

Surface sensitivity of magnetization in the mesoscopic regime

Kevin Moseni¹ and Sinisa Coh^{1,2}

¹*Materials Science and Engineering, University of California Riverside, Riverside, CA 92521, USA*

²*Mechanical Engineering, University of California Riverside, Riverside, CA 92521, USA*

(Dated: December 11, 2023)

We find that in the mesoscopic regime modification of the material's surface can induce an extensive change of the material's magnetic moment. In other words, perturbation of order N^2 atoms on the surface of a 3-dimensional solid can change the magnetic moment proportionally to N^3 . When the solid's surface is perturbed, it triggers two changes in the magnetization. One arises from variations of the electron wavefunction and energy, while the other arises from a modification in the kinetic angular momentum operator. In the macroscopic regime of our model, these two bulk effects cancel each other, resulting in no impact of the surface perturbation on the magnetization — consistent with prior work. In the mesoscopic regime, we find a departure from this behavior, as the cancelation of two terms is not complete.

In a ferromagnet, the magnetic moment primarily arises from the unequal population of electrons with different spin states. A smaller, but significant contribution, known as orbital magnetization, originates from the microscopic spatial motion of electrons throughout the material. Some of these microscopic orbital electron currents flow around individual atoms in the bulk, while other currents traverse the surface of the sample, as demonstrated in Ref. 1 using a framework of localized Wannier states. Although only a fraction of electrons participate in surface currents, their collective effect contributes to the magnetic dipole moment, scaling with the volume of the sample (area in two dimensions).

The question then arises whether the magnetic moment of the ferromagnet could be modified by perturbing surface of the material? For instance, one may wonder if adsorbing atoms to the surface of a solid could induce currents and consequently change the magnetic dipole of the solid, in proportion to the volume of the solid? In other words, we are asking whether perturbing order N^2 atoms on the surface of a 3-dimensional solid could change the magnetic moment proportional to N^3 ? Or, similarly, whether perturbing order N atoms on the *edge* of a 2-dimensional solid could change the magnetic moment in proportion to N^2 ?

The seminal work from Ref. 1 demonstrated that none of these scenarios are possible for insulating systems. In an insulating system, the surface currents are quite remarkably determined by the material properties deep in the bulk of the material! Intuitively, one would expect such a statement to also extend to metallic cases. Reference 2 gives heuristic reasons why magnetization in a metal is equally well determined by the properties of the bulk of the material, as in the case of an insulator. (The same was also suggested for topological insulators in Refs. 2–4.) Additional support is given by the semi-classical formulation of orbital magnetization from Ref. 5 as well as the long-wave perturbation from Ref. 6. A more recent proof that orbital magnetization in a metal is a bulk property relies on a local measure of the orbital

moment from Refs. 7–9.

In this paper, our focus lies on a distinct range of length and temperature scales, one that complements the scope of previous investigations. Previous studies can be applied to the macroscopic regime, which we define as,

$$\frac{L}{v_F} \gg \frac{\hbar}{k_B T}. \quad (1)$$

Here v_F is the electron's Fermi velocity and L is a length of the sample. In other words, in the macroscopic regime, the electron's time of flight across the sample (L/v_F) exceeds the time scale associated with the thermal energy $k_B T$. In the macroscopic regime our findings corroborate the conclusions drawn in Refs. 1–9. Specifically, the surface modifications does not lead to extensive change in the magnetization.

Nevertheless, an intriguing situation emerges when we shift to the opposite regime,

$$\frac{L}{v_F} < \frac{\hbar}{k_B T}, \quad (2)$$

which we refer to as the mesoscopic regime.[10] Our work shows that in the mesoscopic regime the surface can indeed change the overall magnetic moment of the sample, in proportion to the volume of the sample.

Before introducing our numerical model, we first motivate it by considering a continuous one-particle effective Hamiltonian, denoted H_c^0 , for a periodic infinite solid. For simplicity we work in two dimensions, but generalization to higher dimensions is straightforward. When dealing with the two-dimensional models, we will refer to the boundary of this model as *edge* instead of *surface*, which we reserve for three-dimensional solids. To simplify our analysis, throughout this work we neglect spin, self-consistency, many-electron effects, and disorder. Our system is assumed to be in thermal equilibrium. We ignore any temperature effects beyond electron occupation smearing.

The complete basis of the eigenstates of H_c^0 can be expressed in the Bloch form, $\psi_{\mathbf{k}}(\mathbf{r}) = e^{i\mathbf{k}\cdot\mathbf{r}}u_{\mathbf{k}}(\mathbf{r})$. However,

not every eigenstate of H_c^0 has the Bloch form. Generally, we can construct arbitrary linear combinations of states that share the same eigenvalue $E_{\mathbf{k}} = E$, and the resulting function

$$\phi_E(\mathbf{r}) = \int_0^1 e^{if(s)} \psi_{\mathbf{k}(s)}(\mathbf{r}) ds \quad (3)$$

is a valid eigenstate of H_c^0 . Here $s \rightarrow \mathbf{k}(s)$ is a continuous parameterization of a curve in the Brillouin zone along which $E_{\mathbf{k}(s)} = E$. For now we limit $f(s)$ so that $f(0) = f(1)$. We choose $f(s)$ so that $\phi_E(\mathbf{r})$ is as localized as possible in the real space.[11] By selecting a fixed $f(s)$, we create a family of functions, ϕ_{mE} , for any integer m , defined as follows,

$$\phi_{mE}(\mathbf{r}) = \int_0^1 e^{i2\pi ms} e^{if(s)} \psi_{\mathbf{k}(s)}(\mathbf{r}) ds. \quad (4)$$

Note, trivially, that $\langle \phi_{mE} | \phi_{m'E'} \rangle = \delta_{mm'} \delta_{EE'}$. Therefore, ϕ_{mE} for all m and E span the same vector space as the Bloch states.[12] Let us now take H_c^0 to correspond to the free-electron system with mass m_e . In this case $\phi_{mE}(\mathbf{r})$ in cylindrical coordinates is simply $\sim e^{im\varphi} J_m\left(\frac{\sqrt{2m_e E}}{\hbar} r\right)$. Here J_m is the Bessel function of the first kind.

Trivially, the expectation value of the angular momentum operator L_z is

$$\langle \phi_{mE} | L_z | \phi_{mE} \rangle = \hbar m. \quad (5)$$

Therefore, each state ϕ_{mE} carries angular momentum $\hbar m$, and orbital magnetic moment $\mu_B m$. Let us now confine our system to a circular region with radius R . From elementary properties of Bessel functions it follows that states with large enough m , close to $R\frac{\sqrt{2m_e E}}{\hbar}$, are localized near the edge of the sample ($r \approx R$). Edge states therefore carry an angular momentum $\hbar m \sim R^1$. The number of states near the edge also scales as $\sim R^1$. Therefore, one might ask whether tweaking the electron potential V^{edge} near the edge of the sample could modify edge states and induce a net orbital moment that scales as $\sim R^2$? If one could construct an edge potential V^{edge} satisfying

$$\langle \phi_{mE} | V^{\text{edge}} | \phi_{m'E} \rangle \sim m \delta_{mm'} \quad (6)$$

then this would be a good candidate edge perturbation, as it breaks the time-reversal symmetry by differently acting on state with different m . For example, one of the effects of this perturbation would be to push $m < 0$ states below the Fermi level, and $m > 0$ states above the Fermi level, thus inducing a net magnetic dipole. [13]

We now attempt to create edge potential satisfying Eq. 6 in a concrete finite-size model using a numerically convenient tight-binding approach. To construct the tight-binding model, we project our continuous free-electron Hamiltonian H_c^0 on the basis of a $N \times N$ square

mesh of s-like orbitals separated by a distance a (orbitals are sketched as black circles in Fig. 1). We label the orbital at site i as $|i\rangle$. For the position operators x and y , we assume $\langle i | x | j \rangle = x_i \delta_{ij}$ and $\langle i | y | j \rangle = y_i \delta_{ij}$. For convenience, we work with the centered operators $\tilde{x} = x - \sum_i x_i / N^2$ and $\tilde{y} = y - \sum_i y_i / N^2$. We also define the following quantity $\tilde{L}(A)$ for any operator A ,

$$\begin{aligned} \tilde{L}(A) &= \frac{im_e}{\hbar} (\tilde{x}[A, \tilde{y}] - \tilde{y}[A, \tilde{x}]) \\ &= \frac{im_e}{\hbar} (\tilde{x}A\tilde{y} - \tilde{y}A\tilde{x}). \end{aligned} \quad (7)$$

Clearly $\tilde{L}(H)$ corresponds to the angular momentum operator for a system described by the Hamiltonian H .

