Production of HD Molecules in Definite Hyperfine Substates

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Polarized atomic beam sources have been in operation for many years to produce either nuclear polarized atomic hydrogen or deuterium beams. In recent experiments, such a source was used to polarize both isotopes independently at the same time. By recombination of the atoms, hydrogen-deuterium molecules with all possible nuclear spin combinations can be created. Those spin isomers are useful for further applications, like precision spectroscopy, as polarized targets for laser-particle acceleration, polarized fuel for fusion reactors, or as an option for future measurements of electric dipole moments.

Introduction.—The Rabi apparatus [1] is the basis to build modern polarized atomic beam sources (ABSs). Their development started in 1956 [2] and has led to a well-established technology. These ABSs produce a beam of hydrogen or deuterium atoms in dedicated hyperfine substates (HFSs) as shown in Table I. Experiments at storage rings use an ABS either to produce polarized ions for further acceleration [3] or to feed polarized internal storage-cell targets [4–8].

During the last years, it was observed that the polarized hydrogen or deuterium atoms in such storage-cell targets can recombine into molecules in which the nuclear polarization is partially preserved [9–11]. We built a dedicated apparatus for further investigations of the different recombination processes on various surfaces. With this setup, it was possible to produce polarized H_2 and D_2 molecules with parallel nuclear spins. Depending on the surface materials, large polarization values above 0.8 have been obtained [12].

Hydrogen molecules are a combination of fermions and exist as orthohydrogen, where both nuclear spins are coupled to a total spin $F = I_1 + I_2 = 1$, and parahydrogen with $F = 0$. Orthohydrogen has a symmetric spin wave function and an antisymmetric contribution due to the rotational magnetic moment J, which must be odd. The $F = 1$ state has three substates with $m_F = +1$, 0 and -1. Parahydrogen with an antisymmetric wave function due to antiparallel proton spins must have even $J = 0, 2, 4, \ldots$. The substates with $m_F = +1$ or $m_F = -1$ are called "pure"

states, because the proton spins of both atoms in the molecule are well defined, i.e., they must be parallel. The other states with $m_F = 0$ are "mixed" states, either with symmetric (ortho) or antisymmetric (para) combinations of the antiparallel substates. Only the pure states are populated when polarized atoms with a defined nuclear spin from an ABS recombine into molecules.

Deuterons are bosons with $I = 1$. Thus, the total wave function of a D_2 molecule must be symmetric. If both spins are parallel and coupled to a total spin $F = 2$, the spin wave

TABLE I. The definition of the different hyperfine substates in the Zeeman region, described with $F = J + I$, and in the Paschen-Back region, described with I and J. For hydrogen atoms in the $1S$ ground state, the critical field amounts to $B_c(\text{H}) = 50.7$ mT and for deuterium to $B_c(\text{D}) = 11.4$ mT.

Isotope	HFS	$B \ll B_c$	$B \gg B_c$
		$\lvert F, m_F \rangle$	$\lvert m_J, m_I \rangle$
H	$\lvert \text{H1} \rangle$	$\lvert 1, 1 \rangle$	$\lvert 1/2, 1/2 \rangle$
	$\lvert \text{H2} \rangle$	$\lvert 1, 0 \rangle$	$\lvert 1/2, -1/2 \rangle$
	$\lvert \text{H3} \rangle$	$\lvert 1, -1 \rangle$	$\lvert -1/2, -1/2 \rangle$
	$\lvert \text{H4} \rangle$	$\lvert 0, 0 \rangle$	$\lvert -1/2, 1/2 \rangle$
D	$\lvert \text{D1} \rangle$	$\lvert 3/2, 3/2 \rangle$	$\lvert 1/2, 1 \rangle$
	$\lvert \text{D2} \rangle$	$\lvert 3/2, 1/2 \rangle$	$\lvert 1/2, 0 \rangle$
	$\lvert \text{D3} \rangle$	$\lvert 3/2, -1/2 \rangle$	$\lvert 1/2, -1 \rangle$
	$\lvert \text{D4} \rangle$	$\lvert 3/2, -3/2 \rangle$	$\lvert -1/2, -1 \rangle$
	$\lvert \text{D5} \rangle$	$\lvert 1/2, -1/2 \rangle$	$\lvert -1/2, 0 \rangle$
	$\lvert \text{D6} \rangle$	$\lvert 1/2, +1/2 \rangle$	$\lvert -1/2, 1 \rangle$

TABLE II. The hyperfine substates of HD molecules described with the total nuclear spin $F = I_p + I_d$ and its projection m_F along an external magnetic field or directly with the single nuclear spins I and their projections m_I.

