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# Observation of Time-Invariant Coherence in a Nuclear Magnetic Resonance Quantum Simulator

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The ability to live in coherent superpositions is a signature trait of quantum systems and constitutes an irreplaceable resource for quantum-enhanced technologies. However, decoherence effects usually destroy quantum superpositions. It was recently predicted that, in a composite quantum system exposed to dephasing noise, quantum coherence in a transversal reference basis can stay protected for an indefinite time. This can occur for a class of quantum states independently of the measure used to quantify coherence, and it requires no control on the system during the dynamics. Here, such an invariant coherence phenomenon is observed experimentally in two different setups based on nuclear magnetic resonance at room temperature, realizing an effective quantum simulator of two- and four-qubit spin systems. Our study further reveals a novel interplay between coherence and various forms of correlations, and it highlights the natural resilience of quantum effects in complex systems.

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Successfully harnessing genuine nonclassical effects is predicted to herald a new wave of technological devices with a disruptive potential to supersede their conventional counterparts [1]. This prediction is now coming of age, and an international race is on to translate the power of quantum technologies into commercial applications to networked communication, computing, imaging, sensing, and simulation [2]. Quantum *coherence* [3], which incarnates the wavelike nature of matter and the essence of quantum parallelism [4], is the primary ingredient enabling a supraclassical performance in a wide range of such applications. Its key role in quantum algorithms, optics, metrology, condensed matter physics, and nanoscale thermodynamics is actively investigated and widely recognized [5–13]. Furthermore, coherent quantum effects have been observed in large molecules [14] and are advocated to play a functional role in even larger biological complexes [15–18]. However, coherence is an intrinsically fragile property which typically vanishes at macroscopic scales of space, time, and temperature: the disappearance of coherence, i.e., *decoherence* [19], in quantum systems exposed to environmental noise is one of the major hindrances still threatening the scalability of most quantum machines. Numerous efforts have thus been invested in recent years into devising feasible control schemes to preserve coherence in open quantum systems [20], with notable examples including dynamical decoupling [21,22], quantum feedback control [23], and error correcting codes [24].

In this Letter we demonstrate a fundamentally different mechanism. We observe experimentally that quantum coherence in a composite system, whose subsystems are all affected by decoherence, can remain *de facto* invariant for an arbitrarily long time without any external control. This phenomenon was recently predicted to occur for a particular family of initial states of quantum systems of any (however large) even number of qubits [25], and it is here demonstrated in a room temperature liquid-state nuclear magnetic resonance (NMR) quantum simulator [26–30] with two different molecules, encompassing two-qubit and four-qubit spin ensembles. After initialization into a so-called generalized Bell diagonal state [25], the multiqubit ensemble is left to evolve under naturally occurring phase damping noise. Constant coherence in a reference basis (transversal to the noise direction) is then observed within the experimentally considered time scales up to the order of a second. Coherence is measured according to a variety of recently proposed quantifiers [31], and its permanence is verified to be measure independent. We also reveal how coherence captures quantitatively a dynamical interplay between classical and general quantum correlations [32], while any entanglement may rapidly disappear [33]. For more general initial states, we prove theoretically that coherence can decay, yet that it remains above a guaranteed threshold at any time, and we observe this experimentally in the two-qubit instance. The present study advances our physical understanding of the resilience of quantum effects against decoherence.

Quantum coherence manifests when a quantum system is in a superposition of multiple states taken from a reference basis. The reference basis can be indicated by the physics of the problem under investigation (e.g., one may focus on the energy eigenbasis when addressing coherence in transport phenomena and thermodynamics) or by a task for which coherence is required (e.g., the estimation of a magnetic field in a certain direction). Here, for an $N$-qubit system, having in mind a magnetometry setting [34], we can fix the reference basis to be the "plus-minus" basis $\{|\pm\rangle^{\otimes N}\}$, where $\{|\pm\rangle\}$ represents the eigenstates of the $\sigma_1$ Pauli operator, which describes the $x$ component of the spin on each qubit [24]. Any state with a density matrix $\delta$ diagonal in the plus-minus basis will be referred to as incoherent. According to a recently formulated resource theory [3,31,35,36], the degree of quantum coherence in the state $\rho$ of a quantum system can be quantified in terms of how distinguishable $\rho$ is from the set of incoherent states,

