

**Dynamic approach for micromagnetics close to the Curie temperature**O. Chubykalo-Fesenko,<sup>1</sup> U. Nowak,<sup>2</sup> R. W. Chantrell,<sup>2</sup> and D. Garanin<sup>3</sup><sup>1</sup>*Instituto de Ciencia de Materiales de Madrid, CSIC, Cantoblanco, 28049 Madrid, Spain*<sup>2</sup>*Department of Physics, University of York, York YO10 5DD, United Kingdom*<sup>3</sup>*Department of Physics and Astronomy, Lehman College, City University of New York, 250 Bedford Park Boulevard West, Bronx, New York 10468-1589, USA*

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In conventional micromagnetism magnetic domain configurations are calculated based on a continuum theory for the magnetization. This theory assumes that the absolute magnetization value is constant in space and time. Dynamics is usually described with the Landau-Lifshitz-Gilbert (LLG) equation, the stochastic variant of which includes finite temperatures. Using simulation techniques with atomistic resolution we show that this conventional micromagnetic approach fails for higher temperatures since we find two effects which cannot be described in terms of the LLG equation: (i) an enhanced damping when approaching the Curie temperature and, (ii) a magnetization magnitude that is not constant in time. We show, however, that both of these effects are naturally described by the Landau-Lifshitz-Bloch equation which links the LLG equation with the theory of critical phenomena and turns out to be a more realistic equation for magnetization dynamics at elevated temperatures.

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**I. INTRODUCTION**

An increasing amount of research is focusing on the dynamic behavior of ferromagnetic materials at elevated temperatures. The motivations for this are manifold. A major imperative is the understanding of pulsed laser experiments on thin film samples, for example the *all-optical* FMR experiments of van Kampen *et al.*,<sup>1</sup> and the higher laser power experiments of Beaupaire *et al.*,<sup>2</sup> who demonstrated complete demagnetization on a timescale of picoseconds. One of the main issues of the high-temperature magnetization dynamics is the rate of the magnetization relaxation due to different processes involving magnon, phonon and electron interactions that contribute to thermal spin disordering. Preliminary simulations of multispin systems within the micromagnetic approach suggest that the macroscopic relaxation rate is strongly influenced by the spin-spin interaction<sup>3</sup> and increases with temperature.<sup>4</sup>

The basis of most of theoretical investigations of thermal magnetization dynamics is a micromagnetic approach which considers the magnetization of a small particle or a discrete magnetic nanoelement as a vector of a fixed length (referred to here as a macrospin) with the phenomenological Landau-Lifshitz-Gilbert (LLG) equation of motion augmented by a noise term.<sup>5</sup> However, contrary to the situation with atomic spins, there is no reason to assume a fixed magnetization length for nanoelements at nonzero temperature. For instance, the latter can decrease in time upon heating by a laser pulse. Hence, from the point of view of modeling of magnetization dynamics, there is a general need for further development of the micromagnetic theory in terms of its ability to deal with elevated temperatures.

Within this context we note the failure of micromagnetics in general to deal with the high frequency spin waves which give rise to the variation of magnetization with temperature. It has been suggested to treat this problem using scaling approaches.<sup>6,7</sup> A similar problem arises in multiscale model-

ing (with atomistic and micromagnetic discretizations to treat, for example, interfaces<sup>8,9</sup>) which cannot correctly describe the transfer of high energy spin waves from atomistic into the micromagnetic region. An alternative approach is the coarse graining model of Dobrovitski *et al.*,<sup>10</sup> which has the advantage of being able to link the length scales but has been developed for simple systems only.

Some understanding of the pulsed laser experiments could indeed be obtained in terms of a micromagnetic approach taking into account, in an empirical way, the temperature variation of the intrinsic parameters, particularly the saturation magnetization  $M_s$  and the anisotropy energy density  $K$ . Lyberatos and Guslienko<sup>11</sup> have used this macrospin model to investigate the response of nanoparticles during the heat assisted magnetic recording (HAMR) process. The validity of the macrospin approach including the thermal variation of model parameters has further been investigated in Ref. 12 using an atomistic approach. This work demonstrates that, although the macrospin model works well for temperatures far below the Curie temperature  $T_c$ , longitudinal fluctuations of the magnetization become important at elevated temperatures, which cannot be treated within the macrospin model of the corresponding LLG equation of motion. Clearly, some approach to macrospin dynamics beyond the LLG equation is needed.

