Introduction.- In the past decade there has been a keen interest in the thermalization processes of quantum closed systems, partly due to experimental advances in controlling individual quantum systems and partly in order to understand with more rigour the relationship between microscopic and thermodynamic laws [1]. A useful definition in this sense is that thermalization occurs when long-time averages of expected values of observables evolve in such a way as to coincide with the predictions of the standard microcanonical ensemble, and remain close to them for almost any time [2]. One striking exception to this behavior is due to many-body localization (MBL) [3]. This insulating quantum phase of matter emerges in some disordered interacting many-body systems, like the paradigmatic one-dimensional (1D) spin chain, when the disorder is large enough [1, 4–6]. Several experiments in one-dimensional lattice fermions and bosons [7, 8], two-dimensional interacting bosons [9], trapped ultracold ions [10], and superconducting qubits [11, 12] have found its signatures. Notwithstanding, its relevance in the thermodynamic limit (TL) is still under active discussion [13, 14]. Its counterpart, the ergodic phase, in which thermalization is normally expected, is usually not under such scrutiny. However, the transition between the MBL phase and the ergodic phase is not well understood yet. Griffiths effects where anomalously different disorder regions dominate the behavior are supposed to be very relevant close to the transition [15, 16], although recent studies deny that relevance [17]. The possibility of the existence of a non-ergodic but extended phase (a so-called bad metal) between the ergodic and the MBL phases has been proposed but whether it survives in the TL is in doubt [18–21]. Numerical studies of the ergodic phase in spin models showing transitions to a MBL phase for large disorder have found that the ergodic phase shows subdiffusive dynamics and other non-trivial behavior [22, 23]. Yet, it is not clear if these properties are generic or system dependent and they are probably very much affected by intrinsic limitations of the numerical size scaling due to the exponential growth of Hilbert space dimensions.

In this Letter, we explore the Thouless energy (TE) scale [24] in the ergodic region of the disordered Heisenberg spin chain. The TE establishes a limit beyond which the spectral fluctuations separate from the predictions of Random Matrix Theory (RMT), the hallmark of quantum chaos and ergodicity [25–31]. We show that correlations in the fluctuations of observables emerge beyond this scale. This proves that the eigenstate thermalization hypothesis (ETH), which establishes the conditions under which a quantum system thermalizes [32–36], is not exactly fulfilled in the region reported as ergodic in the literature [19]. Hence, thermalization cannot be expected for initial states wider than this scale. Some fundamental questions arise from this fact, like the consequences in the TL. It is usually assumed that the system becomes ergodic in the TL if the TE decreases slower with the system size than the mean level spacing [13]. However, our numerical calculations for the ratio of significantly populated eigenstates by a typical quench suggest that this condition is not enough. The decrease of the TE must be much slower than observed within the ergodic phase. Thus, the absence of equilibration may actually be generic in the TL, and the ergodic phase of our model may disappear altogether in the TL.

Model.- Our results are based on the paradigmatic model for MBL: a 1D chain with two-body nearest-neighbor couplings, \( L \) lattice sites, and onsite magnetic fields with Hamiltonian

\[
\mathcal{H} = \sum_{n=1}^{L} \omega_n \hat{S}_n^z + J \sum_{n=1}^{L-1} \hat{S}_n^x \hat{S}_{n+1}^x + \hat{S}_n^y \hat{S}_{n+1}^y + \lambda \hat{S}_n^z \hat{S}_{n+1}^z. \tag{1}
\]
where $\hat{S}^x_{n,y,z}$ are the total spin operators at site $n$. We choose $J = 1$, $\hbar = 1$, and $\lambda = 1$, the Heisenberg model [37–41]. Periodic boundary conditions are applied, which minimize finite-size effects. Disorder is implemented by the terms $\omega_n$, taken as independent random numbers uniformly distributed over $[-\omega, \omega]$. We allow $\lambda$ to vary to study quenched dynamics.

**Thermalization and its mechanism.** Quantum thermalization occurs if long-time averages of expected values of physical observables, $\langle \hat{O} \rangle \equiv \lim_{\tau \to \infty} 1/\tau \int_0^\tau \text{d}t \langle \psi(t) | \hat{O} | \psi(t) \rangle$, are equal to the microcanonical average,

$$\langle \hat{O} \rangle_{\text{ME}} \equiv \frac{1}{N} \sum_{E\in[E-\Delta E, E+\Delta E]} \langle E \rangle \langle \hat{O} | E \rangle,$$

(2)

