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Magnetic Hyperfine Effects and Electronic Structure of NO*

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The theory of magnetic hyperfine structure in diatomic molecules is briefly reexamined. Previously reported hyperfine effects in the $N^{14}O^{16}$ molecule, both magnetic and electric quadrupole, are interpreted with corrected theoretical expressions in terms of an electronic structure consisting of 65 percent N=O and 35 percent $N^-=O^+$. The unpaired electron is shown to be essentially in a $2p\pi$ orbital but with 2.5 percent s character. Discrepancies of about 8 percent between experimental and calculated values of the magnetic hyperfine constants indicate limitations in the use of atomic wave function approximations for electronic orbitals.

THE NO molecule possesses an unpaired electron and has a $^2\Pi$ ground electronic state. Hyperfine structure due to the magnetic moment and electric quadrupole moment of N^{14} has been observed in both the $\Pi_{\frac{3}{2}}$ and $\Pi_{\frac{1}{2}}$ states.[1–3] This hyperfine structure is discussed and interpreted in terms of molecular constants which give some detailed information about the molecular electronic structure.

THEORY OF MAGNETIC HYPERFINE STRUCTURE AND EXPERIMENTAL RESULTS

The theory of magnetic hyperfine structure in diatomic molecules has been given by Frosch and Foley.[4] They derive the hyperfine interaction from the Dirac equation for the electron. An alternative and simplified derivation of the Hamiltonian is briefly outlined here. The electron spin-nuclear spin part of the interaction, for other than s-states, can be written as the interaction between two point dipoles in the form

$$3(\mathbf{I}\cdot\mathbf{r})(\mathbf{S}\cdot\mathbf{r})/r^5-(\mathbf{I}\cdot\mathbf{S})/r^3, \tag{1}$$

where $\mathbf{r}$ denotes the radius vector from the nucleus to the interacting electron. A coordinate system x', y', z' is taken, fixed in the molecule, with the z' axis coinciding with the internuclear axis. The angle between the internuclear axis and the radius vector from the nucleus to electron is denoted by χ. In cylindrical coordinates, $x'=\rho\cos\varphi$, $y=\rho\sin\varphi$, where $\rho=r\sin\chi$ (the coordinate system and the angle definitions are identical with those of Frosch and Foley). Then expression (1) gives the spin-spin part of the interaction, and on addition of the $\mathbf{I}\cdot\mathbf{L}$ term the total Hamiltonian is obtained as

$$\begin{aligned}H=a\mathbf{I}\cdot\mathbf{L}&+(b+c)I_{z'}S_{z'}+\tfrac{1}{2}b(I^+S^-+I^-S^+)\\&+\tfrac{1}{2}d(e^{2i\varphi}I^-S^-+e^{-2i\varphi}I^+S^+)+e[e^{i\varphi}(S^-I_{z'}+I^-S_{z'})\\&+e^{-i\varphi}(S^+I_{z'}+I^+S_{z'})],\end{aligned} \tag{2}$$

where

$$I^+=I_{x'}+iI_{y'},\qquad I^-=I_{x'}-iI_{y'},\ \text{etc.},$$
$$a=2g_I\mu_0\mu_N(1/r^3)_{\mathrm{Av}},\qquad c=3g_I\mu_0\mu_N[(3\cos^2\chi-1)/r^3]_{\mathrm{Av}},$$
$$d=3g_I\mu_0\mu_N(\sin^2\chi/r^3)_{\mathrm{Av}},\qquad e=3g_I\mu_0\mu_N(\sin\chi\cos\chi/r^3)_{\mathrm{Av}}.$$

μ_0 and μ_N are the values of the Bohr and nuclear magnetons taken as positive; b for non-s-states is $-C/3$. It is to be noted that the term in e above has $\frac{1}{2}$ of the value given by Frosch and Foley.

