Order from Disorder in a Kagomé Antiferromagnet

Andrey Chubukov[a]

Department of Physics, University of Illinois at Urbana-Champaign, 1110 West Green Street, Urbana, Illinois 61801 and Institute for Theoretical Physics, University of California at Santa Barbara, Santa Barbara, California 91106-4030

(Received 9 April 1992)

A Heisenberg antiferromagnet on a kagomé lattice is highly degenerate in the classical limit. I show that quantum fluctuations lift the degeneracy and yield the low-energy branch of spin-wave excitations with a velocity which is a factor $S^{-1/3}$ smaller than for conventional spin waves. The relevance of these results to the experiments on the stacked kagomé antiferromagnet $SrCr_{8-x}Ga_{4+x}O_{19}$ is discussed.

PACS numbers: 75.10.Jm, 75.50.Lk

Many frustrated spin systems experience a phenomenon which Villain *et al.* termed order from disorder [1]. Specifically, in the classical limit, the actual degeneracy of the ground state turns out to be larger than required by the symmetry-breaking pattern. This leads to extra zero modes in the spin-wave spectrum and to divergent fluctuation corrections to the on-site magnetization. However, quantum (or thermal) fluctuations, which normally work against ordering, lift the "accidental" degeneracy and thus *restore* long-range order.

The phenomenon was initially observed in several complicated 3D magnets with competing further-neighbor interactions [2,3]. Recently it was also found in 2D frustrated magnetic systems (e.g., in the J_1-J_2-J_3 model [4–7] and in triangular antiferromagnets [8]) which were intensively studied in a context of high-T_c superconductivity.

The 2D Heisenberg antiferromagnet on a kagomé lattice (KAFM) (Fig. 1) is described by

$$H = J_1 \sum_{n,\Delta} \vec{S}_n \cdot \vec{S}_{n+\Delta}. \tag{1}$$

In this system, the order-from-disorder phenomenon plays a much greater role than in any other frustrated magnet due to an extremely high degeneracy of the ground state in the classical limit. In fact, the only requirement for any particular configuration to be a ground state for classical spins is that the total spin of any elementary triangle in Fig. 1 should be equal to zero. This allows for *local* distortions of the spin configuration with no cost in energy and, for coplanar states, leads to a very specific spin-wave spectrum which contains a whole branch of zero-energy excitations (i.e., $\epsilon = 0$ for all k_x, k_y) [9–12].

The activity in the subject of KAFM was initiated by the observation that in a layered compound $SrCr_{8-x}Ga_{4+x}O_{19}$ (Sr-Cr-Ga-O), the $S = \frac{3}{2}$ Cr^{3+} ions form a stack of dense kagomé lattices separated by more dilute triangular lattices [13,14]. (Another example of a KAFM is a second layer of ^3He on graphite [15].) The magnetothermal [13] and neutron-scattering [14] measurements on Sr-Cr-Ga-O reported the existence of a (presumably) spin-glass transition only at $T_c \sim 3.3$ K which is well below the Curie-Weiss temperature Θ_{CW} of about 500 K, obtained from the susceptibility measurements. The specific heat varies as T^2 below T_c which is consistent with the spin-wave theory in 2D, but requires the spin-wave velocity to be about 25 times smaller than Θ_{CW} to account for the experimental data [13]. The challenge for the theory is therefore to explain how this low-energy scale appears in a KAFM.

In the present paper, I address this issue. I will show that at large S quantum fluctuations lift the local degeneracy in KAFM, restore long-range order at $T = 0$, and produce the dispersion for classically soft excitations. The fluctuation-induced spin-wave velocity scales as $C_{sw} \sim J_1 S^{2/3}$ as compared to $J_1 S$ for ordinary spin waves, yielding a specific heat, $C \sim (T/J_1 S)^2 S^{2/3}$, which is a factor $S^{2/3} \gg 1$ larger than that in a conventional antiferromagnet.

