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Entanglement and phase

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Abstract Entanglement is the cornerstone of all new technological implications of quantum mechanics, which meanwhile extend into the realm of theoretical chemistry. In this contribution, the relation between entanglement and phase factors is investigated in terms of statistical operators. After the introduction of a simplified definition of the separability of a statistical operator, a new way to obtain phase information from experiments is presented, and the emergence of separability from non-separability is explained by the loss of a special phase operator containing the quantum part of the system's information.

Keywords Entanglement · Separability · Phase · Statistical operator

1 Introduction

The physical principle underlying the notion of entanglement has been detected by Schrödinger [1] in his first analysis of the Einstein–Podolsky–Rosen (EPR) problem [2]. Meanwhile, the most fascinating technological implications of quantum mechanics (QM) are based on the said notion. Entanglement is the essential ingredient for both quantum cryptography, quantum computing, and quantum teleportation [3]. While quantum cryptography makes use of photons to transmit messages tap-proof, most feasibility studies of quantum computers rely on quantum dots (clusters) or molecules and therefore already enter the realm of theoretical chemistry, and though the first experiments regarding quantum teleportation have been performed using photons [4–8], teleportation with the

massive particles chemists deal with is, of course, of much higher interest.¹ Therefore, it is evident that the new quantum techniques enter the sphere of interest of chemists, and in consequence theoretical chemistry has to concern itself with entanglement as well. The problem, however, is: What in fact is entanglement? We will address this question in connection with statistical operators which allow for the most general formulation of QM.

Entanglement shows up in cases where a former unit dissociates into simpler sub-systems. Corresponding processes are known quite well in chemistry. The real-space partitioning of a molecule into sub-units is still a challenging problem in theoretical chemistry, because during this process a certain entanglement of the sub-units emerges, and it is very difficult to get rid of it without destroying elementary correlations between the sub-units. So, apart from its evident importance for the foundations of physics, entanglement plays a role in chemistry too. Since the work of Mead and Berry, it is known that such dissections of quantum systems give rise to a non-trivial phase factor (for an introduction to this field, see [9]), i. e., even in chemistry the connection between entanglement and phase is of relevance. The analysis of the said interplay will be the topic of this article.

2 Preliminaries and definitions

An operator ρ on a Hilbert space \mathcal{H} is a *statistical operator* if it is self-adjoint, possesses a non-negative spectrum, and satisfies $\text{Tr } \rho = 1$. The statistical operator defines what we call the *state* of an ensemble. Now, assume that a source produces pairs of entities (A_i, B_i) which dissociate after generation. The A_i are sent to an observer named Alice while the B_i are sent to her colleague Bob. Each of them measures a rotationally variant property type as, e. g., spin or polarization on A_i and B_i , respectively. Let the measurement apparatus

¹Up to now, however, it is an open question whether entanglement can be realized with molecules at all. A corresponding EXPERIMENTUM CRUCIS has been proposed by the author [10].

Dedicated to my doctoral adviser, Prof. Dr. Karl Jug, Hannover, in honor of his 65th birthday

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A (B) be represented by the self-adjoint operator \hat{A} (\hat{B}). We assume that the eigenvalues of both operators are ± 1 , and that \mathcal{H} is the 2×2 -dimensional Hilbert space $\mathcal{H}_A \otimes \mathcal{H}_B$. We further assume that the orientation of A with respect to the laboratory coordinate system is given by the vector \vec{a} , and that

$$\hat{A} = \sigma_3 = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}. \quad (1)$$

Now, let apparatus B differ from A insofar as the vector \vec{b} determining its actual internal status can be obtained from \vec{a} by a rotation around an axis perpendicular to it. Then, \hat{B} emerges from \hat{A} by a rotation around the angle χ between \vec{a} and \vec{b} .

$$\Rightarrow \hat{B} = \begin{pmatrix} \cos \chi & \sin \chi \\ \sin \chi & -\cos \chi \end{pmatrix}. \quad (2)$$

In complete analogy, we define two further operators, \hat{A}' and \hat{B}' , where \hat{A}' represents apparatus A rotated with respect to its first position (determined by \vec{a}) by an angle φ . \hat{B}' stands for B rotated with respect to \vec{a} by an angle ψ . Note that \hat{A} and \hat{A}' as well as \hat{B} and \hat{B}' are in general non-commuting. The determinants of the commutators attain their maximum if $\varphi = \frac{\pi}{2}$ and $\psi = \chi + \frac{\pi}{2}$, respectively.