Our general procedure to construct edge potential V^{edge} for any bulk hamiltonian H^0 consists of the following five steps.

Step 1 : choose H^0 .

Step 2 : construct H^{comm} from H^0 .

Step 3 : construct V^{edge} from $\tilde{L}(H^0)$.

Step 4 : diagonalize $H = H^0 + H^{\text{comm}} + V^{\text{edge}}$.

Step 5 : compute $m_{\text{dip}} = \frac{e}{2m_e} \sum_n \langle \psi_n | \tilde{L}(H) | \psi_n \rangle f_n$.

In *step 1* of our procedure, for now we choose the simplest H^0 , where $H_{ij}^0 = \langle i | H^0 | j \rangle = t < 0$ for the nearest-neighbor orbitals i and j , and 0 for any other pair of orbitals (represented by black lines in Fig. 1).

Now we want to construct an edge potential with the property given in Eq. 6. At first it is not clear how to satisfy Eq. 6 in our model, as eigenvectors of H^0 don't have a well-defined angular momentum (our tight-binding model is projected into a finite square mesh of orbitals which breaks continuous rotational symmetry). Therefore, before discussing the edge perturbation, we in *step 2* of our procedure, construct a commutator correction term H^{comm} which ensures that total bulk Hamiltonian,

$$H^{\text{bulk}} = H^0 + H^{\text{comm}}, \quad (8)$$

at least approximately commutes with the angular momentum operator, $\tilde{L}(H^{\text{bulk}})$. The straightforward but tedious construction of H^{comm} is given in the supplement.

The energy spectrum of H^0 as a function of N exhibits some regularity by having spikes in the density of states separated by $\Delta \sim 1/N$. However, the number of states in between spikes is not strictly zero, and these states don't follow an obvious pattern as a function of increasing N . If we include the H^{comm} in our Hamiltonian, we find that it redistributes the spectrum of the system, creating small gaps in the spectrum (scaling as $\Delta \sim 1/N$), as shown in the supplement. We find that placing a Fermi level E_F within one of these gaps has the additional benefit

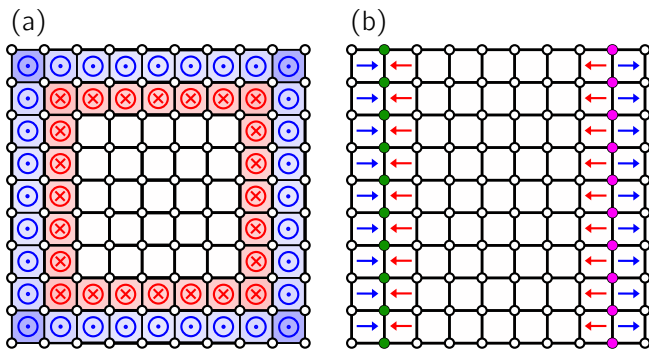


FIG. 1. (a) Perturbation V^{edge} , given by Eq. 9, induces complex phases on hopping elements near the edge. Blue and red colors represent different signs of effective local *magnetic* field on the edge. (b) Perturbation V'^{edge} , given by Eq. 16, changes the onsite energies of green and purple orbitals on the left and right edges. Arrows represent directions of effective local *electric* field at the edge. In both cases (a) and (b) magnitude of effective magnetic and electric fields on the edge is independent of N .

of stabilizing the finite-size effects in our calculations. Related finite-size effects for Landau diamagnetism have also been reported in Refs. 14–18.

Step 3: we now construct edge perturbation

$$V_{ij}^{\text{edge}} = -\frac{eB}{2m_e} S_{ij} \tilde{L}_{ij}(H^0). \quad (9)$$

This term introduces complex phases to the hopping elements on the edge of the model. See panel (a) of Fig. 1 for a sketch of the alternating magnetic flux applied to the edge of the sample.

The S_{ij} term in Eq. 9 ensures that the perturbing potential V^{edge} is zero in the bulk and non-zero only on the edges.[19] Without including S_{ij} in V^{edge} , the resulting V_{ij}^{edge} would represent an approximate interaction term of the orbital magnetic moment with a spatially uniform external magnetic field B , as in the study of Landau diamagnetism. Trivially, the matrix element of such a perturbation is proportional to m , as in Eq. 6.

Step 4: diagonalizing our full Hamiltonian, which includes both bulk and edge contribution,

$$(H^{\text{bulk}} + V^{\text{edge}}) |\psi_n\rangle = E_n |\psi_n\rangle \quad (10)$$

we obtain a set of eigenstates $|\psi_n\rangle$. The largest model we used has $N = 100$, corresponding to a system with 10,000 orbitals.[20] We set the Fermi level E_F to $-2.55|t|$, placing it within a small energy gap Δ in the spectrum.

Step 5: the magnetic dipole moment we compute as

$$m_{\text{dip}} = \frac{e}{2m_e} \sum_n \langle \psi_n | \tilde{L}(H) | \psi_n \rangle f_n. \quad (11)$$

Here f_n is the Fermi-Dirac distribution with effective smearing of electron occupation by $k_B T$. Figure 2 shows

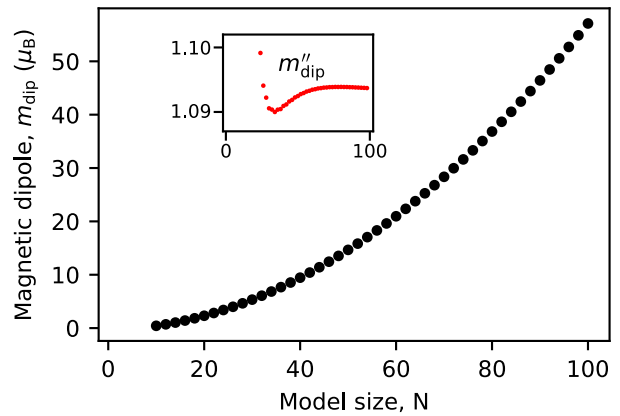


FIG. 2. Changing order N terms in our two-dimensional model induces N^2 change in the computed magnetic dipole m_{dip} . Here, the temperature $k_B T$ in the Fermi-Dirac distribution is set to 0. B is chosen so that $a^2 B = 0.2\hbar/e$. Fermi level E_F is set to $-2.55|t|$ so that the electron density is $\approx 0.12/a^2$. The parameters t and a are set so that the effective mass at low doping is the same as the free electron mass. The inset shows that the second derivative of m_{dip} with respect to N (scaled by 10^2) is constant.

the calculated m_{dip} as a function of N . The computed m_{dip} is clearly extensive for our two-dimensional[21] model, as it scales nearly perfectly as N^2 .

However, as we show in the supplement, we find numerically that the N^2 scaling persists only when

$$k_B T \lesssim 0.2 \Delta \approx 0.6 \frac{|t|}{N}. \quad (12)$$

Since $|t| \sim v_F$ and $N \sim L$ clearly Eq. 12 is equivalent to the definition of the mesoscopic regime given by Eq. 2. In other words, N^2 scaling of m_{dip} in our model persists only in the mesoscopic regime.

