90 YEARS

MAGNETIC MOMENT OF A PROTON

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The magnetic susceptibility of solid hydrogen has been measured at temperatures of 4.22, 2.18, and 1.76 K. It was found that the hydrogen diamagnetic susceptibility diminishes in this temperature interval. This diminution is explained by an increase of the paramagnetism induced by the proton magnetic moment. Using the temperature dependence of the susceptibility, the proton magnetic moment μ_p is calculated. We obtained the values $\mu_p = 2.3 \pm 10\%$ in the nuclear magneton units and $\mu_p = 2.7$ with an error less than 10%.

The possibility to measure the nuclear magnetism by means of the susceptibility shows that the equilibrium between the field and a hydrogen nucleus is established very quickly.

1. Introduction

For the first time, the immediate measurement of the magnetic moment of a proton was conducted with the use of the molecular beam deflection in an inhomogeneous electric field. The measurement was done by Stern, Frisch, and Estermann [1] by using a molecular hydrogen and by Rabi, Kellog, and Zacharias [2] by using an atomic hydrogen.

Those authors obtained the following values for the proton magnetic moment μ_p:

Stern, Frisch, Estermann $\mu_p = 2.5$ ($\pm 10\%$).

Rabi, Kellog, Zacharias $\mu_p = 3.25$ ($\pm 10\%$), in the first paper,

Rabi, Kellog, Zacharias $\mu_p = 2.85 \pm 0.15$ in the second paper with the use of improved method.

The values of μ_p are given in nuclear magnetons.

In 1934, Debye [3] paid attention to another possibility to directly determine the nuclear magnetism. Let us suppose that the paramagnetism emerging due to nuclear moments obeys the Curie law. Then, at temperatures about $0.001 – 0.01$ K, it will have such high value that it can be measured. Naturally, this can occur only in the substances which possess no paramagnetism

of the electron shell, since this magnetism is considerably greater than that of a nucleus.

Later on, Dorfmann [4] pointed out that the presence of the nuclear paramagnetism in the substances with small diamagnetism would be probably determined already at temperatures of about 1 K.

Indeed, let us represent the diamagnetic susceptibility χ in the form $\chi = \chi_d + \chi_p$, where χ_d and χ_p are related, respectively, to the ordinary temperature-independent diamagnetism and the nuclear paramagnetism which increases with decrease in the temperature by the Curie law. Then we should have the possibility to determine the diminution of diamagnetism at sufficiently low temperatures.

2. Measurement Methods

Here, we used the possibility to measure the nuclear magnetism indicated by the above-mentioned authors.

The approximate calculation shows that hydrogen is best suited for measurements due to its small diamagnetic susceptibility $\chi_d = -3.8 \times 10^{-6}$ (per mole). If we set the proton moment χ_p to be about 2–3 nuclear magnetons, then we will get the relation $\chi_p = N\mu^2/3kT \cong 10^{-6}/T$ for hydrogen (per gram-molecule). Hence, χ_p makes up about 20% already at 2 K, and such a change of the susceptibility is already accessible for measurements.

It is worth to mention that other substances, except for the very hydrogen, are unsuitable for these measurements in the range of liquid helium temperatures since relative changes of their susceptibilities are small due to their large magnitudes.

For example, for methane (CH_4) which is the most suitable substance after hydrogen, $\chi_{pCH_4} \cong 10^{-6}/T$ for the nuclear paramagnetism, i.e., we have approximately the same value as that for H_2. But the susceptibility of CH_4, $\chi = -18 \times 10^{-6}$, is much greater than that of H_2,

and χ_p makes up only about 3% of χ at 2 K[1]. For benzene (C_6H_6) at 2 K, $\chi_p/\chi_d \cong 0.01$.

In addition to the insignificance of the effect, there exists another basic reason, by which the compounds are not suitable for measurements. We will discuss it in what follows.

In the present paper, we measure a diminution of the solid hydrogen diamagnetism in the temperature range from 4.22 K and lower. A short communication on the results of investigations was published earlier [5].

The susceptibility was measured using the Guye method by the force, at which the ampoule with hydrogen was pushed out of the magnetic field. The force was measured with the help of a balance.

