[4]E. A. Power, Theoretical Chemistry Institute of the University of Wisconsin, Report No. 186, 1966 (unpublished).

[5]W. Heitler and S. T. Ma, Proc. Roy. Irish Acad. A52, 109 (1949).

[6]W. Heitler, The Quantum Theory of Radiation (Oxford University Press, London, 1954), 3rd. ed., pp. 163-174.

[7]H. F. Hameka, Advanced Quantum Chemistry (Addison-Wesley Publishing Company, Inc., Reading, Massachusetts, 1965).

[8]The coefficient $A(a, b, \alpha)$ is given by M. E. Rose, J. Math. Phys. 37, 215 (1958).

[9]Both γ and Γ are small quantities which are practically independent of E, and whose imaginary parts produce small energy-level shifts.

DISCHARGE NUCLEAR POLARIZATION IN He^3 GAS*

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We report the existence of substantial polarization effects, apparently involving the 2^3S_1 metastable state, produced by maintaining a discharge in a He^3 sample which is placed in a magnetic field. A polarization of 0.06%, which corresponds to an enhancement of 2200, has been produced at 1 kG. A partially successful theoretical explanation is discussed.

He^3 nuclei in gaseous samples have been polarized to some extent at atmospheric pressure by dynamic polarization interactions with optically pumped alkali vapors[1,2] and heavily polarized by optical pumping of the metastable state[3,2] at low pressures. We report the existence of quite substantial polarization effects produced by maintaining a discharge in a He^3 sample when placed in a magnetic field. Except for the fact that nonequilibrium conditions are to be expected in discharges, it might well be thought that the effect of a discharge would be to increase the electron temperature and bring about relaxation to a small Boltzmann population difference. Indication that there is instead some enhancement of nuclear polarization in a discharge has been previously reported.[2] A careful exploration of the magnetic field dependence of this effect has demonstrated that enhancements of nuclear polarization of greater than 2000 may be obtained and that it is possible to have a negative spin temperature or inverted population in certain ranges of the magnetic field.

Samples of He^3 gas at pressures from 2 to 35 cm Hg contained in Pyrex bulbs of 60 to 100 cm^3 and purified sufficiently so as to have intrinsic relaxation times in excess of 1000 sec were placed in a magnetic field and a discharge was produced in the gas. The homogeneity of the field was sufficient so that no inhomogeneous-field relaxation effects[2-6] were important. For dc discharges, some bulbs contained either hot- or cold-cathode tungsten electrodes. Radiofrequency, continuous microwave, or Tesla-coil discharges were used in other bulbs which did not contain electrodes. Nuclear polarization was subsequently measured with nuclear magnetic resonance.[2]

Figure 1 shows the nuclear polarization as a function of applied field which results when an electrodeless discharge is produced by a 2-Mc/sec oscillator in a bulb which was filled to 2 cm pressure at 300°K. Positive polarization corresponds to a positive spin temperature. The complicated structure which appears below 4 kG, the 4.5-kG crossing from positive to negative polarization, and the dip at 6.25 kG are all reproducible, but the second zero crossing at 7.7 kG depends on the discharge intensity and can be moved to higher fields by increasing the power delivered to the sample. Curve 1 was measured at room temperature, and Curve 2 was measured by placing the bulb in liquid nitrogen, allowing the gas to cool, and then striking the discharge. The maximum polarization is only 0.06%, but at 1 kG this corresponds to an enhancement of about 2200 over the polarization given by Boltzmann statistics. An enhancement of this magnitude at, say, 100 kG would give a polarization of 5%, which would be significantly useful.

