Berry Phase Correction to Electron Density of States in Solids

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Liouville's theorem on the conservation of phase-space volume is violated by Berry phase in the semiclassical dynamics of Bloch electrons. This leads to a modification of the phase-space density of states, whose significance is discussed in a number of examples: field modification of the Fermi-sea volume, connection to the anomalous Hall effect, and a general formula for orbital magnetization. The effective quantum mechanics of Bloch electrons is also sketched, where the modified density of states plays an essential role.

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Semiclassical dynamics of Bloch electrons in external fields has provided a powerful theoretical framework to account for various properties of metals, semiconductors, and insulators [1]. In recent years, it has become increasingly clear that essential modification of the semiclassical dynamics is necessary for a proper understanding of a number of phenomena. It was known earlier that global geometric phase effects [2,3] on Bloch states are very important for insulators in our understanding of the quantum Hall effect [4], quantized adiabatic pumps [5], and electric polarization [6,7]. It was shown [8,9] later that geometric phase also modifies the local dynamics of Bloch electrons and thus affects the transport properties of metals and semiconductors. Recently these ideas have been successfully applied to the anomalous Hall effect in ferromagnetic semiconductors and metals [10–13], as well as spin transport [14,15].

In this Letter, we reveal a general property of the Berry phase modified semiclassical dynamics which has been overlooked so far: the violation of Liouville's theorem for the conservation of phase-space volume. Liouville's theorem was originally established for standard classical Hamiltonian dynamics, and its importance cannot be overemphasized as it serves as a foundation for classical statistical physics. The Berry phase makes, in general, the equations of motion noncanonical [8,9,16–18], rendering the violation of Liouville's theorem. Nevertheless, we are able to remedy the situation by modifying the density of states in the phase space.

This modified phase-space density of states enters naturally in the semiclassical expression for the expectation value of physical quantities, and has profound effects on equilibrium as well as transport properties. We demonstrate this with several examples. First, we consider a Fermi sea of electrons in a weak magnetic field, and show that the Fermi-sea volume can be changed linearly by the field. Second, we show how the Berry phase formula for the intrinsic anomalous Hall conductivity may be derived from equilibrium thermodynamics using the Středa formula [19]. Third, we provide a general derivation of an orbital-magnetization formula which is convenient for first-principles calculations.

In addition, we present an effective quantum mechanics for Bloch electrons in solids by quantizing the semiclassical dynamics with the geometric phase. The density of states enters in a nontrivial manner into the commutators of the phase-space coordinates, and relates directly to the minimal uncertainty volume in the phase space.

To begin with, we write down the semiclassical equations of motion for a Bloch electron in weak electric and magnetic fields [9]

$$\dot{\boldsymbol{r}} = \frac{1}{\hbar}\frac{\partial \varepsilon_n(\boldsymbol{k})}{\partial \boldsymbol{k}} - \dot{\boldsymbol{k}} \times \boldsymbol{\Omega}_n(\boldsymbol{k}), \tag{1a}$$

$$\hbar\dot{\boldsymbol{k}} = -e\boldsymbol{E}(\boldsymbol{r}) - e\dot{\boldsymbol{r}} \times \boldsymbol{B}(\boldsymbol{r}), \tag{1b}$$

where $\boldsymbol{\Omega}_n(\boldsymbol{k})$ is the Berry curvature of electronic Bloch states defined by $\boldsymbol{\Omega}_n(\boldsymbol{k}) = \mathrm{i}\langle\boldsymbol{\nabla}_{\boldsymbol{k}} u_n(\boldsymbol{k})| \times |\boldsymbol{\nabla}_{\boldsymbol{k}} u_n(\boldsymbol{k})\rangle$ with $|u_n(\boldsymbol{k})\rangle$ being the periodic part of Bloch waves in the nth band; $\varepsilon_n(\boldsymbol{k})$ is the band energy with a correction due to the orbital magnetic moment [see Eq. (10) and above]. For crystals with broken time-reversal symmetry (such as ferromagnetic materials) or spatial inversion symmetry (such as GaAs), the Berry curvature $\boldsymbol{\Omega}_n(\boldsymbol{k})$ is nonzero.

