Nagel Scaling, Relaxation, and Universality in the Kinetic Ising Model on an Alternating Isotopic Chain

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The dynamic critical exponent and the frequency and wave-vector dependent susceptibility of the kinetic Ising model on an alternating isotopic chain with Glauber dynamics are examined. The analysis provides a connection between a microscopic model and the Nagel scaling curve originally proposed to describe dielectric susceptibility measurements of several glass-forming liquids. While support is given to the hypothesis relating the Nagel scaling to multiple relaxation processes, it is also found that the scaling function may exhibit plateau regions and does not hold for all temperatures.

Experimental work on dielectric relaxation in glass-forming liquids has in recent years been reported in terms of a new (thought to be universal) scaling function [1] which is presumed to be related to multifractal scaling. While the more usual normalized Debye scaling in terms of a single relaxation time is very simple (one chooses to scale the frequency with the inverse of the relaxation time and the real and imaginary parts are then divided by their values at zero and one, respectively), in the so-called Nagel plot the abscissa is $(1 + W)\log_{10}(\omega/\omega_p)/W^2$ and the ordinate is $\log_{10}[\chi''(\omega)\omega_p/\omega\Delta\chi]/W$. Here, χ'' is the imaginary part of the susceptibility, W is the full width at half maximum of χ'', ω is the frequency and ω_p the one corresponding to the peak in χ'', and $\Delta\chi = \chi(0) - \chi_\infty$ is the static susceptibility. Despite its undeniable phenomenological success, such scaling is not quite well understood on a physical basis. The authors of this proposal advance the idea that the presence of more than one relaxation process is not alien to this form and thus suggest that multifractality such as the one present in theories of chaos may well be behind the new scaling. In order to gain some insight into the physical origin of the Nagel scaling, it seems appropriate to consider simple but well established models in which both universal features are unquestionable and more than one relaxation mechanism is present. A good candidate may be found among kinetic Ising models.

The scaling hypothesis of Halperin and Hohenberg [2] relates the time scale τ and the correlation length ξ and introduces the dynamic critical exponent z. In the case of Ising models, z was for a long time believed to be universal, depending on the nature of conserved quantities and of those features—for instance, dimensionality [3]—which determine their static universality class. However, it is now well established that for some simple systems this exponent is nonuniversal [4–10]. In particular, in the case of one-dimensional Glauber dynamics [11] the alternating isotopic chain [8] presents universal behavior (in the sense that it leads to the same value of the dynamic critical exponent as the homogeneous chain), whereas the alternating-bond chain does not [8–19] (see, however, Ref. [20]). Because of the fact of this universality of z, the isotopic alternating Ising chain with Glauber dynamics provides a test model in which to assess the value of the connection between multiple relaxation mechanisms and the Nagel plot. The model consists of a closed linear chain with N sites occupied by two isotopes (characterized by two different spin relaxation times) that are alternately arranged. The Hamiltonian is the usual Ising Hamiltonian given by

$$H = -J \sum_{j=1}^{N} \sigma_j \sigma_{j+1}, \tag{1}$$

where σ_j is a stochastic (time-dependent) spin variable assuming the values ± 1 and J the coupling constant. The configuration of the chain is specified by the set of values $\{\sigma_1, \sigma_2, \ldots, \sigma_N\} \equiv \{\sigma^N\}$ at time t. This configuration evolves in time due to interactions with a heat bath. We assume for this chain the usual Glauber dynamics so that the transition probabilities are given by

$$w_i(\sigma_i) = \alpha_i\left[1 - \frac{\gamma}{2}(\sigma_{i-1}\sigma_i + \sigma_i\sigma_{i+1})\right], \tag{2}$$

where $\gamma = \tanh(2J/k_BT)$, k_B being the Boltzmann constant and T the absolute temperature, and α_i is the inverse

of the relaxation time τ_i of spin i in the absence of spin interaction. If we now let α_1 and α_2 represent the inverses of the free spin relaxation times of chains composed solely of spins of species 1 or species 2, respectively, then we can set $\alpha_i = \overline{\alpha}_1 - (-1)^i \overline{\alpha}_2$, where $\overline{\alpha}_1 = (\alpha_1 + \alpha_2)/2$ and $\overline{\alpha}_2 = (\alpha_1 - \alpha_2)/2$.

