Coexistence of Canted Antiferromagnetism and Bond Order in $\nu = 0$ Graphene

Ankur Das,[1,2,*] Ribhu K. Kaul,[2] and Ganpathy Murthy[2]
[1]Department of Condensed Matter Physics, Weizmann Institute of Science, Rehovot 76100, Israel
[2]Department of Physics and Astronomy, University of Kentucky, Lexington, Kentucky 40506, USA

(Received 22 September 2021; revised 12 December 2021; accepted 14 February 2022; published 11 March 2022)

Motivated by experimental studies of graphene in the quantum Hall regime, we revisit the phase diagram of a single sheet of graphene at charge neutrality. Because of spin and valley degeneracies, interactions play a crucial role in determining the nature of the ground state. We show that, generically within the Hartree-Fock approximation, in the regime of interest there is a region of coexistence between magnetic and bond orders in the phase diagram. We demonstrate this result both in continuum and lattice models, and argue that the coexistence phase naturally provides a possible explanation for unreconciled experimental observations on the quantum Hall effect in graphene.

DOI: 10.1103/PhysRevLett.128.106803

Introduction.—The quantum Hall effect is a fundamental manifestation of topology, quantum mechanics, and many-particle physics in two dimensions [1,2]. Discovered originally in semiconductor heterostructures, it found a new realization in graphene two decades later [3–6]. Graphene brings several tantalizing twists to the original quantum Hall problems that arise due to its honeycomb lattice [6–9]. At low energies graphene has a relativistic linear dispersion leading to an effective Dirac equation near charge neutrality, which leads to a distinct Landau spectrum [10,11]. Additionally, there are two copies of the Landau levels due to valley degeneracy, causing electron-electron interactions to play a crucial role in selecting the ground state even for integer fillings [12–14].

Here we focus on the ground state at charge neutrality ($\nu = 0$), which corresponds to an electron count that fills precisely two of the four (almost) degenerate $n = 0$ Landau levels (LLs). We will call this manifold of states the zero-energy LLs (ZLLs). At the noninteracting level, the Zeeman energy splits the four degenerate $n = 0$ LLs into pairs of twofold degenerate ones, picking a fully polarized ground state [15]. Since the ZLLs have equal contributions from particlelike and holelike states, at the edge one linear combination of the valleys has a particlelike dispersion, while the orthogonal linear combination has a holelike dispersion. The edge of a fully polarized bulk state develops a pair of counterpropagating charged chiral modes protected by spin-rotation symmetry, manifesting the quantum spin Hall effect [15]. The addition of Coulomb interactions gaps the single-particle electron spectrum everywhere, but preserves the two gapless counterpropagating charge modes (protected by S_z conservation), promoting them into a helical Luttinger liquid [16].

From pioneering experiments [12,17–19], we now know that the ground state depends on the balance between the orbital magnetic field, $B_\perp$ (perpendicular to the graphene sheet) and the total field, B_{tot} (which enters via the Zeeman energy E_Z and can be tuned by applying an in-plane field). For E_Z less than a critical value E_Z^* all charge excitations in the bulk and the edges are completely gapped. However, for $E_Z > E_Z^*$, one obtains a gapped bulk with a two-terminal edge conductance of (almost) $2e^2/h$ [20], which is expected of the helical Luttinger liquid. While the nature of the phase for $E_Z < E_Z^*$ has not been conclusively identified in experiment, a continuous phase transition to it from the fully polarized state is observed [20]. Based on a Hartree-Fock (HF) treatment of a continuum model [21] which keeps only the ZLLs with ultra-short-range interactions [22,23], it is believed that the $E_Z < E_Z^*$ phase is a canted antiferromagnet [24]. While this proposal is consistent with recent magnon transmission experiments [25–27] that imply that the state is magnetic, it is in tension with STM studies [28–30] which find evidence for bond order in the $E_Z < E_Z^*$ insulating phase at $\nu = 0$. Note that in a sister material (Bernal-stacked bilayer graphene) bulk Goldstone modes of the CAF phase have recently been observed [31].

