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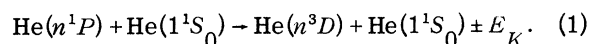
NUMBER 25

## DIRECT DEMONSTRATION OF THE VALIDITY OF THE WIGNER SPIN RULE FOR HELIUM-HELIUM COLLISIONS\*

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There has been considerable discussion in the past thirty years about the validity of the Wigner spin rule<sup>1</sup> for the following collision process:



This process violates the Wigner spin rule which states that the total spin must be conserved in such a process. Early electron and optical excitation experiments by Lees and Skinner<sup>2</sup> and Maurer and Wolf<sup>3</sup> suggested that the above processes occur in helium with high probability. They deduced collision cross sections for such processes as high as ten times the gas-kinetic value of  $1.5 \times 10^{-15} \text{ cm}^2$ .<sup>4</sup> This is difficult to understand as the Wigner spin rule is quite general and spin-orbit coupling is believed to be an excellent approximation for helium.

In 1961, St. John and Fowler<sup>5</sup> suggested a mechanism by which the excitation data could be explained and which was more acceptable on theoretical grounds. They proposed that  $n^1P$  states could transfer excitation by collisions to the  $n^3F$  states and subsequent radiative cascade to the lower  $^3D$  states explains the observed excitation of the  $^3D$  states. This process was acceptable on theoretical grounds because Lin and Fowler<sup>6</sup> showed that for  $nF$  states, spin-orbit coupling breaks down and the spin rule is no longer applicable. Later experiments<sup>7-9</sup> all tended to support the mod-

el of St. John and Fowler.<sup>5</sup>

In the experiment reported here, it is shown directly that processes such as that shown in Eq. (1) are improbable. To our knowledge, this is the first direct demonstration of the validity of the Wigner spin rule for helium. We also confirm Lin and Fowler's prediction by demonstrating that similar processes involving the  $4^3F$  state occur almost as readily as those processes involving no spin change.

Our experimental technique is similar to that used in an experiment performed by Parks and Javan.<sup>10</sup> A helium laser is used to perturb selected excited-state populations in an auxiliary cell containing a pure-helium-gas discharge. The effect of this perturbation on the populations of energy levels other than laser levels is related to the population changes of the laser levels by the equation<sup>11</sup>

$$\Delta N_i = \tau_i \sum_j (A_{ji} + nvq_{ji}) \Delta N_j, \quad (2)$$

where

$$nvq_{ji} = 0.81 \times 10^7 P Q_{ji} \left( \frac{T}{300} \right)^{-1/2} \left( \frac{M_1 + M_2}{M_1 M_2} \right)^{1/2} \\ (\tau_i)^{-1} = A_i + nvq_i, \quad (3)$$

where  $n$  is the density of ground-state helium atoms,  $v$  is the average atomic velocity,  $q_{ij}$  is the velocity-averaged cross section for transfer from level  $i$  to level  $j$ ,  $Q_{ij}$  is the velocity-

averaged cross section for energy transfer from level  $i$  to level  $j$  in units of  $10^{-15}$  cm<sup>2</sup>,  $P$  is the helium pressure in Torr when the discharge is running,  $M_1$  and  $M_2$  are the atomic weights of the colliding species (here  $M_1 = M_2 = 4$ ), and  $A_i$  is the total spontaneous emission probability for the  $i$ th level. The  $A_{ji}$ 's are calculated by the method of Bates and Damgaard<sup>12</sup> and the  $q_i$ 's (total destructive collision cross sections) have been measured by Bennett, Kindlmann, and Mercer.<sup>13</sup> The temperature was inferred by measuring the Doppler width of several spectral lines with a scanning Fabry-Perot interferometer. It was found to be 600°K under the conditions of this experiment.

The population changes of the various levels are monitored by observing the changes in the sidelight emission of the various spectral lines. The population changes are linearly related to the intensity changes through the spontaneous emission rates (Einstein  $A$  coefficients) which are calculated by the method of Bates and Damgaard.<sup>12</sup> The experimental apparatus is shown in Fig. 1. The laser is turned on and off repetitively by the chopper. The sidelight emission is phase-sensitively detected at the chopping frequency. In this manner, the change in sidelight emission in the sample cell due to the laser perturbation is recorded.

Two helium laser transitions were used, the  $4^3P-3^3D$  transition<sup>14</sup> at 1.9543  $\mu$  and the  $4^3F-3^3D$  transition<sup>15</sup> at 1.8685  $\mu$ . When amplifying on one of these two transitions, the population changes of the  $4^3S$ ,  $3^3P$ ,  $3^3D$ ,  $1^3S$ ,  $1^3P$ ,  $1^3D$ , and  $3^3D$  levels were monitored by observing spontaneous emission lines from these levels. This same experiment was performed for each laser transition separately.