The interaction Hamiltonian (2) does not apply to states with an electronic charge density inside the nucleus, i.e., states where ψ at the position of the nucleus has a finite value. For such states (2) would give zero hyperfine energy, except for second-order effects. A term has to be added to (2) to account for the first-order hyperfine energy due to the relativistic part of the spin-spin interaction, which is characteristic of atomic s-states. This interaction has been given for the atomic case[5] as

$$(16\pi/3)g_I\mu_0\mu_N\psi^2(0)\mathbf{I}\cdot\mathbf{S}. \tag{3}$$

This term may be considered to come from the electron spin distribution inside the nucleus. Although the coefficient of $\mathbf{I}\cdot\mathbf{S}$ in (3) for the molecular case should be the same as in the atomic case, the evaluation of $\mathbf{I}\cdot\mathbf{S}$, of course, has to take into account the molecular coupling scheme. The term (3), then, is added to (2) and, since the b term of (2) is also proportional to $\mathbf{I}\cdot\mathbf{S}$, b can be redefined to include both the $\psi^2(0)$ and $[(3\cos^2\chi-1)/r^3]$ terms. This is the meaning of b that will be used, i.e.,

$$b=-g_I\mu_0\mu_N\left(\frac{3\cos^2\chi-1}{r^3}\right)_{\mathrm{Av}}+\frac{16\pi}{3}g_I\mu_0\mu_N\psi^2(0). \tag{4}$$

The terms in (2) with matrix elements diagonal in Λ, Σ, and J, which give the same energy contribution to each member of the Λ doublet, are $aI_{z'}L_{z'}+(b+c)I_{z'}S_{z'}$. The operator $d(e^{2i\varphi}I^-S^-+e^{-2i\varphi}I^+S^+)/2$ has matrix elements connecting the degenerate states $(-\Lambda-\Sigma)$ and $(+\Lambda+\Sigma)$ for specific values of Λ and Σ and gives equal and opposite contributions to each member of the

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[1] R. Beringer and J. G. Castle, Jr., Phys. Rev. **78**, 581 (1950); Beringer, Rawson, and Henry, Phys. Rev. **94**, 343 (1954).

[2] W. Gordy and C. A. Burrus, Phys. Rev. **93**, 419 (1954).

[3] Gallagher, Bedard, and Johnson, Phys. Rev. **93**, 729 (1954).

[4] R. A. Frosch and H. M. Foley, Phys. Rev. **88**, 1347 (1952).

[5] E. Fermi, Z. Physik **60**, 320 (1930); G. Breit and F. W. Doerman, Phys. Rev. **36**, 1732 (1930).

Λ doublet.[4,6,7] This effect, which results from the interaction of a spin on the symmetry axis with spins off the axis, has also been observed in the microwave spectra of OH[8] and ammonia.[9]

The hyperfine energies, to first order, are obtained from (2) using the relation

$$(\alpha J|I^i|\alpha' J)=(\alpha J|J^i|\alpha' J)\mathbf{I}\cdot\mathbf{J}/J(J+1)$$

for matrix elements diagonal in J. For the $\Pi_{\frac{1}{2}}$ state:

$$W=\tfrac{1}{2}\left(a-\frac{b+c}{2}\right)\frac{\mathbf{I}\cdot\mathbf{J}}{J(J+1)}\pm\frac{d(J+\frac{1}{2})\mathbf{I}\cdot\mathbf{J}}{2J(J+1)},\qquad(5)$$

and for the $\Pi_{\frac{3}{2}}$ state:

$$W=\tfrac{3}{2}\left(a+\frac{b+c}{2}\right)\frac{\mathbf{I}\cdot\mathbf{J}}{J(J+1)}.\qquad(6)$$

Expression (5) is identical in form with the one given by Frosch and Foley, except that the term in $\pm d$ is twice as large here.[10] The operator multiplying d in (2) has no matrix elements between states $(\Lambda+\Sigma=\Omega=\frac{3}{2})$ and $(-\Lambda-\Sigma=\Omega=-\frac{3}{2})$, hence this term does not contribute to the hyperfine energy for the $\Pi_{\frac{3}{2}}$ state. Expressions (5) and (6) hold for pure Hund's case (*a*).

