Journal of Statistical Physics, Vol. 24, No. 1, 1981

On the Nonergodicity of the Transverse Magnetization in the Transverse Ising Model

R. Dagonnier[1] and Martine Dumont[1,2]

Received January 8, 1980

The nonergodic behavior exhibited by the transverse spin correlation function $\Gamma^{xx}_{q=0}(t)$ of the transverse Ising model obtained as the solution of approximate kinetic equations (derived on the basis of Résibois and De Leener's method), is shown to be an intrinsic property of the model and not the result of the approximations made in the derivation of the kinetic equations.

KEY WORDS: Transverse Ising model; nonlinear kinetic equations; dynamical transverse spin correlation function; nonergodicity.

1. INTRODUCTION

The transverse Ising model (TIM) is known to give a suitable description of systems in which a dominant role is played by a restricted number of low-lying energy eigenstates.[(1,2),3] Order–disorder ferroelectrics (e.g., KDP),[(3)] Van Vleck paramagnets,[(4,5)] and systems showing a cooperative Jahn–Teller phase transition[(6)] are the most studied systems within the TIM.

The TIM is described by the following spin-1/2 Hamiltonian:

$$H = -\Omega S^x_{q=0} - \frac{1}{N} \sum_q V_q S^z_q S^z_{-q} \tag{1}$$

where Ω represents the strength of the transverse field and V_q stands for the Fourier transform of the interaction between the z-spin-component operators. For instance, in the case of order–disorder ferroelectrics, widely

[1] Faculté des Sciences, Université de l'Etat, Mons, Belgium.

[2] Chargé de Recherches au Fonds National Belge de la Recherche Scientifique.

[3] See Table I of Ref. 2 for an extensive list of applications of the TIM.

0022-4715/81/0100-0059$03.00/0

studied within the TIM, the eigenstates of the *pseudospin* operator $S^z = \pm 1/2$ can be assigned to two polar configurations (dressed dipoles) with a dipole moment in the $+z$ and $-z$ directions. For these systems, Ω represents the tunneling frequency between the two polar configurations.

As usual for spin systems, the dynamics of this model is conveniently formulated in terms of the dynamical spin correlation functions (CF),

$$\Gamma_q^{\alpha\beta}(t) = \langle \Delta S_q^\alpha(t) \Delta S_{-q}^\beta \rangle = \langle S_q^\alpha(t) S_{-q}^\beta \rangle - \langle S_q^\alpha \rangle \langle S_{-q}^\beta \rangle, \qquad \alpha, \beta = x, y, z \tag{2}$$

These canonical ($\langle \cdots \rangle$) averages of the correlations between the fluctuating "coordinates"

$$\Delta S_q^\alpha = \frac{1}{\sqrt{N}} \sum_i e^{iqR_i} \Delta S_i^\alpha \tag{3}$$

provide an essential picture of the dielectric or magnetic susceptibilities and the dynamical form factors for systems described by the TIM.

Among the studies on the dynamics of the TIM let us mention those in which the spin correlation or relaxation functions are obtained (i) by means of a perturbation expansion,[2,7] (ii) as numerical solutions of kinetic equations derived by the authors,[8–11] and (iii) by means of Mori's continued fraction representation[12] applied to these functions.[5,13]

A common result of these studies which cover the whole para region ($\infty \geqslant T \geqslant T_c$) lies in the nonergodic behavior of the transverse CF $\Gamma_{q=0}^{xx}(t)$:

$$\lim_{t\to\infty} \Gamma_{q=0}^{xx}(t) = \text{const} \tag{4}$$

For instance, the numerical resolution of the kinetic equations for the CF obtained for the TIM by using Résibois and De Leener's method[14] shows that, at $T = \infty$ and for $0.3 < \Omega/V_0 \lesssim 0.9$ (i.e., in the resonant regime[13] for the other $\Gamma_q^{\alpha\beta}$), $\Gamma_{q=0}^{xx}$ approaches a positive plateau value p,

$$p = \tfrac{1}{4}\left[1 - \exp(-\lambda\Omega/V_0)\right] \tag{5}$$

in the long-time range.[10] The coefficient λ depends on the nature of the interaction (nearest neighbor, dipolar, or mixed) and on the type of spin configuration considered in the calculation. Equation (5) indicates that p vanishes in the Ising limit, i.e., when $\Omega \to 0$.