With these four apparatus settings, we can perform four experiments, i. e., four large series of single runs. Each single run in one series yields an outcome $O_i = A_i \times B_i$. The result of each experiment is the average over all single runs which, in terms of QM, is given by

$$O(\vec{a}, \vec{b}) = \text{Tr} \left((\hat{A} \otimes \hat{B}) \rho \right). \quad (3)$$

Finally, we calculate the correlation function

$$\Delta \stackrel{\text{def}}{=} |O(\vec{a}, \vec{b}) - O(\vec{a}, \vec{b}')| + |O(\vec{a}', \vec{b}) + O(\vec{a}', \vec{b}')|. \quad (4)$$

The actual value of Δ is contextual, i. e., it depends not only on the statistical operator in question but also on the choice of the apparatus axes.

Let $\{|\alpha_i\rangle|\beta_i\rangle\}$ be an orthonormal basis of \mathcal{H} . Then, in the most general case, ρ is given by

$$\rho = \sum_{i,j,k,l} c_{ij,kl} \hat{A}_{ij} \otimes \hat{B}_{kl}, \quad (5)$$

where $\hat{A}_{ij} = |\alpha_i\rangle\langle\alpha_j|$ and \hat{B}_{kl} defined analogously. In general, the non-diagonal coefficients are complex numbers $r \exp(i\phi)$ where the $\exp(i\phi)$ are called the phase factors of the statistical operator.

The physically most important potential property of a statistical operator acting on a *product* Hilbert space as $\mathcal{H}_A \otimes \mathcal{H}_B$ is its separability. I call a statistical operator *separable* if it can be decomposed according to

$$\rho = \rho_A \otimes \rho_B, \quad (6)$$

with

$$\rho_A = \sum_{i,j} a_{ij} \hat{A}_{ij} \quad (7)$$

on \mathcal{H}_A and ρ_B defined analogously on \mathcal{H}_B . It is important to note that this definition differs from the usual one where an operator is called separable if it can be decomposed into a *convex sum* of direct products as on the right side of Eq. (6). However, the definition used in this article offers a couple of advantages with respect to the usual one, which are discussed in detail in [11].² Moreover, it must be emphasized that the simplified approach to separability is corroborated by Schrödinger's own statements: Suspending entanglement means that each of the sub-systems now is furnished with an independent statistical operator of its own (see [1], p. 559). So, in Schrödinger's view disentanglement and separability (in the sense of Eq. 6) are equivalent descriptions of the same fact. This is reinforced by another quote: "Let x and y stand for all the coordinates of the first and second systems, respectively . . . What constitutes the entanglement is that Ψ is not a product of a function of x and a function of y " (see [1], p. 556). So the usual definition of separability is nothing but the effect of a misreading of Schrödinger's Eq. 1!

Finally, it is instructive to have a look at the result of the diagonalization of ρ from Eq. (5). In the 2×2 case, we obtain

$$\rho^{\text{diag}} = \tilde{c}_{1111} \hat{A}_{11} \otimes \hat{B}_{11} + \tilde{c}_{1122} \hat{A}_{11} \otimes \hat{B}_{22} + \tilde{c}_{2211} \hat{A}_{22} \otimes \hat{B}_{11} + \tilde{c}_{2222} \hat{A}_{22} \otimes \hat{B}_{22}, \quad (8)$$

which can be brought into a formal analogy with the usual separability definition

$$\rho_s^{\text{usual}} = \sum_i p_i \rho_{Ai} \otimes \rho_{Bi}. \quad (9)$$

It is, however, easy to see that ρ^{diag} is non-separable unless $\tilde{c}_{1111} = \tilde{c}_{2211}$ and $\tilde{c}_{1122} = \tilde{c}_{2222}$ which, in general, is not the case.

