Dynamic Polarization of Electron Spins Interacting with Nuclei in Semiconductor Nanostructures

D. S. Smirnov,[1,*] T. S. Shamirzaev,[2,3,†] D. R. Yakovlev,[4,1] and M. Bayer[4,1]
[1]Ioffe Institute, Russian Academy of Sciences, 194021 St. Petersburg, Russia
[2]Rzhanov Institute of Semiconductor Physics, Siberian Branch of the Russian Academy of Sciences, 630090 Novosibirsk, Russia
[3]Ural Federal University, 620002 Yekaterinburg, Russia
[4]Experimentelle Physik 2, Technische Universität Dortmund, 44221 Dortmund, Germany

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We suggest a new spin orientation mechanism for localized electrons: dynamic electron spin polarization provided by nuclear spin fluctuations. The detrimental effect of nuclear spin fluctuations can be harnessed and employed to provide angular momentum for the electrons via the hyperfine interaction in a weak magnetic field. For this, the sample is illuminated by an unpolarized light, which directly polarizes neither the electrons nor the nuclei. We predict that, for the electrons bound in localized excitons, 100% spin polarization can be reached in longitudinal magnetic fields of a few millitesla. The proof of principle experiment is performed on momentum-indirect excitons in (In, Al)As/AlAs quantum dots, where in a magnetic field of 17 mT the electron spin polarization of 30% is measured.

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As the quantum computation era is believed to approach [1], the investigations of the underlying physics drastically intensify. Particular attention is focused on the spin dynamics of localized electrons in semiconductor nanostructures [2], which is at the heart of various quantum computation and quantum cryptography schemes [3,4]. Ultrafast optical orientation [5–7], manipulation [8–10] and readout [11–13] were already demonstrated for the single electrons confined in quantum dots (QDs).

There are two main approaches to spin orientation in nanostructures: optical spin orientation [14] and thermal spin polarization in magnetic field. The first one does not require the external magnetic field as it is based on the transfer of the angular momentum from circularly polarized photons to electrons through the spin-orbit interaction. The second approach does not require optical excitation. It needs lowering of the lattice temperature, so that the thermal energy becomes smaller than the electron Zeeman splitting.

In this Letter, we suggest another approach, which we call the dynamic electron spin orientation. Generally, this concept applies to localized electrons exposed to unpolarized optical excitation. It requires (i) a fine structure splitting of a photogenerated electron-hole pair with different lifetimes of the individual levels and (ii) that these levels are mixed by the random Overhauser field, i.e., the local magnetic field caused by the fluctuations of the host lattice nuclear spins. Previously, the nuclear spin fluctuations were known to be the source of spin relaxation only, while here we demonstrate that they can act as a source of angular momentum for the charge carriers. In III-V and II-VI semiconductors, the hyperfine interaction with nuclei is most pronounced for electrons, while for holes it is an order of magnitude weaker [2]. The typical value of the random Overhauser field scales with the localization volume V as $1/\sqrt{V}$. Therefore, for delocalized or weakly bound states the hyperfine interaction is negligible, while for electrons in QDs it plays the main role in the spin dynamics. We predict that application of a weak external magnetic field of the order of the random Overhauser field (a few millitesla) induces an electron spin polarization that can reach 100%.

In contrast to the optical spin orientation, the proposed mechanism does not require circular polarization of the optical excitation. In contrast to the thermal spin polarization, we consider weak magnetic fields, for which the electron Zeeman splitting is much smaller than the thermal energy. Our approach does not require the resonant excitation of specific states, e.g., as in spin orientation protocols with Λ scheme [15]. The dynamic electron spin polarization is based on the violation of the detailed balance (the equality of the rates of the direct and reverse processes) between spin flips in nonequilibrium conditions in weak magnetic fields. We call this effect dynamic electron spin polarization, in similarity with the dynamic nuclear spin polarization gained in nonequilibrium conditions in weak magnetic fields [16,17].

