

# Polarization of hydrogen atoms by chemodissociation of H<sub>2</sub> molecules

S. P. Dmitriev, R. A. Zhitnikov, V. A. Kartoshkin, G. V. Klement'ev, and V. D. Mel'nikov

*A. F. Ioffe Physicotechnical Institute, USSR Academy of Sciences*

(Submitted 28 January 1983)

Zh. Eksp. Teor. Fiz. **85**, 840–851 (September 1983)

The kinetics of orientation of H atoms in a helium-hydrogen plasma following orientation of He\* ( $2^3S_1$ ) atoms is considered. Evolution equations are obtained for the observable quantities, viz., the populations, orientations, and alignment of He\* and the electron orientation of H. The polarized hydrogen atoms are optically recorded on the basis of the relation between the H and He\* orientations produced by chemoionization and spin exchange. It is shown that the major process that determines the polarization of H atoms in a helium-hydrogen plasma when He\* is oriented in it is the chemodissociation reaction  $\text{He}(2^3S_1) + \text{H}_2 \rightarrow \text{HeH}^+ + \text{H}(1^2S_{1/2}) + e^-$ .

PACS numbers: 35.10.Di, 52.20.Hv

Triplet metastable helium atoms having large reserves of internal energy ( $\sim 20$  eV) can ionize and dissociate most molecules by collisions even at thermal energies of relative motion. The first to report formation of polarized atoms in such a chemoionization process were some of us,<sup>1</sup> having observed polarization of hydrogen atoms produced upon collisions of optically oriented  $2^3S_1$  metastable helium atoms with hydrogen molecules.

Direct optical orientation of hydrogen atoms has not been realized to this day because of the difficulties connected with the short wavelength of the resonant  $1s \rightarrow 2p$  transitions of H atoms ( $\lambda = 1216 \text{ \AA}$ ). "Indirect" optical orientation of hydrogen atoms was initially carried out in the course of spin exchange with optically oriented alkali-metal atoms (Na, Rb).<sup>2,3</sup> As already noted in Ref. 1, and later also in Ref. 4, an entirely different method was developed to obtain polarized hydrogen atoms, consisting of dissociation of hydrogen-containing molecules (H<sub>2</sub>, D<sub>2</sub>, HCl) by optically oriented excited He( $2^3S_1$ ) atoms.

In Refs. 1 and 4 the polarized hydrogen atoms were revealed by the change of the electric conductivity of the plasma in magnetic resonance of the atoms H and D. This method is very sensitive, but quantitative investigations using this method are quite difficult.

In the present work we succeeded in optically recording polarized H atoms and were thus able to investigate in detail the orientation of these atoms in a helium hydrogen plasma. The paper consists of two parts. In the first we obtain the kinetic equations for the experimentally observed quantities—the orientation, alignment, and population of the  $2^3S_1$  states of the He atoms and the orientation of the  $1^2S_{1/2}$  states of H atoms. In the second part we present the results of experiments on the production and registration of polarized hydrogen atoms, which are analyzed by starting with the obtained kinetic equations.

## §1. KINETICS OF THE ORIENTATION OF He\*( $2^3S_1$ ) AND H( $1^2S_{1/2}$ ) IN PLASMA

### 1.1 Experimental conditions

He\* atoms in a helium-hydrogen mixture containing He<sup>4</sup> and H<sub>2</sub> in a ratio 10:1 to 100:1 at a total pressure of

several tenths of a torr were optically oriented in a longitudinal magnetic field<sup>5-7</sup> (Fig. 1). To this end a high-frequency gas discharge was excited in the absorption chamber and as a result a fraction  $\sim 10^{-6}$  of the He atoms turned out to be the metastable  $2^3S_1$  state, and these metastable atoms were acted upon by circularly polarized resonant radiation from a helium lamp with the isotope He<sup>4</sup> ( $\lambda = 10830 \text{ \AA}$ , transition  $2^3S_1 \rightarrow 2^3P_{0,1,2}$ ), directed along the constant magnetic field  $H_0$ . Application of an oscillating magnetic field  $H_1 \cos \omega t$  perpendicular to  $H_0$  made the magnetic-resonance signals observable by the change of the pump-light absorption at the instant of resonance.

We have thus achieved in this experiment longitudinal (diagonal, in density-matrix language) pumping and longitudinal (diagonal) registration.

### 1.2 Quantities observable in experiment

Let  $\rho_{\text{He}}$  be a ( $3 \times 3$ ) density matrix corresponding to the magnetic sublevels  $m_s = 1, 0,$  and  $-1$  of the He\* atoms with spin  $S = 1$ . The electron orientation of the  $2^3S_1$  state of

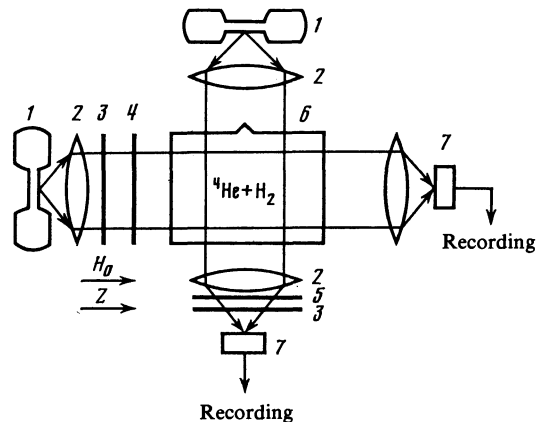


FIG. 1. Experimental setup for optical orientation and magnetic resonance of atoms in an He-H<sub>2</sub> mixture: 1—capillary helium lamp, 2—lens, 3—polarizer, 4—quarter-wave plate, 5—interference filter ( $\lambda = 10830 \text{ \AA}$ ), 6—absorption chamber, 7—FD-24K silicon photodetector.

