We examine how the magnetic susceptibility obtained by the quench experiment on isolated quantum systems is related to the isothermal and adiabatic susceptibilities defined in thermodynamics. Under the conditions similar to the eigenstate thermalization hypothesis (ETH), together with some additional natural ones, we prove that for translationally invariant systems the quench susceptibility as a function of wavevector \( k \) is discontinuous at \( k = 0 \). Moreover, its values at \( k = 0 \) and the \( k \to 0 \) limit coincide with the adiabatic and the isothermal susceptibilities, respectively. We give numerical predictions on how these particular behaviors can be observed in experiments on the XYZ spin chain with tunable parameters, and how they deviate when the conditions are not fully satisfied.

**Introduction**—. Ultracold atoms \(^1\) and molecules \(^3\) in optical lattices offer nearly ideal playgrounds for studying quantum many-body systems experimentally. Various model systems \(^0\) \(^2\) \(^{21}\) are realized on the optical lattices with various geometry \(^22\) \(^27\) and with tunable physical parameters \(^2\) \(^28\) \(^31\). Furthermore, one can isolate the systems from the environments over a reasonably long period, which enables the direct observation of the dynamics of isolated quantum systems induced by suddenly changing a physical parameter \(^23\) \(^38\). After this so-called quench, the system often relaxes to a steady state, where the expectation values of local observables become almost time-independent \(^22\) \(^39\) \(^41\). The nature of such a steady state has been discussed in terms of the eigenstate thermalization hypothesis (ETH) \(^45\) \(^56\). For example, if the ‘strong’ ETH is satisfied, the steady state is an equilibrium state \(^45\) \(^51\).

In this Letter, we study the susceptibility obtained by the quench experiment, and explore whether or not it coincides with a thermodynamic susceptibility. This problem is highly nontrivial since there are two kinds of thermodynamic susceptibilities, the isothermal and the adiabatic ones, which take different values. In other words, it is not even clear which thermodynamic susceptibilities should be compared with the quench one. Furthermore, the wavenumber dependences of these susceptibilities make the problem even more nontrivial, as we will reveal in this paper.

To be concrete, we consider the magnetic susceptibility of a quantum spin system. Suppose that the initial equilibrium state is in a uniform ‘offset’ magnetic field, \( h \), and a weak extra magnetic field of wavenumber \( k \) is suddenly applied. The quench susceptibility, \( \chi_{\text{qch}}(k) \), is defined as the rate of magnetization change induced by such a quench. We explore its relation to the isothermal and the adiabatic thermodynamic susceptibilities, \( \chi^T(k) \) and \( \chi^S(k) \), in the case where \( \chi^T(0) > \chi^S(0) \), which occurs when \( h \neq 0 \).

We reveal that \( \chi_{\text{qch}}^T(k) \) is discontinuous at \( k = 0 \) as a function of \( k \). Due to this discontinuity, both thermodynamic susceptibilities are obtained from the quench one, as \( \chi_{\text{qch}}^T(0) = \chi^S(0) \) and \( \lim_{k \to 0} \chi_{\text{qch}}^T(k) = \chi^T(0) \). The proof requires the conditions similar to the ETH, which hold when the dynamics of the system is complicated enough, as well as the natural conditions that are satisfied except at a phase transition point.

Furthermore, we numerically demonstrate how such anomalous behaviors should be observed in experiments on an isolated quantum spin system when it is nonintegrable. We also predict how the deviation from these behaviors is observed when the physical parameters of the system are tuned so that it becomes integrable.

**Setup**—. We deal with a quantum spin-1/2 system on a \( d \)-dimensional cubic lattice \( \Omega_N \) with linear size \( L \) and \( N = L^d \) spins. The periodic boundary conditions and the invariance under the discrete spatial translations are assumed for the pre-quench Hamiltonian \( \hat{H}(h) \), where \( h \) denotes the uniform offset magnetic field. The density matrix of the initial state is chosen as the canonical Gibbs one, \( \hat{\rho}_{\text{ini}} = e^{-\beta \hat{H}(h)}/Z \).

We are interested in the quantum quench process where the additional magnetic field \( \Delta h(r) \), with wavenumber \( k \) and small magnitude \( \Delta h_k \), is applied suddenly at \( t = 0 \). At \( t > 0 \), the isolated system obeys the Schrödinger dynamics of the post-quench Hamiltonian, \( \hat{H}(h) - \sum_{\mathbf{r} \in \Omega_N} \hat{\sigma}_x r \Delta h(\mathbf{r}) \), where \( \hat{\sigma}_x (\alpha = x, y, z) \) is the Pauli operator on site \( \mathbf{r} \in \Omega_N \). While the previous works regarding the quantum quench focused only on the final state, we here study the quench susceptibility,

\[
\chi_{\text{qch}}^N(k) := \lim_{T \to \infty} \lim_{\Delta h_k \to 0} \frac{\text{Tr}[\hat{\rho}(t) \hat{m}_k] - \text{Tr}[\hat{\rho}_{\text{ini}} \hat{m}_k]}{\Delta h_k},
\]

which quantifies the difference of the expectation values of the \( k \)-component of magnetization, \( \hat{m}_k = \hat{\sigma}_x r \Delta h(\mathbf{r}) \).
\[
\langle v|\bar{\sigma}_0^z|\nu\rangle \propto \Delta E_{\nu}/N + o(1/\sqrt{N}).
\] (8)

This is similar to but different from the ordinary two forms of ETH in the following points. The ordinary strong ETH \cite{48,49,50,51} requires that all \(\langle v|\bar{\sigma}_0^z|\nu\rangle\) behave like a smooth function of \(E_{\nu}/N\), which is often satisfied in nonintegrable systems \cite{68}. Since a smooth function of \(E_{\nu}/N\) can be regarded as linear within the narrow region \(|\Delta E_{\nu}| \lesssim T\sqrt{c_{ch}N}\), any system satisfying the strong ETH also satisfies condition (6), while notice that the converse is not necessarily true. By contrast, the ordinary weak ETH \cite{53,55} requires only that \(\langle v|\bar{\sigma}_0^z|\nu\rangle = o(1)\) for almost all \(\nu\) in the same energy region. For this reason, some models that satisfy the ordinary weak ETH do not satisfy Eq. (6), as will be demonstrated shortly.

Demonstration of (i).—. We now demonstrate how result (i) can be observed in experiments on the XYZ spin chain, which has the pre-quench Hamiltonian,

\[
\hat{H}(h) = -\sum_{j=0}^{N-1} J_{0}\hat{\sigma}_j^\alpha \hat{\sigma}_{j+1}^\alpha - \sum_{j=0}^{N-1} h\hat{\sigma}_j^z,
\] (9)

with periodic boundary condition, \(\hat{\sigma}_N = \hat{\sigma}_0\). Since spin systems \cite{14,21} and a 1D ring \cite{22,23} can be separately realized in ultracold atoms and molecules, we expect this model can also be realized experimentally. This model alone covers three different classes of systems, (a) XYZ, (b) XXZ \((J_x = J_y \neq J_z)\) and (c) XY \((J_z = 0)\) models, by tuning the parameters \(J_{x}, J_{y}\). We here predict the behaviors of the susceptibilities by means of the numerical diagonalization for (a) and (b), and the analytic evaluation for (c), respectively.

Figure 1(a) shows the \(N\) dependence of the \(k = 0\) components \(\chi_{N}^{\text{ch}}(0)\), \(\chi_{N}^{T}(0)\) and \(\chi_{N}^{S}(0)\) in the XYZ model \cite{61}. Since the model has no local conserved quantity for \(h \neq 0\) \cite{62}, it is expected that the condition (6) is fulfilled, so that Eq. (6) holds. In fact, Fig. 1(a) shows that \(\chi_{N}^{\text{ch}}(0)\) approaches \(\chi_{N}^{S}(0)\) as \(N\) increases. Their difference decreases nearly exponentially, as shown in the

\[
\chi_{N}^{\text{ch}}(0) \leq \chi_{N}^{S}(0).
\] (7)
and to the off-diagonal elements between

This is similar to the 'off-diagonal ETH' [44, 49–52], except for the following points. Firstly, the off-diagonal ETH requires not all such off-diagonal elements of \( \overline{\rho} \) to vanish as \( N \to \infty \). By contrast, Eq. (10) refers only to a particular spin operator \( \hat{\sigma} \) and to the off-diagonal elements between specific pairs of states such that

\[
E_v = E_{v'} \quad \text{and} \quad K_v = K_{v'} + k.
\]

Furthermore, it requires not all such off-diagonal elements but most of them to vanish. Secondly, the ordinary off-diagonal ETH [44, 49, 51] requires exponentially fast decay of all the off-diagonal elements, which is not necessarily satisfied in integrable models. By contrast, Eq. (10) is a weaker condition [58] that can be satisfied even in integrable models, as we will demonstrate shortly for the XY model.

