Liquid antiferromagnets in two dimensions

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It is shown that, for proper symmetry of the parent lattice, antiferromagnetic order can survive in two-dimensional liquid crystals and even isotropic liquids of pointlike particles, in contradiction to what common sense might suggest. We discuss the requirements for antiferromagnetic order in the absence of translational and/or orientational lattice order. One example is the honeycomb lattice, which upon melting can form a liquid crystal with quasi-long-range orientational and antiferromagnetic order but short-range translational order. The critical properties of such systems are discussed. Finally, we draw conjectures for the three-dimensional case.

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Ferrofluids, i.e., suspensions of small ferromagnetic particles in a carrier liquid, have been studied quite extensively [1]. These materials are really liquid superparamagnets without long-range magnetic order in the absence of an applied magnetic field. However, there is no fundamental reason why true ferromagnetism should not exist in a liquid. The strong, short-range exchange interactions are not strongly affected by the absence of crystalline order, as shown by the existence of amorphous ferromagnets [2].

The present paper addresses the question of whether *antiferromagnetic liquids*, which one could call "antiferrofluids," are also possible. On first sight, the answer seems to be no. Common sense tells us that the huge frustration in a liquid destroys antiferromagnetic order. To construct an antiferromagnetic liquid one would thus look for liquids that partially retain structural order, i.e., liquid crystals. In fact, antiferroelectric liquid crystals have been studied extensively [3]. These materials consist of long, polar molecules so that antiferroelectric order appears rather naturally in their smectic phases.

The question we want to discuss here is whether liquids (including liquid crystals) consisting of *spherical* particles with a spin degree of freedom can sustain antiferromagnetic order. At least in two dimensions this is possible, as we show below. We consider two-dimensional (2D) systems, since in two dimensions the theory of melting is much further developed than in three. The relevance for three-dimensional systems is briefly discussed afterwards. We introduce spin anisotropy to obtain a finite-temperature phase transition. Specifically, we think of the antiferromagnetic order parameter having either *XY* or Ising symmetry. In the first case there is a Berezinkii-Kosterlitz-Thouless (BKT) transition [4] and the low-temperature phase has quasi-long-range order. In the second case there is an Ising-type transition [5] to a long-range-ordered phase.

Our arguments employ the theory of 2D melting developed by Nelson, Halperin, and Young [6], which is based on the BKT renormalization group theory [4,7]. We first briefly review this theory. Then we discuss melting of a lattice with antiferromagnetic order for the normal case that antiferromagnetism is strongly frustrated by melting [8]. This sets the stage for the discussion of the possibility of antiferromagnetism in the liquid crystal formed upon melting. Surprisingly, for certain lattices melting can even produce an *isotropic* liquid that retains antiferromagnetic order.

The Nelson-Halperin-Young theory [6,9] predicts two distinct melting transitions. The one at the lower temperature separates a 2D solid with quasi-long-range translational order from a liquid crystal with short-range translational but quasi-long-range orientational order [10]. This transition is due to the unbinding of pairs of dislocations. Dislocations are pointlike in 2D and can be thermally created in pairs or multiplets of vanishing total Burgers vector. Pairs of dislocations with opposite Burgers vector have an attractive logarithmic interaction, similar to vortex-antivortex pairs in the 2D *XY* model. The resulting BKT-type transition is characterized by a jump of Young's modulus (the stiffness against tension), which is finite and universal just below the transition and zero above. In the liquid-crystal phase bound pairs of *disclinations* exist, which are defects of the orientational order. This order is destroyed at a higher transition temperature where disclination pairs unbind. Since their interaction is logarithmic in the presence of free dislocations, the transition is also of BKT type. Note that one or both transitions *may* be replaced by a first-order transition.

What happens if the particles carry a spin with a tendency to order antiferromagnetically? We restrict ourselves to bipartite lattices. Then the spins show Néel order in the classical ground state, if frustrating longer-range interactions are not too strong. For most simple lattices such as the square lattice elementary dislocations [11] frustrate the magnetic order, as illustrated by Fig. 1. There is a line of maximally frustrated bonds ending at the dislocation. This line could end at another dislocation of opposite Burgers vector. The energy of such a pair is *linear* in their separation and the pair is confined. This is indeed the case for Ising spins [12]. On the other hand, for two-component (*XY*) spins Fig. 1 does not show the lowest-energy configuration. Rather, the spins relax to spread the frustration more evenly. In effect, the dislocation dresses with *half a vortex* (or antivortex) in the Néel order [8]. The dislocation interaction is now again logarithmic, but with a contribution from the half vortices. The interplay of dislocation-unbinding and magnetic transitions in this case has been studied in Ref. [8]. It is obvious that magnetic order cannot survive the dislocation unbinding,

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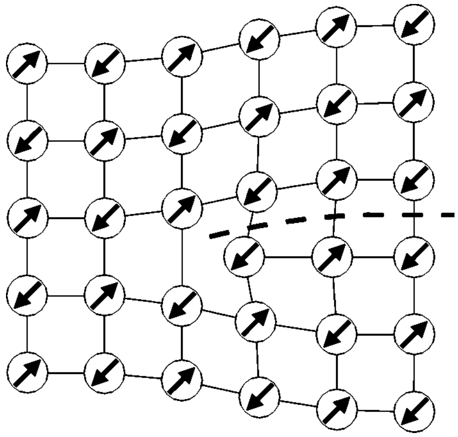