Isotope	HFS	$\lvert F, m_F\rangle$	$\lvert m_{I_p}, m_{I_d}\rangle$
HD	$\lvert HD1\rangle$	$\lvert 3/2, 3/2\rangle$	$\lvert 1/2, 1\rangle$
	$\lvert HD2\rangle$	$\lvert 3/2, 1/2\rangle$	$\lvert 1/2, 0\rangle$
	$\lvert HD3\rangle$	$\lvert 3/2, -1/2\rangle$	$\lvert -1/2, 0\rangle$
	$\lvert HD4\rangle$	$\lvert 3/2, -3/2\rangle$	$\lvert -1/2, -1\rangle$
	$\lvert HD5\rangle$	$\lvert 1/2, 1/2\rangle$	$\lvert -1/2, 1\rangle$
	$\lvert HD6\rangle$	$\lvert 1/2, -1/2\rangle$	$\lvert 1/2, -1\rangle$

function is symmetric and the rotational moment J must be even. The nine possible combinations of the nuclear spins are divided into six ortho- and three paradeuterium substates. Again, only the substates with $m_F = +2$ or -2 are produced during the recombination of polarized atoms. Contrary to hydrogen it might be possible to freeze out these molecules as deuterium ice with $J = 0$.

For a hydrogen-deuterium (HD) molecule, a combination of a boson and a fermion, these selection rules do not apply. The possible substates are shown in Table II. Here, every substate is a pure state and can have any rotational magnetic moment.

Up until now, polarized solid HD ice has been produced with the "brute force" method and was used as a polarized target, e.g., at the CLAS experiment [13]. One advantage of solid HD is the long lifetime of the nuclear polarization, which is in the order of years. However, the polarization values achieved for the proton ($P_z \approx 0.5$) and the deuteron ($P_z \approx 0.27$) are rather small. Moreover, a control of the tensor polarization P_{zz} is not possible. For many important fundamental applications, the other spin combinations are mandatory and larger polarization values would be helpful. Here we present a new method to produce all possible spin

combinations of the proton and the deuteron in the HD molecule.

If an ABS is operated with a hydrogen and deuterium mixture, a beam containing the corresponding atoms in defined hyperfine substates can be generated. When these atoms recombine into HD molecules, it is possible to produce any spin isomer of the HD molecules, because the nuclear spins of the hydrogen and the deuterium atoms can be adjusted separately.

The apparatus.—Our ABS [5] consists of a dissociator in combination with a cooled nozzle to form an atomic hydrogen beam from molecular gas. After shaping the beam with two apertures, cylindrical sextupole magnets are used to focus the atoms with electron spin $m_J = +1/2$ on the beam axis and to defocus the others (Stern-Gerlach effect). A so-called medium magnetic field transition unit (MFT) to induce magnetic dipole π transitions is utilized between the sextupole magnets to exchange the occupation number of the hyperfine substates $\lvert H2\rangle \to \lvert H3\rangle$, $\lvert H1\rangle \to \lvert H3\rangle$, $\lvert D3\rangle \to \lvert D4\rangle$, or $\lvert D1\rangle \to \lvert D4\rangle$. The following sextupole magnets defocus states $\lvert H3\rangle$ and $\lvert D4\rangle$. A weak field transition unit (WFT; again, a π transition) and a strong field transition unit (SFT; magnetic σ transition), placed after the last sextupole magnet, can induce further transitions, which are listed in Table III. The necessary settings for the rf frequency and the magnetic field of the WFT are very similar for hydrogen and deuterium, thus it can be used to induce the transitions for both isotopes simultaneously.

When these atoms are fed into a T-shaped storage cell at temperatures between 40 and 120 K, they can recombine into molecules (see Fig. 1). The ratio of the flux of hydrogen and deuterium into the ABS was tuned to be $2/3$ to obtain an equal amount of hydrogen and deuterium atoms from the ABS in the recombination chamber. In this case, the maximum amount of HD molecules and roughly equal amounts of H_2 and D_2 molecules are produced.

