

Control of Metastable States by Heat Flux in the Hamiltonian Potts Model

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The local equilibrium thermodynamics is a basic assumption of macroscopic descriptions of the out of equilibrium dynamics for Hamiltonian systems. We numerically analyze the Hamiltonian Potts model in two dimensions to study the violation of the assumption for phase coexistence in heat conduction. We observe that the temperature of the interface between ordered and disordered states deviates from the equilibrium transition temperature, indicating that metastable states at equilibrium are stabilized by the influence of a heat flux. We also find that the deviation is described by the formula proposed in an extended framework of the thermodynamics.

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Introduction.—The macroscopic dynamics of a Hamiltonian system driven by nonequilibrium boundary conditions is expected to be described by hydrodynamic equations with local equilibrium thermodynamics [1–6]. However, there are exceptional cases, such as the shear flow near a liquid-gas critical point [7], where suppression of critical fluctuations by the shear flow modifies the thermodynamic properties [8], and as a result, the local equilibrium thermodynamics is violated. A natural question is whether the violation of the local equilibrium thermodynamics occurs except at critical points. The aim of this Letter is to provide a definitive example by studying nonequilibrium dynamics near the first-order transition point.

A first-order phase transition is a different type of singularity than the critical point [9–12]. A characteristic feature of a first-order phase transition is the existence of hysteresis [13]. For an order-disorder transition [14,15], the observed transition temperature when a material is cooled from the disordered state is lower than the transition temperature when the same material is heated from the ordered state. Such transition temperatures, generally both in cooling and heating, deviate from the equilibrium transition temperature T_c . Thus, the supercooled disordered or the superheated ordered states are often observed as a

transient dynamical process. These observations may suggest that the metastable states become steady states when the system sets up at nonequilibrium conditions. To explore this possibility, we consider steady states in heat conduction where two heat baths with temperatures T_1 and T_2 are attached at the left and right sides of the system.

We assume that T_1 and T_2 satisfy $T_1 \leq T_c \leq T_2$ to observe phase coexistence, where ordered and disordered states appear at the low and high temperature sides, respectively, with a unique interface separating the two phases. Our main question in this Letter is whether the temperature of the interface is equal to T_c . If the local equilibrium thermodynamics is assumed to hold at each point of the system, the interface temperature should be equal to the equilibrium transition temperature. However, the validity of this assumption is not obvious because of the existence of metastable states. To our best knowledge, there have been no experimental studies on this question, while a rich variety of nonequilibrium phase-coexistence phenomena have been studied including flow boiling heat transfer and pattern formation in crystal growth [16–21].

We study the interface temperature by numerical simulations of a model that exhibits phase coexistence in steady heat conduction. Since there is no standard model for order-parameter dynamics with conducting energy, in this Letter, we propose the Hamiltonian Potts model in two dimensions. We first confirm the coexistence of ordered and disordered states in an isolated system by numerically solving the Hamiltonian equation. Then, imposing a heat flux at the boundary with the total energy fixed, we produce the phase coexistence in steady heat

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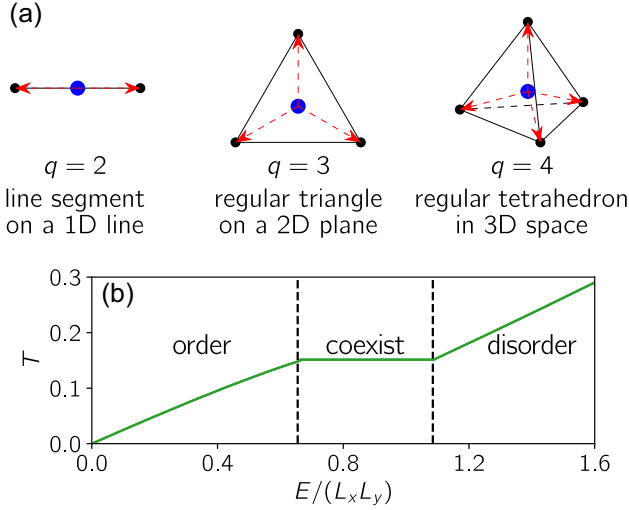


