of as much as 1 Mc, and the amplified leakage voltage in this interval remains less than 1 v r.m.s., causing no appreciable increase in the noise figure. The *u*-mode leakage is considerably less sensitive to frequency change and remains negligible if it is canceled at one frequency; hence, if the spectrometer is adjusted for *v*-mode observation, it will remain adjusted for this mode.

The lock-in amplifier has been described by Dicke.[14] We have used total output time constants of 1.2 and 4.7 sec., corresponding to final band widths of 0.28 c.p.s. and 0.069 c.p.s., and calculated reductions by factors of 130 and 270, respectively, of the noise voltage from the receiver output band pass of 5 kc.

C. Use of the Spectrometer

Before a nuclear resonance frequency can be measured with appreciable accuracy, the occurrence of the signal and its approximate frequency must first be established; consequently, the spectrometer is first used to search for the resonance of the nucleus in question.

Liquid samples are preferred because they usually lead to sharp lines, and because the nuclear relaxation times can be controlled by the addition of paramagnetic ions.[10,12] In these experiments the paramagnetic ion Mn^{++} has been used as catalyst and was brought into the solution by dissolving a suitable salt. Equation (1) may be used to show that $v_{\max}$, the value of v for $\Delta\omega = 0$, as a function of H_1 has a maximum $(v_{\max})_{\max} = (M_0/2)(T_2/T_1)^{\frac{1}{2}}$ for $H_1 = 1/\gamma(T_1T_2)^{\frac{1}{2}}$. To obtain signals of enhanced amplitude, it is helpful to shorten T_1 until it is comparable with T_2 which will normally be the case when the line broadening caused by the addition of paramagnetic ions is of the order of that due to field inhomogeneities. Mn^{++}, having a moment of 5.9 Bohr magnetons, provides a thermal relaxation time T_1 of about 3×10^{-5} sec. for protons in a 1.0 molar aqueous solution of $MnSO_4$. For other nuclei, and for other paramagnetic ions, the thermal relaxation time can be estimated[12] from the above value by taking the relaxation time to be inversely proportional to the product (concentration of paramagnetic atoms) (magnetic moment of paramagnetic atom)^2 (viscosity of sample) (gyromagnetic ratio of nucleus in question).^{2} In view of the rather complicated features underlying the mechanism of relaxation, this relationship can claim only qualitative validity, but it is nevertheless useful as a guide for choosing a suitable concentration of paramagnetic salt.

To obtain suitable conditions for signal observation, the magnitudes of the sweep field and the radiofrequency field must be both chosen of the order of the expected line width, the amplified leakage voltage must be adjusted to about 0.5 v r.m.s. and the gain of the lock-in amplifier made large enough to make the expected signal visible on the tape. Guided by spectroscopic values, as we have been for all nuclei reported on here, a range of frequencies likely to contain the resonance is chosen. It is automatically explored as the clock motor provides a continuous frequency variation while a trace is drawn on the chart by the recording milliammeter. We have usually taken search runs of approximately 10-hr. duration, turning the apparatus on in the evening and examining the milliammeter record for a resonance the next morning.

Once the frequency of a nuclear resonance is known approximately, and it is desired to measure this resonance frequency as exactly as possible, the leakage phase is carefully adjusted for observation of the pure *v*-mode, and the secondary frequency standard is set to generate 25-kc markers. Since the frequency is known approximately, it is possible to set the transmitter to a frequency which is just below the first marker below resonance. Hence, after the motor has begun to turn the condensers, a marker is made, the resonance is drawn, and a second marker is made, whereupon the sought-for resonance frequency can be determined by interpolation between the two markers. To calibrate the gyromagnetic ratio against that of a known nucleus, it is necessary to observe similarly the resonance frequency of the latter by turning the condensers to an appropriate position, without changing the magnetic field.

