Spin Dynamics in a Dissipative Environment: From Quantal to Classical

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We address the problem of spin dynamics in the presence of a thermal bath, by solving exactly the appropriate quantum master equations with continued-fraction methods. The crossover region between the quantum and classical domains is studied by increasing the spin value S, and the asymptote for the classical absorption spectra is eventually recovered. Along with the recognized relevance of the coupling strength, we show the critical role played by the *structure* of the system-environment interaction in the emergence of classical phenomenology.

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Quantum mechanics is one of the most subtle and powerful theoretical constructions of the human mind. Understanding its implications, relation with other theories, and domain of validity has captivated scientists since its advent. This domain has been slowly expanding from the traditional one of atoms and molecules, to condensed matter systems (solids and liquids), and more recently by studies of decoherence, quantum analogs of classical effects (e.g., chaos), and the quantum-to-classical transition [1].

These studies have also brought an increasing awareness of the role of the environment. Thus, the field of *open quantum systems* deals with systems consisting of a few relevant degrees of freedom coupled to the surrounding medium, which has a large number of constituents (photons, phonons, electrons, nuclei, etc.). The coupling produces dissipation, fluctuations, and decoherence; it also enables the system to interchange energy and correlations with the bath and relax to equilibrium [2]. Besides its basic interest, the above generic conditions make this topic relevant in various areas of physics and chemistry.

Spins constitute one of the most paradigmatic quantum systems due to their discrete and finite energy spectrum. Their dynamics is also special and rich because of the underlying commutation relations $[S_i, S_j] = i\epsilon_{ijk}S_k$. Naturally, it is important to take into account environmental effects in spin problems, and this has led to several theories of *spin relaxation*. To deal rigorously with quantum dissipative systems, however, is a difficult task. Path integral propagators and quantum Langevin or master equations can typically be solved in a few simple cases: free particle (or in a uniform field), harmonic oscillator [2], two-state systems (e.g., $S = 1/2$ spins) [3], etc.

The *continued-fraction method*, devised originally for classical Brownian-motion problems [4], has been successfully adapted to solve master equations for some quantum systems [5–8]. Here we shall apply this technique to a spin with *arbitrary* S weakly coupled to a dissipative bath, and monitor its intrinsic dynamics via spin resonance. We investigate how the approach to the classical results takes place (out of reach of previous exact methods due to their limitations in S). We focus on the effects of the environment, not only of the coupling strength, but also of the *structure* of the spin-bath Hamiltonian. Usual studies of open quantum systems overlook the latter and adopt the simplest bilinear interaction. We consider two models with a solid-state motivation: coupling to electron-hole excitations, actually linear in $\boldsymbol{S}$, and to phonons, an even polynomial in $\boldsymbol{S}$. We find that the approach to the classical results depends *qualitatively* on the coupling structure (and the bath spectral properties). This is specially critical for the uniformity of the convergence in the different frequency sectors of the spin absorption spectra. The problem is not merely academic; large-spin molecular clusters are in the focus, while magnetic nanoparticles provide a natural classical limit [9]. Thus, our results could also help in discriminating different proposed couplings in those systems and hence ascertain the microscopic origin of dissipation.

Let us start with the Hamiltonian of a spin $\boldsymbol{S}$ coupled to a bosonic bath (linearly in the bath variables)

$$\mathcal{H}_{\rm tot} = \mathcal{H}(\boldsymbol{S}) + \sum_{\boldsymbol{q}} V_{\boldsymbol{q}} F_{\boldsymbol{q}}(\boldsymbol{S})(a_{\boldsymbol{q}}^{+} + a_{-\boldsymbol{q}}) + \mathcal{H}_{\rm b}. \quad (1)$$

Here $\mathcal{H}(\boldsymbol{S})$ and $\mathcal{H}_{\rm b} = \sum_{\boldsymbol{q}} \omega_{\boldsymbol{q}} a_{\boldsymbol{q}}^{+} a_{\boldsymbol{q}}$ are the spin and bath Hamiltonians, $F_{\boldsymbol{q}}(\boldsymbol{S})$ the spin-dependent part of the interaction, and $V_{\boldsymbol{q}}$ coupling constants. For systems with a discrete and finite spectrum, it is convenient to introduce the Hubbard (level-shift) operators $X_n^m \equiv |n\rangle\langle m|$. Any operator F can then be expanded in this basis $F = \sum_{nm} F_{nm} X_n^m$, with $F_{nm} = \langle n|F|m\rangle$. When coupled to the environment, the spin is not in a pure state and it needs to be described by its density matrix ϱ. Its matrix elements are given in this framework by $\varrho_{mn} = \langle X_n^m \rangle$.

