Glassy Behavior of Light

L. Angelani,[1] C. Conti,[2,3] G. Ruocco,[3,4] and F. Zamponi[3,4]

[1]*Research Center SMC INFM-CNR, c/o Università di Roma "La Sapienza," I-00185, Roma, Italy*
[2]*Centro Studi e Ricerche "Enrico Fermi," Via Panisperna 89/A, I-00184, Roma, Italy*
[3]*Research Center Soft INFM-CNR, c/o Università di Roma "La Sapienza," I-00185, Roma, Italy*
[4]*Dipartimento di Fisica, Università di Roma "La Sapienza," I-00185, Roma, Italy*

(Received 22 September 2005; published 16 February 2006)

We study the nonlinear dynamics of a multimode random laser using the methods of statistical physics of disordered systems. A replica-symmetry breaking phase transition is predicted as a function of the pump intensity. We thus show that light propagating in a random nonlinear medium displays glassy behavior; i.e., the photon gas has a multitude of metastable states and a nonvanishing complexity, corresponding to mode-locking processes in random lasers. The present work reveals the existence of new physical phenomena, and demonstrates how nonlinear optics and random lasers can be a benchmark for the modern theory of complex systems and glasses.

DOI: 10.1103/PhysRevLett.96.065702 PACS numbers: 64.70.Pf, 42.55.Zz, 75.10.Nr

The first marriage between statistical mechanics and lasers dates back to their early development [1,2]. Since the 1970s, many authors have outlined that the threshold for lasing can be interpreted as a thermodynamic phase transition and these ideas spread out in the field of photonics, later embracing also nonlinear optics [for a review see, e.g., [3]]. In recent articles [4–7], the statistical properties of laser light in *homogeneous cavities* have been studied taking into account nonlinear phenomena, like gain saturation and intensity dependent refractive index. This nonlinearity gives rise to an interaction among the oscillation modes, which in turn produces new interesting effects. Specifically, by mapping the dynamics in an *ordered* Hamiltonian problem, the authors of Ref. [7] predicted a critical behavior (a phase transition) of the laser mode-locking process.

In the latter example, the statistical mechanics of *ordered* systems is applied to study light propagation in amplifying homogeneous nonlinear materials. In recent years, the locally inhomogeneous character of matter, and, in particular, the disordered nature of these inhomogeneities, are becoming more and more important. Specifically, relevant attention has been dedicated to light amplification in random media and random lasers (RL) [8–13]. This is a fascinating topic that bridges various fields like light localization and diffusion, thermodynamics, nonlinear physics, and quantum optics, and it has relevant fundamental and applicative perspectives, as in biophysics [14]. Methods of statistical mechanics have not yet been applied to the propagation of light in nonlinear *disordered* active media, but the rich behavior observed in glasses (aging, memory effects, etc.) can be foreseen [15,16].

In this Letter, we analytically study the statistical properties of the modes of a random optical cavity. Various physical settings are embraced by this problem: for example, a microstructured cavity filled by an active soft material, like doped liquid crystals, cavityless RL, or even a standard laser system with a disordered amplifying medium. In all these cases, the disorder varies on time scales much longer than the optical fields [it is "quenched" [15]] and, for any realization of the system, the supported electromagnetic modes interact because of the nonlinearity of the resonant medium.

We first show that light propagation is described by a Hamiltonian with the pumping rate acting as inverse temperature. The mode amplitudes are taken as slowly varying and their phases play the role of "spins" in a Hamiltonian, which turns out to be a generalization of the *XY* model [17] and of the so-called k-trigonometric model [18]. The thermodynamics of the model is studied using the replica trick [15], and a one step replica-symmetry breaking transition is found [18]. Our results show that when the average energy into each mode increases (i.e., the "temperature" is decreased), the system undergoes a glass transition, meaning that its dynamics slows down and an exponentially large number of states appears, with a nonvanishing "complexity" [19]. They correspond to "mode-locked" states of a random laser. This treatment somehow generalizes to disordered systems early works on multimode lasers [see, e.g., [20–22]], and can be also applied to other problems involving multimode interactions in a nonlinear medium. By providing an analytically treatable statistical model, we not only unveil the complex behavior (in the meaning of modern glassy physics) of light in active disordered media, but we also demonstrate the possibility of using them for testing the replica-symmetry breaking transitions. When studying the glass transition of atomic or molecular systems, due to the predicted kinetic arrest, the interesting time scale becomes extremely long with respect to that experimentally accessible. Because of the intrinsically fast *photon* dynamics, we expect that the system can be equilibrated much closer to the transition, and this should provide new and interesting experimental data on kinetically arrested "photon glasses."