In Fig. 1, we show the scheme of the measuring device. On the right end of the balance's beam, quartz ampoule A filled up with hydrogen is hung up. The upper broad part of the ampoule with a volume of about 60 cm^3 serves as a container for hydrogen, and the lower part has form of a capillary with very thin walls (the thickness of walls is about 0.06–0.07 mm), a cross-section of about 3 mm^2, and a length of about 60 cm^2. At room temperature, the pressure of hydrogen in the ampoule is about 3.5 atm.

When cooling down the lower part of the ampoule, hydrogen is condensed and forms a liquid column, as shown in Fig. 2, where K is the capillary, and M, M are the electromagnet poles.

The ampoule is surrounded by glass pipe T which is fixed on the iron base of balance P.

The force arising when a magnetic field is switched on is compensated by a current which causes the pulling of a small constant magnet into coil S. This compensative current I is a measure of the susceptibility.

The preceding calibration of the balance showed a rigorous linear relation between the load and the compensating current.

The balance is covered by a glass bell jar. Having thoroughly been pumped out and blown through by helium, the space under the glass bell jar is filled with helium at a pressure of 3–4 mmHg. Helium for the blowing through and filling-up of the glass bell jar is specially thoroughly purified by coal at a high pressure at the liquid hydrogen temperature.

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[1]In the Dorfmann's paper quoted above, the erroneous calculation is performed for the CH_4 and LiH, which leads to the overstated effect.

[2]Capillaries were produced by the master-glassblower, E.V. Petushkov.
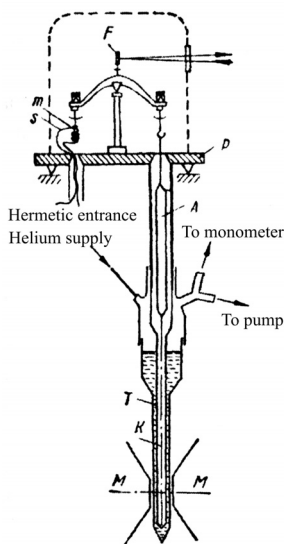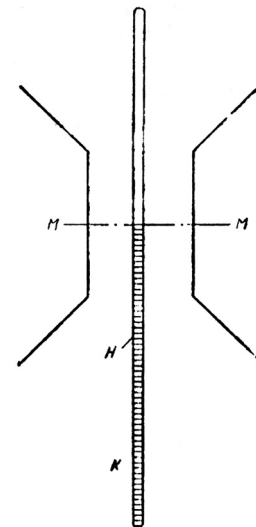

Fig. 1   Fig. 2

The magnetic field is produced by an electromagnet. The measurements were conducted at a field strength of about 20,000 Gs.

The special attention was paid to the purity degree and the composition of hydrogen in the ampoule. The initial hydrogen was very pure. For the repeated purification, hydrogen was liquefied in a vessel cooled down by liquid hydrogen. The ampoule was filled up with hydrogen vaporizing from this vessel. In this way, hydrogen became free from the most dangerous admixture, oxygen. Probably, at such a purification, there could be not more than 10^{-4} mmHg of oxygen in the ampoule, which corresponds to the O_2-vapor pressure at this temperature. This makes up about 10^{-7} of the amount of hydrogen in the ampoule and can cause no considerable error.

The hydrogen composition is controlled by the Schleiermacher method according to work [6]. Hydrogen in the ampoule contained about 75% ortho- and 25% parahydrogen, and the error in the determination of the concentration did not exceed 0.1%.

The measurements were conducted at three temperatures: 4.22, 2.18, and 1.76 K. These temperatures were produced with the help of liquid helium and were determined by a pressure of the vapor over the liquid. These three temperatures correspond to pressures of 760, 37.5, and 12 mmHg which were measured by a mercury manometer. The measurements were conducted by using the empty ampoule or one filled with hydrogen.

Regulating the helium influx speed in a cryostat, hydrogen could be concentrated and freeze in any part

of the capillary. When measuring the "empty" ampoule, hydrogen was condensed very high above the magnetic poles where the field was very weak, so that the vessel can be undoubtedly considered empty. After the "empty" vessel have been measured, helium was vaporized. In this case, hydrogen melted and trickled into the bottom part of the vessel (Fig. 2), where it froze owing to a new helium pouring.

3.   Measurements

At each temperature. the following operations have been done: the zero position of the balance was found, and the magnetic field was switched on; the equilibrium position of the balance was restored by switching on the current in the compensating coil; the magnetic filed was switched off, and the zero position was verified; this procedure was repeated many times.

