Magnetic Saturation and Apparent Molecular Fields of $MnCl_2 \cdot 4H_2O$

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PREVIOUS measurements[1] of magnetic moments of substances with small interionic interactions have demonstrated the applicability of the free spin Brillouin function up to over 99.5 percent saturation. Experimental investigation in the helium range of copper sulfate pentahydrate[2] above and copper chloride dihydrate below the Néel (antiferromagnetic transition[3]) temperature has shown nonsuperposition of the magnetic moment isotherms with M plotted against H/T. The departure of the moment from a Brillouin function (using the applied field,[4] H_0, in the argument of the function) has been attributed to molecular fields[5] seen by the paramagnetic ion. The purpose of this investigation is to measure approximately such molecular fields. Substances, such as $CuSO_4 \cdot 5H_2O$, with Néel temperatures below 1°K are difficult to investigate because of the determination and maintenance of the temperature; on the other hand, high Néel temperatures imply molecular fields which may be large compared with the available applied fields, i.e.,

$$\mu H_m \approx k T_N, \tag{1}$$

where μ is the atomic moment in Bohr magnetons, H_m is the molecular field, k is the Boltzmann constant, and T_N is the Néel temperature.

Manganous chloride tetrahydrate was chosen because the Néel temperature as determined by specific heat measurements[6] is 1.6°K in zero magnetic field. The apparatus for measurement of moments is the same as used in previous studies.[1] A sphere of small crystals was studied. The results are shown in Fig. 1. It is seen that practical saturation is achieved for 58 000 gauss and 1.3°K, as indicated by the nearly vanishing slope. From the point representing the saturation moment, one may now construct a Brillouin function. The approximate molecular field is obtained as follows. An arbitrary test point is chosen on either of the five magnetic moment isotherms. From this arbitrary point, one follows a constant moment line to the Brillouin function where the value of H/T is read. The value obtained by multiplying this value of H/T by the temperature corresponding to the isotherm chosen is taken as the effective field H_{eff}. Employing this method, one finds that the ratio between the apparent molecular field contribution ($H_0 - H_{\text{eff}}$) and the magnetic moment is a constant for all T to within ±10 percent over a wide range, corresponding to quadrupling H/T. Using this ratio and extrapolating the apparent molecular field contribution to saturation, one obtains ~14 000 gauss as the apparent molecular field, in rough agreement with Eq. (1).

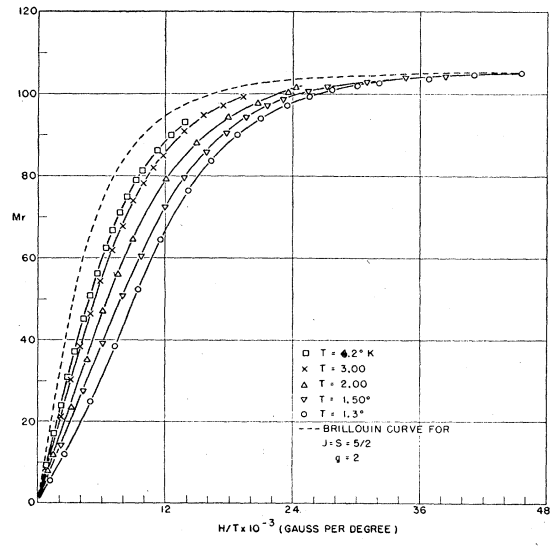

FIG. 1. A plot of relative magnetic moment (M_r) *vs* H/T for manganous chloride tetrahydrate for various temperatures. The broken line is a Brillouin function fitted at saturation.

With our results and neutron diffraction data such as obtained by Shull and Smart[7] to determine the array of sublattices, it would be possible to use the modified Van Vleck[8] method to calculate the antiferromagnetic exchange integral. However, if one assumes a simple model, where the molecular field is γM_0, and if one takes 5 Bohr magnetons as the saturation moment (M_0) per ion, one can compute γ. Now, from the Van Vleck[8] relation,

$$\gamma = zA/Ng^2\beta^2, \tag{2}$$

where N is the number of ions per cm³, $g(=2$ for $J=S)$ is the Landé factor, β is the Bohr magneton, and z is the number of nearest neighbors, one estimates energy density (zA) to be ~10^6 ergs/cm³, $A/2$ being the exchange integral.

