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# A far-from-equilibrium fluctuation-dissipation relation for an Ising-Glauber-like model

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**Abstract.** We derive an exact expression of the response function to an infinitesimal magnetic field for an Ising-Glauber-like model with arbitrary exchange couplings. The result is expressed in terms of thermodynamic averages and does not depend on the initial conditions or on the dimension of the space. The response function is related to time-derivatives of a complicated correlation function and so the expression is a generalisation of the equilibrium fluctuation-dissipation theorem in the special case of this model. Correspondence with the Ising-Glauber model is discussed. A discrete-time version of the relation is implemented in Monte Carlo simulations and then used to study the aging regime of the ferromagnetic two-dimensional Ising-Glauber model quenched from the paramagnetic phase to the ferromagnetic one. Our approach has the originality to give direct access to the response function and the fluctuation-dissipation ratio.

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## 1. Introduction

The knowledge about out-of-equilibrium processes is far from being as advanced as for systems at thermodynamical equilibrium. In particular, the fluctuation-dissipation theorem (FDT) which holds at equilibrium is known to be violated out-of-equilibrium. This theorem states that at equilibrium the response function  $R^{\text{eq}}(t-s)$  at time  $t$  to an infinitesimal field applied to the system at time  $s < t$  is related to the time-derivative of the two-time autocorrelation function  $C^{\text{eq}}(t-s)$ :

$$R^{\text{eq}}(t-s) = \beta \frac{\partial}{\partial s} C^{\text{eq}}(t-s). \quad (1)$$

In the Ising case, the response function reads  $R_{ji}(t,s) = \frac{\delta \langle \sigma_j(t) \rangle}{\delta h_i(s)}$  and the correlation function  $C_{ji}(t,s) = \langle \sigma_j(t) \sigma_i(s) \rangle$ . Based on a mean-field study of spin-glasses, Cugliandolo *et al.* [1] have conjectured that for asymptotically large times the FDT can be generalised by adding a multiplicative factor  $X(t,s)$  which moreover depends on time only through the correlation function:

$$R(t,s) \underset{t \sim s \gg 1}{\sim} \beta X(C(t,s)) \frac{\partial}{\partial s} C(t,s). \quad (2)$$

The quantity  $\beta^{\text{eff}}(t,s) = \beta X(C(t,s))$  is interpreted as an effective inverse temperature. Exact results have been obtained for the ferromagnetic Ising chain [2, 3] that confirm this conjecture. Unfortunately, the response function is rarely so easily accessible for more complex systems. Both numerically and experimentally, only the integrated response function is usually measured by applying a finite magnetic field to the system during a finite time. In the so-called TRM scheme, the magnetic field is applied between the times 0 and  $s$  and the magnetisation is measured at time  $t$ . Assuming the equation (2) valid for any times  $t$  and  $s$ , one can relate the integrated response function to the fluctuation-dissipation (FD) ratio:

$$\chi(t,s) = \int_0^s R(t,u) du \sim \beta \int_{C(t,0)}^{C(t,s)} X(C) dC. \quad (3)$$

The FD ratio  $X(t,s)$  can thus be obtained as the slope of the integrated response function  $\chi(t,s)$  when plotted versus the correlation function  $C(t,s)$ . This method has been applied to the numerical study of many systems: 2d and 3d-Ising ferromagnets [4], 3d Edwards-Anderson model [5, 4], 3d and 4d-Gaussian Ising spin-glasses [6], 2d Ising ferromagnet with dipolar interactions [7], Heisenberg anti-ferromagnet on the Kagome lattice [8] ... The conjecture (2) has also recently been checked experimentally for a spin-glass [9]. More details may be found in the reviews [10, 11]. However, the integrated response function depends linearly on the FD ratio only if the conjecture (2) holds, which has not been demonstrated for any of the previously cited systems. We will see in the case of the homogeneous Ising model that this approach may lead to misinterpretations and erroneous values of  $X(t,s)$ . The generalisation of the equilibrium FDT has recently become an increasingly popular issue. Let us mention two of them: an approximate generalisation of the FDT to metastable systems [12] (limited to dynamics having a transition rate  $W$  with only one negative eigenvalue) that has been successfully

compared to numerical data for the 2D-Ising model and a generalisation of the FDT for trap models [13].

In the present work, we study the dynamics of an Ising-Glauber-like model. In the section 1, we describe the model and its dynamics which are studied analytically in the section 2. The response function to an infinitesimal magnetic field is exactly calculated far-from-equilibrium. It turns out that the response function is no more related to a time-derivative of the spin-spin correlation function but to time-derivatives of a more complicated correlation function. The equilibrium limit is shown to have the usual form. In the section 3, a discrete-time version of this expression is implemented in Monte Carlo simulations. Our approach presents several advantages: (i) we can compute directly the response function and not only the integrated response function, (ii) we obtain the response function to an infinitesimal magnetic field so that we avoid non-linear effects due to the use of a finite magnetic field, (iii) the FD ratio can be computed without resorting to Cugliandolo conjecture (2) and (iv) we can calculate the response function  $R(t, s)$  and the FD ratio  $X(t, s)$  for any time  $t$  and  $s < t$  during one single Monte Carlo simulation. We performed Monte Carlo simulations of the two-dimensional homogeneous Ising model quenched at and below the critical temperature  $T_c$ . In both cases, the expected scaling behaviour of the response function in the aging regime is well reproduced by the numerical data. The value of the exponent  $a$ , still controversial, is estimated and the FD ratio is computed. Our estimate of  $X_\infty$  at  $T_c$  turns out to be compatible with previous work and the scaling behaviour of  $X(t, s)$  below  $T_c$  is well reproduced. In both cases, the FD ratio depend on time not only through the correlation function.

