Geometrical Effects in Orbital Magnetism

Yang Gao,1 Shengyuan A. Yang,2 and Qian Niu1,3

1 Department of Physics, The University of Texas at Austin, Austin, Texas 78712, USA
2 Engineering Product Development, Singapore University of Technology and Design, Singapore 138682, Singapore
3 International Center for Quantum Materials, Peking University, Beijing 100871, China
(Dated: December 3, 2014)

Within the wave-packet semiclassical approach, the Bloch electron energy is derived to second order in the magnetic field and classified into gauge-invariant terms with clear physical meaning, yielding a fresh understanding of the complex behavior of orbital magnetism. The Berry curvature and quantum metric of the Bloch states give rise to a geometrical magnetic susceptibility, which can be dominant when bands are filled up to a small energy gap. There is also an energy polarization term, which can compete with the Peierls-Landau and Pauli magnetism on a Fermi surface. All these, and an additional Langevin susceptibility, can be calculated from each single band, leaving the Van Vleck susceptibility as the only term truly from interband coupling.

PACS numbers: 73.22.-f, 73.20.At, 75.10.Lp, 75.20.-g

The intrinsic geometrical properties of the Bloch band is of great importance in solid state physics. To zeroth order in electromagnetic fields, the effective mass tensor reflects the curvature of band dispersions, which describes the low energy behaviour near band extrema and enters into the carrier density of states and various transport properties[1]. To first order, an accurate description of Bloch electron dynamics not only requires the knowledge of the band dispersion, but also the Berry curvature and the orbital magnetic moment as functions of the crystal momentum[2]. These quantities reflect the intrinsic geometry of the Bloch state fiber bundle and the band dispersion. The importance of these geometrical band properties have been exemplified in the study of anomalous Hall effects of charge and heat and in the investigation of orbital magnetization[3–8].

How does the intrinsic geometry of Bloch bands affect the second order response to electromagnetic fields, such as orbital magnetic susceptibility? Are there additional geometrical quantities emerge in the orbital magnetic susceptibility? In this Letter, we present an exhaustive analysis of the electron wave-packet energy and orbital magnetic susceptibility by making a gauge invariant classification for the first time. We are able to identify three purely geometrical contributions to the susceptibility, i.e. the Pauli orbital paramagnetism, the Peierls-Landau magnetism, and the geometrical magnetism. These terms only involve single band geometrical quantities including the the effective mass tensor, the orbital magnetic moment, the Berry curvature, and the quantum metric. The geometrical magnetism is a novel mechanism for orbital magnetism, which provides the dominant diamagnetic response around the band gaps, and is especially important in strongly spin/pseudospin-orbit coupled systems such as topological insulators, and single layer or bilayer graphene. We propose that by introducing particle-hole symmetry breaking, it is generally possible to enhance the orbital paramagnetism or even turn the orbital susceptibility from diamagnetic to paramagnetic over an extended energy range.

Moreover, we derive a novel Fermi surface contribution, arising from the energy polarization in the Brillouin zone, and competing with Pauli and Peierls-Landau magnetism. To our delight, this effect, together with a Langevin-like magnetism and those geometrical effects, can be calculated based on Bloch states inside a single Bloch band, and the only interband contribution is the Van Vleck paramagnetism. The various terms can dominate over different energy ranges of the Bloch band. There are many other systematic studies of the orbital magnetic susceptibility, mostly based on the spatial perturbation technique[9–15] or Green’s function formalism[16–22]. However, our result has two advantages: (1) it consists of only gauge-invariant terms, which are independent of the phase choice of the Bloch states; (2) it allows simple physical interpretations of each term and hence the precise classification of geometrical and interband effects. In the end, we discuss the generalization of our theory beyond the minimal coupling and for various effective model Hamiltonians.

