3/2→1/2. In each instance, there were at least two strong and well-resolved components. The frequency differences between these components have been measured with good accuracy.

It has been found that the observed Zeeman spectra of the OCS^{33} $J=1\rightarrow2$ hyperfine structure are completely consistent with theory[5,7] if $|g_N|=0.421$, $|g_{mole}|=0.026$, and g_{mole} and g_N have opposite signs. Here, g_N is the S^{33} nuclear g-factor and g_{mole} is the rotational g-factor for OCS. The quantity g_{mole} determined here is consistent with the value of g_{mole} determined directly from the OCS^{32} Zeeman spectrum.

In an attempt to establish the absolute signs of the g-factors, Eshbach[8] has used an arrangement providing a circularly polarized wave and succeeded in proving that the absolute signs of g_N and g_{mole} are respectively positive and negative.

Using $g(N_{33})=0.421$ and $I=3/2$, the final result for the nuclear magnetic moment of S^{33} is

$$\mu(S^{33})=0.632\pm0.010 \text{ n.m.}$$

This result is within the Schmidt limits and is in agreement with the prediction of the single particle model of nuclear shell structure.

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† This work was supported by the U. S. Navy, Signal Corps, and Air Force under ONR contract.
[1] H. L. Poss, *The Properties of Atomic Nuclei*, Brookhaven National Laboratory, October, 1949, p. 11, unpublished.
[2] C. K. Jen, Phys. Rev. **78**, 339 (1950).
[3] J. E. Mack, Rev. Mod. Phys. **22**, 65 (1950).
[4] J. R. Eshbach and R. E. Hillger, Quarterly Progress Report, Research Laboratory of Electronics, M.I.T., pp. 42–43, July, 1950, unpublished.
[5] C. K. Jen, Phys. Rev. **74**, 1396 (1948); Phys. Rev. **76**, 1494 (1949).
[6] C. H. Townes and S. Geschwind, Phys. Rev. **74**, 626 (1948).
[7] F. Coester, Phys. Rev. **77**, 454 (1950).
[8] A complete report of this work will be published later.

Angular Dependence of Hyperfine Structure for the Copper Ion

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SHORTLY after the discovery of hyperfine structure in solids by Penrose, it was found by a number of independent workers[1] that the experimental results are not in agreement with the theoretical deductions, if one assumes that the electron-orbit-nuclear spin and electron-spin-nuclear spin terms are responsible for the hyperfine separation. A careful experimental investigation by Ingram[2] has accentuated the discrepancy between theory and experiment. Recently Abragam[3] has proposed a theory to account for the experimental results. We would like to suggest in this note another term which is perhaps partly responsible for the angular dependence of hyperfine structure.

A new interaction term was suggested to us by the recent work of England and Schneider[4] who investigated the absorption properties of the Mn^{++} ion as activators in zinc sulfide phosphors. The spectroscopic ground state of the ion is $^6S_{5/2}$, and therefore the interaction terms used for the Cu^{++} ion will give no contribution to the separation. To account for the observed hyperfine splitting it is necessary to use a term[5] of the form $a_2S\cdot I$. Since the d electrons are responsible for the spin in both copper and manganese, we have investigated the consequences of assuming that a_2 is approximately the same for copper and manganese. We have used the interaction potential

$$H'=-a_1L\cdot I+a_1\left\{\frac{3(S\cdot r)(I\cdot r)}{r^2}-I\cdot S\right\}-a_2S\cdot I+\gamma H\cdot I.$$

This expression differs from that taken by others in that the signs of the first two terms are different, and that the additional $S\cdot I$ term is introduced.

It follows from fairly general group-theoretical considerations that for a doubly degenerate electron ground state the above

interaction potential leads to the following term values:

$$E^+=(g_{\shortparallel}{}^2\cos^2\theta+g_{\perp}{}^2\sin^2\theta)^{\frac{1}{2}}\mu_BH$$
$$+\left[\left(\frac{\Delta\nu_{\shortparallel}g_{\shortparallel}}{2g}+\gamma H\right)^2\cos^2\theta+\left(\frac{\Delta\nu_{\perp}g_{\perp}}{2g}+\gamma H\right)^2\sin^2\theta\right]^{\frac{1}{2}}m_I,$$

$$E^-=-(g_{\shortparallel}{}^2\cos^2\theta+g_{\perp}{}^2\sin^2\theta)^{\frac{1}{2}}\mu_BH$$
$$-\left[\left(\frac{\Delta\nu_{\shortparallel}g_{\shortparallel}}{2g}-\gamma H\right)^2\cos^2\theta+\left(\frac{\Delta\nu_{\perp}g_{\perp}}{2g}-\gamma H\right)^2\sin^2\theta\right]^{\frac{1}{2}}m_I.$$