One way to lift the degeneracy in a KAFM, which works already in the classical limit, is to consider the interactions between further neighbors, say, second neighbors along basis vectors of the lattice (the J_3 coupling). Besides removing the degeneracy, this interaction also

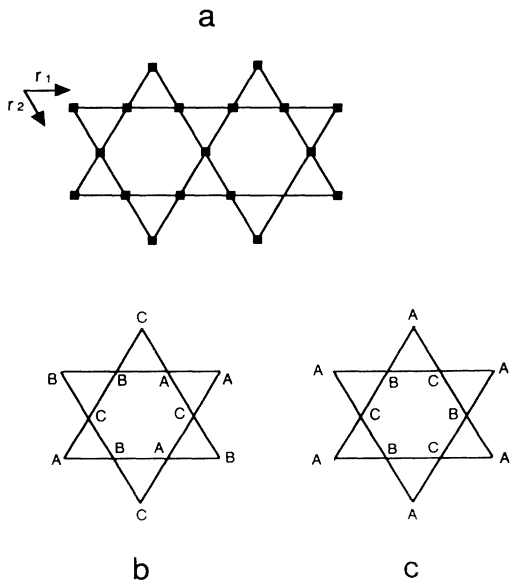

FIG. 1. Two-dimensional kagomé lattice with (a) basis vectors and the two Néel ground states for finite J_3. (b) The $q = 0$ state is selected at $J_3 < 0$ and (c) the $\sqrt{3} \times \sqrt{3}$ state is selected at $J_3 > 0$. A, B, and C label three different spin directions oriented 120° apart.

selects two particular Néel configurations which are the true ground states for nonzero J_3 (Ref. [9]). One of the states is the so-called $q=0$ state of Fig. 1(b) with ferromagnetic ordering of the spins coupled by the J_3 exchange. This configuration is a classical ground state for negative J_3. The other is a conventional 120° antiferromagnet hereafter referred to as a $\sqrt{3}\times\sqrt{3}$ state [Fig. 1(c)]. It has second neighbors along basis vectors oriented 120° apart and is clearly favored when $J_3>0$. My goal is to show that in a quantum KAFM, both states are stable also at $J_3=0$. The analogous results were obtained by Sachdev in the large-N approach to the problem [16].

A standard way to study the role of quantum fluctuations in removing the degeneracy is to calculate the leading corrections to the spin-wave spectrum at $T=0$. These corrections should normally leave only those zero modes which are related to a broken rotational symmetry in the problem. The perturbative approach works well when the "accidental" degeneracy leads to extra points or, at most, lines of zero energy in the spin-wave spectrum [6,7]. However, in a classical KAFM, there is a whole branch of excitations with zero energy. An obvious consequence is that the solution of the quantum problem should be of a self-consistent type, i.e., one should first assume that quantum fluctuations produce a finite stiffness for excitations and then find a self-consistent solution for the energy (alternatively, one can work at finite J_3 and take the limit $J_3\rightarrow 0$ only after renormalization). In principle, the necessity of this procedure raises a question about the validity of restricting to only leading quantum corrections. However, anticipating the result, the additional factor that one picks up by increasing the order in the perturbative expansion is $\alpha=(1/S)(J_1S/C_{sw})\sim S^{-2/3}$. Though α is parametrically larger than the conventional $1/S$, it is still a small parameter which justifies the perturbative approach.

I start with the $q=0$ phase. To obtain the bosonic analog of Eq. (1) is straightforward. I introduce three bosonic fields for the spins labeled as A, B, and C in Fig. 1(b) and use the standard Dyson-Maleev transformation. The spin-wave spectrum has three branches of excitations with the dispersion $\epsilon_k^{(i)}=2J_1S[(A_k^i)^2-(B_k^i)^2]^{1/2}$, where $A_k^i=1+\Delta_i/4$, $B_k=-3\Delta_i/4$, and $i=1,2,3$ labels the branches of excitations. The values of Δ are $\Delta_1=-1$, $\Delta_{2,3}=(1\pm\lambda_k)/2$, where $\lambda_k=(1+8\nu_{k_x}\nu_{k_y}\nu_{k_x+k_y})^{1/2}$, $\nu_{k_j}=\cos k_j$. Two branches of excitations have equal dispersion with a finite spin-wave velocity at $k=0$, $\epsilon_k^{(2,3)}=J_1SQ_k$, where

$$Q_k^2=\tfrac{1}{4}(9-4\lambda_k^2)=\sin^2k_x+\sin^2k_y+\sin^2(k_x+k_y)\,,$$

while the third energy, $\epsilon_k^{(1)}$, is zero for *all* k as a consequence of a local degeneracy in the classical limit [9–11,17].