Since the two resonances are not observed at the same time, one has to anticipate the possibility of drifts in the magnetic field between their observation. We therefore have followed the procedure of alternately repeating, at least three times, the pattern of the two resonances in the fashion described. The time at which each resonance occurs is automatically recorded by the recording milliammeter, since the chart moves at a uniform speed. The resonance frequencies of the two nuclei may be plotted as a function of time, and by interpolation their values can be obtained as they would appear if measured at the same time and hence at the same value of the magnetic field. Typical data, from the measurements on Sn^{115} and Na^{23}, are given in Table I.

III. MEASUREMENTS

We shall give here a detailed description of a series of measurements which have been made, using the methods and apparatus described in the preceding section. Short reports of the results have already been published.[16–18]

A. Measurements on Tl^{203} and Tl^{205}

At the time when the measurements to be described were made, the magnetic moment of Tl^{205} was well known from nuclear induction;[19] the best value for Tl^{203}, however, depended on the spectroscopically determined ratio of the magnetic moments of these two isotopes.[20] It seemed worth while to determine the ratio of these two moments with higher accuracy while the measurement of the magnetic moment of Tl^{205} served merely as a check upon the previously published result.

The sample consisted of an aqueous solution of 2.6 molar $\mathrm{Tl(C_2H_3O_2)}$ with 0.03 molar $\mathrm{MnSO_4}$. Eight experiments were made in which the resonances of Tl^{203}, Tl^{205}, and H^1 from the same sample were in turn observed four times. The first four of these experiments were performed at 1500 gauss and the last four at 1900 gauss.[21] The final frequency ratios were determined to be

$$\nu(\mathrm{Tl}^{203})/\nu(\mathrm{H}^1)=0.5714\pm0.0001 \tag{3}$$

$$\nu(\mathrm{Tl}^{205})/\nu(\mathrm{H}^1)=0.5770\pm0.0001 \tag{4}$$

$$\nu(\mathrm{Tl}^{205})/\nu(\mathrm{Tl}^{203})=1.0098\pm0.0002. \tag{5}$$

The value (4) agrees well with Poss' value,[19] as do the ratios (3) and (5) with those subsequently published by Poss.[22] The ratio (5) is in good agreement with the spectroscopic value

$$\mu(\mathrm{Tl}^{205})/\mu(\mathrm{Tl}^{203})=1.00966\pm0.0004,$$

given by Schüler and Korsching.[20]

With the magnetic moment of the proton taken to be[23] $(2.7935\pm0.0003)\mu_N$ and with the spin of both isotopes having a value $\frac{1}{2}$, one obtains directly from the frequency ratios,

$$\mu(\mathrm{Tl}^{203})=(1.5962\pm0.0003)\mu_N, \tag{6}$$

$$\mu(\mathrm{Tl}^{205})=(1.6118\pm0.0003)\mu_N. \tag{7}$$

The magnitude of the diamagnetic correction to be applied is given in column 3 of Table II, where all of the experimental results are summarized. The corrections indicated are to be applied in such a direction as to increase the magnitudes of the magnetic moments listed in the second column. The diamagnetic correction will be discussed in the final paragraph of this section.

The signs of both thallium nuclear magnetic moments were verified to be positive by the procedure outlined in Section II. The resonances of Tl^{203}, Tl^{205}, and H^1 were observed in a small frequency range, sufficient for their respective line widths, by changing the magnetic field accordingly. Since very little change in frequency takes place, there is no possibility that a leakage phase reversal of 180° could have taken place; hence the relative signs of their magnetic moments were determined upon inspection of the signals on the tape.