Mixing of Bloch states in the wave-packet.— The central concept for the semiclassical theory is the wave-packet constructed from the Bloch states in a single Bloch band (labelled by 0)[2]. Under a weak magnetic field, the full Hamiltonian can be expanded near the central position $r_c$ of the wave packet $|\Psi\rangle$ (assume minimal coupling and set $\epsilon = \hbar = 1$ for simplicity)[2, 23]: $\hat{H} = \hat{H}_c + \hat{H}' + \hat{H}'' + \cdots$. Here $\hat{H}_c(\hat{p}, \hat{q}) = \hat{H}_0(\hat{p} + \frac{1}{2}\hat{B} \times \hat{r}_c, \hat{q})$ is the local Hamiltonian by taking the magnetic vector potential at $r_c$, where $\hat{H}_0$ is the Hamiltonian without external fields, and $\hat{p}$, $\hat{q}$ are the momentum and position operators. $\hat{H}'$ is the first order gradient correction to $\hat{H}_c$: $\hat{H}' = -\frac{1}{2}\hat{B} \cdot [\hat{V} \times (\hat{q} - \hat{r}_c)]$, where $\hat{V} = -i[\hat{q}, \hat{H}_0]$ is the velocity operator. $\hat{H}'' = \frac{1}{2}\Gamma_{ij}[\hat{B} \times (\hat{q} - \hat{r}_c)]_i [\hat{B} \times (\hat{q} - \hat{r}_c)]_j$ is the second order perturbation to $\hat{H}_c$, where $\Gamma_{ij} = \partial_{p_ip_j} \hat{H}_0$ is the Hessian matrix. For nonrelativistic Pauli
Hamiltonians, $\Gamma_{ij} = \delta_{ij}/m$, where $m$ is the bare electron mass. For the relativistic Dirac Hamiltonian, $\Gamma_{ij} = 0$.

In the first order semiclassical theory, the wave-packet is the superposition of local Bloch states of $\hat{H}_c$, i.e. $e^{iqp}|u_0(p + \frac{1}{2}B \times r_c)\rangle$, and is assumed to be localized around some momentum $p_c$ in the Brillouin zone. Then the effective Lagrangian can be calculated for this wave-packet and expressed only in terms of its central position $r_c$, the gauge-invariant central crystal momentum $k_c = p_c + \frac{1}{2}B \times r_c$, and their time derivatives. From the Euler-Lagrange equations of motion, one finds that the dynamics of $k_c$ and $r_c$ contains two geometrical corrections: the orbital magnetic moment $m$ that contributes a Zeeman energy, and the Berry curvature $\Omega$ that modifies the dynamical structure[2].

In the second order theory, the first order correction to the wave-packet is required due to $\hat{H}'$[23]. This correction can be renormalized to a modification to the periodic Hamiltonian $H_0$ schematically in Fig.1. The essential quantity $G_{nm}$ defines a hypersurface in the Brillouin zone, and $\alpha_{k\ell} = \langle u_0|\partial u_0\rangle$ is the intra-band Berry connection. We will see that the momentum shift $\delta p$ enters the description of electrons’ orbital motion through the band geometrical properties, yielding crucial geometrical corrections as discussed later. Since the correction from $\delta p$ mixes Bloch states at neighbouring $k$-points in the same band, we call it the horizontal mixing. We illustrate the two types of mixing of Bloch states schematically in Fig.1.

**Second order wave-packet energy.—**Including the correction to Bloch basis, the wave-packet energy second order in external fields $\tilde{\varepsilon} = \langle \Psi|\hat{H}_c + \hat{H}' + \hat{H}''|\Psi\rangle$ can be arranged into the following compact form[24]

\[
\tilde{\varepsilon} = \varepsilon_0 - B \cdot m + \frac{1}{4}(B \cdot \Omega)(B \cdot m) - \frac{1}{8}\varepsilon_{ik\ell}\varepsilon_{tj\ell}B_xB_tg_{ij}\alpha_{k\ell} - B \cdot (a'_0 \times v_0) + \nabla \cdot P_E + \sum_{n \neq 0} G_{0n}G_{n0} + \frac{1}{8m} (B^2g_{ii} - B_i g_{ij}B_j).
\]

