We study the modification of the second law of thermodynamics for a quantum system interacting with a reservoir regarding quantum coherence. The whole system is isolated so that neither energy nor information is lost. It is discovered that the coherence of the reservoir can serves as a useful resource allowing the system extract more energy from the reservoir; among the coherence measures, only is the relative entropy of coherence feasible to quantitatively characterize energy exchange. We demonstrate that a thermodynamic cycle between two coherent reservoirs can output more work than its classical counterpart. The efficiency of such cycle surpasses the Carnot efficiency, which is the upper bound of heat engine efficiency in classical regime.

Introduction.—Conventional thermodynamics deals with macro-matters in equilibrium states. The thermodynamic limit, introduced as a mathematical approach, restricts this theoretical framework to macroscopic situation [1]. When dealing with the thermodynamic properties and energy conversion for microscopic systems off thermodynamic limit, it is necessary to take into account quantum coherence effects [2–5]. This goes beyond the valid regime of the conventional thermodynamics. Over the past few decades, quantum thermodynamics was developed [6–9] to build a bridge between the macro-world dominated by quantum mechanics and the micro-world by classical thermodynamics.

Recently, quantum resource theories were proposed to quantitatively describe the usefulness of the quantum coherence effects [4, 5, 10–13]. For thermodynamics, an immediate question raises: how quantum coherence effects enhance the energy conversion for non-equilibrium processes in quantitative level [14–17]. Recent studies have shown that quantum coherence resource is a promising candidate for such enhancement role [18–26]. However, how to characterize the role of quantum coherence in thermodynamics with a general approach remains an open problem, although extensive attempts have been made for some specific systems [27–33]. This challenge is not only essential to the foundations of quantum thermodynamics, but also crucial for designing various energy conservation machines operating in the micro-world with high-performance.

In this Letter, we revisit the second law of thermodynamics for a quantum system interacting with a reservoir. Both of the system and reservoir are of finite size so that the quantum coherence can persist in a duration in comparison with the relaxation time and the cycle time. We derive the heat exchange and entropy flow inside such an interacting system which is not thermally equilibrium. The obtained result shows that the second law of thermodynamics is quantitatively modified with the quantum coherence resource, which is characterized by the relative entropy of coherence \( C \) [34]. The modified second law of thermodynamics in quantum regime is further applied to a non-equilibrium thermodynamic process with a finite-sized reservoir. It is discovered that the quantum coherence of the reservoir is converted into heat, which can be extracted as exceeding work. When a quantum Carnot heat engine operates between two coherent reservoirs, the corresponding efficiency will surpass its classical counterpart, namely, the Carnot efficiency.

Thermodynamics in an isolated binary system.—The isolated system \( SE \) of interest is composed of the system \( S \) and the finite-sized reservoir \( E \). The conventional thermodynamics regards \( E \) as an infinite system. The total Hamiltonian of such coupling system is driven by a time-dependent parameter \( \lambda(t) \) as

\[
H(t) = H_S[\lambda(t)] + H_E + V. \tag{1}
\]

Here \( H_S[\lambda(t)] \) and \( H_E \) are the Hamiltonian of \( S \) and \( E \), respectively; \( V \) describes the interaction between \( S \) and \( E \), which will result in entropy exchange and heat exchange between \( S \) and \( E \) during the evolution of SE. Work can be applied to the system \( S \) by tuning \( \lambda(t) \). Hereafter, for simplicity, we consider the reservoir is non-degenerate in energy, and the “coherence” of the reservoir is defined with respect to energy eigen-states of \( E \). Assuming the whole system \( SE \) is initially prepared in the product state \( \rho_{SE}(0) = \rho_S(0) \otimes \rho_E(0) \) with \( \rho_\alpha (\alpha = SE, S, E) \) the density matrix of \( \alpha \). Such a product state \( \rho_{SE}(0) \) implies there is no initial correlation between \( S \) and \( E \). The state of \( SE \) subject to unitary evolution at time \( t \) is \( \rho_{SE}(t) = U(t)\rho_{SE}(0)U(t)^\dagger \), where

\[
U(t) = T \exp \left[ -i/\hbar \int_0^t H(t)dt' \right]
\]

is the evolution operator and \( T \) is the time-order operation. The time evolution of the sub-system \( E \) is described by the reduced density matrix \( \rho_{SE}(t) = T_{E(S)}[\rho_{SE}(t)] \). It is stressed here that in most studies, the reservoir is considered as a macro-system with infinite size, so that its thermal equilibrium state will not change over certain time, namely, \( \rho_E(0) \rightarrow \rho_E(t) = \rho_E(0) = \exp(-\beta_EH_E)/T_E[\exp(-\beta_EH_E)] \). Here \( \beta_E = 1/(k_BT_E) \) is the inverse temperature of \( E \), \( T_E \) the temperature of \( E \), and \( k_B \) the Boltzmann constant.