A measure of the non-separability of a statistical operator ρ_1 with respect to a second one can be defined as the negative difference of the two von Neumann entropies S_1 and S_2 , where $S_i = -\text{Tr}(\rho_i \ln \rho_i)$ [11]. Note that a related approach has been proposed for the partitioning of a molecule's electron density into atom contributions [16].

To sum up: An ensemble consisting of two-sub ensembles is said to be entangled if its statistical operator is non-separable in the sense of a violation of Eq. 6. In consequence, an ensemble consisting of two-sub ensembles is said to be *disentangled* if its statistical operator is given in product form.

Now the importance of Δ becomes clear, because its upper bound differs significantly depending on the statistical

²Furthermore, it is worth noting that there is some ambiguity in these definitions anyway. Abouraddy et al. say that $|\Psi\rangle \in \mathcal{H}_A \otimes \mathcal{H}_B$ is *factorizable* if and only if $|\Psi\rangle = |\Psi_A\rangle \otimes |\Psi_B\rangle$ and *entangled* if not [12]. Tsallis et al., [13] however, divide the set of statistical operators as follows: ρ is *uncorrelated* if $\rho = \rho_A \otimes \rho_B$. It is *separable* if $\rho = \sum_i p_i \rho_{A,i} \otimes \rho_{B,i}$, and it is *entangled* (which is considered equivalent to *non-separable*) if not. But Lomonaco jr. states explicitly [14] that a *pure* ensemble is separable if it satisfies a condition equivalent to the one of Abouraddy et al. while in the case of a *mixed* ensemble "one possible definition" is the one used by Tsallis et al. Obviously, there is no definition which is accepted by all colleagues in common. This situation really demands a simplified approach as presented in [11, 15]

operator in question [17],³ and Δ can be determined experimentally. So it can be decided by experiment whether a given ensemble is entangled or not.

With respect to the (non-)separability of a statistical operator, its phase seems to be of completely inferior importance. Not even the question whether ρ defines a pure or a mixed state depends on ϕ . However, we will see in the following section that the correlation function *strongly* depends on the phase and in consequence can unveil some information about it.

The determination *and* control of phase, the so-called shaping of a wavefunction, has been realized experimentally by Weinacht and coworkers [18,19]. The consequences for theoretical chemistry have been analyzed, on the basis of Wigner and Husimi functions, by Schwarz [20]. Up to now, however, there is no formulation of the phase problem in terms of statistical operators, and it will be shown in the following that the exploitation of the correlation function offers an elegant tool to obtain phase information.

3 Dependence of the correlation function on the phase of ρ

3.1 Maximally non-commuting operators

Using the angle settings $\varphi = \pi/2$ and $\psi = \chi + \pi/2$ which maximize both $\det([\hat{A}, \hat{A}'])$ and $\det([\hat{B}, \hat{B}'])$, and choosing $\chi = \pi/4$ we obtain the operators \hat{A}' , \hat{B} , and \hat{B}' in matrix form as

$$\hat{A}' = \sigma_1 = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \quad (10)$$

$$\hat{B} = \frac{\sqrt{2}}{2} (\sigma_1 + \sigma_3) = \frac{\sqrt{2}}{2} \begin{pmatrix} 1 & 1 \\ 1 & -1 \end{pmatrix}, \quad (11)$$

and

$$\hat{B}' = \frac{\sqrt{2}}{2} (\sigma_1 - \sigma_3) = \frac{\sqrt{2}}{2} \begin{pmatrix} -1 & 1 \\ 1 & 1 \end{pmatrix}. \quad (12)$$

Then, the operator ρ defined by (5) yields

$$\Delta = \sqrt{2} |1 - 2(c_{1122} + c_{2211})| + 2\sqrt{2} |r_1 \cos \phi_1 + r_2 \cos \phi_2|, \quad (13)$$

where we have made use of the representation

$$r_1 \exp(i\phi_1) = c_{1221} \quad \text{and} \quad r_2 \exp(i\phi_2) = c_{1212}. \quad (14)$$

