Magnetic Scattering of Slow Neutrons by Gaseous Oxygen*

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The scattering of neutrons by oxygen was measured as a function of wavelength from 2.5–10 A. By subtracting the calculated nuclear contribution the magnetic scattering is established. The magnitude of the magnetic interaction at long wavelengths is found to be in agreement with the theory of Halpern and Johnson. The measurement is not very sensitive to the details of the spatial distribution of the magnetic electrons. However, the observed wavelength dependence of the magnetic scattering is in agreement with the calculations of Kleiner based on the oxygen wave function derived by Meckler.

I. INTRODUCTION

THE scattering of slow neutrons by gaseous oxygen arises from both nuclear and magnetic interactions. The latter is of particular interest because oxygen is the simplest molecular system exhibiting paramagnetism, and there is a strong possibility that the magnetic scattering can be predicted from fundamental theory. In the past, magnetic measurements (as susceptibility) have dealt with the bulk or macroscopic properties of this gas. High-intensity neutron beams, however, now make it possible to perform experiments that probe the magnetic properties on a molecular (microscopic) scale. Although the scattering of slow neutrons by magnetic substances in general depends in a complicated way on intermolecular forces, in oxygen gas the intermolecular effects are extremely small and the magnetic interaction is between the neutron and the "free" magnetic moment of the oxygen molecule. This latter interaction has been theoretically formulated by Halpern and Johnson.[1]

In order to determine the magnetic scattering cross section it is necessary to subtract the nuclear effects from the total measured cross section. Fortunately the nuclear properties of oxygen are well suited for this purpose. The neutron absorption cross section is very small and the spin and isotope incoherence are negligible (zero spin and almost monoisotopic). For these reasons the coherent nuclear scattering amplitude, which is needed for calculating the nuclear effects, may be determined by a free-atom cross-section measurement at neutron energies of several electron volts,[2] where magnetic scattering is negligible.

II. METHOD

The present experiment is performed by measuring the transmission of neutrons through gaseous oxygen, which gives the total neutron cross section. Because the magnetic moments of the oxygen molecules are randomly oriented, there is no coherence between nuclear and magnetic scattering and the total cross section is separable into magnetic and nuclear terms. The contribution of the nuclear terms has been accurately calculated by Halpern and Appleton.[3] An estimate of the ratio of the magnetic to nuclear cross sections can be arrived at by simple considerations. At wavelengths large (~10 A) compared to the interatomic spacing of the oxygen atoms, the form factor for the nuclear and magnetic scattering approaches unity. For oxygen molecules at rest, in the limit of long wavelength, the nuclear scattering cross section of the molecule is approximately four times σ_b, the nuclear cross section of a bound oxygen atom.

The cross section for the magnetic interaction is given by[3]

$$\sigma_m=\frac{8\pi}{3}\left(\frac{\mu}{m}\right)^2 S(S+1)\left[\frac{\gamma^2e^2}{mc^2}\right]|F|^2, \tag{1}$$

where m is the mass of the neutron, μ is the reduced mass of the oxygen molecule-neutron system; S the electronic spin of the oxygen molecule; γ the magnetic moment of the neutron, expressed in Bohr magnetons and e^2/mc^2 the classical electron radius.

$$|F|^2\equiv\frac{1}{4\pi}\int|f(k)|^2\sin\theta d\theta d\varphi$$

is the integral form factor, normalized to approach unity as the neutron momentum $k\rightarrow 0$. Thus, in the long wavelength limit, the magnetic cross section is given by

$$\sigma_m{}^0=\frac{8\pi}{3}\left(\frac{\mu}{m}\right)^2 S(S+1)\left[\frac{\gamma^2e^2}{mc^2}\right]^2, \tag{2}$$

and the ratio

$$\frac{\sigma_{\text{magnetic}}}{\sigma_{\text{nuclear}}}\xrightarrow{\quad}\frac{2\pi}{3\sigma_b}S(S+1)\left[\frac{\gamma^2e^2}{mc^2}\right]^2=\frac{1}{3.5}. \tag{3}$$

As the Doppler motion (resulting from the finite sample temperature) affects the magnetic and nuclear scattering in the same way, the ratio is not changed by the Doppler effect.

In the long wavelength limit, a one percent error in the total cross-section measurement would result in about a

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[1] O. Halpern and M. H. Johnson, Phys. Rev. **55**, 898 (1939).

[2] E. Melkonian, Phys. Rev. **76**, 1750 (1949).

[3] O. Halpern and G. L. Appleton, Phys. Rev. **90**, 869 (1953).