Furthermore, we find that m_{dip} can be fitted well to the following functional form, either in the macroscopic or the mesoscopic regime,

$$m_{\text{dip}} \sim \frac{N^2}{1 + \exp \left[3.8 \frac{k_B T}{|t|} \left(N - 0.6 \frac{|t|}{k_B T} \right) \right]}. \quad (13)$$

From this functional form it is clear that $m_{\text{dip}} \sim N^2$ in the mesoscopic regime. More precisely, the following mesoscopic limit

$$\lim_{N \rightarrow \infty} \lim_{T \rightarrow 0^+} \frac{m_{\text{dip}}}{N^2} \neq 0 \quad (14)$$

is non-zero. In other words, the N^2 scaling of the magnetic moment continues for all N , as long as the temperature is small enough. On the other hand, if we swap the order of limits, the resulting macroscopic limit

$$\lim_{T \rightarrow 0^+} \lim_{N \rightarrow \infty} \frac{m_{\text{dip}}}{N^2} = 0 \quad (15)$$

is now zero. In other words, for any fixed small positive T there is an N beyond which the magnetic dipole no longer scales as N^2 .

In the supplementary material,[22] we provide explicit numerical values of Hamiltonian matrix elements H_{ij} for different values of N , as well as a computer code that diagonalizes Eq. 10, computes Eq. 11, and performs a range of consistency checks on H_{ij} .

In hindsight, our finding that m_{dip} in a metal is edge sensitive is perhaps not that surprising considering that a similar dependence was found for the electric dipole d_{dip} of a metal. [23] However, importantly, the electric dipole d_{dip} is edge sensitive in a metal even in a macroscopic regime. Therefore, we can naturally ask why, in the macroscopic regime, m_{dip} from our model behaves differently from d_{dip} ?

To establish a parallel between the electric and magnetic dipole, it is instructive to construct an edge potential V^{edge} that changes the bulk *electric* dipole, in analogy to how V^{edge} changed the bulk magnetic dipole. To achieve this, we use the following procedure.

Step 1' : choose H^0 .

Step 2' : (*commutator correction term not needed.*)

Step 3' : construct V^{edge} from \tilde{x} .

Step 4' : diagonalize $H = H^0 + V^{\text{edge}}$.

Step 5' : compute $d_{\text{dip}} = e \sum_n \langle \psi_n | \tilde{x} | \psi_n \rangle f_n$.

In step 1', we take the same H^0 as before. Step 2' is not needed, as we find that a numerically robust N^2 scaling of d_{dip} is present even without commutator correction.

The important difference is in step 3'. Earlier, in the case of the magnetic dipole, we constructed V^{edge} from the angular momentum operator $\tilde{L}(H^0)$, which induced an effective alternating magnetic field at the edge. Now, by analogy, in step 3' we construct the edge potential from the position operator,

$$V_{ij}^{\text{edge}} = -e\mathcal{E}S_i\tilde{x}_i\delta_{ij}, \quad (16)$$

which induced effective *electric* fields on the edge, proportional to \mathcal{E} . Panel (b) of Fig. 1 shows the sketch of the effective electric fields near the edge induced by V^{edge} . In Eq. 16 we use S_i to ensure that the perturbation potential V^{edge} is zero in the bulk. [24]

In the final step (5') of our procedure, we now compute the expectation value of the *electric* dipole moment, $d_{\text{dip}} = e \sum_n \langle \psi_n | \tilde{x} | \psi_n \rangle f_n$. As shown in the supplement, we find that d_{dip} scales as $\sim N^2$, even in the macroscopic regime, as expected based on Ref. [23].

We assign a different behavior of an electrical dipole to that of a magnetic dipole due to the fact that the magnetic dipole in step 5 is computed as a trace over operator $\tilde{L}(H)$ which explicitly includes the edge perturbation

V^{edge} itself,

$$\tilde{L}(H) = \tilde{L}(H^{\text{bulk}}) + \tilde{L}(V^{\text{edge}}). \quad (17)$$

Therefore, the magnetic dipole m_{dip} can be decomposed into two contributions. The first is a partial trace of $\tilde{L}(H^{\text{bulk}})$

$$m_{\text{dip}}^{\text{st}} = \frac{e}{2m_e} \sum_n \langle \psi_n | \tilde{L}(H^{\text{bulk}}) | \psi_n \rangle f_n, \quad (18)$$

and it arises from changes to the electron state (wavefunction and energy) due to edge perturbation V^{edge} . The second term is a partial trace of $\tilde{L}(V^{\text{edge}})$

$$m_{\text{dip}}^{\text{op}} = \frac{e}{2m_e} \sum_n \langle \psi_n | \tilde{L}(V^{\text{edge}}) | \psi_n \rangle f_n. \quad (19)$$

and it originates from the change in the angular momentum operator by inclusion of perturbation V^{edge} in the total Hamiltonian. This term, in the lowest order of perturbation theory, can be computed already from the unperturbed electron wavefunction and energy.

On the contrary, the electric dipole is calculated in step 5' as a trace over the position operator \tilde{x} , which clearly *does not* depend on the edge perturbation V^{edge} . Therefore, the electric dipole is induced in the model only by changes in the electron wavefunction and energy (analogous to $m_{\text{dip}}^{\text{st}}$). In the case of the electric dipole, there are no terms analogous to $m_{\text{dip}}^{\text{op}}$.

Interestingly, we find that both $m_{\text{dip}}^{\text{st}}$ and $m_{\text{dip}}^{\text{op}}$ are extensive in the macroscopic regime, on their own. However, in the macroscopic regime, these two terms exactly cancel each other, resulting in a nonextensive magnetic dipole in the macroscopic regime. In contrast, in the case of the electric dipole, there is only one contribution (the one coming from changes in the electron's state), so there is no cancelation, and the electric dipole remains edge-sensitive in the macroscopic regime.

In our work, we focus on the simplest choice of H^0 , which corresponds to a square lattice with first-neighbor hoppings. However, the procedure presented in this paper can be done for any H^0 . An interesting case is the Haldane model in a topologically nontrivial insulator phase with a nonzero Chern number.[25] Here, even when the Fermi level is within the bulk gap and crosses the topologically protected edge states, we find $m_{\text{dip}} \sim N^2$. This is numerically robust even without including the commutator correction term H^{comm} .

This work was supported by the NSF DMR-1848074 grant. We acknowledge discussions with R. Wilson and L. Vuong on inverse Faraday effect as these discussions have motivated our work.

[1] T. Thonhauser, D. Ceresoli, D. Vanderbilt, and R. Resta, Orbital magnetization in periodic insulators, *Phys. Rev. Lett.* **95**, 137205 (2005).