Many problems in quantum optics, magnetism, or chemical physics involve *weak* system-bath coupling [2]. Then, the dynamical equation for ϱ can be obtained by perturbation theory. In the Hubbard formalism, using the S_z eigenstates as the basis, $S_z|m\rangle = m|m\rangle$, one finds for $\mathcal{H} = \mathcal{H}_{\rm d}(S_z) - \boldsymbol{B} \cdot \boldsymbol{S}$ the following density-matrix equation [10,11]

$$\frac{d}{dt}X_n^m = \mathrm{i}\Delta_{nm}X_n^m + (\mathrm{i}/2)B_+(\ell_m X_n^{m+1} - \ell_{n-1}X_{n-1}^m) + (\mathrm{i}/2)B_-(\ell_{m-1}X_n^{m-1} - \ell_n X_{n+1}^m) + R_n^m. \quad (2)$$

The $\Delta_{nm} \equiv \varepsilon_n - \varepsilon_m$ are the frequencies associated with the $m \to n$ transition, ε_m being the levels of the diagonal part of the spin Hamiltonian (including B_z). The circular components of the transverse field are $B_\pm = B_x \pm \mathrm{i}B_y$ and the $\ell_m = [S(S+1) - m(m+1)]^{1/2}$ are ladder factors.

The first three terms in Eq. (2) give the *unitary* evolution of the isolated spin in the Heisenberg representation. The *relaxation* term R_n^m incorporates the effects of the bath and has a non-Markovian (history dependent) form

$$R_n^m = -\int_{-\infty}^{t} d\tau\{\mathcal{K}(\tau - t)F(\tau)[F, X_n^m] - \mathcal{K}(t-\tau)[F, X_n^m]F(\tau)\}. \quad (3)$$

Here the operators without time argument are evaluated at t whereas $F(\tau) = \sum_{n'm'} F_{n'm'}X_{n'}^{m'}(\tau)$. The memory kernel $\mathcal{K}$ is given in terms of the *spectral density* of bath modes, $J(\omega) \equiv \frac{\pi}{2}\sum_{\boldsymbol{q}}|V_{\boldsymbol{q}}|^2\delta(\omega - \omega_{\boldsymbol{q}})$, and bosonic occupation numbers, $n_\omega = 1/(e^{\omega/T} - 1)$, by

$$\mathcal{K}(\tau) = \int_0^\infty \frac{d\omega}{\pi} J(\omega)[n_\omega e^{+\mathrm{i}\omega\tau} + (n_\omega + 1)e^{-\mathrm{i}\omega\tau}]. \quad (4)$$

To second order in the interaction, and not too strong transverse field, the *retarded* time dependences $X_{n'}^{m'}(\tau)$ can be determined by the dominant term in the conservative evolution $X_n^m(\tau) \simeq e^{-\mathrm{i}\Delta_{nm}(t-\tau)}X_n^m(t)$. Inserting such $X_{n'}^{m'}(\tau)$ in R_n^m, only operators evaluated at t remain and non-Markovian features effectively disappear. Then, the coefficients of the $X_{n'}^{m'}$ include, along with the coupling matrix elements F_{nm}, the relaxation rates $W_{n|m} \equiv W(\Delta_{nm})$, with the universal *rate function* associated with the kernel $W(\Delta) = \mathrm{Re}[\int_0^\infty d\tau e^{-\mathrm{i}\Delta\tau}\mathcal{K}(\tau)]$.

