Experimental search for the chiral glass transition in a ceramic $Bi_2Sr_2CaCu_2O_8$ sample

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The nonlinear susceptibility of a melt-cast $Bi_2Sr_2CaCu_2O_8$ sample, displaying both the paramagnetic Meissner effect and magnetic aging, has been studied experimentally. The presence of magnetic aging in this sample proves that the spontaneous orbital magnetic moments exhibit correlated and frustrated dynamics at low temperatures. The possible existence of a chiral-glass phase transition was investigated by extracting a quantity corresponding to the order parameter susceptibility of the glass phase. It is shown that the expected divergent behavior of this quantity is hampered by relaxation effects and that the sample is out-of-equilibrium already at temperatures slightly below the superconducting transition temperature (T_c). Time dependent magnetization experiments indicate that the magnetic relaxation at temperatures close to but below T_c is due to isolated orbital magnetic moments, not yet being part of the collective orbital moment state that develops at slightly lower temperatures.

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I. INTRODUCTION

It is by now well known that granular high-T_c superconductors can display spontaneous orbital moments. This was first observed in sintered $Bi_2Sr_2CaCu_2O_8$ (Bi-2212) samples,[1,2] but has more recently been observed in artificially engineered grain boundaries in thin films of $YBa_2Cu_3O_7$[3] and $Tl_2Ba_2CuO_{6+\delta}$.[4] These observations give strong support to an explanation based on π junctions arising naturally due to d-wave pairing of the superconducting order parameter;[5,6] one possible candidate for this pairing is the $d_{x^2-y^2}$ state. In case of sintered Bi-2212 samples, the existence of π junctions gives rise to the paramagnetic Meissner effect (PME), implying a positive field cooled (fc) magnetization below the superconducting transition temperature. It should be noted though that a positive fc magnetization is not enough to evidence d-wave pairing of the superconducting order parameter since this has also been observed in conventional superconductors.[7–10] For these superconductors, the explanation instead is based on the idea that during cooling, the surface becomes superconducting before the bulk and hence the magnetic flux in the sample becomes compressed creating an enhanced magnetization.[11]

Recently, magnetic aging was observed in relaxation experiments on a melt-cast Bi-2212 sample.[12] This off-equilibrium property of the dynamics, which has been extensively studied in disorded and frustrated spin systems like spin glasses,[13] strongly suggests a correlated and frustrated behavior of the orbital magnetic moments in such samples. The idea of a collective state for the orbital magnetic moments has recently been corroborated by Monte Carlo simulations on a three-dimensional lattice of Josephson junctions.[14] In these simulations, the Josephson coupling was assumed to be a random variable taking the values J and $-J$ with equal probability, representing 0 and π junctions, respectively. Using this model, it was possible to reproduce all of the experimental findings described in Ref. 12. Moreover, the same model has previously been shown to exhibit a zero field equilibrium phase transition with a broken time-reversal symmetry into the so called chiral-glass state.[15] In this phase, local supercurrents circulating between grains and forming loops carrying a half flux quantum, are frozen in a spatially random manner. It was also argued that the critical exponents associated with the chiral-glass transition are close to those of the three dimensional Ising spin glass.[15] Experimentally, as far as we know, there has been only one attempt to estimate critical exponents in a sample displaying both the PME and magnetic aging.[16] In this particular study, a dynamic scaling analysis was performed finding a value of the critical exponent $z\nu$ in reasonable agreement with theoretical expectations. The experimental uncertainty in that work was however too large to make definite statements about the universality class of the transition.

In this work, we have studied the nonlinear susceptibility of a Bi-2212 sample displaying the PME as well as magnetic aging in relaxation experiments. A quantity corresponding to the order parameter susceptibility in the chiral-glass model was extracted from the low field limit of the nonlinear susceptibility. It is shown that this quantity first increases strongly on decreasing the temperature but its magnitude saturates on a further decrease of the temperature. It is argued that the saturation behavior is due to relaxation effects and therefore it is not possible to probe equilibrium properties. Comparing to measurements of the magnetic relaxation performed on the same sample, we conclude that it will be excessively difficult to fulfill the requirement of equilibrium conditions over any extended temperature range. Differences between real samples and models for the chiral-glass phase are also discussed.