Obviously, a lot of the non-diagonal coefficients of ρ do not enter the result (13), i. e., for our present purposes we may omit them in Eq. (5) so that ρ can be written in the simplified form

$$\rho = \begin{pmatrix} c_{1111} & 0 & 0 & r_2 \exp(i\phi_2) \\ 0 & c_{1122} & r_1 \exp(i\phi_1) & 0 \\ 0 & r_1 \exp(-i\phi_1) & c_{2211} & 0 \\ r_2 \exp(-i\phi_2) & 0 & 0 & c_{2222} \end{pmatrix}. \quad (15)$$

This formula also contains as special cases the four statistical operators which can be defined using the Bell-type basis

³A separable one leads to $\max(\Delta) = \sqrt{2}$ whereas a non-separable one can surmount this limit by a factor of 2.

$$|\Psi_e^\pm\rangle \stackrel{\text{def}}{=} \frac{1}{\sqrt{2}} (|\alpha_1\rangle|\beta_1\rangle \pm |\alpha_2\rangle|\beta_2\rangle), \quad (16)$$

$$|\Psi_o^\pm\rangle \stackrel{\text{def}}{=} \frac{1}{\sqrt{2}} (|\alpha_1\rangle|\beta_2\rangle \pm |\alpha_2\rangle|\beta_1\rangle), \quad (17)$$

according to

$$\rho_{1,2} \stackrel{\text{def}}{=} |\Psi_e^\pm\rangle \langle \Psi_e^\pm| = \frac{1}{2} (\hat{A}_{11} \otimes \hat{B}_{11} \pm \hat{A}_{12} \otimes \hat{B}_{12} \pm \hat{A}_{21} \otimes \hat{B}_{21} + \hat{A}_{22} \otimes \hat{B}_{22}) \quad (18)$$

$$\rho_{3,4} \stackrel{\text{def}}{=} |\Psi_o^\pm\rangle \langle \Psi_o^\pm| = \frac{1}{2} (\hat{A}_{11} \otimes \hat{B}_{22} \pm \hat{A}_{12} \otimes \hat{B}_{21} \pm \hat{A}_{21} \otimes \hat{B}_{12} + \hat{A}_{22} \otimes \hat{B}_{11}). \quad (19)$$

ρ_4 , e. g., which represents the singlet state frequently discussed in the EPR context, is obtained from Eq. (15) if $c_{1111} = c_{2222} = r_2 = 0$. It is easy to see that each of the four operators causes Δ to be equal to $2\sqrt{2}$.

In general, the amplitudes of the non-diagonal elements of Eq. (15) are subject to certain conditions necessary to secure the non-negativity of the spectrum of ρ . The eigenvalues are given by the two following equations:

$$(c_{1122} - \lambda)(c_{2211} - \lambda) = r_1^2, \quad (20)$$

$$(c_{1111} - \lambda)(c_{2222} - \lambda) = r_2^2. \quad (21)$$

It is easy to see that all $\lambda \geq 0$ iff

$$c_{1122}c_{2211} \geq r_1^2 \quad \text{and} \quad c_{1111}c_{2222} \geq r_2^2. \quad (22)$$

In order to maximize the influence of the phase, we have to maximize the coefficients $r_{1,2}$, i. e., in (22) we have to replace the “greater or equals” sign by the simple equals sign. From (13), we then obtain

$$\Delta = \sqrt{2} |1 - 2(c_{1122} + c_{2211})| + 2\sqrt{2} |\sqrt{c_{1122}c_{2211}} \cos \phi_1 + \sqrt{c_{1111}c_{2222}} \cos \phi_2|. \quad (23)$$

Let us consider two extreme situations. We first assume that either $c_{1122} = c_{2211} = 1/2$ or $c_{1111} = c_{2222} = 1/2$, which means that operators similar to the four operators defined in Eqs. (18) and (19), respectively, are realized. Then,

$$\Delta = \sqrt{2} |1 + \cos \phi| \quad (24)$$

so that $\sqrt{2} \leq \Delta \leq 2\sqrt{2}$. Δ can attain its maximal value only if $\exp(i\phi)$, the phase contribution to ρ , is ± 1 . If, on the other hand, all diagonal coefficients are equal to $1/4$, we arrive at

$$\Delta = \frac{\sqrt{2}}{2} |\cos \phi_1 + \cos \phi_2| = \sqrt{2} \left| \cos \frac{2\phi_1 + \delta\phi}{2} \cos \frac{\delta\phi}{2} \right|, \quad (25)$$