We shall consider in the sequel the following family of couplings F: linear in $S_\pm = S_x \pm \mathrm{i}S_y$ but allowing for S_z-dependent "coefficients" $v(S_z)$:

$$F(\boldsymbol{S}) = \eta_+[v(S_z), S_-]_+ + \eta_-[v(S_z), S_+]_+. \quad (5)$$

Here $\eta_\pm$ are some scalars ensuring $F^+ = F$ while $[A, B]_+ \equiv AB + BA$. Then, the matrix elements $F_{nm} = \langle n|F|m\rangle$ read $F_{nm} = L_{m,m-1}\delta_{n,m-1} + L_{m+1,m}^*\delta_{n,m+1}$, where $L_{m,m'} = \eta_+[v(m) + v(m')]\ell_{m,m'}$ and $\ell_{m,m\pm1} = [S(S+1) - m(m\pm1)]^{1/2}$. The relaxation term for these couplings acquires the following (Redfield) form [10–12]

$$\begin{aligned} R_n^m = {} & L_{n,n-1}L_{m,m-1}^*(W_{n|n-1} + W_{m|m-1})X_{n-1}^{m-1} \\ & - (|L_{n+1,n}|^2 W_{n+1|n} + |L_{m+1,m}|^2 W_{m+1|m} \\ & \quad + |L_{n,n-1}|^2 W_{n-1|n} + |L_{m,m-1}|^2 W_{m-1|m})X_n^m \\ & + L_{n+1,n}^* L_{m+1,m}(W_{n|n+1} + W_{m|m+1})X_{n+1}^{m+1}. \end{aligned} \quad (6)$$

Inserting this R_n^m in Eq. (2) we get the master equation for our problem within a fully quantum treatment (no phenomenological relaxation is introduced, no preconceived form of the equation is assumed). Recall finally that handling the spin precession requires solving the full density-matrix equation, because it involves off-diagonal elements, and it is not captured by a (Pauli) master equation for the level populations (ϱ_{mm}).

We mentioned the difficulties in solving our models for quantum dissipation and that the continued-fraction method (a relative of the recursion method and Lanczos tridiagonalization) has been applied to several quantum systems [5–8]. For spin problems [12] one starts writing the master equation compactly as $\dot{X}_n^m = \sum_{n'm'}\mathcal{Q}_{n,n'}^{m,m'}X_{n'}^{m'}$ with $n' = n-1, n, n+1$ and $m' = m-1, m, m+1$. To convert this 2-index differential recurrence into a 1-index one, we introduce appropriate $(2S+1)$ vectors, $\boldsymbol{c}_n$, and $(2S+1)\times(2S+1)$ matrices, $\mathbb{Q}_{n,n'}$, with components and elements [12]

$$(\boldsymbol{c}_n)_m = \langle X_n^m\rangle, \qquad (\mathbb{Q}_{n,n'})_{mm'} = \mathcal{Q}_{n,n'}^{m,m'}, \quad (7)$$

obtaining $\dot{\boldsymbol{c}}_n = \mathbb{Q}_{n,n-1}\boldsymbol{c}_{n-1} + \mathbb{Q}_{n,n}\boldsymbol{c}_n + \mathbb{Q}_{n,n+1}\boldsymbol{c}_{n+1}$. In this 1-index form the recurrence can be tackled by (matrix) continued-fraction methods [4] yielding the solution of the master Eq. (2). We have then the full density matrix $\varrho_{mn} = \langle X_n^m\rangle = (\boldsymbol{c}_n)_m$ and *any* observable (magnetization, susceptibilities, etc.) can be computed from the trace formula $\langle A\rangle = \mathrm{Tr}(\varrho A)$. The matrix associated with the original system, $\dot{\boldsymbol{X}} = \mathcal{Q}\boldsymbol{X}$, had dimensions $(2S+1)^2\times(2S+1)^2$ making the handling even of moderate spins ($S \lesssim 10$) difficult. The continued-fraction approach replaces it by $2S+1$ problems but with matrices $(2S+1)\times(2S+1)$, allowing us to gain some orders of magnitude in S and pursue the classical limit a way longer.