He is then

$$\langle S_{He} \rangle = \text{Tr}(\rho_{He} S), \quad \text{Tr} \rho_{He} = N_{He} = \text{const}, \quad (1a)$$

and the electron-alignment components are

$$\langle Q_{He} \rangle^{\alpha\beta} = \text{Tr}(\rho_{He} Q_{\alpha\beta}), \quad (1b)$$

$$Q_{\alpha\beta} = 3/2 (S_{\alpha} S_{\beta} + S_{\beta} S_{\alpha}) - S^2 \delta_{\alpha\beta}, \quad (1c)$$

where  $\alpha, \beta = x, y, \text{ or } z$ .

Hydrogen atoms in the ground  $1^2S_{1/2}$  state have an electron spin  $S_H = 1/2$ . The nuclear spin of the H atoms is  $1/2$ . The corresponding density matrices are  $\rho_H (2 \times 2)$  and  $\rho_I (2 \times 2)$ . The total angular momentum  $F = I + S_H$  has eigenvalues 1 and 0. The orientation of the hyperfine sublevel  $F = 1$  is then

$$\langle F_1 \rangle = \text{Tr}(P_1 \rho_H \otimes \rho_I P_1 F), \quad \text{Tr} \rho_H = \text{Tr} \rho_I = N_H = \text{const}, \quad (2a)$$

where  $P_1$  is the corresponding projection operator.

In the absence of hyperfine coherence we have

$$\langle F_1 \rangle = \langle S_H \rangle + \langle I \rangle, \quad \langle S_H \rangle = \text{Tr}(\rho_H S_H), \quad \langle I \rangle = \text{Tr}(\rho_I I). \quad (2b)$$

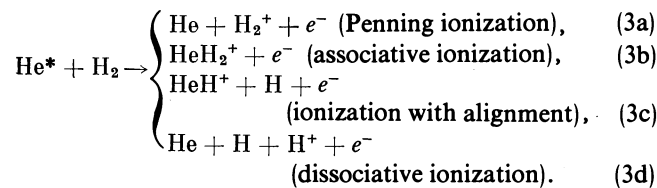
When considering the influence of various elementary processes on the observables (1a), (1b) and (2a) it will be assumed, first, that the interaction time ( $t_{\text{coll}}$ ) is short enough and the nuclear orientation  $\langle I \rangle$  evolves as a result of the hyperfine interaction in the interval between the collisions; second, that there is no stronger nuclear-relaxation mechanism than this interaction. We then have in accord with (2b)

$$\int_{t_{\text{coll}}} d\langle S_H \rangle = \int d\langle F_1 \rangle. \quad (2c)$$

We consider below elementary processes in a helium-hydrogen gas-discharge plasma from the viewpoint of their influence on the quantities  $\langle S_{He} \rangle$ ,  $\langle Q_{He} \rangle$ ,  $\langle S_H \rangle$  considered above and on the population of the  $2^3S_1$  state of helium.

### 1.3.1. Collisions of He\* atoms with H<sub>2</sub> molecules

The quasimolecule He\* – H<sub>2</sub> produced in the collision is unstable to autoionization and decays via the following channels:



The possibility of orientation transfer from the He\* atoms to the products of the chemoionization process (3)—to the H atoms, the electrons, and the molecular ions—is due to the satisfaction of the Wigner rule,<sup>8</sup> i.e., to the conservation of the total electron spin in the process (3).

The rate constant ( $C_1$ ) of this process at 300 K is  $3.4 \times 10^{-11} \text{ cm}^3 \cdot \text{sec}^{-1}$  and was determined in Refs. 6 and 7 for the interval 77–400 K.

The density matrix of an ensemble of atoms will evolve in accord with the equations

$$\frac{d}{dt} \rho_{He} = -\frac{1}{\tau_1} \rho_{He}, \quad \frac{1}{\tau_1} = N_{H_2} C_1, \quad (4a)$$

$$\frac{d}{dt} \rho_H = -\frac{1}{\tau_1'} \text{Tr}_e \rho_H' \otimes \rho_e, \quad \frac{1}{\tau_1'} = \frac{1}{\tau_1} \alpha \frac{N_{He^*}}{N_H}, \quad (4b)$$

$$\alpha = \alpha_a \beta + \alpha_n + \alpha_r, \quad (4c)$$

since the change of the density matrix as a result of the reaction (3) can be represented in the form  $\rho_{He} \rightarrow \rho_H' \otimes \rho_e \rightarrow \rho_H + \rho_e$ , where  $\rho_e$  is the density of the electrons produced in (3).

In (1)–(4),  $N_{H_2}$ ,  $N_{He^*}$ , and  $N_H$  are the densities of the corresponding particles,  $\alpha_k$  are the relative probabilities of the individual channels of (3), and  $\beta$  is the fraction of the electron orientation that is conserved as a result of reaction (3a) and of the secondary reaction of conversion of the molecular hydrogen ions:



From the equations (4) for the evolution of the density matrix we can change to the equations for the observables (1) and (2):

$$\frac{d}{dt} \langle S_{He} \rangle = -\frac{1}{\tau_1} \langle S_{He} \rangle, \quad (6a)$$

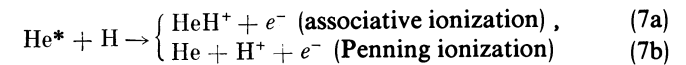
$$\frac{d}{dt} \langle S_H \rangle = \frac{1}{2\tau_1'} \langle S_{He} \rangle. \quad (6b)$$