When the  $4^3P-3^3D$  laser transition is used, the ratio of the  $3^3D$  population change to that

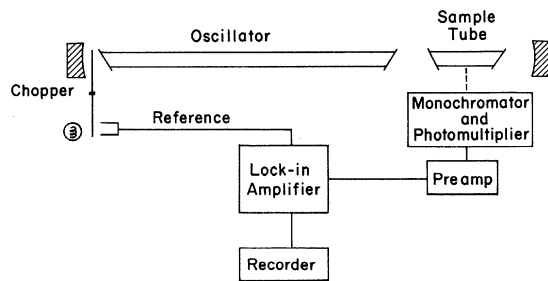


FIG. 1. Experimental apparatus. The sample tube is connected to a vacuum system and the helium pressure is varied from 0.1 to 2.0 Torr.

of the  $4^3P$  level is given by<sup>16</sup>

$$\frac{\Delta N_{3^3D}}{\Delta N_{4^3P}} = -\frac{\tau_{3^3D}}{\tau_{4^3P}} = -\frac{A_{4^3P} + nvq_{4^3P}}{A_{3^3D} + nvq_{3^3D}}. \quad (4)$$

The experimental data for  $\Delta N_{3^3D}/\Delta N_{4^3P}$  versus pressure are shown in Fig. 2. The solid line is the theoretical curve where the values of  $q_{4^3P}$  and  $q_{3^3D}$  are taken from Bennett, Kindlmann, and Mercer<sup>13</sup> and the theoretical Einstein  $A$  coefficients are used. The agreement serves to demonstrate that the technique gives results consistent with previous data.

The population change of the  $4^3D$  level due to amplification on the  $4^3P-3^3D$  transition is given by

$$\frac{\Delta N_{4^3D}}{\Delta N_{4^3P}} = \frac{nvq_{4^3P-4^3D}}{A_{4^3D} + nvq_{4^3D}}. \quad (5)$$

The experimental data for  $\Delta N_{4^3D}/\Delta N_{4^3P}$  are shown in Fig. 3 along with a least-squares fit by Eq. (5). The value of  $q_{4^3P-4^3D}$  determined from these data is

$$q_{4^3P-4^3D} = (0.99 \pm 0.24) \times 10^{-15} \text{ cm}^2, \quad (6)$$

a value comparable in magnitude with the gas-kinetic cross section. Similarly, we determined that

$$q_{4^3P-4^3S} = (0.35 \pm 0.03) \times 10^{-15} \text{ cm}^2. \quad (7)$$

Thus, cross sections for collisional transfer

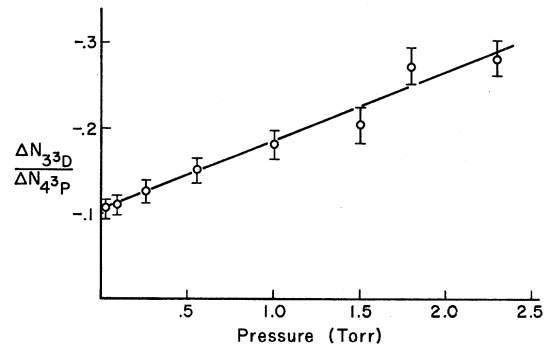


FIG. 2. Experimental values of  $\Delta N_{3^3D}/\Delta N_{4^3P}$  as a function of pressure. The solid curve is the theoretical curve, Eq. (4).

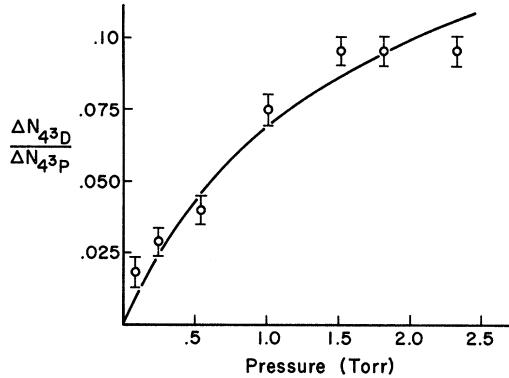


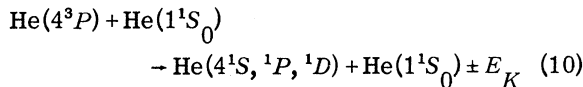
FIG. 3. Experimental values of  $\Delta N_{4^3D}/\Delta N_{4^3P}$  as a function of pressure. The solid curve is the theoretical curve, Eq. (5).