It is noteworthy that if we impose Eqs. (8) and (10), not only on a particular spin operator \( \hat{\sigma} \) but also on all other local operators, we obtain a new necessary condition for thermalization, which is also a sufficient condition as long as the quench parameter \( \Delta h \) is small.

Conditions for (iii) —. We introduce the canonical spin-spin correlation function [58, 64] as \( \phi^T(r) := \beta \langle \hat{\sigma} \hat{\sigma}^\dagger \rangle_{\text{ini}} \). Then, we can show [58] that \( \chi^T_N(k) \) is uniformly continuous on the whole region (including \( k = 0 \)), if \( \phi^T_N(r) \) decays fast enough such that

\[
\lim_{N \to \infty} \sum_{r \in \Omega_N} |\phi^T_N(r)| < \infty \quad (12)
\]

and if finite-size effects are small such that

\[
\lim_{N \to \infty} \sum_{r \in \Omega_N} |\phi^T_N(r) - \phi^T_N(r)| = 0. \quad (13)
\]

Since we exclude phase transition points, condition (12) is expected to be satisfied in most systems. Moreover, it seems normal that the condition (13) holds, since the canonical ensemble well emulates a subsystem in an infinite system [65, 66].

If conditions (10), (12) and (13) are all fulfilled, Eq. (9) follows from results (ii) and (iii). It also follows that \( \chi^T_N(k) \) is discontinuous at \( k = 0 \), as discussed in (iv).

Demonstrations of (ii)-(iv) —. The discontinuity of \( \chi^T_N(k) \) may seem counterintuitive, but can be verified experimentally by adopting the isolated system representation Eq. (9). The observed susceptibility should follow the following results of the numerical simulation.

Figures 2 shows the \( k \)-dependence of \( \chi^T_N(k) \) and \( \chi^T_N(k) \) in the (a) XYZ, (b) XXZ and (c) XY models. Recalling that the condition (10) is weaker than the ordinary off-diagonal ETH [44, 49, 51], we expect that it is fulfilled in all these models. In fact, our data show that Eq. (5), \( \chi^T_N(k) = \chi^T_N(k) = \chi^T_N(k) \) for all \( k \neq 0 \), holds in each model. We also find that \( \chi^T_N(k) = \chi^T_N(k) \) for \( k \neq 0 \) as \( \Theta(1/N) \) in (c). This is because the off-diagonal elements |\( \langle \nu'|\hat{\sigma}^\dagger|\nu \rangle | \) that satisfy Eq. (11) decay not exponentially but algebraically as \( \Theta(1/N) \) for the XY model.

The conditions (12) and (13) are the natural ones that will also be satisfied in all these models. In fact, Figs. 2(a)-(c) indicate Eq. (6), \( \lim_{k \to 0} \chi_N^T(k) = \chi^T_N(0) \), holds and hence \( \chi^T_N(k) \) is discontinuous at \( k = 0 \) while \( \chi^T_N(k) \) is uniformly continuous.

For the parameters presented here, Eqs. (5) and (6) hold in all three cases, while Eq. (4) only in the XYZ one. By further varying \( J_x \) and \( J_y \), we can also construct a model for which none of Eqs. (4)-[6] holds [58]. In such a case, the condition (10) is violated, while the conditions (12) and (13) are still fulfilled.

Physical origin of (iv) —. Among (i)-(v), the result (iv), namely the anomalous behavior of \( \chi^T_N(k) \) at around...
0.15
0.1
0.1
0.2
0
1
2
(a) XYZ
0
0.1
0.15
0.2
0
1
2
3
(b) XXZ
0.1
0.15
0
1
2
3
(c) XY
0.1
0.1
0
1
2
3

FIG. 2. $k$ dependence of $\chi^\text{qch}_N(k)$, $\chi^T_N(k)$ and $\chi^S_N(k)$ in (a) XYZ, (b) XXZ and (c) XY models, with the same parameters as in Fig. 1. We take (a), (b) $N = 12$-17 and $k = 2\pi n_\perp/N$ and (c) $N = 2^n$ with $n = 3$-9 and $k = 2\pi n_\perp/N$, with $n_k = 0$-4. Solid line in (c): $\chi^\text{qch}_N(k) = \chi^S_N(k) - \chi^T_N(k)$ for $k \neq 0$, whereas the dashed line shows its discontinuous jump to $\chi^\text{qch}_N(0)$.

$k = 0$, should be the most nontrivial one. We here give its physical interpretation assuming that Eqs. (4) and (5) hold.

Suppose a huge system enclosed by an adiabatic wall, and its large number of sites, $N_{\text{tot}}$, allows $\chi^*_N(k)$ to be well approximated by $\chi^*_\infty(k)$. Then, we focus on a subsystem of $N$ sites, where $N_{\text{tot}} \gg N \geq 1$, and quasistatically apply an additional field, $\Delta h$, only to the subsystem. Since the rest of the system works as a heat reservoir for the subsystem, the total magnetization of the subsystem changes by $\{N\chi^T_N(0) + o(N)\}\Delta h$. We can also evaluate it as $N E_N[\chi^S_N(k)]\Delta h$ by regarding the same field as the superposition of magnetic fields of wavenumber $k$ in the entire system, where $E_N[\bullet]$ denotes a weighted average over a small but finite region of $k$ such that $|k| \lesssim 2\pi/L$. By equating these two evaluations, we obtain

$$\chi^T_N(0) + o(1) = E_N[\chi^S_N(k)],$$

which yields Eq. (6). From Eqs. (3) and (5), this shows that not only $\chi^S_N(k)$ but also $\chi^\text{qch}_N(k)$ is discontinuous at $k = 0$.

Relation to Kubo formula —. We finally discuss the relation to the susceptibility obtained by the Kubo formula, $\chi^\text{Kubo}_N(k, \omega + i\varepsilon)$, which was derived assuming also that the system is isolated. Here, $\omega$, is the frequency and $\varepsilon$ is an infinitesimal positive number. While we have defined $\chi^\text{qch}_N$ through a sudden quench of $\Delta h(r)$, Kubo derived $\chi^\text{Kubo}_N$ assuming that $\Delta h(r)$ is switched on gradually over a long time scale $\sim 1/\varepsilon$.

It is generally believed that the $\varepsilon \to +0$ limit of $\chi^\text{Kubo}_N$ should be taken after the $N \to \infty$ limit [68] [72]. However, some works took the $\varepsilon \to +0$ limit keeping $N$ finite [73] [75]. For the latter limit, we can show

$$\lim_{\varepsilon \to +0} \chi^\text{Kubo}_N(k, 0 + i\varepsilon) = \chi^\text{qch}_N(k)$$

for all $N$, although LHS and RHS correspond to the slow and fast processes, respectively, which would result in different final states. Therefore, all the statements (i)-(iv) for $\chi^\text{qch}_N(k)$ hold also for $\lim_{\varepsilon \to +0} \chi^\text{Kubo}_N(k, 0 + i\varepsilon)$ [76]. Moreover, the previous results on $\lim_{\varepsilon \to +0} \chi^\text{Kubo}_N(0, 0 + i\varepsilon)$ can be understood more precisely using (i) [58]. However, it is noteworthy that $\chi^\text{Kubo}_N$ is hard to measure in experiments in contrast to $\chi^\text{qch}_N$, since the system cannot be isolated for the infinitely long timescale.

In conclusion, we have revealed the anomalous natures of the quench susceptibility, demonstrating together that experimental verifications are feasible enough.

We thank Y. Yoneta, A. Noguchi and Y. Kato for discussions, and C. Hotta, R. Hamazaki and K. Saito for helpful comments. This work was supported by The Japan Society for the Promotion of Science, KAKENHI No. 19H01810, 15H05700 and 17K05497.