II. EXPERIMENT

The system investigated in this work is a granular $Bi_2Sr_2CaCu_2O_x$ sample, with $x \approx 8.18$, manufactured by a melt-cast process.[17] The sample is single phased apart from a minor fraction of a Bi2223 phase, which is nondetectable in x-ray diffraction, but is revealed from a faint anomaly in the M vs T curves at about 105 K. The onset of superconductiv-

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ity occurs at $T_c \approx 87.5$ K and the transition width, as defined by the low field dc magnetization data, is approximately 5 K. A comparison with typical sintered samples shows that the grains are significantly larger and more densely packed in melt-cast samples. High resolution transmission electron microscopy studies showed that our sample has an extremely polydomain microstructure on a μm length scale.[18] These domains are preferably *c*-axis oriented with atomically sharp interfaces (width $\leqslant 1$ nm), parallel to the *c*-axis. The interface plane can freely rotate around the *c*-axis, resulting in a rather broad distribution of the interface orientations separating domains in the *ab*-plane.

The nonlinear ac susceptibility experiments were performed using a low-field superconducting quantum interference device (SQUID) magnetometer.[19] In this set-up, the dc magnetic field is generated by a small solenoid working in persistent mode during measurements. The sample space is shielded by μ-metal and niobium cans, resulting in, at the position of the sample, a residual field of less than 0.5 mOe. The longitudinal component of this field is further reduced by applying a compensating field yielding a longitudinal residual field of less than 0.1 mOe. The sample was glued to a sapphire rod and was placed in one of the middle coils of a third order gradiometer. During all measurements the sample was kept stationary.

The ac field was generated by a copper coil wound on a sapphire cylinder, placed around the sample. The ac field amplitude and frequency were, unless otherwise stated, 0.8 mOe and 17 Hz, respectively. A second, compensation coil was also wound on the same cylinder. The ac coils were centered in the two middle sections of the gradiometer. The current fed through the compensation coil was adjusted in such a way as for the output of the SQUID electronics to be zero for temperatures above T_c. The in-phase (χ'') and the out-of-phase (χ'') components of the ac susceptibility were simultaneously measured by an EG&G model 7260 lock-in amplifier.

The possible existence of the low temperature chiral-glass phase can be tested by studying the order parameter susceptibility, χ_2, which according to Ref. 15 is expected to show a negative divergence at the chiral-glass transition temperature, T_g,

$$\chi_2 \propto (T/T_g - 1)^{-\gamma} = \epsilon^{-\gamma}, \tag{1}$$

where γ is a critical exponent. Experimentally, χ_2 can be obtained from measurements of the equilibrium nonlinear ac susceptibility, $\chi'_{nl} = \chi_0 - \partial m / \partial H$, where χ_0 is the zero field susceptibility and the magnetization m is written as

$$m = \chi_0 H + \chi_2 H^3 + \chi_4 H^5 + \cdots . \tag{2}$$

In this study, H is composed of an ac field with angular frequency ω and amplitude h superimposed on a dc field H_0, $H(t) = H_0 + h \sin(\omega t)$. For $h \ll H_0$, the lowest order terms of the nonlinear ac susceptibility become

$$\chi'_{nl} = -3\chi_2 H_0^2 - 5\chi_4 H_0^4 - \cdots . \tag{3}$$

Thus, by measuring the nonlinear susceptibility in the limit $H_0 \to 0$, one obtains direct information on χ_2. Another important requirement is to ensure that equilibrium properties are being probed, i.e. close to the transition measurements need to be performed in the $\omega \to 0$ limit.

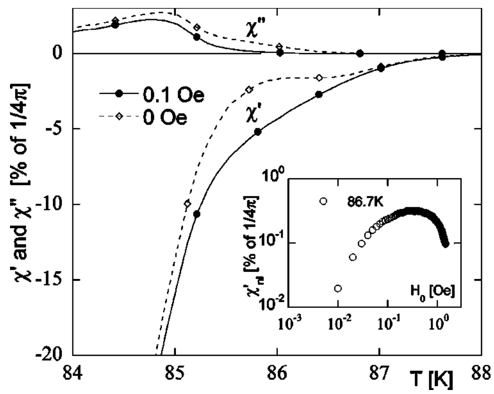

FIG. 1. In-phase χ' and out-of-phase χ'' components of the ac susceptibility versus temperature T. The results are shown for two values of the superimposed dc field, $H_0 = 0$ and $H_0 = 0.1$ Oe. The inset shows the nonlinear susceptibility, χ'_{nl}, vs dc field H_0 at $T = 86.7$ K. $f = 17$ Hz and $h = 0.8$ mOe.