The rate constant (5) amounts to  $1.4 \times 10^{-9} \text{ cm}^3 \cdot \text{sec}^{-1}$  (Ref. 9), so that the lifetime of the molecular ion H<sub>2</sub><sup>+</sup> relative to (5) is  $\sim 10^{-6} \text{ sec}$  at  $N_{H_2} \sim 10^{15} \text{ cm}^{-3}$ . During this time a spin-rotational interaction takes place (its constant is  $\sim 30 \text{ MHz}$ , Ref. 10), causing the electron orientation to decrease strongly, and the more it does, the larger the rotational quantum number  $j$ . In the exothermal process (3), part of the energy goes to excitation of the internal degrees of freedom of the molecular ion, therefore the rotational temperature is considerably higher than the temperature of the absorption chamber. Simple estimates show that  $\beta$  does not exceed several hundredths.

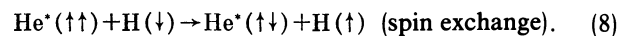
At  $T = 300 \text{ K}$  we have  $\alpha_a = 0.8$ ,  $\alpha_c = 0.15$  (Ref. 11), and apparently  $\alpha_d$  does not exceed 0.01. Thus,  $\alpha \approx 0.2$  and receives the main contribution from the process (3c) of ionization with alignment, which together with the dissociative ionization process (3d) will be named hereafter chemodissociation.

### 1.3.2. Collisions between He\* and H atoms

In this case, besides elastic scattering with spin-orientation conservation, the possible processes are



or



The arrows in (8) show arbitrarily one of the possible orientations of the electron spins of the collision partners relative to the Z axis.

The quasimolecule (HeH)\* has a doublet and a quartet term. Spin exchange is impossible for either, and the effect of spin exchange on an ensemble of atoms is determined by the interference of the amplitudes of the elastic scattering by the

doublet and quartet interaction potentials of the quasimolecule (HeH)\*. At the same time, the course of the chemoionization process (7) depends on the spin state of He\* and H, viz., the reaction is forbidden if the total spin of the initial particles is 3/2, for the Wigner rule<sup>1,4</sup> is not satisfied in this case; the quartet term is therefore not autoionizing.

Examining the evolution of the density matrices  $\rho_{\text{He}}$  and  $\rho_{\text{H}}$  resulting from (7), we find in full analogy with the procedure used in Subsec. 1.3.1 that the orientations of He\* and H vary in the following manner:

$$\frac{d}{dt} \langle S_{\text{He}} \rangle = -\frac{1}{3\tau_2} \left( \langle S_{\text{He}} \rangle - \frac{4}{3} \langle S_{\text{H}} \rangle - \frac{2}{3} \langle S_{\text{H}} \rangle \langle Q_{\text{He}} \rangle \right),$$

$$\frac{1}{\tau_2} = N_{\text{H}} C_2, \quad (9a)$$

$$\frac{d}{dt} \langle S_{\text{H}} \rangle = -\frac{1}{3\tau_2'} \left( \langle S_{\text{H}} \rangle - \frac{1}{2} \langle S_{\text{He}} \rangle \right), \quad \frac{1}{\tau_2'} = N_{\text{He}^*} C_2, \quad (9b)$$

whereas the population of the  $2^3S_1$  state of He varies like

$$\frac{dN_{\text{He}^*}}{N_{\text{He}^*} dt} = -\frac{1}{3\tau_2} (1 - 2 \langle S_{\text{H}} \rangle \langle S_{\text{He}} \rangle). \quad (9c)$$

Here  $C_2$  is the rate constant of (7), equal to  $4 \times 10^{-9} \text{ cm}^3 \cdot \text{sec}^{-1}$  at 300 K and changes insignificantly with temperature.<sup>12</sup>

Equation (9c) is a particular case of the general expressions<sup>13</sup> for the particle ionization probability in a Penning process.

It is likewise easy to show that the evolution of the orientations as a result of the spin exchange satisfies the equations

$$\frac{d}{dt} \langle S_{\text{He}} \rangle = -\frac{1}{\tau_3} \left( \frac{1}{2} \langle S_{\text{He}} \rangle - \frac{4}{3} \langle S_{\text{H}} \rangle + \frac{1}{3} \langle S_{\text{H}} \rangle \langle Q_{\text{He}} \rangle \right),$$

$$\frac{1}{\tau_3} = N_{\text{H}} C_3, \quad (10a)$$

$$\frac{d}{dt} \langle S_{\text{H}} \rangle = -\frac{1}{\tau_3'} \left( \frac{4}{3} \langle S_{\text{H}} \rangle - \frac{1}{2} \langle S_{\text{He}} \rangle - \frac{1}{3} \langle S_{\text{H}} \rangle \langle Q_{\text{He}} \rangle \right),$$

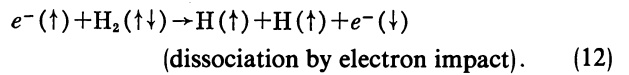
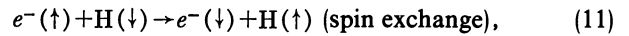
$$\frac{1}{\tau_3'} = N_{\text{He}^*} C_3, \quad (10b)$$

where  $C_3$  is the rate constant of (8) and has not been determined in experiment to this day.

Equations (6), (9), and (10) describe the "longitudinal" experiment and were obtained under the assumption that the total electron spin and its projection on the Z axis is conserved. No account is taken in them of the electron-ion interactions observed during the collision time, since this time is usually much shorter than the hyperfine-interaction time. It is likely, however, that this is not so for the attracting interaction potential realized for the doublet term of the (HeH)\* quasimolecule. The corresponding modification of (9) and (10) entails no difficulty, but so far there are no experiments that show the extent to which it is justified. At any rate, the structure (9), (10) is preserved also when spin is taken into account.