By the procedure outlined in Section II, a comparison of the maximum of the recorded resonances of H^1 and Tl^{205} led to a spin $\frac{1}{2}$ for Tl^{205}. Since the proton signal would otherwise have been about 60 times greater, it was attenuated in the audio system so as to make it comparable to that of Tl^{205}. The ratio of the signal amplitudes of the two thallium isotopes was in good agreement with the ratio of the abundances,[24] so that the spin of Tl^{203} was also indicated to be $\frac{1}{2}$. These results are in agreement with spectroscopic measurements.[25]

B. Measurements on Sn^{115}, Sn^{117}, and Sn^{119}

Spectroscopically the nuclei Sn^{115}, Sn^{117}, and Sn^{119} have been known[26,27] to have a spin of $\frac{1}{2}$, and their magnetic moments were determined to be $-0.86\mu_N$,[27] $-0.9\mu_N$,[28] and $-0.9\mu_N$,[28] respectively. Since the isotopes Sn^{117} and Sn^{119} have, respectively, natural abundances of 7.51 percent and 8.45 percent,[29] a short calculation reveals that a signal-to-noise ratio of about 10 should be available from a 5 molar solution of a tin salt in water and that their respective abundances should be sufficiently different to distinguish between the two resonances. Accordingly, an aqueous 5.3 molar solution of $\mathrm{SnCl_2}$ with 1.0 molar $\mathrm{MnCl_2}$ was used, and their resonances were found about 300 kc apart in the neighborhood of 7.0 Mc, with a magnetic field of 4500 gauss.

The resonances of Sn^{117}, Sn^{119}, and Na^{23} were then observed in turn until about three of each had been

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[16] W. G. Proctor, Phys. Rev. **75**, 522 (1949).

[17] W. G. Proctor, Phys. Rev. **76**, 684 (1949).

[18] W. G. Proctor and F. C. Yu, Phys. Rev. **76**, 1728 (1949).

[19] H. L. Poss, Phys. Rev. **72**, 637 (1947).

[20] H. Schüler and H. Korsching, Zeits. f. Physik **105**, 168 (1937).

[21] The magnet used for the last four experiments was a battery-operated magnet with pole faces 10 in. in diameter, spaced $1\frac{1}{16}$ in. apart. All other measurements reported in this paper have been carried out with the magnet described in Section II.

[22] H. L. Poss, Phys. Rev. **75**, 600 (1949).

[23] H. Taub and P. Kusch, Phys. Rev. **75**, 1481 (1949).

[24] A. O. Nier, Phys. Rev. **54**, 275 (1938).

[25] H. Schüler and J. E. Keyston, Zeits. f. Physik **70**, 1 (1931); H. Schüler and T. Schmidt, Zeits. f. Physik **104**, 468 (1937).

[26] H. Schüler and H. Westmeyer, Naturwiss. **21**, 660 (1933).

[27] M. Gurevitch, Phys. Rev. **75**, 767 (1949).

[28] S. Tolansky, Proc. Roy. Soc. **144**, 574 (1934).

[29] Hintenberger, Mattauch, Seelmann-Eggebert, Zeits. f. Naturforschung **3a**, 413 (1948).

recorded; these observations were repeated once again. The resonance of sodium was obtained by exchanging the sample above with one composed of an aqueous solution of 0.69 molar NaCl with 1.0 molar $MnCl_2$. By the graphical method of interpolation mentioned in Section II to correct for field drifts, we find as the result of both experiments the following frequency ratios:

$$\nu(Sn^{117})/\nu(Na^{23}) = 1.3468 \pm 0.0001, \tag{8}$$

$$\nu(Sn^{119})/\nu(Na^{23}) = 1.4090 \pm 0.0001, \tag{9}$$

$$\nu(Sn^{119})/\nu(Sn^{117}) = 1.0465 \pm 0.0001. \tag{10}$$

Since the abundance of the isotope Sn^{115} is only 0.33 percent,[29] its resonance was not observed with the first sample. It was found, however, about 700 kc below that of Sn^{117} at the same magnetic field, using a sample prepared by dissolving $SnCl_2 \cdot 2H_2O$ in 2.0 molar $MnSO_4$ at the temperature of boiling water. This supersaturated solution of tin would not crystallize at room temperature for several hours; with the search extending over many hours, it was necessary to heat it at intervals. The concentration of tin in this sample proved to be about 2.5 times that of the first sample, and $MnSO_4$ was present to a molar concentration of 0.7. The frequency of resonance of Sn^{115} was compared to that of Na^{23} using a sample containing 0.2 molar NaCl with 0.2 molar $MnSO_4$. The signal-to-noise ratio of the recorded resonance was about two; the resonances of Sn^{115} and Na^{23} were recorded five times each, with the results shown in Table I. From these data we compute that