The rotational energy will mix the $\Pi_{\frac{1}{2}}$ and $\Pi_{\frac{3}{2}}$ states and the correct wave function can be expressed as a linear combination of the two state functions [case intermediate between (*a*) and (*b*)]. Hence, the hyperfine energy will be intermediate between expressions (5) and (6). The amplitudes of the $\Pi_{\frac{1}{2}}$ and $\Pi_{\frac{3}{2}}$ components for the intermediate case depend strongly on J as well as the ratio of the fine structure constant A to the rotational constant B. The hyperfine structure will also be modified by the term $\frac{1}{2}b(I^+S^-+I^-S^+)$ which connects the $\Pi_{\frac{1}{2}}$ and $\Pi_{\frac{3}{2}}$ states.[11] The term in $\pm d$ will be particuarly influenced by the extent of intermediate coupling, since it is completely missing in the $\Pi_{\frac{3}{2}}$ state. Such a dependence of the hyperfine structure on the extent of intermediate coupling has been observed in the microwave spectra of OH and OD. Formulas for the hyperfine structure in the intermediate case will be given in reference 8.

From the observed[2,3] $J=\frac{1}{2}\rightarrow\frac{3}{2}$ transition in the $\Pi_{\frac{1}{2}}$ state of NO, where (5) applies (with the $+$ sign in front of d for the upper member of the Λ doublet, and the $-$ sign for the lower),

$$a-\tfrac{1}{2}(b+c)=92.6\ \text{Mc/sec},$$
$$d=3g_I\mu_0\mu_N(\sin^2\chi/r^3)_{\text{Av}}=112.6\ \text{Mc/sec}.$$

Beringer *et al.*[1] have observed transitions between Zeeman components of the $\Pi_{\frac{3}{2}}$ state in large magnetic fields, which allows them to determine the constant

$$a+(b+c)/2=74.1\ \text{Mc/sec}.$$

From above, using the relation $c=3(a-d)$, we obtain $a=83.4$ Mc/sec, $b=69.2$ Mc/sec, $c=-87.6$ Mc/sec.

INTERPRETATION OF HYPERFINE COUPLING CONSTANTS IN TERMS OF THE STRUCTURE OF NO

The constants determined above from the experimental data can be used to evaluate parameters of the distribution of electronic angular momentum in the molecule. These parameters may be obtained from the experimental constants and the definitions given in (2) and (4) above. Thus one obtains

$$(1/r^3)_{\text{Av}}=14.9\times10^{24}\ \text{cm}^{-3},$$
$$(\sin^2\chi/r^3)_{\text{Av}}=13.4\times10^{24}\ \text{cm}^{-3},$$
$$[(3\cos^2\chi-1)/r^3]_{\text{Av}}=-10.4\times10^{24}\ \text{cm}^{-3},$$
$$(16\pi/3)g_I\mu_0\mu_N\psi^2(0)=39.9\ \text{Mc/sec},$$

which gives

$$\psi^2(0)=0.85\times10^{24}\ \text{cm}^{-3}.$$

These several experimental parameters, as well as the observed quadrupole interaction, give considerable information about electronic wave functions in the molecule, and can be used as critical tests of proposed schemes for the electronic structure of NO.

Pauling[12] has proposed for the electronic structure of NO an equal mixture of the structures $:\dot{\mathrm{N}}=\ddot{\mathrm{O}}:$ and $:\ddot{\mathrm{N}}^-=\dot{\mathrm{O}}:^+$. This may be interpreted as a double bond and a three-electron bond $\ddot{\mathrm{N}}\overset{\cdots}{=}\ddot{\mathrm{O}}$.

The double bond consists of the $2p\ \pi$ and $2p\ \sigma$ electron orbitals. The three-electron bond involves two overlapping $2p\ \pi$ orbitals, one of which is occupied by two electrons with paired spins, whereas the other is occupied by the unpaired electron. According to this structure the unpaired electron spends half of the time in the $2p$ state on the oxygen atom and the other half on the nitrogen.

The contribution to the hyperfine energy from a $2p\ \pi$ electron on the oxygen is small, since the distance between the electron and the interacting nucleus is of the order of the internuclear distance (1.1539 A). Calculated values of $(1/r^3)_{\text{Av}}$ and $(\sin^2\chi/r^3)_{\text{Av}}$ in this case are $0.49\times10^{24}\ \text{cm}^{-3}$ and $0.08\times10^{24}\ \text{cm}^{-3}$, respectively, compared

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[6] Masataka Mizushima, Phys. Rev. **94**, 569 (1954).