[1] M. Lewenstein, A. Sanpera, V. Ahufinger, B. Damski, A. Sen, and U. Sen, Ultracold atomic gases in optical lattices: Mimicking condensed matter physics and beyond, Advances in Physics 56, 243 (2007).
[2] I. Bloch, J. Dalibard, and W. Zwerger, Many-body physics with ultracold gases, Reviews of Modern Physics 80, 885 (2008).
[3] A. Micheli, G. K. Brennen, and P. Zoller, A toolbox for lattice-spin models with polar molecules, Nature Physics 2, 341 (2006).
[4] C. Chin, V. V. Flambaum, and M. G. Kozlov, Ultracold molecules: New probes on the variation of fundamental constants, New Journal of Physics 11, 10.1088/1367-2630/11/5/055048 (2009).
[5] L. D. Carr, D. DeMille, R. V. Krems, and J. Ye, Cold and ultracold molecules: Science, technology and applications, New Journal of Physics 11, 10.1088/1367-2630/11/5/055049 (2009).
[6] D. Jaksch, C. Bruder, J. I. Cirac, C. W. Gardiner, and P. Zoller, Cold bosonic atoms in optical lattices, Physical Review Letters 81, 3108 (1998).
[7] M. Greiner, O. Mandel, T. Esslinger, T. W. Hänsch, and I. Bloch, Quantum phase transition from a superfluid to a Mott insulator in a gas of ultracold atoms, Nature 415, 39 (2002).
[8] T. Stöferle, H. Moritz, C. Schori, M. Köhl, and T. Esslinger, Transition from a strongly interacting 1D superfluid to a Mott insulator, Physical Review Letters 92, 1 (2004).
[9] I. B. Spielman, W. D. Phillips, and J. V. Porto, Mott-insulator transition in a two-dimensional atomic bose gas, Physical Review Letters 98, 1 (2007).
I. B. Spielman, W. D. Phillips, and J. V. Porto, Condensate fraction in a 2D bose gas measured across the mott-insulator transition, Physical Review Letters 100, 1 (2008).

M. Kohl, H. Moritz, T. Stoferle, K. Gunter, and T. Esslinger, Fermionic atoms in a three dimensional optical lattice: Observing Fermi surfaces, dynamics, and interactions, Physical Review Letters 94, 1 (2005) arXiv:0410389 [cond-mat].

M. F. Parsons, A. Mazurenko, C. S. Chiu, G. Ji, D. Greif, and M. Greiner, Site-resolved measurement of the spin-correlation function in the Fermi-Hubbard model, Science 353, 1253 (2016) arXiv:1605.02704.

T. Esslinger, Fermi-Hubbard Physics with Atoms in an Optical Lattice, Annual Review of Condensed Matter Physics 1, 129 (2010).

M. L. Wall, K. Maeda, and L. D. Carr, Realizing unconventional quantum magnetism with symmetric top molecules, New Journal of Physics 17, 10.1088/1367-2630/17/2/025001 (2015).

G. Peligrí, J. Mompart, V. Ahufinger, and A. J. Daley, Quantum magnetism with ultracold bosons carrying orbital angular momentum, Physical Review A 100, 1 (2019).

J. Simon, W. S. Bakr, R. Ma, M. E. Tai, P. M. Preiss, and M. Greiner, Quantum simulation of antiferromagnetic spin chains in an optical lattice, Nature 472, 307 (2011) arXiv:1103.1372.

T. Fukuhara, P. Schauf, M. Endres, S. Hild, M. Cheneau, I. Bloch, and C. Gross, Microscopic observation of magnon bound states and their dynamics, Nature 502, 76 (2013) arXiv:1305.6598.

T. Fukuhara, S. Hild, J. Zeiher, P. Schauf, I. Bloch, M. Endres, and C. Gross, Spatially Resolved Detection of a Spin-Entanglement Wave in a Bose-Hubbard Chain, Physical Review Letters 115, 1 (2015) arXiv:1504.02582.

B. Yan, S. A. Moses, B. Gadway, J. P. Covey, K. R. Hazzard, A. M. Rey, D. S. Jin, and J. Ye, Observation of dipolar spin-exchange interactions with lattice-confined polar molecules, Nature 501, 521 (2013).

K. R. Hazzard, B. Gadway, M. Foss-Feig, B. Yan, S. A. Moses, J. P. Covey, N. Y. Yao, M. D. Lukin, J. Ye, D. S. Jin, and A. M. Rey, Many-body dynamics of dipolar molecules in an optical lattice, Physical Review Letters 113, 1 (2014) arXiv:1402.2354.

A. P. Orioli, A. Signoles, H. Wildhagen, G. Günter, J. Berges, S. Whitlock, and M. Weidmüller, Relaxation of an Isolated Dipolar-Interacting Rydberg Quantum Spin System, Physical Review Letters 120, 63601 (2018) arXiv:1703.05957.

L. Amico, A. Österloh, and F. Cataliotti, Quantum many particle systems in ring-shaped optical lattices, Physical Review Letters 95, 1 (2005).

K. Henderson, C. Ryu, C. MacCormick, and M. G. Boshier, Experimental demonstration of painting arbitrary and dynamic potentials for Bose-Einstein condensates, New Journal of Physics 11, 10.1088/1367-2630/11/4/043030 (2009) arXiv:0902.2171.

G. Wirth, M. Olschläger, and A. Hemmerich, Evidence for orbital superfluidity in the P-band of a bipartite optical square lattice, Nature Physics 7, 147 (2011) arXiv:1006.0569.

P. Soltan-Panahi, J. Struck, P. Hauke, A. Bick, W. Plenkers, G. Meineke, C. Becker, P. Windpassinger, M. Lewenstein, and K. Sengstock, Multi-component quantum gases in spin-dependent hexagonal lattices, Nature Physics 7, 434 (2011) arXiv:1005.1276.

L. Tarruell, D. Greif, T. Uehlinger, G. Jotzu, and T. Esslinger, Creating, moving and merging Dirac points with a Fermi gas in a tunable honeycomb lattice, Nature 483, 302 (2012) arXiv:1111.5020.

G. B. Jo, J. Guzman, C. K. Thomas, P. Hosur, A. Vishwanath, and D. M. Stamper-Kurn, Ultracold atoms in a tunable optical kagome lattice, Physical Review Letters 108, 1 (2012) arXiv:1109.1591.

H. Feshbach, Unified theory of nuclear reactions, Annals of Physics 5, 357 (1958).

U. Fano, Effects of configuration interaction on intensities and phase shifts, Physical Review 124, 1866 (1961).

E. Tiesinga, B. J. Verhaar, and H. T. C. Stoof, Threshold and resonance phenomena in ultracold ground-state collisions, Physical Review A 47, 4114 (1993).

H. T. Stoof, M. Houbiers, C. A. Sackett, and R. G. Hulet, Superfluidity of spin-polarized 6 Li, Physical Review Letters 76, 10 (1996).

M. Greiner, O. Mandel, T. W. Hänsch, and I. Bloch, Collapse and revival of the matter wave field of a Bose-Einstein condensate, Nature 419, 51 (2002).

L. E. Sadler, J. M. Highie, S. R. Leslie, M. Vengalattore, and D. M. Stamper-Kurn, Spontaneous symmetry breaking in a quenched ferromagnetic spinor Bose-Einstein condensate, Nature 443, 312 (2006) arXiv:0605351 [cond-mat].

F. Meinert, M. J. Mark, E. Kirilov, K. Lauber, P. Weimann, A. J. Daley, and H. C. Nägerl, Quantum quench in an atomic one-dimensional Ising chain, Physical Review Letters 111, 1 (2013).

G. Lamporesi, S. Donadello, S. Serafini, F. Dalfovo, and G. Ferrari, Spontaneous creation of Kibble-Zurek solitons in a Bose-Einstein condensate, Nature Physics 9, 656 (2013) arXiv:1306.4523.

C. L. Hung, V. Gurarie, and C. Chin, From cosmology to cold atoms: Observation of Sakharov oscillations in a quenched atomic superfluid, Science 341, 1213 (2013) arXiv:1209.0011.

T. Fukuhara, A. Kwantian, M. Endres, M. Cheneau, P. Schauf, S. Hild, D. Bellem, U. Schollwöck, T. Giamarchi, C. Gross, I. Bloch, and S. Kuhr, Quantum dynamics of a mobile spin impurity, Nature Physics 9, 235 (2013) arXiv:1209.6468.

S. Hild, T. Fukuhara, P. Schauf, J. Zeiher, M. Knap, E. Demler, I. Bloch, and C. Gross, Far-from-equilibrium spin transport in heisenberg quantum magnets, Physical Review Letters 113, 1 (2014).

