energy goes into lattice distortion. It must be remembered that the actual lattice expansion will be less because of the opposing tendency toward contraction due to the electrostatic attraction considered above.

We conclude that the contribution of lattice distortion to the energy of diffusion in the diamond lattice should be negligible (of the order of a few percent), except for much larger impurities than the ones considered here.

Treatment of Ionized Impurity Scattering in Degenerate Semiconductors. Application of the Variational Technique in the Partial-Wave Method

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Using the partial-wave method, ionized impurity scattering in degenerate semiconductors is discussed. It is shown that the scattering process is adequately described by considering only the zeroth-order phase shift. By making use of Kohn's variational principle this quantity is determined analytically and an analytical formula is derived for the resistivity due to ionized impurity scattering alone.

I. INTRODUCTION

IONIZED impurity scattering is an important factor in determining the charge transport in semiconductors. Theoretical treatments[1] of the scattering process make use either of the Born approximation[2] or of the more exact partial-wave method.[3] The latter approach has been used by Blatt[4] for discussing ionized impurity scattering in nondegenerate semiconductors. Unfortunately, the form of his scattering potential did not permit the derivation of analytical formulas for the phase shifts of the partial waves.

The present paper considers ionized impurity scattering in degenerate semiconductors. It is shown that in this case the scattering process is adequately described by considering only the zeroth-order phase shift. By making use of Kohn's variational principle[5] this quantity is determined analytically. As an example, the scattering of electrons by donor ions is discussed and an analytical formula is derived for the resistivity due to ionized impurity scattering alone. The scattering potential used is of the same form as that of Blatt,[4] namely a screened Coulombic potential.[6] It is assumed that in an elastic scattering act only one donor ion participates. As in Blatt's work, anisotropies in the electronic effective mass are ignored and a scalar effective mass is used.

II. CALCULATIONS

1. Calculation of the Total Scattering Cross Section

In the partial-wave method[3] the total scattering cross section for elastic scattering of a particle by spherically symmetric potentials is given by

$$Q' = 2\pi \int_0^\pi \sigma(\vartheta) \sin\vartheta d\vartheta, \tag{1}$$

where

$$\sigma(\vartheta) = \frac{1}{k^2} \left| \sum_{l=0}^\infty (2l+1) e^{i\eta_l} \sin\eta_l P_l(\cos\vartheta) \right|^2 \tag{2}$$

is the differential scattering cross section. If a weighting factor is introduced for large-angle scattering, then Eq. (1) becomes,[7]

$$Q = 2\pi \int_0^\pi (1-\cos\vartheta) \sigma(\vartheta) \sin\vartheta d\vartheta. \tag{3}$$

Using Eq. (2) one finds from Eq. (3) that

$$Q = \sum_{l=0}^\infty Q_l = \frac{4\pi}{k^2} \sum_{l=0}^\infty (l+1) \sin^2(\eta_l - \eta_{l+1}), \tag{4}$$

where η_l is the phase shift of the lth partial wave and k is the wave number of the scattered particle. Assuming a radius R beyond which the scattering potential is "negligible," the summation for practical computations extends[3] from $l=0$ to a maximum l of the order of kR. In what follows it is assumed that the electron's energy is such that $kR \lesssim 1$. In this case Eq. (4) reduces to

$$Q_0 = (4\pi/k^2) \sin^2(\eta_0 - \eta_1). \tag{5}$$

[1] The subject is reviewed by F. J. Blatt, *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1957), Vol. 4, p. 343 ff; H. Brooks, *Advances in Electronics and Electron Physics*, edited by L. Marton (Academic Press Inc., New York, 1955), Vol. VII, p. 156 ff; W. Shockley, *Electrons and Holes in Semiconductors* (D. Van Nostrand Company, Inc., Princeton, New Jersey, 1950), p. 258 ff.

[2] L. I. Schiff, *Quantum Mechanics* (McGraw-Hill Book Company, Inc., New York, 1955), 2nd ed., p. 161 ff.

[3] See reference 2, p. 103 ff.

[4] F. J. Blatt, J. Phys. Chem. Solids **1**, 262 (1957).

[5] W. Kohn, Phys. Rev. **74**, 1763 (1948).

[6] R. B. Dingle, Phil. Mag. **46**, 831 (1955).