[7] C. H. Townes and A. L. Schawlow, *Microwave Spectroscopy* (McGraw Hill Book Company, Inc., New York, to be published), Chap. VIII.

[8] Sanders, Schawlow, Dousmanis, and Townes, Phys. Rev. **89**, 1158 (1953); **94**, 798(A) (1954); and Phys. Rev. (to be published).

[9] Gunther-Mohr, Townes, and Van Vleck, Phys. Rev. **94**, 1191 (1954).

[10] Frosch and Foley have kindly reexamined their result and find that the value of d in their Eq. (6.5) should be corrected to $3g_I\mu_0\mu_N(\sin^2\chi/r^3)_{\text{Av}}$, in agreement with the result here.

[11] In NO the effects of this term have been considered (reference 6); the correction is small, since this is a good case (*a*) molecule for low values of J.

[12] Linus Pauling, *The Nature of the Chemical Bond* (Cornell University Press, Ithaca, 1945).

with 14.9×10^{24} and 13.4×10^{24} obtained from the experimental data. Hence, most of the contribution to the hyperfine energy comes from the unpaired electron on the nitrogen and, in order to compare the experimental values of the constants with calculated ones, a rather accurate value of $(1/r^3)_{\mathrm{Av}}$ in a $2p$ state in the N atom is required.

The value 25.7×10^{24} cm^{-3} has been given by Townes and Dailey,[13] whereas Barnes and Smith[14] obtain 16.6×10^{24}. Both values are based on the fine-structure data, but different values of $Z_{\mathrm{effective}}$ have been used in the two cases. Because of this difference $(1/r^3)_{\mathrm{Av}}$ has been calculated from the radial wave functions given by Hartree and Hartree.[15] The result obtained for the ground state of $\mathrm{N}[(2p)^3\,{}^4S]$ is

$$(1/r^3)_{\mathrm{Av}}=20.8\times10^{24}\ \mathrm{cm}^{-3}.$$

This value is not expected to be extremely accurate, since the wave functions obtained by the Hartree-Fock method are not sufficiently accurate near the nucleus.[16] In the oxygen atom the value obtained from the Hartree wave functions has been compared with a much more accurate value derived from the fine structure data.[16,17] In this case the calculation based on the Hartree-Fock wave functions under estimates $(1/r^3)_{\mathrm{Av}}$ by 8 percent. Applying such a correction to the value calculated above for N, we obtain as probably the best "theoretical" value for a $2p$ electron in N:

$$(1/r^3)_{\mathrm{Av}}=22.5\times10^{24}\ \mathrm{cm}^{-3}.$$

This is intermediate between the two previous values.[13,14]

For a $2p$ electron on the nitrogen atom,

$$[(3\cos^2\chi-1)/r^3]_{\mathrm{Av}}=(1/r^3)_{\mathrm{Av}}(3\cos^2\chi-1)_{\mathrm{Av}};$$

and for a $2p\ \pi$ electron ($m_l=\pm1$),

$$(3\cos^2\chi-1)_{\mathrm{Av}}=-2l/(2l+3)=-\tfrac{2}{5},\quad (\sin^2\chi)_{\mathrm{Av}}=\tfrac{4}{5}.$$

From these and the last values of $(1/r^3)_{\mathrm{Av}}$, the hyperfine structure parameters are obtained as $(1/r^3)_{\mathrm{Av}}=22.5\times10^{24}$ cm^{-3} and $(\sin^2\chi/r^3)_{\mathrm{Av}}=18.0$. This set of values and the other set given above (when the electron was assumed to be on the oxygen) are compared in Table I with the values obtained from the experimental data. The experimental values are intermediate between the values of the two sets. (See Table I.) From the experimental value of $(1/r^3)_{\mathrm{Av}}$ the probabilities of the electron being on the nitrogen and oxygen atoms are 0.65 and 0.35, respectively. A structure consistent with these values, then, would be 65 percent N=O and 35 percent $\mathrm{N^-{=}O^+}$.