$$\nu(Sn^{115})/\nu(Na^{23}) = 1.2362 \pm 0.0001. \tag{11}$$

By dividing (8) by (11), one obtains

$$\nu(Sn^{117})/\nu(Sn^{115}) = 1.0894 \pm 0.0001. \tag{12}$$

With the magnetic moment of Na^{23} taken to be[30] $(2.2166 \pm 0.0004)\mu_N$, one obtains

$$\mu(Sn^{115}) = (-0.9134 \pm 0.0002)\mu_N, \tag{13}$$

$$\mu(Sn^{117}) = (-0.9951 \pm 0.0002)\mu_N, \tag{14}$$

$$\mu(Sn^{119}) = (-1.0411 \pm 0.0002)\mu_N. \tag{15}$$

These values differ from those reported earlier[31] because the diamagnetic correction has not been included in making these calculations and a slightly different value of the magnetic moment of Na^{23} has been taken. The ratios of these magnetic moments are given accurately by the expressions (10) and (12).

The isotopes Sn^{117} and Sn^{119}, with a known abundance ratio[29] 0.89, were identified by the ratio of the amplitudes of their signals. This same ratio, derived from the amplitude ratio of the two signals, was found to be 0.95 ± 0.06, while the opposite assignment would have given 1.05 ± 0.07, showing beyond reasonable doubt that the former assignment is the correct one.

Inspection of the recorded signals showed that the tin nuclei had the same sign of magnetic moment. A comparison of signals for the sign between Sn^{119} and Na^{23} showed these nuclei to have magnetic moments of the opposite sign, thus verifying that all three tin isotopes have negative magnetic moments.[32]

The spin of the tin nuclei were verified to be $\frac{1}{2}$ from a comparison of their signal amplitudes with each other and with that from Na^{23}.

C. Measurements on Pb^{207}

Of all the naturally occurring isotopes of lead, only one, the isotope 207, with an abundance of 21.1 percent, has an odd number of neutrons. Since hyperfine structure measurements have indicated a spin[33] of $\frac{1}{2}$ and a magnetic moment of 0.6 nuclear magnetons,[34] one has sufficient data to calculate that a reasonably concentrated solution of some lead salt would provide a detectable signal. Accordingly, a sample containing an aqueous solution of 1.0 molar $Pb(C_2H_3O_2)_2$ with 0.8 molar $Mn(C_2H_3O_2)_2$ was prepared, and a signal was observed in the neighborhood of 5.8 Mc with a magnetic field of 6600 gauss.

The lead resonance was compared in frequency with that from Na^{23}, which was obtained by exchanging the the sample above with one composed of an aqueous solution of 0.69 molar NaCl and 1.0 molar $MnCl_2$. After four alternate measurements, the following frequency ratio was found:

$$\nu(Pb^{207})/\nu(Na^{23}) = 0.7901 \pm 0.0001, \tag{16}$$

which leads to

$$\mu(Pb^{207}) = (0.5837 \pm 0.0001)\mu_N. \tag{17}$$

The positive sign was verified by comparing the signal trace from the lead resonance to that obtained from the sodium resonance, when they both were made at the same frequency and no possibility of a change of leakage phase could have occurred.

Several measurements were made to determine the relative signal amplitudes of the sodium and lead resonances with the result that the spin of Pb^{207} was verified to be $\frac{1}{2}$.

