To summarize the present situation: attenuation experiments for keV photons are in reasonable agreement with each other. The accuracy of the measurements can be improved by at least one order of magnitude without undue difficulties. The theoretical formulas agree well with the trend of experimental data, but are not too reliable for exact prediction. The procedure of Hall[10] leads to values which are too high by 5% for low values of Z and too high by 35% for high values of Z.

PHYSICAL REVIEW VOLUME 137, NUMBER 3A 1 FEBRUARY 1965

Contributions to the V^{51} Nuclear Magnetic Resonance Frequency Shift and Susceptibility in Vanadium Sesquioxide

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(Received 18 August 1964)

The V^{51} NMR frequency shift $(\Delta\nu/\nu)$ in metallic V_2O_3 has been measured in the temperature range of 175 to 575°K the frequency shift at these two temperatures being -0.61% and $+0.1\%$, respectively. Various contributions to the frequency shift and magnetic susceptibility are determined by constructing a $(\Delta\nu/\nu)$-versus-χ diagram. The d-band spin susceptibility χ_d is found to obey a Curie-Weiss law down to the transition temperature of about 160°K. A large temperature-independent orbital susceptibility is found to be $\chi_{VV}\simeq 1.52\%$. From the temperature dependence of $\chi_d(T)$ and the corresponding frequency shift $(\Delta\nu/\nu)_d$, a negative hyperfine field of $H_d^{\mathrm{hf}}\simeq -140$ kOe/Bohr magneton, resulting from core polarization, a temperature-dependent orbital hyperfine field, and a dipolar hyperfine field, is found. With decreasing temperature the V^{51} NMR signal abruptly disappears at the transition temperature, suggesting that V_2O_3 undergoes antiferromagnetic ordering for temperatures less than 160°K. Contributions to the V^{51} NMR linewidth are discussed.

I. INTRODUCTION

THE series of compounds Ti_2O_3, V_2O_3, Cr_2O_3, and α-Fe_2O_3 have the same crystal structure as Al_2O_3 (corundum). The electrical and magnetic properties of these compounds, however, are markedly different. Two of them, Cr_2O_3 and α-Fe_2O_3, are antiferromagnetic and highly resistive. The two remaining compounds, Ti_2O_3 and V_2O_3, also undergo a phase transition as evidenced by peaks in the measured specific heat[1] and magnetic susceptibility[2] curves, but the susceptibility curve does not exhibit typical antiferromagnetic behavior. In addition, Ti_2O_3 and V_2O_3 undergo a metallic to semiconductor transition,[3] with decreasing temperature, at their respective transition temperatures.

Pearson[4] and Morin[3,5] have proposed that the phase transition for Ti_2O_3 and V_2O_3 is associated with a change from a paramagnetic state to an antiferromagnetic state and this antiferromagnetic phase transition could account for the electrical properties of these compounds. Goodenough[6] suggests that below the transition temperature, a strong cation-cation interaction will lead to direct bond formation between the cations along the trigonal axis and hence the phase transition is not the result of an antiferromagnetic transition. Recently Adler and Feinleib[7] have suggested that the metallic-semiconductor transition is a result of an energy gap in the d band arising from a crystalline distortion due to the crystallographic transformation to lower symmetry (rhombohedral to monoclinic). Their analysis for this mechanism does not rule out antiferromagnetic ordering for Ti_2O_3 and V_2O_3, but that antiferromagnetism need not be associated with the electrical properties of these solids. To date, there has been no experimental confirmation of magnetic ordering in V_2O_3, although antiferromagnetic ordering has been established for Ti_2O_3.[8,9]

Paramagnetic susceptibility measurements[1,10,11] on V_2O_3 have shown that the susceptibility obeys a Curie-Weiss law. However, there is a wide variation in these results concerning the value of the effective number of Bohr magnetons per vanadium atom. For example, μ_{eff} is found to lie in the range of 2.63 to 3.48 Bohr magnetons per vanadium atom. The discrepancy between these measurements has been attributed[10] to the use of nonstoichiometric samples of V_2O_3. However, in the analysis of the susceptibility data there was no correc-

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tion made for the contribution of Van Vleck paramagnetism to the total magnetic susceptibility.