TABLE I. Parameters describing the unpaired electron distribution in NO. In Column *A* are calculated values for an electron in a $2p\ \pi$ state on the oxygen atom. In *B*: calculated values for the electron in a $2p\ \pi$ state on the nitrogen atom. In Column *C* it is assumed that the probabilities are 0.65 and 0.35 for the electron to be on the N and O atoms, respectively. All values are in units of 10^{24} cm^{-3}.

Parameter	*A*	*B*	*C*	Experimental value
$(1/r^3)_{\mathrm{Av}}$	0.49	22.5	14.9	14.9
$(\sin^2\chi/r^3)_{\mathrm{Av}}$	0.08	18.0	11.7	13.4

From this structure the quadrupole coupling constant is calculated by the method of Townes and Dailey.[13] We make two calculations of *eqQ*. The first, according to scheme (*a*) of Townes and Dailey, is based on *eqQ* per *p* electron =24 Mc/sec, 25 percent $s-p$ hybridization of the σ component of the multiple bond, and a quadrupole moment $=0.033\times10^{-24}$ cm^2. It gives an $eqQ=-2.2$ Mc/sec in agreement with the experimental -2.17 Mc/sec.[1,6] The second calculation, according to scheme (*b*), with an *eqQ* per *p* electron =10 Mc/sec, 50 percent hybridization, and a $Q=0.012$, predicts an $eqQ-3.3$ Mc/sec, which is not far from the experimental result. Hence the average value for $Q=0.02\times10^{-24}$ cm^2 assigned by Townes and Dailey to N^{14} is consistent with the observed quadrupole interaction in NO. Mizushima's conclusion,[6] that the NO data favor a smaller value for $Q=0.01$, seems to be based on a value of $(1/r^3)_{\mathrm{Av}}$ for the unpaired electron which is about twice as large as the one obtained here.

As pointed out by Pauling, some amount of $\mathrm{N^+{-}O^-}$ may be expected to be present in the structure. An admixture of $\mathrm{N^+{-}O^-}$ would be consistent with the magnetic hyperfine structure results, provided the total amount of N=O and $\mathrm{N^+{-}O^-}$ (both of which have the unpaired electron on the nitrogen) adds up to 65 percent. Taking, then, as an alternative structure 60 percent N=O 35 percent $\mathrm{N^-{=}O^+}$ and 5 percent $\mathrm{N^+{-}O^-}$, we calculate *eqQ*'s$=-2.6$ and -3.6 according to scheme (*a*) and (*b*), respectively. These values are somewhat larger than the experimental value -2.17 Mc/sec. However, in view of uncertainties as to the extent of hybridization such an admixture of as much as 5 percent of $\mathrm{N^+{-}O^-}$, and possibly some N—O, is not excluded.*

From the above, the wave function for the unpaired electron which gives the observed value of $(1/r^3)_{\mathrm{Av}}$ and of the quadrupole coupling may be approximated as

$$\psi=(0.65)^{\frac{1}{2}}\psi_{2p\pi}(N)+(0.35)^{\frac{1}{2}}\psi_{2p\pi}(\mathrm{O}). \tag{7}$$

Effects of overlapping are neglected.

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[13] C. H. Townes and B. P. Dailey, J. Chem. Phys. **17**, 782 (1949).

[14] R. G. Barnes and W. V. Smith, Phys. Rev. **93**, 95 (1954).

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* *Note added in proof.*—Lin and Van Vleck obtain a new value for $eqQ=-1.6\pm0.3$ Mc/sec (instead of the -2.12 of reference 6) from the $\Pi_{\frac{1}{2}}$ data. The value for the $\Pi_{\frac{3}{2}}$ state is -2.22 mc (reference 1). Hence one should compare our calculated values with an average experimental value of about -1.9 instead of the -2.17 that we used. This slight change does not affect the present conclusions on the structure.

These authors also obtain value for the magnetic constants in NO which are in agreement with the ones given here. (C. C. Lin and J. H. Van Vleck, private communication).