[7] K. Huang, Proc. Phys. Soc. (London) **60**, 161 (1948).

The physical conditions under which a degenerate semiconductor meets the above requirement will be examined later.

A variational principle, formulated by Kohn,[5] states that η_0 can be determined by minimizing the quantity

$$L(c_1,c_2,\cdots,a)=\int \psi_t(H-E)\psi_t{}^*d\tau, \qquad (6)$$

where $\psi_t(c_1,c_2,\cdots,a)$ is a suitably chosen trial wave function of the impinging electron; c_1, c_2, $\cdots$, a are variational parameters; H is the Hamiltonian and E is the energy of the system of an electron and a donor ion. If one minimizes Eq. (6) with the aid of the relations[8]

$$\partial L/\partial a = 4\pi k,$$
$$\partial L/\partial c_r = 0, \quad r=1, 2, \cdots, \qquad (7)$$

then the zeroth-order partial-wave shift is given by

$$\eta_0 = \arc \tan[\bar{a}-\bar{L}/4\pi k], \qquad (8)$$

where $\bar{L}=L(\bar{c}_1,\bar{c}_2,\cdots,\bar{a})$ and the bars on the variational parameters denote values which make L a minimum.

For electrons being scattered on screened donor ions the Hamiltonian, in atomic units,[9] is given by

$$H = -(1/2\gamma)\nabla^2 - (1/\kappa r)e^{-r/R}, \qquad (9)$$

where $\gamma=m^*/m_0$ is the ratio of the effective electronic mass to the real electronic mass, κ is the static dielectric constant of the semiconductor, and R is a screening length. The problem of the screening of donor ions by electrons has been investigated by Dingle,[6] who, for degenerate semiconductors gives the following expression for the screening length

$$R=\left(\frac{\pi}{3}\right)^{1/6}\frac{h\kappa^{\frac{1}{2}}}{4\pi e_0 m_0}\gamma^{-1}n^{-1/6}. \qquad (10)$$

It is seen from Eq. (10) that R is a function of the electron concentration n only.

The choice of a suitable trial wave function is based on the following considerations. Massey and Moiseiwitsch,[8] in discussing the problem of the scattering of slow electrons by hydrogen atoms, obtained very good agreement between the numerically computed exact zeroth-order phase shift and the approximate one that they calculated by assuming a trial wave function for the free electron. In atomic units, the trial wave function they used is of the form

$$\psi_t(a,b) = X/r = [\sin kr + (a+be^{-r})(1-e^{-r})\cos kr]/r, \quad (11)$$

where a and b are variational parameters. Since, in their case, the Hamiltonian is known exactly and since in the wave function for the system of a hydrogen atom and an

electron the only unknown component is the free-electron wave function, the good agreement might be looked upon as evidence that the form of the trial wave function for the free electron is well chosen. It is therefore assumed that the same wave function also will be well suited for the study of slow electrons scattered on donor ions.

With the aid of Eqs. (9) and (11) one can rewrite Eq. (6) as

$$L(a,b) = -\frac{2\pi}{\gamma}\left\{\int_0^\infty XX''dr + k^2\int_0^\infty X^2dr \right.$$
$$\left. +\frac{2\gamma}{\kappa}\int_0^\infty \frac{X^2e^{-\alpha r}}{r}dr\right\}, \quad (12)$$

where the primes on X denote differentiation with respect to r and α stands for $1/R$. Performing the integrations in Eq. (12) one obtains

$$L = -(2\pi/\gamma)W, \qquad (13)$$
where

$$W = a^2F_1 + b^2F_2 + aF_3 + bF_4 + abF_5 + F_6. \qquad (14)$$

The F's in Eq. (14) can be expressed as

$$F_1 = (2\gamma/\kappa)G_{31} - kI_1 + kI_2 - J_1 + J_2,$$
$$F_2 = (2\gamma/\kappa)G_{33} + kI_2 - 3kI_3 + 2kI_4 + J_2 - 5J_3 + 4J_4,$$
$$F_3 = (2\gamma/\kappa)(M_0 - M_1) - \tfrac{1}{2}I_1 - 2kK_1,$$
$$F_4 = (2\gamma/\kappa)(M_1 - M_2) + \tfrac{1}{2}I_1 - 2I_2 + 2kK_1 - 4kK_2, \quad (15)$$
$$F_5 = (4\gamma/\kappa)G_{32} + kI_1 - 4kI_2 + 3kI_3 + J_1 - 6J_2 + 5J_3,$$
$$F_6 = (2\gamma/\kappa)G_1,$$