We consider a dielectric resonator described by a refractive index profile $n(\mathbf{r})$. The time dependence of this quantity is of no interest here, taking place on a time scale much slower than that of the photon propagation. Our approach follows the standard coupled mode theory in the time domain [23]. The Maxwell's equations in the presence of a nonlinear polarization $\mathbf{P}_{NL}$ are:

$$\begin{aligned} \nabla \times \mathbf{H}(\mathbf{r}, t) &= \varepsilon_0 n^2(r)\partial_t \mathbf{E}(\mathbf{r}, t) + \partial_t \mathbf{P}_{NL}(\mathbf{r}, t, \mathbf{E}), \\ \nabla \times \mathbf{E}(\mathbf{r}, t) &= -\mu_0 \partial_t \mathbf{H}(\mathbf{r}, t). \end{aligned} \tag{1}$$

The fields $\mathbf{E}(\mathbf{r}, t)$ and $\mathbf{H}(\mathbf{r}, t)$ are conveniently decomposed in normal modes corresponding to the solutions of the linear problem, $\mathbf{P}_{NL} = 0$ (frequencies ω_m, eigenvectors $\mathbf{E}_m(\mathbf{r})$, $\mathbf{H}_m(\mathbf{r})$, and "amplitudes" a_m). For later convenience, they are cast in the form

$$\mathbf{E}(\mathbf{r}, t) = \Re\left[\sum_m \sqrt{\omega_m}\, a_m \mathbf{E}_m(\mathbf{r}) \exp(-i\omega_m t)\right], \tag{2}$$

(and similarly for $\mathbf{H}$) and the total energy stored in the cavity is $\mathcal{E} = \sum_m \mathcal{E}_m = \sum_m \omega_m |a_m|^2$.

In the presence of nonlinearity, the amplitudes a_m are time dependent and their evolution is described by coupled equations that can be derived using standard perturbation techniques [23] and take the general form

$$\frac{da_m(t)}{dt} = i\frac{\sqrt{\omega_m}}{4}\int_V \mathbf{E}_m^*(\mathbf{r}) \cdot \mathbf{P}_m(\mathbf{r}) dV. \tag{3}$$

The integral in Eq. (3) is extended to the cavity volume where the nonlinear polarization is different from zero. The quantity $\mathbf{P}_m(\mathbf{r})$ is the amplitude of the component of the nonlinear polarization oscillating at ω_m, $\mathbf{P}_{NL} = \Re[\sum_m \mathbf{P}_m \exp(-i\omega_m t)]$ and its explicit expression as function of the fields depends on the considered nonlinearity. For isotropic media the leading cubic terms are written as [24]

$$P_m^\alpha = \sum_{\omega_m = \omega_p + \omega_q - \omega_r} K_{\alpha\beta\gamma\delta} E_p^\beta E_q^\gamma E_r^\delta a_p a_q a_r^*.$$

Here $K_{\alpha\beta\gamma\delta} = \sqrt{\omega_p \omega_q \omega_r}\chi_{\alpha\beta\gamma\delta}(\omega_m; \omega_p, \omega_q, -\omega_r, \mathbf{r})$ and χ is the third order response susceptibility tensor [explicit expressions are known, for example, in the two levels approximation [20]]; the sum over Cartesian indices is implicit. The coupled mode theory equations in a nonlinear cavity read hence as $\dot{a}_m(t) = -\frac{1}{2}\sum_{pqr} g_{pqrm} a_p a_q a_r^*$, where the sum is extended over all the modes and

$$g_{pqrm} = \frac{\sqrt{\omega_m}}{2i}\int_V K_{\alpha\beta\gamma\delta} E_m^\alpha E_p^\beta E_q^\gamma E_r^\delta dV. \tag{4}$$