FIG. 1. (a) Examples of $(q - 1)$ simplexes with q vertices in $(q - 1)$ -dimensional space: a line segment with μ_1 and μ_2 for $q = 2$ (left), a regular triangle with μ_1 , μ_2 , and μ_3 for $q = 3$ (middle), and a regular tetrahedron with μ_1 , μ_2 , μ_3 , and μ_4 for $q = 4$ (right). (b) Phase diagram for the $q = 11$ Hamiltonian Potts model (1). The system shows the first-order phase transition at $T = T_c$ when T is changed, while the ordered and disordered states coexist in the range $E_1 < E < E_2$. T_c , E_1 , and E_2 are numerically estimated to be $T_c \simeq 0.15$, $E_1/(L_x L_y) \simeq 0.66$, and $E_2/(L_x L_y) \simeq 1.09$ [33].

conduction. A remarkable property in this system is that the interface temperature deviates from the equilibrium transition temperature. This indicates that metastable states are stabilized. That is, metastability is controlled by the heat flux. Furthermore, we find that the deviation is well fitted by the formula proposed in an extended framework of thermodynamics, which we call *global thermodynamics* [22–24]. It provides a quantitative prediction of the phase coexistence in the steady heat conduction, in contrast to other extended frameworks of thermodynamics [25–32].

Model and observables.—We study the Hamiltonian Potts model with q -fold symmetry in two dimensions. Let $\phi^a(\mathbf{r})$, $a = 1, 2, \dots, q - 1$, be a $q - 1$ components field defined in a rectangular region $D \equiv [0, L_x] \times [0, L_y]$ with $L_x > L_y$. We express $(\phi^1, \dots, \phi^{q-1})$ as ϕ . The conjugate momentum field of $\phi^a(\mathbf{r})$ is expressed by $\pi^a(\mathbf{r})$, where π represents $(\pi^1, \dots, \pi^{q-1})$. We assume the total Hamiltonian \mathcal{H} as

$$\mathcal{H}(\phi, \pi) = \int_D d^2\mathbf{r} \left\{ \frac{1}{2} \sum_{a=1}^{q-1} [(\pi^a)^2 + |\nabla\phi^a|^2] + V(\phi) \right\}, \quad (1)$$

where the potential $V(\phi)$ possesses q symmetric minima in \mathbb{R}^{q-1} . Let μ_k , $1 \leq k \leq q$, be coordinates of the q vertices for the regular $(q - 1)$ simplex in \mathbb{R}^{q-1} as illustrated in Fig. 1(a) for the cases $q = 2, 3$, and 4 , where the centroid of the simplex is located at the origin. The explicit form of μ_k is

given in Supplemental Material [33]. As a potential with minima at μ_k , we set

$$V(\phi) = \frac{1}{2} \prod_{k=1}^q \sum_{a=1}^{q-1} (\phi^a - \mu_k^a)^2. \quad (2)$$

This potential is regarded as a continuous extension of the standard q -state Potts model. We thus expect that the equilibrium statistical mechanics for \mathcal{H} describes the same phase transitions as those observed in the standard Potts model. As for the standard q -state Potts model, the model exhibits a first-order transition at a temperature T_c for $q \geq 5$ in equilibrium [37]. In this Letter, we numerically study the case $q = 11$ for $L_x = 384$ and $L_y = 64$. The system is spatially discretized with a grid spacing $\Delta x = 1/8$ [33]. The Boltzmann constant is set to unity.

We define the local kinetic energy density per unit degree of freedom and the local order parameter as

$$\hat{T}(\mathbf{r}) = \frac{\sum_{a=1}^{q-1} [\pi^a(\mathbf{r})]^2}{q-1}, \quad \hat{m}(\mathbf{r}) \equiv \sum_{a=1}^{q-1} \phi^a(\mathbf{r}) \mu_1^a, \quad (3)$$

where the direction of symmetry breaking is fixed to the μ_1 direction in our numerical simulations by choosing specific initial conditions [33]. Their average over the y direction with x fixed is expressed as

$$[\hat{A}]_x = \frac{1}{L_y} \int_0^{L_y} dy \hat{A}(\mathbf{r}), \quad (4)$$

where \hat{A} is \hat{T} or \hat{m} .

