Editors' Suggestion

Intrinsic nonlinear Hall effect beyond Bloch geometry

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The theory of the intrinsic Hall effect, both linear and nonlinear, is rooted in a geometry which is defined in the Bloch-vector parameter space; the formal expressions are mostly derived from semiclassical concepts. When disorder and interaction are considered, there is no Bloch vector to speak of; one needs a more general quantum geometry, defined in a different parameter space. The nonlinear Hall effect is a fundamental geometric response of the many-body ground state, not a band-structure peculiarity. The higher-level geometrical formulation of the intrinsic Hall effect provides very compact expressions, which have the additional virtue—in the Bloch special case—of yielding the known results in a straightforward way: The logic is not concealed by the algebra.

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I. INTRODUCTION

The second-order dc conductivity tensor comprises three terms which—in a semiclassical treatment—scale as the zeroth, first, and second powers of the transport lifetime τ. The first two terms are of the Hall kind, i.e., the induced current is normal to the electric field, and their main entries are quantum-geometrical entities: Berry connections and curvatures in Bloch space [1–5]. The main focus in the present work is on the τ^0 term, which—to the best of the author's knowledge—was discovered in Ref. [1], where it was attributed to the field-induced positional shift of Bloch electrons; it will be indicated as $\sigma^{(\mathrm{ps})}_{\alpha\beta\gamma}$ in the following. This term has received much attention in recent times [6–11]; it is shown here that it is not a peculiarity of Bloch electrons: It is a fundamental geometric response of the many-body ground state.

I adopt here an exact quantum-mechanical framework, where by *exact* I mean that I address a nonrelativistic system of interacting electrons and static classical nuclei, possibly in a disordered configuration. No relaxation time τ can enter the theory: In fact, the concept itself does not make sense beyond the semiclassical theory. In the present framework, the response functions are causal but nondissipative: Therefore, the induced current grows with some powers of time. At first order, the intrinsic Hall current is t^0, and the Drude current is t^1 (the electrons undergo free acceleration). At second order, one expects three terms, namely, t^0, t^1, and t^2, in one-to-one correspondence to the three semiclassical terms, having the same symmetry selection rules: The t^0 and t^2 terms require breaking of time-reversal (T) symmetry in addition to breaking of inversion symmetry [5]. While for the t^1 and t^2 terms a many-body formulation exists [12,13], the t^0 term remained elusive so far. Its expression is provided here; for the sake of completeness, the other terms are also addressed.

The parameter space of conventional quantum geometry is defined by the Bloch vector $\mathbf{k}$, and the state vectors are the cell-periodic $\mathbf{k}$-dependent Bloch orbitals; the observables are Fermi-volume integrals. Here, instead, I adopt the many-body formulation of quantum geometry [14–18], where the role of the Bloch vector is played instead by the flux $\boldsymbol{\kappa}$ entering the kinetic term of the many-body Hamiltonian. The main entry of $\sigma^{(\mathrm{ps})}_{\alpha\beta\gamma}$ is cast here as a positional-shift tensor: a Berry curvature of a hybrid kind, whose variables are the flux and the electric field. Geometrical tensors of this kind may appear exotic in the context of conductivities; instead, they are the main entries in the theory of polarization (formerly called *modern*) [19] and of other observables [20].

Many-body quantum geometry addresses, in principle, even systems with disorder and correlation but has the additional virtue that it allows for compact and very transparent notations; all geometrical formulas can be easily converted when needed—e.g., for density functional theory (DFT) implementation—in their more prolix Bloch counterparts. This is shown here in detail for the three terms of second-order conductivity.

II. EXACT THEORY

The flux-dependent many-body Hamiltonian was introduced by Kohn [21]; the most general nonrelativistic Hamiltonian, also accounting for the absence of T symmetry, reads

$$\hat{H} = \frac{1}{2m}\sum_{i=1}^{N}\left[\mathbf{p}_i + \frac{e}{c}\mathbf{A}(\mathbf{r}_i) + \hbar\boldsymbol{\kappa}\right]^2 + \hat{V}. \qquad (1)$$

It addresses a system of N d-dimensional electrons in a cubic box of volume L^d. The flux $\boldsymbol{\kappa}$ is a vector potential

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cast in inverse-length units, constant in space; the potential $\hat{V}$ includes the one-body potential (possibly disordered) and electron-electron interaction.