Unfortunately, the transverse CF of the TIM is not related to any measurable quantity. Although a test of Eq. (4) could be found via the relation[15,16]

$$\lim_{t\to\infty} \Gamma_{q=0}^{xx}(t) = (1/\beta)\left[\chi_T^{xx} - \chi_{q=0}^{xx}(0)\right] \tag{6}$$

where χ_T^{xx} represents the isothermal (thermodynamic) susceptibility and $\chi_{q=0}^{xx}(0)$ stands for the static, isolated (Kubo) susceptibility, the situation is complicated by the fact that the formulation of the transverse electric susceptibility in order–disorder ferroelectrics is entirely different from that of transverse magnetic susceptibility in ferromagnetic crystals.

Indeed, if in both cases the interaction term of the Hamiltonian is $\sum_{i,j} V_{ij} S_i^z S_j^z$, then the transverse susceptibility in the magnetic case comes from the term proportional to $S_{q=0}^x$ (or $S_{q=0}^y$), whereas in the ferroelectric case it comes from the term proportional to $S_{q=0}^z$ [see Eq. (8) below].[17] This particularity can be illustrated in the case of KDP-type ferroelectrics as follows. Let us first recall that, in these ferroelectrics of crystal axes (a, b, c) denoted below by $(\mathcal{X}, \mathcal{Y}, \mathcal{Z})$,[4] the H bonds nearly lie in the plane *ab* perpendicular to the spontaneous polarization *c* axis. The polarization is essentially due to the displacements of the heavy ions (e.g., K^+, P^{5+} ions in KH_2PO_4), which are strongly coupled to the hydrogen ion displacements from one side of their bonds to the other, the phase transition being triggered by the hydrogen ordering. For instance, to obtain the longitudinal susceptibility we assign the quantization z axis of the pseudospin space to the crystal $\mathcal{Z}$ axis. Namely, for an external field in the $\mathcal{Z}$ direction we should add to the TIM Hamiltonian (1) the term

$$H' = -p_{\mathcal{Z}} E_{\mathcal{Z}} S_{q=0}^z \tag{7}$$

where $p_{\mathcal{Z}}$ is the effective value of the $\mathcal{Z}$-axis dipole moment (i.e., essentially the contribution of the heavy-ion displacements). On the other hand, to obtain the transverse susceptibility one assumes that the pseudospin space axis z lies in the crystal plane $\mathcal{X}\mathcal{Y}$ (*ab* for KDP) perpendicular to the $\mathcal{Z}$ axis. Namely, for a field in the $\mathcal{X}$ direction we should add to Eq. (1) the term

$$H' = -p_{\mathcal{X}} E_{\mathcal{X}} S_{q=0}^z \tag{8}$$

where $p_{\mathcal{X}}$ is the effective value of the $\mathcal{X}$-axis dipole moment (i.e., essentially the contribution of the hydrogen-ion displacements).

In spite of this lack of experimental information about $\Gamma_{q=0}^{xx}(t)$, the authors questioned the nonergodicity of S^x. This still-open question may be expressed as follows. Does the nonergodicity of S^x appear *only* as a result of the approximations made in the derivation of the kinetic equations for the dynamical CF, or is it an intrinsic property of the three-dimensional TIM Hamiltonian (1)? In the following sections we shall examine these two aspects of the problem.

2. THE KINETIC EQUATIONS APPROACH

Let us consider the kinetic equations for the CF obtained by application of the method of Résibois and De Leener[14] to the TIM.[8–10,18] These

[4] The crystal axes $(\mathcal{X}, \mathcal{Y}, \mathcal{Z})$ have to be *a priori* distinguished from the pseudospin axes (x, y, z).

kinetic equations give the time evolution, valid in the Weiss limit,[19] of the *normalized* CF $\tilde{\Gamma}_q^{\alpha\beta}(t)$. These normalized CF $\tilde{\Gamma}_q^{\alpha\beta}(t)$ are related to the usual CF $\Gamma_q^{\alpha\beta}(t)$ [see Eq. (2)] and their equilibrium forms $\Gamma_q^{\alpha\beta}(0)$ as follows:

$$\Gamma_q^{\alpha\beta}(t) = \sum_{\gamma} \tilde{\Gamma}_q^{\alpha\gamma}(t)\Gamma_q^{\gamma\beta}(0), \qquad \alpha, \beta, \gamma = x, y, z \tag{9}$$

provided the initial nonequilibrium correlations are neglected.[14] For the transverse CF $\Gamma_q^{xx}(t)$ the kinetic equation reads, at any temperature,

$$\frac{\partial}{\partial t}\Gamma_q^{xx}(t) = 2V_0\langle S^z\rangle\Gamma_q^{yx}(t) + \sum_{\gamma}\int_0^t dt'\, G_q^{x\gamma}(t')\Gamma_q^{\gamma x}(t-t') \tag{10}$$