$\Delta E$ being a small energy window, $\Delta E/E \ll 1$, containing a large number of levels, $N \gg 1$. It is well known that such long-time averages remain close to an equilibrium value under very generic circumstances [42, 43], although some questions remain open [44]. However, this equilibrium value is not necessarily equal to Eq. (2). The key point for this equality is the set of diagonal terms $O_{nn} \equiv \langle E_n | \hat{O} | E_n \rangle$. Regardless of whether the system thermalizes or not, one has

$$O_{nn} = \langle \hat{O} \rangle_{\text{ME}} + \Delta_n,$$

(3)

where $\langle \cdot \rangle_{\text{ME}}$ is given by Eq. (2), and the quantity $\Delta_n$ represents the difference between long-time, $\langle \cdot \rangle_1$, and microcanonical, $\langle \cdot \rangle_{\text{ME}}$, averages; we will call them diagonal fluctuations. The equivalence between microcanonical and long-time averages lies in the ETH [27–29, 32–34]. In its strong version, it requires that all the values of $\Delta_n$ be negligible; for its weak version, it suffices that most $\Delta_n$ fulfill this condition [35, 44]. Generally, $\Delta_n$ is expected to behave like an uncorrelated random noise whose width decreases fast with the system size [36, 45] for thermalizing systems. Contrarily, $\Delta_n$ can show some structure in integrable systems, due to the presence of additional quantum numbers [46, 47].

**Link between Thouless energy and ETH.** The TE is commonly obtained from long range spectral statistics, which measures the correlations between energy levels separated by a certain distance. For spin chains the number variance $\Sigma^2(L)$ [38] and the spectral form factor $K(\tau)$ [13, 14] have been used. A simpler and convenient alternative is given by the $\delta_n$ spectral statistic [48], which is directly linked to the spectral form factor [39, 50]. It measures the distance between the $n$-th unfolded energy level, which is a dimensionless quantity obtained from the smooth part of the cumulative level density $\epsilon_n \equiv \overline{N}(E_n)$, where $E_n$ is the $n$-th energy level [51], and its average value $\langle \epsilon_n \rangle = n$,

$$\delta_n = \epsilon_n - n.$$  

(4)

This equation suggests a remarkable link between spectral statistics and the ETH. A simple comparison with Eq. (3) shows that $\delta_n$ and $\Delta_n$ play a very similar role. The main difference lies in their physical units: due to the unfolding procedure $\delta_n$ is dimensionless; $\Delta_n$ has the same units than the observable to which it refers. This inconvenience is circumvented by normalizing $\Delta_n$ by its standard deviation $\sigma_{\Delta_n}$,

$$\tilde{\Delta}_n = \frac{\Delta_n}{\sigma_{\Delta_n}} = \frac{O_{nn}}{\sigma_{\Delta_n}} - \frac{\langle \hat{O} \rangle_{\text{ME}}}{\sigma_{\Delta_n}}.$$  

(5)

Here we analyze the diagonal fluctuations, Eq. (5), for representative observables in the model Eq. (1) by means of the same techniques used for the $\delta_n$ statistic. We work with $L = 16$ sites in the spin chain, and as observables we choose the full momentum distribution

$$n_k \equiv \frac{1}{L} \sum_{m,n=1}^L \hat{s}_m^+ \hat{s}_n^\pi e^{2\pi i(m-n)k/L},$$  

(6)

where $k \in \{0, 1, \ldots, L - 1\}$, $\hbar = 1$, and $\hat{s}_\pm$ are the usual ladder spin operators. The microcanonical average is accounted for by fitting $O_{nn}$ to a polynomial of degree 4 to avoid spurious effects typical of local unfolding methods [51]. Our conclusions are independent of the degree of the polynomial. We average over 40 realizations of the magnetic field and $L = 16$ observables given by Eq. (6) in each case. We work with the $N = (16)_3/3 = 4290$ central eigenstates to avoid border effects.

We now apply a discrete Fourier transform to the resulting sequence, and obtain its power spectrum, denoted $\langle \hat{P}_k^S \rangle$. The result is shown in panel (a) of Fig. 1 for certain values of $\omega$ up to the Nyquist frequency, $k_{Ny} \equiv N/2$. For each $\omega$, there exists a value of $k$ beyond which the power spectrum behaves as an uncorrelated white noise. Deviations occur for lower frequencies and structure emerges, indicating a failure of the ETH [22, 52]. The lesser deviations happen for intermediate values of the disorder close to $\omega_c = 0.5$.