where the quantities on the right-hand side have the following meaning:

$$G_{31} = \int_0^\infty \frac{e^{-\alpha r}}{r}(1-e^{-r})^2\cos^2 kr\,dr$$
$$= \tfrac{1}{4}\ln\frac{(\alpha+1)^4[(\alpha+1)^2+4k^2]^2}{\alpha^2(\alpha^2+4k^2)(\alpha+2)^2[(\alpha+2)^2+4k^2]}, \quad (16)$$

$$G_{32} = \int_0^\infty \frac{e^{-\alpha r}}{r}e^{-r}(1-e^{-r})^2\cos^2 kr\,dr$$
$$= \tfrac{1}{4}\ln\frac{(\alpha+2)^4[(\alpha+2)^2+4k^2]^2}{(\alpha+1)^2[(\alpha+1)^2+4k^2](\alpha+3)^2[(\alpha+3)^2+4k^2]}, \quad (17)$$

$$G_{33} = \int_0^\infty \frac{e^{-\alpha r}}{r}e^{-2r}(1-e^{-r})^2\cos^2 kr\,dr$$
$$= \tfrac{1}{4}\ln\frac{(\alpha+3)^4[(\alpha+3)^2+4k^2]^2}{(\alpha+2)^2[(\alpha+2)^2+4k^2](\alpha+4)^2[(\alpha+4)^2+4k^2]}, \quad (18)$$

[8] H. S. W. Massey and B. L. Moiseiwitsch, Proc. Roy. Soc. (London) A205, 483 (1951). This paper also reviews various variational approaches for the determination of the phase shift.
[9] $e_0{}^2/a_0 = 27.2$ ev, $a_0 = 0.529$ Å.

TABLE I. Calculated values of the resistivity and several related quantities arising from ionized impurity scattering in degenerate n-type germanium. (An isotropic effective mass of $m^*=0.25m_0$ and a static dielectric constant of $\kappa=16$ was used.)

n (in cm^3)	ρ (in ohm-cm)	k_F (in cm^{-1})	$Q_0{}^F$ (in cm^2)	$\eta_0{}^F$ (in radians)	R (in cm)	$k_F R$
10^{18}	1.14×10^{-2}	3.10×10^6	8.90×10^{-13}	0.969	29.4×10^{-8}	0.911
5×10^{18}	5.23×10^{-3}	5.31×10^6	2.41×10^{-13}	0.830	22.4×10^{-8}	1.19

$$G_1=\int_0^\infty \frac{e^{-\alpha r}}{r}\sin^2 kr\,dr=\tfrac{1}{4}\ln\frac{\alpha^2+4k^2}{\alpha^2}, \tag{19}$$

and

$$I_n=\int_0^\infty e^{-nr}\sin 2kr\,dr=\frac{2k}{n^2+4k^2}, \quad n=1,2,3,4$$

$$J_n=\int_0^\infty e^{-nr}\cos^2 kr\,dr=\frac{1}{n^2+4k^2}\left(n+\frac{2k^2}{n}\right), \quad n=1,2,3,4$$

$$K_n=\int_0^\infty e^{-nr}\sin^2 kr\,dr=\frac{1}{n}J_n, \quad n=1,2$$

$$M_n=\int_0^\infty \frac{e^{-(\alpha+n)r}}{r}\sin 2kr\,dr=\arctan\frac{2k}{\alpha+n}, \quad n=0,1,2.$$

The evaluation of the integrals in Eqs. (16) to (19) is somewhat involved and will be discussed in Appendix I.