The time dependent probability $P(\{\sigma^N\}, t)$ for a given spin configuration satisfies the master equation

$$\frac{dP(\{\sigma^N\}, t)}{dt} = - \sum_{i=1}^{N} w_i(\sigma_i) P(\{\sigma^N\}, t)$$
$$+ \sum_{i=1}^{N} w_i(-\sigma_i) P(T_i\{\sigma^N\}, t), \quad (3)$$

where $T_i\{\sigma^N\} \equiv \{\sigma_1, \sigma_2, \ldots, \sigma_{i-1}, -\sigma_i, \sigma_{i+1}, \ldots, \sigma_N\}$. The dynamical properties we are interested in, namely the dynamic critical exponent and the susceptibility, require the knowledge of some moments of the probability $P(\{\sigma^N\}, t)$. Hence, we introduce the following expectation values and correlation functions defined as

$$q_i(t) = \langle \sigma_i(t) \rangle = \sum_{\{\sigma^N\}} \sigma_i P(\{\sigma^N\}, t), \quad (4)$$

$$r_{i,j}(t) = \langle \sigma_i(t) \sigma_j(t) \rangle = \sum_{\{\sigma^N\}} \sigma_i \sigma_j P(\{\sigma^N\}, t), \quad (5)$$

and

$$c_{i,j}(t', t' + t) = \Theta(t) \langle \sigma_i(t') \sigma_j(t' + t) \rangle$$
$$= \sum_{\{\sigma^N\}, \{\sigma^{N'}\}} \sigma_i' P(\{\sigma^{N'}\}, t')$$
$$\times \sigma_j p(\{\sigma^N\}|\{\sigma^{N'}\}, t), \quad (6)$$

where $\Theta(t)$ is the Heaviside step function and the sums run over all possible configurations. The second equality of Eq. (6), which gives the formal definition of the time-delayed correlation function, involves $p(\{\sigma^N\}|\{\sigma^{N'}\}, t)$, the conditional probability of the chain having the configuration $\{\sigma^N\}$ at time $t' + t$ provided it had the configuration $\{\sigma^{N'}\} = \{\sigma_1', \sigma_2', \ldots, \sigma_N'\}$ at time t'. Multiplying the master equation by the appropriate quantities and performing the required summations we obtain the set of time evolution equations that will be used in our later development. These are given by

$$\frac{dq_j}{dt} = -\alpha_j \left[q_j - \frac{\gamma}{2} (q_{j-1} + q_{j-1}) \right] \quad (7)$$

and

$$\frac{dc_{i,j}(t', t' + t)}{dt} = r_{i,j}(t') \delta(t) - \alpha_j c_{i,j}(t', t' + t)$$
$$+ \frac{\alpha_j \gamma}{2} [c_{i,j-1}(t', t' + t)$$
$$+ c_{i,j+1}(t', t' + t)]. \quad (8)$$

We now impose translational invariance and introduce $\tilde{q}_k$, the (spatial) Fourier transform of q_j, the $t' \to \infty$ limit of the (temporal) Fourier transform of

$c_l(t', t' + t) \equiv c_{i,j}(t', t' + t)$ (with $l = j - i$) denoted by $\hat{c}_l(\omega)$, and $\tilde{C}_k(\omega)$, the spatial Fourier transform of $\hat{c}_l(\omega)$, defined through

$$q_j = \frac{1}{\sqrt{N}} \sum_k \tilde{q}_k \exp(ikj), \quad (9)$$

$$\hat{c}_l(\omega) = \lim_{t' \to \infty} \frac{1}{2\pi} \int_{-\infty}^{\infty} c_l(t', t' + t) \exp(-i\omega t) \, dt, \quad (10)$$

and

$$\tilde{C}_k(\omega) \equiv \langle \sigma_{-k} \sigma_k \rangle_\omega = \frac{1}{N} \sum_l \hat{c}_l(\omega) \exp(-ikl). \quad (11)$$

