Columnar Order and Ashkin-Teller Criticality in Mixtures of Hard Squares and Dimers

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We show that critical exponents of the transition to columnar order in a mixture of 2×1 dimers and 2×2 hard squares on the square lattice depends on the composition of the mixture in exactly the manner predicted by the theory of Ashkin-Teller criticality, including in the hard-square limit. This result settles the question regarding the nature of the transition in the hard-square lattice gas. It also provides the first example of a polydisperse system whose critical properties depend on composition. Our ideas also lead to some interesting predictions for a class of frustrated quantum magnets that exhibit columnar ordering of the bond energies at low temperature.

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Introduction.—In materials that exhibit a continuous transition from a low-density fluid to an ordered high-density crystalline state with spontaneous symmetry breaking, critical properties in the vicinity of the transition are generally independent of microscopic details such as chemical composition and the precise form of the interactions. Indeed, in the standard theory of such critical phenomena, these properties are generally expected to depend only on the symmetries of the ordered state. This universality of critical properties makes it possible to understand such behaviors in terms of simple models. Lattice-gas models of hard-core particles, with different sizes and shapes of the excluded-volume region around each particle, provide many paradigmatic examples of such continuous transitions from a low-density fluid to a high-density ordered state [1–9].

One such simple lattice-gas model of 2×2 hard squares on the square lattice, has long been of special interest and some controversy. Here, the crystalline state has a sliding instability that leads to long-range columnar (stripe) order in the high-density phase [10–18]. General symmetry arguments [19,20] suggest that the transition to this columnar ordered phase should provide an example of "Ashkin-Teller" (AT) critical behavior [21–38]. Such Ashkin-Teller transitions are interesting exceptions to universality, since the correlation length for columnar order is expected to grow with a power-law exponent ν that depends on microscopic details. In light of this, it is surprising that several large-scale Monte Carlo simulations [15–17] found critical properties that are very close to those of a two-dimensional Ising model. Some of these [15] favored an Ising critical point, while others identified small deviations from Ising behavior [16,17].

In this Letter, we show that critical exponents of the transition to columnar order in a more general *mixture* of 2×1 dimers and 2×2 hard squares on the square lattice [Fig 1(a)] depends on the *composition* of the mixture in exactly the manner predicted by the theory of Ashkin-Teller criticality, including in the hard-square limit. This result settles the question regarding the nature of the transition in the hard-square lattice gas. It also provides the first example of a polydisperse system whose critical properties depend on composition. Our ideas also lead to some interesting predictions for a class of frustrated quantum magnets that exhibit columnar ordering of the bond energies at low temperature.

The original hard-square lattice gas corresponds to the boundary line VS in the phase diagram [Fig. 1(b)] of this more general model, while line VD is the well-studied monomer-dimer model [39–46]. For the vacancy-free mixture along line DS [Fig. 1(b)], we show that the power-law columnar order present in the dimer limit D is enhanced by adding hard squares. This eventually leads

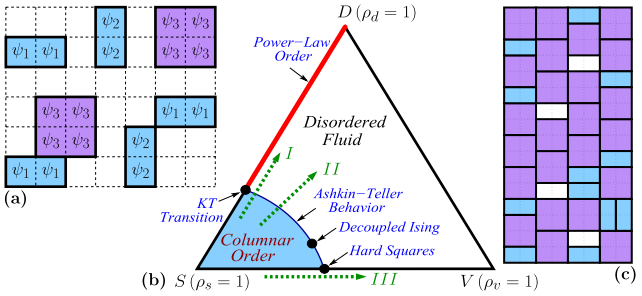

FIG. 1 (color online). (a) Part of a low-density configuration of 2×1 tiles (dimers) and 2×2 tiles (hard squares) on the square lattice, also showing values of the columnar order parameter field $\psi(\vec{r})$ [see Eq. (2)]. (b) Schematic phase diagram of Z_{dsv}. ρ_s, ρ_d, and ρ_v are the densities of squares, dimers, and vacancies, respectively, with $\rho_s + \rho_d + \rho_v = 1$. Monte Carlo results along the cuts I, II, and III are discussed in the text. (c) Columnar ordered high-density configuration, with stripes running in the vertical direction.