- [2] D. Ceresoli, T. Thonhauser, D. Vanderbilt, and R. Resta, Orbital magnetization in crystalline solids: Multi-band insulators, chern insulators, and metals, *Phys. Rev. B* **74**, 024408 (2006).
- [3] K.-T. Chen and P. A. Lee, Effect of the boundary on thermodynamic quantities such as magnetization, *Phys. Rev. B* **86**, 195111 (2012).
- [4] S.-S. Wang, Y. Yu, J.-H. Guan, Y.-M. Dai, H.-H. Wang, and Y.-Y. Zhang, Boundary effects on orbital magnetization for a bilayer system with different chern numbers, *Phys. Rev. B* **106**, 075136 (2022).
- [5] D. Xiao, J. Shi, and Q. Niu, Berry phase correction to electron density of states in solids, *Phys. Rev. Lett.* **95**, 137204 (2005).
- [6] J. Shi, G. Vignale, D. Xiao, and Q. Niu, Quantum theory of orbital magnetization and its generalization to interacting systems, *Phys. Rev. Lett.* **99**, 197202 (2007).
- [7] R. Bianco and R. Resta, Orbital magnetization as a local property, *Phys. Rev. Lett.* **110**, 087202 (2013).
- [8] R. Bianco and R. Resta, Orbital magnetization in insulators: Bulk versus surface, *Phys. Rev. B* **93**, 174417 (2016).
- [9] A. Marrazzo and R. Resta, Irrelevance of the boundary on the magnetization of metals, *Phys. Rev. Lett.* **116**, 137201 (2016).
- [10] Strictly speaking, in the mesoscopic regime we need to require that additionally $k_B T$ is larger than the typical level spacing (scaling as $1/L^2$). If $k_B T$ is smaller than the level spacing, the model is in the microscopic regime. We refer the reader to Ref. 16 where these limits are studied in detail for the related case of Landau diamagnetism. For the present work the distinction between microscopic and mesoscopic regimes is not relevant.
- [11] ϕ_E is only algebraically localized due to integration over part of the Brillouin zone, unlike exponential localization of a Wannier function. Another difference to the Wannier function is that $|\phi_E(\mathbf{r})|^2$ remains stationary in time, in contrast to the Wannier function that disperses in space during its time evolution.
- [12] Transformation defined in Eq. 4 with integer m therefore has similarities to a shift of a Wannier function by a lattice vector \mathbf{R} .
- [13] As we discuss later, there are also other changes to the magnetic moment induced by the edge perturbation. These are changes to the wavefunction, as well as changes to the angular momentum operator itself.
- [14] J. M. van Ruitenbeek and D. A. van Leeuwen, Size effects in orbital magnetism, *Mod. Phys. Lett. B* **07**, 1053 (1993).
- [15] J. M. van Ruitenbeek and D. A. van Leeuwen, Model calculation of size effects in orbital magnetism, *Phys. Rev. Lett.* **67**, 640 (1991).
- [16] E. Gurevich and B. Shapiro, Orbital magnetism in two-dimensional integrable systems, *J. Phys. I France* **7**, 807 (1997).
- [17] A. Aldea, V. Moldoveanu, M. Niță, A. Manolescu, V. Gudmundsson, and B. Tanatar, Orbital magnetization of single and double quantum dots in a tight-binding model, *Phys. Rev. B* **67**, 035324 (2003).
- [18] M. Goldstein and R. Berkovits, Orbital magnetic susceptibility of disordered mesoscopic systems, *Phys. Rev. B* **69**, 035323 (2004).
- [19] We set $S_{ij} = 0$ when orbitals i and j reside in the interior of the sample. When orbitals i and j are on the edge of the model, we set S_{ij} to a non-zero value. As specified in the supplement, the non-zero values of S_{ij} are scaling with system size as $1/N$. This scaling ensures that the complex phase acquired by an electron traversing a closed loop around the edge plaquette (flux) is nearly independent of N and its location along the edge. Our choice of S_{ij} also ensures that the total flux through the entire sample is zero.
- [20] We use even N 's, although odd N 's yields qualitatively similar results with slightly different chemical potential.
- [21] Trivially, stacking of our two-dimensional model to create a three-dimensional solid would result in a $\sim N^3$ scaling of magnetic moment due to perturbing $\sim N^2$ atoms on the surface.
- [22] https://github.com/sinisacoh/supp_morb.
- [23] D. Vanderbilt and R. D. King-Smith, Electric polarization as a bulk quantity and its relation to surface charge, *Phys. Rev. B* **48**, 4442 (1993).
- [24] We set $S_i = 0$ for orbitals i in the bulk, and to a non-zero value, scaling as $\sim 1/N$, on the left and right edges of the model.
- [25] F. D. M. Haldane, Model for a quantum hall effect without landau levels: Condensed-matter realization of the "parity anomaly", *Phys. Rev. Lett.* **61**, 2015 (1988).

Supplement to "Surface sensitivity of magnetization in the mesoscopic regime"

Kevin Moseni¹ and Sinisa Coh^{1,2}

¹*Materials Science and Engineering, University of California Riverside, Riverside, CA 92521, USA*

²*Mechanical Engineering, University of California Riverside, Riverside, CA 92521, USA*

(Dated: December 1, 2023)

I. CONSTRUCTION OF H^{comm}

Given a Hamiltonian H^0 , we wish to construct a commutator correction term H^{comm} such that

$$H^{\text{bulk}} = H^0 + H^{\text{comm}} \quad (1)$$

approximately commutes with the corresponding angular momentum operator, $\tilde{L}(H^{\text{bulk}})$,

$$\left[H^{\text{bulk}}, \tilde{L}(H^{\text{bulk}}) \right] \approx 0. \quad (2)$$

Inserting the definition of H^{bulk} from Eq. 1, and using the linearity of \tilde{L} , we obtain

$$\left[H^0 + H^{\text{comm}}, \tilde{L}(H^0) + \tilde{L}(H^{\text{comm}}) \right] \approx 0. \quad (3)$$

Expanding the second commutator gives us

$$\left[H^0 + H^{\text{comm}}, \tilde{L}(H^0) \right] + \left[H^0, \tilde{L}(H^{\text{comm}}) \right] + \left[H^{\text{comm}}, \tilde{L}(H^{\text{comm}}) \right] \approx 0. \quad (4)$$

If we keep only the lowest order in H^{comm} , and neglect the last term that is quadratic in H^{comm} , we are left with the following,

$$\left[H^0 + H^{\text{comm}}, \tilde{L}(H^0) \right] + \left[H^0, \tilde{L}(H^{\text{comm}}) \right] \approx 0. \quad (5)$$

The unknown matrix H_{ij}^{comm} is generally $N^2 \times N^2 = N^4$ matrix. Therefore, Eq. 5 is a system of N^4 linear equations with N^4 unknowns.

However, we can further restrict H_{ij}^{comm} to zero for distant orbitals i and j , making H^{comm} a local operator. This restriction results in a system of only $\sim N^2$ equations. These equations can be solved using least-square methods. We perform such a minimization of the left-hand side of Eq. 5 while varying the system size N . Our approach produces a purely real H^{comm} that only includes the first-nearest neighbors. The maximum value of $|H_{ij}^{\text{comm}}|$ is $0.5|t|$ independently of N . The operator H_{ij}^{comm} breaks periodicity in the bulk of the sample and resembles the functional form of a parabolic well. The approximate form of H^{comm} is provided in the following section. This form was obtained by fitting the results of our procedure for low N .

II. APPROXIMATE FORM OF H_{ij}^{comm}

The coordinate of orbital i is (x_i, y_i) , as discussed in the main text. The allowed values of x_i and y_i are $0, a, 2a, \dots, (N-1)a$. Now let us introduce the following useful notation,

$$d_i^x = \min[x_i, (N-1)a - x_i], \quad d_i^y = \min[y_i, (N-1)a - y_i]. \quad (6)$$

The quantities d_i^x and d_i^y measure the distance along the x or y axis to the closest edge (either along x or y) of the sample. Next, we define the similar measure of distance for a pair of points i and j ,

$$d_{ij}^x = \frac{1}{2} (d_i^x + d_j^x), \quad d_{ij}^y = \frac{1}{2} (d_i^y + d_j^y) \quad (7)$$

With this notation, we can now give the approximate form of H_{ij}^{comm} . This form was obtained by first explicitly solving for small N the linear system of equations given in the paper. Subsequently, we fit the obtained H_{ij}^{comm} to a simple function that can then be evaluated for any N . To give a fitted approximate form of H^{comm} we first define,

$$h_{ij}^{\min} = \min(d_{ij}^x, d_{ij}^y), \quad h_{ij}^{\max} = \max(d_{ij}^x, d_{ij}^y) \quad (8)$$

Now we set $H_{ij}^{\text{comm}} = 0$ for all (i, j) that are not nearest neighbors. For nearest neighboring (i, j) we set

$$H_{ij}^{\text{comm}} \approx l\left(\frac{h_{ij}^{\text{max}}}{Na}\right)(-t) \quad (9)$$

if h_{ij}^{min}/a is an integer, and

$$H_{ij}^{\text{comm}} \approx l\left(\frac{h_{ij}^{\text{min}}}{Na}\right)(-t) \quad (10)$$

if h_{ij}^{min}/a is not an integer. The function $l(z)$ is defined as $l(z) = 3z^2 - 3z + 1/2$.