## 2. Our Ising-Glauber-like model

### 2.1. Useful relations on Markov processes

We consider a classical Ising model whose degrees of freedom are  $N$  scalar variables  $\sigma_i = \pm 1$  located at the nodes of a  $d$ -dimensional lattice. Let us denote by  $\wp(\{\sigma\}, t)$  the probability to observe the system in the state  $\{\sigma\}$  at time  $t$ . We first define a discrete-time Markov chain by the master equation

$$\wp(\{\sigma\}, t + \Delta t) = (1 - \Delta t)\wp(\{\sigma\}, t) + \Delta t \sum_{\{\sigma'\}} W(\{\sigma'\} \rightarrow \{\sigma\}, t)\wp(\{\sigma'\}, t) \quad (4)$$

where  $W(\{\sigma\} \rightarrow \{\sigma'\}, t)$  is the transition rate per unit time from the state  $\{\sigma\}$  to the state  $\{\sigma'\}$  at time  $t$ . The condition  $\sum_{\{\sigma'\}} W(\{\sigma\} \rightarrow \{\sigma'\}, t) = 1$  ensures the normation of the probability  $\wp(\{\sigma\}, t)$  at any time  $t$ . The system is not forced to make a transition at each time step, i.e. the transition rate may have non-zero diagonal elements  $W(\{\sigma\} \rightarrow \{\sigma\}, t)$ . In the continuous-time limit  $\Delta t \rightarrow 0$ , the master equation (4) goes to

$$\left(1 + \frac{\partial}{\partial t}\right) \wp(\{\sigma\}, t) = \sum_{\{\sigma'\}} W(\{\sigma'\} \rightarrow \{\sigma\}, t)\wp(\{\sigma'\}, t). \quad (5)$$

It is easily shown that the conditional probability,  $\wp(\{\sigma\}, t|\{\sigma'\}, s)$  with  $s < t$ , defined by the Bayes relation

$$\wp(\{\sigma\}, t) = \sum_{\{\sigma'\}} \wp(\{\sigma\}, t|\{\sigma'\}, s) \wp(\{\sigma'\}, s) \quad (6)$$

satisfies the same master equation (4) too:

$$\begin{aligned} \wp(\{\sigma\}, t + \Delta t|\{\sigma'\}, s) &= (1 - \Delta t) \wp(\{\sigma\}, t|\{\sigma'\}, s) \\ &+ \Delta t \sum_{\{\sigma''\}} W(\{\sigma''\} \rightarrow \{\sigma\}, t) \wp(\{\sigma''\}, t|\{\sigma'\}, s) \end{aligned} \quad (7)$$

or in the continuous-time limit

$$\left(1 + \frac{\partial}{\partial t}\right) \wp(\{\sigma\}, t|\{\sigma'\}, s) = \sum_{\{\sigma''\}} W(\{\sigma''\} \rightarrow \{\sigma\}, t) \wp(\{\sigma''\}, t|\{\sigma'\}, s). \quad (8)$$

Moreover, one can work out a master equation for the time  $s$ . It reads

$$\begin{aligned} \wp(\{\sigma\}, t|\{\sigma'\}, s + \Delta t) &= (1 + \Delta t) \wp(\{\sigma\}, t|\{\sigma'\}, s) \\ &- \Delta t \sum_{\{\sigma''\}} W(\{\sigma'\} \rightarrow \{\sigma''\}, s) \wp(\{\sigma\}, t|\{\sigma''\}, s) \end{aligned} \quad (9)$$

and in the continuous-time limit

$$\left(1 - \frac{\partial}{\partial s}\right) \wp(\{\sigma\}, t|\{\sigma'\}, s) = \sum_{\{\sigma''\}} W(\{\sigma'\} \rightarrow \{\sigma''\}, s) \wp(\{\sigma\}, t|\{\sigma''\}, s). \quad (10)$$

This last equation might be obtained simply by using for example the identity  $\frac{\partial}{\partial s} \wp(\{\sigma\}, t) = 0$ .

When the transition rates do not depend on time, the conditional probability  $\wp(\{\sigma\}, t|\{\sigma'\}, s)$  is a function of  $t - s$  only. This can be shown easily by introducing the matrix notation  $\wp(\{\sigma\}, t|\{\sigma'\}, s) = \langle \{\sigma\} | \hat{\wp}(t, s) | \{\sigma'\} \rangle$ . The master equation (8) reads then:

$$\frac{\partial}{\partial t} \hat{\wp}(t, s) = (\hat{W} - \mathbb{1}) \hat{\wp}(t, s) \quad (11)$$

where  $\langle \{\sigma\} | \hat{W} | \{\sigma'\} \rangle = W(\{\sigma\} \rightarrow \{\sigma'\})$ . This equation admits the formal solution

$$\begin{aligned} \wp(\{\sigma\}, t|\{\sigma'\}, s) &= \sum_{\{\sigma''\}} \langle \{\sigma\} | e^{\int_s^t (\hat{W} - \mathbb{1}) dt'} | \{\sigma''\} \rangle \wp(\{\sigma''\}, s|\{\sigma'\}, s) \\ &= \langle \{\sigma\} | e^{(\hat{W} - \mathbb{1})(t-s)} | \{\sigma'\} \rangle \end{aligned} \quad (12)$$

where the initial condition  $\wp(\{\sigma''\}, s|\{\sigma'\}, s) = \delta_{\{\sigma''\}, \{\sigma'\}}$  has been used. This dependence only on  $t - s$ , even far-from-equilibrium, will be used latter in the calculation of the response function.

## 2.2. The model and its dynamics

The Ising model is defined by its equilibrium probability distribution  $\wp_{\text{eq}}(\{\sigma\})$  which reads with general exchange couplings:

$$\wp_{\text{eq}}(\{\sigma\}) = \frac{1}{\mathcal{Z}} e^{-\beta \mathcal{H}(\{\sigma\})} = \frac{1}{\mathcal{Z}} e^{\beta \sum_{k,l < k} J_{kl} \sigma_k \sigma_l}. \quad (13)$$

where ferromagnetic couplings correspond to  $J_{kl} > 0$ . The condition of stationarity  $\frac{\partial}{\partial t} \wp_{\text{eq}}(\{\sigma\}) = 0$  leads according to the master equation (5) to a constrain on the transition rates:

$$\sum_{\{\sigma'\}} \left[ \wp_{\text{eq}}(\{\sigma'\}) W(\{\sigma'\} \rightarrow \{\sigma\}, t) - \wp_{\text{eq}}(\{\sigma\}) W(\{\sigma\} \rightarrow \{\sigma'\}, t) \right] = 0. \quad (14)$$

The equation (14) is satisfied when the detailed balance holds:

$$\wp_{\text{eq}}(\{\sigma'\}) W(\{\sigma'\} \rightarrow \{\sigma\}, t) = \wp_{\text{eq}}(\{\sigma\}) W(\{\sigma\} \rightarrow \{\sigma'\}, t). \quad (15)$$

This last unnecessary but sufficient condition is fulfilled by the heat-bath single-spin flip dynamics defined by the following transition rates:

$$W(\{\sigma\} \rightarrow \{\sigma'\}, t) = \frac{1}{N} \sum_{k=1}^N W_k(\{\sigma\} \rightarrow \{\sigma'\}) \quad (16)$$

where the transition rate for a single spin-flip is

$$W_k(\{\sigma\} \rightarrow \{\sigma'\}) = \left[ \prod_{l \neq k} \delta_{\sigma_l, \sigma'_l} \right] \frac{e^{\beta \sum_{l \neq k} J_{kl} \sigma'_k \sigma'_l}}{\sum_{\sigma = \pm 1} e^{\beta \sum_{l \neq k} J_{kl} \sigma \sigma'_l}}. \quad (17)$$