Under standard approximations [22,24], the tensor g can be taken as real-valued. Introducing the (real-valued) function $H = \frac{1}{4}\sum_{spqr} g_{spqr} \times a_s a_p a_q^* a_r^*$, and taking into account radiation losses and material absorption processes, represented by the coefficients α_m and light amplification through γ_m [23], we have

$$\frac{da_m}{dt} = -\frac{\partial \mathcal{H}}{\partial a_m^*} + \eta_m(t), \tag{5}$$

where $\mathcal{H} = \sum_m(\alpha_m - \gamma_m)|a_m|^2 + H$, and having introduced as usual the noise term [2,25], with $\langle \eta_p(t)\eta_q^*(t')\rangle = 2k_B T_{\text{bath}}\delta_{pq}\delta(t - t')$, weighted by an effective temperature T_{bath} [see Eq. (10)], with k_B the Boltzmann constant. The previous equation is a standard Langevin model for a system of N particles moving in $2N$ dimensions (represented by $\{\Re a_m, \Im a_m\}_{m=1\ldots N}$) [2,6] and its invariant measure is given by $\exp(-\mathcal{H}/k_B T_{\text{bath}})$.

We consider a large number of modes in a small frequency interval $\omega_m \sim \omega_0 = 2\pi c/\lambda$ pumped and put into oscillations. We can write $a_m(t) = A_m(t)\exp[i\varphi_m(t)]$, which is useful as we expect A_m to be slowly varying with respect to φ_m, as discussed below. In previous works, with reference to standard multimode lasers [21,22], the phase-dependent terms in (5) were always averaged out by assuming the φ_m as rapidly varying, independent, and uniformly distributed. The resulting equations determine the oscillation energy $\mathcal{E}_m$ into each mode. However, for an increasing number of modes, nonlinear beatings induce nontrivial light dynamics which is mainly due to the rapidly varying phases, while the amplitudes can still be taken as slowly varying [21,23,26].

Summing up, the Hamiltonian, depending on the relevant dynamic variables, the phases φ_m, is

$$\mathcal{H}(G, \varphi) = \mathcal{H}_o + \sum G_{spqr}\cos(\varphi_s + \varphi_p - \varphi_q - \varphi_r), \tag{6}$$

where $\mathcal{H}_o = \sum_m(\alpha_m - \gamma_m)A_m^2$ is an irrelevant constant term and $G_{spqr} = g_{spqr}A_s A_p A_q A_r$. Hereafter we will consider these G coefficients as quenched (due to the slow t dependence of A_m).

If the cavity is realized by a random medium, as described above, the coupling coefficients G are random variables. Their values depend on the mode profiles, the resonant frequencies, and on the quenched values of the energies $\mathcal{E}_m = \omega_m A_m^2$ in each mode, which vary with each realization of the cavity for a given pumping rate. For these reasons we take G_{spqr} as random Gaussian variables, with zero mean value $\langle G\rangle = 0$. The latter hypothesis can be removed by a suitable, but not trivial, generalization of the treatment below [15].

The Gs roughly scale as $\langle A^2\rangle^2 V^{-3/2}$, as one can derive from Eq. (4) by using that E_m^α is $O(V^{-1/2})$ and $K_{\alpha\beta\gamma\delta}$ is a random variable integrated over V. Recalling that $\omega_m \sim \omega_0$, $\omega_0\langle A^2\rangle$ measures the average energy per mode. By a simple rescaling, the invariant measure can be written as $\exp[-\beta H(J, \varphi)]$, where $J_{spqr} = G_{spqr}/(g_0\langle A^2\rangle^2)$ has standard deviation $\propto 1/V^{3/2} \propto 1/N^{3/2}$ and g_0 is a material-dependent constant. Note that this scaling of the Js guarantees that the Hamiltonian is extensive [15,19]. The parameter that controls the phase transition is then $\beta \equiv 1/T \equiv \langle A^2\rangle^2 g_0/k_B T_{\text{bath}}$. Thus, in what follows, transitions obtained as β are increased (i.e., the adimensional effective temperature T is decreased), can be controlled by increasing the amount of energy stored on average into each mode (i.e., the pumping rate).