Optical orientation of atoms in a plasma produce spin-oriented electrons that can influence the polarization of the

hydrogen via the following reactions:



Estimates show that the influence of these reactions on the polarization of hydrogen is much weaker than that of processes (3), (7), and (8). Likewise inessential are collisions with other polarized particles in the discharge ( $\text{He}^+$ ,  $\text{He}_2^+$ , and others).<sup>1,4</sup>

#### 1.4. Evolution equations for the observable quantities

Under the conditions of the described experiment, the evolution of the longitudinal (relative to the constant magnetic field  $H_0$ ) components of the orientations satisfies, according to (4), (9), and (10), the system of coupled equations

$$\frac{d}{dt} \langle S_{\text{He}} \rangle^z = -\left( \frac{1}{\tau_1} + \frac{1}{3\tau_2} + \frac{1}{2\tau_3} + \frac{1}{\tau_r} + \frac{1}{\tau_p} + \frac{1}{\tau_d} \right) \langle S_{\text{He}} \rangle^z + \left( \frac{4}{9\tau_2} + \frac{4}{3\tau_3} \right) \times \langle S_{\text{H}} \rangle^z + \left( \frac{2}{9\tau_2} - \frac{1}{3\tau_3} \right) \langle S_{\text{H}} \rangle^z \langle Q_{\text{He}} \rangle^{zz} + \frac{L_p}{\tau_p}, \quad (13)$$

$$\frac{d}{dt} \langle S_{\text{H}} \rangle^z = \left( \frac{1}{2\tau_1'} + \frac{1}{6\tau_2'} + \frac{1}{2\tau_3'} \right) \langle S_{\text{He}} \rangle^z + \frac{1}{3\tau_3'} \langle S_{\text{H}} \rangle^z \langle Q_{\text{He}} \rangle^{zz} - \left( \frac{1}{3\tau_2'} + \frac{4}{3\tau_3'} + \frac{1}{\tau_r'} + \frac{2}{\tau_d'} \right) \langle S_{\text{H}} \rangle^z. \quad (14)$$

The evolution of the population and of the alignment of He\* is described by the equations

$$\frac{dN_{\text{He}^*}}{dt} = -\left[ \frac{1}{3\tau_2} (1 - 2 \langle S_{\text{H}} \rangle^z \langle S_{\text{He}} \rangle^z) + \frac{1}{\tau_1} + \frac{1}{\tau_r} + \frac{1}{\tau_d''} \right] N_{\text{He}^*} + K, \quad (15)$$

$$\frac{d}{dt} \langle Q_{\text{He}} \rangle^{zz} = -\left( \frac{1}{\tau_1} + \frac{1}{3\tau_2} + \frac{3}{2\tau_3} + \frac{1}{\tau_r} + \frac{1}{\tau_d} + \frac{1}{\tau_p'} \right) \langle Q_{\text{He}} \rangle^{zz} + \left( \frac{2}{3\tau_2} + \frac{1}{\tau_3} \right) \langle S_{\text{He}} \rangle^z \langle S_{\text{H}} \rangle^z + \frac{L_p'}{\tau_p'}. \quad (16)$$

The terms with  $1/\tau_r$ ,  $1/\tau_r'$ ,  $1/\tau_d$ ,  $1/\tau_d'$ ,  $1/\tau_d''$ ,  $1/\tau_p$ ,  $1/\tau_p'$ ,  $L_p$ , and  $L_p'$  take into account here the influence exerted on the observables of the other processes in the gas discharge, of diffusion to the walls of the absorption chamber, and of the pump light, while  $K$  is a constant that depends on the discharge intensity.

Since the alignment of He\* does not exceed several hundredths, its influence on the orientation of He\* and H, expressed in (13) and (14) by terms of the form  $\langle S_{\text{H}} \rangle^z \langle Q_{\text{He}} \rangle^{zz}$ , can be neglected. This yields the linear system of equations:

$$\frac{d}{dt} \langle S_{\text{He}} \rangle^z = -(p_1 + F_1) \langle S_{\text{He}} \rangle^z + r_1 \langle S_{\text{H}} \rangle^z + \varphi_p, \quad \varphi_p = \frac{L_p}{\tau_p}, \quad (17a)$$

$$\frac{d}{dt} \langle S_{\text{H}} \rangle^z = r_2 \langle S_{\text{He}} \rangle^z - (p_2 + F_2) \langle S_{\text{H}} \rangle^z, \quad (17b)$$

where

$$p_1 = \frac{1}{\tau_1} + \frac{1}{3\tau_2} + \frac{1}{2\tau_3} + \frac{1}{\tau_r} + \frac{1}{\tau_p} + \frac{1}{\tau_d}, \quad r_1 = \frac{4}{9\tau_2} + \frac{4}{3\tau_3}, \quad (17c)$$

$$p_2 = \frac{1}{3\tau_2'} + \frac{4}{3\tau_3'} + \frac{1}{\tau_r'} + \frac{2}{\tau_d'}, \quad r_2 = \frac{1}{2\tau_1'} + \frac{1}{6\tau_2'} + \frac{1}{2\tau_3'}.$$

The diagonal terms  $F_1$  and  $F_2$  were introduced to take into account the influence of the constant and alternating magnetic fields,<sup>14</sup> and their explicit forms are

$$F_i(\omega, \omega_1) = (i\omega_1)^2 \frac{p_i}{p_i^2 + (\omega - i\omega_0)^2}, \quad i=1, 2, \quad (18)$$

where  $\omega_0 = \gamma H_0$ ,  $\omega_1 = \gamma H_1$ , and  $\gamma$  is the gyromagnetic ratio of the H atoms for the hyperfine sublevel  $F = 1$ .