In this last expression, only the single-spin flip  $\sigma_k \rightarrow \sigma'_k$  is allowed. The product of Kronecker deltas ensures that all other spins are not modified during the transition. After the transition, the spin  $\sigma_k$  takes the new value  $\sigma'_k$  chosen according to the equilibrium probability distribution  $\wp_{\text{eq}}(\{\sigma\})$ . In the case of the Ising chain, the transition rates (16) are equivalent to Glauber's ones [14]. We will use a slightly different dynamics consisting in a sequential update of spins. Let us choose a sequence of lattice sites  $\{\kappa(t) \in \{1, \dots, N\}, \forall t = n\Delta t, n \in \mathbb{N}\}$  and let us define the transition rates in discrete time as

$$W(\{\sigma\} \rightarrow \{\sigma'\}, t) = W_{\kappa(t)}(\{\sigma\} \rightarrow \{\sigma'\}). \quad (18)$$

In comparison to Glauber dynamics, only the spin-flip involving the spin  $\sigma_{\kappa(t)}$  is possible at time  $t$ . In the continuous-time limit, the two dynamics are equivalent up to a rescaling of time  $t \rightarrow t/N$  (found for example in the definition of a Monte Carlo step). Indeed, when iterating  $N$  times the master equation (4), one obtains

$$\begin{aligned} \wp(\{\sigma\}, t + N\Delta t) &= (1 - N\Delta t) \wp(\{\sigma\}, t) \\ &+ \Delta t \sum_{\{\sigma'\}} \wp(\{\sigma'\}, t) \sum_{n=0}^{N-1} W_{\kappa(t+n\Delta t)}(\{\sigma'\} \rightarrow \{\sigma\}) + O(\Delta t^2). \end{aligned} \quad (19)$$

and the Glauber dynamics is recovered if  $\{\kappa(t + n\Delta t)\}_{n=0 \dots N-1}$  is any circular permutation of the set of lattice sites  $\{1, \dots, N\}$ . The equivalence of the two dynamics may not hold in the thermodynamic limit  $N \rightarrow +\infty$ . The time-dependence of the transition rates (18) breaks the time-translation invariance of the conditional probabilities. However, the effective transition rate in equation (19) is time-independent and thus the time-translation invariance is restored in the continuous-time limit if  $\{\kappa(t)\}$  is periodic of period  $N\Delta t$ . Again, this may be no more true in the thermodynamic limit. In the following, we will assume that  $\{\kappa(t)\}$  satisfies the two above-presented conditions, i.e. being periodic of period  $N\Delta t$  and that any  $N$  consecutive values are a circular permutation of  $\{1, \dots, N\}$ .

### 3. Fluctuation-dissipation relation

#### 3.1. Far-from-equilibrium fluctuation-dissipation relation

A magnetic field  $h_i$  is coupled to the spin  $\sigma_i$  between the times  $s$  and  $s + \Delta t$ . During this interval of time, the transition rates are changed to

$$W_{k=\kappa(s)}^h(\{\sigma\} \rightarrow \{\sigma'\}) = \left[ \prod_{l \neq k} \delta_{\sigma_l, \sigma'_l} \right] \frac{e^{\beta \left[ \sum_{l \neq k} J_{kl} \sigma'_k \sigma'_l + h_i \sigma'_k \delta_{k,i} \right]}}{\sum_{\sigma = \pm 1} e^{\beta \left[ \sum_{l \neq k} J_{kl} \sigma \sigma'_l + h_i \sigma \delta_{k,i} \right]}} \quad (20)$$

in order to take into account the additional Zeeman term  $\beta h_i \sigma_i$  in the Hamiltonian of the equilibrium probability distribution (13). The transition rates are all identical to the case  $h_i = 0$  apart from the single-spin flip  $W_i^h$  involving the spin  $\sigma_i$ .

Using the Bayes relation and the discrete-time master equation (4), the average of the spin  $\sigma_j$  at time  $t > s$  can be expanded under the following form:

$$\begin{aligned} \langle \sigma_j(t) \rangle &= \sum_{\{\sigma\}} \sigma_j \wp(\{\sigma\}, t) \\ &= \sum_{\{\sigma\}, \{\sigma'\}} \sigma_j \wp(\{\sigma\}, t | \{\sigma'\}, s + \Delta t) \wp(\{\sigma'\}, s + \Delta t) \\ &= \sum_{\{\sigma\}, \{\sigma'\}} \sigma_j \wp(\{\sigma\}, t | \{\sigma'\}, s + \Delta t) \left[ (1 - \Delta t) \wp(\{\sigma'\}, s) \right. \\ &\quad \left. + \Delta t \sum_{\{\sigma''\}} W_{\kappa(s)}^h(\{\sigma''\} \rightarrow \{\sigma'\}) \wp(\{\sigma''\}, s) \right]. \end{aligned} \quad (21)$$

$W_i^h$  being the only quantity depending on the magnetic field in equation (21), only remains the second term when  $\kappa(s) = i$  after derivating with respect to the magnetic field. The derivative leads then to

$$\begin{aligned} \left[ \frac{\partial \langle \sigma_j(t) \rangle}{\partial h_i} \right]_{h_i \rightarrow 0} &= \Delta t \delta_{\kappa(s), i} \sum_{\substack{\{\sigma\}, \{\sigma'\}, \\ \{\sigma''\}}} \sigma_j \wp(\{\sigma\}, t | \{\sigma'\}, s + \Delta t) \\ &\quad \times \left[ \frac{\partial W_i^h}{\partial h_i}(\{\sigma''\} \rightarrow \{\sigma'\}) \right]_{h_i \rightarrow 0} \wp(\{\sigma''\}, s) \end{aligned} \quad (22)$$

This quantity is the magnetisation on site  $j$  at time  $t$  when an infinitesimal magnetic field is applied to the site  $i$  between  $s$  and  $s + \Delta t$ , i.e. an integrated response function that we will denote  $\chi_{ji}(t, [s; s + \Delta t])$ . The derivative of the transition rate  $W_i^h$  defined by equation (20) is easily taken and reads

$$\left[ \frac{\partial W_i^h}{\partial h_i}(\{\sigma''\} \rightarrow \{\sigma'\}) \right]_{h_i \rightarrow 0} = \beta W_i(\{\sigma''\} \rightarrow \{\sigma'\}) \left[ \sigma'_i - \tanh \left( \beta \sum_{k \neq i} J_{ik} \sigma'_k \right) \right]. \quad (23)$$