The leading quantum corrections to the spin-wave spectra are the first-order corrections due to quartic anharmonic terms and the second-order corrections due to cubic terms. Both corrections have an overall factor $1/S$ compared to the bare spectrum. Since the renormalized $\epsilon_k^{(1)}$ is assumed to be parametrically small with respect to $\epsilon_k^{(2,3)}$, the most significant anharmonic contributions are those which contain $\epsilon_k^{(1)}$ in the denominator. Below I will focus only on the renormalization of the soft branch.

The contribution from the quartic terms can be calculated rather easily. In essence, one should simply decouple the fourfold terms in the bosonic version of Eq. (1). This gives corrections to both A_k^1 and B_k^1 in the quadratic form. Since $A_k^1+B_k^1=\frac{3}{2}$ already for classical spins, only corrections to $\delta=A_k^1-B_k^1$ are important. Without renormalization, $\delta=0$. When quartic terms are taken into account, δ acquires a *momentum-independent* contribution,

$$\delta_1=\frac{3}{16SN}\sum_q\frac{2JS}{\epsilon_q^{(1)}}\,. \tag{2}$$

The integration in Eq. (2) is over the Brillouin zone for a triangular lattice. The momentum independence of δ_1 could be anticipated since quartic terms do not distinguish whether the neighboring spins are oriented $+120°$ or $-120°$ apart. In other words, they are identical for any coplanar spin configuration and cannot remove the degeneracy [18].

The evaluation of the diagrams due to the cubic terms requires more effort. Three types of cubic vertices are of equal importance. The first represents the interactions solely between soft bosons while the other two represent the interactions between soft particles and the two other branches of excitations. I will label the corresponding contributions to δ as δ_2 and δ_3, respectively (δ_3 is the total contribution from the last two interactions).

To obtain the vertex functions for triple processes, one should make a transformation to Bose operators which diagonalize the quadratic form. This procedure, as well as the evaluation of diagrams, requires lengthy calculations which will be presented elsewhere. Here I will instead focus only on the results. It is instructive to consider first the point $k=0$. The symmetry-breaking pattern in the $q=0$ phase requires $\epsilon_k^{(1)}$ to have the true Goldstone mode at this point. This is indeed the case when J_3 is finite, $\epsilon_k^{(1)}=2J_1S[\frac{4}{3}\alpha Q_k^2(\frac{3}{2}+\frac{4}{3}\alpha Q_k^2)]^{1/2}\sim|k|$, where $\alpha=|J_3|/J_1$. In the quantum problem, the Goldstone mode is restored only when one combines the contributions from quartic and cubic terms. To see this, I performed the calculations at $k\rightarrow 0$, and obtained

$$\delta_2=-\frac{3}{8SN}\sum_p\frac{2J_1S}{\epsilon_p^{(1)}}\frac{G_p^{(1)}-G_p^{(2)}}{Q_p^4}\,, \tag{3a}$$

$$\delta_3=-\frac{3}{16SN}\sum_p\frac{2J_1S}{\epsilon_p^{(1)}}\frac{4G_p^{(2)}-G_p^{(1)}}{Q_p^4}\,, \tag{3b}$$

where

$$G_p^{(1)}=\sin^4p_x+\sin^4p_y+\sin^4(p_x+p_y)\,,$$

$$G_p^{(2)}=\sin^2p_x\sin^2p_y+\sin^2p_x\sin^2(p_x+p_y)+\sin^2p_y\sin^2(p_x+p_y)\,.$$

When taken together, $\delta_2+\delta_3=-(3/16SN)\sum_p 2J_1S/\epsilon_p^{(1)}$, exactly cancel the contribution from δ_1. After the Goldstone mode is restored, one can expand δ_i near $k=0$ and obtain a self-consistent equation for the spectrum,

$$(\epsilon_k^{(1)})^2=(2J_1S)^2Q_k^2\frac{3}{16SN}\sum_p\frac{2J_1S}{\epsilon_p^{(1)}}\frac{G_p^{(1)}}{Q_p^4}. \tag{4}$$