This behavior resembles that of the TE, $E_{\text{Th}}$, within the ergodic region and across the transition to MBL [13, 14, 38]. To delve into this coincidence, we rely on the $\delta_n$ statistic, Eq. (4). We again focus on the central $N = (16)_3/3 = 4290$ levels and average over 100 different realizations of the magnetic field. We remove 30 energies closest to both spectrum edges before and after unfolding with a polynomial of degree 6. Next, we obtain its power spectrum [39, 50], $\langle \hat{P}_k^S \rangle$. In panel (b) of Fig. 1, we show $\langle \hat{P}_k^S \rangle$ for certain values of $\omega$ in the ergodic region. Accordance between theoretical and simulated curves is best at $\omega = \omega_c$, for which they show agreement for almost all frequencies $k$. As $|\omega - \omega_c|$ is increased, the same phenomenon as in panel (a) is strikingly observed.

To interpret these results, we link the frequency $k$ to the scaled (dimensionless) time $\tau$ of the spectral form...
we plot the difference between the \( L \) does change within the ergodic region with a maximum at \( \tau \). This is, correlations breaking ETH emerge

FIG. 1. Panel (a): Power spectrum \( \langle P_k \rangle \) of the diagonal fluctuations, Eq. (5) as a function of \( k \in \{1, \ldots, N-1\} \). Results are averages over 640 realizations of \( N = 4290 \) expected values. Panel (b): Power spectrum \( \langle P_k^\Delta \rangle \) of the \( \delta \) statistic, Eq. (4), for the system \( \mathcal{H} \), Eq. (1) with \( \lambda = 1 \). Plotted against them are the theoretical power spectra for the chaotic and regular cases \([49, 50]\). Results are averages over 100 spectra with \( N = 4290 \) levels each. For both panels, the disorder parameter is \( \omega \in \{0.3, 0.5, 0.7, 0.9, 1.1, 1.3, 1.5\} \).

factor, \( \tau = k/N \) \([49, 50]\). As the Thouless time is the value, \( \tau_{th} \), below which the spectral form factor deviates from RMT behavior, we can define a Thouless frequency, \( k_{Th} \equiv N\tau_{Th} \), with the same meaning. The inverse of the Thouless time, \( \tau_{Th} \equiv 1/\tau_{Th} = N/k_{Th} \), represents, then, a limiting scale for RMT-like behavior: energy levels with less than \( \tau_{Th} \) in between are correlated like RMT spectra; energy levels with more than \( \tau_{Th} \) in between deviate from this behavior towards integrable-like correlations. As the frequency of panel (a) of Fig. 1 has the same physical meaning, a similar reasoning can be applied.

To determine \( k_{min} \approx k_{Th} \) we choose the first value of \( k \) for which the curve fluctuates below the ergodic expectation, the ergodic curve for \( \langle P_k \rangle \) and zero for \( \langle P_k^\Delta \rangle \). Other procedures give similar qualitative results. Next, we calculate the characteristic length \( \ell_{max} \equiv N/k_{min} \approx \ell_{Th} \). Results are shown in panel (a) of Fig. 2. We find a good quantitative agreement between the results for both power spectra. Diagonal fluctuations cease to be uncorrelated white noise for scales larger than \( \ell_{Th} \) and, therefore, the ETH is broken beyond this scale. In panel (b) of Fig. 2, we show \( \ell_{max}/N \), representing such a critical correlation length normalized by the total number of energy levels. As \( L \) is increased, fluctuations get gradually smoothed, but the general structure remains very similar. Hence, despite the obvious limitations of such a finite-size scaling analysis, this suggests that \( \ell_{max}/N \) does change within the ergodic region with a maximum at \( \omega \approx 0.5 - 0.6 \), but remains approximately constant as we increase \( L \). That is, correlations breaking ETH emerge at quite small scales, even within the region identified as ergodic \([19]\).

**Thermalization in finite systems.** The main consequence of the previous results can be summarized as follows: the key element in predicting whether a particular initial condition is thermalizing is its number of significantly populated eigenstates, \( N \). If \( N < \ell_{Th} \), diagonal fluctuations behave as an uncorrelated white noise at all scales within the populated window, and thermalization may be expected. Otherwise, the emerging structure of diagonal fluctuations can seriously impede thermalization, depending on how the population of eigenstates fluctuate within the window given by \( N \): the larger the ratio \( N/\ell_{Th} \), the more likely to find a non-thermalizing initial condition.