Nuclear-magnetic resonance (NMR) in solids is a convenient method for studying their electrical and magnetic properties. The present investigation is concerned primarily with the V^{51} NMR as a method for studying the paramagnetic properties of metallic V_2O_3. The contributions to the susceptibility by Van Vleck and *d*-band spin paramagnetism are obtained by studying the V^{51} NMR frequency shift as a function of susceptibility.[12] In addition, the V^{51} NMR was followed through the transition temperature of about 160°K and the experimental observations are shown to be consistent with magnetic ordering in V_2O_3 below 160°K.

II. EXPERIMENTAL

Single crystals of V_2O_3 were obtained from the Linde Air Products Company. The crystals were determined by mass analysis to be within 0.1% of the stoichiometric composition. Susceptibility measurements[13] were performed on the V_2O_3 single crystals. In order to allow complete penetration of the rf magnetic field in metallic V_2O_3, the single crystals were subsequently ground into a powder and passed through a #400 mesh screen.

The V^{51} NMR was observed using an induction spectrometer and a conventional 12-in. electromagnet. Sample temperatures between 80 and 575°K were obtained by use of a Varian V-4540 variable temperature control unit. The V^{51} NMR frequency-shift measurements were made at a fixed frequency of 16 Mc/sec and measuring the magnetic field for which resonance occurs with a $NaVO_3$ reference solution. A typical room temperature V^{51} NMR spectrum is shown in Fig. 1. The Al resonance is due to the aluminum in the NMR probe. The peak-to-peak linewidth of the V^{51} NMR is approximately 16 G and was found to be independent of magnetic field strength. In the temperature range for which the V^{51} NMR was observed, there was no discernible indication of anisotropic or quadrupolar interactions contributing to the line shape.

III. CONTRIBUTIONS TO THE SUSCEPTIBILITY AND V^{51} NMR FREQUENCY SHIFT IN V_2O_3

1. Susceptibility

The two principle contributions to the susceptibility are the temperature dependent *d*-band spin paramagnetism $\chi_d(T)$ and the temperature-independent orbital paramagnetism χ_{VV} which is the analog in metals of the Van Vleck paramagnetic susceptibility.[14] The *s* electrons do not contribute to the electrical and magnetic properties of V_2O_3 since they are considered to be localized on the oxygen atoms.[6] The total susceptibility of V_2O_3 also includes a diamagnetic term χ_{dia}, to which the ion cores contribute. For temperature T, the total susceptibility $\chi(T)$ is given by

$$\chi(T)=\chi_d(T)+\chi_{VV}+\chi_{\text{dia}}. \qquad (1)$$

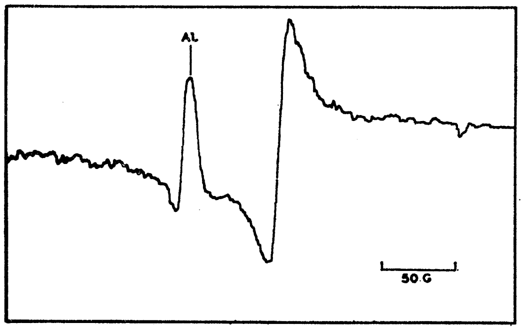

FIG. 1. A room temperature tracing of the V^{51} NMR absorption derivative in powdered V_2O_3.

An estimate of χ_{dia} is now given since the remaining terms are obtained from the graphical procedure to be discussed.

χ_{dia}: The estimated[15] value for χ_{dia} of vanadium is $\chi_{\text{dia}}\simeq-6\times10^{-6}$ emu/mole. The estimated value for O^{2-} is $\chi_{\text{dia}}\simeq-7\times10^{-6}$ emu/mole, which is not unreasonable as may be seen by comparing it to the measured[16] value for neon of $\chi_{\text{neon}}=-6.74\times10^{-6}$ emu/mole. Thus, for $VO_{1.5}$, $\chi_{\text{dia}}\simeq-18\times10^{-6}$ emu/mole. The diamagnetic susceptibility is a small correction when compared to the total susceptibility of $\chi(300°\text{K})\simeq900\times10^{-6}$ emu/mole.