Equilibrium phase coexistence.—We first examine the equilibrium phase diagram by numerically investigating the isothermal dynamics [33]. The first-order transition is observed at $T = T_c \simeq 0.15$, where the energy density changes discontinuously at $T = T_c$. The system is occupied by the ordered and disordered states for $T < T_c$ and $T > T_c$, respectively. The phase diagram is shown in Fig. 1(b).

With the knowledge of the phase diagram, we concentrate on an isolated system with the total energy E fixed. The time evolution is given by the Hamiltonian equation

$$\partial_t \phi^a = \frac{\delta \mathcal{H}}{\delta \pi^a}, \quad \partial_t \pi^a = -\frac{\delta \mathcal{H}}{\delta \phi^a}. \quad (5)$$

Note that

$$\frac{d\mathcal{H}}{dt} = \int_D d^2\mathbf{r} \sum_{a=1}^{q-1} \nabla \cdot (\pi^a \nabla \phi^a). \quad (6)$$

For conserving energy, we assume the Neumann boundary condition in the x direction:

$$\partial_x \phi|_{x=0, L_x} = 0 \quad (7)$$

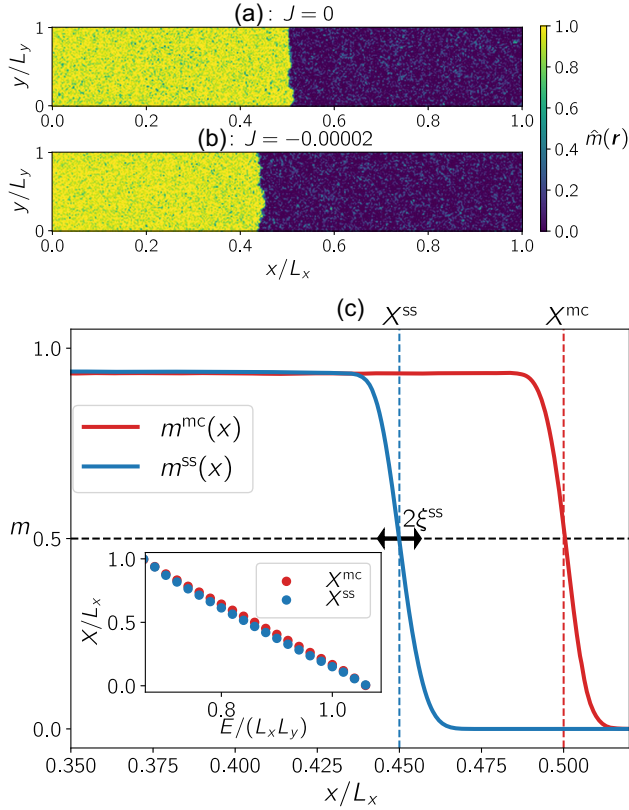


FIG. 2. (a),(b) Snapshot of the order parameter density $\hat{m}(\mathbf{r})$ in equilibrium with $J = 0$ [panel (a)] and the steady state with $J = -0.00002$ [panel (b)] for $E/(L_x L_y) = 0.88$. (c) One-dimensional order parameter profiles $m^{\text{mc}}(x)$ and $m^{\text{ss}}(x)$. The interface positions X^{mc} and X^{ss} are obtained as $m^{\text{mc}}(X^{\text{mc}}) = 0.5$ and $m^{\text{ss}}(X^{\text{ss}}) = 0.5$, respectively. The interface thickness ξ^{ss} is estimated by using $m^{\text{ss}}(x) = a - b \tanh[(x - x_0)/\xi^{\text{ss}}]$. Inset: Dependence of X^{mc} and X^{ss} on the energy density $E/(L_x L_y)$.

for any y , and periodic boundary conditions in the y direction.

We start from initial conditions with a single interface and solve the Hamiltonian equation (5) until an equilibrium state is realized [33]. Figure 2(a) shows a snapshot of the local order parameter $\hat{m}(\mathbf{r})$ in equilibrium with an interface parallel to the y axis minimizing the interfacial energy. We thus discuss the one-dimensional profile by using the average over y defined by (4).