TABLE III. The settings of the ABS transition units to get an atomic beam of hydrogen and deuterium with dedicated nuclear spins to produce all different spin isomers of HD molecules.

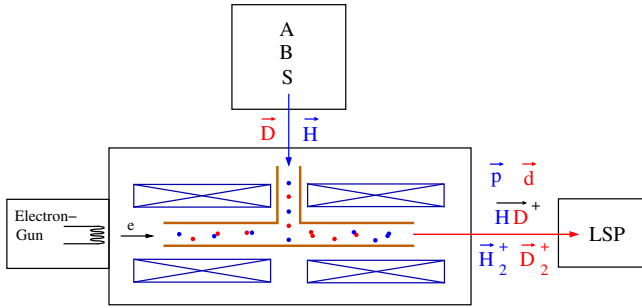| Transition unit	H and D Transition units	Atomic beam	HFSs of HD molecules
MFT: $\lvert D3\rangle \to \lvert D4\rangle$	SFT: $\lvert H2\rangle \to \lvert H4\rangle$ SFT: $\lvert D2\rangle \to \lvert D6\rangle$	$\lvert H1\rangle + \lvert H4\rangle$ $\lvert D1\rangle + \lvert D6\rangle$	$\lvert HD1\rangle$
MFT: $\lvert D1\rangle \to \lvert D4\rangle$	SFT: $\lvert H2\rangle \to \lvert H4\rangle$ SFT: $\lvert D3\rangle \to \lvert D5\rangle$	$\lvert H1\rangle + \lvert H4\rangle$ $\lvert D2\rangle + \lvert D5\rangle$	$\lvert HD2\rangle$
MFT: $\lvert D1\rangle \to \lvert D4\rangle$	WFT: $\lvert H1\rangle + \lvert H2\rangle \to \lvert H2\rangle + \lvert H3\rangle$ SFT: $\lvert D3\rangle \to \lvert D5\rangle$	$\lvert H2\rangle + \lvert H3\rangle$ $\lvert D2\rangle + \lvert D5\rangle$	$\lvert HD3\rangle$
MFT: $\lvert D3\rangle \to \lvert D4\rangle$	WFT: $\lvert H1\rangle + \lvert H2\rangle \to \lvert H2\rangle + \lvert H3\rangle$ WFT: $\lvert D1\rangle + \lvert D2\rangle \to \lvert D3\rangle + \lvert D4\rangle$	$\lvert H2\rangle + \lvert H3\rangle$ $\lvert D3\rangle + \lvert D4\rangle$	$\lvert HD4\rangle$
MFT: $\lvert D3\rangle \to \lvert D4\rangle$	WFT: $\lvert H1\rangle + \lvert H2\rangle \to \lvert H2\rangle + \lvert H3\rangle$ SFT: $\lvert D2\rangle \to \lvert D6\rangle$	$\lvert H2\rangle + \lvert H3\rangle$ $\lvert D1\rangle + \lvert D6\rangle$	$\lvert HD5\rangle$
MFT: $\lvert D3\rangle \to \lvert D4\rangle$	SFT: $\lvert H2\rangle \to \lvert H4\rangle$ WFT: $\lvert D1\rangle + \lvert D2\rangle \to \lvert D3\rangle + \lvert D4\rangle$	$\lvert H1\rangle + \lvert H4\rangle$ $\lvert D3\rangle + \lvert D4\rangle$	$\lvert HD6\rangle$

FIG. 1. Experimental setup taken from Ref. [12] with minor modifications: A storage cell inside a superconducting magnet is fed by the polarized H and D beam from the ABS. Atoms and molecules from recombination are ionized by electron impact. The cell is set to a potential up to +2 keV to accelerate the protons or deuterons and molecular ions into the LSP, which allows one to measure the polarization.

A longitudinal magnetic field along the cell, induced by a superconducting solenoid, increases the lifetime and the nuclear polarization is preserved for several milliseconds, while the molecules stay inside the storage cell. This magnetic field couples to the nuclear spins and, therefore, the spin-spin coupling of the nucleons itself and their coupling with the rotational magnetic moment of the molecule J are suppressed, which are the reasons for depolarization during wall collisions [12]. Up to 50% of the atomic polarization is found in these molecules after the recombination on a gold surface and a magnetic field higher than 0.3 T. A Fomblin surface allows for full vector-polarization preservation of deuterium atoms at 0.5 T. An electron beam is then sent through the cell to ionize the atoms and molecules. The corresponding ions, i.e., protons, deuterons, H_2^+, D_2^+, and HD^+, are accelerated to an energy of 1–2 keV by a positive potential on the gold surface [12].