The symbols $\Delta\nu^{\shortparallel}$ and $\Delta\nu^{\perp}$ represent the hyperfine structure component separations in a parallel and perpendicular magnetic field. For the particular case of Cu^{++} these quantities are given by

$$\Delta\nu_{\shortparallel}=2a_1[-4p+(2/7)-3q/7]+a_2,$$
$$\Delta\nu_{\perp}=2a_1[-q-(1/7)+3q/14]+a_2/2,$$

where the p and q are certain coefficients appearing in Polder's[6] wave functions and are related to the g values by $g_{\shortparallel}=2(1-4p)$ and $g_{\perp}=2(1-q)$.

The results of the above equation can be used to determine the relative values of a_1 and a_2. For the g values we take $g_{\shortparallel}=2.44$ and $g_{\perp}=2.06$. Ingram gives $\Delta\nu_{\shortparallel}/\Delta\nu_{\perp}=g_{\shortparallel}\Delta H_{\shortparallel}/g_{\perp}\Delta H_{\perp}=8.3$. These numerical values lead to $a_1=0.88a_2$. The average separation of adjacent lines in manganese is 68 gauss which corresponds to an energy separation of $2\mu_B\Delta H=1.25\times10^{-18}$ erg. For a copper ion in a parallel field $\Delta\nu_{\shortparallel}=g_{\shortparallel}\mu_B\Delta H=a_1+a_2$. Assuming that a_2 is the same for both copper and manganese we obtain $\Delta H=118$ gauss, which gives an over-all separation of 354 gauss. This value is in good agreement with Ingram's value of 350 gauss.

After completing this investigation we have heard indirectly that J. M. Jauch has come to similar conclusions.

[1] Penrose, Abragam, and Pryce, Nature **163**, 992 (1949); L. J. F. Broer, Physica **15**, 673 (1949); C. Kikuchi and R. D. Spence, Am. J. Phys. **18**, 167 (1950).
[2] D. J. E. Ingram, Proc. Phys. Soc. London **A62**, 664 (1949).
[3] A. Abragam, Phys. Rev. **79**, 534 (1950).
[4] T. S. England and E. E. Schneider, Nature **166**, 437 (1950).
[5] H. A. Bethe, *Handbuch der Physik*, second edition Vol. 24/1, p. 386 (1933).
[6] D. Polder, Physica **9**, 709 (1942).

Spin-Spin Interaction within Paramagnetic Ions

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THAT the magnetic spin-spin interaction (W_{SS}, say) between pairs of electrons in an atom can cause deviations from the Landé interval rule is well known in spectroscopy. The effect is usually relatively small, although in helium it completely distorts the triplet terms. The purpose of this letter is to point out its possible role in the microwave resonance behavior of certain paramagnetic salts. The cases of interest occur where the splitting of the lowest spin multiplet of the paramagnetic ion, due to the combined action of the crystalline potential V and the spin-orbit interaction W_{LS}, either vanishes to second order in W_{LS}, or is very small. The splitting may then be dominated by W_{SS}.

In the salts of the iron transition group the ions Mn^{++} and Fe^{+++}, which are in a $3d^5$ ^6S state, furnish examples of interest. Their splitting has been studied by Van Vleck and Penney,[1] who consider various higher order processes involving V and W_{LS} through intermediate excited electronic states, using order of magnitude arguments to estimate the resulting splitting: but detailed calculation shows that their estimates are too high and that the mechanisms they propose are inadequate to explain the observed splittings. The important mechanisms would actually appear to be (a) a second-order process involving the first powers of W_{SS} and $V_2=H'(2z^2-x^2-y^2)$ via the intermediate state $3d^44s$ ^6D, giving a contribution DS_z^2 to the sub-Hamiltonian, and (b) a fifth-order process, quartic in W_{LS} and linear in $V_4=-G'(x^4+y^4+z^4+3y^2z^2-3z^2x^2-3x^2y^2)$, through intermediate states formed from $3d^5$, giving a contribution $(a/6)(S_x^4+S_y^4+S_z^4)$. Experimental evidence indicates that (a) dominates in manganous salts,[2] and (b) in ferric salts.[3] This changeover is not surprising, for (b) increases with a very high power of the effective nuclear charge of the ion.