In this Letter we offer a possible resolution to this paradox. We propose that the seemingly contradictory observations arise from the coexistence of magnetism and bond order at charge neutrality, which was absent in previous theoretical phase diagrams. Our results are based on the Hartree-Fock approximation, which has been shown to be reliable for broken-symmetry states in the integer quantum Hall effect (IQHE) [1,2,32–39]. We show, both in the continuum and on the lattice, that coexistence is a generic feature in the regime of interest. In the continuum model, justified at weak $B_\perp$ relevant to experiment, we first show that a general HF analysis in the ZLLs depends only on six coupling constants that parametrize the electron-electron interactions. We then show that generic choices of these couplings lead to coexistence. In a complementary, more microscopic, HF analysis on the lattice in a magnetic

field with $1/q$ quanta of flux penetrating each unit cell, we find ubiquitous evidence for coexistence for small and moderate values of q up to 36. Careful extrapolation to large q of our numerical data demonstrates that the coexistence survives in the $B_\perp$ regime relevant to experiments (for reference, $B_\perp = 10$ T gives $q \simeq 10\,000$). Since coexistence is generically present in both limiting cases at the HF level, we argue that it can explain the experimental observations [20,28], especially since disorder, which pins the bond order, will only enhance its presence in the physical system. Very recent experiments have found evidence for the coexistence of bond order with other symmetry breaking [29,30].

A microscopic model for graphene in a magnetic field that is expected to harbor all the phenomena discussed takes the general form,

$$H_{\text{latt}} = -\sum_{\langle ij\rangle} t_{ij} c_{is}^\dagger c_{js} - E_Z \sum_{is} s c_{is}^\dagger c_{is} + H_{\text{int}}^{(4)}, \quad (1)$$

where c_{is} destroys an electron on the ith site of the honeycomb lattice with spin $s = \pm 1$. The Zeeman term, $E_Z = g\mu_B B_{\text{tot}}/2$ and the hopping $t_{ij} = te^{i\int_i^j \vec{A}.d\vec{l}}$ with $\vec{A}$ chosen so $\nabla \times \vec{A} = \hat{z}B_\perp$, together describe the free part of the Hamiltonian. The magnetic field introduces the length scale $\ell = \sqrt{(h/eB)}$, such that an area of $2\pi\ell^2$ is pierced by one flux quantum. Since for $B_\perp = 1T$, $\ell = 25$ nm, it is clear that $\ell \gg a$, where a is the lattice spacing. $H_{\text{int}}^{(4)}$ is a four-fermi electron-electron interaction whose precise form is unknown—we shall discuss specific forms for it below.

Continuum.—In this limit justified for $\ell \gg a$, one restricts attention to low-energy states near the K, K' points, linearizing the band structure to Dirac equations at each valley. Momentum conservation, when applied to two-body interactions, forces the conservation of particle number in the two valleys independently, leading to a U(1) symmetry in the valley space [21]. An orbital B field is introduced by minimal coupling into the Dirac equation, leading to four copies (spin and valley) of a relativistic Landau level spectrum. The interacting Hamiltonian projected into the ZLLs is

$$H_{\text{cont}} = -E_Z \sum_{\alpha,k,s} s c_{\alpha ks}^\dagger c_{\alpha ks} + \sum_{\mathbf{q}\mu} \frac{v_\mu(\mathbf{q}) :\rho_\mu(\mathbf{q})\rho_\mu(-\mathbf{q}):}{2L_x L_y}$$
$$\rho_\mu(\mathbf{q}) = \sum_{k,s,\alpha,\beta} e^{-(\frac{q^2}{4} + iq_x(k - \frac{q_y}{2}))\ell^2} c_{\alpha k - q_y s}^\dagger \tau_\mu^{\alpha\beta} c_{\beta ks}, \quad (2)$$