III. RESULTS

Figure 1 shows the temperature dependence close to T_c of the in-phase and out-of-phase components of the ac susceptibility for two superimposed dc fields, $H_0 = 0$ and $H_0 = 0.1$ Oe. The characteristic and rather anomalous features of a PME sample are that χ' becomes *more diamagnetic* applying a weak dc field while χ'' is suppressed.[20] This is further emphasized in the inset of this figure, where the nonlinear susceptibility vs dc field is shown for $T = 86.7$ K. Here it is seen that the characteristic PME behavior is observed up to dc fields $H_0 \sim 0.3$ Oe, while for larger fields the behavior typical for type-II superconductors is observed, implying that field generated intergranular vortices are penetrating the material making it less diamagnetic. It is known that to probe intrinsic zero-field critical properties of a Josephson junction network, the condition $2\pi H_0 S / \Phi_0 \leqslant 1$, where S is a typical loop area, should be fulfilled.[21] If we take $H_0 = 0.3$ Oe to be the cross-over field, below which intrinsic properties are being probed, the typical loop size becomes $\sqrt{S} \approx 3$ μm, which agrees nicely with the size of the polydomain microstructure observed for our sample. The requirement of probing equilibrium properties (vanishing out-of-phase component of the ac susceptibility) is fulfilled for temperatures $T \gtrsim 86.5$ K, but the magnitude of χ'' remains small down to temperatures slightly below 86 K.

Figure 2 shows χ'_{nl} vs H_0 at different temperatures. The nonlinear susceptibility is shown for fields $H_0 \leqslant 0.1$ Oe, i.e., in the field range where the intrinsic properties of the system are being probed. For low enough fields, χ'_{nl} follows an H_0^{α} dependence reasonably well, with $\alpha \approx 2$. There is a tendency however of decreasing α at temperatures $T \leqslant 86$ K and in the low field limit. Moreover, at temperatures below those included in Fig. 2, the magnitude of χ'_{nl} saturates, something which can be attributed to relaxation effects (cf. Fig. 1).

Figure 3 shows χ_2 vs temperature; the values of χ_2 were extracted from the low field dependence of χ'_{nl} shown in Fig.

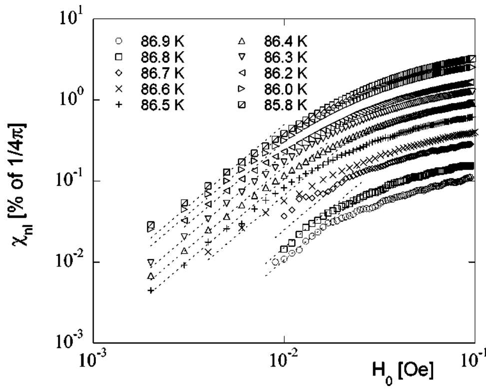

FIG. 2. The nonlinear susceptibility χ'_{nl} versus dc field H_0 for different temperatures (symbols). The dashed curves correspond to fits of a H_0^2-dependence to the low-field χ'_{nl}-data. $f=17$ Hz and $h=0.8$ mOe.

2. Also included in this figure is a fit of the experimental χ_2 data to Eq. (1), excluding data for $T \leqslant 86$ K (dashed line). As can be seen in this figure, the χ_2 data follows the expected behavior reasonably well at temperatures $T \geqslant 86.2$ K, while at lower temperatures the increase of χ_2 levels off, exhibiting clear deviations from the expected divergent behavior. It needs to be emphasized that the margin of error for the critical exponent γ given in Fig. 3 is too large for a precise assessment of the universality class of the chiral–glass phase transition. The deviations from criticality is linked to relaxation in the Josephson junction network, something which will be further discussed below.

A negative divergence of the nonlinear susceptibility at the intergranular transition of a granular $YBa_2Cu_4O_8$ sample was reported in Ref. 22. It was argued that the results were indicative of a chiral–glass phase appearing below this temperature. However, the analyses of the experimental data presented in Ref. 22 are not impeccable. More in particular, the nonlinear susceptibility was extracted from the experimental results without considering effects due to magnetic relaxation and no attempt was made to perform a proper scaling analysis.