The calculation of the thermodynamics goes through the evaluation of the partition function $Z(J) = \int d\varphi \times \exp[-\beta H(J, \varphi)]$ and, more specifically, of the free energy averaged over the quenched disorder $f(\beta) = \lim_{N\to\infty} N^{-1} \overline{\log Z(J)}$. The latter can be written, using the replica trick [15], in terms of the replicated partition function $\overline{Z^n(J)}$, that is a functional of the overlap matrix $q_{ab} = N^{-1}\sum_j e^{i(\varphi_j^a - \varphi_j^b)}$ and can be computed via a saddle-point method using standard techniques [15,19]. In the following, due to rotational symmetry, we will restrict ourselves to real q_{ab} values. The replica symmetric (RS) solution corresponds to $q_{ab} \equiv q$ and as usual it turns out that $q = 0$. The RS free energy is simply $f_{\rm RS} = -\beta/4$ as in the Ising p-spin glass. In this regime the φ_m are uniformly distributed in $[0, 2\pi)$, independent, and rapidly evolving, as originally considered in [21,22].

In the one step replica-symmetry breaking (1RSB) ansatz one divides the matrix q_{ab} in n/m blocks of side m [15,19]. The elements in the off-diagonal blocks are set to 0 while in the diagonal blocks RS is assumed and $q_{ab} = q$. In this case, the free energy is:

$$\beta f(m, q) = -(\beta^2/4)[1 + 3(1-m)q^4 - 4q^3] - m^{-1} \log \int_0^\infty dz\, z\, e^{-z^2/2} I_0(\beta\lambda z)^m, \qquad (7)$$

where I_n indicates the modified Bessel function of first kind, and $\lambda = \sqrt{2q^3}$. The saddle-point values of m and q $(\bar{m}, \bar{q})$ are determined by $\partial_m f = \partial_q f = 0$. The equation for q is

$$q = \frac{\int_0^\infty dz\, z\, e^{-z^2/2} I_0^m(\beta\lambda z) \frac{I_1(\beta\lambda z)^2}{I_0(\beta\lambda z)^2}}{\int_0^\infty dz\, z\, e^{-z^2/2} I_0^m(\beta\lambda z)}. \qquad (8)$$

Starting from Eqs. (7) and (8)—that are identical to the 1RSB free energy for the $(p = 4)$ p-spin model, the only difference being the presence of the Bessel function instead of the hyperbolic cosine—one can derive the full phase space structure of the model at the 1RSB level [27–29].

In Fig. 1 the solution $\bar{q}$ of Eq. (8) for $m = 1$ is reported as a function of T. At high temperature $\bar{q} = 0$ and the RS solution is recovered. On lowering the temperature, a solution $\bar{q} \neq 0$ first appears at T_d (dashed line). However, it becomes stable only below the thermodynamic glass transition temperature $T_c < T_d$ (full line), as in standard first-order transitions. For $T_c < T < T_d$ the phase space is disconnected in an exponential number $\mathcal{N}(T) = \exp[N\Sigma(T)]$ of states. The 1RSB *complexity* $\Sigma(T)$ is reported in the inset of Fig. 1. At $T = T_d$ a *dynamical transition* is expected to take place [17,20].

The stability of the 1RSB solution can be studied (within the assumption that q_{ab} is real). It turns out that the 1RSB solution is thermodynamically stable for all temperatures, so in this case no Gardner transition is present [27–29].

The presence of a dynamical phase transition at a given value of the random laser pump intensity implies different interesting physical phenomena. These could be experimentally investigated by studying (for example, via heterodyne experiments) the self-correlation function of a specific frequency (ω_m) component of the electric field in the cavity:

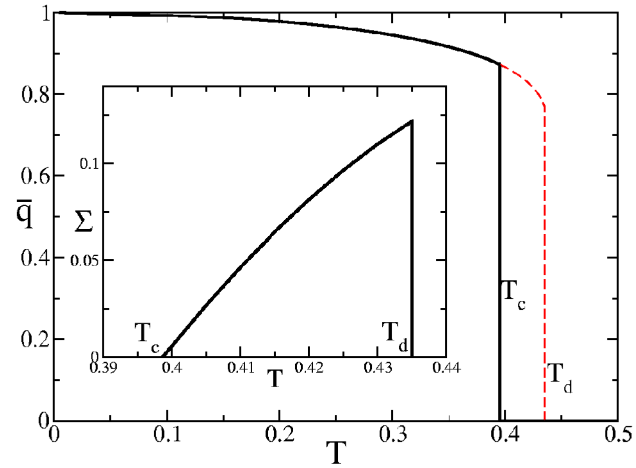

FIG. 1 (color online). The 1RSB overlap $\bar{q}$ as a function of the reduced temperature. The full line is the stable part of the curve; the dashed line is the metastable part. Inset: the complexity $\Sigma(T)$ as a function of the temperature. It jumps discontinuously at T_d where the metastable solution first appears and vanishes at T_c where the glass transition takes place.