III. FORM OF S_{ij}

The object S_{ij} discussed in the main text needs to be zero in the interior and nonzero positive on the edge of the sample. While there are many S_{ij} that could be used to give the same qualitative result, in this work we report results for a specific choice of S_{ij} . First we define

$$D_i = \min(d_i^x, d_i^y), \quad D_{ij} = \frac{1}{2}(D_i + D_j). \quad (11)$$

Therefore, D_i is the distance to the closest edge of the sample, regardless of whether the edge of the sample is on the left, right, top, or bottom side. Then our S_{ij} is

$$S_{ij} = \frac{1}{N} S\left(\frac{D_{ij}}{w}\right). \quad (12)$$

The function $S(z)$ is defined as,

$$S(z) = 16z^2(1-z)^2, \quad (13)$$

when $0 < z < 1$ and $S(z) = 0$ otherwise. The function $S(z)$ has a maximum value of 1 obtained at $z = 1/2$. Since $S(z) = 0$ for $z > 1$, this guarantees that $S_{ij} = 0$ whenever $D_{ij} > w$. All of our calculations are done with $w = 2a$, so that S_{ij} is nonzero only in the two cells closest to the edge.

IV. EDGE CONTRIBUTION TO m_{dip}

The total magnetic dipole of a sample at $k_B T = 0$ we compute as $m_{\text{dip}} = \frac{e}{2m_e} \sum_n^{\text{occ}} \langle \psi_n | \tilde{L}(H) | \psi_n \rangle$. Now we wish to get the contribution of the edge to m_{dip} . For that purpose we define operator $\mathcal{E}(\alpha)$ to project into edge orbitals only,

$$\mathcal{E}(\alpha) = \sum_{D_i \leq \alpha\left(\frac{N}{2}-1\right)a} |i\rangle \langle i|. \quad (14)$$

The thickness of the edge region is $\alpha\left(\frac{N}{2}-1\right)a$. The parameter α is a number between 0 and 1. If α is a small positive number, then only a few sites adjacent to the edge are included in \mathcal{E} . If $\alpha = 1$, the effective edge region is so thick that \mathcal{E} includes the entire sample.

If we further define $\bar{\mathcal{E}} = 1 - \mathcal{E}$ to be a projector into interior orbitals (those that are not on the edge), then by insertion of unity we have $m_{\text{dip}} = \frac{e}{2m_e} \sum_n^{\text{occ}} \langle \psi_n | (\mathcal{E} + \bar{\mathcal{E}}) \tilde{L}(H) (\mathcal{E} + \bar{\mathcal{E}}) | \psi_n \rangle$. Expanding the product we get

$$m_{\text{dip}} = \frac{e}{2m_e} \sum_n^{\text{occ}} \left[\langle \psi_n | \mathcal{E} \tilde{L}(H) \mathcal{E} | \psi_n \rangle + \langle \psi_n | \bar{\mathcal{E}} \tilde{L}(H) \mathcal{E} | \psi_n \rangle + \langle \psi_n | \mathcal{E} \tilde{L}(H) \bar{\mathcal{E}} | \psi_n \rangle + \langle \psi_n | \bar{\mathcal{E}} \tilde{L}(H) \bar{\mathcal{E}} | \psi_n \rangle \right]. \quad (15)$$

We find numerically that the cross-terms (second and third term above) are small in comparison to the first term for most α . We can then use the first term,

$$m_{\text{dip}}(\alpha) = \frac{e}{2m_e} \sum_n^{\text{occ}} \langle \psi_n | \mathcal{E}(\alpha) \tilde{L}(H) \mathcal{E}(\alpha) | \psi_n \rangle, \quad (16)$$

as a measure of contribution of the edge to the magnetic dipole m_{dip} .

V. CONSISTENCY CHECKS

Along with this supplement, we provide numerical values of H_{ij} for various N along with a computer script that finds eigenvectors and computes the magnetic dipole moment. In addition, the same computer script performs the following consistency checks on H_{ij} .

1. The number of occupied electrons, divided by N^2 , is a constant as $N \rightarrow \infty$. Therefore, the order N^2 changes in the magnetic dipole are not due to variations in the number of occupied electronic states.
 2. The only terms with an imaginary part of H_{ij} are near the edge of the sample. In other words, time-reversal breaking edge contributions are really only on the edge.
 3. If we set imaginary terms of H_{ij} to zero, the magnetic dipole moment is also zero. Therefore, time-reversal breaking on the edge is inducing magnetic dipole.
 4. The largest absolute value of the real part of H_{ij} tends to a constant as $N \rightarrow \infty$.
 5. The largest absolute value of the imaginary part of H_{ij} tends to a constant as $N \rightarrow \infty$.
 6. H_{ij} is zero for all pairs (i, j) that are not nearest neighbors. Therefore, H is a local operator.
-