In the stationary case the solution of (17) is obvious:

$$\langle S_{He} \rangle^z = \frac{\Phi_p(p_2 + F_2)}{D}, \quad \langle S_H \rangle^z = \frac{r_2 \Phi_p}{D}, \quad (19)$$

where

$$D = (p_1 + F_1)(p_2 + F_2) - r_1 r_2.$$

### 1.5. Magnetic resonance of the atoms He\* and H

In the case of magnetic resonance we have  $\omega \approx 2\omega_0$  in the system of Zeeman sublevels of He\*, so that  $F_2 \approx 0$ . Then

$$\langle S_H \rangle^z = \frac{r_2}{p_2} \langle S_{He} \rangle^z, \quad (20a)$$

$$\langle S_{He} \rangle^z = \frac{\Phi_p p_2}{d} \left[ 1 - (2\omega_1)^2 \frac{p_1 p_2 / d}{p_1^2 + \Delta\omega^2 + (2\omega_1)^2 p_1 p_2 / d} \right], \quad (20b)$$

where  $d = p_1 p_2 - r_1 r_2$  and  $\Delta\omega = \omega - 2\omega_0$ .

Variation of the magnetic field yields a Lorentzian resonance curve with a half-width  $\Delta\omega_{1/2} = p$  when extrapolated to zero (in terms of the rf magnetic field). This line is analyzed in Refs. 7 and 8.

At resonance we have  $\omega \approx \omega_0$  in the system of Zeeman sublevels of the H atom, so that  $F_1 \approx 0$ . Then

$$\langle S_H \rangle^z = \frac{\Phi_p r_2}{d} \left( 1 - \omega_1^2 \frac{p_1 p_2 / d}{p_2^2 + \Delta\omega^2 + \omega_1^2 p_1 p_2 / d} \right) \quad \Delta\omega = \omega - \omega_0, \quad (21a)$$

$$\langle S_{He} \rangle^z = \frac{\Phi_p}{p_1} \left[ 1 + \frac{r_1 r_2}{d} \left( 1 - \omega_1^2 \frac{p_1 p_2 / d}{p_2^2 + \Delta\omega^2 + \omega_1^2 p_1 p_2 / d} \right) \right]. \quad (21b)$$

Even in the case of rf saturation it is impossible to disorient the helium completely, owing to the presence of the pump light.

According to (21), the changes of the orientations of He\* and H at resonance are connected by the relation

$$\Delta \langle S_H \rangle^z / \Delta \langle S_{He} \rangle^z = p_1 / r_1. \quad (22)$$

Finally, it follows from (15) that at magnetic resonance of the He\* or H atom a change is observed in the population of the  $2^2S_1$  state of He:

$$\frac{\Delta N_{He^*}}{N_{He^*}} = -\frac{2\tau}{3\tau_2} (\langle S_H \rangle_{\max}^z \Delta \langle S_{He} \rangle^z + \langle S_{He} \rangle_{\max}^z \Delta \langle S_H \rangle^z - \Delta \langle S_H \rangle^z \Delta \langle S_{He} \rangle^z) \quad (23)$$

where  $1/\tau = 1/\tau_1 + 1/3\tau_2 + 1/\tau_r + 1/\tau_d$ ;  $\langle S_H \rangle_{\max}^z$ ,  $\langle S_{He} \rangle_{\max}^z$ , while  $\Delta \langle S_H \rangle^z$  and  $\Delta \langle S_{He} \rangle^z$  are the maximum

orientations of He\* and He and their changes with the rf field, as determined from (20) and (21).

In the case of magnetic resonance of the H atoms we have

$$\Delta \langle Q_{He} \rangle^{zz} = \left( 1 + \frac{3\tau_2}{2\tau_3} \right) \frac{\Delta N_{He^*}}{N_{He^*}}. \quad (24)$$

## §2. EXPERIMENTAL STUDY OF POLARIZED H ATOMS

The experimental setup was described in general outline in Subsec. 1.1 (see Fig. 1). The optical registration of the hydrogen-atom magnetic-resonance signals is based on the spin dependence of the probabilities of reactions (7) and (8). The orientation of the He\* atoms in reaction (8), and both their orientation and density in (7), depend on the mutual spin orientations of the He\* and H atoms (see Subsec. 1.3.2). Therefore destruction of the polarization of the H atoms at magnetic resonance changes the absorption of the pump light by the He\* atoms, so that the magnetic resonance of the H atoms can be observed. Besides the longitudinal resonant signal ( $X_{\parallel}$ ), a transverse signal ( $X_{\perp}$ ) can also be observed by using a transverse unpolarized sounding beam from a helium lamp.

In the case of longitudinal observation, magnetic resonance of the H atoms is accompanied by a change of the absorption of the circularly polarized light

$$X_{\parallel} = -A_0 \left| \frac{\Delta N_{He^*}}{N_{He^*}} \right| + A_1 |\Delta \langle S_{He} \rangle^z| + A_2 |\Delta \langle Q_{He} \rangle^{zz}|, \quad (25)$$

where the coefficients  $A_0$ ,  $A_1$ , and  $A_2$  are determined by the relative probabilities of the transitions between the different Zeeman sublevels of the  $2^3S_1$  and  $2^2P_{0,1,2}$  states of the helium, as well as by the intensities of the  $D_0$ ,  $D_1$ , and  $D_2$  lines of the helium pump lamp and by the absorption contour of the cell.<sup>5</sup> The light absorption decreases at the instant of resonance because of the decrease of the He\* density, and increases because of the decrease of the orientation and alignment of the He\*.