FIG. 1. Square lattice antiferromagnet with an elementary dislocation. The magnetic order is maximally frustrated along the heavy dashed line. For XY spins this configuration is unfavorable and the spins will relax to spread out the frustration [8]. As the result, the dislocation dresses with half a vortex.

since free dislocations carry (fractional) vorticity and act like free vortices [4,8]. Of course, the magnetic transition *may* take place at a *lower* temperature than the dislocation unbinding.

However, antiferromagnetism need not be destroyed at the lower melting temperature, if dislocations do not frustrate the magnetic order. One example is the honeycomb lattice. An elementary dislocation [11] does not frustrate the antiferromagnet, as shown in Fig. 2. Since all possible dislocations are superposition of elementary ones, none of them frustrates the order. Consequently, free dislocations above the lower melting temperature do not carry vorticity and thus the existence of free dislocations does not preclude antiferromagnetic (long-range or quasi-long-range) order [13].

When do dislocations not frustrate the magnetic order? This is the case if their Burgers vectors connect two sites with the same spin direction, i.e., on the same sublattice. The Burgers vector can be any lattice vector of the lattice without spins. Hence, all dislocations do not frustrate if any translation by a lattice vector leaves the spins invariant. Or, in other words, if magnetic ordering does not reduce the set of translational symmetry operations of the lattice. This is the case

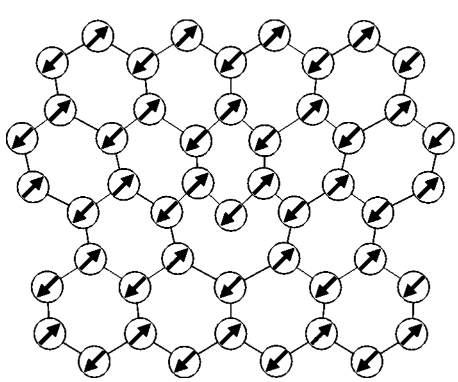

FIG. 2. Honeycomb lattice antiferromagnet with an elementary dislocation. Evidently the defect does not frustrate the magnetic order and, consequently, does not dress with fractional vorticity.

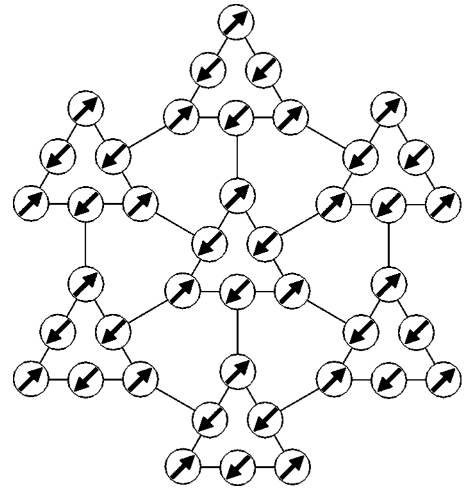

FIG. 3. A more complicated lattice. Here neither dislocations nor disclinations frustrate the magnetic order.

for the honeycomb lattice, which already has a two-site basis. On the other hand, for the square lattice the order reduces the set of translations and dislocations exist that frustrate the magnetic order.

We now turn to the upper, disclination-unbinding transition. For the honeycomb lattice, disclinations are characterized by the angle modulo 2π by which the bond angle changes if one goes around the defect [9,14]. The elementary disclinations [11] of the honeycomb lattice and the corresponding liquid crystal are $\pm 2\pi/6$ disclinations centered at a hexagonal plaquette. Thus, the defects have a five- or seven-sided plaquette at their core, which obviously frustrates the magnetic order. Furthermore, there are paths of arbitrarily large length around the defect that consist of an odd number of bonds. For the XY model, the spins again relax to reduce the energy and the disclinations dress with half vortices. Consequently, the magnetic transition temperature cannot lie above the disclination-unbinding temperature.

The next question is whether there are lattices for which neither dislocations nor disclinations frustrate the magnetic order. The lattice in Fig. 3 satisfies the criterion for nonfrustrating dislocations. Furthermore, elementary disclinations with a change of the bond angle by $\pm 2\pi/3$ do not frustrate either, as illustrated by Fig. 4. If the appearance of magnetic order does not reduce the *orientational* symmetry, i.e., does not remove rotation axes or reduce their multiplicity, all disclinations are compatible with antiferromagnetic order. In this case antiferromagnetic order *can* exist in the isotropic liquid above the upper melting transition. There is another way to express the condition for the existence of non-frustrating dislocations and disclinations for bipartite lattices: Magnetic order in the isotropic liquid is possible if the corresponding lattice does have *two nonequivalent sublattices*, i.e., one cannot be mapped onto the other by any translation or rotation or combination thereof. Then antiferromagnetic ordering does not reduce the lattice symmetry. At higher temperatures the liquid should eventually loose the hidden order that is expressed by the nonequivalence of two