2. V^{51} NMR Frequency Shift

The V^{51} NMR frequency in V_2O_3 is shifted an amount $(\Delta\nu/\nu)$ by interactions between the *d*-band electrons and the nuclear magnetic moment. This NMR frequency shift consists of two contributions: (1) a temperature-dependent frequency shift $(\Delta\nu/\nu)_d$ resulting from hyperfine interactions of the form $A\mathbf{I}\cdot\langle\mathbf{S}\rangle$, where the temperature dependence of these interactions is contained in the time averaged electron spin $\langle S\rangle$; (2) a temperature-independent orbital-frequency shift $(\Delta\nu/\nu)_{VV}$ arising from Van Vleck paramagnetism.[17] Because of the translational motion of the *d* electrons through the crystal, the angular moment l_z is quenched, i.e., $\langle l_z\rangle=0$. Thus the *field-induced* temperature-independent frequency-shift $(\Delta\nu/\nu)_{VV}$ is second order in l_z, as is the field induced orbital paramagnetic susceptibility χ_{VV}. Also in second order in l_z is a *spin-orbit*

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[12] This technique was first fully utilized by A. M. Clogston, A. C. Gossard, V. Jaccarino, and Y. Yafet, Phys. Rev. Letters **9**, 262 (1962). See also A. M. Clogston and V. Jaccarino, Phys. Rev. **121**, 779 (1961).

[13] The author is indebted to R. C. Sherwood for performing the susceptibility measurements.

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[16] *Handbook of Chemistry and Physics* (Chemical Rubber Publishing Company, Cleveland, Ohio, 1963) 44th ed.

[17] T. Moriya, Phys. Chem. Solids **11**, 175 (1959).

induced temperature-dependent orbital-frequency shift which is proportional to $\langle S\rangle$ and therefore will be included in $(\Delta\nu/\nu)_d$. The temperature-dependent frequency shift $(\Delta\nu/\nu)_d$ thus consists of the three following contributions, all of which are proportional to $\langle S\rangle$: (1) a shift resulting from core polarization of the inner s-shell electrons by the d-band electrons; (2) the second-order shift resulting from the spin-orbit interaction; (3) a shift resulting from the dipolar interaction between the nuclear magnetic moment and the conduction electrons since the vanadium site has less than cubic symmetry.

For the case of negligible nuclear quadrupole interactions, the total frequency shift $(\Delta\nu/\nu)$ is written as

$$(\Delta\nu/\nu)=(\Delta\nu/\nu)_d+(\Delta\nu/\nu)_{VV}, \qquad (2)$$

$$(\Delta\nu/\nu)=\alpha_d\chi_d(T)+\beta'\chi_{VV}, \qquad (3)$$

where $\alpha_n=(N\beta)^{-1}H_d{}^{\text{hf}}$, with N Avogadro's number, β the Bohr magneton, and $H_d{}^{\text{hf}}$ the total hyperfine field per Bohr magneton. The susceptibilities are expressed in units of emu/mole. In the case of the free ion, β' is rigourously given as[17]

$$\beta'=(2/N)\langle r^{-3}\rangle, \qquad (4)$$

where $\langle r^{-3}\rangle$ is the expectation value of r^{-3} for the free ion concerned. The electronic configuration of V^{3+} is $3d^2$ and it is estimated[18] that $\langle r^{-3}\rangle=3.2$ a.u. for the V^{3+} ion. Normally a reduction factor $\xi=\langle r^{-3}\rangle_{\text{solid}}/\langle r^{-3}\rangle_{\text{ion}}$ would be included in Eqs. (3) and (4); however, for the present investigation ξ is chosen to be unity.