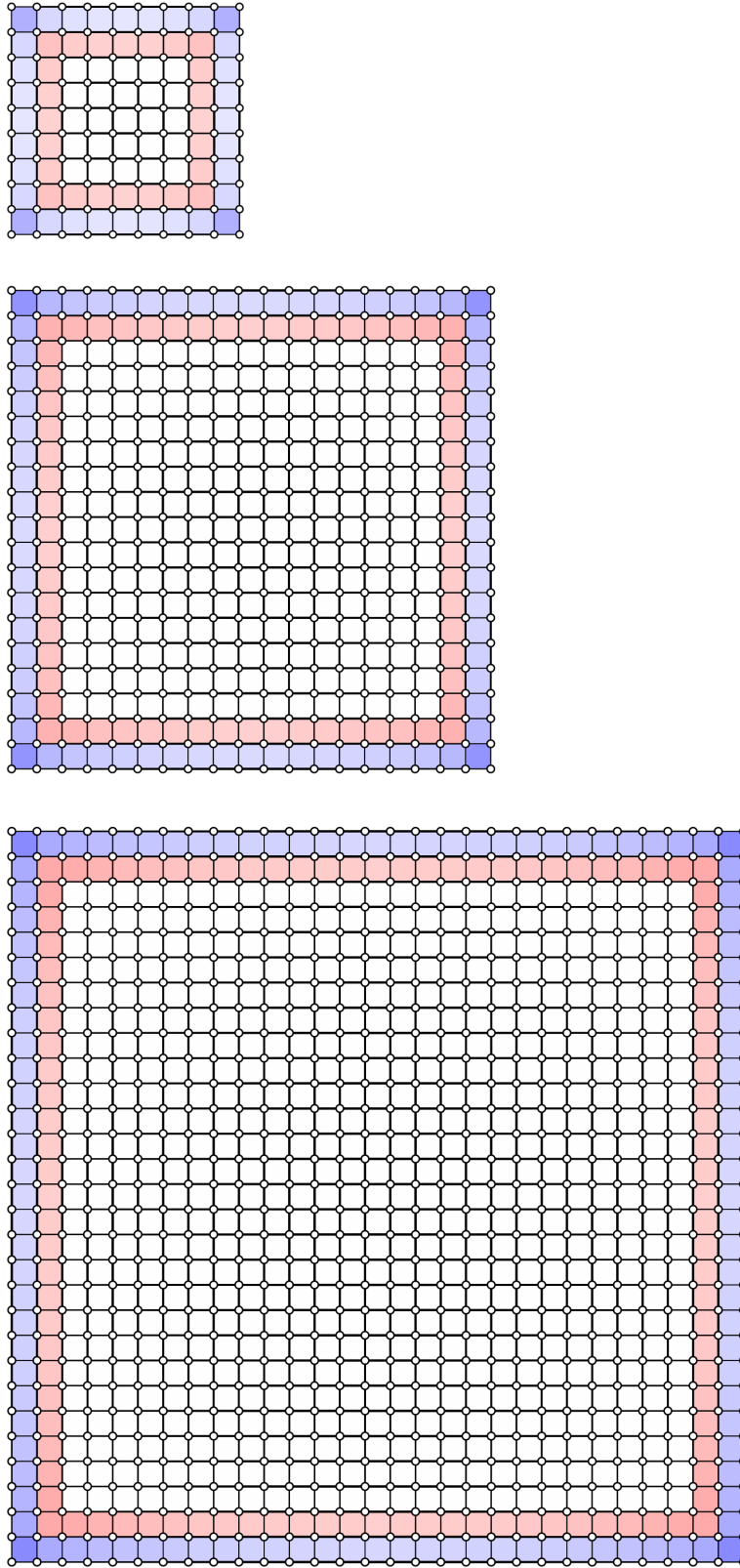


FIG. 1. Sketch of $H = H^{\text{bulk}} + V^{\text{edge}}$ for $N = 10, 20,$ and 30 . Small circles represent orbitals i . The black lines represent the hopping parameters H_{ij} . The thickness of each black line is proportional to $|H_{ij}|$. The variation in the thickness of the black lines is mainly due to H^{comm} . For each square, defined with orbitals i, j, k, l with fixed handedness, we compute the complex phase of $H_{ij}H_{jk}H_{kl}H_{li}$ around the square. If the complex phase is positive, we color the square darker shade of blue, and if it is negative, we color the square darker shade of red. Normalization of the color scale is the same in all three panels.

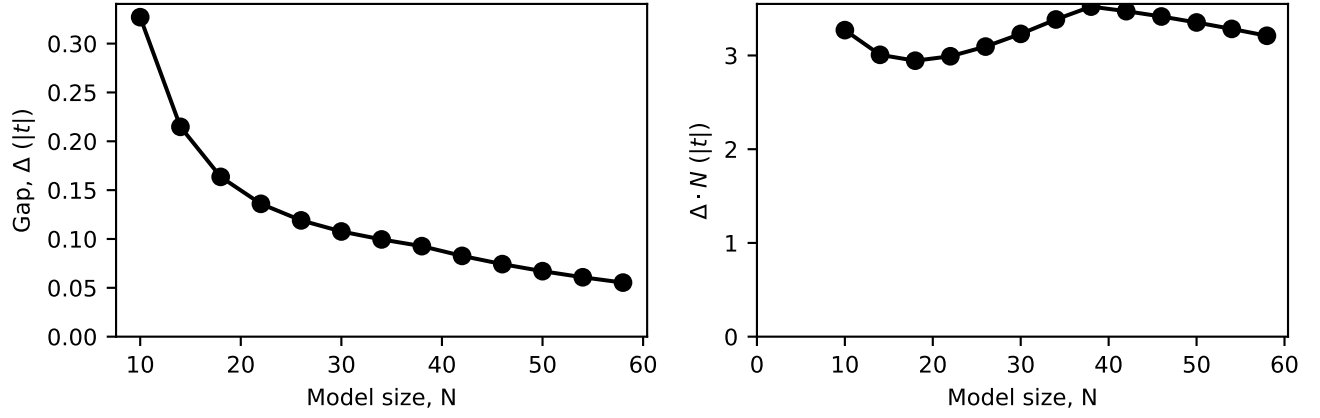


FIG. 2. At $E_F \approx -2.55 |t|$, our model has a gap of size Δ . We find that Δ scales as $1/N$ with model size (N), and therefore $\Delta \rightarrow 0$ when $N \rightarrow \infty$, as expected for a metallic system. The data on the plot fits well to $\Delta \approx \frac{3|t|}{N}$

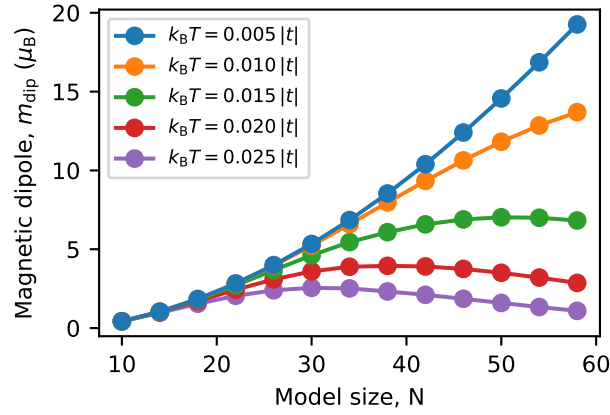


FIG. 3. Dependence of m_{dip} on N for different values of temperature T . As can be seen from the figure, m_{dip} scales as N^2 as long as $k_B T$ is less than $\approx \frac{0.6|t|}{N} \approx 0.2\Delta$.

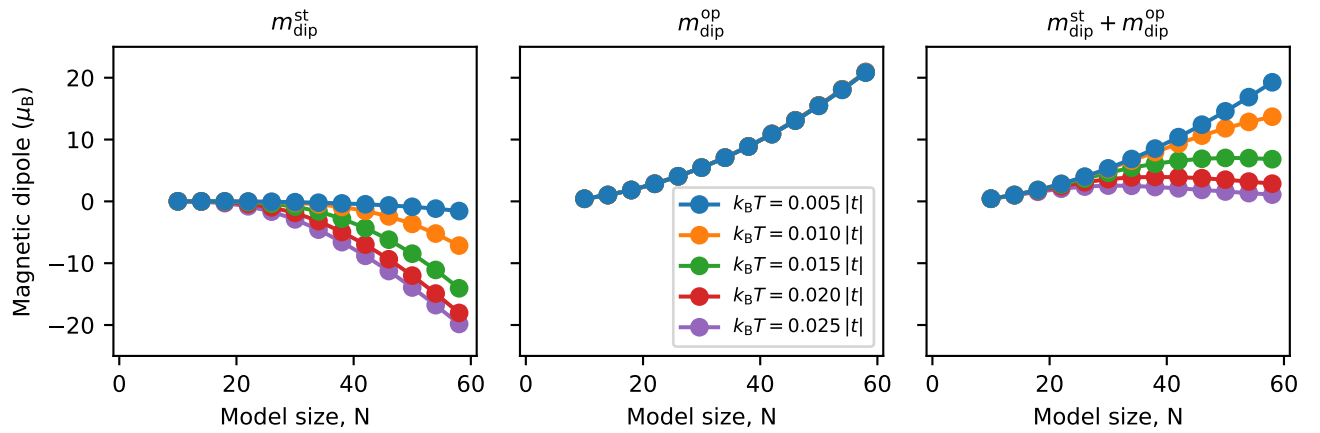


FIG. 4. Two contributions to m_{dip} cancel each other in the macroscopic regime.