of excitation from the  $4^3P$  to the  $4^3S$  and  $4^3D$  levels are comparable in magnitude with the gas-kinetic cross section as expected. The  $4^3P-3^3D$  laser transition is ideal for studying transfer of excitation from  $4^3P$  to  $4^1S$ ,  $4^1P$ , and  $4^1D$ . The primary effect of the laser is to modulate the population of the  $4^3P$  level at the chopping frequency. The modulation of the  $4^1S$ ,  $4^1P$ , or  $4^1D$  level population is greater than or equal to the modulation due to direct collisional transfer from the  $4^3P$  level since two-step transfer processes from  $4^3P$  to  $4^1S$ ,  $4^1P$ , or  $4^1D$  through an intermediate level might also occur. Thus,

$$\Delta N_{4^1(S,P,D)} \geq \tau_{4^1(S,P,D)} [n\nu q_{4^3P-4^1(S,P,D)}] \Delta N_{4^3P}, \quad (8)$$

$$q_{4^3P-4^1(S,P,D)} \leq \frac{1}{n\nu\tau_{4^1(S,P,D)}} \frac{\Delta N_{4^1(S,P,D)}}{\Delta N_{4^3P}}. \quad (9)$$

Application of Eq. (9) to data taken at 1-Torr helium pressure showed that cross sections for processes of the type



were all less than  $2 \times 10^{-17}$  cm<sup>2</sup> or two orders of magnitude less than the gas-kinetic value and considerably less than the cross sections for transfer to the  $4^3S$  and  $4^3D$  states. This is a direct demonstration that the Wigner spin rule is obeyed for collisional transfer of ex-

citation from the  $4^3P$  level to the  $4^1S$ ,  $4^1P$ , and  $4^1D$  levels.

The data taken when amplifying on the  $4^3F-3^3D$  transition did not yield values for cross sections from  $4^3F$  to other levels because the  $4^3F$  level population could not be monitored. However, it was still possible to compare transfer of excitation from  $4^3F$  to  $4^1D$  with the transfer from  $4^3F$  to  $4^3D$ . Analysis of data taken at 0.1 Torr showed that

$$q_{4^3F-4^1D}/q_{4^3F-4^3D} = 0.6. \quad (11)$$

Therefore, there is no strong preference for transfer to the  $4^3D$  state over transfer to the  $4^1D$  state from the  $4^3F$  level. While we have not measured the cross sections for transfer from  $4^3F$  to other levels, the comparison of  $q_{4^3F-4^1D}$  with  $q_{4^3F-4^3D}$  demonstrates directly that the Wigner spin rule does not apply to collisional transfer of excitation from the  $4^3F$  level.

In conclusion, we have demonstrated directly that the Wigner spin rule applies for collisional transfer of excitation from the  $4^3P$  to the  $4^1S$ ,  $4^1P$ , and  $4^1D$  states. We have also shown that the Wigner spin rule does not apply to collisional transfer from the  $4^3F$  state in agreement with the predictions of Lin and Fowler.<sup>6</sup>

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†National Science Foundation Graduate Fellow.

<sup>1</sup>H. S. W. Massey and E. H. S. Burhop, *Electronic and Ionic Impact Phenomena* (Clarendon Press, Oxford, England, 1955).

<sup>2</sup>J. H. Lees and H. W. B. Skinner, *Proc. Roy. Soc. (London)* **A137**, 186 (1932).

<sup>3</sup>W. Maurer and R. Wolf, *Z. Physik* **92**, 100 (1934); **115**, 410 (1940).

<sup>4</sup>The ratio of the collision cross section for a given process to the gas-kinetic cross section is equal to the number of such processes which occur for each gas-kinetic collision. Cross sections for the theoretically forbidden processes should be much less than the gas-kinetic value.

<sup>5</sup>R. M. St. John and R. G. Fowler, *Phys. Rev.* **122**, 1813 (1961).

<sup>6</sup>C. C. Lin and R. G. Fowler, *Ann. Phys. (N.Y.)* **15**, 461 (1961).

<sup>7</sup>R. M. St. John and T. Nee, *J. Opt. Soc. Am.* **55**, 426 (1965).

<sup>8</sup>W. R. Pendleton, Jr., and R. H. Hughes, *Phys. Rev.* **138**, A638 (1965).

<sup>9</sup>R. B. Kay and R. H. Hughes, *Phys. Rev.* **154**, 61

(1967).