The kernels $G_q^{x\gamma}(t)$ are nonlinear functionals of different normalized CF $\tilde{\Gamma}_q^{\mu\nu}(t)$:

$$G_q^{x\gamma}(t) = G_q^{x\gamma}\big(\{\tilde{\Gamma}_q^{\mu\nu}(t)\}\big) = \sum_{n=2}^{\infty}\lambda^n G_q^{x\gamma(n)}(t) \tag{11}$$

and appear as power series of a renormalized interaction[8,9] indicated by the counting parameter λ. It must be clear that the resolution of Eq. (10) requires the simultaneous resolution of similar kinetic equations for all other CF $\tilde{\Gamma}_q^{\mu\nu}(t)$ coupled to $\Gamma_q^{xx}(t)$ (see Refs. 9, 10 for such a system of coupled kinetic equations for the TIM in the region $T \geqslant T_c$).

Assuming that the series in Eq. (11) are convergent (as has been checked[18] numerically up to order λ^4 for $T \gtrsim T_c$), we can approximate Eq. (11) *in the Markovian limit* by

$$\lim_{t\to\infty}\frac{\partial}{\partial t}\Gamma_q^{xx}(t) = \sum_{\gamma}\left\{\int_0^{\infty} G_q^{x\gamma(2)}(t')\,dt'\right\}\Gamma_q^{\gamma x}(t) \tag{12}$$

taking account of the numerical result

$$\lim_{t\to\infty}\Gamma_q^{y\alpha}(t) = 0, \qquad \alpha = x, y, z \tag{13}$$

Now, from the explicit forms of the kernels $G_q^{x\gamma(2)}(t)$ reported in the Appendix, it is simple but tedious to deduce that *at any temperature*

$$\int_0^{\infty} G_{q=0}^{x\gamma(2)}(t)\,dt = 0, \qquad \gamma = x, y, z \tag{14}$$

if one uses the numerical results [Eq. (13)] and

$$\lim_{t\to\infty}\tilde{\Gamma}_q^{z\alpha}(t) = 0, \qquad \alpha = x, y \tag{15}$$

and the *exact* kinetic equations

$$\frac{\partial}{\partial t}\tilde{\Gamma}_q^{z\alpha}(t) = -\Omega\tilde{\Gamma}_q^{y\alpha}(t), \qquad \alpha = x, y, z \tag{16}$$

Consequently, Eqs. (12) and (14) show that Eq. (4) is correct up to $O(\lambda^2)$. This result has been checked numerically for the temperature range $[\infty, T_c - \Delta T]$, where $\Delta T = 10$ deg.[18]

It was first suggested that the use of higher order terms in the kinetic equations should remove this nonergodic behavior of S^x.[9] Unfortunately this suggestion cannot be checked, as the numerical resolution of Eq. (10) up to a higher order than λ^2 is too prohibitive to be tentatively performed. We thus turned to a motion constant approach.

3. THE MOTION INVARIANT APPROACH

The nonergodicity of S^x has been already demonstrated for the *one-dimensional*, spin-1/2 TIM *at any temperature*. Mazur[21] has shown the nonergodicity of the z component of the magnetization in the one-dimensional, spin-1/2, x–y model of Hamiltonian

$$H = \sum_{i=1}^{N} \left[(1+\gamma)S_i^x S_{i+1}^x + (1-\gamma)S_i^y S_{i+1}^y - BS_i^z\right]$$

Since for $\gamma = 1$ this model reduces to the TIM, Mazur's result implies the nonergodicity of S^x in the TIM, Eq. (1), for a linear chain of spin 1/2 whatever T is. This nonergodicity of $\Gamma_{q=0}^{xx}(t)$ has been also obtained as a by-product of an exact calculation of the dynamical properties of the one-dimensional TIM in the limits $T = \infty$ and $T = 0$.[20] Both results[20,21] are based on the diagonalization of one-dimensional Hamiltonians and cannot be extended to spin configurations of higher dimension.