Figure 2 gives the measured polarization for two different samples, one at 3 cm Hg with a microwave discharge applied, and the other at 35 cm Hg with a dc discharge applied. The

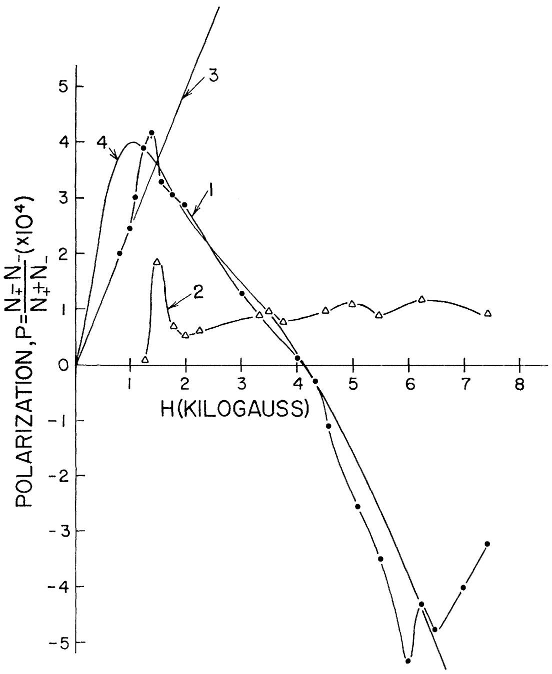

FIG. 1. Nuclear polarization as a function of magnetic field for a sample pressure of 2 cm Hg. Electrodeless discharge produced by a 2-Mc/sec oscillator. Curve 1, measured polarization with initial gas temperature of 300°K. Curve 2, measured polarization when sample was immersed in liquid nitrogen. Curve 3, calculated nuclear polarization assuming that the relative populations of 2^3S_1 hyperfine levels are given by the Boltzmann factor. Curve 4, calculated electron polarization of 2^3S_1 electrons assuming that the relative populations of the 2^3S_1 hyperfine levels are given by the Boltzmann factor.

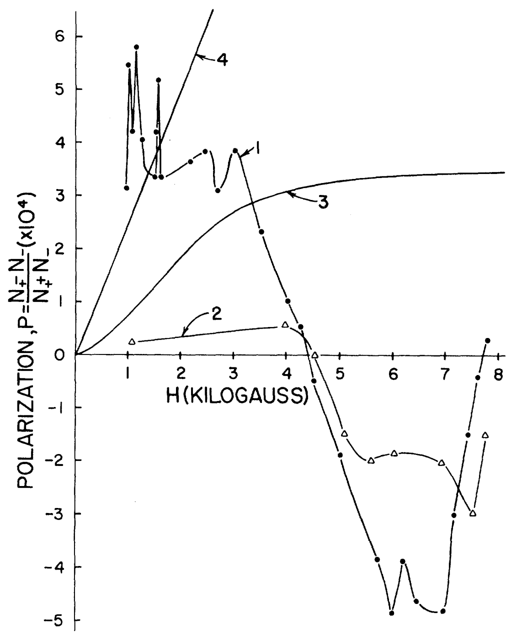

FIG. 2. Nuclear polarization as a function of magnetic field. Curve 1, sample pressure 3 cm Hg. Electrodeless discharge produced by 2450-Mc/sec magnetron. Curve 2, sample pressure 35 cm Hg. Direct-current discharge between cold tungsten electrodes. Curve 3, calculated electron polarization of 2^3S_1 electrons assuming that the relative populations of the 2^3S_1 hyperfine levels are given by the Boltzmann factor. Curve 4, a representative theoretical polarization curve, based on theory described in text. Polarization magnitude multiplied by 4.

dc discharge did not produce a negative spin temperature above 4 kG, but when the same sample was subjected to a pulsed discharge produced by a Tesla coil, a negative-temperature portion appeared for applied fields greater than 4.5 kG. This is the only case in which the type and character of the discharge made any large difference.

Several characteristics of this effect suggest that the 2^3S_1 metastable state may be involved in the polarization process. The decrease in the positive polarization when the gas temperature is reduced to 77°K (Fig. 1) is approximately the same as the decrease in the metastability exchange rate measured by Colegrove, Schearer, and Walters.[7] This implies that the nuclei of the metastables become polarized and then exchange metastability with a ground-state atom. Since the nuclear spin is not affected by this process, a polarized ground state is produced. The polarization rise time was of the order of 10 to 100 sec, which is consistent with a metastability-exchange cross section of 10^{-16} cm^2.[7] Also, the positive peak which appears in the three room-temperature experimental curves occurs very near the field value for which there is a level crossing of two of the 2^3S_1 hyperfine levels; however, no quantitative explanation for this effects has been found. Thirdly, if mercury vapor is added to the He^3, the polarization disappears. Although mercury vapor is known to have little effect on the relaxation time of the ground-state nuclear polarization, it does quench a helium optical-pumping process which uses the 2^3S_1 as the lower level.[2]