D. Measurements on Cd^{111} and Cd^{113}

Various spectroscopic measurements on the hyperfine structure of cadmium were carried out before the publication of the review article by Bethe and Bacher,[34] who conclude that the isotopes Cd^{111} and Cd^{113} each have a spin $\frac{1}{2}$ and the same magnetic moment of -0.65

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[30] The magnitude was computed using $\nu(Na^{23})/\nu(H^1) = 0.26450 \pm 0.0003$ given by F. Bitter, Phys. Rev. **75**, 1326 (1949) and the value of the magnetic moment of the proton given by Taub and Kusch, reference 23.

[31] See references 17 and 18. Furthermore, it will be noted that the uncertainty in the frequency ratios given in (8)–(10), and (16) is smaller than given in the early report, reference 17. The improvement arises from correcting an error made in the previous calculations.

[32] The positive sign of Na^{23} has been taken from J. B. M. Kellogg and S. Millman, Rev. Mod. Phys. **18**, 323 (1946).

[33] H. Kopfermann, Zeits. f. Physik **75**, 363 (1932).

[34] H. A. Bethe and R. F. Bacher, Rev. Mod. Phys. **8**, 82 (1936).

nuclear magnetons. Cadmium has eight naturally occurring isotopes but only Cd^{111} and Cd^{113} have an odd mass number, and these occur with an abundance of 13.0 and 12.3 percent, respectively.[35] The resonances were found at a field of 6600 gauss, with radiofrequencies in the neighborhood of 6.0 Mc, using a saturated sample of $CdCl_2$ in 0.3 molar $MnSO_4$. The frequency ratios were found to be

$$\nu(\mathrm{Cd}^{111})/\nu(\mathrm{Na}^{23})=0.8016\pm0.0001, \quad (18)$$

$$\nu(\mathrm{Cd}^{113})/\nu(\mathrm{Na}^{23})=0.8386\pm0.0001, \quad (19)$$

$$\nu(\mathrm{Cd}^{113})/\nu(\mathrm{Cd}^{111})=1.0461\pm0.0001. \quad (20)$$

Because of the equality of the spins, the last ratio again gives the ratio of the magnetic moments of the cadmium isotopes. The Na^{23} was obtained from an aqueous solution of 0.1 molar NaCl with 0.2 molar $MnSO_4$. These frequency ratios lead to the values

$$\mu(\mathrm{Cd}^{111})=-(0.5923\pm0.0001)\mu_N, \quad (21)$$

$$\mu(\mathrm{Cd}^{113})=-(0.6196\pm0.0001)\mu_N. \quad (22)$$

The signs were determined by comparing, under identical radiofrequency conditions, the cadmium resonances and that of I^{127}, the magnetic moment of which is positive.[36] The cadmium isotopes were identified by their different signal magnitudes, using samples[37] enriched to about 80 percent in Cd^{111} and Cd^{113}. Through comparison of the recorded signal magnitudes from cadmium and sodium, a spin of $\frac{1}{2}$ for both cadmium isotopes has been verified.

The sequence of isotopes, Cd^{111}, Cd^{113}, Sn^{115}, Sn^{117}, Sn^{119} was of interest to us since each of these nuclei differs from the preceding one by the addition of two nucleons. Without change in spin and sign, the magnitude of the moment shows a steady increase, in the ratio 1.000:1.046:1.542:1.680:1.758. While the significance of this fact is not yet capable of direct interpretation in terms of nuclear structures, it is likely to be connected with the circumstance that a "proton shell"[38] comes to a conclusion with tin. It also seems plausible that the relatively much larger increase, from Cd^{113} to Sn^{115}, caused by the addition of two protons is due to their charge, while the addition of two neutrons in the other cases, as in Tl^{203} and Tl^{205}, causes only an increase of a few percent. In this connection it may be worth noticing that the ratio of the magnetic moments of Cd^{113} to Cd^{111} is very nearly equal to that of Sn^{119} to Sn^{117}.