The system is macroscopically homogeneous; the $\boldsymbol{\kappa}$-dependent eigenstates $|\Psi_n\rangle$ are normalized to one in the hypercube of volume L^{Nd}. The thermodynamic limit $N \to \infty$, $L \to \infty$, and N/L^d constant is understood throughout this work; the $\boldsymbol{\kappa}$ derivatives must be evaluated first and the $L \to \infty$ limit taken afterwards [21–23].

Periodic boundary conditions (PBCs) are assumed: The many-body wave functions are periodic with period L over each electron coordinate $\mathbf{r}_i$ independently; the potential $\hat{V}$ enjoys the same periodicity. The vector potential in Eq. (1) summarizes all intrinsic T-breaking terms as, e.g., those due to a coupling to a background of local moments; even $\mathbf{A}(\mathbf{r})$ obeys PBCs. The vector potential could even account for a macroscopic $\mathbf{B}$ field, provided that it is commensurate and that the PBCs are modified accordingly [24]. A time-independent $\boldsymbol{\kappa}$ amounts to a pure gauge transformation, but—as shown by Kohn—owing to PBCs, the gauge-invariance is broken: The eigenvalues E_n depend on $\boldsymbol{\kappa}$ in the metallic case, while they are $\boldsymbol{\kappa}$ independent in insulators [21–23,25].

Kohn's Hamiltonian has the virtue that the macroscopic current density can be cast as

$$\hat{\mathbf{j}} = -\frac{e}{L^d}\hat{\mathbf{v}}, \quad \hat{\mathbf{v}} = \frac{1}{\hbar}\partial_{\boldsymbol{\kappa}}\hat{H}; \tag{2}$$

furthermore, a flux linear in time corresponds to perturbing the system with the dc field

$$\boldsymbol{\mathcal{E}} = -\frac{\hbar}{e}\dot{\boldsymbol{\kappa}}. \tag{3}$$

If $\boldsymbol{\mathcal{E}}$ is adiabatically turned on at time $t = 0$, the current response is

$$\frac{\partial j_\alpha(t)}{\partial \mathcal{E}_\beta} = \frac{e^2}{\hbar L^d}\left[-\Omega(\kappa_\alpha, \kappa_\beta) + \frac{t}{\hbar}\frac{\partial^2 E_0}{\partial\kappa_\alpha \partial\kappa_\beta}\right], \tag{4}$$

where $\Omega(\kappa_\alpha, \kappa_\beta) = -2\,\mathrm{Im}\,\langle\partial_{\kappa_\alpha}\Psi_0|\partial_{\kappa_\beta}\Psi_0\rangle$ is the many-body Berry curvature [16].

The linear conductivity is obtained by setting $\boldsymbol{\kappa} = 0$: The two terms in Eq. (4) are t^0 (transverse) and t^1 (longitudinal), respectively. The longitudinal term in Eq. (4) is rewritten as

$$\partial_t j(t) = \frac{D_{\alpha\beta}}{\pi}\mathcal{E}_\beta, \quad D_{\alpha\beta} = \frac{\pi e^2}{\hbar^2 L^d}\frac{\partial^2 E_0}{\partial\kappa_\alpha\partial\kappa_\beta}, \tag{5}$$

where $D_{\alpha\beta}$ at $\boldsymbol{\kappa} = 0$ is known as the Drude weight [21–23]; the Fourier transform of Eq. (5) is

$$-i\omega j_\alpha(\omega) = \frac{D_{\alpha\beta}}{\pi}\mathcal{E}_\beta(\omega), \tag{6}$$

whose causal inversion yields

$$j_\alpha(\omega) = \frac{D_{\alpha\beta}}{\pi}\frac{i}{\omega + i\eta}\mathcal{E}_\beta(\omega), \quad \eta = 0^+. \tag{7}$$

The famous Kohn's expression [21] for the Drude term in longitudinal conductivity is

$$\sigma^{(\mathrm{Drude})}_{\alpha\beta}(\omega) = \frac{D_{\alpha\beta}}{\pi}\frac{i}{\omega + i\eta} = D_{\alpha\beta}\left[\delta(\omega) + \frac{i}{\pi\omega}\right]. \tag{8}$$