An obvious polarization mechanism involving the metastable state is simply that the ensemble of atoms in this state are thermally relaxed, and the resulting Boltzmann polarization is passed on by metastability exchange[3] to the ground state. If this were the mechanism, the resulting polarization would be as shown in Curve 3 of Fig. 1, and would not account for the negative temperature enhancement. A more promising theoretical explanation has been obtained by general development of a suggestion of Javan[8] concerning inversion of populations in gases undergoing discharge. The $^3S_1$ metastable state is regarded as being "saturated" by collisions with "hot" electrons in the discharge, and undergoing metastablity-exchange collisions characterized by a much cooler kinetic temperature. The set of eight rate equations, six for the six metastable-state hyperfine levels, and two for the ground state, have been solved by computer to give the ground-state polarization as a function of magnetic field.

If it is assumed that the 2^3S_1 levels have a finite width in the discharge conditions, then transitions between levels which overlap can occur with high probability in the low-field region where the levels are not far apart. An enhanced positive polarization occurs in this region. At higher (Bach-Goudsmit) fields the electron collisions produce changes predominantly in the m_S values of the metastable state, and the atomic collisions produce changes predominantly in the m_I values. In this region negative enhancement occurs. Curve 4 in Fig. 2 shows a representative theoretical polarization curve. This curve shows general features well, but in order to fit the actual magnitude of enhancement it has been multiplied by a factor of 4. It should be emphasized that the parameters of this theory are not arbitrary, but are such quantities as electron density, pressure, electron elastic cross section, metastable exchange cross section, gas and electron temperature, metastable decay rates, and level widths which, under the experimental conditions, cannot be measured very well. We believe, therefore, under the circumstances and possible complexities of the situation, that agreement is good. Agreement between theory and experimental at higher pressure and/or lower temperatures is correspondingly good and, in some cases, better as far as magnitudes are concerned.

The theory does not account for most of the fine structure of the effects. The theory also does not predict that the enhancement should return to a small value at the higher fields, but in this case we know that the magnet used began to saturate and produced very high inhomogeneities at the end of the range near 8 kG, and we believe that the small enhancement here results from the rapid shortening of the ground-state nuclear relaxation time due to inhomogeneous-field effects.[2-6]

The theory indicates that substantial polarization should exist at high magnetic fields, and that the effect should be enhanced in this region by the use of low gas temperature, higher discharge current, and high field homogeneity which is required to lengthen the nuclear relaxation time. We intend, when facilities are available, to pursue the effect to higher fields in order to determine whether useful polarization can be produced. The negative-temperature polarization which has already been produced, however, is adequate to run a Zeeman maser.[9]

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[1]M. A. Bouchiat, T. R. Carver, and C. M. Varnum, Phys. Rev. Letters 5, 373 (1960).

[2]R. L. Gamblin and T. R. Carver, Phys. Rev. 138, A946 (1965).

[3]F. D. Colegrove, L. D. Schearer, and G. K. Walters, Phys. Rev. 132, 2561 (1963).

[4]R. L. Gamblin and T. R. Carver, Bull. Am. Phys. Soc. 9, 11 (1964).

[5]G. K. Walters, L. D. Schearer, and F. D. Colegrove, Bull. Am. Phys. Soc. 9, 11 (1964).

[6]L. D. Schearer and G. K. Walters, Bull. Am. Phys. Soc. 10, 74 (1965).

[7]F. D. Colegrove, L. D. Schearer, and G. K. Walters, Phys. Rev. 135, A353 (1964).

[8]A. Javan, in Quantum Optics and Electronics; Lectures Delivered at Les Houches During the 1964 Session of the Summer School of Theoretical Physics, University of Grenoble, edited by C. De Witt, A. Blandin, and C. Cohen-Tannoudji (Gordon and Breach Publishers, Inc., New York, 1965), p. 393.

[9]H. G. Robinson and Than Myint, Appl. Phys. Letters 5, 116 (1964).