E. Discussion of Accuracy

The present spectrometer measures resonance frequencies with accuracies of about one part in 10^4. This corresponds to about 500 c.p.s. for the radio frequencies used, or a distance on the recording tape of 1 mm, since a rate is used such that a frequency interval of 25 kc is equivalent to 60 mm. This limitation is dictated by the signal-to-noise ratios available in the present experiments, uncertainty of leakage phase, and by small, random magnetic field changes. There is little reason to suspect that the magnetic moment ratios of isotopes determined above have any other limitations in accuracy than those just mentioned. Systematic sources of error, such as non-uniform spacing of frequency markers and lag of information recorded on the tape, have been examined and found to be negligible. Since the head only accommodates one sample, samples containing different nuclei have always been accurately located in the same position in the magnetic field.

The situation is different from that met with in the comparison of isotopes when nuclei of different elements are compared, and particularly when one attempts to give the final results in terms of the nuclear magneton μ_N. As far as the latter is concerned, the accuracy depends ultimately on that with which the proton moment is known, and which, at present,[23] is given to be $(2.7935\pm0.0003)\mu_N$. Another source of error arises from the diamagnetic correction, which takes account of the shielding due to extranuclear electrons and has to be applied when comparing moments of different elements. The diamagnetic corrections listed in column 3 of Table II have been linearly interpolated from values for specific atoms given by Lamb[39] using Hartree fields. Calculations[40] were made on six of the atoms reported by Lamb, using hydrogen-like wave functions and empirical shielding constants proposed by Slater.[41] In each case, the Hartree field value and the calculated value agreed to within two percent. This agreement suggests that a smaller error in the magnitude of the diamagnetic correction will be made by using interpolated Hartree values instead of Lamb's formula.

The uncertainties and corrections given in Table II have been arrived at from those mentioned, and it would have seemed that their enumeration was essentially complete. We have recently observed,[42] however, that the resonance lines of N^{14} show a marked dependence on the chemical compound containing nitrogen. For example, in an aqueous solution of NH_4NO_3 there appear two resonances, evidently from the NH_4^+ and NO_3^- ions, separated in frequency by as much as 0.03 percent. Similar, although not quite so pronounced, effects have been found for F^{19} by W. C. Dickinson,[43] and it is possible that they occur for many other nuclei, including those the moments of which have been under

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[35] A. O. Nier, Phys. Rev. **50**, 1041 (1936).

[36] T. Schmidt, Zeits. f. Physik **112**, 199 (1939).

[37] These samples were kindly lent to us by Dr. B. J. Moyer of the Radiation Laboratory of the University of California.

[38] E. Feenberg and K. C. Hammack, Phys. Rev. **75**, 1877 (1949); L. W. Nordheim, Phys. Rev. **75**, 1894 (1949); M. Mayer, Phys. Rev. **75**, 1969 (1949).

[39] W. E. Lamb, Jr., Phys. Rev. **60**, 817 (1941).

[40] These calculations were made by Dr. F. C. Yu, Department of Physics, Stanford University.

[41] J. C. Slater, Phys. Rev. **36**, 57 (1930).

[42] W. G. Proctor and F. C. Yu, Phys. Rev. **77**, 717 (1950).

[43] We are grateful to Professor F. Bitter for sending us a preliminary report of these findings.

investigations in this paper. While these effects are evidently due to extranuclear electrons and possibly of an origin related to that of the hitherto considered diamagnetic effects, they are not sufficiently understood at the present moment to evaluate properly the corresponding corrections in the moment values. Further investigations, now in progress, are necessary before this is possible, and we have to limit ourselves at this stage to the concluding remark that the present estimate of accuracies is under serious doubt.

The author is deeply grateful to Professor Felix Bloch for generous and helpful advice throughout the course of this work. He would also like to express his gratitude to Dr. F. C. Yu who worked with him during the period of the measurements upon the isotopes Cd^{111}, Cd^{113}, and Sn^{115}.

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Negative Energy Neutron Resonance in Cl^{35}

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The neutron scattering cross section of chlorine has been measured at various energies in the range from 0 to 3000 ev. These data have been fitted to a one-level Breit-Wigner scattering formula. A single real level (i.e., one of negative energy with respect to binding) in Cl^{36} at −75 ev is sufficient to account for the observed slow neutron properties of chlorine.