In terms of these quantities, Eqs. (7) and (8) may be rewritten, respectively, as

$$\frac{d\Psi_k}{dt} = \mathbf{M}_k \Psi_k \quad (12)$$

and

$$i\omega \hat{c}_l(\omega) = r_l^\infty$$
$$- \alpha_l \left(\hat{c}_l(\omega) - \frac{\gamma}{2} [\hat{c}_{l-1}(\omega) + \hat{c}_{l+1}(\omega)] \right), \quad (13)$$

where

$$\Psi_k = \begin{pmatrix} \tilde{q}_k \\ \tilde{q}_{k-\pi} \end{pmatrix}, \quad (14)$$

$$\mathbf{M}_k = \begin{bmatrix} -\overline{\alpha}_1(1 - \gamma \cos k) & -\overline{\alpha}_2(1 + \gamma \cos k) \\ -\overline{\alpha}_2(1 - \gamma \cos k) & -\overline{\alpha}_1(1 + \gamma \cos k) \end{bmatrix}, \quad (15)$$

and $r_l^\infty = \lim_{t \to \infty} r_l(t)$ is the value of the pair correlation function corresponding to the stationary solution of the equations of motion in the limit $t \to \infty$.

The solution to Eq. (12), which yields the magnetization, is straightforward, namely

$$\Psi_k(t) = e^{\mathbf{M}_k t} \Psi_k(0). \quad (16)$$

The relaxation process of the wave-vector dependent magnetization is determined by the eigenvalues of $\mathbf{M}_k$. These are given by

$$\lambda_k^{\pm} = -\overline{\alpha}_1 \pm \sqrt{\overline{\alpha}_2^2 + (\overline{\alpha}_1^2 - \overline{\alpha}_2^2)\gamma^2 \cos^2 k}. \quad (17)$$

The inverses of the (k-dependent) relaxation times $\tau_k^{\pm}$ of the $\pm k$th modes are precisely the $\lambda_k^{\pm}$. In the critical region, that is, when $T \to 0$ and $k \to 0$, $\lambda_k^- \to -2\overline{\alpha}_1$ while $\lambda_k^+ \to 0$. This means that the critical mode is the one corresponding to λ_k^+. As for the relaxation time, in this limit one gets

$$\mathrm{Re}(-\lambda_k^+) = -\frac{1}{\tau_k} \sim \frac{(\overline{\alpha}_1^2 - \overline{\alpha}_2^2)}{2\overline{\alpha}_1} \xi^{-2}[1 + (\xi k)^2], \quad (18)$$

where we have identified the correlation length ξ as $\xi = \exp(2J/k_B T)/2$. By comparing the former expression

with the one of the dynamic scaling hypothesis $\tau_k^{-1} \sim \xi^{-z} f(\xi k)$, one finds $z = 2$, so that, as stated above, in the case of one-dimensional Glauber dynamics the alternating isotopic chain [8] leads to the same value of z as the homogeneous chain.

Now we turn to the calculation of the other interesting response function, namely the frequency and wave-vector dependent susceptibility $S_k(\omega)$, which, by virtue of the fluctuation-dissipation theorem [21], is defined by

$$S_k(\omega) = \frac{\langle \sigma_k \sigma_{-k} \rangle_\infty}{k_B T} - \frac{i\omega \langle \sigma_k \sigma_{-k} \rangle_\omega}{k_B T}, \quad (19)$$

where $\langle \sigma_k \sigma_{-k} \rangle_\infty = 1/(1 - \gamma \cos k) \cosh(2J/k_B T)$ is the static correlation function and $\langle \sigma_k \sigma_{-k} \rangle_\omega$ is the Fourier transform of the dynamic one. It should be noted that $\chi \equiv k_B T S_0(\omega)/\langle \sigma_0 \sigma_0 \rangle_\infty$. After some rather lengthy but not too complicated algebraic manipulations starting with Eq. (14) one may arrive at the following result [8], namely

$$S_k(\omega) = \frac{1}{k_B T(1 - \gamma \cos k) \cosh \frac{2J}{k_B T}} \left[1 - \frac{i\omega[i\omega + \overline{\alpha}_1(1 + \gamma \cos k)]}{(i\omega + \overline{\alpha}_1)^2 - \frac{1}{2}\gamma^2 \alpha_1 \alpha_2(1 + \cos 2k) - \overline{\alpha}_2^2} \right], \quad (20)$$

from which, using also Eq. (17) with $k = 0$, χ can be expressed in the form

$$\chi = \frac{(1 - \gamma)(\alpha_1 + \alpha_2)}{4}$$
$$\times \left[\frac{1 - f(\alpha_1, \alpha_2, \gamma)}{i\omega - \lambda_0^+} - \frac{1 + f(\alpha_1, \alpha_2, \gamma)}{i\omega - \lambda_0^-} \right]. \quad (21)$$

