Production of polarized hydrogen atoms in ionization collisions of optically oriented 2^3S_1 helium atoms with hydrogen molecules

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We report observation of H and D atom spin polarization produced as a result of ionization collisions between optically oriented metastable helium atoms and H_2 and D_2 molecules. The registration of the polarized atoms was based on the change of the electric conductivity of the plasma at magnetic resonance.

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Optical polarization of hydrogen atoms in the ground state is a complicated problem, since the $1s\rightarrow 2p$ transition is located in the region of the vacuum ultraviolet ($\lambda = 1216$ Å). For this reason, hydrogen atoms have been oriented up to now only by spin exchange with optically oriented alkali-metal atoms (Na, Rb).[1,2] In the present study, the hydrogen atoms were polarized by an entirely different mechanism—ionization of the hydrogen molecules by optically oriented helium atoms in the metastable 2^3S_1 state.

A weak gas discharge was excited in an absorption chamber containing a mixture of He^4 (0.36 Torr) with H_2 or D_2 (0.019 Torr) (at 300 K) and converted part of the helium atoms (10^{10}–10^{11} cm^{-3}) from the ground 1^1S_0 to the metastable 2^3S_1 state. Collisions of the $He^*(2^3S_1)$ atom and of the hydrogen molecule results in production of the quasimolecule He^*–H_2 (or $He^* - D_2$), which is unstable to autoionization because of the large excitation energy of He^* (19.82 eV), and which decays on one of the following manners:

$$He^* + H_2 \longrightarrow \begin{cases} He + H_2^+ + e^- \text{ (Penning ionization)} & \text{(1a)} \\ HeH_2^+ + e^- \text{ (associative ionization)} & \text{(1b)} \\ HeH^+ + H + e^- \text{ (ionization with realignment)} & \text{(1c)} \\ He + H + H^+ + e^- \text{ (dissociative ionization)} & \text{(1d)} \end{cases}$$

The total cross section of the reaction (1) is quite large ($\sigma = 1.5$ Å^2 at 300 K), and this can lead to effective disruption of the metastable state of the helium atoms. At the H_2 and D_2 concentrations indicated above, however, it is still possible to effect optical orientation of the helium atoms in the 2^3S_1 state.[3,4]

One can expect the electron polarization in reaction (1) to be transferred from the helium atoms to the products of this reaction, i.e., to the electrons, to the molecular ions H_2^+ and HeH_2^+, and to the hydrogen atoms. The polarized hydrogen atoms appear in the reactions (1c) and (1d), and the role of the last reaction is apparently insignificant, inasmuch as its cross section is not more than 1% of the total cross section of the reaction (1). At the same time, the cross section σ_r of the reaction of ionization with realignment (1c) is $\sim 15\%$ of the total cross section σ, i.e., $\sigma_r \cong 0.2$ Å^2 (Ref. 5), which yields, at a helium polarization degree on the order of 0.001, more than 10^{10} polarized hydrogen atoms per second.

The experimental investigations were made with a setup similar to that described previously.[6,7] In an absorption chamber containing helium and hydrogen (or deuterium) we produced, simultaneously with optical orientation of the 2^3S_1 helium atoms by circularly polarized light of wavelength $\lambda = 10830$ Å, magnetic resonance at the frequency of the Larmor precession of the hydrogen or deuterium atoms, using low-frequency modulation of the magnetic field. We registered the change of the high-frequency voltage on the absorption-chamber electrodes which were used to excite the discharge.

The registration of the polarized hydrogen atoms is based on the spin dependence of the probability of the reaction

$$He(2\,^3S_1) + H(1\,^2S_{1/2}) \rightarrow He(1\,^1S_0) + H^+ + e^- . \qquad (2)$$

The yield of the free electrons depends in this case on the mutual orientation of the spin angular momenta of H and He*, inasmuch as the reaction (2) with conservation of the total spin is forbidden if the hydrogen and helium spins have the same orientation, but becomes allowed when the orientation of the hydrogen atoms is disrupted by a magnetic resonance, which leads to an increase of the concentration of the electrons at the instant of resonance and to a change of the electric conductivity of the helium–hydrogen plasma.