Let $\langle \hat{A} \rangle_E$ be the long-term average of $\hat{A}(\mathbf{r})$ after equilibration, which corresponds to the expected value with respect to the microcanonical distribution with E . We show the order parameter profile $m^{\text{mc}}(x) = \langle [\hat{m}]_x \rangle_E$ in Fig. 2(c), which exhibits a typical interface structure. Here, the superscript “mc” indicates “microcanonical.” The interface position X^{mc} defined as $m^{\text{mc}}(X^{\text{mc}}) = 0.5$ decreases approximately linearly with E such that $X^{\text{mc}} = L_x$ for $E = E_1$ and $X^{\text{mc}} = 0$ for $E = E_2$. There is no interface for $E < E_1$ and $E > E_2$.

The one-dimensional temperature profile $T^{\text{mc}}(x) = \langle [\hat{T}]_x \rangle_E$ is homogeneous in x even for the phase coexistence

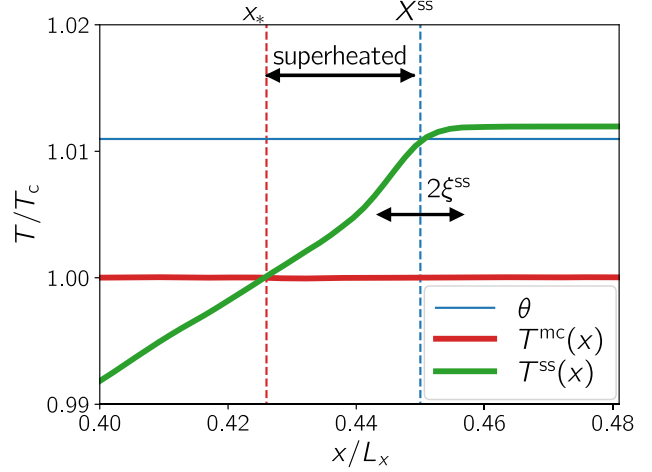


FIG. 3. One-dimensional temperature profile $T^{\text{ss}}(x)$ for $J = -0.00002$ and $T^{\text{mc}}(x)$ for $J = 0$, where $E/(L_x L_y) = 0.88$. The blue dashed line represents the interface position X^{ss} . The red dashed line represents the position x_* for $T^{\text{ss}}(x_*)$ being the equilibrium transition temperature T_c . We note that the position x_* is not at the interface region, i.e., $x_* < X^{\text{ss}} - \xi^{\text{ss}} \simeq 0.444L$.

observed in $E_1 < E < E_2$. This homogeneous temperature is equal to T_c obtained in isothermal systems as shown in Fig. 3 [33]. All these results show that the phase coexistence observed as an equilibrium state for the case $E \in [E_1, E_2]$ is an important feature of the isolated system and that the behavior is equivalent to the discontinuous change observed in the isothermal system, as displayed in the phase diagram in Fig. 1(b).

Nonequilibrium phase coexistence.—We now consider phase coexistence in heat conduction. Recall that the position of the interface is uniquely determined in isolated systems for given $E \in [E_1, E_2]$, whereas it is neutral and not under control in isothermal systems at $T = T_c$. Thus, energy-conserving heat conduction could be preferable to standard heat conduction for a detailed study of thermodynamic properties. From the thermodynamic equivalence for heat conducting states as well as equilibrium states [24,33], we expect that the obtained results for the energy-conserving heat-conduction systems will explain the phase coexistence observed in standard heat-conducting systems.

We construct a heat-conducting system where the energy flows in at $x = L_x$ and flows out at $x = 0$, while keeping the energy \mathcal{H} constant. That is, we impose a constant heat flux $(JL_y, 0)$ at $x = 0$ and $(JL_y, 0)$ at $x = L_x$. Recalling (6), we set

$$\partial_x \phi^a|_{x=0, L_x} = - \frac{JL_y \pi^a|_{x=0, L_x}}{\int_0^{L_y} dy \sum_{b=1}^{q-1} (\pi^b)^2|_{x=0, L_x}}, \quad (8)$$

which is a nonequilibrium generalization of (7).