where we have set $\phi_2 = \phi_1 + \delta\phi$. In this case, the maximum is obtained if *both* phase contributions amount to ± 1 ($\phi_1, \phi_2 \in \{0, 2\pi, 4\pi, \dots\}$). So, we may conclude that, if we could prepare an ensemble either in the state

$$\rho = \frac{1}{2} \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & 1 & \exp(i\phi) & 0 \\ 0 & \exp(-i\phi) & 1 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix} \quad (26)$$

or in

$$\rho = \frac{1}{4} \begin{pmatrix} 1 & 0 & 0 & \exp(i\phi_2) \\ 0 & 1 & \exp(i\phi_1) & 0 \\ 0 & \exp(-i\phi_1) & 1 & 0 \\ \exp(-i\phi_2) & 0 & 0 & 1 \end{pmatrix}, \quad (27)$$

then the measurement of Δ would yield the phase information immediately.

Dephasing is the process when phase coherence gets lost, i. e., if ϕ_1 becomes $\neq \phi_2$. What happens in this case? [Recall that ρ_4 , e. g., cannot be subject to dephasing because of the complete absence of non-diagonal terms.] The effect of dephasing is maximal if Δ becomes equal to 0. Equation 25 tells us that this is the case if either $\delta\phi = \pi$ or $\delta\phi = \pi - 2\phi_1$. Therefore, a Δ -measurement of the state defined by (27) also yields information about the degree of dephasing.

To sum up: Under certain circumstances, a lot of information about the phase of a statistical operator can be obtained.

3.2 Commuting operators

The other extreme is on hand if $\sin \varphi = 0$ and $\psi = \chi + \pi$. We choose $\varphi = \pi$, because otherwise \hat{A}' would be identical to \hat{A} , and set again $\chi = \pi/4$. A lengthy but straightforward calculation yields

$$\Delta = \sqrt{2} |1 - 2(c_{1122} + c_{2211}) + 2r_3 \cos \phi_3 - 2r_4 \cos \phi_4|, \quad (28)$$

where

$$r_3 \exp(i\phi_3) = c_{1122} \quad \text{and} \quad r_4 \exp(i\phi_4) = c_{2212}. \quad (29)$$

Ignoring all those non-diagonal elements which do not appear in (28), ρ can be brought into the block-diagonal form

$$\rho = \begin{pmatrix} c_{1111} & r_3 \exp(i\phi_3) & 0 & 0 \\ r_3 \exp(-i\phi_3) & c_{1122} & 0 & 0 \\ 0 & 0 & c_{2211} & r_4 \exp(i\phi_4) \\ 0 & 0 & r_4 \exp(-i\phi_4) & c_{2222} \end{pmatrix}. \quad (30)$$

In contrast to the previous case, however, the neglect of the non-diagonal elements mentioned above now leads to a significant change in the character of ρ insofar as (30) defines a *separable* operator, because it can be written as

$$\rho = \underbrace{(a_{11}\hat{A}_{11} + a_{22}\hat{A}_{22})}_{=\rho_A} \otimes \rho_B \quad (31)$$

with ρ_B analogous to (7). We therefore always obtain $\Delta \leq \sqrt{2}$.

The diagonalization of the two blocks leads to conditions on $r_{3,4}$ similar to (22), which means that we finally arrive at the same conclusions regarding the availability of phase information as in the previous section.

4 Separability and the loss of phase information

What has happened in the transition from the general, non-separable operator

$$\rho_{\text{non-sep}} = \begin{pmatrix} c_{1111} & r_3 \exp(i\phi_3) & r_5 \exp(i\phi_5) & r_2 \exp(i\phi_2) \\ r_3 \exp(-i\phi_3) & c_{1122} & r_1 \exp(i\phi_1) & r_6 \exp(i\phi_6) \\ r_5 \exp(-i\phi_5) & r_1 \exp(-i\phi_1) & c_{2211} & r_4 \exp(i\phi_4) \\ r_2 \exp(-i\phi_2) & r_6 \exp(-i\phi_6) & r_4 \exp(-i\phi_4) & c_{2222} \end{pmatrix} \quad (32)$$