An equation of motion for macrospins allowing for longitudinal relaxation has been derived in Ref. 13 within the mean-field approximation (MFA) from the classical Fokker-Planck equation for individual spins interacting with the environment. This “Landau-Lifshitz-Bloch (LLB) equation” has been shown to be able to describe linear domain walls, a domain wall type with nonconstant magnetization length. These results are consistent with the measurements of the domain wall mobility in YIG crystals close to  $T_c$  (Ref. 14) and by recent atomistic simulations.<sup>15</sup>

In this article we explore high-temperature dynamic properties using atomistic modeling. These simulations are still based on the LLG equation on the atomic level and, hence,

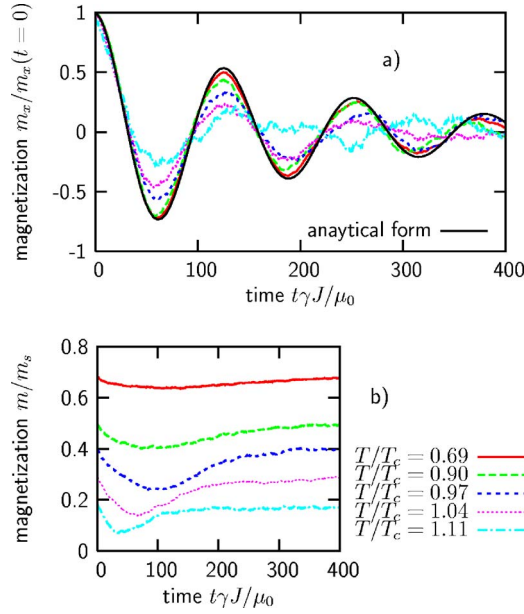


FIG. 1. (Color online) Relaxation of the magnetization for different temperatures using the atomistic modeling: 9a) normalized perpendicular component ( $30^\circ$  excitation); (b) absolute value of the magnetization  $m \equiv |\mathbf{m}|$  ( $135^\circ$  excitation).

still do not provide a microscopic description of the damping itself. Nevertheless they do include thermal degrees of freedom microscopically and demonstrate important phenomena associated with relaxation. We find an enhanced transverse relaxation when approaching the Curie temperature from below and a magnetization magnitude which is not constant in time. Both of these phenomena cannot be understood in terms of conventional micromagnetism but, comparing these predictions with a macrospin model based on the LLB equation, we conclude that here these phenomena are indeed well described by the LLB equation.

## II. ATOMISTIC SIMULATIONS

For our atomistic simulations we use a model in which the dynamic behavior of classical spins  $|\mathbf{s}_i|=1$  on lattice sites  $i$  with magnetic moment  $\mu_0$  is treated at the atomic level with the Langevin form of the LLG equation

$$\dot{\mathbf{s}}_i = -\gamma[\mathbf{s}_i \times \mathbf{H}_i] - \gamma\alpha[\mathbf{s}_i \times [\mathbf{s}_i \times \mathbf{H}_i]], \quad (1)$$

where  $\gamma$  is the gyromagnetic ratio, and  $\alpha$  is the damping parameter,  $\alpha=0.1$  in our simulations. The total field  $\mathbf{H}_i$  contains nearest-neighbor Heisenberg exchange (exchange constant  $J$ ) and Zeeman contributions and it is augmented by a white-noise field  $\zeta_i(t)$  with the correlator  $\langle \zeta_{i\mu}(t)\zeta_{j\nu}(t') \rangle = (2\alpha k_B T / \gamma \mu_0) \delta_{ij} \delta_{\mu\nu} \delta(t-t')$ , where  $\mu, \nu = x, y, z$ . For simplicity, the dipolar interaction is neglected as well as any crystalline anisotropy. A cubic lattice with periodic boundary conditions and system size of  $48^3$  has been considered. In the calculations we first establish thermal equilibrium for a given temperature starting with all magnetic moments parallel to the  $z$  axis and applying a field  $H_z = 0.05J/\mu_0$ . Then, to evaluate the transverse relaxation, all spins were simultaneously

