

Disorder-Induced Delocalization in Magic-Angle Twisted Bilayer Graphene

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Flat bands in moiré systems are exciting new playgrounds for the generation and study of exotic many-body physics phenomena in low-dimensional materials. Such physics is attributed to the vanishing kinetic energy and strong spatial localization of the flat-band states. Here, we use numerical simulations to examine the electronic transport properties of such flat bands in magic-angle twisted bilayer graphene in the presence of disorder. We find that while a conventional downscaling of the mean free path with increasing disorder strength occurs at higher energies, in the flat bands the mean free path can actually increase with increasing disorder strength. This phenomenon is also captured by the disorder-dependent quantum metric, which is directly linked to the ground state localization. This disorder-induced delocalization suggests that weak disorder may have a strong impact on the exotic physics of magic-angle bilayer graphene and other related moiré systems.

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Introduction—Moiré systems such as magic-angle twisted bilayer graphene (MATBLG) have proven to be ideal structures for generating flat bands and strongly correlated physics [1–5]. The unexpected and spectacular observation of superconductivity and Mott insulating phases in MATBLG has been attributed to the vanishing kinetic energy and real-space state localization [6]. This triggers a dominant contribution of the Coulomb interaction, which generates a wealth of emerging many-body physics phenomena [1–18]. Other studies have probed the robustness of this behavior and its relationship with the emerging superlattice potential, which surprisingly occurs for periodic as well as aperiodic (such as quasicrystalline order) systems [19–21].

The role of the moiré superlattice potential in producing such “anomalous electronic spatial localization” and vanishing velocity of the flat bands is now well documented [22–24]. However to date, relatively little is known about the impact of superimposed structural (i.e., twist angle) [25] and electrostatic disorder, which are ubiquitous in real samples and likely play a role in the stability of the flat-band-induced localization and the resilience of strong correlation effects. In particular, the impact of disorder on the transport and localization properties remains to be examined. It has been shown that structural relaxation (which will be substrate-dependent) leads to a renormalized dispersion of the flat bands that may then impact the transport properties [26–28]. Additionally, in the context of general flat-band physics, several theoretical studies

suggest that disorder could produce exotic phenomena such as inverse Anderson localization (observed experimentally in ultracold atoms) [29], supermetallicity [30,31], or deviation from the usual scaling theory of localization [32]. It is thus important to study the impact of disorder in such systems, as some amount of disorder is present in all fabricated samples [33–35].

In the last decade, topological materials have been widely studied due to their nontrivial ground state [36–39]. A particularly fundamental quantity is the quantum geometric tensor, whose imaginary part is the Berry curvature, Ω_{ij} , which drives most studied topological effects, while its real part (g_{ij}) is known as the quantum metric (QM) [40]. Recently g_{ij} has been gaining much attention due to its emergence in linear [41–45] and nonlinear response theory [46–49], although a direct link with an observable remains challenging [41]. In various flat-band systems, the QM seems to play an important role [50–52], especially in the interacting phase [53–57]. In addition, the QM is at the heart of the modern theory of insulators [58,59], where it connects to the real part of the dipole-dipole fluctuations of the ground state, $\text{Re}\langle\hat{r}_i\hat{r}_j\rangle_c = g_{ij}$, indicating if the ground state is metallic (when the fluctuations diverge in the thermodynamic limit) or an insulator. Therefore g_{ij} is directly linked to the electronic localization of the ground state [60,61]. However, despite recent advances, the behavior of the QM in the presence of disorder is still an open question, particularly for disordered MATBLG.

In this Letter, we use numerical simulations to investigate the impact of disorder on electronic transport in MATBLG. We implement a realistic tight-binding Hamiltonian that well captures the weakly dispersive flat bands in this system. With a rotation angle of $\sim 1.1^\circ$ between the two layers, the moiré superlattice has a long range of periodicity, with more than 11 000 atoms in the unit cell. To account for the disorder-induced breaking of translational invariance, we employ a real-space linear-scaling quantum transport methodology that enables the simulation of disordered MATBLG systems containing several million atoms, thus allowing us to reach transport length scales that are relevant to experiments [62]. In addition, we compute the dimensionless quantum metric (from now on just the quantum metric), $\mathcal{G}_{ij} = g_{ij}/V$, which gives us a direct measure of the electronic localization of the ground state, via the Souza-Wilkens-Martin rule [63] and the polynomial expansion of the optical conductivity [62,64]. Our findings reveal a nontrivial evolution of transport characteristics for disorder strengths that do not fully suppress the flat bands. The combination of disorder-induced broadening and scattering leads to delocalization of the flat-band states, which manifests in an increase of the mean free path with increasing disorder. This is opposite to the more conventional behavior observed at higher energies away from the moiré flat bands, where stronger disorder reduces the mean free path. Such unconventional behavior disappears for disorder strong enough to break the intrinsic moiré-induced localization at the magic angle. We have seen emergence of this exotic behavior in both the quantum transport and QM calculations.