to a Kosterlitz-Thouless (KT) phase transition from this power-law ordered phase to a hard-squares-rich phase with long-range columnar order [Fig. 1(c)]. Noting that the power-law ordered phase and the KT point are both characterized by an emergent U(1) symmetry, we show that correlations of the two-sublattice order parameter of hard squares decay in this regime with the same power-law exponent as those of the nematic order parameter. With vacancies allowed, we establish that the phase boundary [Fig. 1(b)] between this columnar ordered phase and the low-density fluid is in the Ashkin-Teller (AT) universality class with a fixed anomalous exponent $\eta = 1/4$ for the columnar order parameter, and a continuously varying correlation length exponent ν. We also demonstrate that the anomalous exponent η_2 for *nematic* order obeys an Ashkin-Teller relation $\eta_2 = 1 - 1/(2\nu)$ along the phase boundary, including at the hard-square transition, thus settling the original question of critical properties at the hard-square transition. These results are made possible by our identification of a detailed correspondence between the microscopic hard-square and dimer variables measured in our Monte Carlo simulations and the XY (Ising) order-parameter fields of a long-wavelength description of KT (AT) criticality. Some of our results on the hard-square lattice gas were summarized earlier in the doctoral thesis of K. Ramola [47] at the TIFR.

Model.—Our analysis begins by defining a lattice gas [Fig. 1(a)] of hard squares that occupy the four elementary plaquettes of a square lattice, dimers that occupy two plaquettes, and vacant single plaquettes (vacancies or monomers). We consider a $L \times L$ square lattice with periodic boundary conditions and associate activities z_s, z_d, and z_v with each square, dimer, and vacancy, respectively. The grand partition function of the system is then given by

$$Z_{dsv} = \sum_{\mathcal{C}_{dsv}} z_s^{N_s} z_d^{N_d} z_v^{N_v}. \tag{1}$$

Here, the sum is over all allowed configurations $\mathcal{C}_{dsv}$ that respect the hard-core constraints [Fig. 1(a)], and N_s, N_d, and N_v, the total numbers of squares, dimers, and vacancies, obey the constraint $4N_s + 2N_d + N_v = L^2$, allowing us to parametrize results in terms of two independent parameters: $v = z_v z_s^{-1/4}$ and $w = z_d/\sqrt{z_s}$.

Line DS.—At $v = 0$, Z_{dsv} reduces to Z_{ds}, the partition function of a vacancy-free mixture of squares and dimers. In the $z_s \to 0$ limit, Z_{ds} further reduces to Z_{dimers}, the partition function of the fully packed dimer model. Z_{dimers} is characterized by a power-law tendency to columnar order manifested in the connected correlation function of horizontal (vertical) dimers, which decays as $(-1)^l/l^2$ for large separation l along the x (y) axis [42]. For small but nonzero w^{-1}, Z_{ds} involves configurations with a small density of squares. Regarding each square as a length-four loop and each dimer as a length-two loop on the dual lattice allows us to use the recursive procedure of Ref. [48] to map Z_{ds} to an interacting dimer model with k-dimer interactions ($k = 2, 3, \ldots$). The leading interaction is a two-body attraction V_2 of strength $\log[1 + 1/(2w^2)]$ between two adjacent dimers whose long sides touch fully. As seen in earlier work [35–37], this interaction enhances the power-law columnar order present in the dimer limit, with power-law exponent $\eta(w)$ decreasing from $\eta(w = \infty) = 2$ as V_2 increases in strength. Furthermore, the net effect of the $k > 2$ interaction terms also favors columnar ordering. Therefore, for w less than a critical value $w_c^{(0)}$, we expect a phase with long-range columnar order. In this columnar state, the symmetry of $\pi/2$ rotations is broken and the unit cell is doubled in the direction perpendicular to the stripes that form [Fig. 1(c)].

This fourfold symmetry breaking is conveniently characterized in terms of a complex order parameter $\psi(\vec{r})$ defined on plaquettes $\vec{r}$ in terms of microscopic variables as follows: $\psi(\vec{r})$ vanishes at $\vec{r}$ if plaquette $\vec{r}$ is vacant. Otherwise, it takes on the values depicted in Fig. 1(a). These values are specified based on the coordinate $\vec{R} \equiv (m, n)$ of the bottom, left corner of the tile covering $\vec{r}$ as follows:

$$\begin{aligned} &\psi_1 = (-1)^m, \qquad \psi_2 = -i(-1)^n, \\ &\psi_3 = [(-1)^m - i(-1)^n]/\sqrt{2}. \end{aligned} \tag{2}$$

With this definition, $\langle\psi\rangle$ takes on values $\pm a, \pm ia$ in the four symmetry-related columnar-ordered states (the magnitude $a > 0$ depends on the composition of the mixture), while $\langle\psi^*(\vec{r})\psi(0)\rangle$ falls off as $1/r^{\eta(w)}$ for large r in the power-law columnar-ordered phase.