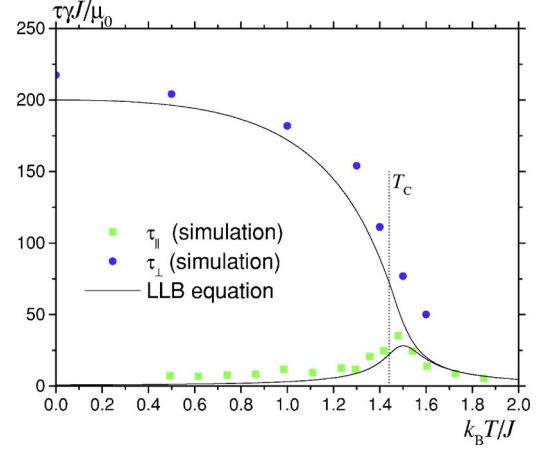


FIG. 2. (Color online) Temperature dependence of longitudinal and transverse relaxation times from the atomistic modeling and the LLB equation, calculated as inverse rates given by Eq. (6).

rotated by an angle of  $30^\circ$ . We have calculated the average spin polarization  $\mathbf{m} = (1/N)\sum_i \langle \mathbf{s}_i \rangle$  per lattice site which is proportional to the experimentally observed magnetization  $\mathbf{M}$ .

Figure 1(a) shows one transverse magnetization component as a function of time for different temperatures. The magnetization is normalized to its initial value and the data show clearly a faster relaxation for higher temperatures. Note that in our simulation even above the Curie temperature  $T_c$  there is still a finite magnetization due to finite-size effects and the fact that the simulations are conducted in an external field. Fitting the curves to an expression  $m_x(t) \sim \cos(t/\tau_{\perp})\exp(-t/\tau_{\perp})$  shows a perpendicular relaxation time  $\tau_{\perp}$  which increases with temperature, deviating from its zero temperature limit  $1/(\alpha\gamma H_z)$  (see Fig. 2).

Figure 1(b) presents the change of the absolute magnetization value as a function of time for a similar simulation but with a large angle of  $135^\circ$ . Note that the magnetization magnitude shows a dip during the relaxation process which is well below its equilibrium value. A dynamic response of this type cannot be described in terms of the macrospin LLG equation which conserves the absolute value of the magnetization, but is consistent with the LLB equation as will be seen below. The magnitude of the magnetization dip can be estimated as the contribution of the magnetic field to the equilibrium magnetization  $m(H, T)$ , i.e., as  $\Delta m \equiv m(H, T) - m_e$ , where  $m_e$  is the zero-field equilibrium magnetization. At equilibrium,  $\Delta m > 0$  and it is not negligible at elevated temperatures. After the magnetization  $\mathbf{m}$  is suddenly turned by  $135^\circ$ , the projection of  $\mathbf{H}$  onto  $\mathbf{m}$  becomes negative. Thus the magnetic field is now opposed to the exchange field that is mainly responsible for the formation of  $m$ . This leads a temporary decrease of  $m$  that is followed by recovery as  $\mathbf{m}$  turns in the direction of  $\mathbf{H}$  in the course of the relaxation.

Furthermore, we investigate the longitudinal relaxation time  $\tau_{\parallel}$  from the relaxation of the initially fully ordered system to thermal equilibrium. The relaxation of the magnetization to equilibrium is found to be approximately exponential on longer time scales which defines the characteristic time  $\tau_{\parallel}$ . Figure 2 shows the variation of the longitudinal relaxation time with temperature. The rapid increase close to  $T_c$  is

known as critical slowing down,<sup>17</sup> a general effect characterizing second order phase transitions. Also shown in Fig. 2 is the perpendicular relaxation time  $\tau_{\perp}$  determined as described above. Approaching the Curie temperature the perpendicular relaxation time  $\tau_{\perp}$  sharply decreases.

As we have demonstrated so far, the atomistic model shows important physical aspects of the behavior of nanoscale magnetic systems, including a temperature dependence of the effective damping, longitudinal fluctuations and critical slowing down. Next, we demonstrate that these effects can be described alternatively by macrospin magnetization dynamics in terms of the Landau-Lifshitz-Bloch equation of motion.<sup>13</sup> This provides not only a deeper understanding of the phenomena but it also suggests that the LLB equation is more suitable than the LLG equation for finite temperature micromagnetics.