Works with quantum resource of coherence

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where, for any thermodynamic variable $S$, the change in internal energy of $E$ with $S$, $E$ and $S$ in dynamics in this scenario. Generally, as the entire system changes, and the effective temperature remains unchanged during the evolution of SE, namely, $\dot{E}_{SE}(t) = \lambda \text{Tr} \left[ \rho_S(t) \frac{\partial H_S}{\partial \lambda} \right] = \dot{W}_S(t)$, is achieved through the outputting work or applied work, that is

$$\dot{E}_{SE}(t) \approx \frac{d}{dt} \langle H_S \rangle + \frac{d}{dt} \langle H_E \rangle$$

Here, the change in the internal energy of $E$, i.e., $d \langle H_E \rangle / dt$, is defined as the heat exchange between $S$ and $E$, i.e., $\text{Tr} \left[ \dot{\rho}_E(t) H_E \right] = \dot{Q}_E(t) = -\dot{Q}_S(t)$. It follows from Eqs. (2) and (3) that

$$\dot{E}_S(t) = \frac{d}{dt} \langle H_S \rangle = \dot{W}_S(t) + \dot{Q}_S(t).$$

With the obtained first law of thermodynamics in Eq. (4), we go further to study the second law of thermodynamics in this scenario. Generally, as the entire system SE evolves, $S$ and $E$ will become entangled, resulting in correlation between $S$ and $E$. Such correlation can be measured by the von Neumann entropy $I = I(S : E)$ as $[36, 37, 39–41]$. Notice that $S_\alpha(t) = -\text{Tr} \left[ \rho_\alpha(t) \ln \rho_\alpha(t) \right]$ ($\alpha = \text{SE, S, E}$) being the time derivative of the above equation, we can express entropy flow of the system as

$$\dot{S}_S(t) = \dot{I}(t) - \dot{S}_E(t),$$

where the relation $\dot{S}_SE(t) = 0$ is used since the evolution of SE is unitary. The entropy flow of $E$, i.e., $\dot{S}_E(t)$, can be re-written with the heat flow $\dot{Q}_S$ as (see Supplementary Materials (SM) for detailed derivation $[38]$)

$$\dot{S}_E = -\dot{\rho}_E(t) || \rho_E^{\text{eq}}(0) || - \beta_E \dot{Q}_S.$$

Here, $S[\rho_E(t)||\rho_E^{\text{eq}}(0)] \equiv \text{tr} \left[ \rho_E(t) \ln \rho_E(t) - \rho_E(t) \ln \rho_E^{\text{eq}}(0) \right]$ is the quantum relative entropy between the state $\rho_E(t)$ and $\rho_E^{\text{eq}}(0); \rho_E^{\text{eq}}(0) \equiv e^{-\beta_E H_E}/(e^{-\beta_E H_E})$ is the initial effective equilibrium density matrix of the reservoir with $\beta_E = \beta_E(0)$ the corresponding effective initial inverse temperature of $E$. It follows from Eqs. (6) and (7) that

$$\Delta S_S = \beta_E \Delta Q_S + \Delta I + \Delta S[\rho_E(t)||\rho_E^{\text{eq}}(0)],$$

where $\Delta(\bullet) \equiv \int_0^t (\bullet) dt = (\bullet) - (\bullet(0))$. The second and third terms in the right hand of the above equation relate to the irreversible entropy generation $\Delta S_S(\alpha) = \Delta S_S - \beta_E \Delta Q_S$, $[37, 39–41]$. Notice that the mutual information is non-negative, namely, $I(t) \geq 0 \ [36]$, and there is no initial correlation between the system and reservoir, i.e., $I(0) = 0$, then $\Delta I \geq 0$. Therefore, the second law of thermodynamics is obtained as

$$\Delta S_S \geq \frac{\Delta Q_S}{T_E} + \Delta S[\rho_E(t)||\rho_E^{\text{eq}}(0)],$$

where $T_E = \beta_E^{-1}$ is the initial effective temperature of the reservoir.