where $c_{\alpha ks}$ destroys an electron with spin s in valley α and y momentum k, and τ_μ are Pauli matrices in the valley space. We work in the Landau gauge $\vec{A} = (0, B_\perp x)$ on an $L_x \times L_y$ sample with periodic boundary conditions in y. Since the valley and sublattice indices are tied in the ZLLs, no sublattice index appears. The functions $v_\mu(\mathbf{q})$ are the Fourier transforms of the effective interactions (in the ZLLs) in the $\mu = 0$, x, y, and z valley channels (τ_0 is the unit matrix). The U(1) valley symmetry forces $v_x(\mathbf{q}) = v_y(\mathbf{q})$. The phase diagram of Eq. (2) can be calculated in the HF approximation with the averages $\langle c_{\alpha ks}^\dagger c_{\alpha' k' s'}\rangle = \delta_{kk'}\Delta_{\alpha\alpha'}^{ss'}$ preserves translation invariance up to an intervalley coherence. Intervalley coherence signifies incipient bond order, though to realize a bond-ordered state breaking lattice translation symmetries requires physics beyond the continuum model (as we explore below). Building on previous work [21–23] assuming ultra-short-range interactions in real space [$v_\mu(\mathbf{q}) \equiv v_\mu$ constant], Kharitonov [24] found a comprehensive HF phase diagram exhibiting four phases: canted antiferromagnetic [CAF, characterized by the order parameter $\text{Tr}(\tau_z\sigma_x\Delta) \neq 0$ and $\text{Tr}(\sigma_z\Delta) < 2$], fully polarized [F, characterized by $\text{Tr}(\sigma_z\Delta) = 2$], charge-density-wave [CDW, characterized by $\text{Tr}(\tau_z\Delta) \neq 0$], and bond ordered [BO, characterized by $\text{Tr}(\tau_x\Delta) \neq 0$]. There is no coexistence of order parameters in this model, and all transitions except for CAF to F are first order. Experimental graphene samples are believed to be in the CAF regime for purely perpendicular fields, which needs $v_x = v_y < 0$, and $v_z > |v_x|$. Kharitonov found in his model that $E_Z^* = |v_x|/\pi\ell^2$, leading to the conclusion that increasing E_Z while keeping $B_\perp$ fixed will eventually lead to a fully polarized bulk state for $E_Z > E_Z^*$ via a second-order phase transition, consistent with experiment [20].

We now show that relaxing the ultra-short-range assumption leads generically to coexistence between the canted antiferromagnet and bond-ordered states near their phase boundary in the ultra-short-range model. While the functions $v_\mu(\mathbf{q})$ have an infinite number of degrees of freedom, the ground state energy of any translation-invariant HF state depends only on six coupling constants; two specific numbers for each v_μ: The Hartree coupling $g_{\mu,H} = [v_\mu(\mathbf{0})/2\pi l^2]$ and the Fock coupling $g_{\mu,F} = \int [d\mathbf{q}/(2\pi)^2] v_\mu(\mathbf{q}) e^{-q^2 l^2/2}$. The assumption in previous work [24] that the interactions remain short range on the lattice scale $a \ll \ell$ even in the effective theory in the ZLLs forces $g_{\mu,H} = g_{\mu,F}$, and leads to the lack of coexistence in the phase diagram [24].

In the regime of coupling constants of interest in real graphene samples, where the ground states are CAF and/or BO, we find that three of the couplings $g_{0,H}, g_{0,F}, g_{z,H}$ play no role in selecting the ground state. We are left with just three independent couplings $g_{z,F}, g_{xy,H}, g_{xy,F}$. We assume an ansatz for the two occupied orbitals that interpolates between the CAF and the BO states [40].

$$|a\rangle = \frac{1}{\sqrt{2}}(c_a|K\uparrow\rangle - s_a|K\downarrow\rangle + c_a|K'\uparrow\rangle + s_a|K'\downarrow\rangle), \quad (3)$$

$$|b\rangle = \frac{1}{\sqrt{2}}(-c_b|K\uparrow\rangle + s_b|K\downarrow\rangle + c_b|K'\uparrow\rangle + s_b|K'\downarrow\rangle), \quad (4)$$