There is no frequency shift due to diamagnetism included in Eqs. (2) and (3) since the incremental-diamagnetic shift between V_2O_3 and the reference compound ($NaVO_3$) used to measure the frequency shift is negligible.

IV. CONSTRUCTION OF THE $(\Delta\nu/\nu)$ VERSUS χ DIAGRAM

Single crystal susceptibility measurements were made in the temperature range of 175 to 300°K. Within experimental error there was no anisotropy observed in the susceptibility measurements for various crystalline orientations with respect to the magnetic field.

The measured frequency shift $(\Delta\nu/\nu)$ as a function of susceptibility χ, with temperature the implicit variable, is shown in Fig. 2. The temperature range for the frequency shift measurements was 175 to 575°K. The solid portion of the line shown in Fig. 2 is the data for which the susceptibility χ was measured. Since $(\Delta\nu/\nu)$ versus χ is linear in this temperature region, it is plausible to assume that $(\Delta\nu/\nu)$ versus χ will be linear in the higher temperature region, i.e., 300 to 575°K. In this manner the susceptibility $\chi(T)$ in the 300 to 575°K temperature range is determined by measurement of the V^{51} NMR frequency shift. From Eqs. (2) and (3), it is seen that the slope of $(\Delta\nu/\nu)$ versus χ determines α_d. The value so obtained is $\alpha_d=-25.0(\text{emu/mole})^{-1}$ yielding a hyperfine field $H_d{}^{\text{hf}}=-140$ kOe/Bohr magneton. Usually the hyperfine field $H_d{}^{\text{hf}}$ is expressed in units of kOe/unit spin,[12] thereby necessitating knowledge of the electronic g value since, for the hyperfine field expressed in these units, $\alpha_d=(Ng\beta)^{-1}H_d{}^{\text{hf}}$. The g value for V_2O_3, at the present time, however, has not been measured. For the case of no spin-orbit interaction, $g=2$ and the resulting hyperfine field would be $H_d{}^{\text{hf}}=-280$ kOe/unit spin.

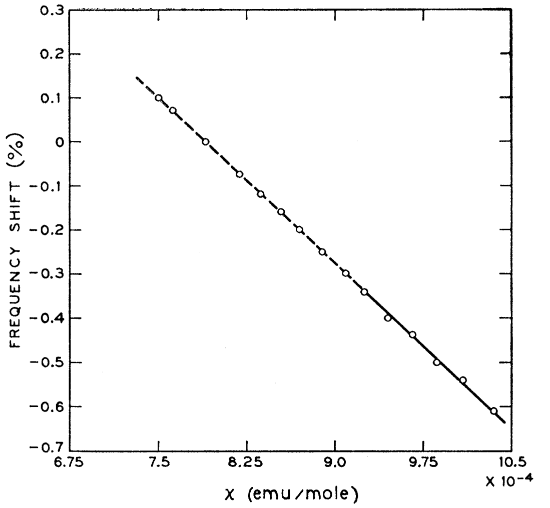

FIG. 2. The experimentally observed V^{51} NMR frequency shift $(\Delta\nu/\nu)$ in V_2O_3 plotted as a function of the susceptibility χ, with temperature the implicit parameter. The solid line is drawn through the data for which the susceptibility χ was measured. The straight line is a best fit of the data.

The g values have been measured[19] for the dilute system $V^{3+}:Al_2O_3$. However, V_2O_3 is metallic while $V^{3+}:Al_2O_3$ is an insulator and hence, there may be no correlation between the results obtained from the dilute system ($V^{3+}:Al_2O_3$) and the concentrated system (V_2O_3). For example, the susceptibility of $V^{3+}:Al_2O_3$ has been measured[19] with the result that extrapolating from the dilute crystal to the concentrated crystal predicts $(\chi_{VV})_\perp\simeq600\times10^{-6}$ emu/mole and $(\chi_{VV})_\parallel\simeq200\times10^{-6}$ emu/mole. Such a large anisotropic susceptibility was not observed for V_2O_3. Furthermore, evidence of an anisotropic g value and an anisotropic Van Vleck susceptibility would be reflected in the line shape of the V^{51} NMR in powdered V_2O_3. As previously mentioned, there was no indication of anisotropic contributions to the NMR line shape, thus leading to the conclusion that the magnetic properties of V_2O_3 are markedly different from those of the "concentrated" $V^{3+}:Al_2O_3$ crystal.