S. Trotzky, Y. A. Chen, A. Flesch, I. P. McCulloch, U. Schollwöck, J. Eisert, and I. Bloch, Probing the relaxation towards equilibrium in an isolated strongly correlated one-dimensional Bose gas, Nature Physics 8, 325 (2012).

T. Kinoshita, T. Wenger, and D. S. Weiss, A quantum Newton’s cradle, Nature 440, 900 (2006).

M. Gring, M. Kuhwert, T. Langen, T. Kitagawa, B. Rauer, M. Schreitl, I. Mazets, D. A. Smith, E. Demler, and J. Schmiedmayer, Relaxation and Prethermalization in, Science 337, 1318 (2012).

H. Tsuchi, From quantum dynamics to the canonical distribution: General picture and a rigorous example, Physical...
P. Reimann, Foundation of statistical mechanics under experimen-
tally realistic conditions, Physical Review Letters 101, 1 (2008) [arXiv:0810.3092]

T. Mori, T. N. Ikeda, E. Kaminishi, and M. Ueda, Thermalization and prether-
malization in isolated quantum systems: A theoretical overview, Journal of Physics B: Atomic, Molecular and Optical Physics 51, 10.1088/1361-6455/aabcd (2018).

J. von Neumann, Proof of the ergodic theorem and the H-
theorem in quantum mechanics, The European Physical-
Journal H 35, 201 (2010)

J. M. Deutsch, Quantum statistical mechanics in a closed
system, Physical Review A 43, 2046 (1991)

M. Srednicki, Chaos and quantum thermalization, Phys-
ical Review E 50, 888 (1994)

T. Kuwahara and K. Saito, Ensemble equivalence and
thermodynamics, Physical Review E - Statistical, Nonlin-
erar, and Soft Matter Physics 91, 1 (2015)

D. Iyer, M. Srednicki, and M. Rigol, Optimization of
finite-size errors in finite-temperature calculations of un-
ordered phases, Physical Review E - Statistical, Nonlin-
erar, and Soft Matter Physics 90, 1 (2014)

R. Kubo, Statistical Mechanic Theory of Irreversible
Processes. I. General Theory and Simple Applications to Magnetic and Conduction Problems, Journal of the
Physical Society of Japan 12, 570 (1957) [arXiv:0211006]

D. Pines and P. Nozieres, The Theory of Quantum Li-
quids, Vol I: Normal Fermi Liquids (W.A.Benjamin, Inc.
New York, 1966).

G. Giuliani and G. Vignale, Quantum Theory of the Elec-
tron Liquid (Cambridge University Press, 2005).

D.N.Zubarev, Nonequilibrium Statistical Thermodynam-
ics (Consultants Bureau, New York, 1974).

D. Zubarev, V. Morozov, and G. Röpke, Statistical Me-
chanics of Nonequilibrium Processes, Volume 1: Bas-
cial Concepts, Kinetic Theory, Vol. 1 (Akademie-Verlag
Berlin, 1996).

D. Zubarev, V. Morozov, and G. Röpke, Statistical Me-
chanics of Nonequilibrium Processes, Volume 2: Relax-
ation and Hydrodynamic Processes, Vol. 2 (Akademie-
Verlag Berlin, 1997).

H. Falk, Lower Bound for the Isothermal Magnetic Sus-
ceptibility, Physical Review 165, 602 (1968)

R. M. Wilcox, Bounds for the isothermal, adiabatic,
and isolated static susceptibility tensors, Physical Review
174, 624 (1968)

M. Suzuki, Ergodicity, constants of motion, and bounds for
susceptibilities, Physica 51, 277 (1971)

For the former order of limits (ε → +0 after
N → ∞), the conventional wisdom [63, 69] is that
lim k→0 ε→+0 N→∞ χ_{N}^{ab}(k, 0+ εi) = \chi_{N}^{T}(0),
which however does not always hold. Our results (ii)-(iv) suggest
the condition for the validity of this wisdom, although
we have not yet proved \lim \epsilon \rightarrow +0 N \rightarrow \infty \chi_{N}^{ab}(k, 0+εi) = \chi_{N}^{ab}(k).

S. Sugiuara and A. Shimizu, Canonical thermal pure
quantum state, Physical Review Letters 111, 1 (2013)
[arXiv:1302.3138]

A. Shimizu and K. Fujikura, Quantum violation of fluctuation-dissipation theorem, Journal of Statistical Mechanics: Theory and Experiment 2017, 10.1088/1742-5468/aa5a6f (2017) [arXiv:1610.03161]

H. Endo, C. Hotta, and A. Shimizu, From Linear to Nonlinear Responses of Thermal Pure Quantum States, Physical Review Letters 121, 220601 (2018) [arXiv:1806.02054]
Anomalous behavior of magnetic susceptibility obtained by quench experiments in isolated quantum systems: Supplemental Material

Yuuya Chiba* and Akira Shimizu†
Komaba Institute for Science, The University of Tokyo, 3-8-1 Komaba, Meguro, Tokyo 153-8902, Japan and
Department of Basic Science, The University of Tokyo, 3-8-1 Komaba, Meguro, Tokyo 153-8902, Japan

Kenichi Asano‡
Center for Education in Liberal Arts and Sciences, Osaka University, Toyonaka, Osaka 560-0043, Japan
(Dated: December 2, 2019)

A. Quench susceptibility

We deal with a quantum spin system on a $d$-dimensional hypercubic lattice $\Omega_N$ with linear size $L$ and $N = |\Omega_N| = L^d$ spins centered at $r = 0$. The unit of length is taken as the lattice constant. We consider a quantum quench process where the weak additional field $\Delta h(r)$, with wavenumber $k$ and magnitude $\Delta h_k$, is applied suddenly at $t = 0$ and after that the expectation value of $\hat{\sigma}_r^z$ evolves in time as

$$\langle \hat{\sigma}_r^z \rangle_{\text{qch}}^{\text{qch}}(t) = \langle \hat{\sigma}_r^z \rangle_{\text{ini}} + \sum_{r' \in \Omega_N} \phi_N^{\text{qch}}(r - r'; t) \Delta h(r') + O(\Delta h^2_k),$$

(S1)

where $(\bullet)_{\text{ini}} = \text{Tr}[\hat{\rho}_{\text{ini}} \bullet]$. Here, $\phi_N^{\text{qch}}(r; t) = \beta \langle \delta \hat{\sigma}_0^z; \delta \hat{\sigma}_r^z \rangle_{\text{ini}} - \beta \langle \delta \hat{\sigma}_0^z; \delta \hat{\sigma}_r^z(t) \rangle_{\text{ini}}$ is a periodic function of $r$ with period $L$, where $\hat{X}(t) = e^{iH(t)} \hat{X} e^{-iH(t)}$ is the Heisenberg operator and $\langle \hat{X}; \hat{Y} \rangle_{\text{ini}} = \frac{1}{\beta} \int_0^\beta \text{d}u \langle e^{uH}\hat{X} e^{-uH}\hat{Y} \rangle_{\text{ini}}$ is the canonical correlation. Then, the response of $\hat{m}_k$ at time $t$ reads $\Delta \langle \hat{m}_k \rangle_{\text{qch}}(t) = \langle \hat{m}_k \rangle_{\text{qch}}(t) - \langle \hat{m}_k \rangle_{\text{ini}} = \chi_N^{\text{qch}}(k; t) \Delta h_k + O(\Delta h^2_k)$, where

$$\chi_N^{\text{qch}}(k; t) = \sum_{r \in \Omega_N} e^{-ik \cdot r} \phi_N^{\text{qch}}(r; t) = \beta N \langle \delta \hat{m}_k^{\text{qch}}(t) \rangle_{\text{ini}} - \beta N \langle \delta \hat{m}_k^{\text{qch}} + \delta \hat{m}_k^{\text{qch}} \rangle_{\text{ini}}.$$ 

(S2)

Since we are only interested in the relaxed value of $\Delta \langle \hat{m}_k \rangle_{\text{qch}}(t)$, we define the quench susceptibility $\chi_N^{\text{qch}}(k)$ as the long time average of $\chi_N^{\text{qch}}(k; t)$,

$$\chi_N^{\text{qch}}(k) = \lim_{T \to \infty} \frac{\chi_N^{\text{qch}}(k; t)^T}{T} = \beta N \langle \delta \hat{m}_k^{\text{qch}}(t) \rangle_{\text{ini}} - \beta N \langle \delta \hat{m}_k^{\text{qch}} + \delta \hat{m}_k^{\text{qch}} \rangle_{\text{ini}}.$$ 

(S3)

Here the energy diagonal part of an operator $\hat{X}$ is given as $\hat{X}^0 = \lim_{T \to \infty} \hat{X}(t) = \sum_{\nu, \nu'} \delta_{E_{\nu}, E_{\nu'}} |\nu\rangle \langle \nu| \hat{X} |\nu\rangle \langle \nu'|$.