To understand the nature of the transition at $w_c^{(0)}$ along DS [Fig. 1(b)], we use the fact that Z_{ds} admits a height representation; i.e., the microscopic configurations are uniquely specified in terms of a single-valued scalar height $H(\vec{R})$ defined on lattice sites $\vec{R}$ as follows: Set $\eta_{mn} \equiv (-1)^{m+n}$ and the height at the origin $H(\vec{O}) = 0$. To construct the height field $H(\vec{R})$, traverse any sequence of links of the square lattice to go from $\vec{O}$ to $\vec{R} \equiv (m, n)$. When traversing a vertical link from (m, n) to $(m, n + 1)$ [horizontal link from $(m + 1, n)$ to (m, n)], H increases by $3\eta_{mn}/4$ if this link is fully covered by a dimer, by $\eta_{mn}/4$ if fully covered by a square, and by $-\eta_{mn}/4$ otherwise. When there are no squares, this reduces to the well-known height representation for the fully packed dimer model [37,49–56].

In the $w > w_c^{(0)}$ power-law ordered phase, long-wavelength fluctuations of the height field are well described by the effective action [37,49–52,54,56]:

$$S_{\text{eff}} = \int_\Lambda d^2x \left[\pi g(\nabla h)^2 + \sum_{n=4,8,12\ldots} u_n \cos(2\pi n h)\right]. \tag{3}$$

Here h is a coarse-grained version of the microscopic height field $H(\vec{R})$, the values of the stiffness g and n-fold anisotropy terms u_n at the coarse-graining scale Λ are phenomenological parameters, and the form of the cosine terms in the action are fixed [37,54,56] by the transformation properties of h under lattice symmetries of the original partition function.

The utility of $S_{\rm eff}$ lies in two observations: First, since $e^{2\pi i h(\vec{r})}$ transforms [37,54,56] under lattice symmetries in the same way as $\psi(\vec{r})$, we expect long-distance properties of correlators of $\psi(\vec{r})$ in Z_{dsv} to correspond to those of $e^{2\pi i h(\vec{r})}$ in the coarse-grained theory $S_{\rm eff}$. Second, $S_{\rm eff}$ with all u_n set to zero represents a line of critical fixed points parametrized by a variable stiffness g. All allowed cosine terms u_n are irrelevant perturbations of this fixed line for $g < 4$ [24]. Along this fixed line [24], $\langle e^{2\pi i[h(\vec{r})-h(0)]}\rangle$ falls off as $1/r^{1/g}$. This implies power-law columnar order with exponent $\eta = g^{-1}$, since correlations of $\psi(\vec{r})$ and $e^{2\pi i h(\vec{r})}$ have the same long-distance behavior. Therefore, we may identify the $w \to \infty$ limit of Z_{ds} with the point [37] $g = 1/2$ on this fixed line, consistent with $\eta(\infty) = 2$. Since we have already argued that $\eta(w)$ reduces as w^{-1} is increased from 0, we expect that the corresponding value of g increases on this fixed line until it hits $g = 4$, corresponding to $\eta = 1/4$. At this point, u_4 becomes marginally relevant, driving a Kosterlitz-Thouless (KT) transition to a fourfold symmetry-breaking state with long range order for $e^{2\pi i h(\vec{r})}$, i.e., a columnar ordered state with nonzero $\langle\psi\rangle$.

This irrelevance of all cosine terms in the power-law ordered phase implies that the phase of $\Psi_L \equiv \sum_r \psi(\vec{r})$ for large L will be uniformly distributed in $(0, 2\pi)$ throughout the power-law ordered phase and at the KT point, reflecting the presence of an emergent U(1) symmetry. From their microscopic expressions, we note that $\mathrm{Re}[\psi^2(\vec{r})]$ measures nematic order in terms of orientations of dimers, while $\mathrm{Im}[\psi^2(\vec{r})]$ is the two-sublattice order parameter of hard squares. This U(1) symmetry implies that η_s, the anomalous exponent governing the power-law correlations of $\mathrm{Im}[\psi^2(\vec{r})]$, equals η_2, the corresponding exponent for $\mathrm{Re}[\psi^2(\vec{r})]$. The Gaussian nature of height fluctuations further ensures that both η_2 and η_s equal 4η throughout this power-law phase and at the KT point.

