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Novel magnetic structure of a vortex lattice in adsorbed ^{3}He films

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Stimulated by a recent finding of a sharp specific-heat anomaly at 2.5 mK in a second layer of ^{3}He adsorbed on Grafoil by Greywall *et al.*, we studied a lattice-gas model or Hubbard model on a triangular lattice. We found a novel type of magnetic ordering of a vortex lattice, which is noncollinear with triple **Q** wave vectors and holds threefold symmetry, for nearly half-fillings. The size of the unit cell varies with ^{3}He coverages. This is a generic periodic pattern for neatly accommodating defects always present in an experiment.

A mysterious phase transition signaled by a sharp specific-heat peak around $T=2.5$ mK in ^{3}He films adsorbed on Grafoil was found by Greywall and Busch[1] and aroused much attention.[2] The experiment was done at surface coverages of $\rho=0.160$, 0.178, 0.184 atoms/Å^2 above the first layer ($\rho\sim0.114$), but before third-layer promotion ($\rho\sim0.24$). The transition entropy is only $(k_B/2)\ln 2$ per second-layer spin. Since Franco, Rapp, and Godfrin[3] already found a discernible negative deviation of the magnetization expected from the free-spin value in this coverage regime, this phenomenon is certainly related to some kind of nuclear magnetic ordering (possibly antiferromagnetic) of the second-layer ^{3}He atoms located on the first layer which itself forms a triangular lattice incommensurate with the underlying graphite.

To construct a model to understand this intriguing discovery, we summarize the important relevant facts: (i) A particular value of the second-layer density $\rho_2=\rho-\rho_1=0.064$ ($\rho_1=0.114$; first-layer density) used[1] fits nicely with the so-called $\sqrt{7}\times\sqrt{7}$ registry structure on the first layer, as noticed by Elser;[4] namely, that the relative density $\rho_2/\rho_1=0.56$ almost equals $\frac{4}{7}$, as expected. However, it is only approximate. In fact, the other density $\rho_2=0.07$ ($\rho_2/\rho_1=0.61$) exhibits the same anomaly, which is very insensitive to areal density, as emphasized by the authors.[1] (ii) Van Sciver and Vilches[5] found evidence for solidification of the second layer ($\rho_2=0.072$) in their specific-heat experiment. Abraham *et al.*[6] found a registry solid phase of the low-density second layer by a numerical Monte Carlo simulation. On the other hand, a neutron experiment[7] on $\rho=0.203$ failed to detect solidification of the second layer. These facts indicate that, depending upon the density, different types of registry solid phases exist for very narrow ranges of coverage, which are incommensurate with the first-layer triangular lattice, such as 3×3, $\sqrt{7}\times\sqrt{7}$, $\sqrt{3}\times\sqrt{3}$, etc. as fundamental unit cells. These lower commensurate solid phases play a role as a reference system for nearby densities. For a given ρ_2 (say $\rho_2=0.064$) the system finds an appropriate registry phase (say $\sqrt{7}\times\sqrt{7}$) which forms a triangular lattice. Since the density ρ_2 is rather randomly chosen in an actual experimental situation, the ^{3}He atoms would never be precisely accommodated in an appropriately selected registry solid phase. There generally exist excess or deficit atoms in this registry phase. This situation coincides with a suggestion by Greywall and Busch[1] that the heat-capacity anomaly is due to antiferromagnetic polarons formed around zero-point vacancies. This is to be contrasted with the situation envisaged by Elser[4] and Roger,[8] who take up a localized spin model on a triangular lattice where no vacancies are present.

As for the magnetic interactions between the ^{3}He atoms, Elser[4] considers two-body Heisenberg interaction while Roger[8] stresses three or more (4,6,...) particle ring exchange processes to investigate this phase transition of a frustrated antiferromagnetic system in two dimensions. In view of the recent progress of understanding this exchange process in the ^{3}He bulk solid due to Ceperley and Jacucci,[9] where higher-order cyclic exchanges make more significant contributions than previously thought, it is worth examining this two-dimensional magnetic phenomenon from the opposite viewpoint, or the itinerant picture rather than the localized one.