It immediately follows from Eq. (4) that for small k, $\epsilon_k^{(1)}=2J_1S\bar{C}_{sw}Q_k$ as if the J_3 coupling was finite. The kernal in Eq. (4) is *positive* over the whole Brillouin zone and hence the spin-wave frequencies are real. The spin dependence of the spin-wave velocity follows from $\bar{C}_{sw}^2\sim(1/S)/\bar{C}_{sw}$, which yields $\bar{C}_{sw}\sim S^{-1/3}\ll 1$. Note that to get an exact solution for the spectrum, one should know the kernel in Eq. (4) for arbitrary k which is rather difficult to obtain. However, one can estimate the magnitude of $\bar{C}_{sw}$ by assuming that the whole spectrum preserves the same form ($\epsilon_k^{(1)}\sim Q_k$) as in the classical problem at a finite J_3. Within this assumption, one can perform the integration over the Brillouin zone on the right-hand side of Eq. (4) and obtain $\bar{C}_{sw}=0.42/S^{1/3}$.

The calculations for the $\sqrt{3}\times\sqrt{3}$ phase proceed along the same lines. I preserve the description in terms of the three bosonic fields, only now the neighboring spins coupled by J_3 are oriented 120° apart (this procedure is equivalent to a one-sublattice description of a conventional triangular antiferromagnet). For nearest-neighbor KAFM, the unrenormalized spin-wave spectrum is identical to that in the $q=0$ phase (as well as in any other planar configuration) and involves a branch of zero-energy excitations. At the same time, the symmetry-breaking pattern in the $\sqrt{3}\times\sqrt{3}$ phase is different from that in the $q=0$ phase and, in particular, it requires the true zero modes in $\tilde{\epsilon}_k^{(1)}$ to be located at $\pm k_0$ rather than at $k=0$ (hereafter I use $\tilde{\epsilon}$ to label the excitations in the $\sqrt{3}\times\sqrt{3}$ phase). This is indeed the case when J_3 is finite and positive: $\tilde{\epsilon}_k^{(1)}=2J_1S\alpha^{1/2}[(\frac{9}{4}-Q_k^2)(2+\frac{8}{9}\alpha Q_k^2)]^{1/2}$ touches zero at $k=\pm k_0$, where $Q_{k_0}^2=\frac{9}{4}$. Hence, the first thing one has to prove about the $\sqrt{3}\times\sqrt{3}$ phase is that the combined corrections from quartic and cubic terms preserve the Goldstone modes at $k=\pm k_0$ in the quantum problem as well. The quartic terms are identical for any planar configuration and δ_1 is thus given by Eq. (2). The contributions from the triple processes at $k\to\pm k_0$ are

$$\delta_2=-\frac{3}{8SN}\sum_p\frac{2J_1S}{\tilde{\epsilon}_p^{(1)}}\frac{1}{Q_p^2Q_{2\pi/3-p}^2}(Q_{2\pi/3-p}^2+Q_p^2-\tfrac{9}{4})^2, \tag{5a}$$

$$\delta_3=-\frac{3}{8SN}\sum_p\frac{2J_1S}{\epsilon_p^{(1)}}\frac{1}{Q_p^2Q_{2\pi/3-p}^2}(Q_{2\pi/3-p}^2+Q_p^2-\tfrac{9}{4})\times(\tfrac{9}{4}-Q_p^2). \tag{5b}$$

Here $Q_{2\pi/3-p}=Q_{2\pi/3-p_x,2\pi/3-p_y}$. One can immediately check that $\delta_1+\delta_2+\delta_3=0$ and the Goldstone mode at $k=\pm k_0$ survives the effect of quantum fluctuations. The expansion around $\pm k_0$ then produces a dispersion in $\tilde{\epsilon}_k^{(1)}$ which is linear in deviations from $\pm k_0$:

$$\tilde{\epsilon}_k^{(1)}=2J_1S\tilde{C}_{sw}(\tfrac{9}{2}-2Q_k^2)^{1/2}, \tag{6}$$

where

$$\tilde{C}_{sw}^2=\frac{1}{4SN}\sum_p\frac{2J_1S}{\tilde{\epsilon}_p^{(1)}}\frac{(\frac{9}{4}-Q_{2\pi/3-p}^2)(\frac{9}{4}-Q_p^2)}{Q_p^2Q_{2\pi/3-p}^2}. \tag{7}$$

The kernel in this equation is again *positive* over the whole Brillouin zone and hence the spin-wave velocity $\tilde{C}_{sw}$ is real. The magnitude of $\tilde{C}_{sw}$ can be estimated in the same way as for the $q=0$ state, i.e., by assuming that the whole spectrum preserves the same shape [$\tilde{\epsilon}_k^{(1)}\sim(\frac{9}{2}-2Q_k^2)^{1/2}$] as in the classical problem at a small but finite J_3. This gives $\tilde{C}_{sw}\approx 0.40/S^{1/3}$, which is slightly less than the spin-wave velocity for the $q=0$ state [19].