As we mentioned before, in the usual treatment, the reservoir is considered as a classical infinite thermal reservoir, thus the state of $E$ will not be affected by $S$ and will remain unchanged during the evolution of SE, namely, $\rho_E(t) = \rho_E^{\text{eq}}(0)$. In this sense, $\Delta S[\rho_E(t)||\rho_E^{\text{eq}}(0)] = 0$, and then Eq. (9) reduces to the familiar form $\Delta S_S \geq \beta_E \Delta Q_S$, which is the second law of thermodynamics in classical regime. In following discussion, we will analyze the second term of Eq. (9), i.e., $\Delta S[\rho_E(t)||\rho_E^{\text{eq}}(0)]$, in detail. The role of the reservoir’s finite size and coherence in the modified second law of thermodynamics will be distinguished and clearly presented.

**Finite size reservoir with coherence.** To further explore the quantum properties of $\Delta S[\rho_E(t)||\rho_E^{\text{eq}}(0)]$, we first divide $\rho_E(t)$ into the diagonal part $\rho_E^d(t)$ and non-diagonal part $\rho_E^{nd}(t)$, namely, $\rho_E(t) = \rho_E^d(t) + \rho_E^{nd}(t)$, where $\rho_E^{nd}(t)$ is non-vanishing for coherent reservoirs. Then, $\Delta S[\rho_E(t)||\rho_E^{\text{eq}}(0)]$ can be divided into three parts as $[38]$

$$S[\rho_E(t)||\rho_E^{\text{eq}}(0)] = \mathcal{C}_E(t) + S[\rho_E^d(t)||\rho_E^d(0)] + \frac{\Delta Q_S^2}{2C_E T_E^2},$$

Here, $\mathcal{C}_E(t) \equiv \text{tr} \left[ \rho_E(t) \ln \rho_E(t) - \rho_E^d(t) \ln \rho_E^d(t) \right]$ is the relative entropy of coherence, which is a measure of quantum coherence in the resource theory of coherence $[5, 34, 42–44]$. $\rho_E^{eq}(t) \equiv e^{-\beta_E(t) H_E}/\text{tr} \left[ e^{-\beta_E(t) H_E} \right]$ is the effective equilibrium density matrix of $E$ at time $t$, the effective inverse temperature $\beta_E(t)$ is determined by the internal energy $U_E(t)$, $\Delta Q_S = \int_0^t \dot{Q}_S dt$ is the heat exchange between $S$ and $E$, and $\mathcal{C}_E$ is the heat capacity of $E$. Physically, $\mathcal{C}_E(t)$ represents the contribution of coherence, and $S[\rho_E^d(t)||\rho_E^d(0)]$ describes the degree to which the reservoir deviates from equilibrium. The term $\Delta Q_S^2/ \left( 2C_E T_E^2 \right)$ is caused by the back-action of $S$ to $E$ under the constraint of energy conservation $[45–47]$. Specifically, with the heat exchange between $S$ and $E$, the internal energy of $E$ changes, and the effective temperature
of E thus changes accordingly. This effect will reduce the efficiency at maximum work of a heat engine operating between finite-sized heat reservoirs [48, 49]. From Eq. (8) and Eq. (10), we obtain

$$\Delta S_s = \frac{\Delta Q_S}{T_E} + \frac{\Delta Q_S^2}{2C_E T_E^2} + \Delta \mathcal{E}_E + \Delta I, \quad (11)$$

where $\Delta \mathcal{E}_E = \mathcal{E}_E(t) - \mathcal{E}_E(0)$ is the change in coherence of the reservoir during the whole process. In order to compare the current result with that of the conventional thermodynamics, we further assume that the diagonal elements the reservoir’s density matrix follows the time-dependent Boltzmann distribution, namely, $\rho_E^{di}(t) = \rho_E^{ci}(t)$, and thus $S[\rho_E^{di}(t)||\rho_E^{ci}(t)] = 0$. In this case, Eq. (11) becomes

$$\Delta S_s = \frac{\Delta Q_S}{T_E} + \frac{\Delta Q_S^2}{2C_E T_E^2} + \Delta \mathcal{E}_E + \Delta I. \quad (12)$$