Both $\Omega(\kappa_\alpha, \kappa_\beta)$ and $D_{\alpha\beta}$ acquire a time dependence when expanded to the next order in $\boldsymbol{\kappa}$

$$\begin{aligned}\partial_t\Omega(\kappa_\alpha, \kappa_\beta) &= \partial_{\kappa_\gamma}\Omega(\kappa_\alpha, \kappa_\beta)\dot{\kappa}_\gamma,\\ \partial_t D_{\alpha\beta} &= \frac{e^2}{\hbar^2 L^d}\frac{\partial^3 E_0}{\partial\kappa_\alpha\partial\kappa_\beta\partial\kappa_\gamma}\dot{\kappa}_\gamma.\end{aligned} \tag{9}$$

Therefore, owing to Eqs. (3) and (4), one has

$$\begin{aligned}\frac{\partial^2 j_\alpha(t)}{\partial\mathcal{E}_\beta\partial\mathcal{E}_\gamma} = \frac{\partial^2 j_\alpha(0)}{\partial\mathcal{E}_\beta\partial\mathcal{E}_\gamma} &+ \frac{e^3 t}{\hbar^2 L^d}\partial_{\kappa_\gamma}\Omega(\kappa_\alpha, \kappa_\beta)\\ &- \frac{e^3 t^2}{\hbar^3 L^d}\frac{\partial^3 E_0}{\partial\kappa_\alpha\partial\kappa_\beta\partial\kappa_\gamma},\end{aligned} \tag{10}$$

where all derivatives are evaluated at $\boldsymbol{\kappa} = 0$.

The three terms are parsed by their t dependence: t^0, t^1, and t^2, respectively. The t^2 term accounts for the quadratic Drude conductivity and coincides with the expression found by Watanabe and Oshikawa [12]; the t^1 term is the many-body formulation of the nonlinear Hall effect previously established by the present author [13]. The t^0 term is the main focus of this work: As said above, it will be indicated as $\sigma^{(\mathrm{ps})}_{\alpha\beta\gamma}$.

To address this term, it is expedient to make the Hamiltonian $\boldsymbol{\mathcal{E}}$ dependent in the scalar-potential gauge, i.e.,

$$\hat{V} \to \hat{V}_0 + e\boldsymbol{\mathcal{E}}\cdot\hat{\mathbf{r}}, \quad \hat{\mathbf{r}} = \sum_{i=1}^{N}\mathbf{r}_i; \tag{11}$$

notice that $\boldsymbol{\kappa}$ and $\boldsymbol{\mathcal{E}}$ are independent variables. The $\hat{\mathbf{r}}$ operator does not make sense within PBCs, but its off-diagonal elements are defined as

$$\langle\Psi_0|\,\hat{r}_\alpha\,|\Psi_n\rangle = i\langle\Psi_0|\partial_{\kappa_\alpha}\Psi_n\rangle = -i\hbar\frac{\langle\Psi_0|\,\hat{v}_\alpha\,|\Psi_n\rangle}{E_0 - E_n}. \tag{12}$$

The t^0 current obtains from the $\boldsymbol{\mathcal{E}}$ derivative of the first term in Eq. (4). Incidentally, this is consistent with some alternative nomenclature [5,10]: *Berry-curvature polarizability*. The concept of field-induced positional shift—within the present beyond-Bloch geometry—can be understood as follows. The electronic term in the polarization of an insulator can be expressed as

$$\mathbf{P}^{(\mathrm{el})} = -\frac{ie}{L^d}\langle\Psi_0|\partial_{\boldsymbol{\kappa}}\Psi_0\rangle, \quad \boldsymbol{\kappa} = 0, \tag{13}$$

augmented with a prescription for fixing the gauge [26]; the position of the electrons—i.e., the Berry connection $\mathcal{A}_\alpha = i\langle\Psi_0|\partial_{\kappa_\alpha}\Psi_0\rangle$—is ill defined unless the gauge is fixed, yet the shift—i.e., its $\boldsymbol{\mathcal{E}}$ derivative—is gauge invariant and well defined, both in insulators and metals.