Here $m = -\frac{1}{2}\text{Im}(|\partial u_0| \times (\varepsilon_0 - \hat{H}_c)|\partial u_0\rangle$ is the orbital magnetic moment, $\Omega = -\text{Im}(\partial u_0 \times |\partial u_0\rangle$ is the Berry curvature, $g_{ij} = \text{Re}(\partial_i u_0(\partial_j u_0^\dagger) - a_i a_j)$ is the quantum metric of $k$-space[25, 26], $\alpha_{k\ell} = \partial_{k\ell}\varepsilon_0$ is the inverse of effective mass tensor, $a'_0 = \sum_{n \neq 0} G_{0n}A_{n0}/(\varepsilon_0 - \varepsilon_n)\rangle + \frac{1}{2}\partial_{k\ell}([B \times A_{n0}], A_{n0}) + c.c.$ is the field-induced positional shift of the wave-packet center (its second term is a geometrical quantity from the horizontal mixing and is proportional to the Christoffel symbol[23]), and $P_E = (1/4)[[(B \times \hat{D})u_0](\nabla + v_0) \cdot (B \times \hat{D})u_0 + c.c.$ is a single band quantity representing the energy polarization density in $k$-space. Indices $i$, $j$, $k$, $t$, and $c$ refer to Cartesian coordinates and repeated indices are summed over. All physical quantities in Eq.(2) should be understood as functions of the gauge-invariant crystal momentum $k_c$, and the partial derivatives are with respect to $k_c$.

The two terms in the first line of Eq.(2) are the band energy plus the magnetic dipolar energy, which is the result obtained in the first order semiclassical theory. The two terms in the second line are the geometrical energies, in the sense that they only consist of single band geometrical quantities. The energy dispersion $\varepsilon_0$ defines a hypersurface in the Brillouin zone, and $\alpha_{k\ell}$ reflects its curvature[1]. On the Brillouin zone,
the Hilbert space with single band Bloch states $|u_0\rangle$ forms a fiber bundle, and its curvature is characterized by the Berry curvature $\Omega[2]$. It is interesting to note that $\alpha_{ij}$ and $m$ actually form a conjugate pair: they are proportional to the real and the imaginary part of $\delta_{ij}/m + 2i(\partial_t u_0)(\varepsilon_0 - \hat{H}_c)(\partial_j u_0)$, respectively. So are the quantum metric and the Berry curvature, with respect to the quantity $\langle \partial_t u_0 | \partial_j u_0 \rangle - \langle a_0 \rangle_a \langle a_0 \rangle_j$. Thus the less obvious geometric meaning of $\chi_{ij}$ and $m$ can be understood from their well studied conjugate partners $\Omega$ and $\alpha_{ij}$. The geometrical energy is solely due to the horizontal mixing in the correction to the wave-packet [24], which reflects the property of neighboring $k$-points in the same band.

The two terms in the third line of Eq.(2) are caused by the combined effects of vertical mixing and horizontal mixing. The first term is a real space polarization energy. The magnetic field shifts the wave-packet center by $\alpha_0^T[23]$, hence modifying the magnetic dipole moment and the wave-packet energy. The next term is a $k$-space polarization energy. This can be understood by noticing that the momentum shift $\delta p$ gives rise to a second order energy polarization in $k$-space, $(1/2)(\hat{H}\delta p + c.c.)$. Similar to the relation between electric polarization and wave-vector, the divergence of such energy polarization yields a local energy correction. We find that this term is a single band quantity, and is proportional to the quadrupole moments of the velocity operator[24].

In the fourth line of Eq.(2), the first term is a second order correction due to the vertical mixing, i.e. from virtual interband transitions. The last term in Eq.(2) is from the perturbation of $\hat{H}_c$. Note that this term vanishes for the Dirac Hamiltonian, and for the nonrelativistic Pauli Hamiltonian, it comes with a pre-assumed global parameter $m$.

**Orbital magnetic susceptibility.**—The various second order energy corrections in Eq.(2) are indispensable for the evaluation of the orbital magnetic susceptibility. The general approach is to evaluate the thermodynamic grand potential $G = \text{Tr}[g(\hat{H})]$, where $g(\varepsilon) = -k_B T \ln(1 + \exp(\mu - \varepsilon)/k_B T)$, $T$ is the temperature and $\mu$ is the chemical potential. Under external magnetic field, the semiclassical limit of the grand potential written in terms of physical variables is given by[13]:

$$G = V \int_{\text{BZ}} [Dg(\varepsilon) + g_\text{L}] \frac{d^3k_c}{8\pi^3}. \quad (3)$$