TABLE I. Values of ρ from spectroscopic data.

Ion	State	ρ/cm^{-1}	Reference
Ti^{++}	$3d^2\,{}^3F$	0.24$\pm$0.03	a
V^{++}	$3d^3\,{}^4F$	0.4 $\pm$0.1	a
V^{+++}	$3d^2\,{}^3F$	0.24$\pm$0.05	a
Cr^{++}	$3d^4\,{}^5D$	0.42$\pm$0.05	b
Cr^{+++}	$3d^3\,{}^4F$	0.44$\pm$0.05	c
Mn^{+++}	$3d^4\,{}^5D$	0.8 $\pm$0.2	c
Fe^{++}	$3d^6\,{}^5D$	0.95$\pm$0.1	d
Co^{++}	$3d^7\,{}^4F$	1.50$\pm$0.05	e
Ni^{++}	$3d^8\,{}^3F$	5.31$\pm$0.03	e

a Atomic Energy Levels, Circular 467, National Bureau of Standards (1949).
b F. L. Moore, Princeton thesis (1950).
c I. S. Bowen, Phys. Rev. 52, 1153 (1937).
d B. Edlen and P. Swings, Astrophys. J. 95, 532 (1942).
e A. G. Shenstone, unpublished.

Detailed calculation shows that the coefficient D arising from (a) is given by

$$D=\frac{6}{25}\left(\frac{eh}{mc}\right)^2\frac{eH'}{\Delta E}(3w-2v)\int r^4\psi_d(r)\psi_s(r)dr,\qquad(1)$$

where

$$v=\int_0^\infty \frac{1}{r}\psi_d(r)\psi_s(r)\int_0^r r'^2\psi_d^2(r')dr'dr,$$

$$w=\int_0^\infty \frac{1}{r^3}\psi_d^2(r)\int_0^r r'^4\psi_d(r')\psi_s(r')dr'dr.$$

Here ψ_d and ψ_s are the radial wave functions of the $3d$ and $4s$ electrons, and ΔE is the energy of $3d^4 4s\,{}^6D$ above $3d^5\,{}^6S$. It is difficult to estimate the integrals, which depend strongly on the overlap of ψ_d and ψ_s, but reasonable assumptions give the right order of magnitude ($D\sim 0.02$ cm^{-1}).

In the other ions of the iron group W_{SS} has diagonal elements within the ground term, representable by a sub-Hamiltonian quadratic in $\mathbf{S}$, and which from arguments of rotational covariance must have the form

$$W_{SS}=-\rho\{\tfrac{1}{2}L_iL_j+\tfrac{1}{2}L_jL_i-\tfrac{1}{3}L(L+1)\delta_{ij}\}S_iS_j$$
$$=-\rho\{(\mathbf{L}\cdot\mathbf{S})^2+\tfrac{1}{2}(\mathbf{L}\cdot\mathbf{S})-\tfrac{1}{3}LS(L+1)(S+1)\}.\qquad(2)$$

One finds, for the lowest term,

$$\rho=\frac{1}{7S(2L-1)}\left(\frac{eh}{mc}\right)^2\left\{(4S-5)p+\frac{1}{7}(100-62S)q\right\},\qquad(3)$$

where

$$p=\int_0^\infty \frac{1}{r}\psi_d^2(r)\int_0^r r'^2\psi_d^2(r')dr'dr,$$

$$q=\int_0^\infty \frac{1}{r^3}\psi_d^2(r)\int_0^r r'^4\psi_d^2(r')dr'dr.$$

If hydrogen-like wave functions with effective nuclear charge Z' are used to calculate p and q, the result is

$$p=12q/7=(11/25920)(Z'me^2/\hbar^2)^3.$$

Second-order effects from W_{LS} via other terms of the $3d^n$ configuration will also give contributions of the form (2), which will add to (3), and in the second half of the transition series they may be larger than the W_{SS} contribution.

In chromous and manganic salts ($3d^4\,{}^5D$) the splitting of the spin quintet which is left lowest by the crystalline field is probably mainly determined[4] by W_{SS}. In chromic and vanadous salts ($3d^3\,{}^4F$), and nickel salts ($3d^8\,{}^3F$), the term in ρ, and also a mechanism analogous to (a), may contribute appreciably to the splitting, and previous estimates of the magnitude of V_2 from the splitting should be treated with reserve.