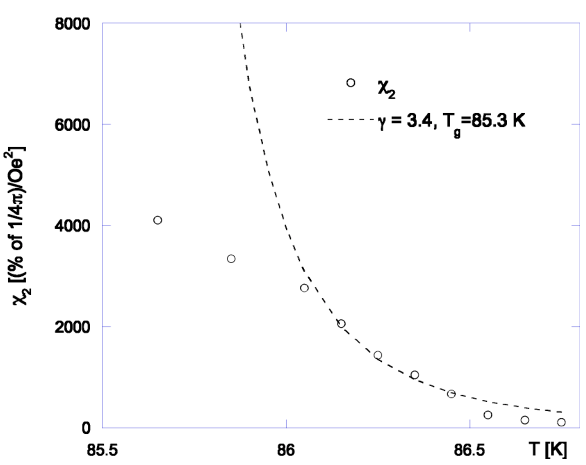

FIG. 3. The order parameter susceptibility χ_2 versus temperature T. The dashed line corresponds to a fit of the χ_2 data to Eq. (1). $f=17$ Hz and $h=0.8$ mOe.

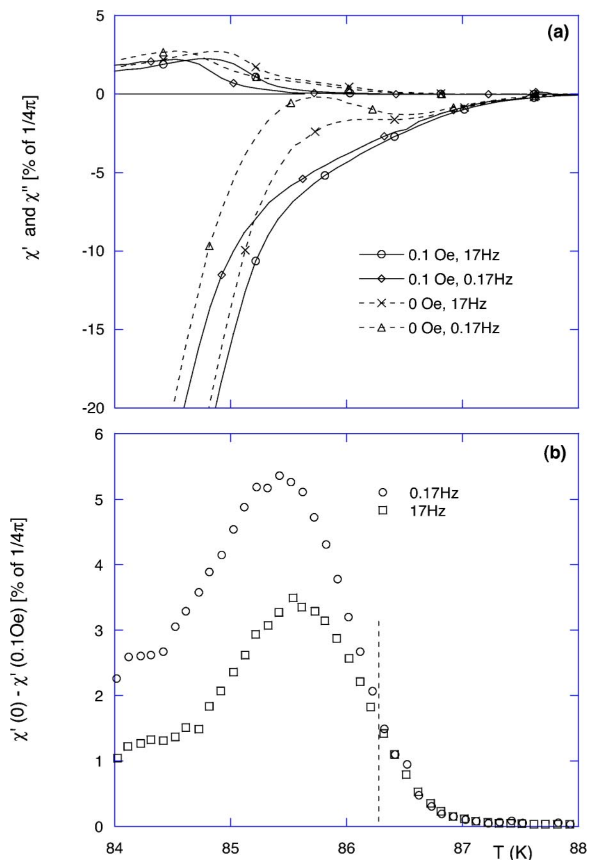

FIG. 4. (a) In-phase χ' and out-of-phase χ'' components of the ac susceptibility versus temperature T. The results are shown for two frequencies, $f=0.17$ Hz and $f=17$ Hz, and two values of the superimposed dc field, $H_0=0$ and $H_0=0.1$ Oe. (b) $\chi'(0)-\chi'(0.1\text{ Oe})$ vs temperature T for two frequencies, $f=0.17$ Hz and $f=17$ Hz. $h=0.8$ mOe.

Figure 4(a) shows the temperature dependence of the in-phase and out-of-phase components of the ac susceptibility for two frequencies, $f=0.17$ Hz and $f=17$ Hz, and for the same superimposed dc fields as used in Fig. 1. These results indicate that there is a noticeable frequency dependence of $\chi'(0)$ at temperatures $T \leqslant 86.4$ K. Moreover, it is below this temperature where $\chi''(0)$ increases in magnitude, thus indicating that the system is out-of-equilibrium for temperatures below 86.4 K. This is further emphasized in Fig. 4(b) where $\chi'(0)-\chi'(0.1\text{ Oe})$, which can be taken as a measure of the nonlinear response applying a dc field of $H_0=0.1$ Oe, is shown vs temperature. A clear frequency dependence is seen for temperatures $T \leqslant 86.4$ K.