### III. LANDAU-LIFSHITZ-BLOCH EQUATION

The LLB equation following from Eq. (1) in the spatially homogeneous case can be written in the form<sup>13</sup>

$$\dot{\mathbf{m}} = -\gamma[\mathbf{m} \times \mathbf{H}_{\text{eff}}] + \gamma\alpha_{\parallel} \frac{(\mathbf{m} \cdot \mathbf{H}_{\text{eff}})\mathbf{m}}{m^2} - \gamma\alpha_{\perp} \frac{[\mathbf{m} \times [\mathbf{m} \times \mathbf{H}_{\text{eff}}]]}{m^2}, \quad (2)$$

where  $\mathbf{m} = \langle \mathbf{s} \rangle$  is the spin polarization and  $\alpha_{\parallel}$  and  $\alpha_{\perp}$  are dimensionless longitudinal and transverse damping parameters given by

$$\alpha_{\parallel} = \alpha \frac{2T}{3T_c^{\text{MFA}}}, \quad \alpha_{\perp} = \alpha \left[ 1 - \frac{T}{3T_c^{\text{MFA}}} \right] \quad (3)$$

for  $T < T_c^{\text{MFA}}$  and the same with  $\alpha_{\perp} \Rightarrow \alpha_{\parallel}$  for  $T > T_c^{\text{MFA}}$ , where  $T_c^{\text{MFA}}$  is the mean-field Curie temperature. Here,  $\alpha$  is the same damping parameter that enters Eq. (1). The effective field  $\mathbf{H}_{\text{eff}}$  is assumed to be much weaker than the exchange interaction and it is given by

$$\mathbf{H}_{\text{eff}} = \mathbf{H} + \mathbf{H}_A + \begin{cases} \frac{1}{2\tilde{\chi}_{\parallel}} \left( 1 - \frac{m^2}{m_e^2} \right) \mathbf{m}, & T \lesssim T_c^{\text{MFA}} \\ \frac{J_0}{\mu_0} \left( \epsilon - \frac{3}{5} m^2 \right) \mathbf{m}, & T \gtrsim T_c^{\text{MFA}} \end{cases}. \quad (4)$$

Here  $\mathbf{H}$  and  $\mathbf{H}_A$  are applied and anisotropy fields and  $m_e$  is the zero-field equilibrium spin polarization in the MFA that satisfies the Curie-Weiss equation

$$m = B[\beta(mJ_0 + \mu_0 H)] \quad (5)$$

with  $H=0$  and  $\epsilon \equiv 1 - T/T_c^{\text{MFA}}$ .  $B$  is the Langevin function,  $\beta = 1/(k_B T)$ , and  $J_0$  the zero Fourier component of the exchange interaction related to  $T_c^{\text{MFA}}$  as  $k_B T_c^{\text{MFA}} = J_0/3$ . In Eq. (4)  $\tilde{\chi}_{\parallel} = \partial m(H, T) / \partial H$  is the longitudinal susceptibility at zero field that can be obtained from Eq. (5). The anisotropy field  $\mathbf{H}_A$  due to the uniaxial anisotropy is related to the zero-field transverse susceptibility  $\tilde{\chi}_{\perp}$  as  $\mathbf{H}_A = -(m_x \mathbf{e}_x + m_y \mathbf{e}_y) / \tilde{\chi}_{\perp}$ .<sup>13</sup> The equilibrium solution of the LLB equation satisfies  $\mathbf{H}_{\text{eff}} = 0$ . For  $T \ll T_c^{\text{MFA}}$  the longitudinal susceptibility  $\tilde{\chi}_{\parallel}$  becomes very

small in which case it can be shown that  $m \cong m_e$ . This means that the longitudinal relaxation vanishes and Eq. (2) reduces to the standard LLG equation with  $\alpha_{\perp} = \alpha$ .

In the damping parameters  $\alpha_{\parallel}$  and  $\alpha_{\perp}$  of Eq. (3)  $\alpha$  is noncritical at  $T_c^{\text{MFA}}$ . Its temperature dependence cannot be established within our semiphenomenological approach, so we assume it to be a constant, for the sake of comparison with the results of our atomistic simulations. The LLB equation also can be written in terms of the vector  $\mathbf{n} = \mathbf{m}/m_e$ .<sup>18</sup> This form provides a link to the micromagnetic anisotropy constants but becomes inconvenient above  $T_c$  where  $m_e$  disappears.