The AT phase boundary.—The KT transition at $(w = w_c^{(0)}, v = 0)$ represents the beginning of an Ashkin-Teller critical line in the phase diagram of Z_{dsv} [Fig. 1(b)], at whose other end $(w = 0, v = v_c^*)$ lies the density-driven transition of the hard-square lattice gas. To establish this, we first note that it is enough to keep a nonzero u_4 and set all other u_n in $S_{\rm eff}$ to zero in the vicinity of this KT transition at $g = 4$ [24]. Thus, the $v = 0$ KT transition can be thought of as a transition to long-range order in a vortex-free XY model with fourfold anisotropy. Next, we note that an isolated vacancy on plaquette $\vec{r} = (m + 1/2, n + 1/2)$ causes the phase of the XY order parameter $\psi(\vec{r})$ to wind by $2\pi \times (-1)^{m+n}$ along a circuit that encloses the vacant plaquette once. On the vacant plaquette itself, $\psi = 0$, as befits the core of a vortex in an XY order parameter. Thus, a nonzero density of vacancies in Z_{dsv} corresponds to perturbing this vortex-free, fourfold anisotropic XY model with a nonzero density of vortices and antivortices. As is well known from the work of Kadanoff and others on such XY models with fourfold anisotropy [24–28,30,31,35–37], vorticity and fourfold anisotropy "balance" each other along a line of fixed points that starts at this vortex-free KT point. This fixed line describes the continuously varying critical properties of the Ashkin-Teller universality class [21–38], i.e., the critical behavior of two Ising models coupled via their energy densities. For Z_{dsv}, this implies that the $(w = w_c^{(0)}, v = 0)$ KT transition represents the start of an AT critical line that separates a square-rich columnar-ordered phase from a low-density fluid phase [Fig. 1(b)]. The density-driven transition at $(w = 0, v = v_c^*)$ in the hard-square lattice gas thus represents the other end of this AT line. The two real scalar fields σ and τ of this alternate Ashkin-Teller description are defined in terms of the XY order parameter ψ [defined in Fig. 1(a)] by the equation

$$\psi(\vec{r}) \equiv \frac{\sigma(\vec{r}) + \tau(\vec{r})}{2} + i\frac{\sigma(\vec{r}) - \tau(\vec{r})}{2}. \tag{4}$$

From their expressions in terms of microscopic variables, it is clear that lattice symmetries only guarantee

$$\langle\sigma(\vec{r}_1)\tau(\vec{r}_2)\rangle = 0, \qquad \langle\sigma(\vec{r})\sigma(0)\rangle = \langle\tau(\vec{r})\tau(0)\rangle. \tag{5}$$

In particular, $\langle\sigma^2(\vec{r})\tau^2(0)\rangle$ is not constrained to vanish even in the pure hard-square limit, and there is no symmetry reason to expect that the Ising fields σ and τ are asymptotically decoupled.

Numerics.—These ideas, in conjunction with our knowledge [24–28,30,31,35–37] of the long-wavelength physics of the Ashkin-Teller universality class, lead to three key predictions that can be tested via numerical simulations: All along the AT phase boundary, $\langle\psi^*(\vec{r})\psi(0)\rangle$ is predicted to fall off as $1/r^{1/4}$, while $\langle\mathrm{Re}[\psi^2(\vec{r})]\mathrm{Re}[\psi^2(0)]\rangle$ is expected to decay as $1/r^{\eta_2(v)}$, where $\eta_2(v)$ varies continuously, starting from the $v = 0$ value $\eta_2(v = 0) = 1$. Thus, η_2 is a natural coordinate in terms of which one can specify the position along the AT phase boundary. Moreover, the correlation-length exponent ν is related to η_2 via an Ashkin-Teller relation:

$$\eta_2 = 1 - 1/(2\nu). \tag{6}$$

In the power-law ordered phase at full packing, our earlier results imply, via finite-size scaling, that $\mathcal{C}(L) = \langle|\Psi_L|^2\rangle/L^2$ scales as $L^{2-\eta(w)}$, while $\mathcal{R}(L) = \langle\{\sum_{\vec{r}}\mathrm{Re}[\psi^2(\vec{r})]\}^2\rangle/L^2$ and $\mathcal{I}(L) = \langle\{\sum_{\vec{r}}\mathrm{Im}[\psi^2(\vec{r})]\}^2\rangle/L^2$ scale as $L^{2-4\eta(w)}$. In the vicinity of the AT phase boundary, finite-size scaling implies that $\mathcal{C}(L)$ and $\mathcal{R}(L)$