In transverse observation of the absorption of unpolarized light, the magnetic-resonance signal is

$$X_{\perp} = -A_0 \left| \frac{\Delta N_{He^*}}{N_{He^*}} \right| + A_2' |\Delta \langle Q_{He} \rangle^{zz}|. \quad (26)$$

Magnetic resonance of the H atoms was produced in a magnetic field  $H_0 \sim 3.5$  Oe. Two well-resolved lines were observed with a signal/noise ratio 100 and larger, corresponding to the transitions  $m_F = 0 \leftrightarrow F = 1$ ,  $m_F = -1$  and  $F = 1$ ,  $m_F = 0 \leftrightarrow F = 1$ ,  $m_F = 1$  (Fig. 2). The line positions agree with those calculated by the Breit-Rabi formula.<sup>15</sup> By virtue of the statements made in Subsec. 1.2 we have

$${}^{1/2} \Delta \langle F_1 \rangle^z(\omega, \omega_1) = \Delta \langle S_H \rangle^z(\omega, \omega_1).$$

Since the degree of polarization of He\*, and all the more of H, does not exceed usually several hundredths, and  $He^4$ ,  $A_0 \gg A_2, A_2'$ , for optical orientation is by a pump lamps containing also the isotope He<sup>4</sup> one can expect, taking (24) into account, a small effect of the longitudinal and transverse signals from the change of alignment by the resonance of the atoms. This premise was verified by us experimentally by

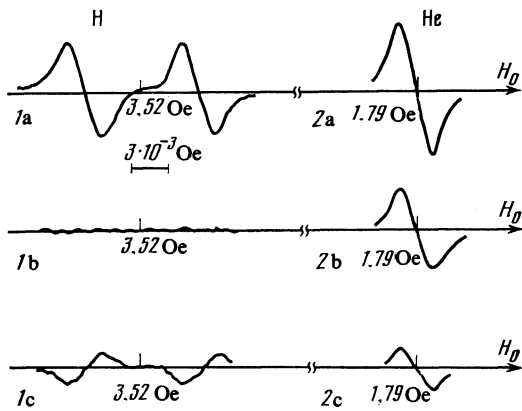


FIG. 2. Derivatives of the change of the absorption of  $1.08 \mu\text{m}$  light at magnetic resonance of the H atoms (1) and He\* atoms (2)  $a - \Delta I/I = 25\%$ ,  $b - \Delta I/I = 20\%$ ,  $c - \Delta I/I = 15\%$ . The gain for He\* resonance is smaller by a factor 500.

placing a polarizer in the path of the transverse ray and plotting the resonance signal as a function of the angle between the direction of the field  $H_0$  and the electric field of the light wave (by the procedure of Ref. 16). No change of the signal was observed in this case. It is therefore meaningless to speak of any influence exerted by the alignment of He\* on the signals  $X_{\parallel}$  and  $X_{\perp}$  in the case of magnetic resonance of the H atoms.

Figure 3 shows plots of the amplitudes of the hydrogen-atom magnetic-resonance signals vs the relative absorption of  $1.08 \mu\text{m}$  light ( $\Delta I/I$ ). It can be noted, first, that a transverse signal is of the same order of magnitude as the longitudinal. Second, the resonance signal reverses sign when the light absorption by the working chamber changes, i.e., with change of the He\* density which increases with increasing intensity of the high-frequency discharge in the absorption chamber.

This reversal of the sign of  $X_{\parallel}$  agrees with the fact that the signs of the terms in the right-hand side of (25) are opposite, but is still surprising, for at first glance it might appear

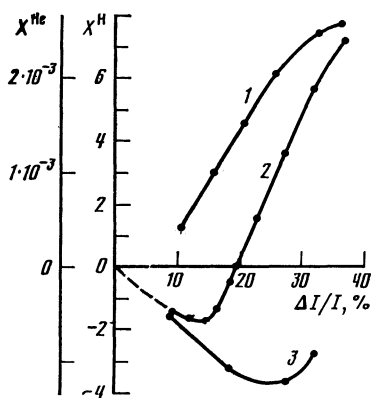


FIG. 3. Dependence of the magnetic-resonance signals (in arbitrary units) on the relative absorption of  $1.08 \mu\text{m}$  light ( $\text{H}_2$  and He pressures 0.015 and 0.36 Torr,  $T = 300 \text{ K}$ , regime close to saturation): 1—He\* resonance signal, 2—longitudinal signal  $X_{\parallel}$  of the H atoms, 3—transverse signal  $X_{\perp}$  of the H atoms.

that the first term of (25) should be considerably smaller than the second, since it contains [according to (23)] the product of the orientations of He\* and H, quantities much smaller than unity. It should also be noted here that the longitudinal signal connected with the magnetic resonance of He\* does not reverse sign (curve 1 of Fig. 3).