In some cases ρ can be inferred from the departures from the interval rule in spectroscopic data. These are listed in Table I.

I wish to thank Mr. Martin Redlich for help in the calculations of the coefficients in (1) and (3).

1 J. H. Van Vleck and W. G. Penney, Phil. Mag. 17, 961 (1934).
2 B. Bleaney and D. J. E. Ingram, Proc. Roy. Soc. A (to be published).
3 Weidner, Weiss, Whitmer, and Blosser, Phys. Rev. 76, 1727 (1949); D. Bijl, Leiden thesis, Excelsiors Foto Offset, s'Gravenhagen (1950).
4 A. Abragam and M. H. L. Pryce, Proc. Roy. Soc. A (to be published).

The New Isotope Pu^{242} and Additional Information on Other Plutonium Isotopes

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INVESTIGATION of the higher isotopes of plutonium produced by neutron irradiation has revealed the existence and properties of a new plutonium isotope, namely Pu^{242}, and has also yielded some additional information about the previously reported isotopes Pu^{240} and Pu^{241}. This letter will give only a brief report of these new results and a detailed description of the experiments will be deferred until a later date. The investigation of these isotopes was made possible by the intensive irradiation with neutrons of plutonium and americium samples originally consisting essentially completely of the isotopes Pu^{239} and Am^{241}, respectively. The production of Pu^{240} and Pu^{241} by the neutron irradiation of Pu^{239} has been reported previously.[1] Following the irradiation of the samples, which occurred over periods up to a few years, the plutonium, americium, and curium were chemically separated from each other and from fission products and impurities. The isotopic composition of each plutonium sample was then determined by use of a mass spectrograph which employed a thermal ionization source. Lines corresponding to plutonium isotopes of masses 241 and 242 produced by (n,γ) reactions were observed in addition to lines due to the other well-known plutonium isotopes (Pu^{238}, Pu^{239}, and Pu^{240}).

Pu^{240}.—Using very thin samples formed by volatilization from a hot filament, the width of the observed alpha-pulse analysis peak corresponding to the alpha-particles in a sample containing the two isotopes Pu^{239} and Pu^{240} was such that there is not more than about 20-kev difference in the alpha-particle energies of these two isotopes. Therefore, the alpha-particle energy[2] of Pu^{240} is 5.16$\pm$0.02 Mev.

Pu^{241}.—A low abundance group of alpha-particles at the energy 4.91$\pm$0.03 Mev was observed in neutron-bombarded plutonium using the 48-channel differential alpha-energy pulse analyzer. This group is present in the amount expected if it is due to the isotope Pu^{241}, i.e., best agreement with the alpha-decay systematics[3] is obtained if it is due to Pu^{241}, and it cannot be due to Pu^{242} in view of the results discussed below. To be a low energy alpha-group of the isotope, Pu^{240}, its abundance would apparently be greater by a factor[4] of 5 to 10. If this alpha-particle group is ascribed to Pu^{241}, its alpha-intensity and isotopic abundance lead to a partial half-life of Pu^{241} for alpha-particle decay of roughly 4×10^5 years in agreement with previous results.[5]

The half-life of Pu^{241} for beta-particle decay was estimated from the growth of the daughter Am^{241} using tracer Cm^{242} to determine chemical yield in the separations of the Am^{241} from the plutonium. The separations were made with measured amounts of plutonium in which the daughter had been allowed to grow over successively measured intervals of time. Using the mass spectrographically determined value for the isotopic abundance of Pu^{241} in the plutonium, and a value of 475 years for the half-life[6] of Am^{241}, the half-life of the Pu^{241} for beta-decay was calculated as 14 years, which is in rough agreement with the previous value[6] ($\sim$10 years).

A rough estimate of the cross section for the reaction Pu$^{241}(n,\gamma)$Pu^{242} with pile neutrons was obtained using an estimated value for the neutron flux and the isotopic compositions determined in the mass spectrograph. The cross section so obtained was very roughly 250 barns, but it is subject to large error owing to uncertainty in the estimation of the neutron flux.

Pu^{242}.—The isotope Pu^{242} was observed in a mass spectrographic analysis of neutron-bombarded plutonium, but its abundance was too small to make it possible to identify the radioactive decay properties in such samples. However, O'Kelley, Crane, Barton, and Perlman[7] have observed that the 16-hour Am^{242m} undergoes appreciable branching decay through the electron capture process. Advantage was taken of this by separating a plutonium fraction from a sample of americium (Am^{241}) which had been bombarded