IV. DISCUSSION AND CONCLUSIONS

The results presented above expose some serious difficulties met when attempting to explore the possibility of a chiral–glass transition. Why is it that Monte Carlo (MC) simulations[15] on the three dimensional lattice model, introduced in Ref. 23 to describe the PME, find evidence of a chiral–glass transition, while the same evidence continuous to elude the experimentalist? To be able to address to this question, one will have to compare the chiral–glass model with a real sample displaying both the PME and collective behavior as evidenced by magnetic aging in relaxation experiments.

The MC simulations described in Ref. 15 were performed on rather small and fully equilibrated lattices. Moreover, the simulations used temperature independent coupling constants $\pm J$, which implies that spontaneous orbital moments can arise in multi-junction loops even for arbitrarily weak coupling constants.[21] As a consequence, the equilibrium zero field susceptibility is paramagnetic at all temperatures.[15] This is in contrast with the single-junction (π-junction) model,[5,6] where there appears a critical value for the parameter $\widetilde{\mathcal{L}} = 2\pi\mathcal{L}J/\Phi_0$; a spontaneous orbital moment only exists for $\widetilde{\mathcal{L}} > 1$. It is also in contrast with a multi-junction model where the coupling constants are $-J$ and ϵJ and where $\epsilon > 3$, since also in this case there appears a critical $\widetilde{\mathcal{L}}$-value below which spontaneous orbital moments cannot exist.[21]

In a real sample, temperature dependent coupling constants are a reality and there exists a distribution of coupling constants both for 0 and π junctions. A likely scenario is as follows. Below T_c, the grains become superconducting and the sample displays diamagnetic behavior because of intragranular screening currents. Some of the superconducting loops made up of connected grains will be frustrated and carry weak spontaneous orbital moments ($\propto \widetilde{\mathcal{L}}$ for weak coupling constants[21]), while for other loops there exists a critical $\widetilde{\mathcal{L}}$-value, implying that for these loops no spontaneous moments will arise until on decreasing temperature, J has become sufficiently large. As a consequence, in a real sample, the Josephson junction network at high temperature is dilute with respect to spontaneous orbital moments but the number of loops that carry spontaneous orbital moments increases with decreasing temperature. The equilibrium susceptibility is therefore diamagnetic close to T_c, but will become paramagnetic as the network develops on decreasing the temperature. Close to T_c, it may even be that there will exist isolated orbital moments not being part of the collective state. The evidence for this comes from magnetic relaxation measurements performed on the same sample, studied in Ref. 12. Even at temperature as high as $T = 86$ K, the relaxation effects extend over time scales much larger than 10^4 s. In fact, the relaxation is close to logarithmic in time and shows no tendency to approach equilibrium. Moreover, the relaxation of the magnetization makes the susceptibility paramagnetic for observation times $t > 30$ s, which clearly indicates that the magnetic relaxation is due to the time dependent polarization of spontaneous orbital moments. Still, at this temperature, there is no sign of magnetic aging; it is not possible to detect magnetic aging until the temperature is below $T \simeq 85.5$ K. This can be contrasted to spin glasses that exhibit both magnetic aging and a phase transition to a low temperature glass state. For spin glasses and for temperatures $T > T_g$, long time relaxation is due to correlated spin dynamics and magnetic relaxation ends at a well defined temperature dependent maximum relaxation time $\tau_m(T)$. Moreover, if the spin system is equilibrated for a waiting time $t_w \geqslant \tau_m(T)$ magnetic aging effects are absent. The absence of aging in conjunction with a logarithmic relaxation in the case of the PME sample therefore indicates that the relaxation at $T = 86$ K is due to isolated orbital moments with a rather broad distribution of energy barriers to yield a logarithmic relaxation. The isolated moments exhibit magnetic relaxation as well as a nonlinear response, which may mask the equilibrium behavior of the collective state of the orbital moments. Whether or not these effects will destroy the chiral–glass phase transition is left for future work to resolve.

In summary, the requirement of performing experiments on a sample in equilibrium turns out to be the most difficult to fulfill. The origin of these difficulties can be traced back to the complexity of the Josephson junction network in real samples, including temperature dependent coupling constants and the influence of magnetic relaxation from isolated orbital moments not being part of the collective state. The mixture of collective and isolated orbital moment effects, and the nonequilibrium state of the latter, makes it difficult to evidence a chiral–glass transition in the presently investigated Bi-2212 sample. However, further experimental studies on other melt-cast samples exhibiting collective behavior, as evidenced by, e.g., magnetic aging, are needed to make a more definite statement regarding the possible existence of a low-temperature chiral–glass phase in ceramic high-T_c samples.