Figures S1(a) and (b) show the time dependence of $\chi_N^{\text{qch}}(0; t)$ and $\chi_N^{\text{qch}}(\pi/2; t)$ in 1D XYZ model, respectively. For $t \geq 5$, i.e., after the transient regime, $\chi_N^{\text{qch}}(k; t)$ fluctuates in time around the quench susceptibility $\chi_N^{\text{qch}}(k)$, which is shown by the solid line. When the system size $N$ is increased as 8, 12 and 16, this time fluctuation gets small. Therefore, if $\chi_N^{\text{qch}}(k; t)$ is measured after the transient regime in the system with sufficiently large spin number, $N$, the measured value of $\chi_N^{\text{qch}}(k; t)$ will be close to $\chi_N^{\text{qch}}(k)$.

B. Thermodynamic susceptibilities

We consider the isothermal quasistatic process in which the weak additional field is applied gradually and the final state of the system is the canonical Gibbs one, $\rho^G_{\text{fin}} \propto \exp \left(-\beta (H(h) - \sum_{r \in \Omega_N} \hat{\sigma}_r^z \Delta h(r)) \right)$, with the same inverse

* chiba@as.c.u-tokyo.ac.jp
† shmz@as.c.u-tokyo.ac.jp
‡ asano@celas.osaka-u.ac.jp
temperature as the initial one. Then, the expectation value of $\hat{\sigma}_r^z$ changes by

$$
\Delta \langle \hat{\sigma}_r^z \rangle^T = \text{Tr}[\hat{\rho}_{\text{fin}} \hat{\sigma}_r^z] - \langle \hat{\sigma}_r^z \rangle_{\text{ini}} = \sum_{r' \in \Omega_N} \phi_N^r(r - r') \Delta h(r') + O(\Delta h_k^2), \tag{S4}
$$

where $\phi_N^r(r) = \beta (\Delta \hat{\sigma}_r^z)_{\text{ini}}$ is defined as a periodic function of $r$ in the same way as $\phi_N^{qch}(r; t)$. From Eq. (S4), the response of $\hat{m}_k$ is given as $\Delta \langle \hat{m}_k \rangle^T = \text{Tr}[\hat{\rho}_{\text{fin}} \hat{m}_k] - \langle \hat{m}_k \rangle_{\text{ini}} = \chi_N^T(k) \Delta h_k + O(\Delta h_k^2)$, where

$$
\chi_N^T(k) = \sum_{r \in \Omega_N} e^{-ikr} \phi_N^r(r) = \beta N \langle \hat{\delta}_r \hat{m}_k \rangle_{\text{ini}} \tag{S5}
$$

is the isothermal susceptibility.

We also consider the adiabatic quasistatic process in which the weak additional field is applied gradually and the final state of the system is the canonical Gibbs one $\hat{\rho}_{\text{fin}} = \exp(-\beta_{\text{fin}} \hat{H}(h) - \sum_{r \in \Omega_N} \hat{\sigma}_r^z \Delta h(r))$ with the same entropy as the initial one, $-\text{Tr}[\hat{\rho}_{\text{ini}} \ln \hat{\rho}_{\text{fin}}]/N = -\text{Tr}[\hat{\rho}_{\text{ini}} \ln \hat{\rho}_{\text{fin}}]/N$. From this condition, the final inverse temperature $\beta_{\text{fin}}^S$ is determined as

$$
\beta_{\text{fin}}^S = \beta + \sum_{r \in \Omega_N} \frac{\langle \Delta \hat{H}(h) \hat{\delta}_r^z \rangle_{\text{ini}}}{\langle \Delta \hat{H}(h)^2 \rangle_{\text{ini}}} \Delta h(r) + O(\Delta h_k^2). \tag{S6}
$$

The change of the expectation value of $\hat{\sigma}_r^z$, $\Delta \langle \hat{\sigma}_r^z \rangle^S = \text{Tr}[\hat{\rho}_{\text{fin}}^S \hat{\sigma}_r^z] - \langle \hat{\sigma}_r^z \rangle_{\text{ini}}$, is given as

$$
\Delta \langle \hat{\sigma}_r^z \rangle^S = \Delta \langle \hat{\sigma}_r^z \rangle^T - (\beta_{\text{fin}}^S - \beta) \langle \Delta \hat{H}(h) \hat{\delta}_r^z \rangle_{\text{ini}} + O(\Delta h_k^2) = \sum_{r' \in \Omega_N} \phi_N^r(r - r') \Delta h(r') + O(\Delta h_k^2), \tag{S7}
$$

where

$$
\phi_N^S(r) = \phi_N^r(r) - \beta \frac{(\Delta \hat{H}(h) \hat{\delta}_0^z)^2_{\text{ini}}}{\langle \Delta \hat{H}(h)^2 \rangle_{\text{ini}}}. \tag{S8}
$$

Then, the response of $\hat{m}_k$ is also given as $\Delta \langle \hat{m}_k \rangle^S = \text{Tr}[\hat{\rho}_{\text{fin}}^S \hat{m}_k] - \langle \hat{m}_k \rangle_{\text{ini}} = \chi_N^S(k) \Delta h_k + O(\Delta h_k^2)$, where

$$
\chi_N^S(k) = \sum_{r \in \Omega_N} e^{-ikr} \phi_N^S(r) = \chi_N^T(k) - \beta N \frac{(\Delta \hat{H}(h) \hat{\delta}_0^z)^2_{\text{ini}}}{\langle \Delta \hat{H}(h)^2 \rangle_{\text{ini}}} \tag{S9}
$$

is the adiabatic susceptibility.

### C. Relations between the susceptibilities

From Eq. (S9), we have $\chi_N^S(k) = \chi_N^T(k) - \frac{1}{c_h}(\Delta m_k/\partial T)_{\text{ini}}^2$, where $c_h = \beta^2 \langle \Delta \hat{H}(h)^2 \rangle_{\text{ini}}/N$ is the specific heat at constant magnetic field and $(\Delta m_k/\partial T)_h = -\beta (\Delta \hat{H}(h) \hat{\delta}_0^z)_{\text{ini}}$. In contrast to $k = 0$ component, $(\Delta m_k/\partial T)_h = 0$ hold for all $k \neq 0$ due to the translation invariance of $\hat{H}(h)$, yielding

$$
\chi_N^S(k) = \chi_N^T(k) \quad \text{for all } k \neq 0. \tag{S10}
$$

FIG. S1. Time dependence of (a) $\chi_N^{qch}(0; t)$ and (b) $\chi_N^{qch}(\pi/2; t)$ in XYZ model, with the parameters, $J_x + J_y = 0.6$, $J_x - J_y = 1.2$, $J_z = 1.0$, $h = 0.8$ and $\beta = 0.15$. We take $N = 8, 12, 16$. The solid lines in (a) and (b) show $\chi_N^{qch}(0)$ and $\chi_N^{qch}(\pi/2)$ for each $N$, respectively. As the system size $N$ is increased, the time fluctuation of $\chi_N^{qch}(k; t)$ from its time average $\chi_N^{qch}(k)$ gets small in both (a) and (b).
Comparing Eqs. (S3) and (S9), we have
\[
\chi_N^S(0) - \chi_{N(q)}^c(0) = \beta N (\delta \hat{m}_0^0; \delta \hat{m}_0^0)_{ini} - \beta N |(\delta \hat{H}(h) \delta \hat{m}_{k=0}^0)_{ini}|^2 \tag{S11}
\]
\[
= \beta N \left( \sum_{\nu} \frac{e^{-\beta E_{\nu}}}{Z} \langle \nu | \delta \hat{\sigma}_0^z | \nu \rangle^2 \right) - \beta N \left( \sum_{\nu} \frac{e^{-\beta E_{\nu}}}{Z} \delta E_{\nu} \langle \nu | \delta \hat{\sigma}_0^z | \nu \rangle^2 \right) / \left( \sum_{\nu} \frac{e^{-\beta E_{\nu}}}{Z} \delta E_{\nu}^2 \right) \geq 0 \tag{S12}
\]
from the Cauchy-Schwarz inequality. Here \( \langle \nu | \hat{m}_0^0; \delta \hat{m}_0^0 | \nu \rangle = \delta_{\nu,\nu'} \langle \nu | \hat{m}_0^0; \delta \hat{m}_0^0 | \nu \rangle \) is the simultaneous eigenstate of \( \hat{H}(h) \), translation operators and \( \hat{m}_0^0 \). This yields the general relation (7) [1-3]. The equality for finite \( N \) holds if and only if \( \langle \nu | \delta \hat{\sigma}_0^z | \nu \rangle \propto \delta E_{\nu}/N \) for all \( \nu \), which is not satisfied in almost all systems. In the thermodynamic limit \( N \to \infty \), the condition for the equality is relaxed as follows.