A further check on the wave function is allowed by the experimentally measured parameter $[\sin^2\chi/r^3]_{\text{Av}}$. From (7), one calculates a value $[\sin^2\chi/r^3]_{\text{Av}}=11.7\times10^{24}$ cm^{-3} (see column C, Table I), which is 13 percent lower than the experimental value. Another expression of the same results can be obtained by calculating $(\sin^2\chi)_{\text{Av}}$ under the assumption of an atomic p orbital as in (7). The result is for a $p\pi(m_l=\pm1)$ orbital, $(\sin^2\chi)_{\text{Av}}=0.8$ and for a $p\sigma(m_l=0)$ orbital $(\sin^2\chi)_{\text{Av}}=0.40$. The value obtained from experimental values of $(1/r^3)_{\text{Av}}$ and $(\sin^2\chi/r^3)_{\text{Av}}$, assuming r and χ are independent, is $\sin^2\chi=0.90$. This is in fair agreement with the value expected from a $p\,\pi$ orbit, but is about 13 percent higher. This indicates that the $p\,\pi$ orbit is somewhat "compressed" in a plane perpendicular to the internuclear axis. Part of this discrepancy can be accounted for by effects of overlapping which have been omitted from the calculation. A similar discrepancy partly due to overlapping effects has been reported by Miller, Townes, and Kotani in the case of molecular oxygen.[17] If a similar amount of overlapping is assumed for NO, it would account for about 5 percent of the discrepancy. This leaves an unaccounted 8 percent discrepancy, which could indicate a deviation of the molecular wave function from the pure atomic $p\,\pi$ type.

The value of $\psi^2(0)=0.85\times10^{24}$ cm^{-3} obtained from the data shows that some s character must be attributed to the unpaired electron, which is primarily in a $2p\,\pi$ state. An estimate of $\psi^2(0)$ for a $2s$ state in the nitrogen atom is 34×10^{24} cm^{-3}. Hence the molecular wave function possesses $0.85/34=2.5$ percent s character. Neglect of electrostatic interactions between the electrons in an atom can cause a mixing of configurations. In the case of O^+ as much as 4 percent of configuration mixing has been calculated.[18] Hence the 2.5 percent of s character found in the molecular wave function in NO is perhaps not surprising. Although this amount of s state is small, its contribution to the magnetic hyperfine energy is comparable to that of the classical dipole-dipole part of the interaction. For instance, this relativistic term contributes 40 Mc/sec to $(b+c)$, whereas the classical part contributes -58 Mc/sec.

CONCLUSION

There is fair agreement between the observed hyperfine structure constants a, b, c, d, eqQ, and those expected from a relatively simple model of the electronic structure in NO. There are, however, substantial disagreements between the experimentally determined values and those calculated from simple molecular wave functions. Part of the discrepancy can be accounted for by neglected effects of overlapping. A remaining discrepancy of about 8 percent in the hyperfine coupling constants may give interesting information on the deviation of the molecular wave functions from atomic orbital approximations.

ACKNOWLEDGMENT

I wish to express my gratitude to Professor Charles H. Townes for his valuable advice, many stimulating discussions on the subject of this paper, and for reading and commenting on the manuscript.

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Sum Rules for Photodisintegration of the Deuteron*

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The experimental total cross section for photodisintegration of the deuteron is integrated numerically to 155 Mev for three moments of the curve weighting with W^{-1}, 1 and W, respectively: the bremsstrahlung-weighted cross section, the integrated cross section, and $\bar{W}\sigma_{\text{int}}$. The integrated cross section is compared with the phenomenological sum-rule calculation, and also with the dispersion-theoretic calculation; all three values are in reasonable agreement, giving an integrated cross section about 30 percent higher than that of 30 Mev-mb for the Thomas-Reiche-Kuhn dipole sum rule. There is good agreement between the experimental value of 3.9 mb and the phenomenological calculation of 3.78 mb for the bremsstrahlung-weighted cross section.

INTRODUCTION

RECENT experiments at the University of Illinois[1] and California Institute of Technology[2] have filled in and extended our knowledge of the total cross section for photodisintegration of the deuteron. Combining these results with earlier measurements,[3–6] we

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Note added in proof.—Recalibration of the ThC″ sources gives a 6 percent smaller photodisintegration cross section. [Marin, Bishop, and Halban, Proc. Phys. Soc. (London) A67, 1113 (1954).]

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