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[1] P. Svedlindh, K. Niskanen, P. Nordblad, L. Lundgren, B. Lönnberg, and T. Lundström, Physica C **162–164**, 1365 (1989).

[2] W. Braunisch, N. Knauf, V. Kataev, S. Neuhausen, A. Grutz, A. Kock, B. Roden, D. Khomskii, and D. Wohlleben, Phys. Rev. Lett. **68**, 1908 (1992).

[3] C. C. Tsuei, J. R. Kirtley, C. C. Chi, Lock See Yu-Jahnes, A. Gupta, T. Shaw, J. Z. Sun, and M. B. Ketchen, Phys. Rev. Lett. **73**, 593 (1994).

[4] C. C. Tsuei, J. R. Kirtley, M. Rupp, J. Z. Sun, A. Gupta, M. B. Ketchen, C. A. Wang, Z. F. Ren, J. H. Wang, and M. Bhushan, Science **271**, 329 (1996).

[5] M. Sigrist and T. M. Rice, J. Phys. Soc. Jpn. **61**, 4283 (1992).

[6] M. Sigrist and T. M. Rice, Rev. Mod. Phys. **67**, 503 (1995).

[7] D. J. Thompson, M. S. M. Minhaj, L. E. Wenger, and J. T. Chen, Phys. Rev. Lett. **75**, 529 (1995).

[8] P. Kostic, B. Veal, A. P. Paulikas, U. Welp, V. R. Todt, C. Gu, U. Geiser, J. M. Williams, K. D. Carlson, and R. A. Klemm, Phys. Rev. B **53**, 791 (1996).

[9] L. Pust, L. E. Wenger, and M. R. Koblischka, Phys. Rev. B **58**, 14 191 (1998).

[10] A. K. Geim, S. V. Dubonos, J. G. S. Lok, M. Henini, and J. C. Maan, Nature (London) **396**, 144 (1998).
[11] A. E. Koshelev and A. I. Larkin, Phys. Rev. B **52**, 13 559 (1995).
[12] E. L. Papadopoulou, P. Nordblad, P. Svedlindh, R. Schöneberger, and R. Gross, Phys. Rev. Lett. **82**, 173 (1999).
[13] L. Lundgren, P. Svedlindh, P. Nordblad, and O. Beckman, Phys. Rev. Lett. **51**, 911 (1983).
[14] M. S. Li, P. Nordblad, and H. Kawamura, Phys. Rev. Lett. **86**, 1339 (2001).
[15] H. Kawamura and M. S. Li, Phys. Rev. Lett. **78**, 1556 (1997); J. Phys. Soc. Jpn. **66**, 2110 (1997).
[16] E. L. Papadopoulou, P. Nordblad, and P. Svedlindh, Physica C **341–348**, 1379 (2000).
[17] W. Braunisch, N. Knauf, G. Bauer, A. Kock, A. Becker, B. Freitag, A. Grutz, V. Kataev, S. Neuhausen, B. Roden, D. Khomskii, D. Wohlleben, J. Bock, and E. Preisler, Phys. Rev. B **48**, 4030 (1993).
[18] B. Freitag, B. Büchner, N. Knauf, B. Roden, H. Micklitz, A. Freimuth, and V. Kataev, Europhys. Lett. **45**, 393 (1999).
[19] J. Magnusson, C. Djurberg, P. Granberg, and P. Nordblad, Rev. Sci. Instrum. **68**, 3761 (1997).
[20] J. Magnusson, E. Papadopoulou, P. Svedlindh, and P. Nordblad, Physica C **297**, 317 (1998).
[21] H. Kawamura and M. S. Li, Phys. Rev. B **54**, 619 (1996).
[22] M. Matsuura, M. Kawachi, K. Miyoshi, M. Hagiwara, and K. Koyama, J. Phys. Soc. Jpn. **64**, 4540 (1995).
[23] D. Dominguez, E. A. Jagla, and C. A. Balseiro, Phys. Rev. Lett. **72**, 2773 (1994).