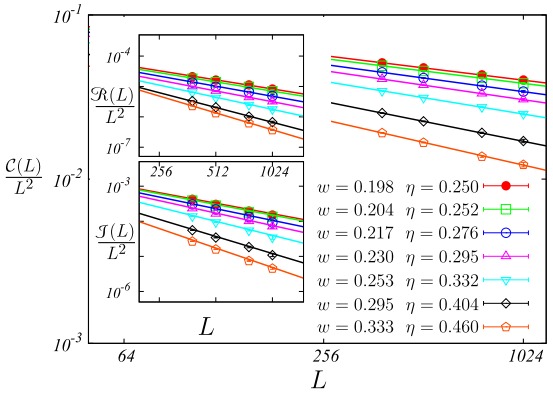

FIG. 2 (color online). $\mathcal{C}(L)/L^2 \sim L^{-\eta(w)}$ with variable exponent $\eta(w)$ in the power-law ordered phase at full packing. Insets: $\mathcal{R}(L)/L^2$ and $\mathcal{I}(L)/L^2$ both scale as $L^{-4\eta(w)}$ with the same $\eta(w)$.

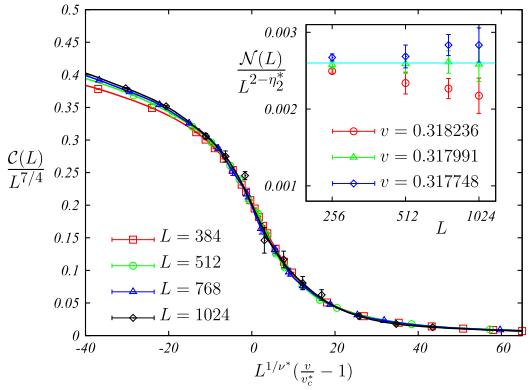

FIG. 3 (color online). Scaling collapse of $\mathcal{C}(L)/L^{7/4}$ for various L at the hard-square transition yields the estimate $\nu^* = 0.92(3)$ and $v_c^* = 0.31799(30)$. Inset: $\mathcal{N}(L)/L^{2-\eta_2^*}$ is a constant for $v = v_c^*$ with $\eta_2^* \approx 0.46(3)$.

are expected to satisfy the scaling forms $L^{7/4} f_{\mathcal{C}}(\delta L^{1/\nu})$ and $L^{2-\eta_2(v)} f_{\mathcal{R}}(\delta L^{1/\nu})$ respectively, where δ denotes the deviation from criticality and the f are finite-size scaling functions. Close to the density-driven hard-square transition, it is more convenient to measure η_2 using an alternate nematic order parameter $T(\vec{r})$, which keeps track of the orientations of vacancy pairs and dimers adjacent to hard squares: $T(\vec{r}) = 0$ when $\vec{r}$ is not covered by a hard square. Otherwise, $T(\vec{r}) \equiv T_H(\vec{r}) - T_V(\vec{r})$, where $T_H(\vec{r})$ $[T_V(\vec{r})]$ equals one-quarter the total number of horizontal (vertical) vacancy pairs or dimers immediately adjoining the hard square that covers $\vec{r}$. $T(\vec{r})$ transforms in the same way as $\mathrm{Re}[\psi^2(\vec{r})]$, and $\langle T(\vec{r})T(0)\rangle$ is predicted to also decay as $1/r^{\eta_2}$ at criticality. By finite-size scaling, this implies that $\mathcal{N}(L) \equiv \langle[\sum_{\vec{r}} T(\vec{r})]^2\rangle/L^2$ is expected to have the scaling form $L^{2-\eta_2} f_{\mathcal{N}}(\delta L^{1/\nu})$ in the vicinity of the hard-square transition.