It turns out to involve the transition rate of the zero-field dynamics (17). Due to this property, the integrated response function can be expressed in terms of thermodynamic averages of the zero-field dynamics. Inserting (23) into (22), the integrated response function is rewritten as

$$\chi_{ji}(t, [s; s + N\Delta t]) = \beta \Delta t \delta_{\kappa(s), i} \sum_{\substack{\{\sigma\}, \{\sigma'\}, \\ \{\sigma''\}}} \sigma_j \wp(\{\sigma\}, t | \{\sigma'\}, s + \Delta t)$$

$$\times \left[ \sigma'_i - \tanh \left( \beta \sum_{k \neq i} J_{ik} \sigma'_k \right) \right] W_i(\{\sigma''\} \rightarrow \{\sigma'\}) \wp(\{\sigma''\}, s) \quad (24)$$

The summation over  $\{\sigma''\}$  can be performed by using the discrete-time master equation (4). One obtains

$$\begin{aligned} \chi_{ji}(t, [s; s + N\Delta t]) &= \beta \delta_{\kappa(s), i} \sum_{\{\sigma\}, \{\sigma'\}} \sigma_j \wp(\{\sigma\}, t | \{\sigma'\}, s + \Delta t) \left[ \sigma'_i - \tanh \left( \beta \sum_{k \neq i} J_{ik} \sigma'_k \right) \right] \\ &\quad \times \left[ \wp(\{\sigma'\}, s + \Delta t) - (1 - \Delta t) \wp(\{\sigma'\}, s) \right] \end{aligned} \quad (25)$$

Using a Taylor-expansion of  $\wp(\{\sigma'\}, s)$  in the vicinity of  $s + \Delta t$ , equation (25) can be rewritten to lowest order in  $\Delta t$  as

$$\begin{aligned} \chi_{ji}(t, [s; s + N\Delta t]) &= \beta \Delta t \delta_{\kappa(s), i} \sum_{\{\sigma\}, \{\sigma'\}} \sigma_j \wp(\{\sigma\}, t | \{\sigma'\}, s + \Delta t) \left[ \sigma'_i - \tanh \left( \beta \sum_{k \neq i} J_{ik} \sigma'_k \right) \right] \\ &\quad \times \left[ \wp(\{\sigma'\}, s + \Delta t) + \frac{\partial \wp}{\partial s}(\{\sigma'\}, s + \Delta t) \right]. \end{aligned} \quad (26)$$

The time-translation invariance of conditional probabilities being restored in the continuous-time limit, they are function of  $t - s$  only and thus satisfy the property

$$\frac{\partial \wp}{\partial s}(\{\sigma\}, t | \{\sigma'\}, s) = -\frac{\partial \wp}{\partial t}(\{\sigma\}, t | \{\sigma'\}, s). \quad (27)$$

The term involving the time-derivative in equation (26) can thus be rewritten in the continuous-time limit as

$$\begin{aligned} \wp(\{\sigma\}, t | \{\sigma'\}, s) \frac{\partial \wp}{\partial s}(\{\sigma'\}, s) &= \frac{\partial}{\partial s} \left[ \wp(\{\sigma\}, t | \{\sigma'\}, s) \wp(\{\sigma'\}, s) \right] \\ &\quad - \underbrace{\frac{\partial \wp}{\partial s}(\{\sigma\}, t | \{\sigma'\}, s) \wp(\{\sigma'\}, s)}_{= + \frac{\partial \wp}{\partial t}(\{\sigma\}, t | \{\sigma'\}, s)}. \end{aligned} \quad (28)$$

Moreover, the integrated response function  $\chi_{ji}(t, [s; s + \Delta t])$  goes to the response function  $R_{ji}(t, s)$  in the continuous-time limit :

$$\chi_{ji}(t, [s; s + \Delta t]) = \int_s^{s+\Delta t} R_{ji}(t, u) du = R_{ji}(t, s) \Delta t + O(\Delta t^2). \quad (29)$$

Combining equations (26), (28) and (29), the response function reads in the continuous-time limit

$$R_{ji}(t, s) = \beta \left( 1 + \frac{\partial}{\partial s} + \frac{\partial}{\partial t} \right) \langle \sigma_j(t) [\sigma_i(s) - \sigma_i^{\text{Weiss}}(s)] \rangle \delta_{\kappa(s), i} \quad (30)$$

where  $\sigma_i^{\text{Weiss}}(s) = \tanh \left( \beta \sum_{k \neq i} J_{ik} \sigma_k(s) \right)$  is the equilibrium value of the spin  $\sigma_i$  in the Weiss field created by all other spins at time  $s$ . Relation (30) generalises equation (1). The response function  $R_{ji}(t, s)$  turns out to be related to time-derivatives of the correlation function of the spin  $\sigma_j$  at time  $t$  with the fluctuations of the spin  $\sigma_i$  at time  $s$  around the equilibrium average  $\sigma_i^{\text{Weiss}}(s)$  of this spin in its Weiss field. In this sense, this relation is still a fluctuation-dissipation relation but valid far-from-equilibrium. No assumption has been made on the dimension of the space or on the set of exchange couplings  $J_{kl}$  during the calculation. Moreover, it applies for any initial conditions  $\wp(\{\sigma\}, 0)$ . The appearance of the prefactor  $1 + \frac{\partial}{\partial s} + \frac{\partial}{\partial t}$  is not related to the equilibrium



probability distribution of the model but comes from the Markovian properties of the dynamics. Generalised response functions are easily calculated along the same lines than equation (30). The second-order term for example reads

$$\begin{aligned} R_{kji}^{(2)}(t, s, r) &= \left( \frac{\delta^2 \langle \sigma_k(t) \rangle}{\delta h_j(s) \delta h_i(r)} \right)_{h \rightarrow 0} \quad (t > s > r) \\ &= \beta \left( 1 + \frac{\partial}{\partial s} + \frac{\partial}{\partial t} \right) \left( 1 + \frac{\partial}{\partial r} + \frac{\partial}{\partial s} + \frac{\partial}{\partial t} \right) \langle \sigma_k(t) \delta \sigma_j(s) \delta \sigma_i(r) \rangle \delta_{\kappa(r), j} \delta_{\kappa(s), i} \end{aligned} \quad (31)$$

where  $\delta \sigma_j(s) = \sigma_j(s) - \sigma_j^{\text{Weiss}}(s)$ . Calculation of non-linear terms requires higher-order derivatives of the transition rate as for example

$$\begin{aligned} R_{jii}^{(2)}(t, s, s) &= \left( \frac{\delta^2 \langle \sigma_j(t) \rangle}{\delta h_i^2(s)} \right)_{h \rightarrow 0} \quad (t > s) \\ &= -2\beta^2 \left( 1 + \frac{\partial}{\partial s} + \frac{\partial}{\partial t} \right) \langle \sigma_j(t) \sigma_i^{\text{Weiss}}(s) \delta \sigma_i(s) \rangle \delta_{\kappa(s), i}. \end{aligned} \quad (32)$$

These relations are moreover easily extended to other models. The relations (30) to (32) hold for the  $O(n)$  or the  $q$ -state Potts for example where  $\sigma_i(s)$  has to be replaced by the local order parameter at time  $s$  on the site  $i$  and  $\sigma_i^{\text{Weiss}}(s)$  by its average value in the Weiss field. Since equations (30) to (32) involve a constraint on the sequence of spin-flips, their generalisation to the Ising-Glauber model is not trivial. However, they will be of great interest for Monte Carlo simulations.