We study here a lattice-gas model or Hubbard model at $T=0$ on a triangular lattice[10] (the lattice constant is unity): $H=-t\sum_{\langle ij\rangle\sigma}c_{i\sigma}^{\dagger}c_{j\sigma}+U\sum_i n_{i\uparrow}n_{i\downarrow}$, where $t(=1)$ is the hopping integral between the nearest-neighbor sites and U is the on-site repulsion due to the hard core. We confine our discussion to coverages well before the second layer is completed, beyond which the situation differs due to the presence of the third layer.[11] This may mediate other types of exchange processes, such as the Ruderman-Kittel-Kasuya-Yosida interaction between the second-layer ^{3}He atoms. We ignore the first layer and consider only the second layer of ^{3}He atoms. If the number of ^{3}He atoms is equal to the number of lattice sites of a registered triangular lattice, it corresponds to "half-filling." A deviation from that atom number inevitably introduces a few excess or deficit atoms, namely defects. This corresponds to the "nearly half-filling case" of the Hubbard model,[12] which is the case of general experimental situations mentioned above. Since to our knowledge there is no systematic study for this problem, we take here the simplest approach, a mean-field approximation under the assump-

tion that there is long-range order in the ground state. In fact, as stressed by Friedman *et al.*[13] the dipole interaction, although its coupling energy is extremely weak ($\sim 0.1\ \mu$K), guarantees its existence and keeps the nuclear spins confined in the plane of the substrate. [We choose the spin components (S^z, S^x) and ignore the third component.] The mean-field Hamiltonian is

$$H = -t\sum_{\langle ij\rangle\sigma} c_{i\sigma}^{\dagger}c_{j\sigma} + \frac{U}{2}\sum_i [(n_i - S_i^z)n_{i\uparrow} + (n_i + S_i^z)n_{i\downarrow} - S_i^x(c_{i\uparrow}^{\dagger}c_{i\downarrow} + \text{H.c.})] - \frac{U}{4}\sum_i [n_i^2 - (S_i^z)^2 - (S_i^x)^2], \quad (1)$$

where the order parameters, the moments S_i^z, S_i^x, and atom density n_i depend on the site i to allow for spatial variation ($\mu_B = 1$). These are determined self-consistently by $S_i^z = \langle n_{i\uparrow}\rangle - \langle n_{i\downarrow}\rangle$, $S_i^x = \langle c_{i\uparrow}^{\dagger}c_{i\downarrow}\rangle + \langle c_{i\downarrow}^{\dagger}c_{i\uparrow}\rangle$, and $n_i = \langle n_{i\uparrow}\rangle + \langle n_{i\downarrow}\rangle$.

Our basic strategy is to seek a stable spin and density configuration by increasing the unit-cell size of a periodic pattern under a given filling. To do this, we numerically solve (1) in a self-consistent way, starting with an appropriate initial configuration given by the set $\{S_i^z, S_i^x, n_i\}$ ($i = 1, \ldots, N$; the total number of sites is N) under periodic boundary conditions. For larger unit-cell cases we restrict ourselves to the case that the system holds threefold symmetry; namely the pattern is characterized by the triple-**Q** wave vector whose unit cell forms a triangular lattice, such as $\sqrt{3}\times\sqrt{3}$, 2×2, $\sqrt{7}\times\sqrt{7}$, $3\times 3, \ldots$ sublattices in order of size. As for the initial condition, we consider both the collinear and noncollinear spin patterns which are constructed by the fundamental triple-**Q** wave vectors. We note, however, that the $\sqrt{3}\times\sqrt{3}$ sublattice pattern is not characterized by the triple-**Q** but single-**Q** vector because the three vectors with $|\mathbf{Q}| = 4\pi/3$ are equivalent.

Let us first examine the half-filling case where the atom number per site $n = 1$. There are several possible magnetic phases: (i) antiferromagnetic phase (AF) in which the ferromagnetically aligned lines are alternated along one of the triangular axes, (ii) ferromagnetic phase (FM), (iii) ferrimagnetic phase (FR) where two of the three sublattice moments point in the same direction, but opposite to the remaining moment, and (iv) and 120° structure (V_2) in which the three spins on a unit triangle take three distinctive directions, which differ by 120° from each other (see the inset of Fig. 1). The first three are collinear while the last is noncollinear.

It is rather a trivial task to examine the relative stability among these states because the number of the order parameters to be determined are quite small: It is found that V_2 is the lowest among them for a wide range of U ($U \gtrsim 5.5$; note that the bandwidth is nine in our unit). This conclusion is also confirmed by the usual momentum-space diagonalization method. For example, in $U = 10.0$ the relative energies per site are -2.9231 (V_2), -2.8925 (FR), -2.8817 (AF), and -2.4997 (FM). We mention that a few other types of the collinear phase with a larger unit cell are also found but they have higher energy than the AF. Since V_2 belongs to a three-sublattice system the eigenvalues ω are determined by