Here, the (temperature dependent) function $f(\alpha_1, \alpha_2, \gamma)$ is given by

$$f(\alpha_1, \alpha_2, \gamma) = \frac{(\alpha_1 - \alpha_2)^2 - 4\alpha_1 \alpha_2 \gamma^2}{(\alpha_1 + \alpha_2)\sqrt{(\alpha_1 - \alpha_2)^2 + 4\alpha_1 \alpha_2 \gamma^2}}. \quad (22)$$

If we set $\alpha_1 = \alpha_2$ in Eq. (21), i.e., we take the uniform chain, then of course the resulting susceptibility has the simple Debye form. Although not shown, we have checked that this form does not lead to Nagel scaling. On the other hand, for the case $\gamma = 0$, we get

$$\chi_{\gamma=0} = \frac{1}{2}\left[\frac{\alpha_1}{i\omega + \alpha_1} + \frac{\alpha_2}{i\omega + \alpha_2} \right], \quad (23)$$

so that the general structure of the result for the susceptibility of the alternating isotopic chain is preserved irrespective of the value of γ (i.e., of the temperature), namely a linear combination of two Debye-like terms.

In Figs. 1–3 we present Nagel plots for the cases $\alpha_1 = 1$ and $\alpha_2 = 2$, $\alpha_1 = 1$ and $\alpha_2 = 10$, and $\alpha_1 = 1$ and $\alpha_2 = 1000$, respectively, and different values of $1/T^* \equiv 2J/k_B T$. In the insets we include the plots $\chi''(\omega)/\chi''(\omega_p)$ against ω/ω_p which are the natural variables of the Debye scaling. We note that as soon as the relaxation times become different, except for very low values of T^*, the agreement with the Nagel scaling improves significantly (cf. Fig. 1) until such scaling is virtually perfect as depicted in Figs. 2 and 3. On the other hand, the almost perfect Debye scaling in Fig. 1 is completely lost in Fig. 3. It should be noted that if the two time scales are very different, plateau regions eventually appear in the Nagel plot as clearly seen in Fig. 3. Whether the presence of more than two relaxation times, even if not as widely separated as in the case of Fig. 3, would also lead to the same type of results is something requiring future assessment. Also, the precise

location of the critical value of T^* above which the Nagel scaling holds as well as the nature of the crossover and of the "low T^*" regime are worth investigating.

It is important to point out that the experiments in which the Nagel plots have been more successful concern glass-forming systems in which topological constraints are assumed to be crucial. However, a clear-cut connection between such constraints and the different relaxation mechanisms has not been established. In this sense, it is rewarding that the alternating isotopic Ising chain, which is relatively simple with regards to relaxation phenomena but shows, nevertheless, universal behavior in terms of the dynamic critical exponent, provides perhaps the *first microscopic model* in which this scaling is shown to arise. We further want to mention that in order to include some of the features present in systems such as the above mentioned glass-forming liquids, we have also considered in the present context a generalization to an alternating isotopic chain of our quasi one-dimensional kinetic Ising-like model of linear polymeric chains [22], in which the Hamiltonian was chosen as to reduce to the one giving the
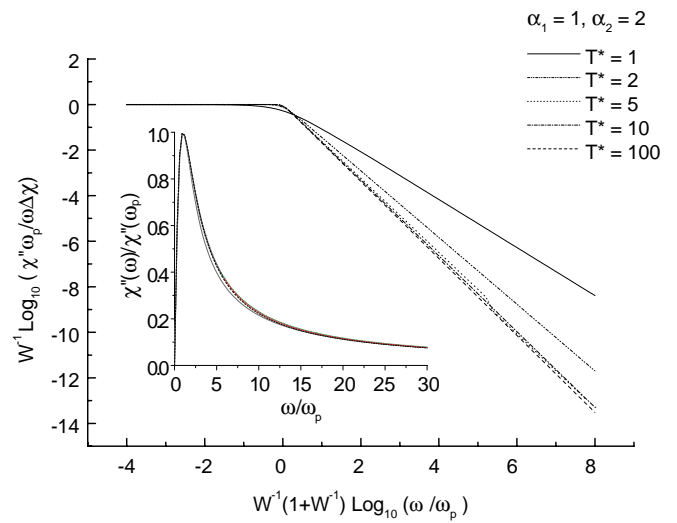