To ascertain the cause of the reversal of the sign of the longitudinal signal of the H atoms let us examine in greater detail the structure of (25). Assume for simplicity that the conditions are close to rf saturation ( $\omega_1 \rightarrow \infty$ ). At resonance of the H atoms we have then

$$X_{\parallel} = \frac{\tau}{3\tau_2} |\Delta \langle S_{\text{H}} \rangle^z| (-2A_0 \langle S_{\text{He}} \rangle_{\text{max}}^z + \varepsilon A_1), \quad (27a)$$

$$\varepsilon \approx \frac{4}{3} + 4C_3/C_2, \quad (27b)$$

and at resonance of the He\* atoms

$$X_{\parallel}' \approx |\Delta \langle S_{\text{He}} \rangle^z| \left( -A_0 \langle S_{\text{H}} \rangle_{\text{max}}^z \frac{4\tau}{3\tau_2} + A_1 \right). \quad (28)$$

It follows from (27a) that at the instant when the longitudinal resonant signal of the H atoms goes through zero we have

$$A_0/A_1 = \varepsilon/2 \langle S_{\text{He}} \rangle_{\text{max}}, \quad (29)$$

whence it follows that the coefficient  $A_0$  should be considerably larger (by 10—20 times) than  $A_1$ , since the degree of polarization of He\* is several hundreds and  $\varepsilon \approx 2$ . This relation between  $A_0$  and  $A_1$  can hold because the absorption of the  $D_0$  and  $D_1$  helium lines decreases, while that of the  $D_2$  line increases in the presence of He\* orientation,<sup>17</sup> whereas the absorption of all three lines increases with increasing population of the  $2^3S_1$  state of He. Relation (29) is indeed satisfied under the conditions of our experiment, so that there is no  $X_{\parallel}$  signal at a certain charge intensity (Fig. 3, curve 2). At low intensities of the high-frequency discharge (and hence at small absorption of the resonant light), a greater role is assumed by the first term, and a signal with negative phase (relative to the He\* resonance signal) is observed, while at higher discharge intensities the second term of (25) is more important and a resonance signal of the same phase as in the resonance of the He\* atoms is observed. The relative role of the terms (27a) varies with the discharge intensity, both because of the change of the coefficients  $A_0$  and  $A_1$  and because of the decrease of the He\* orientation with increasing absorption of the  $1.08 \mu\text{m}$  pump light.

As for the possibility of reversal of the longitudinal-signal light when the He\* atoms are resonant, it can hardly occur under the conditions of our experiment [see (28)], for usually  $\tau_1 \ll \tau_2$ , i.e.,  $N_{\text{H}} C_2 \ll N_{\text{He}} C_1$ .

It can be seen from Fig. 3 that in the case of weak absorption of the light the predominant contribution to the resonance signal comes from the change of the He\* population, while for strong absorption it comes from the change of the He\* orientation. The dependences of the corresponding signals on the density of the  $\text{H}_2$  molecules are shown in Fig. 4. According to (21a) we have close to saturation

$$\Delta \langle S_{\text{He}} \rangle^z = \frac{\Phi p r_1 r_2}{p_1 (p_1 p_2 - r_1 r_2)}. \quad (30)$$

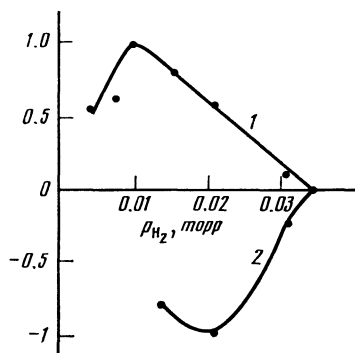


FIG. 4. Magnetic-resonance signal of hydrogen atoms ( $X^H/X_{\max}^H$ ) vs the density of the  $H_2$  molecules ( $T = 300$  K): 1 —  $\Delta I/I = 35\%$ , 2 —  $\Delta I/I = 15\%$ .

At low  $H_2$  pressures (less than 0.005 Torr at 300 K) the resonance signal therefore increases with  $H_2$  density, since  $p_1$  is almost independent of  $N_{H_2}$ , whereas  $r_1 \sim N_H$  and  $r_2 \sim (\alpha C_1 N_{H_2} + 1/6 C_2 N_H + 1/2 C_3 N_H)$  [see (17c)]. At relatively high  $H_2$  densities (0.02 Torr and higher) we have  $p_1 \sim N_{H_2} C_1$ , and the signal decreases with increasing  $N_{H_2}$ . In the intermediate density region, a maximum signal is observed at  $p = 0.01$  Torr (curve 1 of Fig. 4).

The explanation of the pressure dependence of the negative-phase signal (curve 2 of the same figure) is perfectly similar. The maximum signal is observed at  $p = 0.02$  Torr.

The results shown in Fig. 4 agree thus with the theoretical analysis in §1. They make it possible to choose the optimal experimental conditions.

Figure 5 shows the dependence of the H-atom magnetic-resonance signal on the temperature of the absorption chamber at constant absorption of pump light and at relatively low density of the  $H_2$  molecules. The magnetic-resonance signal decreases noticeably with decreasing temperature, and the temperature dependence of the rate constant  $C_1$  is well duplicated here. In accord with (30) and (17c) we find then that  $\alpha C_1 N_{H_2} \gg (1/6 C_2 + 1/2 C_3) N_H$ , i.e., the hydrogen is polarized mainly via chemodissociation of the  $H_2$  molecules as the collide with the metastable  $2^3S_1$  helium atoms, while

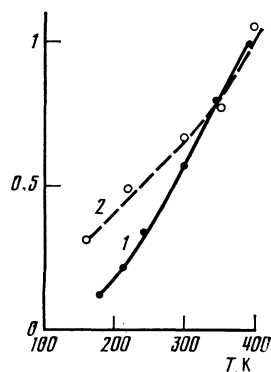


FIG. 5. Temperature dependence of the magnetic-resonance signal of H atoms ( $X_{\parallel}^H/X_{\parallel}^H_{400K}$ , curve 1),  $\Delta I/I = 53\%$ ,  $p_H = 0.004$  Torr at 300 K. Also shown is the temperature dependence of the rate constant  $C_1$  of the process (3) (Refs. 6 and 7) ( $C_1/C_{400K}$ , curve 2).

the processes (7) and (8) play a much lesser role in the production of the polarized H atoms. At the same time, owing to these two processes (chemoionization of the atomic hydrogen and spin exchange) a connection is established between the orientations of H and  $He^*$ , so that the magnetic resonance of hydrogen atoms is revealed by the change of the orientation (or population) of the  $2^3S_1$  state of the helium.