We study the case $J = -0.00002$ [38]. Figure 2(b) is a snapshot of the order-parameter density field $\hat{m}(\mathbf{r})$ in the

steady state, which can be compared with the equilibrium case shown in Fig. 2(a). The interface in the steady state is shifted slightly to the left of the equilibrium interface. In Fig. 2(c), we display the order-parameter profile $m^{\text{ss}}(x) = \langle [\hat{m}]_x \rangle_{E,J}$, where $\langle \cdot \rangle_{E,J}$ represents the long-term average in the steady state with E and J . For the interface position X^{ss} defined by $m^{\text{ss}}(X^{\text{ss}}) = 0.5$, we find that $|X^{\text{ss}} - X^{\text{mc}}|/L \simeq 0.05$. The inset plots X^{ss} for an energy density $E/(L_x L_y)$. The relative difference $|X^{\text{ss}} - X^{\text{mc}}|/L$ becomes smaller as E approaches E_1 or E_2 .

In Fig. 3, we plot the temperature profile $T^{\text{ss}}(x) = \langle [\hat{T}]_x \rangle_{E,J}$. An important feature is that the position x_* satisfying $T^{\text{ss}}(x_*) = T_c$ is inside the ordered regions, i.e., $x_* < X^{\text{ss}} - \xi^{\text{ss}}$, where ξ^{ss} is the interface thickness as indicated in Fig. 2(c). That is, superheated ordered states appear in a region where $T(x) > T_c$ and $m(x) > 0.5$. This means that the local equilibrium thermodynamics is violated due to the heat flux. In other words, the temperature at the interface $\theta \equiv T^{\text{ss}}(X^{\text{ss}})$ deviates from T_c .

To study the violation more quantitatively, in Fig. 4(a), we plot θ as a function of E with $J = -0.00002$ fixed. It is notable that the interface temperature θ deviates from the equilibrium transition temperature T_c for $E \in [E_1, E_2]$. The deviation becomes a maximum around the midpoint of $[E_1, E_2]$, where the interface position is around $L_x/2$. The superheated regions disappear for $E \rightarrow E_1$ or E_2 , where the system becomes occupied by an ordered or disordered state. To examine the parameter dependence, we compare the numerical results for the violation with the formula

$$\theta^{\text{Th}} = T_c + |J| \left(\frac{1}{\kappa^{\text{o}}} - \frac{1}{\kappa^{\text{d}}} \right) \frac{X(L_x - X)}{2L_x}, \quad (9)$$

which was proposed in the global thermodynamics framework [23], where κ^{o} and κ^{d} are the heat conductivities of ordered and disordered states, respectively, and X is the position of the interface. The formula (9) can be derived for the order-disorder transition [33]. In Fig. 4(a), we simultaneously plot θ and θ^{Th} , where the latter is obtained by substituting X^{mc} into X in (9). We find that θ directly measured in numerical simulations is in qualitative agreement with θ^{Th} determined by the formula (9). The discrepancy is due to a nonlinear effect of $|J|$ as shown in the inset of Fig. 4(a).

Finally, we investigate the finite size effects. We find that the violation of local equilibrium becomes weaker for smaller L_x or larger Δx . Instead of the original system consisting of 3073×512 grid points with $\Delta x = 1/8$, we study smaller systems consisting of 1537×256 grid points and 768×128 grid points, keeping the value of Δx fixed. The results are shown in Fig. 4(b). The interface temperature deviates from the theoretical curve and eventually reaches the equilibrium transition temperature. This indicates that long-wavelength fluctuations play an important role in the violation of local equilibrium properties.