The interpretation of the results of this investigation, in which manganous chloride tetrahydrate is magnetized up to saturation, enables one to explain the dispersion in the magnetic moment isotherms on the basis of the estimated molecular fields and suggests a means of calculating antiferromagnetic exchange integrals. A more detailed analysis will be presented later.

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A Proposed Experiment to Detect the Free Neutrino*

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THE success of the neutrino hypothesis[1,2] in explaining the observed facts of beta-decay provides reasonably convincing evidence for the existence of the neutrino. Nevertheless, the observation of an effect produced by a neutrino at a location other than its place of origin would be interesting because (1) any doubts as to its existence would be resolved, and (2) further information as to its properties and place in the nature of things might be obtained. It is with these aims in mind that we have developed the following technique which we plan to apply to the problem of detecting the free neutrino.

The neutrinos produced in the beta-decay of fission fragments[3] in a powerful chain reacting pile are to be allowed to pass through a large volume (10-ft³) liquid scintillator. The protons in the scintillator have a cross section of about 10^{-44} cm² for conversion by the fission fragment neutrinos to neutrons with the emission of a positron. It seems feasible to obtain some tenths such events per minute in the detector. Loading the scintillator solution with boron or cadmium compounds and counting the neutron capture gamma-pulse in delayed coincidence with the positron and annihilation radiation pulse assists in an important way in the reduction of background. Also necessary to the reduction of background in our experiment is the use of thick boron paraffin shielding, massive composite lead-steel shields, and Geiger tube anticoincidence umbrellas, the latter to discriminate against double pulses arising from μ-meson decay, stars, etc.

An essential feature of the experiment is the large scintillation detector required. As described in the following letter, such a detector has been developed which is energy sensitive and of good efficiency for the events in which we are interested. In this way it becomes possible to discriminate against background on an energy basis as well. By means of the foregoing techniques it appears possible to reduce the background to an acceptable figure. The required electronics have been developed. Our thanks and appreciation are due our colleagues: E. C. Anderson, L. J. Brown, F. B. Harrison, F. N. Hayes, C. W. Johnstone, R. L. Schuch, and Captain W. A. Walker for their whole-hearted cooperation in all phases of the work.

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Large Liquid Scintillation Detectors*

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THE technique outlined in the preceding letter for the detection of the free neutrino demands a detector having the following properties: (1) the provision of a large number of target protons in the sensitive volume; (2) a good efficiency for the counting of beta-particles, gamma-rays, and neutrons; (3) fair energy resolution; and (4) a controlled short mean detection (capture) time for the neutrons produced in the neutrino capture.

These requirements have been met in the design of a liquid scintillation counter in the shape of a cylinder containing 10.7 cubic feet of solution. A pilot model containing 2 cubic feet of solution was first constructed of brass, followed by the building of two full-scale counters of stainless steel and cold-rolled steel, respectively. The tanks were baked after fabrication to remove absorbed oils which would poison the solutions. The inside surfaces are coated with white Tygon paint.

Three filling solutions were developed:

(1) Toluene containing terphenyl, 2-(1-naphthyl)-5-phenyloxazole (αNPO), and methyl borate (or cadmium propionate in methanol).
(2) Triethylbenzene containing 2,5-diphenyloxazole, αNPO and methyl borate.
(3) Selected pure mineral oil containing 2,5-diphenyloxazole, αNPO, and methyl borate.

These solutions provide proton densities ranging from 4.6×10^{22} to 7.2×10^{22} per cubic centimeter. Control of boron or cadmium content permits selected mean neutron capture times down to about 5 microseconds. The capture times were computed by a Monte Carlo calculation using the Los Alamos MANIAC for a range of primary neutron energies and several cadmium concentrations. The results were checked experimentally with the detector in a preliminary manner by using spontaneous fission pulses in delayed coincidence with the neutrons emitted in such fissions. The optical absorption of the solutions for the scintillation light was reduced by purification and by use of αNPO as a scintillation spectrum shifter. The minimum absorption length accepted was several meters.