$$C(t, \omega_m) = \omega_m A_m^2 \langle \exp\{i[\varphi_m(t+\tau) - \varphi_m(\tau)]\}\rangle_\tau. \qquad (9)$$

On approaching T_d from above (i.e., on increasing the pump power), the dynamics of phase variable $\varphi_m(t)$ becomes slower and slower and $C(t, \omega_m)$ is expected to decay towards zero in longer and longer times. At the dynamical transition, the dynamics of the φ's becomes nonergodic; they are no longer able to explore the whole phase space and the function $C(t, \omega_m)$ decays towards a plateau. In other words, the mode's phases $\varphi_m(t)$—beside from small oscillation—are locked to some "equilibrium" values ("random mode locking"). Because of the nonvanishing value of the complexity Σ at T_d, however, an exponentially large number of such equilibrium positions exist, so giving rise to many different time structures of the electric field in random lasers. On further increasing the pump power, the complexity Σ decreases and the ideal glassy state is reached at $T = T_c$. Below this value the number of equilibrium states is not exponential in N $(\Sigma = 0)$. The equilibrium states below T_d are difficult to reach because the needed time scale diverges [16]: interesting phenomena as aging, memory effects, and history dependent responses are expected to take place for $T < T_d$. All these nonequilibrium phenomena are theoretically predicted and, to some extent, experimentally verified [16] in material systems (structural glasses, spin glasses, ...). The absence of conclusive experiments is mainly due to the long time needed to "equilibrate" real glasses below T_d, a time which is dictated by the relaxation time of the atomic or molecular dynamics in condensed matter (seconds in the interesting glass transition region). *Photon* dynamics in cavities is orders of magnitude faster. This observation drives us to propose lasers in disordered media as a benchmark to test experimentally the outcome of those theories, as the replica-symmetry breaking.

As outlined above, the previous analysis holds for a variety of physical systems. As an example we consider recent experiments on RL [e.g., [10,11]] with some scatterers dispersed in an host medium. Either the former or the latter can play the role of the amplifying medium; as a result, the nonlinear susceptibility varies on scales comparable with the dimensions of the scatterers. We hence take for the generic component of the nonlinear susceptibility, with typical value χ_0, $\langle\chi(\mathbf{r})\chi(\mathbf{r}')\rangle = \chi_0^2 L_r^3 \delta(\mathbf{r}-\mathbf{r}')$, with L_r a typical length of the systems (e.g., the dimension of the scatterers) and δ a coarse-grained Dirac delta. Considering N modes oscillating in a wavelength range $\Delta\lambda$, and taking for the density of modes the standard expression for a box [26], with average refractive index n_0, the scaling arguments reported above lead to an explicit expression of g_0, from which the RSB threshold average energy per mode is obtained

$$\mathcal{E}_{\mathrm{RSB}} = \omega_0\langle A^2\rangle = \sqrt{\frac{k_B T_{\mathrm{bath}} \epsilon_0^2 \lambda^6}{8\pi^{3/2} n_0^{1/2} T_d \chi_0 (L_r \Delta\lambda)^{3/2}}}. \quad (10)$$

The noise temperature can be taken as due to the spontaneous emission, which is typically the dominant contribution, following [26] $2k_B T_{\mathrm{bath}} = \hbar[N_2/(N_2 - N_1)]_t/\tau \cong \hbar/\tau$, with τ the average lifetime per mode, and $[N_2/(N_2 - N_1)]_t$ the population inversion at lasing threshold. Taking typical values from the reported experiments ($\Delta\lambda = 100$ nm, $\lambda = 630$ nm, $n_0 = 2$, $L_r = 10$ nm, $\tau = 100$ fs) and for the susceptibility $\chi_0 = 10^{-27}$ C m V^{-3} [24] it is $\mathcal{E}_{\mathrm{RSB}} \cong 10^{-16}$ J. Assuming a pumping beam with peak power $P_{\mathrm{RSB}} \cong N\mathcal{E}_{\mathrm{RSB}}/\tau$ gives $P_{\mathrm{RSB}} \cong 0.1$ W with $N = 100$, which focused on the typical area of 100 μm^2 provides the typical values for the peak pump intensities used in the experiments ($\cong$100 kW/cm^2). Thus we expect that the glass transition can be observed within the currently available experimental framework.