All ions with the same kinetic energy reach the first component of the Lamb-shift polarimeter (LSP), i.e., a Wien filter that is used in two different ways: It is able to separate the different velocities, i.e., the different ions according to their masses. Only deuterons and H_2^+ ions, both with mass 2, cannot be distinguished. At the same time, the transversal magnetic field induces a Larmor precession of the longitudinally oriented nuclear spin. If the magnetic field is chosen in a way that the time of flight of the ions through the Wien filter allows a spin rotation of 180°, the orientation of the spin direction is changed, but not its projection onto the beam axis. The coupling of the nuclear spins to the spin of the single electron of the molecular ions is responsible for an adiabatic realignment along the radial magnetic field. Afterward, the ions undergo a charge exchange with cesium vapor to form metastable atoms in the first excited state $2S$. A strong magnetic field along this cesium cell reorients the spins of the molecular ions back on the beam direction and preserves the nuclear

polarization, until the field is much larger than the critical field of these metastable atoms ($B_c = 6.34$ mT for hydrogen and $B_c = 1.54$ mT for deuterium). With a longitudinal magnetic field around 57 mT, a radial electric field of about 10 V/cm and a radio frequency of 1.609 75 GHz inside a cavity in the TM_{010} mode, the spin filter is able to quench all metastable atoms into the ground state. Only at special resonances at different magnetic fields can metastable atoms in single HFSs pass through. In the last section, these excited atoms are quenched into the ground state with a strong electric field (Stark effect) and the produced Lyman-α photons are registered with a photomultiplier tube (PMT) as a function of the magnetic field in the spin filter. The amount of measured photons N_+ and N_- at the resonances correspond linearly to the number of incoming ions with the same nuclear spin. Therefore, the nuclear vector-polarization P_z of the protons or deuterons can be deduced by comparing the photon numbers $P_z = (N_+ - N_-)/(N_+ + N_-)$ [14,15]. The resonances of the HFSs of the metastable hydrogen and deuterium are well separated. Thus, even when the Wien filter is not able to separate H_2^+ ions and deuterons, or when HD^+ molecules will reach the cesium cell, the LSP can measure the nuclear polarization of both nucleons independently.

Experimental results.—The standard settings of our ABS allow only the production of the spin isomer $|HD4\rangle$; i.e., the proton and deuteron spin are antiparallel to the quantization axis (see Table III). In this case, the first set of sextupole magnets will defocus the hydrogen atoms in the HFSs $|H3\rangle$ and $|H4\rangle$ and the deuterium atoms in the substates $|D4\rangle$, $|D5\rangle$, and $|D6\rangle$. The MFT is used to transfer deuterium atoms from HFS $|D3\rangle$ into $|D4\rangle$. The next set of sextupole magnets defocuses the deuterium atoms in state $|D4\rangle$ again, so that only the HFSs $|H1\rangle$, $|H2\rangle$, $|D1\rangle$, and $|D2\rangle$ are left in
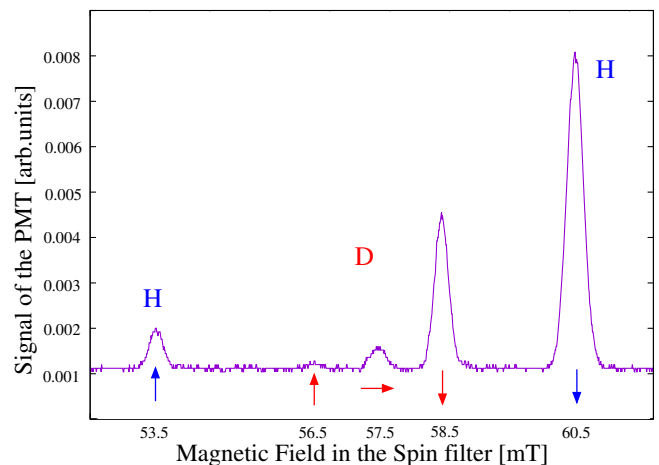