With the aid of Eq. (7), one finds from Eq. (13) that the values of the variational parameters which minimize L are

$$\bar{a}=(F_4F_5-2F_2F_3-4k\gamma F_2)/(4F_1F_2-F_5{}^2),$$
$$\bar{b}=(F_3F_5-2F_1F_4+2k\gamma F_5)/(4F_1F_2-F_5{}^2). \tag{20}$$

Substituting η_0 from Eq. (8) into Eq. (5), the total scattering cross section is given by

$$Q_0=(4\pi/k^2)\sin^2[\arctan(\bar{a}-\bar{L}/4\pi k)-\eta_1]. \tag{21}$$

If one assumes now that $|\eta_1|$ is negligibly small compared to $|\eta_0|$, which, as will be shown later, is actually the case, then one finds from Eq. (21) that

$$Q_0=\frac{4\pi}{k^2}\frac{(\bar{a}-\bar{L}/4\pi k)^2}{1+(\bar{a}-\bar{L}/4\pi k)^2}, \tag{22}$$

where $\bar{L}=L(\bar{a},\bar{b})$ is given by Eqs. (13) and (20). It is mentioned that as a consequence of using atomic units, the total scattering cross section in Eq. (22) is measured in units of $a_0{}^2$, where a_0 is the first Bohr radius.

2. Calculation of the Resistivity

If one's aim is the calculation of the resistivity ρ due to ionized impurity scattering alone, then one has to compute the relaxation time τ at first. This quantity, in terms of the total scattering cross section Q_0, the number of scatterers N^+, and the velocity of the electrons v, is

given[10] by

$$\tau=1/(N^+vQ_0). \tag{23}$$

Since Q_0 and v are functions of the electron energy, an average value of τ must be used. In case of degenerate semiconductors the averaging over the electron distribution shows[11] that the average relaxation time $\langle\tau\rangle$ is a function of the Fermi energy[12] E_F only. Accordingly, Eq. (23) becomes

$$\langle\tau\rangle=1/(N^+v^FQ_0{}^F), \tag{24}$$

where the superscript F indicates that the corresponding quantities are to be taken at the Fermi energy. The wave number of an electron at the Fermi level, k_F, is calculated from the expression

$$\frac{h^2}{8m^*}\left(\frac{3n}{\pi}\right)^{\tfrac{2}{3}}=\frac{h^2k_F{}^2}{8\pi^2m^*}, \tag{25}$$

where the left-hand side is the Fermi energy and the right-hand side is the kinetic energy of the electron.

According to theory[13] the electron mobility μ is given by

$$\mu=(e_0/m^*)\langle\tau\rangle, \tag{26}$$

and the resistivity by

$$\rho=1/(ne_0\mu). \tag{27}$$

Resistivity values calculated from Eq. (27) are shown in Table I. The k_F, $Q_0{}^F$, $\eta_0{}^F$, R, and $k_F R$ values are also listed as functions of the electron concentration n.

The resistivity in Eq. (27) does not depend on temperature. Measured values at low temperatures, where ionized impurity scattering is the dominant scattering process, are also known to be fairly independent on temperature. Since other scattering processes are not considered in this paper no strict comparison can be made between experimental resistivity values and the calculated values based on ionized impurity scattering alone. It should, however, be remarked that Fritzsche's[14] measurements show that at 77°K germanium samples doped with 10^{17} to 10^{18} antimony atoms per cm^3 have resistivities in the 10^{-2} to 10^{-3} ohm-cm region. This

[10] C. Erginsoy, Phys. Rev. 79, 1013 (1950).
[11] See p. 280 of third work in reference 1.
[12] A. H. Wilson, The Theory of Metals (Cambridge University Press, New York, 1953), 2nd ed., p. 16.
[13] See p. 240 of first work in reference 1.
[14] H. Fritzsche, J. Phys. Chem. Solids 6, 69 (1958).

suggests that the calculated resistivity values in Table I can be considered fairly reliable. Unfortunately, no comparison can be made between Blatt's work[4] and the present work because Blatt considered weakly doped semiconductors and his graphs do not cover the range in which the present parameters are.

III. DISCUSSION

To estimate the range of validity of the present treatment, three questions must be answered. First, how well justified is the use of only the zeroth-order phase shift η_0 in computing the leading term Q_0 of the total scattering cross section Q? Second, how good an approximation is the consideration of Q_0 only in computing Q? Third, how well justified is the picture of individual scattering of the electrons by the donor ions?

The first and second questions can be answered easily. The maximum available energy for an electron in a degenerate semiconductor is the Fermi energy. Therefore, the magnitude of $k_F R$ will determine how many partial waves one has to consider in Eq. (4).

It is seen from Table I that $k_F R$ is approximately equal to unity at the electron concentrations considered.