In conclusion, the multimode dynamics in a random laser cavity has been investigated by statistical physics techniques, and a one step replica-symmetry breaking phase transition has been found. Our results emphasize two important points: (i) the light propagation in nonlinear disordered media shows the same complex behavior of the dynamics of glassy systems (aging, memory, ...) and (ii) due to the faster photon dynamics with respect to the atomic one, it is possible to use the random lasers as systems for experimentally testing the replica-symmetry breaking transitions.

We are pleased to thank T. Castellani, L. Leuzzi, and F. Ricci-Tersenghi for useful discussions.

[1] H. Haken, *Synergetics* (Springer-Verlag, Berlin, 1978).
[2] H. Risken, *The Fokker-Planck Equation* (Springer-Verlag, Berlin, 1989).
[3] F. T. Arecchi, S. Boccaletti, and P. L. Ramazza, Phys. Rep. **318**, 1 (1999).
[4] B. Vodonos, R. Weill, A. Gordon, A. Bekker, V. Smulakovsky, O. Gat, and B. Fischer, Phys. Rev. Lett. **93**, 153901 (2004).
[5] A. Gordon, B. Vodonos, V. Smulakovski, and B. Fischer, Opt. Express **11**, 3418 (2003).
[6] A. Gordon and B. Fischer, Opt. Commun. **223**, 151 (2003).
[7] R. Weill, A. Rosen, A. Gordon, O. Gat, and B. Fischer, Phys. Rev. Lett. **95**, 013903 (2005).
[8] V. S. Lethokov, Sov. Phys. JETP **26**, 835 (1968).
[9] N. M. Lawandy, R. M. Balachandran, A. S. L. Gomes, and E. Sauvain, Nature (London) **368**, 436 (1994).
[10] D. S. Wiersma, M. P. Vanalbada, and A. Lagendijk, Nature (London) **373**, 203 (1995).
[11] H. Cao, X. Jiang, Y. Ling, J. Y. Xu, and C. M. Soukoulis, Phys. Rev. B **67**, 161101(R) (2003).
[12] D. S. Wiersma and S. Cavalieri, Nature (London) **414**, 708 (2001).
[13] L. Florescu and S. John, Phys. Rev. Lett. **93**, 013602 (2004).
[14] R. C. Polson and Z. V. Vardeny, Appl. Phys. Lett. **85**, 1289 (2004).
[15] M. Mézard, G. Parisi, and M. A. Virasoro, *Spin Glass Theory and Beyond* (World Scientific, Singapore, 1987).
[16] L. Cugliandolo, *Dynamics of Glassy Systems, in "Slow Relaxation and Nonequilibrium Dynamics in Condensed Matter"*, Proceedings of the Les Houches Summer School, session LXXVII, edited by J.-L Barrat *et al.* (Springer-Verlag, Berlin, 2002).
[17] J. Kosterlitz and D. Thouless, J. Phys. C **5**, L124 (1972).
[18] L. Angelani, L. Casetti, M. Pettini, G. Ruocco, and F. Zamponi, Europhys. Lett. **62**, 775 (2003).
[19] T. Castellani and A. Cavagna, cond-mat/0505032.
[20] W. E. Lamb, Jr., Phys. Rev. **134**, A1429 (1964).
[21] J. Ducuing and N. Bloembergen, Phys. Rev. **133**, A1493 (1964).
[22] I. C. L. O'Bryan and I. M. Sargent, Phys. Rev. A **8**, 3071 (1973).
[23] H. A. Haus, *Waves and Fields in Optoelectronics* (Prentice-Hall, Englewood Cliffs, NJ, 1984).
[24] R. W. Boyd, *Nonlinear Optics* (Academic, New York, 2002), 2nd ed..
[25] C. W. Gardiner and P. Zoller, *Quantum Noise* (Springer-Verlag, Berlin, 2004), 3rd ed.
[26] A. Yariv, *Quantum Electronics* (Saunders College, San Diego, 1991).
[27] A. Montanari and F. Ricci-Tersenghi, Eur. Phys. J. B **33**, 339 (2003).
[28] E. Gardner, Nucl. Phys. **B257**, 747 (1985).
[29] A. Crisanti, L. Leuzzi, and T. Rizzo, Phys. Rev. B **71**, 094202 (2005).