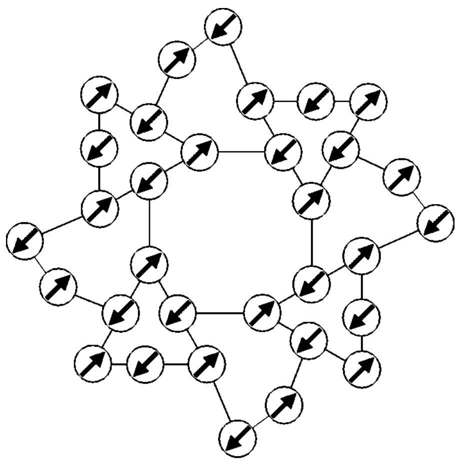

FIG. 4. The core of a $+2\pi/3$ disclination for the lattice shown in Fig. 3. The defect does not frustrate the magnetic order.

subsystems. Note, however, that this cannot happen through disclination unbinding, but will probably take place at a first-order transition.

Even if dislocations (or disclinations) do not dress with vorticity, their energies depend on the magnetic order, since part of the interaction is of magnetic origin. Conversely, due to frustration of the magnetic interaction at larger distances structural order affects the vortex energies. We now argue why this subdominant coupling leaves the principal picture unchanged, focusing on dislocations and vortices. The interaction energy of dislocations is proportional to Young's modulus, which we expect to be a continuous function of the vortex density. Since the vortex density itself is a continuous function of temperature through the vortex-unbinding transition [4,15], the parameters entering in the BKT theory of dislocation unbinding are continuous through the magnetic transition. A similar argument can be made for the change of the vortex energy due to dislocations. If one tunes the strength of magnetic vs nonmagnetic interactions, the dislocation-unbinding and vortex-unbinding transitions thus cross in a tetracritical point and both the structural and the magnetic order show a universal BKT jump at this point. This is drastically different from the normal case of, e.g., the square lattice, where for strong magnetic interactions the two transitions merge into a single one, at which only one order parameters shows a *universal* jump [8].

Next, we briefly commend on the low-energy collective excitations of liquids with antiferromagnetic quasi-long-range order. First, there is the usual longitudinal acoustic phonon branch. The liquid crystal phases differ from the isotropic liquid in that they have massive topological excitations, i.e., the disclinations. In addition, for XY spins there is a linearly dispersing spin wave mode in all liquid phases. Its presence is one main characteristic of an antiferromagnetic liquid. This mode leads to the characteristic behavior of the magnetic susceptibility of an antiferromagnet [16].

How can these considerations be applied to three-dimensional systems? In three dimensions melting typically proceeds by a first-order transition directly to the isotropic liquid. Nevertheless a dislocation-unbinding mechanism may apply [14,17–19]. To obtain an isotropic liquid, disclinations also have to unbind [14]. They usually do so at the same temperature, but this does not invalidate our criterion for antiferromagnetic fluids. Note also that our arguments never took advantage of the two dimensionality. Thus it may be inferred that also in three dimensions antiferromagnetic liquids can exist if the underlying lattice has *two inequivalent sublattices*.

Finally, we turn to possible experimental realizations. A soft 2D XY antiferromagnet is the Skyrmion crystal in the quantum Hall system close to filling factor $\nu=1$ [8,20–22]. Skyrmions are topological excitations of the ferromagnetic quantum Hall state, which carry a quantized electric charge. Upon changing the filling factor away from $\nu=1$, the extra charge appears in the form of Skyrmions. The in-plane magnetization of a Skyrmion has a vortexlike structure. Its direction can be characterized by a single XY angle θ, which couples antiferromagnetically [23,24]. The classical ground states of the Skyrmion system are various lattice types [8,25]. One is a honeycomb lattice, albeit probably outside of the realistic parameter range.

A straightforward realization of an Ising pseudospin model is a binary alloy (in 2D or three dimensional 3D). Another example is a system of vortices and antivortices, which are prevented from annihilating, e.g., by an additional Coulomb repulsion. The vorticity then constitutes the Ising degree of freedom. It has been suggested that such a vortex system is formed when holes are doped into the antiferromagnetic cuprates [26,27]. These charged vortices might form a strongly anisotropic (stripe) crystal at low temperatures [28,29,27]. It should be interesting to apply the ideas of the present paper to its melting [29].

To conclude, we have shown that there is no fundamental reason why 2D, and possibly 3D, antiferromagnetic liquids should *not* exist. Their existence is determined by the structure of the underlying lattice: If dislocations do not frustrate the antiferromagnetic order, antiferromagnetic liquid-crystal phases are possible. One example is the honeycomb lattice. If, in addition, disclinations also do not frustrate the magnetic order, it can even survive in isotropic liquids. The crystal phase must have two inequivalent sublattices for this to be possible. The resulting ''antiferrofluids'' would support spin waves with linear dispersion besides longitudinal phonons. It would be worthwhile to search for experimental realizations of this new phase of matter.

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