What emerges from the above analysis is that if one scans over J_3/J_1, the stability regions for the two possible Néel ground states in a KAFM overlap in a finite region $\sim S^{-2/3}$ around the nearest-neighbor KAFM. In both states the Néel long-range magnetic order survives at large S with the fluctuation corrections being of the order of $(1/S)\sum_p 2J_1S/\epsilon_p^{(1)}\sim S^{-2/3}$. The behavior at small S may be different as suggested by several authors [11,20,21], but to investigate it is beyond the scope of the present paper.

Inherent to the studies of the order-from-disorder phenomena is that the renormalized spin-wave theory allows one to find whether a particular state becomes a *local* minimum in the presence of fluctuations. More effort is required to find a global minimum. High-temperature [9] and large-N [16] expansions predict that it should be the $\sqrt{3}\times\sqrt{3}$ state. This is consistent with our observation that the induced spin-wave velocity for the $\sqrt{3}\times\sqrt{3}$ phase is smaller than that for the $q=0$ phase. However, to properly address the question of the global stability within $1/S$ expansion, one should calculate $\epsilon_k^{(1)}$ for all k. This problem is not solved at the moment.

To summarize, I have shown that in a Heisenberg antiferromagnet on a kagomé lattice, quantum fluctuations lift the local degeneracy, restore long-range magnetic order at $T=0$ in both ground-state candidates, and produce a new energy scale for the spin-wave velocity, $C_{sw}=2J_1S\tilde{C}_{sw}$, which at large S is a factor $S^{1/3}$ smaller than the conventional energy scale inferred from the nearest-neighbor exchange [22]. This yields a large specific heat $C(T)\approx 2.06(T/C_{sw})^2\sim(T/J_1S)^2S^{2/3}$.

The T^2 form of $C(T)$ was observed in the $S=\frac{3}{2}$ kagomé antiferromagnet Sr-Cr-Ga-O [13]. The nearest-neighbor exchange was extrapolated from the high-temperature data [14] to be $J_1=57$ K (Ref. [9]). For this value of J_1, C_{sw} should be about $0.2J_1S$ to account for the experimental data for $C(T)$. The extrapolation of the large-S expansion to $S=\frac{3}{2}$ gives a somewhat larger value, $C_{sw}\approx 0.7J_1S$. However, the solution for C_{sw} is only approximate and more work is necessary to conclude

to what extent the new scale produced by quantum fluctuations is relevant to the magnetothermal experiments on Sr-Cr-Ga-O. The study of the neutron-scattering data deserves a separate consideration.

I acknowledge useful discussions with G. Aeppli, I. Affleck, A. J. Berlinsky, P. Coleman, D. Frenkel, C. L. Henley, D. L. Huber, D. Huse, C. Kallin, A. P. Ramirez, S. Sachdev, E. F. Shender, J. Reimers, N. Read, and A. P. Young. I am also grateful to the staff at the Institute for Theoretical Physics in Santa Barbara for the hospitality during the completion of this work. The research was supported by NSF Grants No. DMR 88-09854 through the Science and Technology Center for Superconductivity at UIUC and No. PHY 89-04035 (ITP).

---

[a]Also at the P. L. Kapitza Institute for Physical Problems, Moscow, Russia.

[1] J. Villian, R. Bidaux, J. P. Carton, and R. Conte, J. Phys. (Paris) **41**, 1263 (1980).

[2] See, e.g., E. Rastelli and A. Tassi, J. Phys. C **19**, L423 (1986), and references therein.

[3] E. F. Shender, Zh. Eksp. Teor. Fiz. **83**, 326 (1982) [Sov. Phys. JETP **56**, 178 (1982)]; C. L. Henley, Phys. Rev. Lett. **62**, 2056 (1989).

[4] N. Read and S. Sachdev, Phys. Rev. Lett. **62**, 1694 (1989); S. Sachdev and N. Read, Int. J. Mod. Phys. B **5**, 219 (1991).