To test this conclusion, we perform the following numerical experiments. We start from the central state of a certain initial value for \( \lambda \) in Eq. (1), \( \lambda_i > 1 \), and let it evolve under a Hamiltonian with the same values for the random magnetic field, \( \omega_n \), \( \forall n \), and \( \lambda_M = 1 \). The size of this quench, \( \Delta \equiv |\lambda_i - \lambda_f| \), determines the width, \( N \), of the resulting state. We perform these experiments with five different values of \( \omega \) and five different quench sizes. In panel (a) of Fig. 3 we plot the difference between the long-time, \( \langle n_k \rangle_t \), and the microcanonical, \( \langle n_k \rangle_{ME} \), averages. We have averaged over all 16 observables and 20 realizations of the random magnetic field. The microcanonical average is obtained with 41 eigenstates around the expected energy of the initial state. In panel (b) of the same figure, a disorder average of the eigenstate populations for the five quench sizes is shown. The results are compatible with our previous statement. Agreement between long-time and microcanonical averages is favoured.
by large $\ell_{\text{max}}$. The case with the greatest $\ell_{\text{Th}}$, $\omega = 0.6$, is the least sensible to $\Delta \lambda$, and the one showing minimal $\langle |\langle n_k \rangle_t - \langle n_k \rangle_{\text{ME}} \rangle \rangle$. The cases with $\omega = 0.3$ and $\omega = 1.2$ show a fast increase of the difference between these two averages from $\Delta \lambda = 0.1$ to $\Delta \lambda = 0.3$. This means that, because $\ell_{\text{Th}}$ is smaller, thermalization is more sensible to $\Delta \lambda$. For $\Delta \lambda \gtrsim 0.3$ the behavior is not monotonic, probably because the populated window is still significantly covering integrable-like scales. The case $\omega = 0.9$ shows a monotonic increase of this difference, leading to a very similar value to $\omega = 0.3$ and $1.2$ for $\Delta \lambda = 0.8$. Finally, $\omega = 1.5$ shows the largest difference as $\ell_{\text{Th}}$ is very small.

**Challenges in the TL.-** Fig. 3 shows that thermalization is not guaranteed within the ergodic region for finite systems. Here, we investigate the implications in the TL by considering the number of populated eigenstates by a typical quench. We have checked that the excitation energy at the band center is an extensive quantity and proportional to the size of the system, as expected in ordinary statistical mechanics. Then, we assume that the width of a typical quench is equal to that of a canonical equilibrium state [53], $\sigma_E \propto \sqrt{L}$. We now study the number of levels $N$ populated in an energy window of width $\sigma_E = \Gamma \sqrt{L}$. In Fig. 4 we show the ratio $N/N$ for different $\Gamma$ as a function of $L$. Quite surprisingly, this ratio remains approximately constant for all values of $L$, meaning that the number of typically populated eigenstates shows a fast linear growth $N \propto L$. Since $N/N$ shows *almost perfect* constant behavior in $L$, we suspect that this may actually be the case in the TL. Thus, results of panel (b) of Fig. 3, showing that typical quenches are wider than the TE, may be representative of the TL.

Therefore, the absence of thermalization in the ergodic phase, panel (a) of Fig. 3, may remain in the TL for typical initial states.

**Discussion.-** In this Letter we have studied the spectral statistics and the validity of the ETH within the usually identified as ergodic region of a paradigmatic 1D spin chain which transits to the MBL phase. We have found a strong link between the TE — the scale beyond which the spectral statistics deviate from the expected behavior for ergodic systems — and the failure of the ETH measured through the fluctuations of expected values of observables in the Hamiltonian eigenbasis. The power spectrum of these fluctuations shows deviations from results corresponding to an uncorrelated random noise at scales larger than the TE. As opposed to RMT spectra, in which these fluctuations are totally random, they show a certain structure beyond this scale. We have performed a numerical experiment on a finite chain with $L = 16$ sites to show that, the shorter this scale, the less accurate the microcanonical ensemble is in describing long-time averages. Previous studies of long range correlations and the TE scale in disordered many-body quantum systems have focused on large values of the disorder and the transition to the MBL phase and, in this sense, our results are quite different [13, 14, 38]. The relevance of the diagonal fluctuations has gone quite unnoticed in the past.

Results shown in Fig. 3 show that the TE is narrower than the width of typical quenches in finite (and small) systems. A natural question that emerges from this result regards the implications in the TL. Despite the obvious limitations of our finite-size scaling, we can make a solid conjecture. Results from Fig. 4 appear to suggest that the observed absence of thermalization may not be a finite-size effect. Since the number of populated levels by a typical quench grows linearly with the system size, our findings for finite $L$ may be reflecting the real situation in the TL, meaning that ergodicity would perhaps disappear. We call for future research on this matter, both theoretical and experimental. The hardship that large $L$...
This work has been supported by the Spanish Grants Nos. FIS2015-63770-P (MINECO/FEDER) and PGC2018-094180-B-100 (MCU/AEI/FEDER, EU), CAM/FEDER Project No.S2018/TCS-4342 (QUITEMAD-CM) and CSIC Research Platform PTI-001.

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