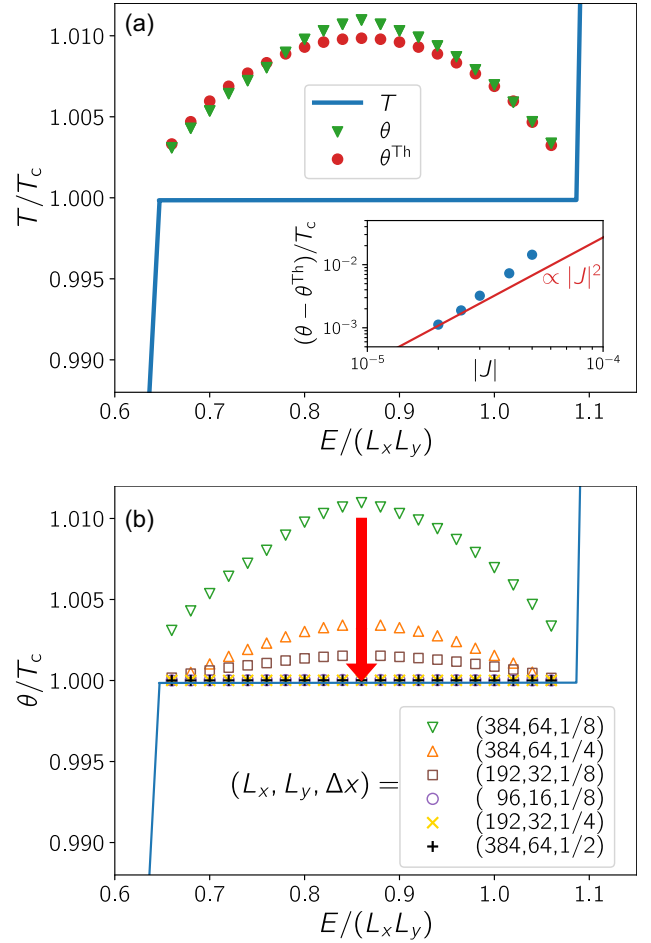


FIG. 4. (a) Interface temperature as a function of the energy density $E/(L_x L_y)$ for $J = -0.00002$. The green points are numerical values $\theta \equiv T^{\text{ss}}(X^{\text{ss}})$ and the red points are the theoretical predictions θ^{Th} in (9), substituting the numerically determined X^{mc} for X . The blue line corresponds to the green line in Fig. 1(b). Inset: $\theta - \theta^{\text{Th}}$ are plotted for $|J|$ at $E/(L_x L_y) = 0.86$ that gives the peak value. $\theta - \theta^{\text{Th}}$ approaches the red line representing a relationship proportional to $|J|^2$. (b) Finite size effects of the numerically determined θ for $(L_x, L_y, \Delta x)$ depicted in the box with $J = -0.00002$. Several symbols are overlapped [33].

A similar trend is observed for rough systems with larger values of Δx for the same system size. The interface temperature becomes closer to T_c as Δx increases from $1/8$ to $1/2$. This implies that regularity of the short wavelength fluctuations of the continuum fields is also necessary for the violation, which is in stark contrast to the properties in equilibrium. We do not observe the violation of local equilibrium with smaller or rougher systems. These observations suggest that hydrodynamic fluctuations on some scales play an important role for the violation of the local equilibrium at the interface.

Concluding remarks.—In this Letter, we studied non-equilibrium phase coexistence under a heat flux using the two-dimensional Hamiltonian Potts model with $q = 11$.

We found that a super-heated ordered region stably appears in the heat-conducting state. This indicates that the metastable states are controlled by the heat flux. Our numerical results also quantitatively support the validity of global thermodynamics. We hope this work triggers further experimental and theoretical research, such as engineering thermodynamic metastable states. In conclusion, we present three remarks.

Although we expect that the violation of local equilibrium thermodynamics is generically observed in other models and experiments, we need to carefully choose the system conditions. For example, liquid-gas coexistence under heat conduction was studied using molecular dynamics simulations, and no violation was observed [39,40]. We conjecture that the system sizes were too small to allow long-wavelength fluctuations that would lead to the violation of local equilibrium thermodynamics. So far, we do not estimate a crossover system size beyond which the violation is observed. As a reference, we remark that a long-range correlation of hydrodynamic fluctuations in a sheared system is observed only when the particle number exceeds a crossover value of 10^7 [41]. We thus expect that the same order of particles are necessary for the violation. To confirm this conjecture is left for future study.

From a theoretical viewpoint, an important first study is to derive the stationary distribution of the heat conduction system. In the linear response regime, the Zubarev-McLennan representation is a generalization of the micro-canonical distribution, where the correction term is expressed in terms of entropy production [3,4], as discussed for phase coexistence in heat conduction [42]. Developing a method for estimating this correction term explicitly for microscopic or mesoscopic models is an important goal.

The most critical challenge is the observation of the violation of local equilibrium thermodynamics in laboratory experiments. As an example, for the liquid-gas coexistence for water at 1 atm pressure, the interface temperature was calculated to be 96 °C when the temperatures of the heat baths attached to the left and right sides were 95 and 105 °C [23]. This means that super-cooled gas stably appears near the interface due to the heat flux. We believe that observation of this phenomenon is of fundamental importance.

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