Since $\Delta I \geq 0$, the explicit form of the second law in quantum regime can be derived directly from the above equality as

$$\Delta S_s \geq \frac{\Delta Q_S}{T_E} + \frac{\Delta Q_S^2}{2C_E T_E^2} + \Delta \mathcal{E}_E, \quad (13)$$

where the equal sign is hold if and only if $I(t) = 0$, namely, the final correlation between S and E can be ignored in comparison with other terms. We note that a relevant result was obtained recently in Ref. [32], however, only limited to the case where the reservoirs are described by collisional models. The result presented here is applicable to the systems coupled with generic reservoirs.

With the main result of this work illustrated in Eq. (13), we summarize the second law of thermodynamics in different regime in Tab. I. In this table, for the case of finite reservoir without coherence, similar results were obtained in some recent studies with specific examples [45–47]. In the case that the reservoir is coherent, our result shows that, in addition to the change in entropy of the system, the change in coherence of the reservoir also contributes to the amount of heat exchange between the system and the reservoir. This implies that the coherence can be used as a quantum resource for energy extraction in thermodynamic process. Next, we will further show how to take advantages of this quantum resource to make a heat engine working between two coherent reservoirs output more work with higher efficiency than its classical counterpart.

**Surpassing the Carnot efficiency with coherent reservoirs.** Consider a general Carnot cycle, consisting of two isothermal and two adiabatic processes. In the high (low) temperature isothermal process, the working substance is in contact with the heat reservoir of effective temperature $T_h$ ($T_c$). Here, we do not limit the specific characteristics of the working substance. To use Eq. (12) to describe the heat change in the isothermal processes, the composite system of the heat engine and the heat reservoir need to be isolated from the outside world during the corresponding isothermal process. In the high temperature and low temperature isothermal process, it follows from Eq. (12) that the heat absorbed and heat released is

$$\Delta Q_h = T_h \Delta S_s - T_h (\Delta \mathcal{E}_h + \Delta I_h),$$

and

$$\Delta Q_c = - [T_c (-\Delta S_s) - T_c (\Delta \mathcal{E}_c + \Delta I_c)],$$

respectively. Here, we have assumed that the reservoirs are large enough in comparison with the working substance, thus the finite-size effect corresponded to the $C_E^{-1}$ term is ignored. $\Delta S_s > 0$ is the entropy change of the working substance in the high temperature isothermal process, and $\Delta \mathcal{E}_a (a = h, c)$ is the change in relative entropy of coherence of reservoir $a$. To focus on the effect of coherence, we further assume that the correlation change between the working substance and the reservoir can be ignored in comparison with the change in coherence of the reservoir, i.e., $\Delta I \ll \Delta \mathcal{E}$ [50]. In this sense, according to the definition of the efficiency $\eta \equiv (\Delta Q_h - \Delta Q_c) / \Delta Q_h$, we obtain

$$\eta = \eta_C - (1 - \eta_C) \frac{\Delta \mathcal{E}_h + \Delta \mathcal{E}_c}{\Delta S_s - \Delta \mathcal{E}_h}, \quad (14)$$

where $\eta_C = 1 - T_c / T_h$ is the Carnot efficiency, known as the upper bound of efficiency in classical thermodynamics. Basically, due to the existence of decoherence effect, the coherence of the reservoirs at the end of the isothermal process will be smaller than that before the isothermal process [19], which means $\Delta \mathcal{E}_h < 0$ and $\Delta \mathcal{E}_c < 0$ (we will illustrate this fact with a specific example below). In this case, one has

$$\eta = \eta_C + (1 - \eta_C) \frac{\Delta \mathcal{E}_h + \Delta \mathcal{E}_c}{\Delta S_s - \Delta \mathcal{E}_h} > \eta_C, \quad (15)$$

which shows the efficiency of the cycle can surpass the Carnot efficiency. Correspondingly, the work output per

| Infinite Reservoir | Finite Reservoir |
|--------------------|-----------------|
| $T_h \Delta S_s \geq \Delta Q_h + \Delta \mathcal{E}_h$ | $T_E \Delta S_s \geq \Delta Q_E + \frac{\Delta Q_E^2}{2C_E T_E}$ [45–47] |
| $T_E \Delta S_s \geq \Delta Q_E$ [1] | $T_E \Delta S_s \geq \Delta Q_E + \frac{\Delta Q_E^2}{2C_E T_E}$ [45–47] |