To make contact with the existing literature, I define the positional-shift tensor as the derivative with respect to $e\boldsymbol{\mathcal{E}}$, i.e.,

$$\begin{aligned}\mathcal{G}_{\alpha\gamma} &= \frac{1}{e}\partial_{\mathcal{E}_\gamma}\mathcal{A}_\alpha = \frac{i}{e}\big\langle\partial_{\mathcal{E}_\gamma}\Psi_0|\partial_{\kappa_\alpha}\Psi_0\big\rangle + \mathrm{c.c.}\\ &= \frac{i}{e}\Big(\big\langle\partial_{\mathcal{E}_\gamma}\Psi_0|\partial_{\kappa_\alpha}\Psi_0\big\rangle - \big\langle\partial_{\kappa_\alpha}\Psi_0|\partial_{\mathcal{E}_\gamma}\Psi_0\big\rangle\Big)\\ &= \frac{1}{e}\Omega\big(\mathcal{E}_\gamma, \kappa_\alpha\big).\end{aligned} \tag{14}$$

Here, $\mathcal{G}_{\alpha\gamma}$ is therefore cast as an hybrid geometrical tensor of the same family as those entering polarization theory [19]. In

fact, it is shown elsewhere [20] that the curvature $\Omega(\mathcal{E}_\gamma, \kappa_\alpha)$, evaluated at $\boldsymbol{\kappa} = 0$, yields the linear polarizability of an insulator

$$\chi_{\alpha\gamma} = -\frac{e}{L^d}\Omega\big(\mathcal{E}_\gamma, \kappa_\alpha\big). \tag{15}$$

Notice that $\Omega(\mathcal{E}_\gamma, \kappa_\alpha)$ is antisymmetric for the exchange $\mathcal{E}_\gamma \leftrightarrow \kappa_\alpha$ and is symmetric for the exchange $\gamma \leftrightarrow \alpha$.

In the metallic case—considered here—$\chi_{\alpha\gamma}$ diverges, yet $\mathcal{G}_{\alpha\gamma}$ retains a physical meaning. In fact, the imaginary part of the longitudinal linear conductivity in a metal is

$$\mathrm{Im}\,\sigma_{\alpha\gamma}(\omega) = \frac{D_{\alpha\gamma}}{\pi\omega} + \mathrm{Im}\,\sigma^{(\mathrm{regular})}_{\alpha\gamma}(\omega), \tag{16}$$

where the regular term is linear in ω; in insulators, the expression is the same but with a vanishing Drude term. It is then easy to show that, in both insulators and metals, one has

$$\lim_{\omega\to 0}\frac{\mathrm{Im}\,\sigma^{(\mathrm{regular})}_{\alpha\gamma}(\omega)}{\omega} = \frac{e}{L^d}\Omega\big(\mathcal{E}_\gamma, \kappa_\alpha\big). \tag{17}$$

In metals, this regular term is not observable because it is obliterated by the divergent imaginary part of $\sigma^{(\mathrm{Drude})}_{\alpha\beta}(\omega)$; it manifests itself at second order via its $\boldsymbol{\kappa}$ derivatives.

By evaluating $\mathcal{G}_{\alpha\gamma}$ in the parallel-transport gauge, one gets (again both in metals and insulators)

$$\begin{aligned}\mathcal{G}_{\alpha\gamma} &= -2\,\mathrm{Re}\sum_{n\neq 0}\frac{\langle\Psi_0|\,\hat{r}_\alpha\,|\Psi_n\rangle\langle\Psi_n|\,\hat{r}_\gamma\,|\Psi_0\rangle}{E_0 - E_n}\\ &= -2\hbar^2\,\mathrm{Re}\sum_{n\neq 0}\frac{\langle\Psi_0|\,\hat{v}_\alpha\,|\Psi_n\rangle\langle\Psi_n|\,\hat{v}_\gamma\,|\Psi_0\rangle}{(E_0 - E_n)^3}.\end{aligned} \tag{18}$$

Finally, the sought-for expression for the positional-shift conductivity is

$$\sigma^{(\mathrm{ps})}_{\alpha\beta\gamma} = \frac{e^3}{\hbar L^d}\big(\,\partial_{\kappa_\alpha}\mathcal{G}_{\beta\gamma} - \partial_{\kappa_\beta}\mathcal{G}_{\alpha\gamma}\,\big); \tag{19}$$

even this expression is evaluated at $\boldsymbol{\kappa} = 0$. Equations (18) and (19) are reminiscent of the corresponding Bloch expressions in the literature: Eqs. (24) and (25) below.