<sup>10</sup>J. H. Parks and A. Javan, Phys. Rev. **139**, A1351 (1965).

<sup>11</sup>The laser directly perturbs the populations of two states within the summation. The remaining terms in the summation arise from radiative and collisional coupling of the levels.

<sup>12</sup>D. R. Bates and A. Damgaard, Phil. Trans. Roy. Soc. London, Ser. A **242**, 101 (1950).

<sup>13</sup>W. R. Bennett, Jr., P. J. Kindlmann, and G. N. Mercer, Appl. Opt. Suppl. **2**, 34 (1965).

<sup>14</sup>R. Cagnard, R. Agobian, R. Echard, and J. Otto,

Compt. Rend. **257**, 1044 (1963).

<sup>15</sup>R. L. Abrams and G. J. Wolga, IEEE J. Quant. Electron. **QE3**, 368 (1967).

<sup>16</sup>If the rate at which atoms are pumped from the upper laser level to the lower laser level by stimulated emission is much larger than the rate at which the collisional and radiative transfer processes occur, then this equation is justified by solution of a simple rate equation. Experimentally it was found that the population changes of nonlaser levels were less than  $\frac{1}{10}$  the laser-level population changes, thereby satisfying the above condition.

### EXCITATION OF LOWER HYBRID OSCILLATIONS AT UPPER HYBRID RESONANCE BY MICROWAVES

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Parametric excitation of plasma waves has been of considerable interest.<sup>1-3</sup> In the absence of a dc magnetic field, a nonlinear coupling process has been observed<sup>1</sup> between two basic collective modes, the electron plasma wave and the ion acoustic waves. There are also several theories<sup>2</sup> related to the parametric excitation of plasma oscillations by means of transverse electromagnetic radiation. In the presence of a dc magnetic field, the excitation of Alfvén waves by a small low-frequency oscillation has been studied theoretically.<sup>3</sup>

In this paper we report the possible observation of lower hybrid oscillations excited by microwaves in the presence of a dc magnetic field. The crucial point is that the parametric coupling is observed only when the microwave frequency  $f_0$  satisfies the condition of upper hybrid resonance,  $f_0^2 = f_{ce}^2 + f_{pe}^2$ , where  $f_{pe}$  and  $f_{ce}$  are the electron plasma frequency and the electron cyclotron frequency, respectively. Figure 1 shows the radiation pattern of microwaves for fixed receiving frequency 4100 MHz as a function of the dc magnetic field. The radiation is in the extraordinary mode for propagation at right angles to the magnetic field. The emission peaks move toward the lower magnetic field region as the current is increased. Further increase of the current yields the second-harmonic radiation at  $f_{ce}/f_0 = 0.5$ . The characteristics of the upper hybrid resonance have been well explained by Bernstein's longitudinal mode.<sup>4,5</sup>

In the present experiment the magnetic field

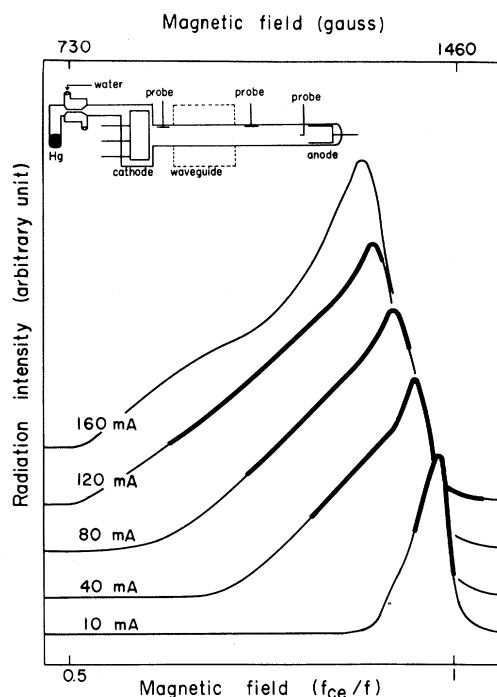


FIG. 1. Microwave radiation (in relative unit) in an extraordinary mode at 4100 MHz from a plasma column of mercury vapor as a function of magnetic field with discharge current as a parameter. The curves for current are displaced for display purposes. The bold line indicates the region where the low-frequency oscillations are excited by the external microwave irradiation. Plasma is produced in mercury vapor at  $3.4 \times 10^{-3}$  Torr. The discharge tube is inserted in a waveguide ( $TE_{10}$ ) with the electric field parallel to the tube axis.