Here $V$ is the system volume and $D = 1 + \mathbf{B} \cdot (\mathbf{\Omega} + \nabla \times \alpha_0^T)$ is the modified density of states[23]. The first term in the bracket of Eq.(3) is from the semiclassical grand potential density with second order energy correction, which yields the semiclassical free energy. The second term $g_\text{L}$ is the Peierls-Landau magnetic energy: $g_\text{L} = -(f'/48) B_c B_r \epsilon_{ijk} \epsilon_{ijk} \alpha_{ik}\alpha_{jk}$, where $f'$ is the energy derivative of the Fermi distribution function $f$. For isotropic bands, the effective mass tensor $\alpha$ takes a diagonal form, and $g_\text{L}$ reduces to its familiar form[1]. This term originates from the discreteness of the Landau levels, and appears when we transform the free energy from the quantum version to its semiclassical limit[13].

We combine Eq.(2) with Eq.(3) and expand the free energy to second order: $G = V \int_{\text{BZ}} (g_0 + g' + g''v) d^3k_c/(8\pi^3)$. At zeroth order, $g_0 = g(\varepsilon_0)$. At first order, $g' = -\mathbf{B} \cdot m f + \mathbf{B} \cdot \mathbf{\Omega} g_0$, which yields the same magnetization as in Ref.8. The second order $g''$ is required for the magnetic susceptibility $\chi_{ij} = -(1/V)(\partial^2 G/\partial B_i \partial B_j)_{\mu,T,V}$, and reads

$$g'' = g_\text{L} + \frac{f'}{2} (\mathbf{B} \cdot \mathbf{m})^2 - \frac{f'}{4} \varepsilon_0 \cdot \mathbf{P}_E$$

$$+ f \frac{G_{0n} G_{n0}}{\varepsilon_0 - \varepsilon_n} + \frac{f}{8m} (B^2 g_{ii} - B_i g_{ij} B_j)$$

$$- \frac{3f}{4} (\mathbf{B} \cdot \mathbf{\Omega}) (\mathbf{B} \cdot \mathbf{m}) - \frac{f}{8} \varepsilon_{sik} \epsilon_{ijl} B_i B_j g_{lk} \alpha_{kl}. \quad (4)$$

Magnetisms in the first line of Eq.(4) are contributions from the Fermi surface. The first two contributions are the Peierls-Landau magnetism, and the Pauli paramagnetism for the orbital moment $m$. In solids, the Peierls-Landau magnetism could become paramagnetic, especially near the band saddle points[22]. The third term is due to the $k$-space energy polarization in Eq.(2), and is first identified here. This term could compete with the Pauli orbital and Peierls-Landau magnetisms, except at the band maxima or minima where $|u_0|$ vanishes.

The other terms in Eq.(4) are Fermi sea contributions. The first term in the second line is the Van Vleck paramagnetism originated from the vertical mixing energy in Eq.(2). It is always paramagnetic after summing over all the occupied bands, similar to the Van Vleck paramagnetism in atomic systems. The second one is a Langevin-like magnetism from the last term in Eq.(2). It can be expressed in a compact form using the quantum metric $g_{ij}$, which describes the intrinsic fluctuation of position-position operators ($q_i q_j$) in the Bloch representation: $g_{ij} = \text{Re}((A_i^\dagger a_n(A_j^\dagger)_{mn})[24]$. For Pauli Hamiltonian with constant mass, this term yields diamagnetic response along directions that diagonalize $g_{ij}$. Its expression will change for effective Hamiltonians with a general Hessian matrix $\Gamma_{ij}$, as will be discussed later.

Magnetisms in the third line in Eq.(4) have no analogs as in atomic physics or free particles, and are first identified here. We call these two contributions the geometrical magnetisms, because they are due to the geometrical energies in Eq.(2) from the horizontal mixing of Bloch states and the geometrical correction to the density of states in Eq.(3). Note that the first term requires either broken time reversal symmetry or broken spatial inversion symmetry[2], while the second term does not. For two band systems with the particle-hole symmetry, band geometrical magnetisms always yield a diamagnetic susceptibility[24] and is the dominant contribution in the
Eq. (4) and Eq. (2) are the main results of this work. By developing a gauge-invariant second order semiclassical theory, we successfully obtain the complete orbital magnetism in a compact form, with each term gauge-invariant and having a clear physical meaning. Of all the terms in Eq. (4), only the Van Vleck contributions involve interband processes, while other terms are single band properties. Particularly, we are able to cast these single band terms (except for energy polarization and the Langvin-like term in general case) in a form only involving the geometrical quantities $\alpha_{ij}$, $m$, $\Omega$, and $g_{ij}$, clearly demonstrating their intrinsic geometrical identity.