To show the violation of Liouville's theorem, we consider the time evolution of a volume element $\Delta V = \Delta\boldsymbol{r}\Delta\boldsymbol{k}$ in the phase space. The equation of motion for ΔV is given by $(1/\Delta V)d\Delta V/dt = \boldsymbol{\nabla}_{\boldsymbol{r}} \cdot \dot{\boldsymbol{r}} + \boldsymbol{\nabla}_{\boldsymbol{k}} \cdot \dot{\boldsymbol{k}}$ [20]. A straightforward but somewhat tedious calculation shows that the right-hand side is equal to $-d\ln(1 + e\boldsymbol{B} \cdot \boldsymbol{\Omega}/\hbar)/dt$, which is a total time derivative. Therefore we can solve for the time evolution of the volume element and obtain

$$\Delta V = \Delta V_0/(1 + e\boldsymbol{B} \cdot \boldsymbol{\Omega}_n/\hbar). \tag{2}$$

The fact that the Berry curvature is generally $\boldsymbol{k}$ dependent (and the magnetic field can also depend on $\boldsymbol{r}$) implies that the phase-space volume element changes during time evolution of the state variables $(\boldsymbol{r}, \boldsymbol{k})$.

Nevertheless, we have a remedy to this breakdown of Liouville's theorem. Equation (2) shows that the volume element is a local function of the state variables (through

the magnetic field and the Berry curvature) and has nothing to do with the history of time evolution. We can thus introduce a modified density of states

$$D_n(\boldsymbol{r}, \boldsymbol{k}) = (2\pi)^{-d}(1 + e\boldsymbol{B} \cdot \boldsymbol{\Omega}_n/\hbar), \tag{3}$$

such that the number of states in the volume element, $D_n(\boldsymbol{r}, \boldsymbol{k})\Delta V$, remains constant in time, where d is the spatial dimensionality of the system. The prefactor $(2\pi)^{-d}$ is obtained by demanding that the density of states $D_n(\boldsymbol{r}, \boldsymbol{k})$ reduces to the conventional form when the Berry curvature vanishes. As will be shown later, this density of states corresponds to the minimal quantum uncertainty volume of the state variables. Therefore, it does serve as the semiclassical measure for the number of quantum states per unit volume in the phase space. Based on this understanding, we write the classical phase-space probability density as

$$\rho_n(\boldsymbol{r}, \boldsymbol{k}, t) = D_n(\boldsymbol{r}, \boldsymbol{k}) f_n(\boldsymbol{r}, \boldsymbol{k}, t), \tag{4}$$

with $f_n(\boldsymbol{r}, \boldsymbol{k}, t)$ being the occupation number of the state labeled by $(\boldsymbol{r}, \boldsymbol{k})$. Probability conservation demands that $\rho_n(\boldsymbol{r}, \boldsymbol{k}, t)$ satisfies the continuity equation in phase space. On the other hand, our density of states satisfies $dD_n/dt = -(\boldsymbol{\nabla}_{\boldsymbol{r}} \cdot \dot{\boldsymbol{r}} + \boldsymbol{\nabla}_{\boldsymbol{k}} \cdot \dot{\boldsymbol{k}})D_n$. It then follows that the occupation number introduced above has the desired property of being invariant along the trajectory, i.e., $df_n/dt = 0$ [21].