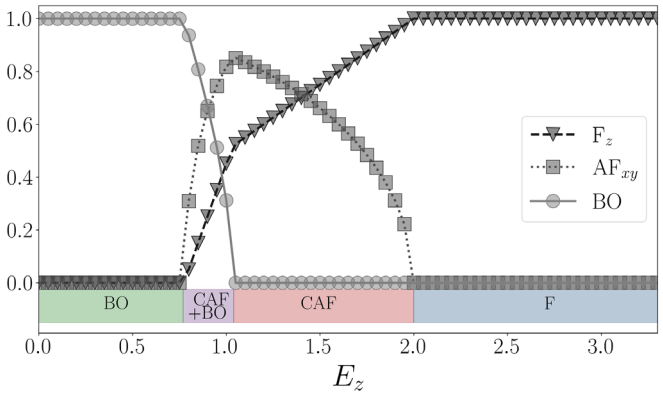

FIG. 1. Order parameters obtained from our generalized HF study of the continuum theory, Eq. (2) plotted as a function of the Zeeman energy, E_Z. We have chosen the interaction parameters $g_{z,F} = 0.1$, $g_{xy,H} = -0.75$, and $g_{xy,F} = -1$. The bar at the bottom shows the phase the system is in based on which orders have condensed. For $E_z = 0$ the system is in the BO (bond ordered) phase. For E_z very large the system is in the (F) ferromagnetic phase. Varying E_Z between these limits, the system goes through two intermediate phases, a canted antiferromagnet (CAF) without and with bond order coexistent (CAF + BO) (all three order parameters are non-zero). All the transitions are continuous in our HF theory.

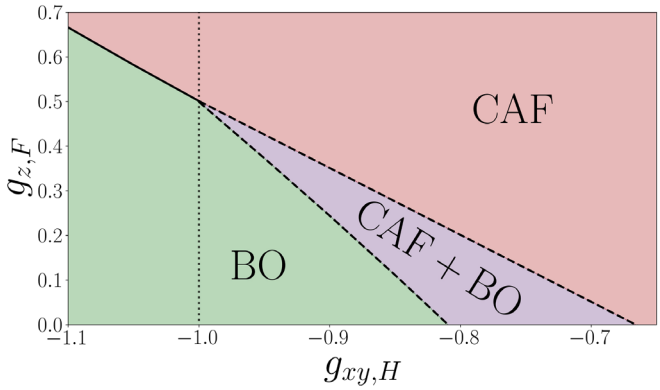

FIG. 2. A section of the HF phase diagram obtained from our continuum theory, Eq. (2). Coexistence between CAF and BO at fixed E_Z can be seen in a robust region. The plots are made for $g_{xy,F} = -1$, $E_z = 1.0$. Two necessary conditions for coexistence are $0 > g_{xy,H} > g_{xy,H}$ and $E_Z > 0$. The ultra-short-range result is the dotted vertical line at $g_{xy,H} = -1$.

where $c_\alpha = \cos(\psi_\alpha/2)$ and $s_\alpha = \sin(\psi_\alpha/2)$. The CAF state corresponds to $\psi_a = \psi_b = \theta$, the canting angle, and the BO state corresponds to $\psi_a = 0$, $\psi_b = \pi$. In a generic state, these two angles are independently minimized. We have verified that this ansatz correctly describes the states of interest by numerically carrying out iterative HF starting from random "seed" Δ matrices. We find two necessary conditions for coexistence: $|g_{xy,F}| > |g_{xy,H}|$ and $E_Z > 0$. Figure 1 shows the order parameters for the BO, CAF, and F states as a function of E_Z for a particular choice of our parameters. With this choice, the system starts in the BO phase at zero E_Z, undergoes a phase transition to a phase with coexistence between BO and CAF for intermediate E_Z, goes through another transition to a pure CAF phase, and finally to the F phase. All transitions are second order. Figure 2 is a section of the phase diagram at constant $g_{xy,F} = -1$, $E_Z = 1$, clearly showing that coexistence is absent with the usual ultra-short-range assumption $g_{xy,H} = g_{xy,F}$, but appears when $g_{xy,H} - g_{xy,F} > 0$. Evidently, $g_{xy,H} - g_{xy,F}$ determines the sign of the energy-energy coupling between the two order parameters [41] in a Landau theory of the phase transition.

In order for $g_{\mu,H}$ to be significantly different from $g_{\mu,F}$ one needs the relevant function $v_\mu(\mathbf{q})$ to vary on the scale of the magnetic length ℓ in real-space and be nonmonotonic. The Dirac-Landau quantization of energy levels, in combination with LL-mixing induced by the Coulomb interaction [42], naturally introduces this scale into the effective interactions. We show an explicit model calculation of this effect in the Supplemental Material [43].