Table I. The second law of thermodynamics in different regime. Here, in the considered thermodynamic process, $T_E$ and $C_E$ are respectively the effective temperature and heat capacity of the reservoir. $\Delta S_s$ is the change in von Neumann entropy of the system, $\Delta Q_E$ is the heat absorbed from the reservoir, and $\Delta \mathcal{E}_E$ represents the change in coherence of the reservoir with $\mathcal{E}_E$ being the relative entropy of coherence.
cycle \( W = W_c + \sum_{\alpha=h,c} T_\alpha |\Delta \mathcal{E}_\alpha| \) is larger than its classical counterpart \( W_c = (T_h - T_c) \Delta S_h \). This means that extra work \( W_e = W - W_c = \sum_{\alpha=h,c} T_\alpha |\Delta \mathcal{E}_\alpha| \) can be extracted from the reservoirs by utilizing the quantum resource of coherence.

Before proceeding further, we give three remarks to the above result:

At first, even there is no effective temperature difference between the two reservoirs, i.e., \( \eta_c = 0 \), the efficiency of the cycle \( \eta = |\Delta \mathcal{E}_h + \Delta \mathcal{E}_c|/\Delta S_h - \Delta S_c > 0 \) can be non-zero due to the coherence of the reservoirs. This is a pure quantum effect, because in classical thermodynamics, the heat engine operating between two heat reservoirs with same temperature cannot output work. This implies that the quantum coherence alone can be used to output work. Similar result has been discovered with a specific model by Scully et al. [18]. However, their investigation is limited to the cavity-QED system, and their main finding that the efficiency is higher than the Carnot efficiency is based on the assumption that the working substance is in the equilibrium state with a coherence modified temperature [18, 19]. In the current work, we obtain the result from the modified second law of thermodynamics in quantum regime, without relying on the assumption that the working substance is in the thermal equilibrium state. Our results are universal for generic working substances and reservoirs, and the effect of the reservoirs’ coherence is directly demonstrated.

Secondly, a recent study [31] connected the coherence of a driven non-equilibrium quantum system to its irreversible entropy generation. In this work, we demonstrate that the coherence of the reservoir, which the system is in contact with, allows the system to extract more energy from the reservoir to output work. In general, combining our current work and Ref. [31] together, we can conclude that in the thermodynamics of a (interaction) non-equilibrium quantum system, both of the reservoir’s coherence and the system’s coherence are of great significance.

At last, we emphasize here that this result does not violate the second law thermodynamics in classical regime. After a cycle, although the state of the working substance returns to its initial state, the states of the heat reservoirs do not return to their initial states due to the loss of coherence. The price of the heat engine outputting more work is the sacrifice of the coherence of the heat reservoirs. If a larger thermodynamic cycle is considered, namely, the working substance and the heat reservoirs are all included, we believe that the Carnot bound would not be violated. Similar idea has been successfully used to explain the Maxwell’s paradox, in which case the Landauer’s principle [51, 52] states that erasing the memory of the demon requires extra work [53]. In our case, the preparation of reservoirs’ coherence requires additional energy in principle.

Example.- We further illustrate the above discussion with a specific example. Inspired by the studies in [18, 19], we specific our isolated system as a cavity QED system. The whole system is illustrated in the schematic graph in Fig. 1. As shown in the figure, \( N \) atoms pass through the photon field in the cavity with a rate \( r \), and the duration of this thermodynamic process is \( t_f \). We specify the atoms as the reservoir, and the photon field as the system. We stress here, different from the model considered in [18, 19], for simplicity, the atoms are assumed to be two-level systems rather than three-level systems with degeneracy in this work. The atom-photon interaction is described by the Hamiltonian \( V = \hbar \omega |e\rangle \langle g| a + h.c \), where \( |e\rangle \) (\( |g\rangle \)) is the excited (ground) state of the atom, \( a \) is the annihilation operator of the photon field, and \( g \) is the coupling constant. The energy spacing of the two-level atom is \( \omega \). For each atom interact with the photon field in a time interval \( \tau = t_f/N = r^{-1} \), the dynamics of the whole system is dominated by the Hamiltonian of the Jaynes-Cummings model at resonance [54]. Suppose each atom is initialized in