FIG. 1. Nagel plot for $\alpha_1 = 1$ and $\alpha_2 = 2$ and for $T^* = 1, 2, 5, 10,$ and 100. There is reasonable agreement with the scaling form for this choice except for low T^*. The inset contains the plot of $\chi''(\omega)/\chi''(\omega_p)$ vs ω/ω_p in order to test the Debye-like behavior.
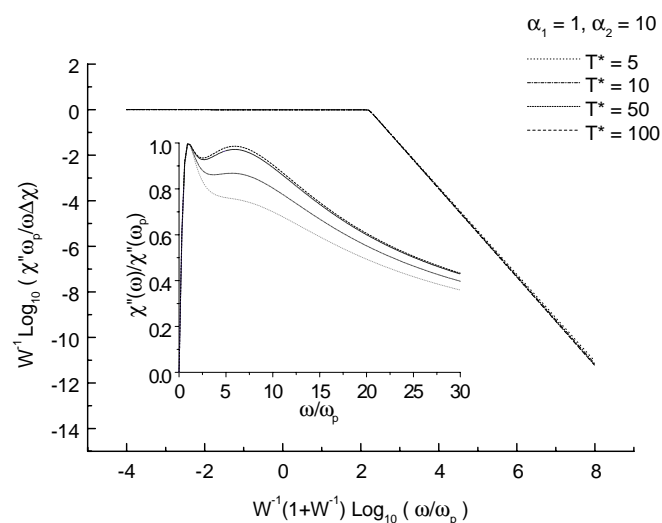
FIG. 2.   The same as Fig. 1 but with the choice $\alpha_1 = 1$, $\alpha_2 = 10$, and $T^* = 5, 10, 50$, and 100. The improvement in the agreement with the Nagel scaling is rather noticeable, while the opposite trend is observed with respect to the Debye scaling.

intramolecular energy of the Gibbs–di Marzio lattice model [23]. Interestingly enough in this model, in which the stochastic dynamics implied a rule of transition for the configurational changes which was tied to the creation or disappearance of flexes and so only some states were selected (in the magnetic language this means that the domain wall motion is through a biased random walk), similar conclusions concerning the Nagel scaling readily follow. These will be reported elsewhere. Finally, it would be interesting to test whether the same kind of scaling is present in other Ising models related to glassy systems, such as the spin facilitated kinetic Ising model originally introduced by Fredrickson and Andersen [24] and recently studied in connection with glassy dynamics [25].
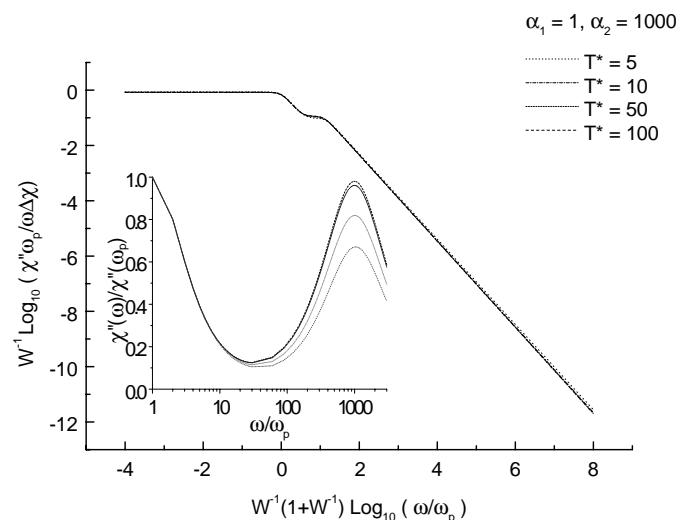

FIG. 3.   The same as Figs. 1 and 2 but for $\alpha_1 = 1$, $\alpha_2 = 1000$, and $T^* = 5, 10, 50$, and 100. A plateau region in the Nagel plot is clearly present in this case. Here, the behavior is definitely non-Debye.

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