Lattice.—One logical approach to determine the effective $g_{\mu,H(F)}$ interactions in the ZLLs is through a renormalization group calculation that starts from a microscopic model and then taking the LL-mixing into account [42], includes structure at all $\mathbf{q}$. Carrying out this explicitly is difficult and many simplifications have to be made: current RG treatments [24] are restricted to the ultra-short-range assumption (neglecting the $\mathbf{q}$ dependence), and do not treat the discreteness of the Dirac-Landau levels. To capture this crucial missing physics, we will proceed instead by carrying out a lattice HF calculation for an explicit microscopic model in the presence of an orbital flux (for the noninteracting limit, see, for example, Refs. [44,45]) per unit cell [46–53]. Since no projection to the low-energy manifold is performed, all LL-mixing effects are automatically included. Furthermore, lattice scale physics (C_3 symmetry, reciprocal lattice vectors, etc) that plays an important role in the bond order is kept fully, while it is absent in the continuum.

We use

$$H^{(4)}_{\text{int}} = \frac{U}{2}\sum_i (n_i)^2 - 2g\sum_{\langle ij\rangle} \vec{S}_i \cdot \vec{S}_j, \tag{5}$$

where $n_i = \sum_s c^\dagger_{is} c_{is}$ and $\vec{S}_i = \frac{1}{2}\sum_{s,s'} c^\dagger_{is}\vec{\sigma}_{ss'}c_{is'}$. The first term is the Hubbard interaction, and the second is a nearest-neighbor Heisenberg spin exchange. We treat this model in HF approximation allowing for translation symmetry breaking [43].

As expected, the phase diagram we find is much richer than that found in the continuum, with several different types of magnetic order and bond order making their appearance in different ranges of parameters. The full phase diagram appears in the Supplemental Material [43]. Here, we focus on the issue of interest, coexistence

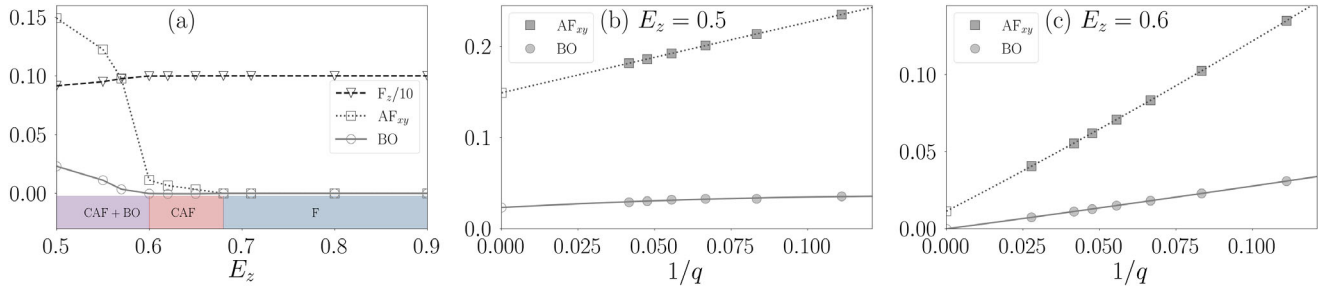

FIG. 3. Illustrative HF results for the lattice model defined by Eqs. (1) and (5). The numerical results are obtained on lattices with a flux of $1/q$ quanta per unit cell and then extrapolated to the weak field regime ($q \to \infty$). (a) The extrapolated order parameters for $g = 0.3$, $U = 3.5$ as a function of E_Z. Note that there are two phase transitions from zero to large Zeeman coupling, consistent with our continuum result. The phases are labeled in the bottom bar. (b),(c) Examples of the extrapolations of the AF_{xy} and BO order parameters to the $q \to \infty$ limit used to produce (a). For finite-q coexistence between magnetism and BO is ubiquitous, but the BO vanishes at intermediate Zeeman as $q \to \infty$, resulting in a pure CAF phase for $E_Z > 0.6$.

of BO and CAF orders. Computational resources limit us to a maximum q of 36, which corresponds to $B_\perp$ much larger than experimentally accessible fields. We circumvent this shortcoming by extrapolating our data to the large-q limit, which corresponds to experimentally realizable fields. The extrapolated order parameters are shown in Fig. 3(a) for a particular choice of couplings U, g. There are two distinct phase transitions at E_{Z1} and E_{Z2} as E_Z is increased. The other two panels show how the extrapolation is done for representative points in the CAF/BO coexistent phase (b), and for the pure CAF phase (c). For $E_Z < E_{Z1}$, as shown in Fig. 3(b), the order parameters of the CAF and BO both saturate to nonzero values in the limit $q \to \infty$. However, for $E_{Z1} < E_Z < E_{Z2}$ [Fig. 3(c)] the bond order vanishes in the continuum limit, while the CAF saturates to a nonzero value.