to the separable operator ρ_{sep} defined by (30)? The two non-diagonal blocks have been disregarded, and this obviously must be responsible for the loss of the factor of 2 between $\max(\Delta_{\text{non-sep}}) = 2\sqrt{2}$ and $\max(\Delta_{\text{sep}}) = \sqrt{2}$. What is the deeper reason for this loss? The non-separable operator can be written as the sum of its separable counterpart and an additional operator \hat{R} :

$$\rho_{\text{non-sep}} = \rho_{\text{sep}} + \begin{pmatrix} 0 & 0 & r_5 \exp(i\phi_5) & r_2 \exp(i\phi_2) \\ 0 & 0 & r_1 \exp(i\phi_1) & r_6 \exp(i\phi_6) \\ r_5 \exp(-i\phi_5) & r_1 \exp(-i\phi_1) & 0 & 0 \\ r_2 \exp(-i\phi_2) & r_6 \exp(-i\phi_6) & 0 & 0 \end{pmatrix} \quad (33)$$

This operator, which is canceled in the transition to ρ_{sep} , is special insofar as its trace is equal to 0, the eigenvalues may be negative, and they depend on the phase sum $\Phi = \phi_1 + \phi_2 - \phi_5 - \phi_6$. So, strictly speaking, (3) does not make sense if we replace ρ by \hat{R} , but let us nevertheless calculate $\Delta(\hat{R})$ with the operator settings as in subsection 3.1. We then obtain

$$\Delta(\hat{R}) = 2\sqrt{2} |r_1 \cos \phi_1 + r_2 \cos \phi_2|. \quad (34)$$

We may use the restrictions imposed on r_1 and r_2 as given in (22),

$$\Rightarrow \Delta(\hat{R}) = 2\sqrt{2} |\sqrt{c_{1122}c_{2211}} \cos \phi_1 + \sqrt{c_{1111}c_{2222}} \cos \phi_2|, \quad (35)$$

and it is easy to see that $\Delta(\hat{R}) \leq \sqrt{2}$. This means that both ρ_{sep} and the additional operator \hat{R} contribute equally ($\sqrt{2}$) to the final result $\max(\Delta_{\text{non-sep}}) = 2\sqrt{2}$, and if \hat{R} is omitted, the maximum of the correlation function obviously drops by a factor of 2.

Let us view this situation from another point. We perform a measurement of, say, \hat{A} on Alice's sub-ensemble $\{A_i\}$. After the measurement, Bob's sub-ensemble is in the state

$$\text{Tr}_A \left((\hat{A} \otimes \hat{I}_B) (\rho_{\text{sep}} + \hat{R}) \right) = \text{Tr}_A \left((\hat{A} \otimes \hat{I}_B) \rho_{\text{sep}} \right) + \text{Tr}_A \left((\hat{A} \otimes \hat{I}_B) \hat{R} \right). \quad (36)$$

The partial trace over $(\hat{A} \otimes \hat{I}_B) \hat{R}$ is equal to 0, i. e., by the measurement the complete information contained in \hat{R} has been eliminated. This is equivalent to the situation in the double-slit experiment if an additional detector is inserted into the course of the beam in front of the double-slit arrangement in order to obtain the which-way information. As soon as the path of the particles is determined, no interference

pattern can be observed any more. So, we may identify \hat{R} with a phase operator carrying the essential quantum part of the complete information about the ensemble. In this way, the transition from non-separability to separability (and therefore from entanglement to disentanglement) may be construed as a fundamental dephasing process as has recently been proposed by Sanctuary [21].

5 Summary

In the present contribution, the following results have been achieved:

- A statistical operator acting on a product Hilbert space $\mathcal{H}_A \otimes \mathcal{H}_B$ is separable if and only if it can be decomposed into a direct product of a statistical operator acting on \mathcal{H}_A and another one acting on \mathcal{H}_B . This simplified definition is in accordance with Schrödinger's original view on disentanglement.
- The correlation function Δ , familiar in the context of EPR-type experiments, is an elegant tool to obtain phase information about statistical operators.
- In the realm of statistical operators, the transition from entanglement to disentanglement can be viewed as the loss of a special phase operator carrying the *quantum* part of the complete ensemble information.

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