We can thus write the real space density of a physical observable $\hat{O}$ in the form [22]

$$\bar{O}(\boldsymbol{R}) = \sum_n \int d\boldsymbol{k} D_n(\boldsymbol{r}, \boldsymbol{k}) f_n(\boldsymbol{r}, \boldsymbol{k}, t) \langle \hat{O}\delta(\hat{\boldsymbol{r}} - \boldsymbol{R}) \rangle_{\boldsymbol{r}\boldsymbol{k}n} \tag{5}$$

where $\langle \cdots \rangle_{\boldsymbol{r}\boldsymbol{k}n}$ denotes the expectation value in the wave-packet state centered at $(\boldsymbol{r}, \boldsymbol{k})$ with the band index n. In the spatially homogeneous case, it reduces to

$$\bar{O} = \sum_n \int d\boldsymbol{k} D_n(\boldsymbol{k}) f_n(\boldsymbol{k}) O_n(\boldsymbol{k}), \tag{6}$$

where $O_n(\boldsymbol{k})$ is the expectation value of $\hat{O}$ in a Bloch state. For simpler notation, we will drop the band index n and assume that the integral over $\boldsymbol{k}$ includes the sum over n.

We now discuss the magnitude of the correction term $e\boldsymbol{B} \cdot \boldsymbol{\Omega}/\hbar$ to the density of states in Eq. (3). The Berry curvature for several materials has been calculated before using the first-principles method [11,12]. Over large regions of the Brillouin zone, its magnitude is on the order of a^2 with a being the lattice constant. Thus, $e\boldsymbol{B} \cdot \boldsymbol{\Omega}/\hbar \sim eBa^2/\hbar$ is the ratio of the magnetic flux through a unit cell to the magnetic flux quantum, and can be 10^{-2} to 10^{-3} for a magnetic field of 1 T. In the vicinity of some isolated points, the Berry curvature can be several orders of magnitude higher, leading to bigger effects for measurement. In the following, we will present a number of applications of our formula Eq. (6).

In our first example, we consider the quantity of electron density and show that the Fermi-sea volume can be changed linearly by a magnetic field when the Berry curvature is nonzero. Assuming zero temperature and using Eq. (3), we have the electron density as

$$n_e = \int^{\mu} \frac{d\boldsymbol{k}}{(2\pi)^d} \left(1 + \frac{e\boldsymbol{B} \cdot \boldsymbol{\Omega}}{\hbar}\right), \tag{7}$$

where the upper limit means that the integral is over states with energies below the chemical potential μ. Noting that the electron density is fixed by the background charge density, we conclude that the Fermi volume must change with the magnetic field. To first order, this change is given by

$$\delta V_F = -\int^{\mu_0} d\boldsymbol{k} \frac{e\boldsymbol{B} \cdot \boldsymbol{\Omega}}{\hbar}. \tag{8}$$

We note that while Landau levels make the Fermi-sea volume oscillate with the field, the effect described above gives an overall shift on average. Such a shift has important implications to those Fermi-surface related behaviors such as transport properties. For instance, in metals, it can induce a magnetoresistance linearly depending on the magnetic field. On the other hand, in band insulators, the $\boldsymbol{k}$ space is limited to the Brillouin zone. Electrons must populate a higher band if $(e/\hbar)\int_{\mathrm{BZ}} d\boldsymbol{k}\boldsymbol{B} \cdot \boldsymbol{\Omega}$ is negative. When this quantity is positive, holes must appear at the top of the valence bands. Discontinuous behavior of physical properties in a magnetic field is therefore expected for band insulators with a nonzero integral of the Berry curvatures (Chern numbers).

In our second example, we show a connection between our phase-space density of states to the intrinsic anomalous Hall effect, which is due to spin-orbit coupling in the band structure of a ferromagnetic crystal. In the context of the quantum Hall effect, Středa derived a formula relating the Hall conductivity to the field derivative of the electron density at a fixed chemical potential [19], $\sigma_{xy} = -e(\partial n_e/\partial B_z)_{\mu}$. There is a simple justification of this relation by a thermodynamic argument by considering the following adiabatic process in two dimensions. A time dependent magnetic flux generates an electric field with an emf around the boundary of some region; and the Hall current leads to a net flow of electrons across the boundary and thus a change of electron density inside. This argument can be straightforwardly applied to the case of anomalous Hall effect and to three dimensions. By taking the derivative of the electron density (7) with respect to $\boldsymbol{B} = B\hat{\boldsymbol{z}}$ at fixed chemical potential, we find that

$$\sigma_{xy} = -\frac{e^2}{\hbar} \int^{\mu} \frac{d\boldsymbol{k}}{(2\pi)^d} \Omega_z. \tag{9}$$

This is an intrinsic effect because it is independent of scattering, and thus differs from conventional skew scattering and side jump mechanisms [10–13].