The diagram shown in Fig. 3 is now constructed. The diamagnetic contribution to the susceptibility χ_{dia} corresponds to the negative displacement from the origin

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[18] R. E. Watson (private communication).

[19] See W. H. Brumage, C. R. Quade, and C. C. Lin, Phys. Rev. **131**, 949 (1963) and references therein.

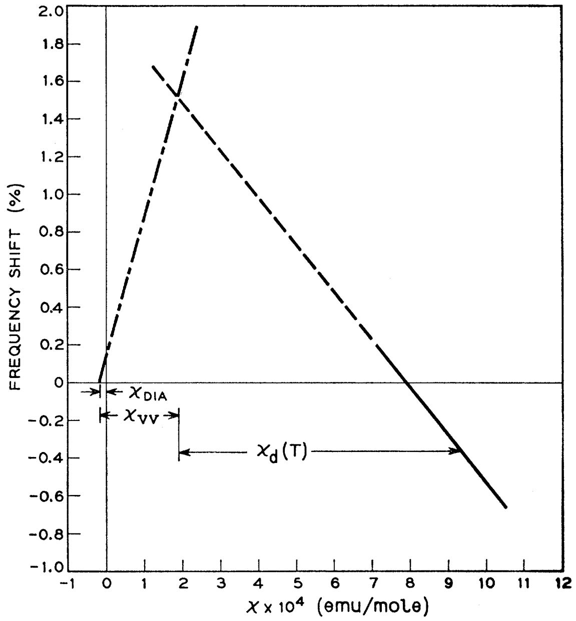

FIG. 3. A diagram of the V^{51} NMR frequency shift $(\Delta\nu/\nu)$ in V_2O_3 versus the susceptibility χ.

of the $(\Delta\nu/\nu)-\chi$ axes. A line with slope β' is drawn from the end of the χ_{dia} contribution. The observed $(\Delta\nu/\nu)$ versus $\chi(T)$ data is shown in the lower right hand portion of the figure and is extrapolated until it intersects the line with slope β'. This extrapolation procedure provides all of the information concerning χ that is needed. The magnitudes of the contributions to the susceptibility are indicated by the horizontal lines shown in Fig. 3. From Fig. 3, the Van Vleck temperature-independent susceptibility is determined to be $\chi_{VV} \simeq 210 \times 10^{-6}$ emu/mole corresponding to a frequency shift $(\Delta\nu/\nu)_{VV} = 1.52\%$. It is interesting to note that χ_{VV} for V_2O_3 is of the same order of magnitude as χ_{VV} found in the V_3X intermetallic compounds.[12] As can be seen from Fig. 3, $(\Delta\nu/\nu)$ becomes positive when $(\Delta\nu/\nu)_{VV} > |(\Delta\nu/\nu)_d|$. Table I lists the relevant parameters used in the construction of the $(\Delta\nu/\nu)$ versus χ diagram.

TABLE I. Parameters used in the analysis of the V^{51} NMR frequency shift.

α_d	-25.0 (emu/mole)$^{-1}$	H_d^{hf}	-140 kOe/Bohr magneton
β'	71.7 (emu/mole)$^{-1}$	$2\beta\langle r^{-3}\rangle$ $\langle r^{-3}\rangle = 3.2$ a.u.	395 kOe

The first column gives the parameters defined in Eqs. (3) and (4). The second column gives the hyperfine field per Bohr magneton H_d^{hf}, obtained from the analysis of the frequency shift, and the hyperfine field per unit field induced orbital angular momentum $2\beta\langle r^{-3}\rangle$.