$$C_D(\rho) = \inf_{\delta\,\text{incoherent}} D(\rho, \delta), \tag{1}$$

where the distance $D$ is assumed to be jointly convex and contractive under the quantum channels, as detailed in the Supplemental Material [37]. In general, different measures of coherence induce different orderings on the space of quantum states, as happens, e.g., for entanglement or other resources. A consequence of this is that, for states of a single qubit, it is impossible to find a nontrivial noisy dynamics under which coherence is naturally preserved when measured with respect to all possible choices of $D$ in Eq. (1). As predicted in Ref. [25], such a counterintuitive situation can occur instead for larger composite systems. Here, we observe this phenomenon experimentally.

Our NMR setup realizes an effective quantum simulator in which the $N$-qubit states $\rho$ can be prepared by manipulating the deviation matrix from the thermal equilibrium density operator of a spin ensemble [26,28], via application of radio frequency pulses and evolution under spin interactions [24,30]. The scalability of the setup relies on the availability of suitably large controllable molecules in liquid-state solutions.

We first encoded a two-qubit system in a chloroform ($\mathrm{CHCl_3}$) sample enriched with $^{13}$C, where the $^1$H and $^{13}$C spin-$\frac{1}{2}$ nuclei are associated with the first and second qubits, respectively. This experiment was performed in a Varian 500 MHz liquid-NMR spectrometer at room temperature, according to the plan illustrated in Fig. 1(a). The state preparation stage allowed us to initialize the system in any state obtained as a mixture of maximally entangled Bell states—that is, any Bell diagonal (BD) state [47]. These states take the form

$$\rho = \frac{1}{4}\left(\mathbb{I}\otimes\mathbb{I} + \sum_{j=1}^{3} c_j \sigma_j \otimes \sigma_j\right), \tag{2}$$

where $\{\sigma_j\}$ represents the Pauli matrices and $\mathbb{I}$ is the identity operator on each qubit; they are completely specified by the spin-spin correlation functions $c_j = \langle \sigma_j \otimes \sigma_j \rangle$ for $j = 1, 2, 3$, and they can be conveniently represented in the space spanned by these three parameters, as depicted in Fig. 1(b). We aimed to prepare specifically a BD state with the initial correlation functions $c_1(0) = 1$, $c_2(0) = 0.7$, and $c_3(0) = -0.7$, by first initializing the system in the pseudopure state $|00\rangle\langle 00|$ as described in Refs. [24,26], and then implementing the sequence of rf pulses shown in Fig. 1(a) with $\theta = \pi$ and $\alpha = \arccos(-0.7) \approx 134°$.

After state preparation, the system was allowed to evolve freely during a period of time $t$, with $t$ increased for each trial in increments of $2/J$ from 0 to 0.5 s (where $J \approx 215$ Hz is the scalar spin-spin coupling constant [37]), in order to obtain the complete dynamics. In the employed setup, the two main sources of decoherence can be modeled as Markovian phase damping and generalized amplitude damping channels acting on each qubit, with characteristic relaxation times $T_2$ and $T_1$, respectively [37]. For our system, the relaxation times were measured as $T_1^{\mathrm{H}} = 7.53$ s, $T_2^{\mathrm{H}} = 0.14$ s, $T_1^{\mathrm{C}} = 12.46$ s, $T_2^{\mathrm{C}} = 0.90$ s, which implies that $T_1^{\mathrm{H,C}} \gg T_2^{\mathrm{H,C}}$. Therefore, considering also the time domain of the experiment, only the phase damping noise can be seen to have a dominant effect.

The final stage consisted of performing full quantum state tomography for each interval of time $t$, following the procedure detailed in Refs. [37,48]. Instances of the reconstructed experimental states at $t = 0$ and $t = 0.25$ s are presented in Figs. 1(c)–1(f). The fidelity of the initial state with the ideal target was measured at 99.1%, testifying to the high degree of accuracy of our preparation stage. We verified that the evolved state remained in the BD form (2) during the whole dynamics with fidelities above 98.5%: we could then conveniently visualize the dynamics, focusing on the evolution of the spin-spin correlation triple $\{c_j(t)\}$, as indicated by the magenta points in Fig. 1(b). The time evolution of the triple $\{c_j(t)\}$ is detailed in Fig. 1(g).