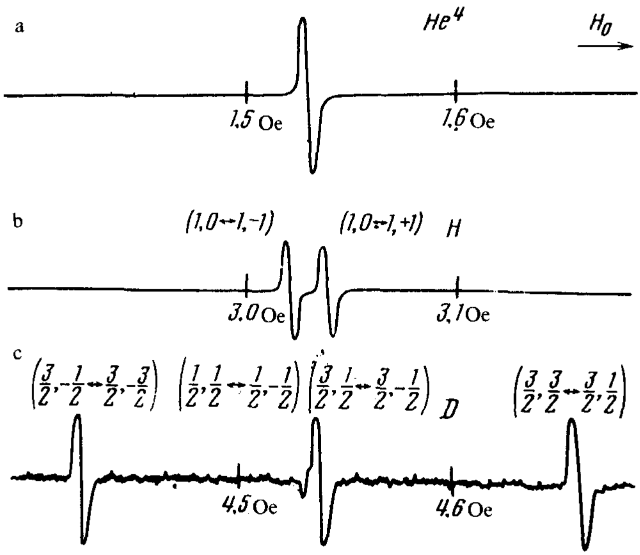

FIG. 1. Signals due to change of the electric conductivity of the plasma in magnetic resonance: (a) In the metastable state 2^3S_1 of the He^4 atoms; (b) in the ground state $1^2S_{1/2}$ of the hydrogen atoms; (c) in the ground state $1^2S_{1/2}$ of the deuterium atoms. The frequency of the RF magnetic field is $f_0 = 4242$ kHz.

Figures 1(b) and 1(c) show the experimentally obtained changes of the electron density at magnetic resonance in the $1^2S_{1/2}$ state of the hydrogen or deuterium atoms. Two lines are observed in the case of hydrogen and four lines in the case of deuterium. The line positions correspond to those calculated by the Breit–Rabi formula for nuclear spins 1/2 (H) and 1(D). Figure 1(a) shows for comparison the change of the electron density in resonance at the Larmor frequency of the He* atoms. The relative changes of the high-frequency voltage on the electrodes were 10^{-4}, 3×10^{-5}, and 2×10^{-6} for He^4, H, and D, respectively.

The complicated shape of the signal at the center of Fig. 1(c) is due to superposition of two incompletely resolved signals of opposite polarity, corresponding to the transitions $F=1/2$, $m_F = +1/2 \leftrightarrow F=1/2$, $m_F = -1/2$ and $F=3/2$, $m_F = +1/2 \leftrightarrow F=3/2$, $m_F = -1/2$. The signals in Fig. 1 correspond to a decrease of the high-frequency voltage and, consequently, to an increase of the electric conductivity of the plasma at magnetic resonance. An exception is the incompletely resolved signal from the sublevel with $F=1/2$ in deuterium, which corresponds to a decrease of the electric conductivity of the plasma and whose explanation calls for an additional investigation.

It should be noted that the polarized hydrogen atoms can appear also in a reaction that is secondary relative to the reaction (1a):

$$H_2^+ + H_2 \rightarrow H_3^+ + H \,. \qquad (3)$$

According to Ref. 8, the rate constant of the reaction (3) is quite high ($C = 1.4 \times 10^{-9}$ $cm^3\ sec^{-1}$), so that this reaction makes an appreciable contribution to the production of the hydrogen atoms; the effectiveness of polarization of the hydrogen atoms via the reaction (3) should decrease substantially as a result of the presence of a coupling between the electron spin and the rotational motion of the molecular ion H_2^+, a coupling that leads to disorientation of the spin angular momentum.

Polarization of the hydrogen atom can be produced, besides in reactions (1) and (3), also in collisions of hydrogen atoms and molecules with other polarized particles in the discharge. Estimates have shown that these processes contribute little to the electron polarization of the hydrogen atoms compared with reaction (1c).

We have thus established that hydrogen atoms become polarized by ionization collisions between optically oriented metastable helium atoms and hydrogen molecules. According to estimates, the main contribution to the polarization is due to the reaction (1c) of ionization with realignment.

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