\[
\rho_a(t) = p_e |e\rangle \langle e| + p_g |g\rangle \langle g| + \mu |e\rangle \langle g| + \mu^* |g\rangle \langle e|, \quad (16)
\]

and the photon field is prepared in a thermal state, thus the evolution of the whole model can be solved analytically [38]. In this case, the change in coherence of all the \( N \) atoms during the isothermal process \( \Delta \xi_E(t_f) = \xi_E(t_f) - \xi_E(0) \) is \( \Delta \xi_E(t_f) = -\Gamma \xi_a(0)t_f \) (See [38] for details). Here, \( \xi_a(0) \) is the initial relative entropy of coherence of each atom. The coefficient \( \Gamma = g^2r^{-1}(1 + 2 \langle n \rangle) \) is an increasing function of the coupling constant \( g \) and the mean particle number \( \langle n \rangle \) of the photon field, and a decreasing function of the passing rate \( r \) of the atoms.
Then, according to Eq. (14), the efficiency of this cycle is explicitly obtained as [38]

\[ \eta = \eta_c + (1 - \eta_c) \frac{\Gamma \xi_h t_h}{\Delta S_t + \Gamma \xi_h t_h}. \tag{17} \]

Here, we have assumed that in the high-temperature isothermal process of duration \( t_h \), the relative entropy of coherence of each initial atom is \( \xi_a(0) = \xi_h \), while the atoms in the low-temperature isothermal process have vanish coherence, namely, \( \xi = 0 \). \( \Delta S_t \) is the change in entropy of the photon field per cycle. Obviously, Eq. (17) indicates that the efficiency of such photon-cavity Carnot engine can surpass the Carnot efficiency, which is defined by the effective temperature of the atoms in the corresponding Carnot efficiency. Besides, note that for a given \( t_r \), the coherence correction term in Eq. (17) decreases until it reaches zero.

**Conclusions and discussions.** As a summary, we studied the second law of thermodynamics in quantum regime, which involves a system interacting with a finite-sized reservoir with quantum coherence characterized by off-diagonal terms in the density matrix. It is shown that the coherence, qualified by relative entropy, can serve as a useful resource for improving the output efficiency of heat engines. Particularly, even there is no temperature difference between the two reservoirs, the heat engine can also output non-vanishing work resulting from the reservoir’s coherence. The above observations were further demonstrated with a photon-cavity-Carnot engine, where the reservoir consists of two-level atoms with coherence. It should be mentioned that, besides the relative entropy of coherence [34], other coherence measures had also been proposed, such as the \( l_1 \) norm of coherence [34], the robustness of coherence [55], and the coherence of formation [56]. Nevertheless, our current studies revealed that only the relative entropy of coherence is feasible to describe energy exchange in the thermodynamics with quantum coherence. The coherence-enhancement in energy conversion processes predicted above can be tested in some state-of-art experiments for quantum thermodynamics [57–59].

The general findings in the current study are expected to be applied to optimize the operation of quantum heat engine in finite-time cycles, although the optimizations for classical heat engine have been extensively investigated [8, 60, 61]. We suggest to consider the relevant issues listed as follows: (i) the influence of the reservoir coherence on the engine efficiency at maximum power [62], the power-efficiency constraint relation [40], and the efficiency fluctuation [63]; (ii) whether the coherence will change the optimal operation scheme of the heat engine [41, 64–66]; (iii) the information correlation [67, 68] result from the finite-sized reservoir with non-canonical statistics [46, 68–71]. The above mentioned investigations in future would deepen the understanding of the information-assisted thermodynamic processes of energy conservation in micro-scale.

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See Supplemental Material for the derivation of Eqs. (7) and (10), and the detailed solution of the dynamic evolution of the Jaynes-Cummings model.

Here, the information correlation $\Delta \mathcal{I}$ is related to the irreversible entropy generation [37], as which is vanish in the reversible processes in quasi-static limit.

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