We now apply the above formalism to the problem of spin dynamics in a magnetic-anisotropy plus Zeeman potential $\mathcal{H} = -DS_z^2 - \boldsymbol{B}\cdot\boldsymbol{S}$. This Hamiltonian may also be viewed as the minimal model for superparamagnets [9]. The anisotropy term has two minima at $S_z = \pm S$ with a barrier at $S_z = 0$. The coupling to the environment provokes quantum Brownian motion of the spin, which may overcome the potential barriers. We consider two basic solid-state mechanisms [2,3]: (i) Coupling to *electron-hole* excitations near the Fermi surface (a bosonizable bath); then $F(\boldsymbol{S}) = \frac{1}{2}(\eta_+S_- + \eta_-S_+)$ [i.e., $v(S_z) = \text{const}$ in Eq. (5)] while the bath is *Ohmic*, $J(\omega) = \lambda\omega$. (ii) Coupling to *phonons*; now $v(S_z) \propto S_z$ and the environment is *super-Ohmic*, $J(\omega) = \lambda\omega^3$ (in 3D). Classically these couplings yield field-type and anisotropy-type fluctuations, respectively, in the Langevin equations. The rates $W_{n'|m'}$ required in R_n^m can be obtained from the spectral density by $W(\Delta) = J(\Delta)n_\Delta + J(-\Delta)(n_{-\Delta} + 1)$, understanding $J(\omega < 0) \equiv 0$. We start with the super-Ohmic nonlinear case, which has received less attention in the context of quantum dissipative systems than the Ohmic bilinear coupling; we will see that it also has a rich physics.

The Zeeman term and $F \propto S_\pm$ have nonzero matrix elements between the states $|m\rangle$, producing transitions

between them. In an oscillating field, they result in peaks in the imaginary part $\chi''(\Omega)$ of the dynamical susceptibility (absorption line shape) located at the transition frequencies $\Delta_{m,m+1} = \varepsilon_m - \varepsilon_{m+1} = D(2m+1) + B_z$ (Fig. 1). The transitions at the potential wells ($|m| \sim S$) correspond to the largest frequencies ($\Delta \sim 2DS$ at $B_z = 0$), while those near the barrier top ($m \sim 0$) appear at low frequencies ($\sim D$). Then, going from high to low Ω, the intensity of the peaks decreases, as they involve transitions between higher levels, which are thermally less populated.

The peaks have finite width and height due to the damping λ and the temperature, as the interaction with the bath "blurs" the spin energy levels. Thus, a lowering of λ or T makes the peaks narrower and higher (phenomenology akin to that of a damped oscillator). There is an extra narrowing of the low Ω peaks, because the spin-phonon coupling $F \sim S_z S_\pm$ leads to an effective damping decreasing with m [$\lambda_{\rm eff} \sim \lambda(2m \pm 1)^2$]. This enters in R_n^m via the modified ladder factors $|L_{m,m\pm1}|^2 \sim (2m \pm 1)^2 \ell^2_{m,m\pm1}$ and it is a spin analogue of position-dependent damping in translational Brownian motion.

Next, let us briefly discuss the corresponding *classical* behavior. The actual line shape will depend on the phenomenological relaxation model considered (Bloch equations, Landau-Lifshitz, etc.). Nevertheless, the result in the limit of zero damping is universal [13,14]

$$\chi''(\Omega) = \frac{\mu^2}{T}\frac{\pi}{2\mathcal{Z}}\Omega[1 - (\Omega/\Omega_{\rm a})^2]\exp[\sigma(\Omega/\Omega_{\rm a})^2]. \quad (8)$$

Here $\mathcal{Z}$ is the partition function, $\Omega_{\rm a}$ the resonance frequency at the wells, and σ the barrier over T. Physically, the anisotropy $\mathcal{H}_{\rm d} = -DS_z^2$ leads to S_z-dependent precession frequencies $\propto \partial\mathcal{H}/\partial S_z$ and the ensuing *spreading* of the line shape (inset of Fig. 1). The population of the different S_z orbits changes with T, modifying $\chi''(\Omega)$. Note that this dissipationless limit provides a good description for weak enough coupling in most of the Ω range.