From the acquired state tomographies during the relaxation progress, we measured the dynamics of quantum coherence in our states by adopting all of the known geometric coherence monotones proposed in the literature, as shown in Fig. 1(h). All quantifiers were found to be simultaneously constant within the experimental confidence levels, revealing a universal resilience of quantum coherence in the dynamics under investigation. Note that

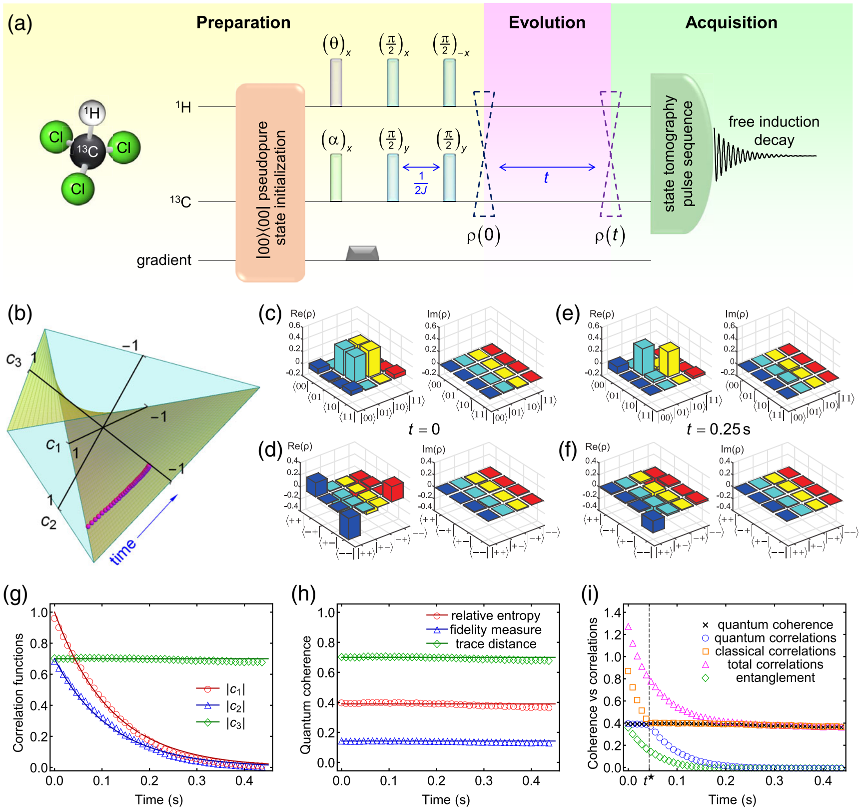

FIG. 1. (a) Pulse sequence to prepare two-qubit BD states encoded in the $^1$H and $^{13}$C nuclear spins of chloroform. The rf pulse $(\theta)_\mu$ realizes a qubit rotation by $\theta$ around the spin-$\mu$ axis, $J$ is the scalar spin-spin coupling, and time flows from left to right. (b) Dynamics of the experimental states $\rho(t)$ (the magenta points) in the space of the spin-spin correlation triple $c_j = \langle \sigma_j \otimes \sigma_j \rangle$, $j = 1, 2, 3$. All of the BD states (2) fill the light blue tetrahedron, while the subclass of states spanning the inscribed green surface is predicted to have time-invariant coherence in the plus-minus basis according to any measure of Eq. (1) [25]. (c)–(f) Full tomographies of the experimental states as prepared (c),(d) at the time $t = 0$ and (e),(f) after $t = 0.25$ s of free evolution, recorded in the computational basis (the top row) and in the plus-minus basis (the bottom row). (g) Evolution of (absolute values of) the correlation functions $|c_j|$ in the experimental states (the points), along with theoretical predictions (the solid lines) based on phase damping noise with our measured relaxation times. (h) Experimental observation of time-invariant coherence (in the plus-minus basis), measured by relative entropy (the red circles) [31], fidelity-based measure (the blue triangles) [35], and normalized trace distance (the green diamonds) [25], equal to the $l_1$ norm [31] in the BD states. The slight negative slope is due to the subdominant effect of amplitude damping. (i) Experimental dynamics of coherence and all forms of correlations [46] measured via relative entropies (the theoretical curves are omitted for graphical clarity). In (g)–(i), experimental errors due to small pulse imperfections (0.3% per pulse) result in error bars within the size of the data points.