### 3.2. Equilibrium limit

We will show in this section that the usual expression of the FDT (1) is recovered in the equilibrium limit. At equilibrium, the probability distribution  $\wp_{\text{eq}}(\{\sigma\})$  does not depend on time. As a consequence, the integrated response function can be written according to equation (26) as

$$\begin{aligned} \chi_{ji}^{\text{eq}}(t, [s; s + \Delta t]) &= \beta \Delta t \delta_{\kappa(s), i} \sum_{\{\sigma\}, \{\sigma'\}} \sigma_j \wp(\{\sigma\}, t | \{\sigma'\}, s + \Delta t) \\ &\quad \times \left[ \sigma'_i - \tanh \left( \beta \sum_{k \neq i} J_{ik} \sigma'_k \right) \right] \wp_{\text{eq}}(\{\sigma'\}) \end{aligned} \quad (33)$$

The hyperbolic tangent can be expressed in terms of the transition ratio of the zero-field dynamics (17):

$$\begin{aligned} \tanh \left( \beta \sum_{k \neq i} J_{ik} \sigma'_k \right) \wp_{\text{eq}}(\{\sigma'\}) &= \sum_{\{\sigma''\}} \sigma''_i \frac{\left[ \prod_{k \neq i} \delta_{\sigma''_k, \sigma'_k} \right] e^{\beta \sum_{k \neq i} J_{ik} \sigma''_i \sigma'_k} e^{\beta \sum_{k, l < k} J_{kl} \sigma''_k \sigma''_l}}{\sum_{\sigma = \pm 1} e^{\beta \sum_{k \neq i} J_{ik} \sigma \sigma'_k}} \frac{1}{\mathcal{Z}} \\ &= \sum_{\{\sigma''\}} \sigma''_i W_i(\{\sigma''\} \rightarrow \{\sigma'\}) \wp_{\text{eq}}(\{\sigma''\}). \end{aligned} \quad (34)$$

Inserting in equation (33), the integrated response function reads

$$\begin{aligned} \chi_{ji}^{\text{eq}}(t, [s; s + \Delta t]) &= \beta \Delta t \delta_{\kappa(s), i} \left[ \sum_{\{\sigma\}, \{\sigma'\}} \sigma_j \wp(\{\sigma\}, t | \{\sigma'\}, s + \Delta t) \sigma'_i \wp_{\text{eq}}(\{\sigma'\}) \right. \\ &\quad \left. - \sum_{\substack{\{\sigma\}, \{\sigma'\}, \\ \{\sigma''\}}} \sigma_j \wp(\{\sigma\}, t | \{\sigma'\}, s + \Delta t) \sigma''_i W_i(\{\sigma''\} \rightarrow \{\sigma'\}) \wp_{\text{eq}}(\{\sigma''\}) \right]. \end{aligned} \quad (35)$$

The first term can be expressed as a thermodynamic average while in the second, one needs to get rid first of the transition rate. The Kronecker delta constrains the only possible spin-flip to involve site  $i$  at time  $s$ . As a consequence,  $W_i(\{\sigma''\} \rightarrow \{\sigma'\})$  can be replaced by  $W_{\kappa(s)}(\{\sigma''\} \rightarrow \{\sigma'\})$  and the master equation (9) can be applied to equation (35). Moreover, one can show that

$$\begin{aligned} \wp(\{\sigma\}, t | \{\sigma''\}, s) &= (1 - \Delta t) \wp(\{\sigma\}, t | \{\sigma''\}, s + \Delta t) \\ &+ \Delta t \sum_{\{\sigma'\}} W(\{\sigma''\} \rightarrow \{\sigma'\}, s) \wp(\{\sigma\}, t | \{\sigma'\}, s + \Delta t) + O(\Delta t^2). \end{aligned} \quad (36)$$

This relation is obtained by first putting alone  $\wp(\{\sigma\}, t | \{\sigma''\}, s)$  in the left member of the master equation (9) and then by iterating the relation to make disappear  $\wp(\{\sigma\}, t | \{\sigma''\}, s)$  in the right member. Equation (36) is then used to eliminate the transition rate from equation (35) :

$$\begin{aligned} \chi_{ji}^{\text{eq}}(t, [s; s + \Delta t]) &= \beta \delta_{\kappa(s), i} \left[ \Delta t \sum_{\{\sigma\}, \{\sigma'\}} \sigma_j \wp(\{\sigma\}, t | \{\sigma'\}, s + \Delta t) \sigma'_i \wp_{\text{eq}}(\{\sigma'\}) \right. \\ &- \sum_{\{\sigma\}, \{\sigma'\}} \sigma_j \wp(\{\sigma\}, t | \{\sigma'\}, s) \sigma'_i \wp_{\text{eq}}(\{\sigma'\}) \\ &\left. + (1 - \Delta t) \sum_{\{\sigma\}, \{\sigma'\}} \sigma_j \wp(\{\sigma\}, t | \{\sigma'\}, s + \Delta t) \sigma'_i \wp_{\text{eq}}(\{\sigma'\}) \right]. \end{aligned} \quad (37)$$

The two terms of order  $\Delta t$  cancel and it remains only

$$\chi_{ji}^{\text{eq}}(t, [s; s + \Delta t]) = \beta \delta_{\kappa(s), i} \langle \sigma_j(t) [\sigma_i(s + \Delta t) - \sigma_i(s)] \rangle_{\text{eq}} \quad (38)$$

and in the continuous-time limit, one obtains equilibrium fluctuation-dissipation:

$$R_{ji}^{\text{eq}}(t, s) = \beta \delta_{\kappa(s), i} \frac{\partial}{\partial s} \langle \sigma_j(t) \sigma_i(s) \rangle_{\text{eq}} = \beta \delta_{\kappa(s), i} \langle \sigma_j(t) [\sigma_i(s) - \sigma_i^{\text{Weiss}}(s)] \rangle_{\text{eq}} \quad (39)$$

where the last member is simply equation (30) at equilibrium. One recovers the usual equilibrium fluctuation-dissipation relation up to a Kronecker delta due the fact that the response function is non-zero only for times at which a spin-flip involving the spin connected to the magnetic field occurs.