As for the possibility of separating the processes (7) and (8), it encounters no difficulty in principle. According to (29), to find the value of  $\epsilon$ , which is determined by the ratio of the rate constants  $C_2$  and  $C_3$ , it is necessary to measure the  $He^*$  orientation and the coefficients  $A_0$  and  $A_1$  (e.g., with a Fabry-Perot interferometer) at a high-frequency discharge intensity corresponding to a zero longitudinal magnetic-resonance signal of the H atoms.

## CONCLUSION

1. We have considered the kinetics of the orientation of H atoms in a helium-hydrogen plasma in which  $He^*$  atoms are optically oriented. Equations were obtained for the evolution of the observed quantities—the population, orientation, and alignment of  $He^*$  and the electron orientation of H.

2. It is shown that the orientations of H and  $He^*$  are interconnected because of the chemoionization and spin-exchange processes (7) and (8). This has made it possible to detect the magnetic resonance of polarized hydrogen atoms, and furthermore by two methods:

a) on a longitudinal beam of circularly polarized light—from the change of the density and orientation of the  $He^*$  atoms;

b) on a transverse beam of unpolarized light—from the change of the density of the  $2^3S_1$  atoms of  $He^*$ .

3. The experimental investigations of polarized hydrogen atoms (see Figs. 3 and 5) confirm the kinetics, described in §1, of the orientation of  $He^*$  and atoms in a gas-discharge plasma.

4. It was shown that the principal process that leads to formation of polarized H atoms in a helium-hydrogen plasma in which  $He^*$  atoms are oriented is the chemodissociation reaction (3c)



This follows from the character of the temperature dependence of the intensity of the resonance signal polarized H atoms, a dependence close to the temperature dependence determined in Refs. 6 and 7 for the rate constant  $C_1$ . The number of polarized H atoms is seen to increase with temperature in this case, whereas in the case when the H atoms are predominantly polarized as a result of (7) or (8), or via spin exchange with the electrons, a different dependence should have been observed: either a decrease of the polarization of the hydrogen atoms with increasing temperature, or practically no dependence on temperature.

5. A method is proposed for separating the ionization (7) and the spin exchange (8) which take place when  $He(2^3S_1)$  and  $H(1^2S_{1/2})$  collide. This method can also be used for other systems, for example  $He^*$  plus an alkali metal.

In conclusion, the authors thank A. I. Okunevich for a helpful discussion of the results.

- <sup>1</sup>S. P. Dmitriev, R. A. Zhitnikov, V. A. Kartoshkin, G. V. Klement'ev, and A. I. Okunevich, *Pis'ma Zh. Eksp. Teor. Fiz.* **28**, 442 (1978) [*JETP Lett.* **28**, 409 (1978)].
- <sup>2</sup>L. W. Anderson, F. M. Pipkin, and J. C. Baird, *Phys. Rev. Lett.* **1**, 229 (1958).
- <sup>3</sup>L. W. Anderson, F. M. Pipkin, and J. C. Bird, *Phys. Rev.* **120**, 1279 (1960).
- <sup>4</sup>S. P. Dmitriev, V. A. Kartoshkin, G. V. Klement'ev, V. D. Mel'nikov, and A. I. Okunevich, *Opt. Spektrosk.* **51**, 221 (1981).
- <sup>5</sup>L. D. Schearer, *Adv. in Quant. Electron.* 1961, p. 239.
- <sup>6</sup>R. A. Zhitnikov, V. A. Kartoshkin, and G. V. Klement'ev, *Pis'ma Zh. Eksp. Teor. Fiz.* **26**, 651 (1977) [*JETP Lett.* **26**, 496 (1977)].
- <sup>7</sup>G. V. Klement'ev, V. D. Mel'nikov, and V. A. Kartoshkin, *Zh. Eksp. Teor. Fiz.* **77**, 52 (1979) [*Sov. Phys. JETP* **50**, 25 (1979)].
- <sup>8</sup>E. Wigner, *Gött. Nachr.* **4**, 375 (1927).
- <sup>9</sup>R. Johnsen and M. A. Biondi, *J. Chem. Phys.* **61**, 212 (1974).
- <sup>10</sup>K. B. Jefferts, *Phys. Rev. Lett.* **23**, 1476 (1969).
- <sup>11</sup>L. T. Specht, K. D. Foster, and E. E. Muschlitz, *J. Chem. Phys.* **63**, 1582 (1975).
- <sup>12</sup>W. H. Miller, C. A. Slocumb, and H. F. Schaefer, *J. Chem. Phys.* **56**, 1347 (1972).
- <sup>13</sup>A. I. Okunevich, *Zh. Eksp. Teor. Fiz.* **70**, 899 (1976) [*Sov. Phys.* **43**, 467 (1976)].
- <sup>14</sup>J. Dupont-Roc, M. Leduc, and F. Laloë, *J. Phys.* **36**, 961, 977 (1973).
- <sup>15</sup>J. G. Breit and I. I. Rabi, *Phys. Rev.* **38**, 2082 (1931).
- <sup>16</sup>S. P. Dmitriev and A. I. Okunevich, 15th ICPOG, Minsk, 1981, 14–18 July, Abstracts of Contributed Papers, p. 403.
- <sup>17</sup>L. D. Schearer, *Phys. Rev. A* **10**, 1380 (1974).

Translated by J. G. Adashko