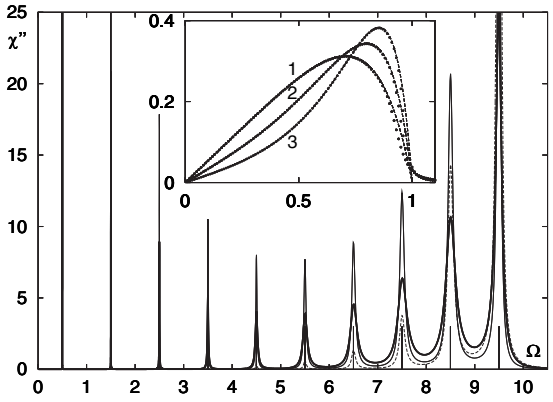

FIG. 1. Absorption line shape $\chi''(\Omega)$ for a spin $S = 10$ with $D = 0.5$ at $B_z = 0$. Thick line: $\sigma = DS^2/T = 5$ and spin-*phonon* coupling $\lambda = 3 \times 10^{-8}$. Thin lines: effects of halving the damping at the same T (solid), and of halving T keeping λ (dashed). Vertical lines: loci of the transition frequencies $\Delta_{m,m+1} = D(2m+1)$. Inset: *classical* dampingless asymptote (8) for $\sigma = 1$, 2, and 3 (lines), and exact Fokker-Planck results for *finite* Landau-Lifshitz damping ($\lambda_{\rm LL} = 0.003$; symbols).

We thus see that the classical phenomenology looks quite different from the multipeaked structure of the quantum case. This poses the following questions: (i) How does quantum mechanics manage to join those two behaviors? and (ii) Which are the main factors determining the way in which the classical phenomenology emerges? We now try to answer these questions by solving the density-matrix equation (2) for increasing S and getting as close as possible to the classical domain.

Recall, however, that limiting procedures in physical problems (e.g., lattice to continuous limit in field theories, thermodynamic limit in statistical mechanics, etc.) require us to define clearly: (i) which quantities are kept constant when taking the limit and (ii) which scaled variables are needed to monitor the evolution. We fix the reduced anisotropy and field parameters $\sigma = DS^2/T$ and $\xi = SB/T$. At constant T this entails keeping the anisotropy-barrier height and amount of Zeeman energy constant (and hence finite) while introducing more levels with S (the spacing decreases as $\Delta \sim 1/S$). As for the scaled quantities, guided by the classical result (8), we use χ/χ_0 with $\chi_0 = S(S+1)/T$ (corresponding to μ^2/T) and $\Omega/2DS$ (which tends to $\Omega/\Omega_{\rm a}$). Finally, we also scale the bare coupling strength λ with S. The reason is that in the density-matrix equation the Hamiltonian coefficients go as $\Delta \sim 1/S$ while the relaxation ones decrease as $\lambda D^2 L^2 \Delta^2 \sim \lambda/S^2$ (we include a D^2 dependence arising in the coupling to phonons [11]). Thus, fixing λ/S we can study the effects of going to large S while maintaining the relative "weights" of the conservative and relaxation terms in the quantum master equation.

Proceeding in this way, we compute the transverse dynamical response for various S (Fig. 2). For moderate spins we clearly recognize the quantum features of Fig. 1. As S is increased more peaks are introduced into the same interval $\Omega/\Omega_{\rm a}$. Because of their finite width they start to coalesce and a limit curve progressively emerges. However, the approach is far from uniform in Ω. At low frequencies the peaks merge slowly with S; they are sharp and narrow due to the m-dependent damping associated with S_z in $F \sim S_z S_\pm$. This is less relevant at high frequencies [large $|m|$, $\delta\lambda_{\rm eff}/\lambda_{\rm eff} \sim 4/(2m \pm 1)$] and a smooth peakless line shape arises there. For a fixed S, in addition, one would expect that larger spin-bath coupling will "accelerate" the classical convergence. Figure 2 actually shows that the wildly peaked part is then pushed further into the low Ω sector and that the "oscillations" around the limit curve are reduced. It is remarkable that this limit curve is indeed Gekht's classical prediction (8).

The finite width of the absorption peaks has been essential to reconstruct the classical curve. Here it has been provided by the coupling to the environment; in other situations different broadening mechanisms may contribute [15]. The form of the interaction, on the other hand, has led to a highly nonuniform approach to the classical