#### 4. Monte Carlo simulations of the 2d-Ising model

The discrete-time analogous of expression (30) of the response function enables to study the aging displayed by the Ising-Glauber model more accurately than in previous works that were based on the numerical estimate of the integrated response function. In the first part of this section, the algorithm is given. In the second part, simulations of the Glauber dynamics of the two-dimensional Ising model during a quench from the paramagnetic phase to the ferromagnetic one are presented. The system is expected to display aging, associated with the existence of growing domains corresponding to competing ferromagnetic states [15]. Reversible processes occur in the bulk of domains while domain wall rearrangements are irreversible. We will distinguish between quenches at the critical temperature  $T_c$  and below. In both cases, lattice sizes  $128 \times 128$ ,  $256 \times 256$

and  $362 \times 362$  were simulated and the data averaged over 3000, 10000 and 5000 initial configurations respectively. For all data, error bars were estimated as the standard deviation around the average value.

#### 4.1. Discrete response function

During a Monte Carlo simulation, the time is a discrete variable and the time step is set to  $\Delta t = 1$ . Monte Carlo simulations implement indeed the Markov process defined by the master equation (4) with the choice  $\Delta t = 1$ . Since simulations are always made on finite systems, dynamics with sequential and parallel updates are equivalent in the large-time limit up to a time-renormalisation corresponding to the definition of a Monte Carlo Step (MCS). The response function can only be defined for continuous time processes. However, the integrated response function during one spin-flip  $\sigma_i \rightarrow \sigma'_i$  is the best estimator for the response function  $R_{ij}(t, s)$  that we can define. Inserting  $\Delta t = 1$  into equation (25), the estimator of the response function is simply

$$\chi_{ji}(t, [s; s+1]) = \beta \delta_{\kappa(s), i} \langle \sigma_j(t) [\sigma_i(s+1) - \sigma_i^{\text{Weiss}}(s+1)] \rangle \quad (40)$$

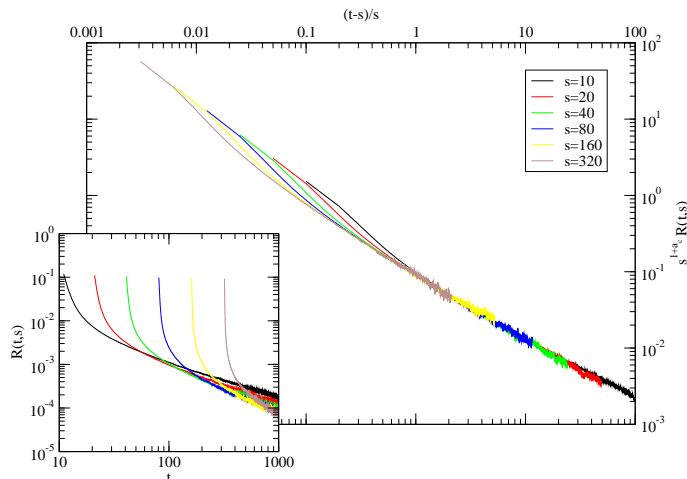
where  $\sigma_i^{\text{Weiss}}(s) = \tanh\left(\beta \sum_{k \neq i} J_{ik} \sigma'_k(s)\right)$ . In the following, we will be interested only on response functions of the form  $R_{ii}(t, s)$ . In order to reduce statistical fluctuations, we have then estimated the response function  $R(t, s)$  as the average over all spin-flips during one MCS:

$$R(t, s) = \frac{1}{N} \sum_{n=0}^{N-1} \chi_{\kappa(s+n)\kappa(s+n)}(t, [s+n; s+n+1]) \quad (41)$$

The calculation of this quantity is quite simple. Let evolve the simulation until time  $s$ . For each of the  $N$  next spin-flips  $\sigma_i \rightarrow \sigma'_i$ , store the quantity  $\sigma'_i - \sigma_i^{\text{Weiss}}$ . Note that  $\sigma'_i$  may be equal to  $\sigma_i$  meaning that the system has not changed during this time step. However, in strict application of equation (40), one has nevertheless to store  $\sigma_i - \sigma_i^{\text{Weiss}}$ . After  $N$  spin-flips, let the system evolve again until time  $t$ . Calculate the response function for each site  $i$  by multiplying the quantity  $\sigma'_i - \sigma_i^{\text{Weiss}}$  stored by the new value of the spin  $\sigma_i$  and add all these one-site response functions. Repeat the simulation as many times as necessary and average the results. The integrated response function can be easily calculated by numerical integration of the response function.

The time-derivative of the correlation function  $\frac{\partial}{\partial s} C_{ji}(t, s)$  at time  $s$  can be estimated by  $\langle \sigma_j(t) [\sigma_i(s+1) - \sigma_i(s)] \rangle$ . Again, this quantity is averaged over all spin-flips during one MCS. The FD ratio (2) can be estimated as

$$X(t, s) = \frac{R(t, s)}{\beta \frac{\partial}{\partial s} C(t, s)} = \frac{\sum_{n=0}^{N-1} \langle \sigma_{\kappa(s+n)}(t) [\sigma_{\kappa(s+n)}(s+1) - \sigma_{\kappa(s+n)}^{\text{Weiss}}(s+1)] \rangle}{\beta \sum_{n=0}^{N-1} \langle \sigma_{\kappa(s+n)}(t) [\sigma_{\kappa(s+n)}(s+1) - \sigma_{\kappa(s+n)}(s)] \rangle}. \quad (42)$$



**Figure 1.** Response function of the 2D-Ising model during a quench at the critical temperature (inset) and collapse of the scaling function for different values of  $s$ . The data were obtained with a lattice of size  $362 \times 362$  and averaged over 5000 initial configurations. Each curve is surrounded by a clouds of dots corresponding to the lower and upper bounds of the error bars. The value  $a_c = 0.115$  was used [17].