To see if the nonergodic behavior of $\Gamma_{q=0}^{xx}(t)$, Eq. (4), is an intrinsic property of the *three-dimensional* TIM *at any temperature*, we use a theorem derived by Suzuki.[22] This theorem expresses the time average of a CF (i.e., its infinite-time value) in terms of canonical averages involving *all* the constants of motion of the system considered.

Let us recall the usual definition of a general CF:

$$\Gamma^{AB}(t) = \langle \Delta A(t) \Delta B \rangle \tag{17}$$

and say that a dynamical variable A is ergodic if

$$\lim_{t\to\infty} \Gamma^{AA}(t) = 0 \tag{18}$$

Then Suzuki's theorem may be expressed as follows: if $\{I_\mu\}$ are all the motion invariants of the Hamiltonian H considered, i.e., $[I_\mu, H] = 0$, then

$$\lim_{t\to\infty} \Gamma^{AB}(t) = \sum_{\mu=1}^{\infty} \frac{\langle (\Delta A) I_\mu \rangle \langle (\Delta B) I_\mu \rangle}{\langle I_\mu^2 \rangle} \tag{19}$$

where, without loss of generality, all I_μ are assumed Hermitian ($I_\mu^+ = I_\mu$) and orthogonal to one another in the sense that

$$\langle I_\mu I_\nu \rangle = \delta_{\mu\nu} \langle I_\mu^2 \rangle \tag{20}$$

Now applying this theorem to the transverse CF $\Gamma^{xx}_{q=0}(t)$, one readily gets

$$\lim_{t\to\infty}\Gamma^{xx}_{q=0}(t)=\sum_{\mu=1}^{\infty}\frac{\langle(\Delta S^x_{q=0})I_\mu\rangle^2}{\langle I_\mu^2\rangle}\geqslant\sum_{\mu=1}^{m}\frac{\langle(\Delta S^x_{q=0})I_\mu\rangle^2}{\langle I_\mu^2\rangle}\tag{21}$$

if one confines oneself to any subset $\{I_\mu\}_{\mu=1,m}$ of constants of motion. In Eq. (21) we have used the definition

$$\Gamma^{\alpha\beta}_q(t)=\langle\Delta S^\alpha_q(t)\,\Delta S^\beta_{-q}\rangle=\frac{1}{N}\sum_{i,j}\langle\Delta S^\alpha_i(t)\,\Delta S^\beta_j\rangle\exp(iqR_{ij})\tag{22}$$

Equation (21) clearly shows that the nonergodicity of S^x in the TIM is proved provided there exists *at least* one motion invariant I_1 such that

$$\lim_{t\to\infty}\Gamma^{xx}_{q=0}(t)\geqslant\langle\Delta S^x_{q=0}I_1\rangle^2/\langle I_1^2\rangle\neq 0\tag{23}$$

It should be specified that such an inequality [here a "corollary" of Eq. (19)] was derived and used by Mazur[21] in his treatment of the one-dimensional x–y model mentioned above.

The simplest subset of motion invariants that can be defined to apply Eq. (21) or Eq. (23) to our purpose is

$$I_\mu=(\Delta H)^\mu\tag{24}$$

Then, since

$$\langle(\Delta S^\alpha_i)(\Delta H)\rangle=-\frac{\partial}{\partial\beta}\langle S^\alpha\rangle,\qquad \alpha=x,y,z\tag{25}$$

relation (23) may be cast into ($I_1=\Delta H$) the form

$$\lim_{t\to\infty}\Gamma^{xx}_{q=0}(t)\geqslant\frac{N}{\langle(\Delta H)^2\rangle}\left[\frac{\partial}{\partial\beta}\langle S^x\rangle\right]^2\overset{?}{\neq}0\tag{26}$$

Consequently, if $\langle S^x\rangle$ is temperature dependent in a given temperature region (and for a given spin lattice dimensionality), the transverse spin component S^x for the TIM is not ergodic in this temperature range.