In order to effect a comparison we analyze the relaxation rates derived from the LLB equation. First we note from Eq. (3) a linear increase of  $\alpha_{\parallel}$  with  $T$ , whereas the behavior of  $\alpha_{\perp}$  is nonmonotonic, changing from a linear decrease below  $T_c^{\text{MFA}}$  to a linear increase above  $T_c^{\text{MFA}}$ . However, it is important to note that  $\alpha_{\parallel}$  and  $\alpha_{\perp}$  are noncritical for all finite temperatures, and that the variation of  $\alpha_{\perp}$  is weak. With this background, we now consider the relaxation rates from the linearized LLB equation which have the form

$$\Gamma_{\parallel} = \frac{\gamma\alpha_{\parallel}}{\tilde{\chi}_{\parallel}(H, T)}, \quad \Gamma_{\perp} = \frac{\gamma\alpha_{\perp}}{\tilde{\chi}_{\perp}(H, T)}, \quad (6)$$

where  $\tilde{\chi}_{\parallel}(H, T)$  is the longitudinal susceptibility at nonzero field that follows from Eq. (5) or simply from  $\mathbf{m} \cdot \mathbf{H}_{\text{eff}} = 0$ , in our approximation.

The longitudinal relaxation rate is, in general, very fast as  $\Gamma_{\parallel} \sim J_0$ . Since  $\tilde{\chi}_{\parallel}(H, T)$  is large near  $T_c^{\text{MFA}}$ ,  $\Gamma_{\parallel}$  shows critical slowing down which is a result of the critical behavior of  $\tilde{\chi}_{\parallel}(H, T)$  rather than the variation of  $\alpha_{\parallel}$ . The transverse susceptibility for the isotropic model is simply given by  $\tilde{\chi}_{\perp}(H, T) = m(H, T)/H$  so that  $\Gamma_{\perp} \sim H$  is much smaller than  $\Gamma_{\parallel}$  below  $T_c^{\text{MFA}}$ . However, it increases with temperature, as was observed in the atomistic modeling presented above and its critically behavior close to  $T_c$  is  $\Gamma_{\perp} \sim 1/m(H, T)$ . For temperatures below  $T_c$  a corresponding behavior was found for the linewidths of FMR experiments.<sup>16</sup>

At  $T = T_c^{\text{MFA}}$  the rates are given by

$$\Gamma_{\parallel} \cong \frac{6}{5} \frac{\gamma\alpha J_0}{\mu_0} m_H^2, \quad \Gamma_{\perp} \cong \frac{2}{5} \frac{\gamma\alpha J_0}{\mu_0} m_H^2, \quad (7)$$

where  $m_H = [(5/3)(\mu_0 H / J_0)]^{1/3}$  is the induced magnetization at  $T_c^{\text{MFA}}$ . Above  $T_c^{\text{MFA}}$  both rates merge:

$$\Gamma_{\parallel} \cong \Gamma_{\perp} \cong \frac{2}{3} \frac{\gamma\alpha J_0}{\mu_0} \frac{T}{T_c^{\text{MFA}}} \left( \frac{T}{T_c^{\text{MFA}}} - 1 \right). \quad (8)$$

Finally, in the presence of uniaxial anisotropy  $\Gamma_{\perp}$  is given by Eq. (6) with  $1/\tilde{\chi}_{\perp}(H, T) = H/m(H, T) + 1/\tilde{\chi}_{\perp}$ , where  $\tilde{\chi}_{\perp}$  is only weakly temperature dependent within mean-field theory below  $T_c^{\text{MFA}}$ .

To compare the LLB results with the predictions of the atomistic model, Fig. 2 includes the inverse relaxation rates calculated using Eq. (6) with rescaled temperature to fit the exact value  $k_B T_c = 1.44J$  for a simple cubic lattice. The agreement between Eq. (6) and the numerical results is remarkable given the MFA used in the derivation of Eq. (6).