The proposed concept is applicable to various systems. Here, we theoretically describe and experimentally demonstrate the dynamic spin polarization of electrons in excitons. It is most pronounced for excitons with the long lifetimes and small splittings between bright and dark states. These conditions are valid for the excitons that are indirect either in real or in momentum space. As a test bed we use momentum-indirect excitons confined in

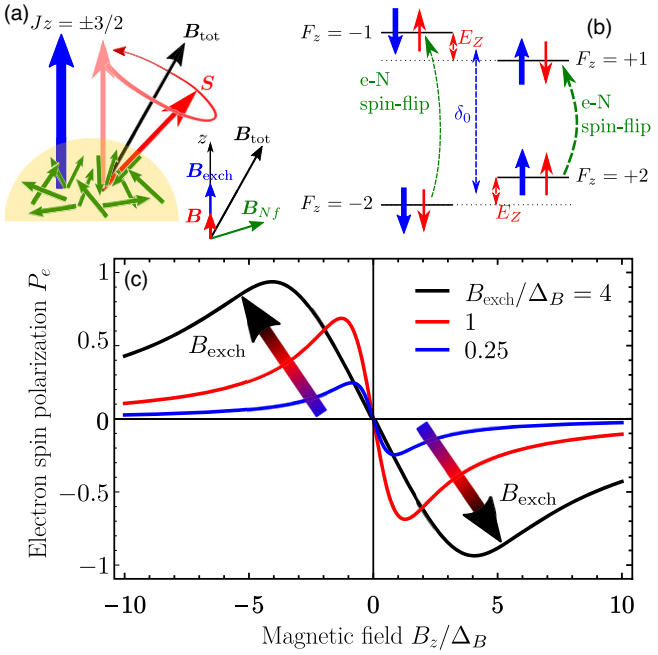

FIG. 1. (a) Sketch of the QD (yellow) with randomly oriented nuclear spins (green arrows) and the spins of electron (red) and hole (blue) in a localized exciton. The electron spin precesses in the total field $\boldsymbol{B}_{\rm tot}$. (b) Fine structure of the exciton levels with electron and heavy hole spins denoted by red and blue arrows, respectively (for $g_h = 0$ and $B_{\rm exch} > B_z \gg \Delta_B$). The splitting of the bright ($F_z = \pm 1$) and dark ($F_z = \pm 2$) states due to the exchange interaction is changed in the external magnetic field by $E_Z = g_e \mu_B B_z$. As a result, the rates of nuclei-assisted spin flips in these pairs of states (green arrows) are different. (c) Electron spin polarization degree as a function of the external magnetic field for the different exchange interaction strengths indicated in the legend, calculated after Eq. (S8) with the parameters $g_e \mu_B \Delta_B \tau_b/\hbar = 10^3$ and $\tau_s^e/\tau_b = 10^3$, see Supplemental Material [20].

(In, Al)As/AlAs QDs, where we get an electron spin polarization of 30% in a magnetic field of 17 mT.

Microscopic mechanism.—We consider an exciton localized in a QD, which consists of an electron with spin projection on the growth z axis $S_z = \pm 1/2$ and the heavy hole with spin $J_z = \pm 3/2$ [18] [Fig. 1(a)]. We neglect the possible valley degeneracy of the states as well as the interaction between the excitons. The exciton Hamiltonian in the external longitudinal magnetic field $\boldsymbol{B} = (0, 0, B_z)$ (Faraday geometry) has the form

$$\mathcal{H} = g_e \mu_B (\boldsymbol{B} + \boldsymbol{B}_{Nf}) \boldsymbol{S} + g_h \mu_B B_z J_z - \frac{2}{3} \delta_0 S_z J_z. \quad (1)$$

Here g_e and g_h are the electron and hole longitudinal g factors, respectively, μ_B is the Bohr magneton, $\boldsymbol{B}_{Nf}$ is the random Overhauser field, and $\boldsymbol{S}$ is the spin of electron in exciton. The exchange interaction splits the four exciton states by δ_0 into two upper bright states with the total spin $F_z = S_z + J_z = \pm 1$ and the lower dark ones with $F_z = \pm 2$. The exciton fine structure is sketched in Fig. 1(b). The bright excitons can radiatively recombine and have the lifetime τ_b, while the radiative recombination of the dark excitons is spin forbidden. We account for the short-range exchange interaction only, while the long-range one can lead to a splitting between the linearly polarized excitonic states, which would suppress the dynamic electron spin polarization. We also neglect the hole hyperfine interaction, which is small due to the p type of the Bloch wave functions [2]. The key difference of our model with the standard description of the exciton states [18,19] is the hyperfine interaction of electrons in excitons with nuclei, which can be comparable with the exciton exchange splitting δ_0.