III. KOHN-SHAM THEORY

In the special case of a crystalline system of noninteracting electrons, the $L \to \infty$ limit can be performed analytically thanks to translational symmetry: All intensive observables are then expressed as Fermi-volume integrals.

In a Kohn-Sham framework, the ground-state $|\Psi_0\rangle$ is a Slater determinant of single-particle orbitals. Whenever an operator $\hat{O}$ is the sum of one-body operators $\tilde{O}$, its expectation value is given by the sum of the expectation values of $\tilde{O}$ over the occupied orbitals. Here, the orbitals are the Bloch orbitals $|\psi_{j\mathbf{k}}\rangle = \exp(i\mathbf{k}\cdot\mathbf{r})|u_{j\mathbf{k}}\rangle$ with eigenvalues $\epsilon_{j\mathbf{k}}$, normalized in the crystal cell of volume V_{cell}—they are normalized differently from $|\Psi_0\rangle$. The formulas below are given per spin channel, for spinless electrons in jargon.

Quantum geometry deals with $|u_{j\mathbf{k}}\rangle$, which are eigenstates of the Kohn-Sham $\boldsymbol{\kappa}$-dependent Hamiltonian

$$H_{\mathbf{k}} = \frac{1}{2m}\Big[\mathbf{p} + \frac{e}{c}\mathbf{A}(\mathbf{r}) + \hbar\mathbf{k} + \hbar\boldsymbol{\kappa}\Big]^2 + V(\mathbf{r}); \tag{20}$$

hence, a $\boldsymbol{\kappa}$ derivative evaluated at $\boldsymbol{\kappa} = 0$ coincides with a $\mathbf{k}$ derivative. The one-body version of the curvature of Eq. (14) is then

$$G_{j\alpha\gamma} = \frac{i}{e}\big(\langle\partial_{\mathcal{E}_\gamma}u_{j\mathbf{k}}|\partial_{k_\alpha}u_{j\mathbf{k}}\rangle - \langle\partial_{k_\alpha}u_{j\mathbf{k}}|\partial_{\mathcal{E}_\gamma}u_{j\mathbf{k}}\rangle\big), \tag{21}$$

and the positional-shift conductivity is

$$\sigma^{(\mathrm{ps})}_{\alpha\beta\gamma} = \frac{e^3}{\hbar V_{\mathrm{cell}}}\sum_{j\mathbf{k}}\theta\big(\epsilon_{\mathrm{F}} - \epsilon_{j\mathbf{k}}\big)\big(\,\partial_{k_\alpha}G_{j\beta\gamma} - \partial_{k_\beta}G_{j\alpha\gamma}\,\big), \tag{22}$$

where ϵ_{F} is the Fermi energy. In the $L \to \infty$ limit,

$$\frac{1}{V_{\mathrm{cell}}}\sum_{\mathbf{k}} \to \int_{\mathrm{BZ}}\frac{d\mathbf{k}}{(2\pi)^d}\,, \tag{23}$$

$$\sigma^{(\mathrm{ps})}_{\alpha\beta\gamma} = \frac{e^3}{\hbar}\sum_j\int_{\mathrm{BZ}}\frac{d\mathbf{k}}{(2\pi)^d}\,\theta\big(\epsilon_{\mathrm{F}} - \epsilon_{j\mathbf{k}}\big)\big(\,\partial_{k_\alpha}G_{j\beta\gamma} - \partial_{k_\beta}G_{j\alpha\gamma}\,\big). \tag{24}$$

This is identical to the expressions in the semiclassical literature [1,5,7] once $G_{j\alpha\gamma}$ is evaluated in its equivalent sum-over-states form

$$G_{j\alpha\gamma} = -2\hbar\,\mathrm{Re}\sum_{j'\neq j}\frac{\langle u_{j\mathbf{k}}|\,v_\alpha\,|u_{j'\mathbf{k}}\rangle\langle u_{j'\mathbf{k}}|\,v_\gamma\,|u_{j\mathbf{k}}\rangle}{(\epsilon_{j\mathbf{k}} - \epsilon_{j'\mathbf{k}})^3}. \tag{25}$$

It is worth observing that Eq. (21) is better suited to computational implementation: In fact, modern computer codes implement density-functional perturbation theory [27], which directly evaluates $|\partial_{\boldsymbol{\mathcal{E}}}u_{j\mathbf{k}}\rangle$ more efficiently than performing sums over states.