Generally $\langle S^x\rangle$ cannot be evaluated exactly. For instance, in the MFA (0) we have[3]

$$\langle S^x\rangle_0=\tfrac{1}{2}\tanh(\beta\Omega/2),\qquad T\geqslant T_c\tag{27a}$$

$$\langle S^x\rangle_0=\Omega/2V_0,\qquad T<T_c\tag{27b}$$

According to Eq. (27a), Eq. (26) leads to

$$\inf\lim\Gamma^{xx}_{q=0}(t\to\infty)\sim\Omega^2\operatorname{sech}^2(\beta\Omega/2),\qquad T\geqslant T_c\tag{28}$$

For the region $T<T_c$ we should take into account at least the first correction $\langle S^x\rangle_1$ to the MFA value $\langle S^x\rangle_0$, Eq. (26b). This value $\langle S^x\rangle_1$ of order Z^{-1} (Z being the number of interacting nearest neighbors) was

calculated by Stinchcombe,[23]

$$\langle S^x\rangle_1 = \frac{\Omega}{F}\frac{1}{N}\sum_q \left\{ \frac{V_q R_1 \coth(\beta\omega_q/2)}{\omega_q} + \frac{2V_q}{\beta}\left[\frac{R_1 - \beta F(1/4 - R_1^2)}{\omega_q^2 - 8\beta V_q V_0^2 \langle S^z\rangle^2 (1/4 - R_1^2)} - \frac{R_1}{\omega_q^2}\right]\right\} \tag{29}$$

where

$$F^2 = \Omega^2 + (2V_0\langle S^z\rangle_0)^2 \tag{30a}$$

$$R_1 = F/2V_0 \tag{30b}$$

and the RPA pseudospin wave frequency ω_q is defined by

$$\omega_q^2 = F^2 - 2\Omega V_0 \langle S^x\rangle_0 \tag{30c}$$

Equation (29) clearly shows that $\langle S^x\rangle \approx \langle S^x\rangle_0 + \langle S^x\rangle_1$ is T dependent in the region $T < T_c$. Therefore, up to order Z^{-1}, we have

$$\lim_{t\to\infty} \Gamma_q^{xx}(t) \neq 0, \qquad T < T_c \tag{31}$$

The rigorous proof of the nonergodicity of S^x requires a general demonstration either of the property [see Eq. (25)]

$$\frac{\partial}{\partial\beta}\langle S^x\rangle \neq 0 \qquad \text{all} \quad T \tag{32}$$

or equivalently of the diagonality of S^x with respect to the energy [see Eq. (25)].

A convincing argument about the validity of Eq. (32) may be found in the examination of the values of the lower x_L and upper x_U bounds for $\langle S^x\rangle$ determined by Lam and Bunde.[24] These authors show that

$$x_U = \inf(1/2, T/4\Omega) \tag{33a}$$

$$x_L = \Omega^{-1}(\lambda_L T - V_0/2) \tag{33b}$$

where λ_L is the root of the equation

$$f(\lambda_L) = (2T/\Omega^2)(2T\lambda_L - V_0) \tag{34}$$

with

$$f(x\tanh x) = (\tanh x)/x \tag{35}$$

In Eqs. (33a) and (33b) the unique T-independent bound $x_U = 1/2$ for $T < T_c$ (see Fig. 1 in Ref. 24) is the MFA result [see Eq. (27b) with the necessary phase-transition condition $\Omega/V_0 \leqslant 1$ for the TIM[3]]. As this MFA result overestimates $\langle S^x\rangle$ by a T-dependent quantity [see Eq. (29) above and Fig. 3 in Ref. 24] we may conclude that, all other bounds of $\langle S^x\rangle$ being actually T dependent for $0 \leqslant T \leqslant \infty$, Eq. (32) is valid.

In conclusion, we have shown that the nonergodic behavior of $\Gamma^{xx}_{q=0}(t)$ is an *intrinsic* property of the three-dimensional TIM at any temperature T. It should be added that the determination of all the motion invariants I_μ of the TIM nonorthogonal to S^x should lead to the plateau value reached by $\Gamma^{xx}_{q=0}(t)$ in the long-time limit. Further work in this direction is underway.

APPENDIX

We report here the explicit forms of the kernels $G_q^{x\gamma(2)}(t)$ ($\gamma = x, y, z$) in terms of the *normalized* CF $\tilde{\Gamma}_q^{\alpha\beta}(t)$ defined in Eq. (9) and of the following equilibrium CF evaluated up to order Z^{-1} (Z being the number of interacting nearest neighbors):

$$\Gamma_q^{zz}(0) = \frac{2\Omega R}{4\omega_q \tanh(\beta\omega_q/2)} + \frac{\omega_1^2 F^2}{\omega_q^2(4\omega_q^2 - 2\beta V_q \omega_1^2)} \tag{A1}$$