Table 1 gives the values of I in terms of the device scale readings for three temperatures for the ampoule with hydrogen.

The measurements with empty and filled-up with hydrogen ampoules were conducted during one experiment in the following order: at 4.22, 2.18, 1.76, and again at 4.22 K. The data for 4.22 K are in agreement with each other. The first four values in Table 1 for 4.22 K were taken from the first measurement run at this temperature, the next four from the second one.

The average values of I for the measurements with the empty ampoule, ampoule with hydrogen, and hydrogen itself are shown in Table 2.

It is necessary to mention that the measurement accuracy on the transition to temperatures lower than the transformation point of helium I into helium II (2.2 K) is considerably increased.

This transition has a sharp effect on the behavior of the balance: in the temperature range of helium I, some "walk" of readings was revealed. When the temperature falls lower than 2.2 K, this "walk" stops, and the measurement is conducted as quietly as that at room temperature. Probably, this phenomenon is caused by a sudden huge increase of the heat conductivity of helium when the transition between states I and II takes place [7].

We do not obtain the absolute values of the hydrogen magnetic susceptibility, because the following should be guaranteed for this purpose: first, the known and always identical cross-section of the cylinder with solid hydrogen, and, second, its identical properties over the whole length. These requirements would additionally

complicate the already quite involved conditions of measurements.

For our purposes it is sufficient to know the temperature dependence of the susceptibility of hydrogen determined by values of the compensating current.

The obtained values of the compensating current allow us to calculate the Curie constant of the nuclear paramagnetism and then the proton magnetic moment.

The calculation can be carried out using the data of either all three measurement groups including the measurements performed with small accuracy at 4.22 K, or only two measurement groups, at 2.18 and 1.76 K performed with a greater accuracy.

The starting equation for the calculation has the form

$$\chi = \chi_d + \chi_p = \chi_d + \frac{C}{T}.$$

Here, we assume that χ_d remains constant in the whole temperature interval, whereas the paramagnetic term obeys the Curie law.

In the experiment, we determined the compensating currents proportional to the susceptibility, for which an analogous relation

$$I = I_d + I_p = I_d + \frac{aC}{T}$$

T a b l e 1. Compensating current I (ampoule with the hydrogen)

4.22 K	2.18 K	1.76 K
107.5	104.3	100.3
113.3	102.9	101.2
103.5	103.2	101.3
112.5	103.4	100.0
111.5	104.2	100.3
115.4	104.2	100.7
101.4	103.1	100.0
109.9	105.0	100.3
	104.6	101.4
		100.3
		(102.1)
		(101.9)
		100.3
		99.1
109.4 ± 3.8	103.8 ± 0.6	100.4 ± 0.4

T a b l e 2

T, K	I		H$_2$
	Empty ampoule	Ampoule with hydrogen	
4.22	−21.5	109.4	130.9 ± 7.5
2.18	−19.5	103.8	123.3 ± 1.2
1.76	−15.6	100.4	116.0 ± 0.8

is valid. If we use three measurement groups, then we cannot apply the statistical method to them due to the systematic error at 4.22 K. Nevertheless, the use of the least squares method will be most correct. In this case, using the obtained values of I, we can calculate the average value

$$x = \frac{C}{\chi_d} = \frac{n \sum \frac{1}{T} - \sum I \cdot \sum \frac{1}{T}}{\sum I \cdot \sum \frac{1}{T^2} - \sum \frac{I}{T} \sum \frac{1}{T}}$$

and its limiting relative error $\Delta x / x$ by determining

$$\left(\frac{\Delta x}{x} \right)^2 = \frac{\delta^2 \left(n - 1 + \frac{\sum \bar{I}^2}{I_d^2} \right)}{C_I \left[n \sum \frac{1}{T^2} - \left(\sum \frac{1}{T} \right)^2 \right]}.$$

Here, n is the number of measurements, and δ, C_I, $\bar{I}$, and I_d are determined by the following expressions:

$$\delta^2 = \frac{\sum \left(I - \bar{I} \right)^2}{n - 2}, \quad \bar{I} = I_d + \frac{C_I}{T},$$

$$I_d = \frac{\sum \frac{1}{T^2} \cdot \sum I - \sum \frac{1}{T} \cdot \sum \frac{I}{T}}{n \sum \frac{1}{T^2} - \left(\sum \frac{1}{T} \right)^2},$$

$$C_I = \frac{n \sum \frac{I}{T} - \sum I \cdot \sum \frac{1}{T}}{n \sum \frac{1}{T^2} - \left(\sum \frac{1}{T} \right)^2}.$$