FIG. 2. Lyman-α spectrum of HD molecules in the spin isomer $|HD4\rangle$ after recombination on a Fomblin surface. The nuclear polarization deduced from this spectra is $P_z = -0.77 \pm 0.01$ for hydrogen and $P_z = -0.79 \pm 0.01$ and $P_{zz} = +0.69 \pm 0.01$ for deuterium.
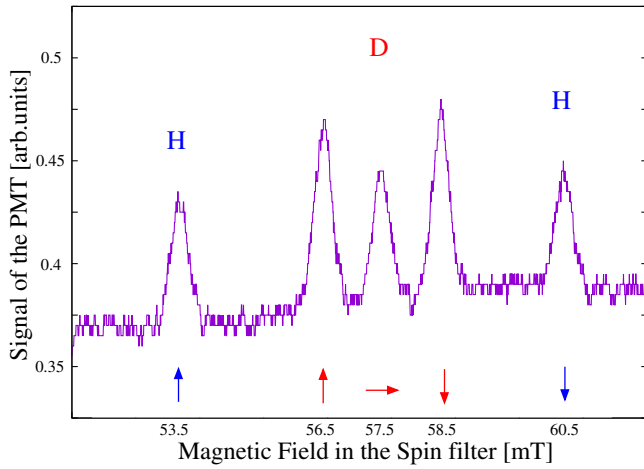
FIG. 3. Lyman-α spectrum of HD molecules when unpolarized hydrogen atoms recombine on a gold surface mostly with deuterium atoms in the HFS $|D3\rangle$ or $|D6\rangle$. The protons are unpolarized and the deuterons with $m_I = 0$ are suppressed. The observed tensor-polarization of $P_{zz} = +0.22 \pm 0.02$ is rather low due to some technical problems with one transition unit of the ABS during these measurements.

the beam. Now, the WFT will induce the corresponding transitions for hydrogen and deuterium at the same time ($|H1\rangle + |H2\rangle \rightarrow |H2\rangle + |H3\rangle$ and $|D1\rangle + |D2\rangle \rightarrow |D3\rangle + |D4\rangle$). Thus, the beam includes mostly atoms with antiparallel nuclear spins, i.e., the HFSs $|H2\rangle + |H3\rangle$ for hydrogen and $|D3\rangle + |D4\rangle$ for deuterium. These atoms recombine on a Fomblin (perfluoropolyether) surface into H_2, D_2, and HD molecules. After ionization by electron impact, acceleration of the molecular ions into the LSP and mass separation by the Wien filter, the polarization of the HD$^+$ ions is deduced from the Lyman-α spectra (see Fig. 2). The polarization can be calculated from the peak ratios to $P_z = -0.77 \pm 0.01$ for hydrogen and $P_z = -0.79 \pm 0.01$ and $P_{zz} = +0.69 \pm 0.01$ for deuterium.

The necessary settings for the production of the other spin isomers of HD molecules are listed in Table III. With the simultaneous use of a WFT and a SFT, or two SFTs, all possible spin directions of the proton and the deuteron are possible and in range after minor modifications of the existing ABS.

Instead of Fomblin, most measurements were carried out on a gold surface. As expected from Ref. [12], Fomblin allows twice as large polarization values for the molecules, because this surface preserves the nuclear polarization during the recombination process. Nevertheless, most measurements are made with a gold surface, because the use of Fomblin oil pollutes all surfaces in the vacuum chamber and reduces the pumping speed of the internal cryopanels by orders of magnitude.

When the MFT transfers HFS $|D1\rangle$ into $|D4\rangle$ and the SFT is used at the end, it is possible to produce only tensor-polarized deuterium. If the SFT transfers $|D2\rangle \rightarrow |D6\rangle$ (σ transition), the deuterium atoms are in the HFSs
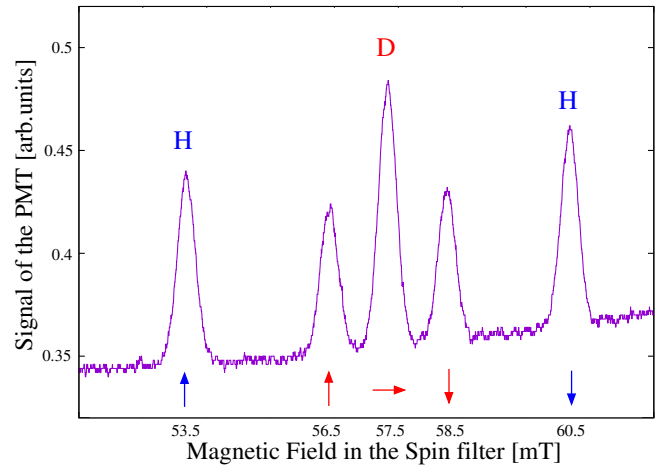