Strikingly, this is the same sequence of phases with increasing E_Z as in the continuum. Even though the two calculations approach the problem from opposite limits they converge on the same generic nature of the coexistence between CAF and BO. As anticipated, even though U and J are ultra-short-range interactions, the LL-mixing inherent in the full lattice calculation has succeeded in generating structure in the effective $v_\mu(\mathbf{q})$ on the scale of ℓ.

Discussion.—The coexistence of magnetism and bond order we propose can reconcile disparate experiments [26,28–30]. We theoretically find coexistence in two independent ways: First, we find that HF studies on a generic lattice model with short-range interactions Eq. (5) finds evidence for a coexistence phase even when extrapolated to the experimentally relevant weak field limit. Second, in a HF study of the symmetry-allowed continuum model restricted to the ZLLs, if the ultra-short-range assumption of previous studies [22–24] is relaxed, the same coexistence phase arises. One of the central ideas of our Letter is that even if the microscopic interactions (other than Coulomb) are short range on the lattice scale [such as Eq. (5)], the effective interactions generated by RG in the ZLLs will have nontrivial structure on the length scale ℓ. While such a computation is too complicated to implement, we substantiate this argument by an RG calculation on a toy model [43].

An important aspect of the experiment not in our study is disorder. We generically expect disorder to enhance bond order, though it will have other effects as well [54]. While bond order breaks translational invariance spontaneously, disorder breaks this symmetry explicitly, favoring the bond-ordered state over the translation-invariant CAF state. Thus, we can expect STM experiments to see bond order over a wider range of E_Z than we found theoretically. While technically, based on the mapping to a random-field Ising model [55–57], one may conclude that long-range bond order is destroyed by disorder, this clearly does not have implications for STM experiments, which measure the local strength of bond-order.

The HF calculations we have used here are based on the single Slater-determinant mean-field approximation. How much are these results affected by quantum and thermal fluctuations? At $T = 0$, based on past experience in the IQHE, deep within a phase, we expect quantum fluctuations to reduce the order parameter but not destroy it completely [24,32–35,37]. The nature of quantum phase transitions can depend profoundly on fluctuation effects; a study of these is exciting but beyond the scope of this work. For $T > 0$ any order parameter that breaks a discrete symmetry, such as CDW or BO, will continue to have long-range order up to some critical temperature. The CAF order parameter breaks a continuous symmetry at $T = 0$, and is forbidden from having long-range order for $T > 0$ by the Mermin-Wagner theorem [58]. Instead, it will have a phase with correlations decaying as a power law at low T, with a Kosterlitz-Thouless transition [59,60] to a phase with exponentially decaying correlations at a critical temperature.

In summary, we have presented a possible resolution to a seeming contradiction in the nature of the low-Zeeman charge-neutral state of graphene in the quantum Hall regime. By two complementary methods we find that

coexistence between CAF and BO orders is generic. From the theoretical side, the neighborhood of the phase transition between the CAF and the BO phases in $\nu = 0$ graphene is interesting, because it may host an approximate SO(5) symmetry [61,62] and field theories for this transition contain topological terms [63] which allow certain excitations in either phase to carry the quantum numbers of the other. These intriguing ideas provide further motivation for future experimental and theoretical work on bond order in $\nu = 0$ graphene.

The authors acknowledge supported from NSF DMR-1611161, German-Israeli Foundation (GIF) Grant No. I-1505-303.10/2019, Minerva Foundation, Dean of Faculty WIS, Israel planning and budgeting committee for financial support, and Koshland Foundation for Koshland Fellowship (A. D.), NSF DMR-2026947 (R. K. K.), and US-Israel Binational Science Foundation Grant No. 2016130 (G. M.). The authors thank Benjamin Sacépé, Alexis Coissard, Adolfo Grushin, and Cécile Repellin for stimulating conversations, and the Aspen Center for Physics (NSF Grant No. PHY-1607611) (R. K. K., G. M.) where this work was completed. We would also like to thank the University of Kentucky Center for Computational Sciences and Information Technology Services Research Computing for their support and use of the Lipscomb Compute Cluster and associated research computing resources.

*ankur.das@weizmann.ac.il

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