$$\Gamma_q^{z+}(0) = \frac{R}{2} - \frac{V_0 PR}{4\omega_q \tanh(\beta\omega_q/2)} + \frac{1}{\beta}\left[\frac{V_0 PR}{2\omega_q^2} - \frac{\beta\Omega\omega_1^2 - 2V_0^2P^2R}{V_0P(4\omega_q^2 - 2\beta V_q\omega_1^2)}\right] \tag{A2}$$

where

$$R = 2\langle S^x\rangle \tag{A3}$$

$$P = 2\langle S^z\rangle \tag{A4}$$

In Eqs. (A1) and (A2), ω_q (the RPA pseudospin wave frequency) and F are defined in Eqs. (30a) and (30c), and

$$\omega_1^2 = (V_0^2 - F^2)P^2 \tag{A5}$$

The expressions of $G_q^{x\gamma(2)}(t)$ ($\gamma = x, y, z$) are, respectively,

$$\begin{aligned}
G_q^{xx(2)}(t) = &- \frac{4}{N}\sum_{q'} V_{q'}^2 \Gamma_{q'}^{zz}(0)\tilde{\Gamma}_{q-q'}^{yy}(t)\tilde{\Gamma}_{q'}^{zz}(t) \\
&- \frac{4}{N}\sum_{q'} V_{q'}V_{q-q'}\Gamma_{q'}^{zz}(0)\tilde{\Gamma}_{q-q'}^{zy}(t)\tilde{\Gamma}_{q'}^{yz}(t) \\
&- \frac{4}{N}\sum_{q'} V_{q'}^2\left\{\left[\Gamma_{q'}^{z+}(0) - \frac{R}{4}\right]\tilde{\Gamma}_{q-q'}^{yy}(t)\tilde{\Gamma}_{q'}^{zx}(t)\right. \\
&\left.- \frac{PR}{4}\tilde{\Gamma}_{q-q'}^{yx}(t)\tilde{\Gamma}_{q'}^{zy}(t)\right\} \\
&- \frac{4}{N}\sum_{q'} V_{q'}V_{q-q'}\left\{\left[\Gamma_{q'}^{z+}(0) - \frac{R}{4}\right]\tilde{\Gamma}_{q-q'}^{zy}(t)\tilde{\Gamma}_{q'}^{yx}(t)\right. \\
&\left.- \frac{PR}{4}\tilde{\Gamma}_{q-q'}^{zx}(t)\tilde{\Gamma}_{q'}^{yy}(t)\right\}
\end{aligned} \tag{A6}$$

$$
\begin{aligned}
G_q^{xy(2)}(t) = {} & \frac{4}{N}\sum_{q'} V_{q'}^2 \Gamma_{q'}^{zz}(0)\tilde{\Gamma}_{q-q'}^{yx}(t)\tilde{\Gamma}_{q'}^{zz}(t) \\
& + \frac{4}{N}\sum_{q'} V_{q'}V_{q-q'}\Gamma_{q'}^{zz}(0)\tilde{\Gamma}_{q-q'}^{zx}(t)\tilde{\Gamma}_{q'}^{yz}(t) \\
& + \frac{4}{N}\sum_{q'} V_{q'}^2\left\{\left[\Gamma_{q'}^{z+}(0) - \frac{R}{4}\right]\tilde{\Gamma}_{q-q'}^{yx}(t)\tilde{\Gamma}_{q'}^{zx}(t)\right. \\
& \left. + \frac{PR}{4}\tilde{\Gamma}_{q-q'}^{yy}(t)\tilde{\Gamma}_{q'}^{zy}(t)\right\} \\
& + \frac{4}{N}\sum_{q'} V_{q'}V_{q-q'}\left\{\left[\Gamma_{q'}^{z+}(0) - \frac{R}{4}\right]\tilde{\Gamma}_{q-q'}^{zx}(t)\tilde{\Gamma}_{q'}^{yx}(t)\right. \\
& \left. + \frac{PR}{4}\tilde{\Gamma}_{q-q'}^{zy}(t)\tilde{\Gamma}_{q'}^{yy}(t)\right\} \qquad \text{(A7)}
\end{aligned}
$$