#### 4.2. Quench at the critical temperature

During a quench at the critical temperature  $T_c$ , the asymptotic decay of the correlation function has been conjectured to be [16, 17]

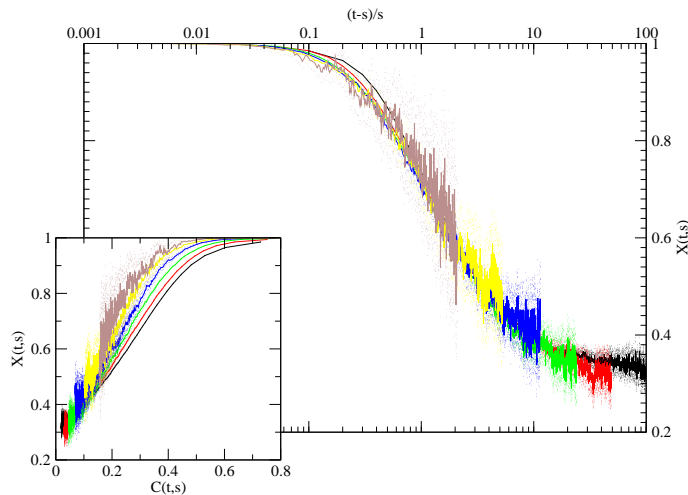
$$C(t, s) \underset{t, s \gg 1}{\sim} s^{-a_c} \mathcal{C}_c(t/s) \quad (43)$$

where  $a_c = \frac{2\beta}{\nu z_c}$  and  $\mathcal{C}_c(x)$  is a scaling function that asymptotically behaves as  $\mathcal{C}_c(x) \underset{x \gg 1}{\sim} x^{-\lambda_c/z_c}$ .  $\lambda_c$  is the critical autocorrelation exponent [18] and  $z_c$  the dynamical exponent. Similarly, the asymptotic behaviour of the response function is

$$R(t, s) \underset{t, s \gg 1}{\sim} s^{-1-a_c} \mathcal{R}_c(t/s) \quad (44)$$

where the scaling function  $\mathcal{R}_c(x)$  behaves asymptotically as  $\mathcal{R}_c(x) \underset{x \gg 1}{\sim} x^{-\lambda_c/z_c}$  too. By integrating over  $s$ , one obtains a relation similar to (44) for the integrated response function that has been checked by large-scale Monte Carlo simulations [19]. However, the relation (44) is asymptotic so is not expected to hold for the response function  $R(t, s)$  with small values of  $s$  that are the main contribution to the integrated response function. As a consequence, it is difficult to test the asymptotic behaviour of the response function in this way. Our approach permits us to avoid these problems and to test directly the relation (44).

The numerical data are plotted in figure 1. For the largest lattice size ( $L = 362$ ) and the smallest value of  $s$  ( $s = 10$ ), errors bars are at most 6 % of the value of the response function while for the largest ( $s = 320$ ), they increase up to 12 %. Indeed, in the last case, the response function is of order of  $1/N_{\text{config}}$  and so can not be sampled accurately. Nevertheless, a fairly good collapse of the data is observed indicating that



**Figure 2.** FD ratio  $X(t, s)$  versus  $t$  for different values  $s$  for the Ising-Glauber model quenched at the critical temperature  $T_c$ . The data were obtained for a lattice  $362 \times 362$  and averaged over 5000 initial configurations. Each curve is surrounded by a clouds of dots corresponding to the lower and upper bounds of the error bars.

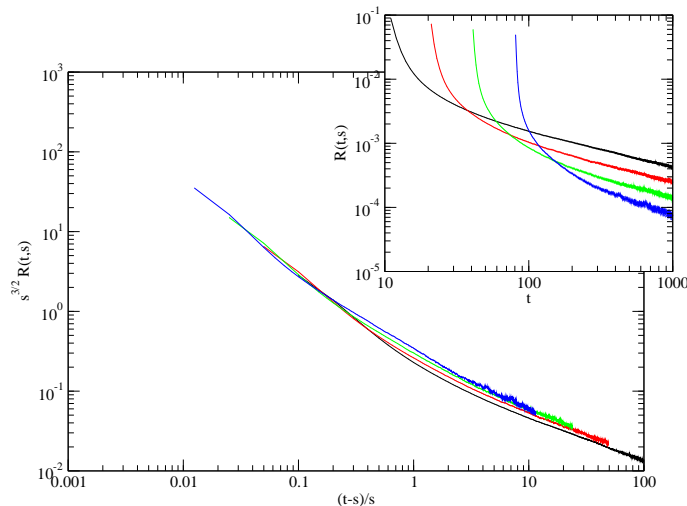
$\mathcal{R}_c(t/s) = s^{1+a_c} R(t, s)$  is indeed a scaling function (actually we have used  $(t - s)/s$  instead of  $t/s$  but this has no consequence on the asymptotic behaviour).

We computed the FD ratio  $X(t, s)$  using the estimator previously derived and whose expression is given by equation (42). The error bars are quite large. The numerical data are plotted in figure 2 for the largest lattice size ( $L = 362$ ). In contradistinction to Cugliandolo conjecture (2), the inset of figure 2 shows that the FD ratio does not depend on time only through the correlation function. However, it seems that it may be the case in the limit  $C(t, s) \rightarrow 0$ . On the other hand, it seems that the FD ratio depends on time only through  $t/s$  and reach a plateau for large enough values of  $t$  that we may estimate roughly to be  $X_\infty \simeq 0.33(2)$ . The same value is obtained for  $L = 256$  and  $L = 362$  excluding any possibility of finite-size effects. The limit  $X_\infty$  has been conjectured to be universal [17] but incompatible values have been given by different groups:  $X_\infty = 0.26(1)$  [20] and  $X_\infty = 0.340(5)$  [21] by Monte Carlo simulations and  $X_\infty \simeq 0.35$  [22] for the O(1)-model in dimension  $d = 4 - \epsilon$ . Our estimate is compatible with the last two ones. The estimate  $X_\infty = 0.26(1)$  has probably been measured for a too-short time  $t$ , far from the region where Cugliandolo conjecture (2) and thus equation (3) hold. This puts stress upon the danger of using equation (3) to compute the FD ratio.

#### 4.3. Quench below the critical temperature

The same analysis can be done below  $T_c$ . In this regime, The correlation function decays as [16, 17]

$$C(t, s) \underset{t, s \gg 1}{\sim} M_{\text{eq}}^2 \mathcal{C}(t/s) \quad (45)$$