4.5 percent error in the magnetic cross section, because of their relative magnitude. As the wavelength is decreased, the magnetic scattering decreases more rapidly than the nuclear scattering until at wavelengths <1 A the magnetic scattering is less than 2 percent of the nuclear scattering. For this reason the calculations of Halpern and Appleton were made in regions between 5–10 A, where one may hope to obtain the magnetic scattering with good accuracy from a total cross-section measurement. For the sake of completeness the experimental data were taken over a wavelength range from 2.5–10 A. The data for wavelengths less than 5 A can be compared with existing measurements.

III. EXPERIMENTAL DETAILS

Figure 1 shows the experimental apparatus. A collimated beam of neutrons from the Brookhaven pile is chopped into a series of approximately 200-μsec bursts by the "slow chopper," a rotating cadmium shutter. The beam passes through a filter which consists of four inches of Be maintained at liquid N_2 temperature. The Be transmits, with little attenuation, neutrons of wavelengths longer than 3.9 A but is essentially opaque to neutrons of shorter wavelength. Thus the filter eliminates neutrons of thermal and higher energies from the beam and thereby greatly reduces background. Next the beam passes through the sample vessel which is 3.5 meters long. The vessel may either be evacuated or filled to 10 atmospheres with O_2 and the total cross section of the O_2 is obtained by measuring the attenuation of the beam upon filling the vessel. The neutrons are detected in the shielded BF_3 counter, which is located 5.0 meters from the chopper, in a good geometry arrangement with respect to the sample vessel. The velocities or wavelengths of the detected neutrons are determined by measuring the flight time over the 5.0-meter path.

The chopped beam is monitored by a BF_3 counter which is in a beam obtained from the same region of the pile as the chopped beam. The monitor beam is filtered through graphite so that the mean wavelength of the neutrons detected by the monitor lies within the wavelength range 5 to 10 A that is being investigated with the chopper.

The oxygen used to fill the sample vessel is obtained from the electrolysis of H_2O, the only impurity being $\sim$1 percent hydrogen. The hydrogen is removed by passing the gas over heated CuO which catalyzes the oxidation of H_2 to form H_2O. The gas is than passed through a dry ice trap which removes the H_2O. The remaining impurities in the sample are traces of unoxidized H_2, and H_2O resulting from the vapor pressure at dry ice temperatures. Typical samples of gas from the target vessel were analyzed for H_2 and H_2O. These impurities were found to be present to the extent of 0.01 mole percent. Since the molecular cross sections of H_2 or H_2O are about ten times the molecular cross sections of O_2 at these wavelengths, the error in the measured O_2 cross section due to sample impurities is 0.2 percent.

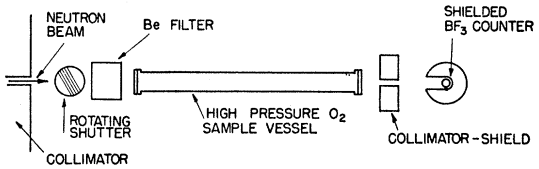

FIG. 1. Experimental arrangement of apparatus.

To calculate the cross section from the measured transmission the number of molecules/cm^2 in the sample is required. Measurements of the temperature and pressure of the gas and the length of the target vessel were made with $\frac{1}{4}$ percent accuracy. The molecular density was computed by means of the Beattie-Bridgeman equation of state. At the operating pressure of 10 atmospheres this equation of state indicates a 0.7 percent deviation from the perfect gas law.

The time-of-flight spectra of the neutrons in the beam is analyzed by a 12-channel delayed-coincidence analyzer. Flight times are measured relative to a one-megacycle crystal oscillator. The zero of time is set by a calibration procedure referring to the crystal lattice spacing of graphite[4] which is known from x-ray diffraction experiments. The neutron time of flight is measured with an accuracy of about $\frac{1}{4}$ percent.

The transmission is given by the ratio of the counting rate less background with O_2 in the beam divided by the counting rate less background of the open beam. Backgrounds are measured by inserting a 0.010-in. cadmium foil in the beam. This foil absorbs more than 99 percent of the timed neutrons but does not affect the epicadmium background neutrons. The data were obtained by repeatedly running the four measurements involved in the measurement of the transmission. The cross section was calculated from the measured transmission and molecular density of the sample. The results are shown in Fig. 2. The abscissa is neutron wavelength including a small correction for the finite resolution of the spectrometer and the ordinate is the total cross section per O_2 molecule. The indicated errors range from ± 1.6 percent at 10 A to ± 0.1 percent at 5 A. In order to compare the results of the present experiment with existing cross section information, which extends up to 5 A, the cross section was measured from 2.5 A to 5 A with somewhat poorer statistics. These data are in good agreement with the measurements of Melkonian.[2]

IV. CONCLUSIONS

Figure 3 shows the experimental magnetic scattering in the region from 5–10 A, obtained by subtracting the calculated nuclear scattering[3] from the measured points. The solid line is a theoretical prediction of the magnetic

[4] Carter, Palevsky, Myers, and Hughes, Phys. Rev. **92**, 716 (1953).