When all three terms are included, the quadratic Hall conductivity in the ω domain is

$$\begin{aligned}\sigma_{\alpha\beta\gamma}(\omega) =& \frac{e^3}{\hbar}\sum_j\int_{\mathrm{BZ}}\frac{d\mathbf{k}}{(2\pi)^d}\,\theta(\epsilon_{\mathrm{F}} - \epsilon_{j\mathbf{k}})\\ &\times\Bigg[\partial_{k_\alpha}G_{j\beta\gamma} - \partial_{k_\beta}G_{j\alpha\gamma} + \frac{1}{\hbar}\partial_{k_\gamma}\tilde{\Omega}_j\big(k_\alpha, k_\beta\big)\frac{i}{\omega + i\eta}\\ &+ \frac{1}{\hbar^2}\frac{\partial^3\epsilon_{j\mathbf{k}}}{\partial k_\alpha\partial k_\beta\partial k_\alpha}\left(\frac{i}{\omega + i\eta}\right)^2\Bigg],\end{aligned}$$

where $\tilde{\Omega}_j(k_\alpha, k_\beta)$ is the Berry curvature of band j [19].

Finally, when the infinitesimal η is heuristically replaced with the inverse of a relaxation time τ, one gets the semiclassical result in the form reported, e.g., in Ref. [5]. The zero-temperature Fermi function $\theta(\epsilon_{\mathrm{F}} - \epsilon)$ can also be heuristically replaced by its finite-temperature counterpart.

IV. DISCUSSION

I have shown that both terms of the intrinsic Hall conductivity have a geometrical formulation beyond the Bloch setting, which also applies to a larger class of materials: those where disorder and interaction play an important role. When the system is crystalline, the theory is formulated à la Kohn-Sham; the resulting expressions coincide with the semiclassical ones in the $\tau \to \infty$ limit and for zero temperature. This agrees with the common wisdom that the semiclassical

approximation is exact when addressing dc transport properties in a crystalline system of noninteracting electrons. In this context, the semiclassical approximation is not an approximation after all.

The major result of this work is the exact many-body expression for $\sigma^{(\mathrm{ps})}_{\alpha\beta\gamma}$, given as the curl in the $\boldsymbol{\kappa}$ variable (the flux) of a positional-shift tensor: a Berry curvature whose variables are the flux and the electric field, having an equivalent sum-over-states expression. Hybrid geometrical quantities of the same family are at the heart of polarization theory [19]. This tensor yields indeed the linear static polarizability of a metal, where the divergent Drude term has been discounted.

When the system is crystalline, the main entry of $\sigma^{(\mathrm{ps})}_{\alpha\beta\gamma}$ is an analogous band curvature whose variables are instead the Bloch vector and the electric field. The equivalent sum-over-states form coincides with what one finds in the literature [1,5,7], while the compact curvature form is possibly new. The latter, as explained above, could be computationally more appealing.

Finally, a short digression about the extrinsic effects: In the case of the linear Hall conductivity, they come in two kinds: τ^0 (called *side jump*) and τ^1 (called *skew scattering*) [28], while the terms *intrinsic* and *geometrical* are used synonymously (the Bloch geometry of the pristine crystal is intended). When the disordered system is addressed by means of a supercell (ideally in the $L \to \infty$ limit), the effects of disorder become by construction intrinsic; we previously argued in Ref. [29] that the geometrical conductivity of the disordered system includes the side-jump contributions in addition to the pristine-crystal geometrical response. A similar statement holds for the quadratic Hall conductivity: The τ^0 extrinsic effects become intrinsic and included in the present generalized geometrical formulation of $\sigma^{(\mathrm{ps})}_{\alpha\beta\gamma}$.

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DATA AVAILABILITY

No data were created or analyzed in this study.

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