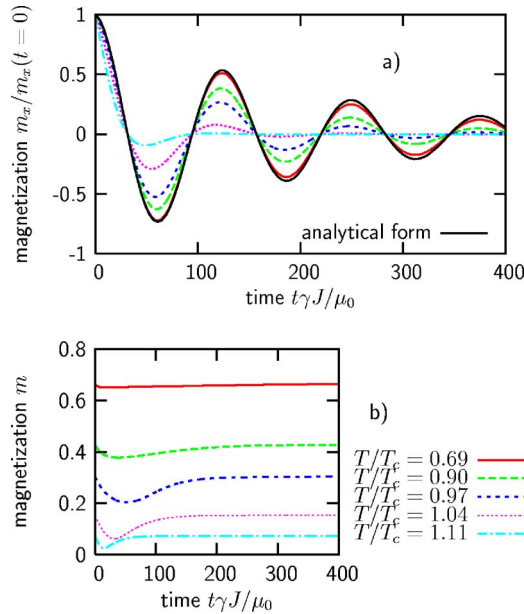


FIG. 3. (Color online) Relaxation of the magnetization for different temperatures as in Fig. 1 but using the macrospin LLB modeling.

Also, we have integrated numerically Eq. (2) for a macrospin to give the time evolution of the magnetization components for comparison with the numerical results of Fig. 1. The results are presented in Fig. 3. Comparison with Fig. 1 shows that the LLB equation reproduces essential physical processes which govern the magnetization dynamics at elevated temperatures and thus it can be used as an alternative to micromagnetics in this region. However, this comparison could still be improved if one evaluates the macrospin parameters directly from an atomistic simulation. Furthermore, if the LLB equation is to be used as an alternative to micromagnetics, the corresponding parameters could as well be extracted from experiment.

#### IV. CONCLUSIONS AND OUTLOOK

In conclusion, performing atomistic simulations of thermal magnetization dynamics we observe an increase of the macroscopic transverse damping approaching the Curie temperature. This increase is determined by the thermal dispersion of magnetization and would exist independently from any other possible thermal dependence of internal damping mechanisms such as phonon-magnon coupling. This effect explains the broadening of the resonance linewidth in classical FMR experiments.<sup>16</sup> Furthermore, the magnetization vector turns out not to be constant in length. Instead during relaxation one can observe a dip of the magnetization which is more pronounced when approaching the Curie temperature. Finally, the magnetization dynamics has important con-

tributions from longitudinal relaxation. This relaxation shows critical slowing down at temperatures close to  $T_c$ . Importantly, the observed dynamics is in agreement with the dynamics of a macrospin described by the Landau-Lifshitz-Bloch equation which contains both longitudinal and transverse relaxation.

The LLB equation could serve in future as a basis for an improved micromagnetics at elevated temperature. This suggestion is based on the remarkable degree to which the LLB equation captures the complex physics revealed by the atomistic model, specifically the variation of the magnetization magnitude during reversal, and the increase of damping with temperature. Both of these are nontrivial effects and are reproduced extremely well by the LLB equation. The longitudinal and transverse relaxation times calculated from the LLB equation also agree well with those calculated from the atomistic model. This is in itself remarkable given that the parameters for the LLB equation were calculated using mean field theory. Our suggestion is in practice to improve the LLB equation using parameters such as the susceptibilities, magnetization and anisotropy calculated numerically from atomistic simulations. Alternatively one could take the parameters from experiments.

The LLB equation then reproduces the temperature dependence of both the static properties such as  $M(T)$  and also the longitudinal and transverse relaxation, and is naturally suited to micromagnetic simulations. We note especially that the use of the LLB equation would open the possibility to enable micromagnetic simulations of HAMR experiments, including the rapid decrease of the magnetization induced by the laser pulse. In order to extend the approach to micromagnetic systems with the LLB equation, it will be necessary to understand the variation of the micromagnetic exchange parameter with temperature.

An interesting further use of the LLB equation is in the area of multiscale simulations<sup>8,9</sup> where atomistic simulations of areas are linked to micromagnetic regions to extend the calculations to macroscopic lengthscales. Current simulations<sup>8</sup> use the LLG equation for the micromagnetic cells, which has the disadvantage that high frequency spin waves within the atomistic region will be reflected at the atomistic/micromagnetic boundary. The LLB equation relaxes the constraint of constant magnetization which represents a significant advance in the physical basis of multiscale simulations of magnetic systems.

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