As a concrete example to show how various terms contribute to the total magnetic susceptibility, we consider the following tight-binding model defined on a honeycomb lattice[30]:

$$\hat{H} = -t \sum_{\langle i,j \rangle} c^\dagger_i c_j - t' \sum_{\langle \langle i,j \rangle \rangle} c^\dagger_i c_j + \Delta \sum_i \xi_i c^\dagger_i c_i,$$  \hspace{1cm} (5)

where $c_i (c^\dagger_i)$ is the electron annihilation (creation) operator on site $i$, the first and the second terms are the nearest-neighbor and second-neighbor hopping terms, the third term is a staggered potential with $\xi_i = \pm 1$ for the two sublattices, and $t$, $t'$ and $\Delta$ are the strengths of the terms. The staggered potential breaks the inversion symmetry and generates a gap of $2\Delta$ in the spectrum. The second-neighbor hopping is introduced for breaking the particle-hole symmetry.

The various contributions to the orbital magnetic susceptibility are plotted in Fig. 2 with (a) and without (b) particle-hole symmetry. In the presence of particle-hole symmetry ($t' = 0$), the energy polarization and the Van Vleck contributions vanish identically. From Fig. 2(a), one observes that in the gap the Fermi surface terms vanish and the geometrical magnetism dominates, which leads to a large diamagnetic response. The magnitude of the geometrical magnetism decreases rapidly away from the gap and it (along with the Peierls-Landau term) is compensated largely by the Pauli orbital paramagnetism which is peaked at the band edges where $m$ takes its largest value[28]. Two noticeable paramagnetic peaks are observed around the band saddle points due to the Peierls-Landau contribution, which is a general feature as discussed before[22]. Further away from the gap region, the susceptibility decreases gradually to zero. Our result of $\chi$ agrees with that from the exact quantum treatment[19].

The physics around gap can be described by the gapped Dirac model $\hat{H} = v k_i \sigma_1 + v k_2 \sigma_2 + \Delta \sigma_3$ with $v = 3at/2$ where $a$ is the nearest-neighbor bond length[30]. This model is widely used in the study of graphene, MoS$_2$, topological insulator surfaces and thin films[29–32]. Here $\sigma$'s are the Pauli matrices. Near the band edge ($|\mu| > \Delta$), the three competing magnetisms (Pauli $\chi_P$, Peierls-Landau $\chi_L$, and geometrical $\chi_{Geom}$) read explicitly

$$\chi_P = \frac{e^2 \Delta^2 v^2}{8\pi|\mu|^3}, \quad \chi_L = -\frac{\chi_P}{3}, \quad \chi_{Geom} = \frac{e^2 \Delta^2 v^2}{12\pi|\mu|^3}. \hspace{1cm} (6)$$

We emphasize that for systems with two valleys connected by time reversal operation, such as graphene or MoS$_2$, the geometrical magnetisms from the two valleys have the same sign. Note that in this low energy model the total susceptibility vanishes identically outside gap, which seems contradicting to the result in Fig. 2(a) where one sees a finite paramagnetic plateau. The difference is due to two Fermi sea contributions including the Langvin and a term in geometrical magnetism resulting from the nonzero Hessian $\Gamma_{ij}$ (whereas $\Gamma_{ij}$ vanishes in the low energy model) which produce an overall shift of $\chi$. This was known as the “lattice contribution” in previous studies[18]. This agreement demonstrates the validity of our theory.