If we insert the values of temperature T and the corresponding compensating currents I into these expressions, we obtain

$$x = \frac{C}{\chi_d} = 0.306; \quad C = 0.306 \, \chi_d,$$

$$\left(\frac{\Delta x}{x} \right)^2 = 0.048; \quad \frac{\Delta x}{x} = \frac{\Delta C}{C} \cong 0.2.$$

To carry out the transition from the Curie constant C to the proton magnetic moment μ_p, let us dwell upon the nuclear paramagnetism in more details. If a molecule has nuclear magnetic moment μ, then, according to the Curie law, its paramagnetism is characterized by the quantity

$$\chi_p = \frac{N\mu^2}{3kT}.$$

One can simply express μ in terms of μ_p. If a proton moment is μ_p then

$$\mu = (2\mu_p) \, j; \quad \mu^2 = (2\mu_p)^2 j \, (j + 1).$$

For a proton, $j = 1/2$. Hence, for two protons in orthohydrogen, $j = 1$ and

$$\mu^2 = 2(2\mu_p)^2 = 8\mu_p^2.$$

When hydrogen consists of orthohydrogen and of parahydrogen in the ratio 3:1, we have

$$\chi_p = \frac{3}{4} \frac{N}{3} \frac{8\mu_p^2}{kT} = 2 \frac{N\mu_p^2}{kT}.$$

To this paramagnetism, it is necessary to add a rotational paramagnetism of nuclei which, as we consider, follows the Curie–Weiss law, because between the rotational magnetic moments interact with one another. This additional paramagnetism is characterized by the formula $\chi_p' = N\mu_R^2 / 3k \, (T + Q)$ (where μ_R is the rotational magnetic moment), so we get

$$\chi_p = \frac{2N}{k} \frac{\mu_p^2}{T} + \frac{N\mu_R^2}{3k \, (T + Q)}.$$

If we take into account that $\mu_R \cong 0.85$ of the nuclear magneton [1] (whereas $\mu_p \cong 2.5$ of the nuclear magneton), it is easy to see that the addition caused by the rotational moment accounts for only about 2 % of χ_p and can be neglected in the first approximation, which corresponds to our measurement.

Thus, we have

$$C = 2N\mu_p^2 / k; \quad \mu_p^2 = Ck / 2N.$$

For the gram-molecule, $\chi_d = -3.8 \times 10^{-6}$ and $C = 0.306 \times 3.8 \times 10^{-6} = 1.16 \times 10^{-6}$; hence, we get

$$\mu_p^2 = \frac{1.16 \times 10^{-6} \times 1.37 \times 10^{-16}}{2 \times 6.06 \times 10^{23}} = 1.32 \times 10^{-46},$$

$$\mu_p = 1.15 \times 10^{-23} \, \text{CGSM} = \frac{1.15 \times 10^{-23}}{5 \times 10^{-24}} = 2.3 \text{ of the}$$
nuclear magneton.

The error of the determination of C is $\Delta C / C \cong 0.2$. Hence,

$$\Delta \mu / \mu = \frac{1}{2} \Delta C / C \cong 0.1, \text{ approximately 10\%, and}$$

$\mu_p = 2.3 \pm (0.2 - 0.3)$ of the nuclear magneton.

This result was obtained by the averaging of C for all three temperatures. One can see from Table 1 that the measurement accuracy at 2.18 and 1.76 K is considerably higher than that at 4.22 K. If we calculate C by using the values of I for both these groups, the error of μ_p will be less than that in the previous calculation. From the ratio I_{T_1}/I_{T_2}, one can easily obtain

$$C = \chi_d \frac{1 - \dfrac{I_{T_1}}{I_{T_2}}}{\dfrac{I_{T_1}}{I_{T_2}} \left(\dfrac{1}{T_2} - \dfrac{1}{T_1} \right)}.$$

With the values of I at 1.76 and 2.18 K, we obtain $C = 1.63 \times 10^{-6}$ and $\mu_p = 2.7$ of the nuclear magneton.