Scintillations in the solutions are detected by photomultiplier tubes spaced uniformly around the cylinder walls. Thirty-two tubes are employed in the pilot model and ninety in each full-scale counter. The tubes are balanced for uniform gain by means of their voltage dividers and are connected in parallel in one or more banks as needed. Linear pulse amplifiers, pulse-height analyzers, delayed coincidence analyzers and scalers comprise the associated electronic equipment.

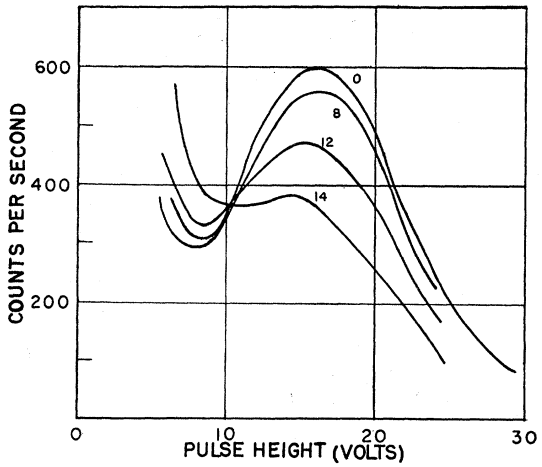

FIG. 1. Pulse-height distribution due to 0.41-Mev γ-rays from Au^{198} as a function of source position in the large detector. The source was on the axis of the cylinder, and the number on each curve indicates the distance in inches above the center. One-volt pulse-height channel widths were used.

Appropriate pipe connections, pumping systems, and reservoirs have been provided for filling the counters. Automatic float-switch systems control the liquid levels, and electronic vapor detectors guard against dangerous spills resulting from leaks and tube breakage. A nitrogen atmosphere is maintained over the solutions to retard hydrolysis and exclude air-borne radioactive contaminants.

The performance of these counters has been investigated by analyzing their output pulse rate and pulse-height spectrum with alpha-, beta-, and gamma-sources immersed at various points throughout their volumes and by using external neutron sources. Their response was found to be rather insensitive to source position (see Fig. 1), and their uniformity was considerably better in the full-scale counter than in the pilot model. It was found that the efficiency for counting 0.5-Mev gamma-radiation is about 75 percent. Figure 2 is a plot of data obtained with an external neutron source, a 6-inch thick lead shield around the detector, and with no cadmium or boron in the solution. The $p(n,\gamma)d$ peak is well resolved from the recoil proton background and that arising from capture gammas in the lead shield.

The performance of the system for delayed coincidence counting was tested by measuring the decay time of the cosmic ray μ-mesons stopping in the scintillator. Delayed coincidences falling

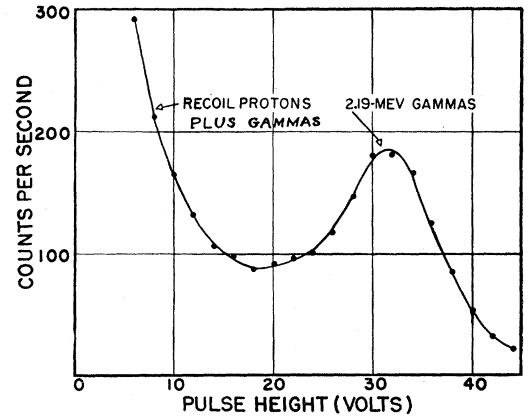

FIG. 2. Pulse-height distribution of scintillations in the large detector from an external neutron source. Pulses resulting from recoil protons and the gamma-rays from the capture of neutrons in the shield account for the rising portion on the left. The peak indicated is due to pulses from the gamma-rays from the reaction $n+H^1\rightarrow H^2+\gamma$ in the solution. The pulse-height scale is different from that in Fig. 1.