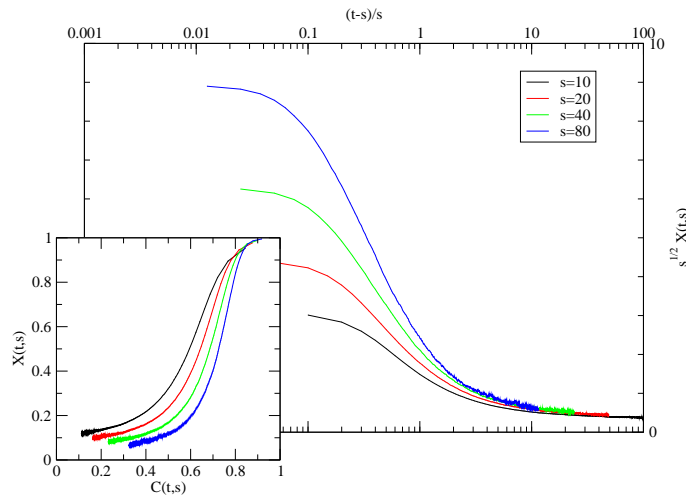
**Figure 3.** Response function of the 2D-Ising model during a quench at the temperatures  $T = J/0.6 \simeq \frac{3}{4}T_c$  (inset) and collapse of the scaling function for different values of  $s$ . The data were obtained for a lattice  $256 \times 256$  and averaged over 10000 initial configurations. Each curve is surrounded by a clouds of dots corresponding to the lower and upper bounds of the error bars.

where  $M_{\text{eq}}$  is the equilibrium magnetisation and  $\mathcal{C}(x)$  a scaling function that asymptotically behaves as  $\mathcal{C}(x) \underset{x \gg 1}{\sim} x^{-\lambda/z}$ . The autocorrelation exponent  $\lambda$  and the dynamical exponent  $z$  are expected to take values which are different from those at  $T_c$ . The response function is expected to scale as [16, 17]

$$R(t, s) \underset{t, s \gg 1}{\sim} s^{-1-a} \mathcal{R}(t/s) \quad (46)$$

where  $\mathcal{R}(x) \underset{x \gg 1}{\sim} x^{-\lambda/z}$ . A controversy exists concerning the value of  $a$  that has been estimated to be either  $1/4$  [23] or  $1/2$  [24, 25]. Our numerical data are presented in the figure 3. We studied lattice sizes only up to  $L = 256$  but calculations were made for two temperatures:  $J/0.6 \simeq \frac{3}{4}T_c$  and  $J/0.9 \simeq T_c/2$ . The error bars are much smaller than in the critical case for small values of  $s$ . The relative error is at most 2.6 % for  $s = 10$  at  $\frac{3}{4}T_c$  but increase faster with  $s$ : the relative error increases up to 11 % for  $s = 80$ . As a consequence, the study was limited to the values of  $s$  ranging from  $s = 10$  to  $s = 80$ . The response function displays the expected scaling behaviour (46) with  $a = 1/2$ . However, the collapse is not perfect, especially for the smallest values of  $s$  but a very small variation of  $a$  does not improve it significantly. The value  $a = 1/4$  in particular improves the collapse for the small values of  $s$  only. The response function has probably strong corrections to scaling. Note that corrections have already been taken into account for the study of the scaling behaviour of the integrated response function [19, 26].

Combining the relations (45) and (46), the FD ratio  $X(t, s) = R(t, s)/\beta \frac{\partial}{\partial s} C(t, s)$  is predicted to vanish as  $s^{-a}$  below  $T_c$ . Our numerical estimates for  $T = J/0.6 \simeq \frac{3}{4}T_c$  are plotted in figure 4. The statistical errors decrease with the temperature so that the



**Figure 4.** FD ratio  $X(t, s)$  versus  $t$  for different values  $s$  for the Ising-Glauber model quenched at the temperature  $J/0.6k_B \simeq \frac{3}{4}T_c$ . The data were obtained for a lattice  $256 \times 256$  and averaged over 10000 initial configurations. Each curve is surrounded by a clouds of dots corresponding to the lower and upper bounds of the error bars.

data are less fluctuating than at  $T_c$ . As expected, the FD ratio is equal to 1 for small values of  $t - s$ , signalling that the main contribution to the response function is due to equilibrium processes. On the other hand, it vanishes in the limit  $t \sim s \gg 1$  as  $s^{-1/2}$ . As shown in figure 4, the data for  $s^{1/2}X(t, s)$  collapse for large values of  $t/s$ . Moreover, figure 4 shows unambiguously that the FD ratio does not depend on time only through the correlation function. This makes the relation (3) invalid. The study of the violation of the equilibrium FDT by the usual method relying on the equation (3) would have led to erroneous values of the FD ratio.

## 5. Conclusion

Using a formalism similar to Kubo's one in the quantum case, we derive an exact expression of the response function of an Ising-Glauber-like model far-from-equilibrium (equation (30)). At least for finite systems, the dynamics of our model is equivalent to the Glauber dynamics up to a time-renormalisation  $t \rightarrow t/N$ . The derivation is possible because the dynamics consists in a sequential update of the spins and the transition rate under a magnetic field can be written as a product of the transition rate without magnetic field and of a term depending only on the final spin configuration. The response function turns out to be related to time-derivatives of a correlation function involving the fluctuations of the spin excited by the magnetic field around its equilibrium average in its Weiss field. In this sense, the expression is a generalisation of the equilibrium fluctuation-dissipation. Our expression is quite general: no assumption has been made during its derivation on the dimension of the space, the set of exchange couplings or the initial conditions. Moreover, it can be easily extended to other classical models. Generalised and non-linear response functions can be obtained analogously. However,

the continuous-time expression (30) may not hold in the thermodynamic limit. Analytic results would be desirable. Unfortunately, the response function calculated for the Ising-chain by Glauber itself in his original paper [14] does not help because the magnetic field was coupled differently to the system (by a multiplicative factor to make the calculation feasible while we coupled the field by a modification of the transition rate corresponding to the addition of the Zeeman interaction in the equilibrium probability distribution). Generalisation to the Ising-Glauber model is not trivial because equation (39) sets a constrain on the sequence of spin-flips. Nevertheless, It is tempting to imagine that like the equilibrium FDT (39), equations (30) to (32) hold for the Ising-Glauber model when suppressing this constrain on the sequence of spin-flips.

The expression (30) of the response function is then implemented in Monte Carlo simulations. Our approach gives access to the response function and the FD ratio directly. In particular, the FD ratio can be obtained without assuming the validity of the Cugliandolo conjecture (2). We then study numerically the homogeneous two-dimensional Ising-Glauber model quenched from the paramagnetic phase to the ferromagnetic one. Both the response function and the FD ratio display the expected scaling behaviour both at  $T_c$  and below  $T_c$ . The values, still controversial, of  $a$  and  $X_\infty$  are estimated to be equal to  $1/2$  and  $0.33(2)$  respectively, in agreement with some previous works. The Cugliandolo conjecture (2) does not hold for this model apart perhaps at  $T_c$  in the limit of vanishing correlation functions. This would explain discrepancies of previous estimates of  $X_\infty$  relying on Cugliandolo conjecture. The above-presented numerical procedure may be extended to many different systems and would provided a unambiguous test of Cugliandolo conjecture. We are currently studying the dynamics of spin-glasses in this framework.

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