I. INTRODUCTION

No positive energy neutron resonance below 3000 ev is known for chlorine. However, there is abundant evidence for the existence of a negative energy neutron resonance (i.e., a real level) in Cl^{35}. A level just below neutron binding in Cl^{36} is consistent with the following facts: (a) the scattering cross section of chlorine markedly decreases with increasing neutron energy up to at least 3000 ev, (b) the thermal and coherent scattering cross sections σ_s and σ_{coh} of chlorine are unequal[1] and the thermal scattering phase of chlorine is positive, (c) the thermal scattering cross section, σ_s, of chlorine is much larger than $4\pi R^2$, where R is the nuclear radius, (d) the thermal capture cross section, σ_{tha}, of Cl^{35} (75 percent abundant[2] −42.0b) is large, (e) the deep potential well of the odd Z-odd N, Cl^{36} nucleus ($\sim$9.8 Mev) implies an unusually high level density near neutron binding,[3] and (f) Cl^{37} (25 percent abundant) is a closed shell nucleus;[4] therefore, it is expected to have a large level separation.[5] This in turn makes the existence of a neutron level near binding in Cl^{37} improbable.

II. THEORY

The Breit-Wigner[6] one-level scattering cross section, $\sigma_s(E)$, as a function of energy is given by:

$$\sigma_s(E)=\sigma_p+\pi pg\bar{\lambda}_0\Gamma_n\frac{\bar{\lambda}_0\Gamma_n+4R(E-E_0)}{(E-E_0)^2+\Gamma^2/4}.$$

where σ_p is the potential scattering cross section; p is the fractional abundance of the isotope having the resonance, R is the nuclear radius, and $g=\frac{1}{2}[1\pm 1/(2i+1)]$, where i = spin of initial nucleus. The other symbols have their usual meanings. For a negative energy level in which Γ is small compared to the resonance energy, the scattering cross section can be written as

$$\sigma_s(E)=\sigma_p+\pi pg\bar{\lambda}_0\Gamma_n[\bar{\lambda}_0\Gamma_n+4R(E+E_0)]/(E+E_0)^2.$$

E_0 in this expression is positive, and

$$\bar{\lambda}_0=[(2.60\times 10^6)/4\pi E_0]^{\frac{1}{2}}.$$

This work will be concerned with fitting measured values of the scattering cross section, $\sigma_s(E)$, at different neutron energies to the Breit-Wigner expression in order to obtain the parameters of the resonance.

First, the quantity $\bar{\lambda}_0\Gamma_n$ can be evaluated in terms of E_0. This relation is given in terms of the thermal $[\sigma_s(0)=14.2\text{ b}]$ and the coherent $[\sigma_{coh}(0)=11.5\text{ b}]$ scattering cross sections.[1]

$$(\sigma_s-\sigma_{coh})/4\pi=pg(1-pg)(\bar{\lambda}_0\Gamma_n/2E_0)^2.$$

For Cl^{35}, $p=3/4$ and $g=5/8$ or $3/8$ (spin of $Cl^{35}=3/2$).

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[1] E. O. Wollan and C. G. Shull, Phys. Rev. **73**, 830 (1948).

[2] Harris, Muehlhause, Rasmussen, Schroeder, and Thomas, Phys. Rev. (to be published).

[3] The depth of the well can be calculated from mass values. Evidence of a deep well is indicated by the facts that the n,p reaction on Cl^{35} is exoergic, and that the average number γ-rays per neutron capture from Cl^{35} is high. C. O. Muehlhause, Phys. Rev. (to be published).

[4] M. G. Mayer, Phys. Rev. **75**, 1969 (1949).

[5] Harris, Muehlhause, and Thomas, Phys. Rev. **79**, 11 (1950).

[6] H. A. Bethe, Rev. Mod. Phys. **9**, 69 (1937).