The typical timescale of the nuclear spin dynamics is milliseconds, so $\boldsymbol{B}_{Nf}$ can be considered as "frozen" for short times [2,45,46]. This makes the electron spin relaxation non-Markovian (due to the long nuclear spin memory time), which, as we demonstrate below, leads to the dynamic electron spin polarization. The electron spin precesses in the total magnetic field $\boldsymbol{B}_{\rm tot} = \boldsymbol{B} + \boldsymbol{B}_{Nf} + \boldsymbol{B}_{\rm exch}$, see Fig. 1(a). Here the exchange field $\boldsymbol{B}_{\rm exch}$ is directed along the heavy hole spin quantization axis z: $B_{{\rm exch},z} = -(2/3)\delta_0 J_z/(g_e \mu_B)$ [47]. The electron spin precession can be described as a classical precession of the magnetic moment, but the exchange field $\boldsymbol{B}_{\rm exch}$ is essentially quantum. It cannot be described in the mean field approach, but must be treated as a quantum operator with the two eigenvalues with the opposite signs [see, e.g., Eqs. (2) below].

The incoherent processes in the exciton spin dynamics can be described in the density matrix formalism. For simplicity, we consider only two such processes: exciton nonresonant generation with the rate G and bright exciton recombination with the time τ_b. The more elaborate model of the spin dynamics is described in the Supplemental Material [20]. Moreover, we assume that the electron spin precesses around $\boldsymbol{B}_{\rm tot}$ much faster than the bright exciton recombines, so the average electron spin is parallel to $\boldsymbol{B}_{\rm tot}$. We denote the total occupancies of the states with $F_z = +2, +1$ $(-2, -1)$ as N^+ (N^-) and introduce the average electron spins $S_\parallel^\pm$ in the corresponding Hilbert spaces. The kinetic equations for these quantities read [20]

$$\frac{dN^\pm}{dt} = \frac{G}{2} - \frac{N^\pm}{2\tau_b} \pm \frac{S_\parallel^\pm \cos(\theta_\pm)}{\tau_b}, \quad (2a)$$

$$\frac{dS_\parallel^\pm}{dt} = -\frac{S_\parallel^\pm}{2\tau_b} \pm \frac{N^\pm \cos(\theta_\pm)}{4\tau_b}, \quad (2b)$$

where $\theta_\pm$ are the angles between $\boldsymbol{B}_{\rm tot}$ and the z axis in the corresponding Hilbert spaces.

In the steady state we solve Eqs. (2) and obtain the average electron spin along the z axis $S_z = \sum_\pm S_\parallel^\pm \cos(\theta_\pm)$. For a

QD ensemble, the spin polarization has to be averaged over the Gaussian distribution function of the random Overhauser field $\propto \exp(-B_{Nf}^2/\Delta_B^2)$, with Δ_B describing the dispersion [45]. Δ_B depends only on the hyperfine interaction constant and QD volume. After averaging we obtain the simple expression for the degree of the dynamic electron spin polarization [see Eq. (S6) in the Supplemental Material [20]]

$$P_e = \frac{-2B_z B_{\text{exch}}}{B_{\text{exch}}^2 + \Delta_B^2/2 + B_z^2}. \tag{3}$$

This is the main theoretical result of this Letter.

In Fig. 1(c) we show the electron spin polarization as a function of B_z calculated in the extended model accounting for the finite bright and dark exciton lifetimes [20]. It is noteworthy that it agrees with the simple Eq. (3) within 25% accuracy. Generally, the electron spin polarization is an odd function of B_z in agreement with the time reversal symmetry. The polarization reaches maximum at $B_z \approx \sqrt{\Delta_B^2/2 + B_{\text{exch}}^2}$ and vanishes in large magnetic fields. This is in stark contrast with the thermal spin polarization, which monotonically saturates in strong fields. We note that the fact that P_e stays finite in the limit of small random Overhauser field $\Delta_B \to 0$ is related with the above-mentioned assumption, that the spin precession frequency in $\boldsymbol{B}_{\text{tot}}$ is faster than the bright exciton recombination. If the exchange splitting exceeds the typical hyperfine interaction energy (or, equivalently, $B_{\text{exch}} \gtrsim \Delta_B$), then the electron spin polarization can approach 100%.