As a third example of application, we now derive a semiclassical formula for orbital magnetization. In the semiclassical picture, a Bloch electron is modeled by a wave packet in a Bloch band, which is found to rotate about

its center of mass in general, yielding an intrinsic magnetic moment given by $\boldsymbol{m}(\boldsymbol{k}) = -\mathrm{i}(e/2\hbar)\langle\boldsymbol{\nabla}_{\boldsymbol{k}}u| \times [\hat{H}_0(\boldsymbol{k}) - \varepsilon_0(\boldsymbol{k})]|\boldsymbol{\nabla}_{\boldsymbol{k}}u\rangle$, where $\hat{H}_0$ is the Hamiltonian [23]. In the presence of a weak magnetic field $\boldsymbol{B}$, the electron band structure energy $\varepsilon_0(\boldsymbol{k})$ (which may already include Zeeman energy from spin magnetization) acquires a correction term from this intrinsic orbital moment [8,9], $\varepsilon(\boldsymbol{k}) = \varepsilon_0(\boldsymbol{k}) - \boldsymbol{m}(\boldsymbol{k}) \cdot \boldsymbol{B}$. For an equilibrium ensemble of electrons, the total orbital magnetization can be found from the total energy, which is given by Eq. (6) as,

$$E = \int^{\mu} \frac{d\boldsymbol{k}}{(2\pi)^d}\left(1 + \frac{e\boldsymbol{B}\cdot\boldsymbol{\Omega}}{\hbar}\right)\Big(\varepsilon_0(\boldsymbol{k}) - \boldsymbol{m}(\boldsymbol{k})\cdot\boldsymbol{B}\Big). \quad (10)$$

Taking the differential of E with respect to $\boldsymbol{B}$, we obtain the magnetization at zero magnetic field to be

$$\begin{aligned}\boldsymbol{M} &= \int^{\mu_0} \frac{d\boldsymbol{k}}{(2\pi)^d}\left(\boldsymbol{m}(\boldsymbol{k}) + \frac{e\boldsymbol{\Omega}}{\hbar}[\mu_0 - \varepsilon_0(\boldsymbol{k})]\right) \\ &= \frac{e}{2\hbar}\int^{\mu_0} \frac{d\boldsymbol{k}}{(2\pi)^d}\mathrm{i}\left\langle\frac{\partial u}{\partial \boldsymbol{k}}\right| \times [2\mu_0 - \varepsilon_0(\boldsymbol{k}) - \hat{H}_0]\left|\frac{\partial u}{\partial \boldsymbol{k}}\right\rangle. \end{aligned} \quad (11)$$

In the upper line of the above expression, the first term is the contribution from the intrinsic orbital moment of each Bloch electron, and the second term comes from the explicit field dependence of the density of states and the resulting change in the Fermi volume in Eq. (8). We expect this effect to be important in ferromagnetic materials with strong spin-orbit coupling.

Gat and Avron obtained an equivalent result for the special case of the Hofstadter model [24]. Our derivation provides a more general formula that is applicable to other systems. Following the discussions on band insulators in our first example, there will be a discontinuity of the orbital magnetization if the integral of the Berry curvature over the Brillouin zone, or the anomalous Hall conductivity, is nonzero and quantized. Depending on the direction of the field, the chemical potential μ_0 in the above formula should be taken at the top of the valence bands or the bottom of the conduction bands. The size of the discontinuity is given by the quantized anomalous Hall conductivity times E_g/e, where E_g is the energy gap. For insulators with zero Chern numbers, the orbital magnetization can be directly evaluated from Wannier functions, with results consistent with our general formula [25]. Our general formula can also be derived from a full quantum mechanical linear response analysis [26].