the observed time-invariant coherence is a nontrivial feature which only occurs under particular dynamical conditions. A theoretical analysis [25,37] predicts, in fact, that, for all BD states evolving such that their spin-spin correlations obey the condition $c_2(t) = -c_1(t)c_3(t)$ [corresponding to the lime green surface in Fig. 1(b)], any valid measure of coherence as defined in Eq. (1) with respect to the plus-minus basis should remain constant at any time $t$. As evident from the placement of the data points in Fig. 1(b), our setup realized precisely the predicted dynamical conditions for time-invariant coherence, with no further control during the relaxation. Our experiment thus demonstrated a nontrivial spontaneous occurrence of long-lived quantum coherence under Markovian dynamics.

We remark that the observed effect is distinct from the physical mechanism of long-lived singlet states also studied in NMR [49], and from an instance of decoherence-free subspace [50]. In the latter case, an open system dynamics can act effectively as a unitary evolution on a subset of quantum states, automatically preserving their entropy and other informational properties. In our case, the states are instead degraded with time, but only their coherence in the considered reference basis remains unaffected. We verified this by measuring other indicators of correlations [46] in our states as a function of time. Figure 1(i) shows the dynamics of entanglement, classical, quantum, and total correlations (defined in Ref. [37]), as well as coherence. While entanglement is found to undergo a sudden death [33,51] at ≈0.21 s, a sharp transition between the decay of classical and quantum correlations is observed at the switch time $t^\star = (T_2^{\mathrm{H}} T_2^{\mathrm{C}} / T_2^{\mathrm{H}} + T_2^{\mathrm{C}}) \ln|c_1(0)/c_3(0)| \approx 0.043$ s. Such a puzzling feature was previously reported theoretically [32,52] and experimentally [47,53,54], but here we reveal the prominent role played by coherence in this dynamical picture. Namely, coherence in the plus-minus basis is found to be equal to quantum correlations before $t^{\star}$ and to classical ones after $t^{\star}$, thus remaining constant at all times. This novel interplay between coherence and correlations, observed in our natural decohering conditions, is expected to manifest for any valid choice of geometric quantifiers used to measure the involved quantities [25]; for instance, in Fig. 1(i) we picked all measures based on relative entropy.

One might wonder how general the reported phenomena are if the initial states differ from the BD states of Eq. (2). In Ref. [37] we prove that, given an arbitrary state $\rho$ with spin-spin correlation functions $\{c_j\}$, its coherence with respect to any basis is always larger than the coherence of the generalized BD state defined by the same correlation functions. This entails that, even if coherence in arbitrary states may decay under noise, it will stay above a threshold guaranteed by the coherence of corresponding BD states. To demonstrate this, we modified our preparation scheme to engineer more general two-qubit states [Fig. 2(a)]. We prepared two different pseudopure states, $\rho_1$ and $\rho_2$, with the matching initial correlation triple $c_1(0) = 0.95$, $c_2(0) = 0.62$, $c_3(0) = -0.65$, within the experimental accuracy. We then measured their coherence dynamics under natural evolution as before. For both of them, we observed a decay of coherence (albeit with different rates) towards a common time-invariant lower bound, which was determined solely by the evolution of the spin-spin correlation functions, regardless of the specifics of the states [Fig. 2(b)].