These findings suggest that weak disorder in MATBLG, by driving delocalization of the flat-band states, may have a significant impact on the relative strength of the Coulomb interaction at the heart of the exotic physics in this material. In this context, a metric for characterizing the delocalization of flat-band states may serve as an important quantity for characterizing the stability of exotic phases in such systems. In addition we expect this result to be general for other systems with flat bands.

Structural and tight-binding Hamiltonian models—To build realistic magic-angle twisted graphene superlattices, we use molecular dynamics simulations with classical potentials to relax the structures [27,65–68]. We start with uniform Bernal-stacked bilayer graphene, twisted to an angle of $\sim 1.1^\circ$, and optimize the structure until all the force components are smaller than 0.5 meV/atom. Intralayer forces are computed using the optimized Tersoff and Brenner potentials [66], whereas interlayer forces are modeled using the Kolmogorov-Crespi potentials [67,68]. The electronic properties of the twisted graphene systems are then computed using the p_z tight-binding Hamiltonian,

$$\hat{\mathcal{H}} = \sum_n \varepsilon_n |\phi_n\rangle \langle \phi_n| + \sum_{n,m} t(\vec{r}_{nm}) |\phi_n\rangle \langle \phi_m|, \quad (1)$$

where $|\phi_n\rangle$ describes the p_z orbital on carbon site n with position $\vec{r}_n$, ε_n is the electrostatic potential at carbon site n , and $\vec{r}_{nm} = \vec{r}_m - \vec{r}_n$. The hopping energies $t(\vec{r}_{nm})$ between carbon sites are given by the standard Slater-Koster expression [27,65,69,70]

$$t(\vec{r}_{nm}) = \cos^2(\phi_{nm}) V_{pp\sigma}(r_{nm}) + \sin^2(\phi_{nm}) V_{pp\pi}(r_{nm}), \quad (2)$$

where the direction cosine of $\vec{r}_{nm}$ along the z axis is $\cos(\phi_{nm}) = z_{nm}/r_{nm}$. The distance-dependent Slater-Koster parameters are [70]

$$V_{pp\pi}(r_{nm}) = V_{pp\pi}^0 \exp \left[q_\pi \left(1 - \frac{r_{nm}}{a_0} \right) \right] F_c(r_{nm}), \\ V_{pp\sigma}(r_{nm}) = V_{pp\sigma}^0 \exp \left[q_\sigma \left(1 - \frac{r_{nm}}{d_0} \right) \right] F_c(r_{nm}), \quad (3)$$

with a smooth cutoff function $F_c(r_{nm}) = [1 + \exp((r_{nm} - r_c)/\lambda_c)]^{-1}$. To model the flat electronic bands of relaxed twisted bilayer graphene at the magic angle $\sim 1.1^\circ$, the tight-binding parameters are adjusted to $V_{pp\pi}^0 = -\gamma_0 = -2.7$ eV, $V_{pp\sigma}^0 = 367.5$ meV, $q_\pi/a_0 = q_\sigma/d_0 = 22.18$ nm $^{-1}$, $a_0 = 0.1439$ nm, $d_0 = 0.33$ nm, $r_c = 0.614$ nm, and $\lambda_c = 0.0265$ nm [65]. Finally, disorder is introduced via an Anderson potential by assigning random values to the on-site energies within a uniform distribution with interval $\varepsilon_n \in [-W/2, W/2]$. We consider values of W from $\gamma_0/6$ to $2\gamma_0$, corresponding to bulk mean free paths from a few tens to a few hundred nanometers [71], covering the range typically measured in graphene on SiO $_2$ [72].