4. Measurements Results

Thus, the following values for μ_p were obtained: $\mu_p = 2.3 \pm (0.2 - 0.3)$ from three measurement groups and a more accurate value of 2.7 from the measurements at 1.76 and 2.18 K with considerably less error.

The results obtained are in satisfactory agreement with the data of other authors [1, 2], particularly taking the difference in methods into account.

By the time of the completion of this experiment, the paper by Heitler and Teller [8] was appeared, where it is shown that the transition time of nuclear moments to an equilibrium state in the magnetic field for the diamagnetic substances (nonmetals) is very large; at 0.1 K, this time is about 10^6 years.

However, our experiments indicate that the duration of the transition to an equilibrium state is at most a second. The further calculations made by Fröhlich and Heitler [9] showed that the transition time to the equilibrium state for rotational diamagnetic molecules is about 0.1 s at our temperatures.

This conclusion is exceptionally important. It requires that the substances, whose nuclear magnetism should be measured, satisfy one of the two conditions: they should be either metals or they should have rotational moments up to the lowest temperatures.

The second requirement is hardly satisfied by any other substance except for hydrogen. It is evident that it is impossible to conduct the measurements with heavy hydrogen as with ordinary one, since the orthostate of heavy hydrogen has no rotational moment at low temperatures.

Thus, the determination of the proton magnetic moment from the magnetic susceptibility gave the correct value. Moreover, it was shown that the transition time to the equilibrium state of nuclear moments for the hydrogen in the magnetic field is very short.

In conclusion, we acknowledge L. Landau for his participation in the discussion of the experimental results.

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LAZAREV BORIS GEORGIEVICH
(06.08.1906–20.03.2001)

Boris Georgievich Lazarev was the outstanding physicist-experimentalist, Full Member of the National Academy of Sciences (NAS) of Ukraine (1951), winner of the State prize of the USSR (1951) and UkrSSR (1982). He was born in the Myropol'e village (now the Sumy region) in Ukraine. From 1928 till 1932, B.G. Lazarev had worked at the Leningrad Physical-Technical Institute, then he had worked at the Ukrainian Physical-Technical Institute (now National Science Center "Kharkov Institute of Physics and Technology" of NAS of Ukraine) from 1937 till 2001. He was Head of a laboratory at UPhTI (after L.V. Shubnikov, 1938) and Head of a department of KIPT (till 1988). B.G. Lazarev and his colleagues carried out a series of works concerned with the electronic properties of metals, liquid helium properties, cryogenic technique for the creation of high vacuum. He had proposed and realized the method of helium isotope separation (1950) [1], discovered the (2+1/2)-phase transition (1963), had measured the magnetic moment of a proton in solid hydrogen (together with L.V. Shubnikov, 1936), quantum oscillations of the magnetic susceptibility for a number of metals at low temperatures [2]. B.G. Lazarev is a founder of the scientific school in low-temperature physics.

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SHUBNIKOV LEV VASIL'EVICH
(29.09.1901–10.11.1937)

Lev Vasil'evich Shubnikov was the outstanding physicist-experimentalist, the co-author of some Nobel-prize-level

publications, had worked at Leiden University during 1926–1930; from 1930 till 1937, he worked at the Ukrainian Physical-Technical Institute (UPhTI), Kharkov (now NSC "Kharkov Institute of Physics and Technology" of the NAS of Ukraine). L.V. Shubnikov was Professor of the Kharkov State University (1934–1937). L.V. Shubnikov was a founder and the first head of the cryogenic laboratory at UPhTI.

In 1930, L.V. Shubnikov and W. De Haas have discovered the effect of electric resistance oscillations of bismuth at low temperatures (the Shubnikov–de Haas effect). During his work at UFTI, L.V. Shubnikov together with his colleagues had performed a number of high-priority works on superconductivity (1934–1938) and antiferromagnetism (1936–1938) that gained a worldwide recognition. Together with I.V. Obreimov, he had developed a new method for growing single crystals of metals (the Obreimov–Shubnikov method) [1, 2]. For the first time in the USSR, he had performed the researches of liquid helium, hydrogen, and other liquefied gases (1931–1934). In 1937, Shubnikov was subjected to repressions without any reasons, and therefore his name was absent in papers [6, 7] under the conditions of that time. He was a founder of the scientific school of low-temperature physics.

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