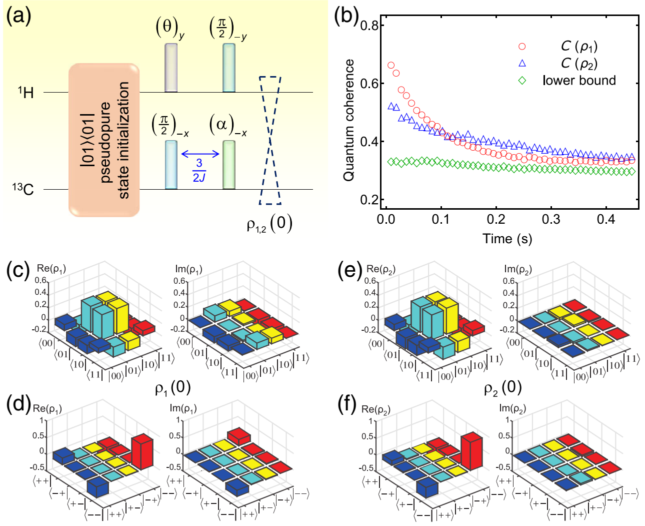

FIG. 2. (a) Modified preparation stage to engineer non-BD two-qubit states. We prepared two states, $\rho_1$ and $\rho_2$, with purity 0.92 and 0.93, respectively, setting phases $\theta \approx 0.94$ rad, $\alpha = \pi/3$ for $\rho_1$, and $\theta \approx 0.78$ rad, $\alpha = \pi/2$ for $\rho_2$. The evolution and acquisition stages were as in Fig. 1(a). Full tomographies of the produced states at $t = 0$ are presented in (c) $\rho_1$, computational basis; (d) $\rho_1$, plus-minus basis; (e) $\rho_2$, computational basis; and (f) $\rho_2$, plus-minus basis. (b) Dynamics of the relative entropy of coherence in the prepared states, along with the lower bound inferred from the evolution of their spin-spin correlation functions. The experimental errors are estimated as in Fig. 1.

Finally, we investigated experimentally the resilience of coherence in a larger system, composed of four logical qubits. To this aim, we performed a more advanced NMR demonstration in a Bruker AVIII 600 MHz spectrometer equipped with a prototype six-channel probe head, allowing full and independent control of up to five different nuclear spins [27,29]. We used the $^{13}C^{O}$-$^{15}$N-diethyl-(dimethylcarbamoyl)fluoromethyl-phosphonate compound [29], whose coupling topology is shown in Fig. 3(a). This molecule contains five NMR-active spins ($^1$H, $^{19}$F, $^{13}$C, $^{31}$P, and $^{15}$N); therefore, we chose to decouple $^{15}$N and encode our four-qubit system in the remaining spins. Each pair of spins $k, l = \{\mathrm{H, F, C, P}\}$ was coupled to each other by suitable scalar constants $J_{kl}$ [37]. We employed an "insensitive nuclei enhanced by polarization transfer" (INEPT)-like procedure [55] to prepare a generalized BD state $\rho(0) = \frac{1}{16}[\mathbb{1}^{\otimes 4} + c_1(0)\sigma_1^{\otimes 4} + c_2(0)\sigma_2^{\otimes 4} + c_3(0)\sigma_3^{\otimes 4}]$ with initial correlation functions $c_1(0) = 1$, $c_2(0) = c_3(0) = 0.7$ [37], as detailed in Fig. 3(a). After evolution in a natural phase damping environment, as before, the coherence dynamics was measured by a nontomographic detection method similar to what was done in Ref. [56], reading out the correlation triple [see Fig. 3(b)] from local spin observables on the $^1$H nucleus, whose spectrum exhibited the best signal-to-noise ratio [37]. The results in Fig. 3(c) demonstrate—albeit with a less spectacular accuracy than the two-qubit case—time-invariant coherence in the plus-minus basis in our four-qubit complex, as measured by the normalized trace distance and the relative entropy of coherence; the latter quantity also coincides with the global discord, a measure of multipartite quantum correlations [57,58], in generalized BD states.

In conclusion, we demonstrated experimentally in two different room temperature NMR setups that coherence, the quintessential signature of quantum mechanics [3], can resist decoherence under particular dynamical conditions, in principle, with no need for external control. While only certain states feature exactly time-invariant coherence in theory, more general states were shown to maintain a guaranteed amount of coherence within the experimental time scales. These phenomena, here observed for two- and four-qubit ensembles, are predicted to occur in larger systems composed by an arbitrary (even) number of qubits [25]. It is intriguing to wonder whether biological systems such as light-harvesting complexes, in which quantum coherence effects persist under exposure to dephasing environments [16–18], might have evolved towards exploiting natural mechanisms for coherence protection