Let us qualitatively describe the origin of the dynamic electron spin polarization. The bright excitons recombine during the characteristic time τ_b, while the dark excitons can recombine only due to the nuclei-assisted mixing with the bright states. The transverse components of the random Overhauser field $B_{Nf,x}$ and $B_{Nf,y}$ lead to the electron-nuclear spin flips. The larger the energy difference between bright and dark states, the smaller the mixing. Note that this is in contrast with the phonon-assisted spin relaxation and is a direct consequence of the non-Markovian spin relaxation. Thus, one can see from Fig. 1(b) that the mixing is different for the dark states with $F_z = -2$ and $+2$, so one of them recombines faster. It is noteworthy that this requires both exchange splitting between bright and dark states and the longitudinal magnetic field [20]. The difference in the lifetimes of the exciton states with spin-up and spin-down electron results in the dynamic polarization of electron spin.

If the bright exciton lifetime is shorter than the typical spin precession period in the random Overhauser field, $\tau_b g_e \mu_B \Delta_B/\hbar \ll 1$, then the bright exciton states have large homogeneous broadening, and this leads to the suppression of dynamic electron spin polarization. In the opposite limit, we can use perturbation theory to calculate the lifetimes of the dark excitons with $F_z = \pm 2$,

$$\frac{1}{\tau_{\pm 2}} = \frac{1}{\tau_b} \frac{B_{Nf,x}^2 + B_{Nf,y}^2}{(B_z + B_{Nf,z} \mp B_{\text{exch}})^2}. \tag{4}$$

The smaller $B_{Nf,x}$ and $B_{Nf,y}$, the longer $\tau_{\pm 2}$, so they can be much longer than τ_b. In the steady state, the occupancies of the dark states are $N_{\pm 2} = (G/4)\tau_{\pm 2}$, which are on average much larger than the occupancies of the bright states. As a result, the polarization degree of electron spins is $(N_{+2} - N_{-2})/(N_{+2} + N_{-2})$, which yields Eq. (3). The spin polarization degrees of electrons, holes, and excitons in this case coincide.

From the above derivation it follows that, for pulsed excitation, the dynamic polarization will arise not immediately, but with a delay, e.g., only after the recombination of the bright excitons. This is in stark contrast with the usual optical orientation.

To summarize the theory predictions, the dynamic electron spin polarization for excitons requires (i) the exciton lifetime to be longer than the typical electron spin precession period in the random Overhauser field, $\tau_b g_e \mu_B \Delta_B/\hbar > 1$ and (ii) the exchange interaction between electron and hole to be smaller than the thermal energy. Otherwise, the dynamic electron spin polarization mechanism will smoothly transform into the thermal spin polarization [20,48]. Both requirements can be met using separation between electron and hole either in real or in momentum space.

Experiment.—For experimental demonstration of the suggested mechanism, we choose the momentum-indirect (In, Al)As/AlAs QDs. Recently, we showed that in these QDs at low temperatures the exciton spin relaxation is dominated by the hyperfine interaction with Δ_B being a few millitesla [49], while the exciton lifetime reaches hundreds of microseconds [50,51]. The QDs have type-I band alignment (both electron and hole are localized inside the QD) [52,53]. In large QDs, the lowest electron and hole states are in the Γ valley, so the excitons are momentum direct [20], but with the decrease of the QD size the Γ valley of the conduction band shifts to higher energies faster than the X valley, due to the smaller effective mass and the strain [53]. As a result, the electron ground state in small QDs is in the X valley [see inset in Fig. 2(a)], so the excitons in these QDs are momentum indirect. The Hamiltonian (1) is valid in this case, provided the splitting between the linearly polarized bright excitonic states can be neglected. The excitons have finite radiative lifetime due to their mixing with the direct excitons at QD interfaces. The spectral distribution of exciton lifetimes allows us to identify the indirect QDs in the inhomogeneous ensemble [20]. For photoluminescence (PL) studies we used nonresonant pulsed optical excitation at 3.49 eV by linearly polarized light. In absence of the magnetic field, the exciton PL is unpolarized.