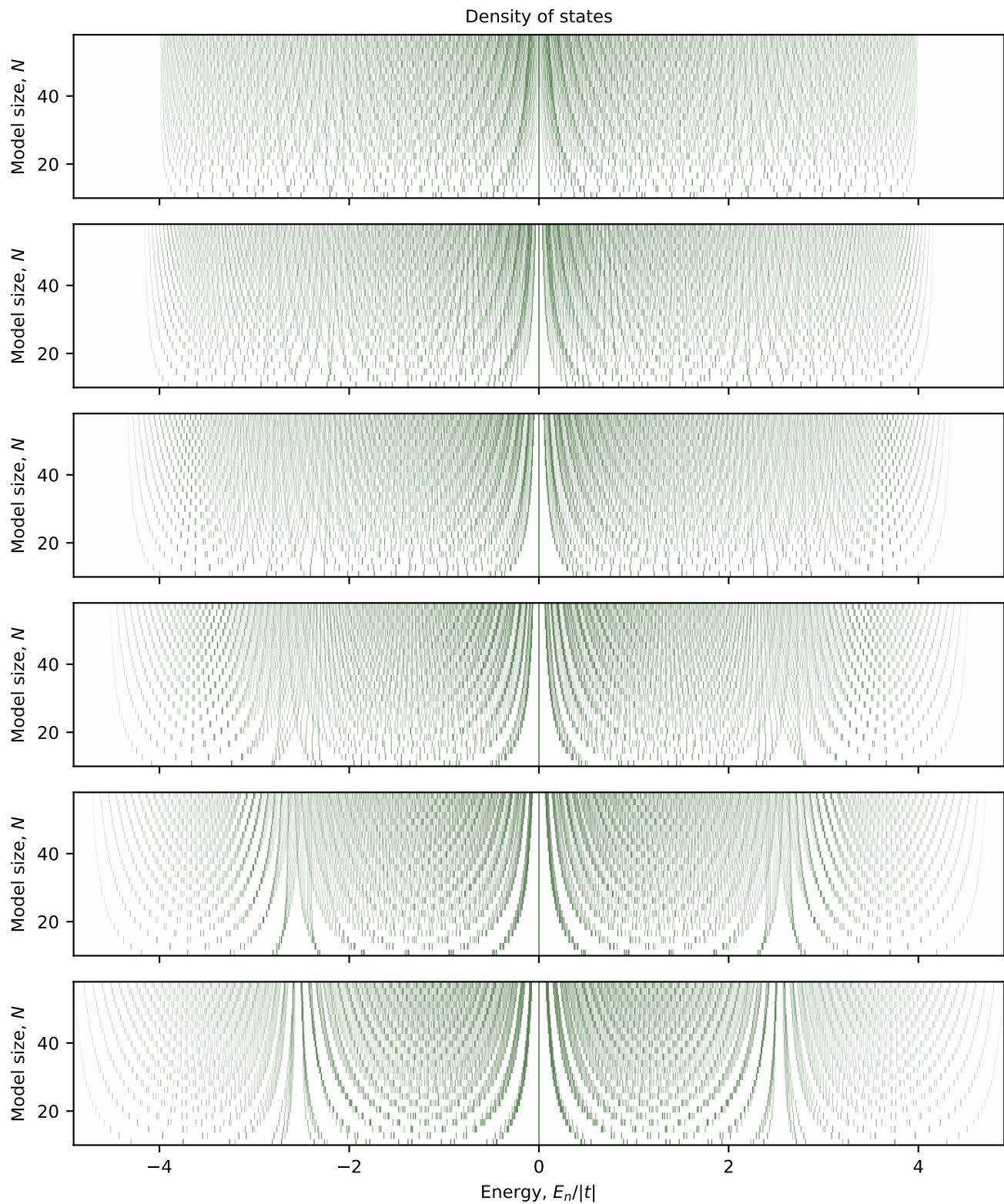


FIG. 5. Density of states for Hamiltonian $H^0 + \beta H^{\text{comm}}$ as a function of N . Parameter β is varied from 0 (top panel) to 1 (bottom panel). Clearly, addition of H^{comm} rearranges the spectrum by opening a set of small gaps $\Delta \sim 1/N$.

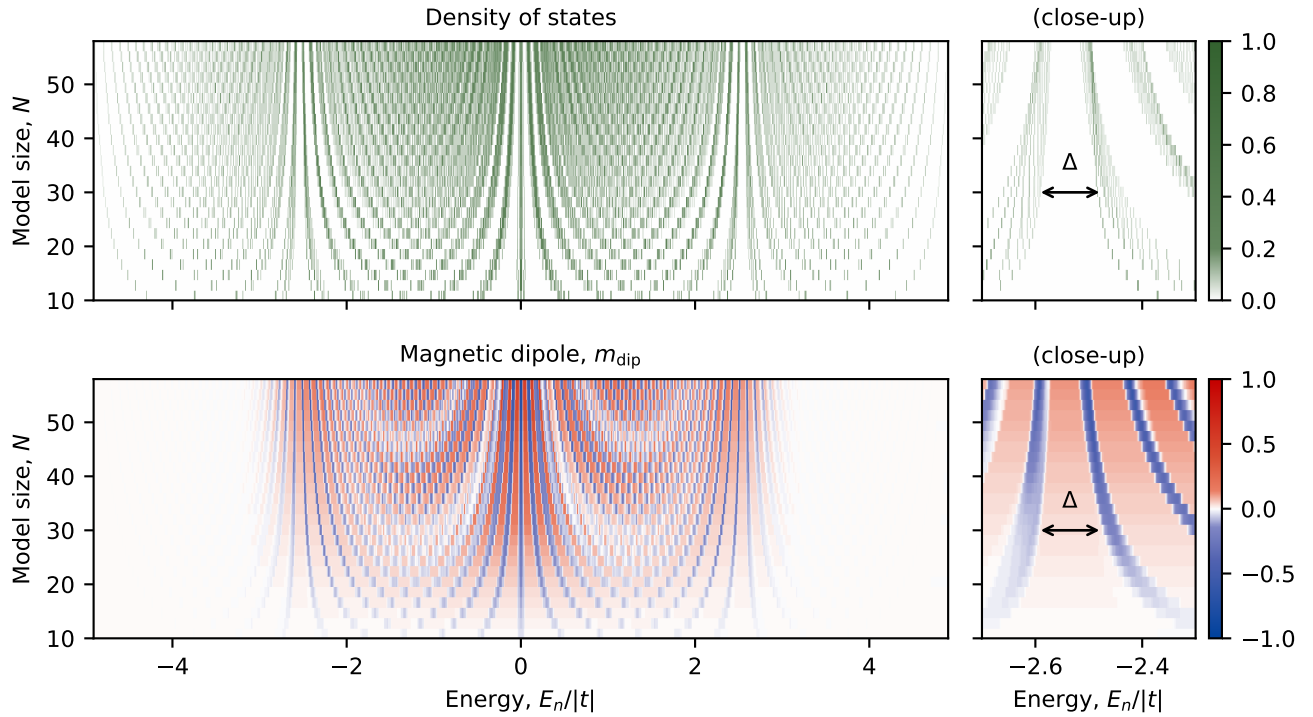


FIG. 6. Density of states (top, green) as a function of energy (E_n) and N . Close-up of the region close to $E_F \approx -2.55|t|$ is shown on the right-hand side. The gap Δ is indicated with an arrow. Bottom panels show the magnetic dipole m_{dip} as a function of Fermi level E_F and N . Red and blue regions indicate positive and negative m_{dip} .