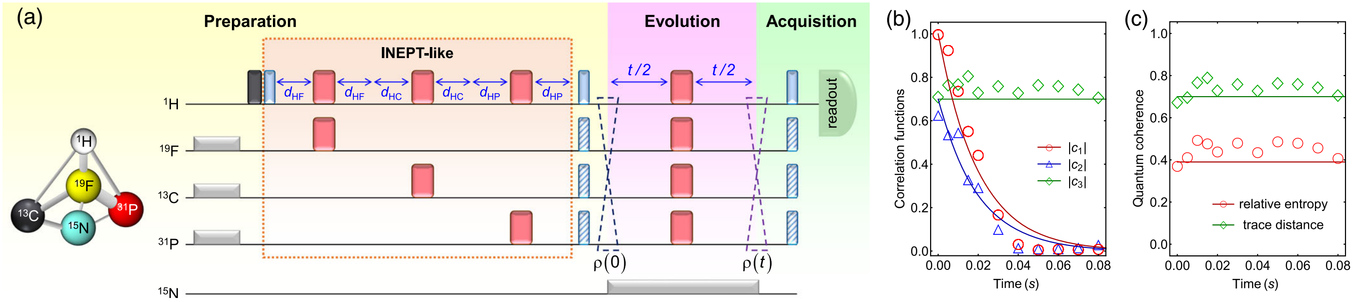

FIG. 3. (a) Pulse sequence (with time flowing from left to right) to prepare four-qubit generalized Bell states encoded in the $^1$H, $^{19}$F, $^{13}$C, and $^{31}$P nuclear spins of the $^{13}$C$^O$-$^{15}$N-diethyl-(dimethylcarbamoyl)fluoromethyl-phosphonate molecule (whose coupling topology is illustrated as an inset) by an INEPT-like procedure, where $d_{kl} = 1/(4J_{kl})$ and $J_{kl}$ is the scalar coupling between the spins $k$ and $l$. The light-gray rectangles denote continuous-wave pulses, used to decouple the $^{15}$N nucleus. The dark gray bar denotes a variable pulse, applied to set the desired correlation triple $\{c_j(0)\}$. Thicker (red) and thinner (blue) bars denote the $\pi$ and $\pi/2$ pulses, respectively; the phases of the striped $\pi/2$ pulses were cycled to construct each density matrix element. After the preparation stage, the system was left to decohere in its environment; $\pi$ pulses were applied in the middle of the evolution to avoid $J_{kl}$ oscillations. The final $\pi/2$ pulses served to produce a detectable NMR signal in the $^1$H spin channel. (b) Evolution of (absolute values of) the correlation functions $|c_j|$ in the experimental states (the points), along with theoretical predictions (the solid lines) based on phase damping noise with an effective relaxation time $T_2 \approx 0.04$ s. (c) Experimental observation of time-invariant coherence (in the plus-minus basis) in the four-qubit ensemble, measured by the relative entropy (the red circles) and the normalized trace distance (the green diamonds), along with theoretical predictions (the solid lines). In (b) and (c), experimental errors due to pulse imperfections and coupling instabilities result in error bars within the size of the data points.

similar to the ideal one reported here; this is a topic for further investigation [15].

While this Letter realizes a proof-of-principle demonstration, our findings can impact on practical applications, specifically on noisy quantum and nanoscale technologies. Particularly in quantum metrology [7], coherence in the plus-minus basis is a resource for precise estimation of frequencies or magnetic fields generated by a Hamiltonian aligned along the spin-$x$ direction. When decoherence with a preferred transversal direction (e.g., phase damping noise) affects the estimation, as in atomic magnetometry [34,59], a quantum enhancement can be achieved by optimizing the evolution time [34,60] or using error correcting techniques [61,62]. Here, we observed instances in which coherence is basically unaffected by transversal dephasing noise. This suggests that the states prepared here (or others in which similar phenomena occur, such as GHZ states; see also Ref. [63]) could be used as metrological probes with a sensitivity immune to decoherence. Furthermore, it was recently shown that the quantum advantage in discriminating phase shifts generated by local spin-$x$ Hamiltonians is given exactly by the "robustness of coherence" [13] in the plus-minus basis, a measure equal to the trace distance of coherence for BD states: this implies that the performance of such an operational task can, in principle, run unperturbed, if the probes are initialized as in our demonstration, in the presence of a natural dephasing environment. We will explore these applications experimentally in future works.

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