$$
\begin{aligned}
G_q^{xz(2)}(t) = {} & -\frac{4}{N}\sum_{q'} V_q V_{q'}\left\{\left[\Gamma_{q'}^{z+}(0) - \frac{R}{4}(1-P)\right]\tilde{\Gamma}_{q-q'}^{yy}(t)\tilde{\Gamma}_{q'}^{zz}(t)\right. \\
& + \left[\Gamma_{q'}^{xx}(0) - \Gamma_{q-q'}^{yy}(0) + \frac{R^2}{4}\right]\tilde{\Gamma}_{q-q'}^{yy}(t)\tilde{\Gamma}_{q'}^{zx}(t) \\
& \left. - \left[\Gamma_{q'}^{yy}(0) - \Gamma_{q-q'}^{xx}(0) - \frac{R^2}{4}\right]\tilde{\Gamma}_{q-q'}^{yx}(t)\tilde{\Gamma}_{q'}^{zy}(t)\right\} \\
& - \frac{4}{N}\sum_{q'} V_q V_{q'}\left[\Gamma_{q-q'}^{z+}(0) - \frac{R}{4}(1-P)\right]\tilde{\Gamma}_{q-q'}^{yz}(t)\tilde{\Gamma}_{q'}^{zy}(t) \\
& + \frac{4}{N}\sum_{q'} V_{q'}^2\frac{R}{2}\left[P\tilde{\Gamma}_{q-q'}^{yz}(t) + \frac{R}{2}\tilde{\Gamma}_{q-q'}^{yz}(t)\right]\tilde{\Gamma}_{q'}^{zy}(t) \\
& + \frac{4}{N}\sum_{q'} V_{q'}V_{q-q'}\frac{R}{2}\left[P\tilde{\Gamma}_{q-q'}^{zz}(t) + \frac{R}{2}\tilde{\Gamma}_{q-q'}^{zx}(t)\right]\tilde{\Gamma}_{q'}^{yy}(t) \qquad \text{(A8)}
\end{aligned}
$$

REFERENCES

1. R. J. Elliot and A. P. Young, *Ferroelectrics* **7**:23 (1974).
2. R. B. Stinchcombe, *J. Phys. C* **6**:2459 (1973).
3. R. Blinc and B. Zeks, *Adv. Phys.* **21**:693 (1972); *Soft Modes in Ferroelectrics and Antiferroelectrics* (North-Holland, Amsterdam, 1975); P. Schuster, G. Zundel, and G. Sandorfy, *The Hydrogen Bond*, Vol. 3 (North-Holland, Amsterdam, 1976).
4. P. Fulde and I. Peschel, *Adv. Phys.* **21**:1 (1972).
5. T. H. Cheung, *Phys. Stat. Sol. B* **58**:567 (1973); *Z. Phys.* **267**:251 (1974).
6. E. Pytte, *Phys. Rev. B* **8**:3954 (1973).
7. S. Takada, I. Ohnari, H. Kurosawa, and Y. Ohmura, *Prog. Theor. Phys.* **55**:986 (1976).
8. D. P. Chock, P. Résibois, G. Dewel, and R. Dagonnier, *Physica (Utr.)* **53**:364 (1971).
9. D. P. Chock and R. Dagonnier, *Physica (Utr.)* **53**:393 (1971).
10. M. Dumont and R. Dagonnier, *Phys. Rev. B* **16**:363 (1977).
11. M. A. Moore and H. C. W. L. Williams, *J. Phys. C* **5**:3168, 3185, 3222 (1972).
12. H. Mori, *Prog. Theor. Phys.* **34**:399 (1965).

13. T. N. Tommet and D. L. Huber, *Phys. Rev. B* **11**:1971 (1975).
14. P. Résibois and M. De Leener, *Phys. Rev.* **152**:305, 318 (1966); **178**:806, 819 (1969).
15. R. Kubo, *J. Phys. Soc. Japan* **12**:570 (1957).
16. M. Suzuki, *Prog. Theor. Phys.* **43**:882 (1970).
17. S. Havlin and E. Litov, *Phys. Rev. B* **9**:1024 (1974).
18. M. Dumont, Thesis, State University of Mons, Belgium (1979).
19. R. Brout, *Phase Transitions* (Benjamin, New York, 1965).
20. T. N. Tommet and D. L. Huber, *Phys. Rev. B* **11**:450 (1975).
21. P. Mazur, *Physica (Utr.)* **43**:533 (1969).
22. M. Suzuki, *Physica (Utr.)* **51**:277 (1971).
23. R. B. Stinchcombe, *J. Phys. C* **6**:2507 (1973).
24. L. Lam and A. Bunde, *J. Phys. C* **10**:693 (1977).