V. *d*-BAND SPIN SUSCEPTIBILITY $\chi_d(T)$

The magnitude and temperature dependence of the *d*-band spin susceptibility $\chi_d(T)$ is found by subtracting the diamagnetic and Van Vleck susceptibilities from the total susceptibility, i.e.,

$$\chi_d(T) = \chi(T) - \chi_{VV} - \chi_{\text{dia}}, \tag{5}$$

Fig. 4 shows the results obtained from plotting $(1/\chi_n)$ as a function of temperature T. As is evident from Fig. 4, $\chi_d(T)$ obeys a Curie-Weiss law, i.e.,

$$\chi_d(T) = C/(T+\theta) \tag{6}$$

with $C = 0.657$ (emu/mole)°K and $\theta = -600$°K. The Curie constant C is given by $C = (N\mu_{\text{eff}}^2\beta^2)/3k$ with N Avogadro's number, β the Bohr magneton, μ_{eff} the effective number of Bohr magnetons per vanadium atom and k Boltzman's constant. For $C = 0.657$ (emu/mole)°K as found from Fig. 4, the effective number of Bohr magnetons per vanadium atom is $\mu_{\text{eff}} = 2.37$, a value to be compared with the free ion value of $\mu_{\text{eff}} = 2.83$. The difference between the observed μ^{eff} and the ion value is thus an indication of the strength of the spin-orbit interaction. The value of $\theta = -600$°K is to be compared with previously published data[10,11] of $\theta \simeq -1100$°K. This large value for θ was obtained from susceptibility measurements with no correction for the contribution of the temperature-independent Van Vleck susceptibility to the total susceptibility. Hence for V_2O_3, $-\theta/T_N \simeq 3.75$ for $T_N \simeq 160$°K.

VI V^{51} NMR THROUGH THE PHASE TRANSITION AND NMR LINEWIDTHS

The V^{51} NMR is observable down to a temperature of about 160°K. At this temperature the resonance abruptly disappears. Below 160°K no V^{51} resonance was observed in the range of 0 to 20 kOe for a frequency of 16 Mc/sec. A search for the V^{51} NMR was also conducted at 4.2 and 20°K with negative results. Past experience indicates that such an abrupt disappearance of the NMR resonance is usually associated with magnetic ordering. The assumption that the static quadrupolar interaction is larger than the Zeeman interaction is improbable since it would necessitate an unusually large value of $e^2qQ/h \simeq 50$ Mc/sec. Furthermore, since

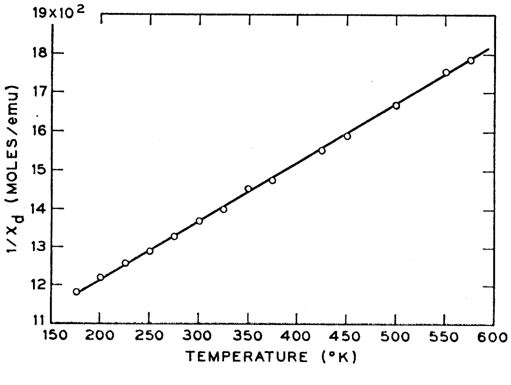

FIG. 4. The temperature dependence of the *d*-band spin susceptibility $\chi_d(T)$. The straight line is a best fit of the data.

the symmetry at the vanadium site is less than cubic for $T>160°\mathrm{K}$, it is unlikely that the crystallographic transformation from rhombohedral to monoclinic symmetry will lead to a quadrupolar broadened V^{51} NMR line shape for $T<160°\mathrm{K}$. Thus, it is not inconsistent to conclude that V_2O_3 undergoes an antiferromagnetic transition at 160°K; and for temperatures less than 160°K, the V^{51} NMR would occur at higher frequencies (200–350 Mc/sec).