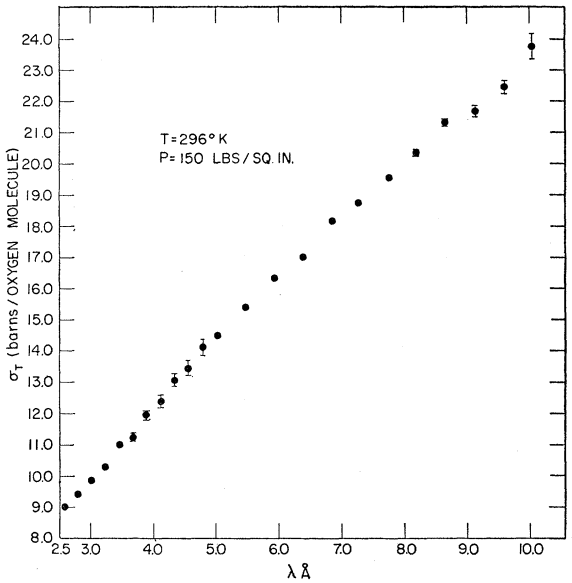

FIG. 2. Total cross section of oxygen in the wavelength region 2.5–10 A. Where no error is shown, the statistical error is equal to or less than the size of the plotted point.

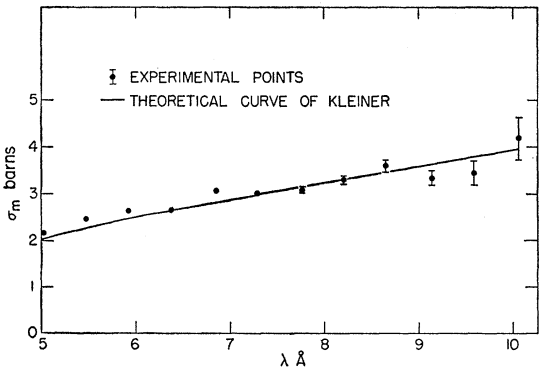

FIG. 3. Magnetic scattering of oxygen in the wavelength region 5–10 A. Where no error is shown, the statistical error is equal to or less than the size of the plotted point.

scattering made by Kleiner[5] using the oxygen wave function of Meckler.[6] In the region 7.5 to 10 A, Kleiner's calculations indicate that for any reasonable oxygen wave functions the magnetic form factor is very nearly unity. In this wavelength interval the slope of the magnetic scattering depends only on the limiting cross section σ_m^0 [Eq. (2)] and the temperature of the gas. Evaluation of Eq. (2) gives $\sigma_m^0=4.58$ barns, and use of this value in calculating the Doppler effect gives a slope of 0.356 barn/A.

In order to compare the experimental results with those theoretically expected, a least squares fit to the six experimental points was made between 7.7 and 10.1 A gives an experimental slope of 0.340 ± 0.007 barn/A, which is about 4.5 percent lower than the theoretical prediction. This discrepancy is to be compared to the 2 percent standard deviation in the least-squares fit of the experimental data.

The magnetic scattering between 5 and 7.5 A depends not only on the σ_m^0 and temperature but also to a small extent on the details of the spatial distribution of the magnetic electrons in the molecule, i.e., in this wavelength region there is a departure of the form factor from unity. The experimental points here are known with about one percent (standard deviation) accuracy; however, they all lie above the theoretical curve of Kleiner. The average deviation is seen (Fig. 3) to be about 6 percent. Even though the form factor in this wavelength region is not unity, the work of Kleiner indicates that the approximate wave functions he used should accurately predict the magnetic scattering. Both the discrepancy in the slope and magnitude of the experimental cross sections as compared to theory could be explained by a small (~1 percent) error in the calculation of the nuclear scattering.

This experiment establishes the magnetic scattering of neutrons by oxygen gas. The slope of the magnetic cross section curve verifies that at long wavelengths σ_m^0 approaches the value predicted by theory for the interaction of the neutron and a "free" magnetic moment. The measurement is rather insensitive to the details of the spatial distribution of the magnetic electrons because a transmission experiment depends on the integral of the form factor over all angles. The details are further smeared out by the Doppler motion of the gas. In the region between 5 and 10 A, these two effects make it very difficult to obtain with any accuracy properties of the distribution of the magnetic electrons in oxygen. The smearing arising from the Doppler motion becomes less serious at shorter wavelengths and it would be possible to learn more about the magnetic moment distribution in the molecule from data between 2 and 5 A if the nuclear contributions could be calculated.

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[5] W. Kleiner, Phys. Rev. **97**, 411 (1955).
[6] A. Meckler, J. Chem. Phys. **21**, 1750 (1953).