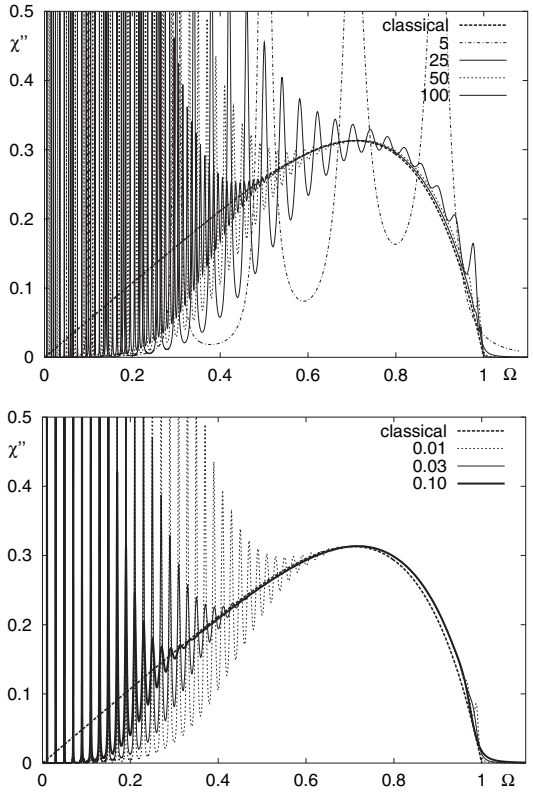

FIG. 2. Spectral line shape $\chi''(\Omega)$ for $\sigma = 1$ at $B_z = 0$ (see the text for the scaled quantities). The thick dashed line is the classical Eq. (8). Top: $S = 5$, 25, 50, and 100 with ''constant'' coupling $\lambda/S = 10^{-2}$. Bottom: fixed $S = 50$ with $\lambda/S = 10^{-2}$ (as in top), 3×10^{-2}, and 10^{-1}.

asymptote. This shows that not only the strength, but also the structure of the coupling Hamiltonian can play an important role in the approach to the classical regime.

This can be further supported by comparing with the ''electron-hole'' coupling model, where $F \sim S_{\pm}$. To assess the different contributions we proceed in two steps, adjusting λ to get the same intensity for the ground-state transitions (rightmost peaks); Fig. 3. First, we go from the phonon-coupling $F \sim S_z S_{\pm}$ to a hybrid model with $F \sim S_{\pm}$, but still super-Ohmic spectral density. This F greatly tames the low-frequency sharp peaks, but some nonuniformity still remains, due to $W_{m|m+1} \sim J(\Delta) n_{\Delta} \sim \Delta_{m,m+1}^{3-1}$ in the relaxation term. Second, we add the Ohmic bath $J \propto \omega$ to the bilinear coupling. Then $W_{m|m+1} \sim \Delta_{m,m+1}^{1-1} \sim$ const and the approach to the classical behavior becomes quite uniform in most of the Ω range, in spite of the moderate spin value considered ($S = 10$).

In summary, we have addressed the problem of spin dynamics in a dissipative thermal bath. Solving exactly the quantum master equation by a continued-fraction method for increasing S has allowed us to approach the classical prediction for the absorption spectra. We have investigated the effects of the spin-bath interaction on the quantum-to-classical crossover. The coupling strength, as usual in quantum dissipative systems, accentuates the attainment of the classical phenomenology. However, the approach is qualitatively affected by the structure of the interaction, as illustrated with the different convergences in the different sectors of the absorption spectra for two important solid-state mechanisms. Although the relevance of dissipation, specially in mesoscopic systems, is amply recognized, only studies of decoherence and approach to equilibrium had paid due attention to the structure of the coupling Hamiltonian. Here we have shown its relevance also in the features of the quantum-classical border and in the emergence of classical behavior.

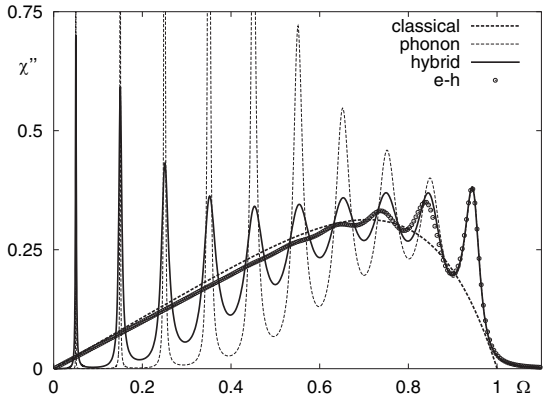

FIG. 3. Line shape for $S = 10$ at $\sigma = 1$ with (i) the phonon-coupling model and (ii) bilinear coupling, both with super-Ohmic (hybrid) and Ohmic (electron-hole) spectral densities.

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