Result (i) : From Eq. (S12), the necessary and sufficient condition for Eq. (4) is given as
\[
\lim_{N \to \infty} N \left( \sum_{\nu} \frac{e^{-\beta E_{\nu}}}{Z} \langle \nu | \delta \hat{\sigma}_0^z | \nu \rangle^2 \right) = \lim_{N \to \infty} N \left( \sum_{\nu} \frac{e^{-\beta E_{\nu}}}{Z} \delta E_{\nu} \langle \nu | \delta \hat{\sigma}_0^z | \nu \rangle^2 \right) / \left( \sum_{\nu} \frac{e^{-\beta E_{\nu}}}{Z} \delta E_{\nu}^2 \right), \tag{S13}
\]
which holds if and only if Eq. (8) is fulfilled for almost all \( |\nu\rangle \) in a narrow energy region \( |\delta E_{\nu}| \lesssim T \sqrt{\epsilon_{ch} N} \).

We can relate condition (8) with the ordinary ETH more directly. Let us introduce the microcanonical average over the energy shell \( (E - \delta, E) \) as \( (\bullet)_{mc}(E/N) \) and the number of states in \( (E - \delta, E) \) as \( W(E/N) \), assuming that the energy width \( \delta \) can be taken as \( \delta_N = \Theta(1/N^{1+\alpha}) \), where \( \alpha \) is a small positive number. Then we can evaluate \( \langle \delta \hat{\sigma}_0^z \rangle_{ini} \) as
\[
\langle \delta \hat{\sigma}_0^z \rangle_{ini} = \sum_{\nu} \frac{e^{-\beta E_{\nu}}}{Z} \langle \nu | \delta \hat{\sigma}_0^z | \nu \rangle = \frac{\int \, d\epsilon \, \text{exp}(N(s_N(\epsilon) - \beta e) \langle \delta \hat{\sigma}_0^z \rangle_{mc}(\epsilon))}{\int \, d\epsilon \, \text{exp}(N(s_N(\epsilon) - \beta e))} + O(\delta_N), \tag{S14}
\]
where \( s_N(\epsilon) = \log W(\epsilon/N) \). Except at a phase transition point, we can use the saddle point method and obtain
\[
\langle \delta \hat{\sigma}_0^z \rangle_{ini} = \langle \delta \hat{\sigma}_0^z \rangle_{mc}(\epsilon^*) + O(1/N), \tag{S15}
\]
where \( \epsilon^* \) is determined by \( s_N'(\epsilon^*) = \frac{ds_N}{d\epsilon}(\epsilon^*) = \beta \). In the same way,
\[
\langle \hat{H}(h) \rangle_{ini}/N = \epsilon^* + O(1/N), \tag{S16}
\]
\[
\sum_{\nu} \frac{e^{-\beta E_{\nu}}}{Z} \delta E_{\nu}^2 / N = 1/|s_N''(\epsilon^*)| + o(1), \tag{S17}
\]
\[
\sum_{\nu} \frac{e^{-\beta E_{\nu}}}{Z} \delta E_{\nu} \langle \nu | \delta \hat{\sigma}_0^z | \nu \rangle = \frac{\frac{d}{d\epsilon} \langle \delta \hat{\sigma}_0^z \rangle_{mc}(\epsilon^*)}{|s_N''(\epsilon^*)|} + o(1), \tag{S18}
\]
\[
N \sum_{\nu} \frac{e^{-\beta E_{\nu}}}{Z} \langle \nu | \delta \hat{\sigma}_0^z | \nu \rangle^2 = N \sum_{\nu} \frac{e^{-\beta E_{\nu}}}{Z} |\langle \nu | \delta \hat{\sigma}_0^z | \nu \rangle - \langle \delta \hat{\sigma}_0^z \rangle_{mc}(E_{\nu}/N)|^2 + \frac{\left( \frac{d}{d\epsilon} \langle \delta \hat{\sigma}_0^z \rangle_{mc}(\epsilon^*) \right)^2}{|s_N''(\epsilon^*)|} + o(1) \tag{S19}
\]
can be shown. From Eqs. (S17), (S18) and (S19), the following result holds.

Result (i') : Eq. (4) or its equivalent condition (8) holds if and only if
\[
N \sum_{\nu} \frac{e^{-\beta E_{\nu}}}{Z} \langle \nu | \delta \hat{\sigma}_0^z | \nu \rangle - \langle \delta \hat{\sigma}_0^z \rangle_{mc}(E_{\nu}/N)^2 = o(1), \tag{S20}
\]
which is similar to the weak ETH [4-6] in that it requires almost all \( \langle \nu | \delta \hat{\sigma}_0^z | \nu \rangle \) should be close to \( \langle \delta \hat{\sigma}_0^z \rangle_{mc}(E_{\nu}/N) \). Condition (S20) will be satisfied in nonintegrable systems, where \( \langle \nu | \delta \hat{\sigma}_0^z | \nu \rangle \) is often exponentially close to \( \langle \delta \hat{\sigma}_0^z \rangle_{mc}(E_{\nu}/N) \) [7, 8]. Note that, there are some integrable models which satisfy the ordinary weak ETH [4, 5, 9] but do not satisfy condition (S20). This fact can be confirmed by the violation of its equivalent Eq. (4), \( \chi_N^{mc}(0) = \chi_N^c(0) \). (See main text.) Indeed, condition (S20) is more stringent than the ordinary weak ETH [4, 5, 9] in that condition (S20) requires \( \langle \nu | \delta \hat{\sigma}_0^z | \nu \rangle - \langle \delta \hat{\sigma}_0^z \rangle_{mc}(E_{\nu}/N)^2 \) to be typically \( o(1/N) \), while the ordinary weak ETH [4, 5, 9] allows this quantity to be larger than \( \Theta(1/N) \). Here, functions of \( N, f_N \) and \( g_N \), satisfy \( g_N = \Theta(f_N) \), if there are positive constants \( 0 < c_1 \leq c_2 < \infty \) such that \( c_1 f_N \leq g_N \leq c_2 f_N \) holds for sufficiently large \( N \).

Eqs. (S3) and (S5) give a relation between \( k \neq 0 \) components,
\[
\chi_N^T(k) - \chi_N^{qch}(k) = \beta N \langle \hat{m}_0^k; \hat{m}_0^k \rangle_{ini} = \beta N \sum_{\nu} \frac{e^{-\beta E_{\nu}}}{Z} \sum_{\nu'} \delta E_{\nu,\nu'} \delta K_{\nu,\nu} + k \langle \nu' | \delta \hat{\sigma}_0^z | \nu \rangle^2, \tag{S21}
\]
where the crystal momentum $\mathbf{K}_\nu$ is defined so that the eigenvalue of $r$ sites translation operator is written as $e^{-i\mathbf{K}_\nu \cdot \mathbf{r}}$ and we used $|\langle \nu'| \hat{m}_{\mathbf{k}} | \nu \rangle | = \delta_{\mathbf{K}_\nu, \mathbf{K}_\nu + \mathbf{k}} |\langle \nu'| \hat{\sigma}_0 | \nu \rangle |$. Therefore Eqs. (S10) and (S21) yield the following.

Result (ii) : Eq. (5) holds if and only if the off-diagonal elements are small so that

$$
\sum_{\nu} e^{-\beta E_{\nu}} \frac{N}{Z} \sum_{\nu'} \delta_{E_{\nu}, E_{\nu'}} \delta_{\mathbf{K}_\nu, \mathbf{K}_\nu + \mathbf{k}} |\langle \nu'| \hat{\sigma}_0 | \nu \rangle |^2 = o(1) \quad \text{for all } \mathbf{k} \neq \mathbf{0}.
$$

(S22)

This condition can be rephrased as Eq. (10), which is weaker than the ordinary off-diagonal ETH [10–13] as explained below using XY model.