An estimate for the NMR linewidth δH for a nucleus of a paramagnetic ion has been given by Moriya[20] with the result

$$\delta H \simeq \left(\frac{2\pi}{3}\right)^{1/2} \frac{A^2 S(S+1)}{3\gamma \hbar^2 \omega_e}, \qquad (7)$$

where A is the hyperfine coupling constant, S the electron spin, γ the nuclear gyromagnetic ratio and ω_e is defined by

$$\omega_e^2 = \tfrac{2}{3}(J/\hbar)^2 Z S(S+1), \qquad (8)$$

where Z is the number of neighbors and $-J$ is the antiferromagnetic exchange constant. The molecular field approximation gives a relationship between the exchange constant J and the Curie temperature θ as[6]

$$\frac{3k\theta}{2S(S+1)ZJ} \simeq 1. \qquad (9)$$

From Eqs. (8) and (9), and taking $Z \simeq 6$,

$$\omega_e \simeq (\tfrac{1}{8})^{1/2}(k\theta)/\hbar \qquad (10)$$

or for $|\theta| \simeq 600°\mathrm{K}$, $\omega_e \simeq 3\times 10^{13}\ \mathrm{sec}^{-1}$. An estimate for the hyperfine coupling constant A can be made by combining the previously determined values of H_d^{hf} and μ_{eff} with the result $A \simeq 100\times 10^{-4}\ \mathrm{cm}^{-1}$. The line width δH is thus calculated from Eq. (7) to be

$$(\delta H)_{\mathrm{calc}} \simeq 20\ \mathrm{G} \qquad (11)$$

which is in satisfactory agreement with the observed $\delta H \simeq 16$ G. For comparison, the dipolar linewidth in V_2O_3 is of the order of 3 G.

ACKNOWLEDGMENTS

The author wishes to thank C. Britz for experimental assistance and S. Geschwind, A. C. Gossard, and V. Jaccarino for many helpful discussions.

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[20] T. Moriya, Progr. Theoret. Physics (Kyoto) 16, 641 (1956).

PHYSICAL REVIEW VOLUME 137, NUMBER 3A 1 FEBRUARY 1965

Magnetic Properties of CoF_2

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(Received 14 September 1964)

CoF_2 is a simple two-sublattice antiferromagnet and has a rutile crystal structure. Each Co^{++} ion is surrounded by a rhombically distorted octahedron of fluorine anions and the crystal-field parameters are known from an analysis of infrared absorption measurements. A good description of a single Co^{++} system can be obtained in terms of a spin Hamiltonian with $S=\frac{3}{2}$. In this paper, an exchange Hamiltonian for the entire lattice is determined in which the only unknowns are the exchange parameters J_1 and J_2 describing interactions between nearest- and next-nearest-neighbor Co^{++} ions, respectively. The resulting intermediate coupling problem is attacked by spin-wave methods, introducing operators which represent deviations of spin from the molecular-field ground state. The parameters J_1 and J_2 are determined by calculating the antiferromagnetic resonance frequency and the temperature dependence of sublattice magnetization, and by fitting theory to experiment. We find that J_1 is very small and probably ferromagnetic. With the exchange Hamiltonian now completely determined, a molecular-field treatment is shown to reproduce closely the measured parallel and perpendicular magnetic susceptibilities in the temperature range 0 to 300°K (except in the immediate vicinity of the Néel temperature), and is used to discuss the nuclear magnetic resonance of Co^{59} in CoF_2.

1. INTRODUCTION

SOME years ago, an attempt was made by Nakamura and Taketa[1] to obtain a theoretical understanding of the somewhat unusual magnetic properties of the antiferromagnetic salt CoF_2. At that time, however, the experimental information was scant, consisting of little more than a knowledge of the magnetic structure of the crystal and some rather incomplete susceptibility data. Even so, they found it possible to obtain a qualitative explanation of the unusual features of the susceptibility curves and were able to demonstrate the origin of the large anisotropy which exists in the salt and to obtain some measure of the exchange interaction present. Since that time, a considerable amount of new experimental data has been obtained and we are now in a position where a more quantitative approach to the problem can be attempted.

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[1] T. Nakamura and H. Taketa, Progr. Theoret. Phys. (Kyoto) 13, 129 (1955).