This means, according to partial-wave theory,[3] that the terms of importance in Eq. (4) are those with $l=0$ and $l=1$. According to an approximate formula[3] the higher-order phase shifts can be calculated from

$$\eta_l \approx -\epsilon_l 2^{2l}(l!)^2(k_F R)^{2l+2}/[(2l+1)!]^2, \qquad (28)$$

where ϵ_l must satisfy the condition

$$|\epsilon_l| \ll l/k_F R.$$

From Eq. (28) one obtains

$$\eta_1 \approx -4\epsilon_1(k_F R)^4/36, \quad \eta_2 \approx -\epsilon_2(k_F R)^6/225,$$

which, assuming $|\epsilon_1| = |\epsilon_2| = 0.1$ and using $k_F R = 1$, leads to

$$|\eta_1| \approx 0.011, \quad |\eta_2| \approx 0.0044.$$

Since $|\eta_1|$ is small compared to $|\eta_0|$ (see Table I), and $|\eta_2|$ is small compared to $|\eta_1|$, the first two terms in Eq. (4) can be written, to a good approximation, as

$$Q_0 \approx (4\pi/k_F^2) \sin^2\eta_0,$$
$$Q_1 \approx (4\pi/k_F^2)2 \sin^2\eta_1 \approx (8\pi/k_F^2)\eta_1^2. \qquad (29)$$

This answers the first question raised above.

To answer the second question, one has to compute Q_1^F. One finds from Eq. (29), using $k_F = 4 \times 10^6$ cm^{-1}, that $Q_1^F \approx 1.89 \times 10^{-16}$ cm^2. Comparing this value with the Q_0^F values in Table I one sees that $Q_1^F/Q_0^F < 10^{-3}$. Since this ratio is small, compared with unity, one concludes that the consideration of Q_0^F alone is justified.

No such clear-cut answer can be given for the third question, namely to what doping level is the scattering of an electron on a donor ion independent of the surrounding other donor ions. From an analysis of the

problem[15] it has been concluded that, at electron concentrations of $n \sim 10^{18}$/cc, about half of the scattering acts are independent while the rest are influenced to variable degrees by the nearest neighbor ions. In view of this fact, one may conclude that at the electron concentrations considered in this paper the assumption of individual scattering is still a reasonable one.

APPENDIX I. EVALUATION OF THE INTEGRALS G_{31}, G_{32}, G_{33}, AND G_1

The integral in Eq. (16) is defined as

$$G_{31} = \int_0^\infty \frac{e^{-\alpha r}}{r}(1-e^{-r})^2 \cos^2 kr\, dr. \qquad (A1)$$

It is seen that the integrand has a singularity at $r=0$. To evaluate Eq. (A1) one considers G_{31} as a function of α. Differentiation of the integral with respect to α results in

$$\frac{d}{d\alpha}G_{31}(\alpha) = -\int_0^\infty e^{-\alpha r}(1-e^{-\alpha r})^2 \cos^2 kr\, dr. \qquad (A2)$$

The integrand in Eq. (A2) is nonsingular at both limits of integration and Eq. (A2) can easily be evaluated as

$$\frac{d}{d\alpha}G_{31}(\alpha) = -\frac{1}{\alpha^2+4k^2}\left[\alpha+\frac{2k^2}{\alpha}\right]$$
$$+2\frac{1}{(\alpha+1)^2+4k^2}\left[(\alpha+1)+\frac{2k^2}{(\alpha+1)}\right]$$
$$-\frac{1}{(\alpha+2)^2+4k^2}\left[(\alpha+2)+\frac{2k^2}{(\alpha+2)}\right]. \qquad (A3)$$

If one now integrates Eq. (A3) with respect to α, the resulting expression is

$$G_{31}(\alpha) = -g_1(\alpha)+2g_2(\alpha)-g_3(\alpha)+C_{31}, \qquad (A4)$$

where

$$g_1(\alpha) = \int \frac{1}{\alpha^2+4k^2}\left[\alpha+\frac{2k^2}{\alpha}\right]d\alpha$$
$$= \tfrac{1}{4}\ln\alpha^2[\alpha^2+4k^2]+C_1,$$

$$g_2(\alpha) = \int \frac{1}{(\alpha+1)^2+4k^2}\left[(\alpha+1)+\frac{2k^2}{(\alpha+1)}\right]d\alpha \qquad (A5)$$
$$= \tfrac{1}{4}\ln(\alpha+1)^2[(\alpha+1)^2+4k^2]+C_2,$$

$$g_3(\alpha) = \int \frac{1}{(\alpha+2)^2+4k^2}\left[(\alpha+2)+\frac{2k^2}{(\alpha+2)}\right]d\alpha$$
$$= \tfrac{1}{4}\ln(\alpha+2)^2[(\alpha+2)^2+4k^2]+C_3,$$