### D. Analysis of the quench process using thermodynamics

In this section, we assume that thermalization occurs after the quench process, where the small uniform magnetic field $\Delta h_0$ is applied. From this assumption, the state of the system, which evolves from the initial equilibrium state, relaxes to another equilibrium state. Since the expectation value of the post-quench Hamiltonian $\hat{H}(h + \Delta h_0)$ does not change before and after the quench, the initial and the final equilibrium states satisfy

$$
e + \Delta e = \langle \hat{H}(h + \Delta h_0) \rangle_{\text{ini}} / N = \langle \hat{H}(h + \Delta h_0) \rangle_{\text{ini}} / N = \langle \hat{H}(h) \rangle_{\text{ini}} / N - \langle \hat{m}_0 \rangle_{\text{ini}} \Delta h_0 = e - m_0 \Delta h_0, $$

where $e$ and $e + \Delta e$ are the initial and the final equilibrium values of the energy per site and $m_0$ is the initial equilibrium value of the $\mathbf{k} = \mathbf{0}$ component of the magnetization. From Eq. (S23), the change of the entropy is

$$
\Delta s = \frac{\partial s}{\partial e} (e, h) \Delta e + \frac{\partial s}{\partial h} (e, h) \Delta h_0 + \mathcal{O}((\Delta h_0)^2) = \beta \Delta e + \beta m_0 \Delta h_0 + \mathcal{O}((\Delta h_0)^2) = \mathcal{O}((\Delta h_0)^2),
$$

where $\beta$ is the initial inverse temperature. Eq. (S24) is consistent with Eq. (4). Note that the change of the energy per site $\Delta e = -m_0 \Delta h_0$ and the change of the inverse temperature

$$
\Delta \beta = \beta \left( \frac{\partial m_0}{\partial T} \right)_h \Delta h_0 + \mathcal{O}((\Delta h_0)^2)
$$

are $\mathcal{O}(\Delta h_0)$ because $h \neq 0$. This results in $\chi^{\text{qh}}(0) < \chi^T(0)$.

### E. Proof of (iii)

From condition (12), we can define $\chi^{\text{inf}}(\mathbf{k}) = \lim_{N \to \infty} \sum_{\mathbf{r} \in \Omega_N} e^{-i \mathbf{k} \cdot \mathbf{r}} \phi^T_{\infty}(\mathbf{r})$, which is uniformly continuous in $\mathbf{k}$ by the property of Fourier transform. From Eq. (S5),

$$
|\chi^T_{\infty}(\mathbf{k}) - \chi^{\text{inf}}(\mathbf{k})| \leq \left| \sum_{\mathbf{r} \in \Omega_N} e^{-i \mathbf{k} \cdot \mathbf{r}} (\phi^T_{\infty}(\mathbf{r}) - \phi^T_{\infty}(\mathbf{r})) \right| + \left| \lim_{N' \to \infty} \sum_{\mathbf{r} \in \Omega_{N'} \setminus \Omega_N} e^{-i \mathbf{k} \cdot \mathbf{r}} \phi^T_{\infty}(\mathbf{r}) \right| \leq \left| \sum_{\mathbf{r} \in \Omega_N} \phi^T_{\infty}(\mathbf{r}) - \phi^T_{\infty}(\mathbf{r}) \right| + \left| \lim_{N' \to \infty} \sum_{\mathbf{r} \in \Omega_{N'} \setminus \Omega_N} \phi^T_{\infty}(\mathbf{r}) \right|. \quad \text{(S27)}
$$

In the $N \to \infty$ limit, the first term and the second term of Eq. (S27) converges to 0 from condition (13) and (12), respectively. As a result, $\chi^T_{\infty}(\mathbf{k})$ converges to $\chi^{\text{inf}}(\mathbf{k})$ in the $N \to \infty$ limit, $\chi^T_{\infty}(\mathbf{k}) = \chi^{\text{inf}}(\mathbf{k})$ for all $\mathbf{k}$, which implies that $\chi^T_{\infty}(\mathbf{k})$ is also uniformly continuous in $\mathbf{k}$. \(\square\)

Note that condition (13) is essential for the uniform continuity of $\chi^T_{\infty}(\mathbf{k})$. Since $\phi^T_{\infty}(\mathbf{r}) = \phi^T_{\infty}(\mathbf{r})$ follows from Eq. (S8), condition (12) holds also for $\phi^S$. However condition (13) does not hold for $\phi^S$:

$$
\lim_{N \to \infty} \sum_{\mathbf{r} \in \Omega_N} |\phi^S_{\infty}(\mathbf{r}) - \phi^S_{\infty}(\mathbf{r})| = \lim_{N \to \infty} \sum_{\mathbf{r} \in \Omega_N} |\phi^S_{\infty}(\mathbf{r}) - \phi^T_{\infty}(\mathbf{r}) + \phi^T_{\infty}(\mathbf{r})| = \chi^T_{\infty}(0) - \chi^S_{\infty}(0) > 0, \quad \text{(S28)}
$$

which is consistent with the discontinuity of $\chi^S_{\infty}(\mathbf{k})$ at $\mathbf{k} = \mathbf{0}$.

In Fig. S2, we verify the conditions for (iii), (a) $\phi^T_{\infty}(\mathbf{r})$ decays fast enough and (b) finite size effects of $\phi^T_{\infty}(\mathbf{r})$ are small, in XYZ model. To this end, we introduce two quantities, (a) $D_N = \sum_{\mathbf{r} \in \Omega_N} |\phi^T_{\infty}(\mathbf{r})|$ and (b) $F_N = \sum_{\mathbf{r} \in \Omega_N} |\phi^T_{\infty}(\mathbf{r}) - \phi^T_{\infty}(\mathbf{r})|$, where $N_{\text{max}}$ is taken as large as possible. Fig. S2 (a) shows $N$ dependence of $D_N$ in XYZ model. As $N$ increases, $D_N$ is saturated, suggesting that condition (12) holds. Fig. S2 (b) shows $N$ dependence of $F_N$ in the same system. As $N$ increases, $F_N$ decreases, suggesting that condition (13) holds.
FIG. S2. Verification of conditions for (iii) in XYZ model, with the same parameters as in Fig. S1. We investigate the $N$ dependence of (a) $D_N$, the sum of $|\phi^T_{N_{max}}(r)|$ over all $r \in \Omega_N$, and (b) $F_N$, the sum of $|\phi^T_N(r) - \phi^T_{N_{max}}(r)|$ over all $r \in \Omega_N$. We take $N_{max} = 16$.

F. Analytic solutions in 1D XY model

We here describe the analytic solutions $\chi^{\text{ch}}(k)$, $\chi^{S}(k)$ and $\chi^{T}(k)$ in 1D XY model and verify whether the above relations hold or not in this model. By defining $J_s = J_x + J_y$, $J_a = J_x - J_y$ and $\varepsilon_k = \sqrt{(J_s \cos k + h)^2 + J_a^2 \sin^2 k}$, we can write the results as follows.

For the $k = 0$ components, we have

$$\chi^{\text{ch}}(0) = \frac{1}{2\pi} \int_0^{2\pi} dk' \frac{J_s^2 \sin^2 k' \tanh \beta \varepsilon_{k'}}{\varepsilon_{k'}^2},$$

(S29)

$$\chi^{T}(0) = \chi^{\text{ch}}(0) + \frac{\beta}{2\pi} \int_0^{2\pi} dk' \frac{(J_s \cos k' + h)^2}{\varepsilon_{k'}^2} \frac{1}{\cosh^2 \beta \varepsilon_{k'}},$$

(S30)

$$\chi^{S}(0) = \chi^{T}(0) - \left( \frac{\beta}{2\pi} \int_0^{2\pi} dk' \frac{J_s \cos k' + h}{\cosh^2 \beta \varepsilon_{k'}} \right)^2 / \left( \frac{\beta}{2\pi} \int_0^{2\pi} dk' \frac{\varepsilon_{k'}^2}{\cosh^2 \beta \varepsilon_{k'}} \right) < \chi^{T}(0).$$

(S31)

From Eqs. (S29) and (S31), Eq. (4) is violated except at the case of free spin model ($J_s = J_a = 0$) or critical point of transverse field Ising model ($|J_s| = |J_a| = |h|$). Therefore, condition (8) does not hold, whereas the ordinary weak ETH [4, 5, 9] is satisfied in this model.