To dynamically polarize electron spins, we apply the longitudinal magnetic field of 17 mT (Faraday geometry).

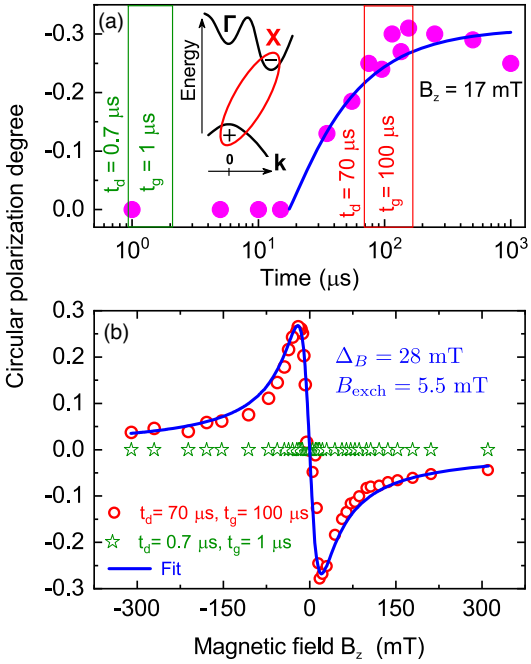

FIG. 2. (a) Dynamics of PL circular polarization degree measured at $B_z = 17$ mT and $T = 2$ K; the integration time is 5 μs. Vertical lines show time-integration windows for (b). Blue line is a fit after Eqs. (2) with parameters $\Delta_B = 28$ mT, $B_{\text{exch}} = 6.6$ mT, and $\tau_b = 2$ μs. (Inset) The band diagram of the momentum-indirect (In, Al)As/AlAs QDs. (b) Magnetic field dependencies of the polarization degree measured at 0.7 (green stars) and 70 μs (red circles) with the integration windows of 1 and 100 μs, respectively. Blue line is a fit after Eq. (3) with $\Delta_B = 28$ and $B_{\text{exch}} = 5.5$ mT.

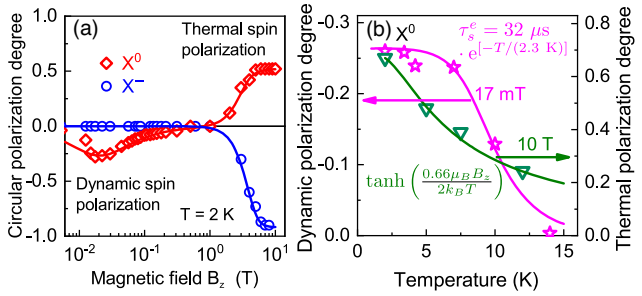

FIG. 3. (a) PL circular polarization degree as a function of the longitudinal magnetic field for excitons (X^0) and negatively charged trions (X^-). (b) Temperature dependence of the dynamic polarization of excitons measured in time window 70–170 μs at $B_z = 17$ mT (magenta stars) and the thermal polarization at $B_z = 10$ T (green triangles). The fit details for (a) and (b) are described in the Supplemental Material [20]. In particular, the magenta curve in (b) is calculated using the same parameters as for the fit in Fig. 2(a) and an activation law for the electron spin relaxation time given in the panel.

In Fig. 2(a) we show the PL circular polarization as a function of time, detected at the energy of 1.70 eV (see the Supplemental Material [20] for details). The degree of circular polarization is defined as $P_c = (I_+ - I_-)/(I_+ + I_-)$, where $I_\pm$ are the intensities of $\sigma^\pm$ polarized emission. The polarization appears with a delay of 15 μs after the pump pulse and saturates after 100 μs. It is in line with model prediction that the dynamic polarization appears only after recombination of the bright excitons. It is noteworthy that the PL stays polarized up to 1 ms.