Quantum transport methodology—To investigate electronic transport in the relaxed twisted bilayer graphene structure, we use the linear-scaling quantum transport methodology detailed in Ref. [62]. Specifically, we calculate the mean square displacement of an initial electronic state $|\psi(0)\rangle$ as a function of Fermi energy and time,

$$\Delta X^2(E, t) = \frac{\langle \psi_X(t) | \delta(E - \hat{\mathcal{H}}) | \psi_X(t) \rangle}{\rho(E)}, \quad (4)$$

where $|\psi_X(t)\rangle = [\hat{X}, \hat{U}(t)] |\psi(0)\rangle$, $\hat{X}$ is the position operator, $\hat{U}(t) = \exp(-i\hat{\mathcal{H}}t/\hbar)$ is the time evolution operator, and $\rho(E) = \langle \psi(0) | \delta(E - \hat{\mathcal{H}}) | \psi(0) \rangle \langle \psi(0) | \delta(E - \hat{\mathcal{H}}) | \psi(0) \rangle$ is the density of states. The time evolution operator and the energy projection operator $\delta(E - \hat{\mathcal{H}})$ are both expanded in a numerically efficient way using Chebyshev polynomials [62,73]. Here, we use 3500 polynomials, corresponding to a Gaussian energy broadening of 23 meV. We use a time step of 10 fs for transport in the clean system and 1 fs for the disordered cases. The initial state $|\psi(0)\rangle$ is chosen to be a random-phase state, which is a standard approach for the efficient calculation of material properties over the entire

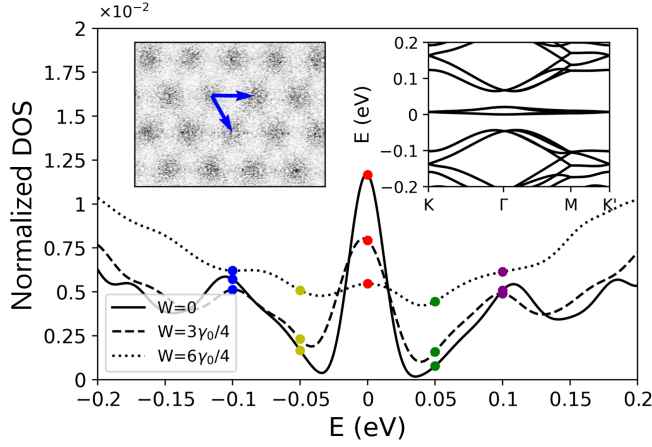


FIG. 1. Total density of states of MATBLG for disorder strengths of $W = 3\gamma_0/2$ (dotted line), $W = 3\gamma_0/4$ (dashed line), and no disorder (solid line). Left inset: local density of states (LDOS) of the clean system at charge neutrality where higher LDOS (darker zones) is concentrated in the AA moiré regions. The blue arrows indicate the moiré unit cell lattice vectors. Right inset: band structure of MATBLG, with the flat bands clearly present around $E = 0$.

Hamiltonian spectrum; see Refs. [62] and [73] for more details.

From the mean square displacement we calculate the time-dependent diffusion coefficient $D(E, t)$ and extract the mean free path $\ell(E)$ from its saturated value at long times, $D_{\max}(E)$, according to

$$D(E, t) = \frac{1}{2} \frac{d}{dt} \Delta X^2(E, t), \quad (5)$$

$$\ell(E) = \frac{2D_{\max}(E)}{v(E)}, \quad (6)$$

where $v(E)$ is the Fermi velocity of disorder-free MATBLG. In our transport simulations, a system with a 16×16 tiling of the MATBLG unit cell is considered, containing about 2.9×10^6 atoms.