[15] P. Csavinszky, J. Phys. Soc. Japan 16, 1865 (1961).

and $C_{31}=C_1+C_2+C_3$ is a constant of integration which has to be determined. With the aid of Eq. (A5), one rewrites Eq. (A4) as

$$G_{31}(\alpha)=\tfrac{1}{4}\ln\frac{(\alpha+1)^4[(\alpha+1)^2+4k^2]^2}{\alpha^2(\alpha^2+4k^2)(\alpha+2)^2[(\alpha+2)^2+4k^2]}+C_{31}.$$

(A6)

To determine C_{31} one may assume that $\alpha=\infty$. In this case Eq. (A1) shows that G_{31} is zero. The logarithmic term in Eq. (A6) is also zero as can be seen by rewriting both numerator and denominator as a power series in α. Denoting only the leading terms, one obtains

$$0=\tfrac{1}{4}\ln\left\{\frac{\alpha^8+\cdots}{\alpha^8+\cdots}\right\}+C_{31}.$$

(A7)

Using l'Hospital's rule[16] repeatedly, one finds that the bracket has the value of unity and therefore the logarithmic term is zero. For this reason Eq. (A7) can only be satisfied if $C_{31}=0$.

The evaluation of the integrals G_{32}, G_{33}, and G_1, displayed in Eqs. (17), (18), and (19), is done in a quite similar manner.

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[16] P. Franklin, *Methods of Advanced Calculus* (McGraw-Hill Book Company, Inc., New York, 1944), p. 15 ff.

Double Quantum Transition in Electron Spin Resonance of Gamma-Irradiated Acetyl-d,l-Alanine*

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Double quantum transitions were observed at high rf fields in hyperfine spectra of electron spin resonance of the free radical produced by the gamma irradiation of acetyl-d,l-alanine. The presence of interaction between the free radicals give rise to the double quantum transitions. A theoretical explanation is given.

DOUBLE quantum transitions have been observed in the electron spin resonance of Mn^{++} and Ni^{++} ions as impurities in a single crystal of MgO.[1] The absorption lines were interpreted as arising from the simultaneous absorption of two photons, the absorption being excited from the $S_z=M$ to the $S_z=M+2$ state. Three consecutive energy levels participate in this transition.

In the present article the absorption due to a different kind of double quantum transition is reported in the electron spin resonance of a gamma-irradiated single crystal of acetyl-d,l-alanine. Here, the four energy levels of two free radicals are taking part in the transition.

The analysis of the electron spin resonance spectra of the free radical produced by gamma irradiation from acetyl-d,l-analine[2] shows that the free radical has

the following form:
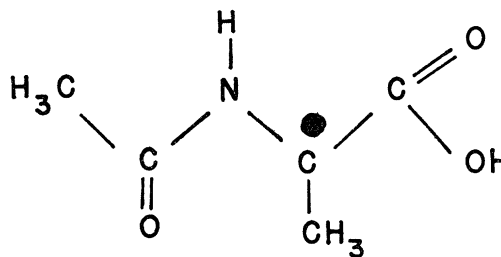
The interaction between the unpaired electron and the CH_3 protons is responsible for the hyperfine structure of these spectra.

Figure 1 shows the hyperfine spectra measured at 9 kMc/sec and 23 kMc/sec with varying microwave power. The spectrum measured at 9 kMc/sec using 6 mw of power consists of a quartet, the intensity ratio being $1:3:3:1$. As greater microwave power was applied, three weak lines were observed at the midpoints of the main lines. The intensities of these lines increase when the amplitude of the microwave H_1 is raised. The weak lines were observed to have the same separation at 23 kMc/sec as at 9 kMc/sec.

The appearance of forbidden transitions, the simul-

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