$$\omega^6 - (6|\epsilon|^2 + \tfrac{3}{4}m^2)\omega^4 - 2(\epsilon^3 + \epsilon^{*3})\omega^3 + (9|\epsilon|^4 + \tfrac{9}{4}m^2|\epsilon|^2 + \tfrac{3}{16}m^4)\omega^2 + [6(\epsilon\epsilon^{*4} + \epsilon^4\epsilon^*) + \tfrac{3}{4}m^2(\epsilon^3 + \epsilon^{*3})]\omega + 2|\epsilon|^6 + \epsilon^6 + \epsilon^{*6} - \tfrac{3}{16}|\epsilon|^2 m^4 - (m^6/64) = 0, \quad (2)$$

with $\epsilon = e^{ik_x} + 2e^{-i(k_x/2)}\cos(\sqrt{3}/2)k_y$ and $m = (U/2)S^z$. As shown in Fig. 1, the six bands group together into two separate bands for $m \gtrsim 2$, making V_2 energetically favorable for $n = 1$ but definitely not for $n \neq 1$. The conclusion that in the exactly half-filling case V_2 is the lowest for $U > U_{cr} \sim 5.5$, below which the two groups of the bands become overlapped, destabilizing V_2, coincides with the classical localized spin (Heisenberg) picture as a strong-coupling limit of the Hubbard model where the intrinsic frustration due to the nonbipartite triangular lattice leads to V_2, but this coincidence may be accidental. The obtained solution of V_2 is stable against local-density fluctuations, and thus n_i is uniform. The moment is almost saturated when U is large, and the chirality κ defined by $\mathbf{S}_i\times\mathbf{S}_j + \mathbf{S}_j\times\mathbf{S}_k + \mathbf{S}_k\times\mathbf{S}_i$ on a unit triangle is completely ordered.

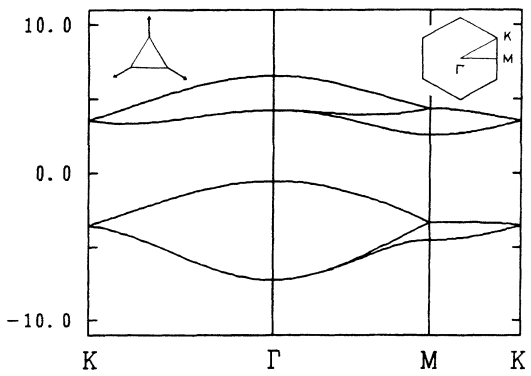

FIG. 1. Dispersion relation for the V_2 phase calculated by (2) along the symmetry lines in reciprocal space shown in the inset ($U = 8.0$). The other inset indicates the V_2 phase.

Having seen that the noncollinear V_2 is stable over the other collinear states in $n = 1$, we seek a generic magnetic structure with larger unit-cell size for the nearly half-filling cases where defects should be neatly accommodated. We display a typical example of the stable magnetic ordering periodic pattern we found in Fig. 2(a) and associated density profile in Fig. 2(b) for $N = 9\times 9$ with the number of atoms 93, i.e., $n = \frac{93}{81} \sim 1.148$ when $U = 8.0$. It is seen that (i), as shown in Fig. 2(c), there exist two kinds of three vortices contained in a unit cell consisting of 27 sublattices; one is characterized by the vorticity (or winding number) $+1$ situated at the center and three corners of the hexagon while the other is -2. The total vorticity vanishes. (ii) The gross feature of this vortex-lattice structure (VLS) can be constructed by superposing three