To test these predictions, we have performed Monte Carlo simulations of Z_{dsv} on $L \times L$ periodic lattices (with L up to 1024) using a variation [57] of an algorithm [58] that generates, in a single move, an equilibrium configuration of an entire row (or column), given the configuration of the rest of the system. Our method does not suffer from jamming even at full packing, and can be generalized to a large class of similar problems. More details are provided in the Supplemental Material [57]. For $w > w_c^{(0)} \approx 0.198(2)$ along DS, we find clear evidence of a $v = 0$ power-law ordered phase, in which $\mathcal{C}(L)/L^2$ decays as $1/L^{\eta(w)}$, while $\mathcal{R}(L)/L^2$ and $\mathcal{I}(L)/L^2$ both decay as $1/L^{4\eta(w)}$, with $\eta(w_c^{(0)}) = 1/4$ (Fig. 2). For the hard-square lattice gas, we estimate that the transition point is located at $v_c^* = 0.3180(3)$. Our data for $\mathcal{C}(L)$ is well fit by $\eta = 1/4$, and $\nu^* \approx 0.92(3)$, consistent with some of the earlier studies [16,17], while $\mathcal{N}(L)$ diverges as $L^{2-\eta_2^*}$ at criticality, with $\eta_2^* \approx 0.46(3)$ (Fig. 3), consistent with the Ashkin-Teller relation, providing conclusive evidence of the AT nature of the hard-square transition, and emphasizing that the hard-square transition lies beyond the decoupled Ising point [Fig. 1(b)] on the AT phase boundary. Additionally, at an intermediate point [Fig. 1(b)] on the phase boundary, our data for $\mathcal{C}(L)$ is fit well by $\eta = 1/4$ and $\nu \approx 1.70(5)$, while $\mathcal{R}(L)$ grows as $L^{2-\eta_2}$ at criticality, with $\eta_2 \approx 0.70(5)$ (Fig. 4), consistent with the Ashkin-Teller relation. This provides the first test of this relation in a microscopic lattice model with continuously varying exponents.

Outlook.—Given that columnar ordering is ubiquitous in a wide variety of strongly correlated systems [59–64], the ideas discussed here are of immediate relevance in a variety of other contexts. For instance, the emergent U(1) symmetry at full packing is closely related to the U(1) symmetry that is expected to emerge in the zero temperature limit [65,66] of the thermal AT transition [59] to columnar valence-bond solid (VBS) order in a class of frustrated square-lattice antiferromagnets that have been

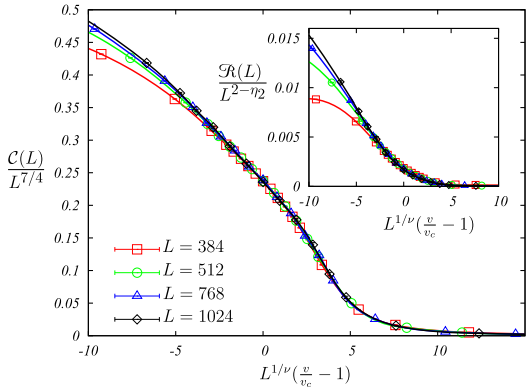

FIG. 4 (color online). Scaling collapse of $\mathcal{C}(L)/L^{7/4}$ for various L along a cut that crosses the AT boundary at an intermediate point $w_c = 0.1600(1)$, $v_c = 0.0623(1)$ yields the estimate $\nu = 1.70(5)$. Inset: Scaling collapse of $\mathcal{R}(L)/L^{2-\eta_2}$ yields the estimate $\eta_2 \approx 0.70(5)$.

the focus of many recent studies [67–77]. The ideas developed here predict that this emergent U(1) symmetry constrains the behavior of certain subdominant orders at this "deconfined" quantum critical point [65,66]. More precisely, with $\psi(\vec{r})$ now representing the complex VBS order parameter, we predict that correlations of $\mathrm{Re}[\psi^2(\vec{r})]$, the valence-bond nematic order parameter, decay with power-law exponent η_{VBN} that equals the power-law decay exponent for correlations of $\mathrm{Im}[\psi^2(\vec{r})]$, the wave vector (π,π) component of the next-nearest-neighbor bond energy, at this quantum critical point. Additionally, we predict that η_{VBN} and ν, the correlation length exponent for VBS order parameter correlations, are related all along the AT phase boundary via the Ashkin-Teller relation discussed here.

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