When particle-hole symmetry is broken by a finite $t'$, as shown in Fig. 2(b), the geometrical magnetism still dominates in the gap and one notes that the paramagnetic plateau near gap is suppressed in the valence band while enhanced in the conduction band. Now the energy polarization and the Van Vleck contributions are finite. With current parameters, the Van Vleck paramagnetism takes a small value around the gap region, while the energy polarization contribution takes opposite signs between the two bands. The energy polarization term, along with the enhanced Langevin and Pauli terms are the main contributions to a large paramagnetic response between the conduction band edge and the saddle point. This is different from the usual orbital paramagnetism resulting from the Peierls-Landau contribution. In fact, the contribution from Peierls-Landau is less important even in the region near and above the saddle point in conduc-

FIG. 2. (color online) Orbital magnetic susceptibility for the lattice model (5) as a function of $\mu$. $\chi$ is in units of $\chi_0 = e^2 v^2 \alpha^2/(4\pi^2 h^2)$. $\alpha$ is the bond length. Here $\Delta = 0.2t$, (a) $t' = 0$ and (b) $t' = 0.1t$. Here P-L, E Polar, and SP stand for the Peierls-Landau, energy-polarization, and saddle point, respectively.
tion band. The total susceptibility there is more affected by the competition between geometrical, Langevin, and energy polarization terms. This is in contrast to valence band where the Peierls-Landau dominates while other contributions are suppressed.

This example illustrates that: (1) the geometrical magnetism is an important contribution, especially around the band gap; (2) different terms in our classification could dominate over different energy ranges; (3) it is possible to enhance the paramagnetic susceptibility by breaking the particle-hole symmetry. In addition, we note that for a generic two band model \( \hat{h} = h_0 + \mathbf{h} \cdot \mathbf{\sigma} \), \( G_{n_0} = -\mathbf{B} \cdot (\partial h_0 \times A_{n_0}) \) is finite when particle-hole symmetry is broken. From Eq.(4), we find that only the Van Vleck paramagnetism depends quadratically on \( \partial h_0 \), while all other terms are either independent or only have linear dependence[24]. This implies that Van Vleck could in principle dominate for large \( |\partial h_0| \), which also leads to an strong paramagnetic response.

Discussions.—The wave packet energy in Eq.(2) and hence the susceptibility in Eq.(4) apply in general if we start from the Pauli Hamiltonian with a scalar particle mass \( m \). If we start from an effective Hamiltonian we should instead compute its Hessian matrix \( \Gamma_{ij} \) first, and the last term in Eq.(2) becomes[24]: \( \varepsilon r = (1/8)(B \times D)_{i_0} \Gamma_{ij} (B \times D)_{j_0} - (1/16)(B \times D)_{i_0} \partial_j (B \times D)_{j_0} \Gamma_{ij} \). The form of the Langevin-like magnetism changes accordingly. For example, we verify that in the low energy model of the bilayer graphene, this modified Langevin-like magnetism yields the correct logarithmic behavior of the susceptibility[16].

Our theory could also be generalized beyond the minimal coupling. Based on the Foldy-Wouthuysen transformation in solid state context, the correction beyond minimal coupling to the Hamiltonian is[34, 35]: \( \Delta \hat{H} = \hat{B} \cdot \mu + B_\parallel \hat{n}_i B_y \), where \( \mu \) and \( \hat{n}_i \) are appropriate matrix operators as functions of \( (\mathbf{p} + i \mathbf{B} \cdot \mathbf{r}_c) \). Then the expectation value of the second term directly adds to the second order wave-packet energy, while the first term modifies the orbital magnetic moment. For example, \( \hat{\mu} \) could stand for the electron spin magnetic moment, whose diagonal and off-diagonal parts add to the corresponding orbital moment \( m \) and \( G_{n_0} \), respectively.

Acknowledgment.—We acknowledge valuable discussions with S. Guan, Z. Qiao, H. Chen, J. Zhou, X. Li, R. Cheng, and L. Zhang. We especially acknowledge J.-N. Fuchs, A. Raoux, F. Piéchon and G. Montambaux for pointing out the numerical mistake in our previous manuscript. QN is supported by NBRPC (No.2012CB921300 and No.2013CB921900), and NSFC (No.91121004). SAY is supported by SUTD-SRG-EPD-2013062. YG is supported by DOE (DE-FG03-02ER45958, Division of Materials Science and Engineering) and Welch Foundation (F-1255).

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