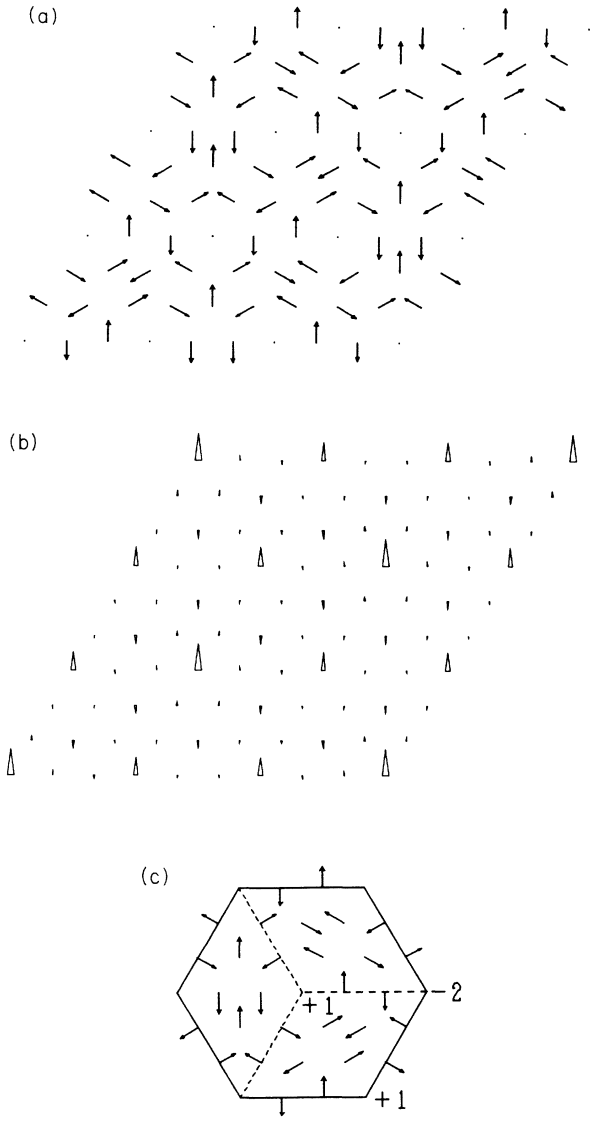

FIG. 2. (a) Result for the spin configuration with the sublattice size $\sqrt{27}\times\sqrt{27}$ where the system size $N=9\times9$, $U=8.0$, and $n=\frac{93}{81}$. The maximum moment per site is 0.8239. (b) Associated density profile for the same $N=9\times9$ system. The size of a triangle is proportional to the deviation from the average density. The large (small) triangles pointing up (down) denote excess (deficit) density. The maximum density deviation from the average one is 0.2461 situated at a -2 vorticity site, while 0.1710 is at a $+1$ vorticity site. (c) Unit cell of the above vortex lattice. The $+1$ vortices are at the center and three corners and -2 vortices at the other corners of the hexagon. The total moment vanishes in each subdivision indicated by the dotted lines.

plane waves with the triple-**Q** wave vectors whose directions are 120° apart and whose magnitudes $|\mathbf{Q}|=8\pi/9$ in this filling, making the unit cell $\sqrt{27}\times\sqrt{27}$ sublattices. (This is indeed an input configuration in our computation). (iii) All the moments on each site are almost equal except for the vortex sites where the moment vanishes and an excess (or deficit) density exclusively accumulates, as seen from Fig. 2(b), forming a localized object or a triple-**Q** soliton lattice.[14] This implies that higher harmonics play an important role. (iv) We subdivide the unit cell into three sections so as to preserve the threefold symmetry, as shown by the dotted lines in Fig. 2(c). In each subsection the total moment added up vanishes. Thus there is no net ferromagnetic component in the system. (v) The spin configuration on a unit triangle far from a vortex tends to form the V_2 configuration. (vi) At the vortex sites the nearest-neighbor molecular field is completely canceled out. (vii) Depending upon the vorticity, accommodated defects are different.

We have checked that at other fillings near $n=1$ we can always find a stable VLS with similar topological features, differing by the size of the pattern, provided that the combination of the system size N and filling n is properly chosen so as to fit the unit cell of a VLS with the system size restricted under periodic boundary conditions.

The VLS which are featured by the triple-**Q** wave vectors and noncollinear spin arrangement is quite "generic" as a stable magnetic pattern near the half-filling when the spin direction is confined in the substrate plane by the dipole interaction: The local spin arrangement tends to refer to V_2 which is most stable at the half-filling. As the deviation from $n=1$ becomes small or the number of defects decreases, the unit cell of the VLS becomes large by tuning the fundamental wave vector $|\mathbf{Q}|$ to accommodate defects. Thus VLS is a global V_2 phase and is characterized by a soliton lattice structure with the triple-**Q** vectors whose isolated nodal points accommodate defects.

In conclusion, the second ^{3}He layer is found to be an interesting testing ground for the study of a two-dimensional spin-$\frac{1}{2}$ system on a frustrated triangular lattice with defects whose number and interaction strength (U/t) can be controlled by varying coverages. We have found a type of periodic ordering pattern of the vortex-lattice structure for accommodating defects which normally exist in an actual experimental situation involving adsorbed ^{3}He films on Grafoil. The pattern is generic in the sense that the topological feature is insensitive to the number of defects. We urge further experimental investigation into the nature of the "2.5-mK anomaly" in order to examine our proposal. Magnetic neutron scattering is best suited for this purpose if feasible, but other indirect methods such as NMR are interesting. This study might help with the understanding of the magnetism in itinerant electron systems such as fcc three-dimensional transition-metal alloys,[15] where because of the nonbipartite crystalline structure there appear various noncollinear types of magnetic patterns with triple-**Q** wave vectors. The physical origin for this largely remains unexplored.

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