Quantum metric—To compute the quantum metric we make use of the Souza-Wilkens-Martin rule [63], which relates it to the optical conductivity (via the fluctuation-dissipation theorem) as

$$\int_0^\infty \frac{d\omega}{\omega} \text{Re}\{\sigma_{xx}(\omega)\} = \frac{\pi e^2}{h} \mathcal{G}_{xx}, \quad (7)$$

where $\mathcal{G}_{xx}$ is the dimensionless quantum metric, e is the electric charge, h is the Planck constant, $\sigma(\omega)$ is the optical conductivity, and ω is the frequency. We note that $\mathcal{G}_{xx}$, which we refer to here as the quantum metric, is the k -space integral of the real part of the quantum geometric tensor. This QM is linked to the invariant part of the spread of the Wannier functions [60], $\Omega_1 = V/(2\pi)^2 \text{Tr}\{\mathcal{G}_{xx}\}$, where the trace is over the Cartesian indices. Here, a small QM relates

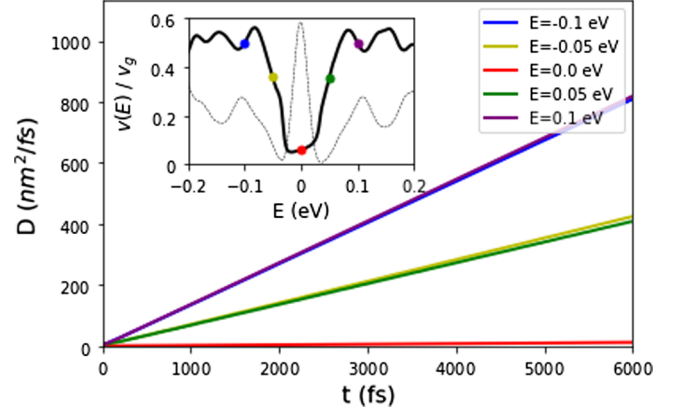


FIG. 2. Diffusion coefficient as a function of time for clean MATBLG at energies of $E = -100$ meV (blue), -50 meV (yellow), charge neutrality (red), 50 meV (green), and 100 meV (purple). Inset: the corresponding energy-dependent Fermi velocity (solid line), superimposed with the DOS of the clean case (dotted line, rescaled for clarity).

to a small Wannier spread, and thus to a strongly localized ground state. On the other hand, when the QM increases continuously with system size, it indicates delocalization of the Wannier function and the ground state. Here, we compute $\sigma(\omega)$ from the Kubo formula [74], making use of a Chebyshev polynomial expansion [64] with a broadening of 66 meV and obtain convergence of $\mathcal{G}_{xx}$ with system size and number of polynomials [75].

Electronic properties of disordered MATBLG—We first start by analyzing the impact of disorder on the electronic structure of MATBLG through its impact on the total (DOS) and local density of states (LDOS). In Fig. 1 we plot the DOS for Anderson disorder strengths of $W = 0$, $3\gamma_0/4$, and $3\gamma_0/2$. For reference, we show the band structure of the clean case in the right inset and the LDOS at charge neutrality ($E = 0$) in the left inset. The flat bands and corresponding localization of states in AA regions are well reproduced in the absence of disorder. In the main panel, the presence of a strong peak in the DOS at $E = 0$ highlights the presence of the moiré-induced flat bands. The role of disorder is to broaden and reduce this peak, which remains clearly visible at $W = 3\gamma_0/4$, before finally being washed out for $W \geq 3\gamma_0/2$, coinciding with the disappearance of AA spatial localization (see the right inset, Fig. 4, for $W = 2\gamma_0$).

Quantum transport in clean and disordered MATBLG—In Fig. 2 we plot the time evolution of the diffusion coefficient of clean MATBLG. In absence of disorder, transport is ballistic and the diffusion coefficient increases linearly with time, $D(E, t) = v^2(E)t$. This behavior is seen at all energies (different colored curves) in Fig. 2. From the slope of these curves we then extract the Fermi velocity of the MATBLG system, which we plot in the inset, relative to the velocity of single-layer graphene v_g . Note that while the Fermi velocity may not be uniform around the Fermi surface, here we are plotting its average over the Fermi