The central result of this Letter, Eq. (3), can be extended to the more general case when Berry curvature includes the components of $\vec{\Omega}^{\boldsymbol{kr}}$ as well as $\vec{\Omega}^{\boldsymbol{kk}}$ and $\vec{\Omega}^{\boldsymbol{rr}}$ [9]. In this case, we introduce the Berry curvature in phase space,

$$\vec{\Omega} = \begin{pmatrix} \vec{\Omega}^{\boldsymbol{rr}} & \vec{\Omega}^{\boldsymbol{rk}} \\ \vec{\Omega}^{\boldsymbol{kr}} & \vec{\Omega}^{\boldsymbol{kk}} \end{pmatrix}, \quad (12)$$

where each block is a 3×3 matrix; $\vec{\Omega}^{\boldsymbol{rk}} = -(\vec{\Omega}^{\boldsymbol{kr}})^T$. The phase-space density of states then reads,

$$D = (2\pi)^{-d}\sqrt{\det(\vec{\Omega} - \vec{\mathbf{J}})}. \quad (13)$$

with

$$\vec{\mathbf{J}} = \begin{pmatrix} 0 & \vec{1} \\ -\vec{1} & 0 \end{pmatrix}.$$

In the special case of electromagnetic perturbations with $\vec{\Omega}^{\boldsymbol{kk}}_{ab} = \epsilon_{abc}\Omega_c$, $\vec{\Omega}^{\boldsymbol{rr}}_{ab} = -(e/\hbar)\epsilon_{abc}B_c$ and $\vec{\Omega}^{\boldsymbol{kr}} = 0$, it reduces to (3). On the other hand, when either $\vec{\Omega}^{\boldsymbol{kk}}$ or $\vec{\Omega}^{\boldsymbol{rr}}$ vanishes, it has a simpler form

$$D = (2\pi)^{-d}\det(\vec{1} - \vec{\Omega}^{\boldsymbol{rk}}). \quad (14)$$

This result has found application in the study of spin-force induced charge-Hall effect [27].

Finally, we show how the density of states emerges naturally in the effective quantum mechanics of Bloch electrons. Although our system is not canonical, it can nevertheless be quantized following a standard procedure developed for nonholonomic systems with second class constraints [28,29]. First, one redefines the Poisson bracket $\{f, g\}^* = (\partial f/\partial\xi^a)M_{ab}(\partial g/\partial\xi^b)$, where ξ^a are the components of phase-space coordinates $\boldsymbol{\xi} \equiv (\boldsymbol{r}, \boldsymbol{k})$ and $\vec{\mathbf{M}} = (\vec{\boldsymbol{\Omega}} - \vec{\mathbf{J}})^{-1}$. Our equations of motion (1) can then be written as $\dot{\xi}^a = \{\xi^a, \varepsilon\}^*$, where the energy $\varepsilon(\boldsymbol{\xi})$ plays the role as the Hamiltonian function. Then, one promotes the Poisson brackets into quantum commutators:

$$[\hat{\xi}^a, \hat{\xi}^b] = \mathrm{i}M_{ab}, \quad (15)$$

where $\hat{\xi}^a$ is the quantum operator corresponding to the phase-space coordinates. It then follows that a phase-space point acquires a minimal uncertainty volume given by [30]

$$\min\left(\prod_a \Delta\xi^a\right) = 2^{-d}[\det(\vec{\boldsymbol{\Omega}} - \vec{\mathbf{J}})]^{-1/2}. \quad (16)$$

This can be understood as the phase-space volume occupied by a single quantum state, therefore Eq. (13), which is proportional to the reciprocal of this volume, can naturally be regarded as the semiclassical expression for the number of quantum states per unit volume in the phase space.