The magnetic field dependence of the dynamic polarization integrated for two time windows is shown in Fig. 2(b). This is the main experimental result of this Letter. The absolute value of $P_c(B_z)$ increases in weak fields, reaches maximum of about 0.3 at $B_z = 17$ mT, and then monotonously decreases tending to zero in high fields. Fitting this dependence with Eq. (3) we find two parameters: $\Delta_B = 28$ and $B_{\text{exch}} = 5.5$ mT. The strength of the hyperfine interaction is in good agreement with measurements of optical spin orientation in transverse and longitudinal magnetic fields in a similar sample [49,54], which supports our interpretation. Using the electron g factor $g_e = 2$ [55,56], we find the splitting between bright and dark states $\delta_0 = 0.6$ μeV. Note that for momentum-indirect excitons the long-range exchange interaction is suppressed, because it can be described as a result of the virtual electron-hole recombination. However, the short-range interaction is not expected to be suppressed, so we additionally verified the small value of the short-range exchange interaction constant measuring the PL dynamics in weak transverse magnetic fields [20].

To evidence the role of the thermal spin polarization, we measured the polarized PL in magnetic fields up to 10 T. The results are shown by the red diamonds in Fig. 3(a). One can see that the dynamic electron spin polarization takes place in fields ~10 mT only, while the thermal polarization appears in fields larger than 1 T and has the opposite (positive) sign.

The blue circles in Fig. 3(a) show for comparison the PL polarization for an ensemble of negatively charged (In, Al)As/AlAs QDs; see details in the Supplemental Material [20]. Photoexcitation of singly charged QDs generates negatively charged excitons (trions) [51]. In its ground singlet state, the electron-hole exchange interaction is absent and, therefore, dynamic electron spin polarization in weak fields does not form, in agreement with the theory. At the same time, the thermal spin polarization has the negative sign [51], opposite to the exciton thermal polarization. Note that the excitation of the triplet trion state would allow one to observe the dynamic electron spin polarization for trions and to transfer it to the resident charge carriers, as we show in the Supplemental Material [20].

Additionally, dynamic and thermal spin polarization differ by their temporal dynamics [20] and temperature dependencies. The dynamic polarization is temperature independent, as long as the electron spin relaxation is dominated by the hyperfine interaction. By contrast, the

thermal polarization is controlled by the factor E_Z/k_BT and decreases with raising temperature. This is experimentally proven in Fig. 3(b), where the thermal polarization at 10 T (green line) decays rapidly with temperature increase, while the dynamic polarization is constant in the range 2–7 K. We assume that with further temperature increase the phonon-assisted electron spin relaxation time τ_s^e shortens, so the dynamic polarization decays at the timescales longer than τ_s^e. As a result, when τ_s^e becomes shorter than the delay after the pump pulse, the spin polarization decreases.

Realization of the dynamic electron spin polarization in typical direct GaAs QDs is prevented by the large splitting between bright and dark excitons and by short exciton lifetimes. These limitations can be overcome in type-II QDs or core-shell colloidal nanocrystals [57]. Another promising platform for the implementation of the dynamic spin polarization involves twisted heterobilayers of transition metal dichalcogenides. The moiré pattern in these structures creates a superlattice potential with the typical period of about 5 nm [58]. For excitons localized in this potential, the hyperfine interaction leads to the spin-valley relaxation time on the order of tens of nanoseconds [59], while the exciton lifetime due to the confinement and spatial separation of electron and hole can be as long as 100 ns [58,60]. The fine structure of spin singlet and spin triplet excitons confined in a moiré potential remains poorly investigated [61,62]. Nevertheless, due to the electron-hole separation in heterobilayers, the fine structure splitting is expected to be small and can be on the order of the hyperfine interaction strength. Therefore, we expect the dynamic spin polarization of moiré trapped excitons.

In the process of the dynamic electron spin polarization, the angular momentum for the electrons is gained from the nuclear spin bath via the hyperfine interaction. This looks surprising, as commonly this interaction is considered as a source of spin relaxation only [45,63]. The dynamic electron polarization can be further transferred to nuclei [64] or magnetic impurities. Additionally, the dynamic spin polarization can be transferred to the resident electrons in the charged QDs [20]. The dynamic electron spin polarization has the following advantages: (i) It requires weak magnetic fields, which can be easily modulated. (ii) It uses nonresonant and unpolarized optical excitation. (iii) It is temperature independent, as long as the spin relaxation is dominated by the hyperfine interaction. (iv) 100% electron spin polarization is feasible. All that makes dynamic spin polarization very attractive for the spin orientation in nanodevices for quantum information processing.

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*smirnov@mail.ioffe.ru
†sha_tim@mail.ru

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