FIG. 4. Lyman-α spectrum of HD molecules when unpolarized hydrogen atoms recombine on a gold surface mostly with deuterium atoms in $|D2\rangle$ and $|D5\rangle$. The protons are unpolarized and the deuteron spin is perpendicular to the quantization axis with a tensor-polarization of $P_{zz} = -0.61 \pm 0.01$.

$|D3\rangle$ and $|D6\rangle$. Recombination with unpolarized hydrogen atoms gives equal amounts of the substates $|HD1\rangle$, $|HD4\rangle$, $|HD5\rangle$, and $|HD6\rangle$, i.e., only states with positive tensor-polarization of the deuteron (see Fig. 3). Negative tensor-polarization can be produced if the SFT $|D3\rangle \rightarrow |D5\rangle$ (σ transition) is induced to get $|D2\rangle$ and $|D5\rangle$. These atoms can recombine with unpolarized hydrogen to produce the states $|HD2\rangle$ and $|HD3\rangle$ with equal amounts (Fig. 4).

Discussion and outlook.—As in previous works on the production of polarized H_2 and D_2 molecules due to the recombination of polarized H or D atoms, the creation of polarized HD molecules in single spin isomers was investigated. Depending on the spin states of the different isotopes, defined with the corresponding settings of the ABS, all possible combinations of the proton and deuteron spin are accessible with an ABS.

In former polarization measurements of the H_2 and D_2 molecules recombined on a gold surface, polarization values up to $P_z = 0.45$ were observed, about 50% of the atomic polarization [12]. During these measurements, the polarization of the H_2, the D_2, and the nucleons in the HD molecules does not exceed 0.35. The reason is still not completely understood, but seems to depend on the production method of the self-made gold cells that had to be changed recently.

Compared to the measurements with protons and deuterons at the entrance of the cesium cell, the molecular ions have more options to interact with the cesium. For example, the stripping process into two protons or the charge exchange into fast molecules are responsible for a larger background in the Lyman-α spectra. The interaction of the ions in the beam with the magnetic field of the spin filter is responsible for the increase of the background at higher magnetic fields.

In the next step, the polarized D_2 and HD molecules will be frozen as polarized ice on a cold surface inside a strong magnetic field, exploiting the higher evaporation temperature of the H_2 molecules. The goal is to build polarized targets for laser-acceleration experiments to produce polarized proton and deuteron beams [16].

Polarized H_2^+ and HD^+ ions in single spin isomers are a perfect tool for precision spectroscopy of these most simple molecular ions, because the influence of the spin-dependent part of the transition frequencies can be determined separately [17].

Another option to use these molecules can be the search for a static electric dipole moment (EDM) with oscillating electric fields [18] or even experiments searching for an oscillating nuclear EDM produced by the axion dark matter field [19]. For both methods, the use of highly polarized HD molecules is beneficial, because the EDM effect would be proportional to the inverse nucleon charge z^{-1} that is at minimum for the hydrogen isotopes.

Another promising application of nuclear polarized hydrogen molecules and their isotopes would be the use as polarized fuel for fusion reactors [20]. Besides D_2 molecules, polarized DT would be needed, either with parallel nuclear spins (Fig. 2) to increase the reaction rate of the fusion reactions or with only tensor-polarized deuterons (Fig. 4) to focus the neutrons on special blanket areas, e.g., to increase the lifetime of the blanket or to breed new tritium via the $^6Li + n \rightarrow {}^4He + t$ reaction. To avoid the radioactive tritium, the HD molecules are a perfect training ground to study the production, storage, and handling of these molecules for further use, because the hyperfine structures of HD and DT are almost identical.

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