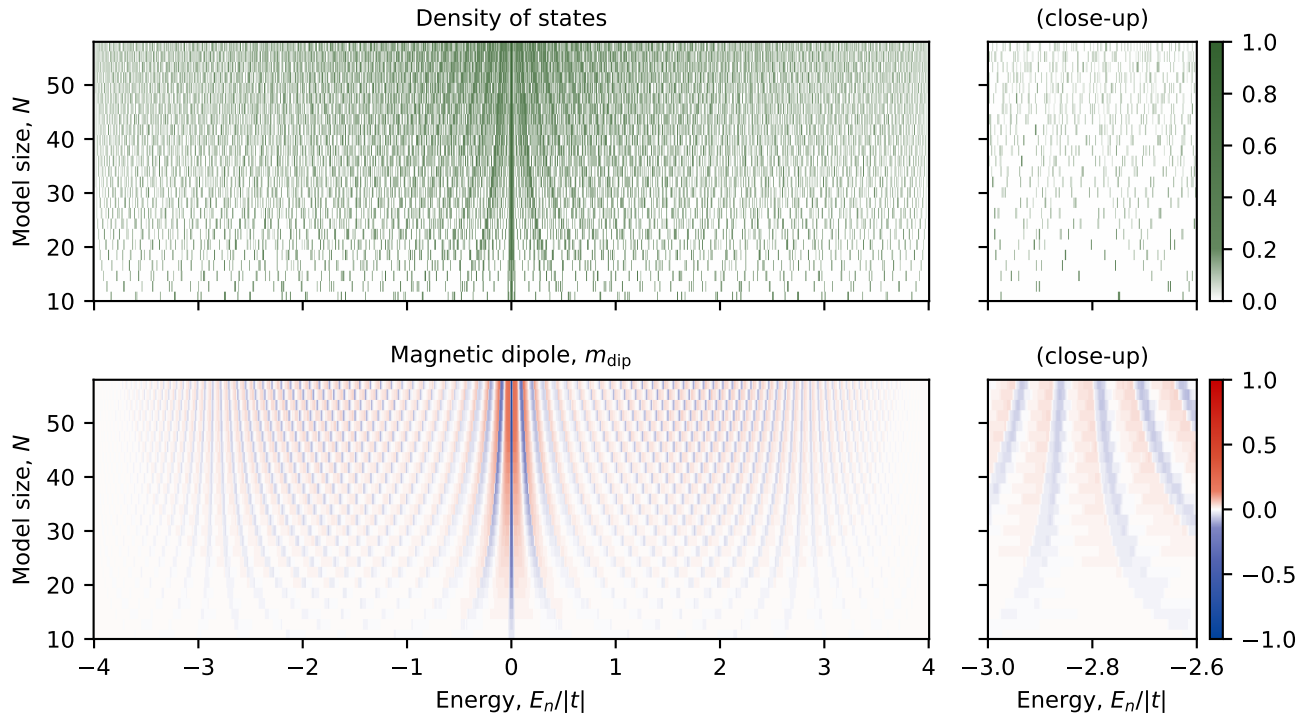


FIG. 7. Same as Fig. 6 but now the Hamiltonian is $H^0 + V^{\text{edge}}$, without the commutator correction term H^{comm} . Clearly, the density of states now no longer have a clearly defined gap, but we still find spikes in density of states.

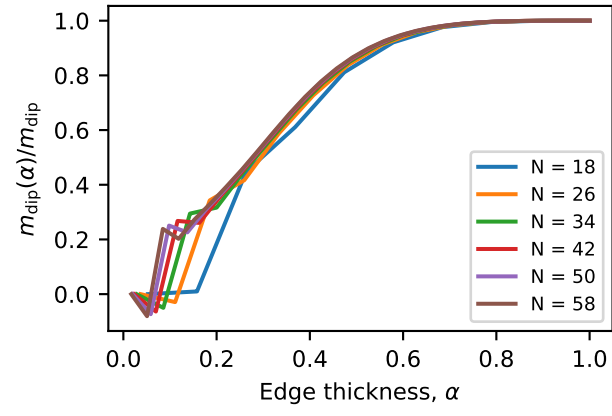


FIG. 8. Quantity $m_{\text{dip}}(\alpha)$ measures contribution of edge to m_{dip} . Thickness of the edge region is parameterized with α , as defined in the text of the supplement. About half of the m_{dip} is recovered when $\alpha \approx 0.3$, regardless of N .

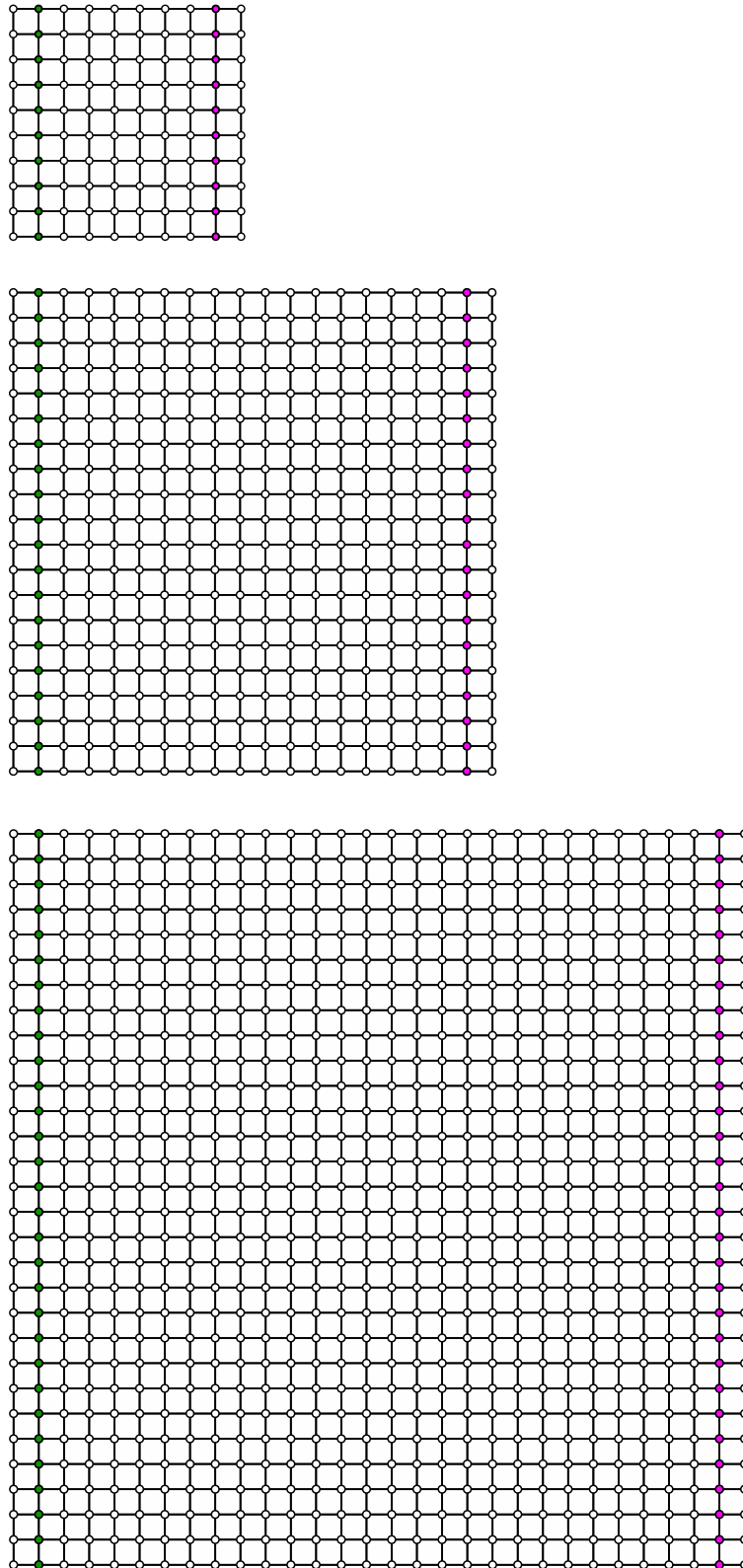


FIG. 9. Sketch of the model using the construction for the electrical dipole V'^{edge} in analogy to our construction for the magnetic dipole. Small circles represent orbitals. The black lines represent the hopping parameters. Here, in the case of the electrical dipole, all hopping parameters have the same magnitude. The edge perturbation for the dipole moment introduces a constant (independent of N) change of the onsite energy at the left and right edge. The different sign of change in the onsite energy on the left versus right edge is indicated with green and purple color.

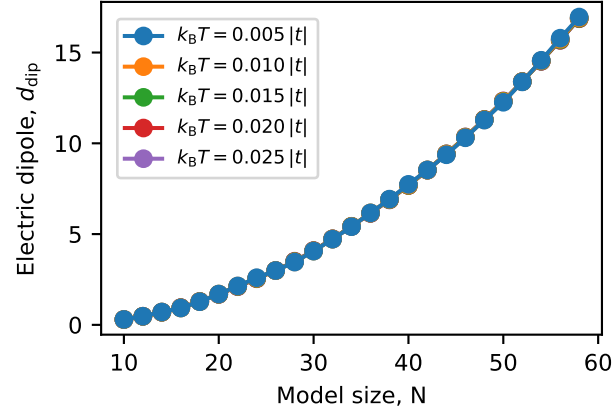


FIG. 10. Dependence of the *electric* dipole d_{dip} on N for different values of temperature T due to edge modification V^{edge} . Here d_{dip} scales as N^2 for any $k_B T$.