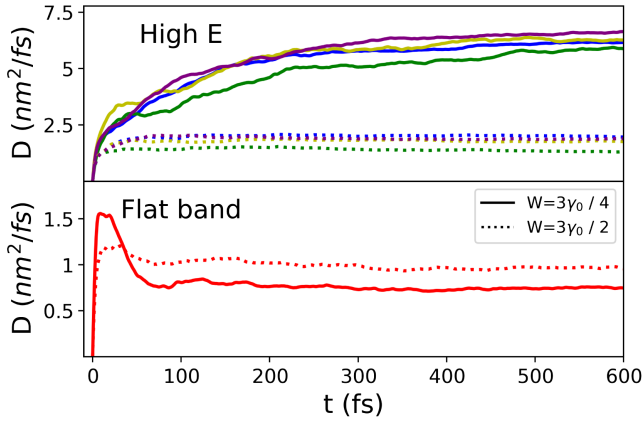


FIG. 3. Time-dependent diffusion coefficient at energies of $E = -100$ meV (blue), -50 meV (yellow), charge neutrality (red), 50 meV (green), and 100 meV (purple) for MATBLG under Anderson disorder strengths of $W = 3\gamma_0/4$ (dashed) and $W = 3\gamma_0/2$ (dotted).

surface at the indicated energies E . Here, we see that around charge neutrality the Fermi velocity is very low, $v < 0.1 v_g$, characteristic of the flat nature of the moiré bands.

Next, in Fig. 3 we examine electronic transport in disordered MATBLG. In the presence of Anderson disorder, the diffusion coefficients now saturate at long times for all energies. However, we observe a qualitative difference between transport within the flat bands compared to that at higher energies. When increasing disorder strength from $W = 3\gamma_0/4 \rightarrow 3\gamma_0/2$, D decreases by a factor of ~ 4 for all energies except at charge neutrality (red curve), where D actually increases, opposite to typical behavior. This is illustrated further in Fig. 4, where we plot the mean free path $\ell(E)$ for three different disorder strengths. In the energy range corresponding to the moiré flat bands, for weaker disorder we see a clear increase of the mean free path with increasing disorder strength, opposite to the scaling behavior at higher energies. This increase in $\ell(E)$ actually coincides with a delocalization of the LDOS around charge neutrality in the presence of disorder, as highlighted in the left and right insets of Fig. 4. Here, we note a slight electron-hole asymmetry in the mean free path, arising from an asymmetry in the band structure (inset of Fig. 1) and correspondingly in the Fermi velocity (inset of Fig. 2). A similar behavior of the electron-phonon coupling in MATBLG has been reported in Refs. [80,81].

We remark that stronger disorder will eventually suppress any remnant of the flat bands and thus reduce the mean free path following the scaling behavior $\ell \sim (\gamma_0/W)^2 a_{cc}$, where a_{cc} is the carbon-carbon spacing. This is seen when increasing disorder from $W = 3\gamma_0/2$ to $W = 2\gamma_0$ in Fig. 4.

Therefore, the observed anomalous “disorder-induced delocalization” exists over a finite range of disorder strengths, and is maintained when disorder is low enough to preserve the moiré-induced flat-band states. This effect is driven by the