Equation (15) presents the effective quantum mechanics of Bloch electrons. As a demonstration for the validity of the quantization scheme as well as the quantum effect of the phase-space density of states, we consider a simple toy model of a two-dimensional electron system with a constant Berry curvature, subjected to a uniform magnetic field. The commutators read,

$$\begin{aligned}&[\hat{x}, \hat{y}] = \mathrm{i}\frac{\Omega}{1 + (e/\hbar)B\Omega}, \qquad [\hat{k}_x, \hat{k}_y] = -\mathrm{i}\frac{(e/\hbar)B}{1 + (e/\hbar)B\Omega}, \\ &[\hat{x}, \hat{k}_x] = [\hat{y}, \hat{k}_y] = \mathrm{i}\frac{1}{1 + (e/\hbar)B\Omega}. \end{aligned} \quad (17)$$

In the absence of the Berry curvature, we reduce the problem to a known case with the familiar nontrivial commutator $[\hat{k}_x, \hat{k}_y] = -\mathrm{i}(e/\hbar)B$. In the absence of the B field, we have the nontrivial commutator $[\hat{x}, \hat{y}] = \mathrm{i}\Omega$ discussed extensively in the literature on noncommutative geometry. It is interesting to see that in the presence of both fields, we do not just have a combination of these nontrivial commutators. Instead, we have a nontrivial density of states which enters into all of the commutators.

Assuming $\varepsilon(\boldsymbol{k}) = \hbar^2\boldsymbol{k}^2/2m$, the system can be solved algebraically to yield the energy spectrum and degeneracy. We found that the spectrum consists of a set of Landau levels with the renormalized cyclotron frequency $\omega_c = \omega_c^0/[1 + (e/\hbar)B\Omega]$, where $\omega_c^0 = eB/m$ is the usual cyclotron frequency [31]. At the same time, it is more important to note that each Landau level still has the same degeneracy of eB/h as in the absence of the Berry curvature. It is known that this degeneracy is directly related to the quantized Hall conductance e^2/h for a filled Landau level [32]. Had the density of states not entered in the commutators, the Landau level degeneracy would be modified, violating the topological requirement that the Hall conductance for a filled Landau level is quantized.

Before closing, we note that the phase-space density of states also enters naturally in the alternative quantization scheme with the Feynman path integral. The S matrix is calculated by [28]

$$\langle \mathrm{out}|S|\mathrm{in}\rangle = \int \prod_t [D(\boldsymbol{\xi})d\boldsymbol{\xi}] \exp\left[\frac{\mathrm{i}}{\hbar}\int L dt\right]. \tag{18}$$

where L is the Lagrangian for our system [9],

$$L = \frac{1}{2}\dot{\xi}^a J_{ab}\xi^b - \varepsilon(\boldsymbol{\xi}) + \dot{\xi}^a \mathcal{A}_a(\boldsymbol{\xi}) \tag{19}$$

with $\mathcal{A}_a(\boldsymbol{\xi}) \equiv \mathrm{i}\langle u(\boldsymbol{\xi})|\nabla_\alpha u(\boldsymbol{\xi})\rangle$ being the phase-space gauge potentials associated with the Berry curvature field $\vec{\vec{\boldsymbol{\Omega}}}$.

In summary, we have found a Berry phase correction to the phase-space density of states for Bloch electrons. This correction emerges naturally in both semiclassical and quantum mechanics of Bloch electrons, and has profound effects on the equilibrium and transport properties. Because of the fundamental change introduced by this correction, it could have important implications on other aspects of condensed matter physics, such as the Fermi liquid theory. For instance, in the presence of a magnetic field, interaction between electrons can change the Fermi-sea volume by modifying the Berry curvature and thus the phase-space density of states.

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$$\frac{df_n}{dt} \equiv \left(\frac{\partial}{\partial t} + \dot{\boldsymbol{r}}\cdot\nabla_{\boldsymbol{r}} + \dot{\boldsymbol{k}}\cdot\nabla_{\boldsymbol{k}}\right) f_n(\boldsymbol{k}, \boldsymbol{r}, t) = \left(\frac{\partial f_n}{\partial t}\right)_{\mathrm{coll}}.$$

The right-hand side denotes the collision contribution.
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