For the $k \neq 0$ components, Eq. (5) is satisfied as

$$\chi^{T}(k) = \chi^{S}(k) = \chi^{\text{ch}}(k)$$

$$= \frac{1}{2\pi} \int_0^{2\pi} dk' \frac{\varepsilon_{k'} \varepsilon_{k' + k} - (J_s \cos k' + h)(J_s \cos (k' + k) + h) + J_a^2 \sin k' \sin (k' + k)}{2 \varepsilon_{k'} \varepsilon_{k' + k}} 
\times \tanh \beta \varepsilon_{k'} + \tanh \beta \varepsilon_{k' + k}$$

$$+ \frac{1}{2\pi} \int_0^{2\pi} dk' \frac{\sinh \beta (\varepsilon_{k'} - \varepsilon_{k' + k})}{\varepsilon_{k'} - \varepsilon_{k' + k}} \frac{1}{\cosh \beta \varepsilon_{k'} \cosh \beta \varepsilon_{k' + k}}$$

$$\times \frac{\varepsilon_{k'} \varepsilon_{k' + k} + (J_s \cos k' + h)(J_s \cos (k' + k) + h) - J_a^2 \sin k' \sin (k' + k)}{2 \varepsilon_{k'} \varepsilon_{k' + k}}.$$

(S32)

This indicates condition (S22) is satisfied in this model. From Eqs. (S30) and (S32), Eq. (5) holds and $\chi^{T}(k)$ is uniformly continuous in $k$, while $\chi^{\text{ch}}(k)$ is discontinuous at $k = 0$. Moreover, for $k \neq 0$, $\chi^{T}(k) - \chi^{\text{ch}}(k)$ scales as

$$\chi^{T}(k) - \chi^{\text{ch}}(k) = \frac{\beta}{2N} \left( \frac{1}{\cosh^2 \beta \varepsilon_{k/2}} + \frac{1}{\cosh^2 \beta \varepsilon_{-k/2}} \right) + \exp(-\Theta(N)) = \Theta(1/N),$$

(S33)

because some off-diagonal elements $|\langle \nu | \hat{\sigma}_z^\dagger | \nu' \rangle|$ that are appeared in Eq. (S22) scale as $\Theta(1/N)$. That indicates the ordinary off-diagonal ETH [10–13], which requires exponentially fast decay of all off-diagonal elements, is not satisfied in this model.
G. Additional demonstrations of (ii)-(iv)

Although condition (S22) is weaker than the ordinary off-diagonal ETH [10–13] as mentioned above, there are some models which do not satisfy it such as the longitudinal field Ising model ($J_x = J_y = 0$). Fig. S3 (a) shows $k$ dependence of $\chi^\text{qch}_N(k)$, $\chi^T_N(k)$ and $\chi^S_N(k)$ in this model. Since $\hat{m}_k$ is conserved, $\chi^\text{qch}_N(k) = 0$ holds, while $\chi^T_N(k)$ and $\chi^S_N(k) > 0$ for all $k$, resulting in the violation of Eq. (5) or equivalent condition (S22). In contrast, Fig. S3 (b) shows how the susceptibilities behave when a small nonintegrability ($J_x + J_y = 0.006$, $J_x - J_y = 0.012$) is added to this system. For the $k = 0$ component, each susceptibility, $\chi^\text{qch}_N(0)$, $\chi^T_N(0)$ and $\chi^S_N(0)$, in (b) is almost the same as one in (a), and Eq. (4) is not satisfied in both (a) and (b). On the other hand, the $k \neq 0$ component $\chi^\text{qch}_N(k)$ differs dramatically between (a) and (b), and Fig. S3 (b) indicates Eq. (5) is satisfied in (b). These results suggest that Eq. (5) is easily satisfied as in (b), while we need more nonintegrability for Eq. (4). Reflecting these facts, $\chi^\text{qch}_N(k)$ is discontinuous at $k = 0$ in only (b), while $\chi^T_N(k)$ is uniformly continuous in both (a) and (b).

![Fig. S3. $k$ dependence of $\chi^\text{qch}_N(k)$, $\chi^T_N(k)$ and $\chi^S_N(k)$ in (a) longitudinal field Ising model ($J_x = J_y = 0$) and (b) XYZ model with small $J_x$ and $J_y$ ($J_x + J_y = 0.006$, $J_x - J_y = 0.012$). $J_z = 1.0$, $h = 0.8$ and $\beta = 0.15$ are fixed. We take $N = 12$-14 and $k = 2\pi n_k/N$ with $n_k \in \mathbb{Z}$.

H. Relation to Kubo formula

The susceptibility obtained by Kubo formula [14, 15] is given as

$$\chi^\text{Kubo}_N(k, \omega + i \varepsilon) = \int_0^\infty dt e^{i\omega t - \varepsilon} N \langle [\hat{m}_k(t), -\hat{m}_k] \rangle_{\text{ini}}$$

(S34)

$$= \chi^T_N(k) + (i\omega - \varepsilon) \int_0^\infty dt e^{i\omega t - \varepsilon} \beta N \langle \delta \hat{m}_k; \delta \hat{m}_k \rangle_{\text{ini}},$$

(S35)

where $[\hat{X}, \hat{Y}] = \hat{X}\hat{Y} - \hat{Y}\hat{X}$ is the commutator, $\omega$ is the angular frequency and $\varepsilon$ is a small positive number.

From Eqs. (S35) and (S3), the following holds for all $N$ and for all $k$,

$$\lim_{\varepsilon \to +0} \chi^\text{Kubo}_N(k, 0 + i \varepsilon) = \chi^T_N(k) - \beta N \langle \delta \hat{m}_k^0; \delta \hat{m}_k^0 \rangle_{\text{ini}} = \chi^\text{qch}_N(k).$$

(S36)

[1] R. M. Wilcox, Bounds for the isothermal, adiabatic, and isolated static susceptibility tensors, Physical Review 174, 624 (1968).
[2] P. Mazur, Non-ergodicity of phase functions in certain systems, Physica 43, 533 (1969).
[3] M. Suzuki, Ergodicity, constants of motion, and bounds for susceptibilities, Physica 51, 277 (1971).
[4] G. Biroli, C. Kollath, and A. M. Läuchli, Effect of rare fluctuations on the thermalization of isolated quantum systems, Physical Review Letters 105, 1 (2010).
[5] E. Iyoda, K. Kaneko, and T. Sagawa, Fluctuation Theorem for Many-Body Pure Quantum States, Physical Review Letters 119, 1 (2017).
[6] T. Mori, Weak eigenstate thermalization with large deviation bound, arXiv , 1 (2016), arXiv:1609.09776.
[7] W. Beugeling, R. Moessner, and M. Haque, Finite-size scaling of eigenstate thermalization, Physical Review E - Statistical, Nonlinear, and Soft Matter Physics 89, 1 (2014).
[8] R. Steinigeweg, A. Khodja, H. Niemeyer, C. Gogolin, and J. Gemmer, Pushing the limits of the eigenstate thermalization hypothesis towards mesoscopic quantum systems, Physical Review Letters 112, 1 (2014).
[9] T. Kuwahara and K. Saito, Ensemble equivalence and eigenstate thermalization from clustering of correlation, arXiv (2019), arXiv:1905.01886.
[10] M. Srednicki, The approach to thermal equilibrium in quantized chaotic systems systems, Journal of Physics A : Mathematical and General 32, 1163 (1999).
[11] L. D’Alessio, Y. Kafri, A. Polkovnikov, and M. Rigol, From quantum chaos and eigenstate thermalization to statistical mechanics and thermodynamics, Advances in Physics 65, 239 (2016).
[12] T. Mori, T. N. Ikeda, E. Kaminishi, and M. Ueda, Thermalization and prethermalization in isolated quantum systems: A theoretical overview, Journal of Physics B: Atomic, Molecular and Optical Physics 51, 10.1088/1361-6455/aabcdf (2018).
[13] F. Anza, C. Gogolin, and M. Huber, Eigenstate Thermalization for Degenerate Observables, Physical Review Letters 120, 150603 (2018).
[14] R. Kubo, Statistical Mechanical Theory of Irreversible Processes. I. General Theory and Simple Applications to Magnetic and Conduction Problems, Journal of the Physical Society of Japan 12, 570 (1957), arXiv:0211006 [cs].
[15] R. Kubo, M. Toda, and N. Hashitsume, Statistical Physics II, 2nd ed. (Springer-Verlag Berlin Heidelberg, 1991).