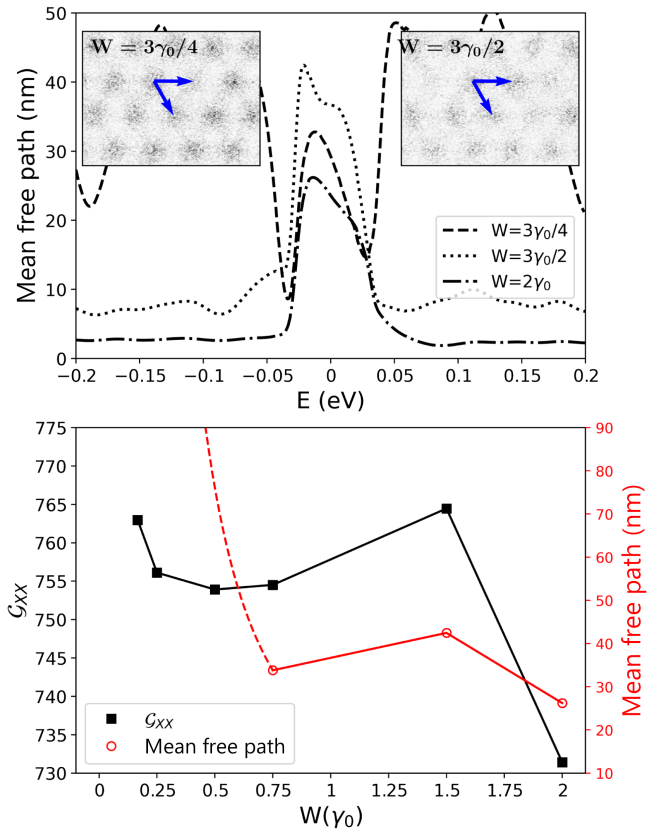


FIG. 4. Top panel: mean free path for Anderson strengths of $W = 3\gamma_0/4$ (dashed line), $W = 3\gamma_0/2$ (dotted line), and $W = 2\gamma_0$ (dash-dotted line). The insets show the LDOS at charge neutrality with disorders of $W = 3\gamma_0/4$ (left) and $W = 3\gamma_0/2$ (right). Bottom panel: quantum metric $\mathcal{G}_{xx}$ and mean free path at the flat band for different disorder strengths. The red dashed line represents the expected mean free path computed using the Fermi golden rule for smaller disorders.

disorder-induced broadening of the flat bands and the corresponding delocalization of states in real space. Using a simple argument based on the Fermi golden rule, the increase in the mean free path is driven by the reduction of the DOS and the corresponding scattering rate. For weaker disorder, following the scaling theory of localization, one expects that near the flat bands the localization length (related to the mean free path through the Thouless relationship) will reach values on the order of 100 nanometers for a disorder strength corresponding to the effect of electron-hole puddles generated by a silicon oxide substrate [82]. Finally, we note that in the strong Anderson disorder limit, the mean free paths in disordered MATBLG are similar to those found in disordered monolayer graphene [76].

Quantum metric in disordered MATBLG—Figure 4 (bottom panel) shows $\mathcal{G}_{xx}$ for different disorder strengths, and we compare its disorder-dependent evolution with the mean free path. Interestingly, the evolution between $W = 0.75\gamma_0$ and $W = 2\gamma_0$ is qualitatively similar for $\mathcal{G}_{xx}$ and ℓ , indicating a disorder-induced delocalization mechanism.

The increase of the QM for lower disorder is expected for weakly disordered cases, since the cleaner the system is the longer the corresponding mean free path and localization length. Note that the precise value of the mean free path is not shown for lowest disorders due to computational difficulty accessing the diffusive regime in the simulations. However, we can make a rough estimate by using the Fermi golden rule when disorder only weakly affects the band structure, for which $l \propto 1/W^2$. Taking the smallest value of disorder with a converged mfp ($W = 3/4$), for which $l = 33.8$ nm, we extrapolate $l \approx 76$ nm for $W = 1/2$ and $l \approx 306$ nm for $W = 1/4$. Here, it is important to remark that such disorder-induced changes in the quantum metric could also have an impact on MATBLG superconductivity, following some recent works relating the contribution of the quantum metric to the superfluid weight and the critical temperature [54,57].

Conclusion and perspective—Our findings show how disorder can interfere with the inherent superlattice-driven localization effect in MATBLG, giving rise to a nontrivial evolution of the transport length scales, consistent with the evolution of the quantum metric, as a function of disorder and energy. Although our model neglects the Coulomb interaction at the origin of the exotic many-body physics of MATBLG, understanding the impact of disorder on single-electron properties in flat-band systems could prove relevant to gauge the stability of exotic interaction-driven physics. Indeed, disorder-induced broadening of flat bands, as seen in the DOS of Fig. 1, should result in a “softening” of the Coulomb interactions, such that a possible regime of non-interacting electrons might be dominant for the considered Anderson disorder strengths, with Coulomb interactions playing a perturbative role driven by screening effects.

On the other hand, we predict an increment of the quantum metric with the mean free path, which could translate to an extension of the wave packet and enhance the geometrical contribution to the superconductivity [54,57]. So we would expect the geometrical part of the superconductivity to gain importance for a certain range of disorder strength, as well as see a competition between these two contributions in stabilizing the superconducting state. This behavior could be sample-dependent and modulated by effects seen in most fabricated samples, such as substrate-induced corrugation, edge roughness, or twist angle disorder [33–35]. Thus, the quantification of disorder-induced transport characteristics shown here should serve as an interesting metric to explore the stability and nature of the exotic transport phases reported in magic-angle twisted layered materials. Moreover, the work could be extended, applying this methodology to study the Coulomb interaction in such disordered systems (at least in the mean field).

Given the generality of the competition between localization due to geometry and delocalization induced by disorder, we expect these results to also be relevant to other types of flat-band systems in moiré structures or otherwise.

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