[5] M. P. Gelfand, R. R. P. Singh, and D. A. Huse, Phys. Rev. B **40**, 10801 (1989); P. Chandra, P. Coleman, and A. I. Larkin, J. Phys. Condens. Matter **2**, 7933 (1990); P. Chandra and P. Coleman, Phys. Rev. Lett. **66**, 100 (1991); A. Moreo, E. Dagotto, Th. Jolicoeur, and J. Riera, Phys. Rev. B **42**, 6283 (1990).

[6] A. V. Chubukov, Phys. Rev. B **44**, 392 (1991); F. Mila, D. Poliblanc, and C. Bruder, Phys. Rev. B **43**, 7891 (1991).

[7] M. P. Gelfand, Phys. Rev. B **42**, 8206 (1990); S. Sachdev and R. N. Bhatt, Phys. Rev. B **41**, 4502 (1990); A. V. Chubukov and Th. Jolicoeur, Phys. Rev. B **44**, 12050 (1991).

[8] H. Kawamura and S. Miyashita, J. Phys. Soc. Jpn. **54**, 4530 (1985); A. V. Chubukov and D. I. Golosov, J. Phys. Condens. Matter **3**, 71 (1991); Th. Jolicoeur, E. Dagotto, E. Gagliano, and S. Bassi, Phys. Rev. B **42**, 4800 (1990); Q. Sheng and C. L. Henley, J. Phys. Condens. Matter **4**, 2937 (1992); S. E. Korshunov (to be published); A. V. Chubukov and Th. Jolicoeur, Phys. Rev. B (to be published).

[9] A. B. Harris, C. Kallin, and A. J. Berlinsky, Phys. Rev. B **45**, 2889 (1992).

[10] J. T. Chalker, P. S. Holdsworth, and E. F. Shender, Phys. Rev. Lett. **68**, 855 (1992).

[11] I. Ritchey, P. Chandra, and P. Coleman, "Spin Origami: The 2D Heisenberg Kagomé Antiferromagnet" (to be published).

[12] J. N. Reimers, A. J. Berlinsky, and A.-C. Shi, Phys. Rev. B **43**, 865 (1991).

[13] A. P. Ramirez, G. P. Espinosa, and A. S. Cooper, Phys. Rev. Lett. **64**, 2070 (1990); Phys. Rev. B **45**, 2505 (1992).

[14] C. Broholm, G. Aeppli, G. P. Espinosa, and A. S. Cooper, Phys. Rev. Lett. **65**, 3173 (1990).

[15] V. Elser, Phys. Rev. Lett. **62**, 2405 (1989); D. S. Greywall and P. A. Busch, Phys. Rev. Lett. **65**, 2788 (1990).

[16] S. Sachdev, Phys. Rev. B **45**, 12377 (1992).

[17] C. Zeng and V. Elser, Phys. Rev. B **42**, 8436 (1990).

[18] Q. Sheng and C. L. Henley, Bull. Am. Phys. Soc. **37**, 603 (1992).

[19] Actually, to prove that both states are local minima in a KAFM, one should show that the spectrum is real not only near zero modes but over the whole Brillouin zone. For arbitrary k, the calculation of the dispersion is a rather difficult problem. However, I have explicitly calculated the self-energy corrections at $k=(\pm\pi/3,\pm\pi/3)$ for the $q=0$ state and at $k=0$ for the $\sqrt{3}\times\sqrt{3}$ state and in both cases found that the excitation energies are real because the kernels in the corresponding integral equations are positive over the whole Brillouin zone.

[20] R. R. P. Singh and D. Huse, Phys. Rev. Lett. **68**, 1706 (1992).

[21] J. B. Marston and C. Zeng, J. Appl. Phys. **69**, 5692 (1991); T. S. Hsu and A. J. Schofield, J. Phys. Condens. Matter **3**, 8067 (1991).

[22] The analogous results can be obtained if we consider the effects of thermal rather than quantum fluctuations. The induced spin-wave velocity scales with the temperature as $C_{sw}\sim(T/J_1S)^{1/4}$. The long-range magnetic order at $T\rightarrow 0$ was also obtained in numerical simulations for classical spins: D. A. Huse and A. D. Rutenberg, Phys. Rev. B **45**, 7536 (1992).