Quantumness of correlations and Maxwell's demons in elementary scattering processes—Energetic consequences

C A Chatzidimitriou-Dreismann[1] and A Dreismann[2]

[1] Institute of Chemistry, Sekr. C2, Technical University of Berlin, D-10623 Berlin, Germany
[2] Department of Physics, Cavendish Laboratory, University of Cambridge, Cambridge, CB3 0HE, UK

E-mail: `dreismann@chem.tu-berlin.de`

Abstract. The interactions between physical systems generally lead to the formation of correlations. In this paper we consider the phenomena of entanglement and "quantumness of correlations", such as quantum discord, with particular emphasis on their energetic consequences for the participating systems. We describe a number of theoretical models that are commonly employed in this context, highlighting the general character of one of their most intriguing results: In contradiction to conventional expectations, erasure (decay, consumption) of quantum correlations may be a source of work, i.e. may have "negative energetic costs". We report experimental evidence of this surprising effect obtained within the framework of an elementary scattering experiment, namely ultrafast neutron Compton scattering from normal-state liquid ^{4}He. The general theory of quantumness of correlations provides a natural way of interpreting the reported results, which stand in blatant contrast to the conventional theory of scattering, where neutron-atom-environment quantum correlations and decoherence play no role. Moreover, they provide a new operational meaning of discord and related measures of quantumness.

1. Introduction: "Information is Physical"

Correlations between physical (chemical, biological etc.) systems are created by interactions and thus are ubiquitous in Nature. They play a significant role in classical and quantum physics. The emergent behaviour of many complex systems cannot simply be derived from their individual components, since their properties fundamentally depend on the delicate correlations between their subsystems; cf. [1]. Such a situation might be expressed by the old saying: "the whole is more than the sum of its parts", which expresses a holistic in contrast to reductionist view of nature.

To prevent possible confusion, it should be emphasized that the well-known quantum mechanical correlations due to the indistinguishability of identical particles (as treated in most textbooks of quantum mechanics) play no role in the following investigations. Namely we will deal with quantum correlations between *distinguishable quantum systems* only. For instance, below we will consider specific quantum correlations between a neutron (which is a fermion) and a ^{4}He atom (a boson). Additionally, classical correlations (e.g. as appearing in classical statistical mechanics and condensed matter theory) are also excluded from our considerations.

In quantum mechanics, correlations can be essential for the description of quantum systems, in ways that, in some cases, are in blatant contrast to every expectation based on classical physics. A particular kind of quantum correlations which have been shown to exhibit distinctive non-classical features are known as quantum entanglement [2, 3, 4, 5, 6, 7], and are related to the phenomenon of non-locality [8]. Nowadays entanglement is recognized to be an indispensable resource for quantum information, computing and communication, and thus plays a central role in the associated field of quantum information processing (QIP); see e.g. [9].

The correlations between two quantum systems (say, A and B) are associated with information, as for instance in the case of DNA, where the correlations between base pairs encode genetic information. In statistical physics and thermodynamics, the crucial role of correlations became obvious when Maxwell [10], in an attempt to clarify the limitations of thermodynamics, introduced his famous demon, which also plays an important role in information theory [11]. Generally, thermodynamics and information theory possess several links, as e.g. indicated by the formal similarity of the formulas of the Shannon and the von Neumann entropies. Finally Landauer [12] succeeded to "exorcize" Maxwell's demon through his information erasure theorem; see also the related work by Bennett [13, 14]. There is no doubt that the intricate information-thermodynamics relation also implies a thermodynamic cost for information acquisition. Likewise, any information acquisition process is expected to be ultimately limited by the Second Law of thermodynamics. In short, as Landauer puts it: "Information is physical". For recent comprehensive works about the physics of (classical and quantum) Maxwell's demons and their relation to correlation, information and thermodynamics, see Refs. [15, 16, 11].

In 2001, the latest chapter in the story of the relation between correlations and information theory began when the pioneering works by Ollivier and Zurek [17] and Henderson and Vedral [18] revealed that, beyond entanglement, there exists another, more general kind of correlation in quantum physics. These correlations are popularly known as "quantum discord" [19], or more generally "quantumness of correlations". Discord and related concepts (like e.g. quantum (work) deficit, measurement-induced disturbance [20] etc.) have recently entered the field of QIP and are subject to numerous investigations regarding their operational meaning; see e.g. the review articles [21, 22]. As they concern the physics of Maxwell's demon, they also have entered the foundations of (quantum) thermodynamics.

In the present paper, we consider the possibility of applying these phenomena (i.e., entanglement, discord, decoherence, etc.) and their dynamics in a concrete experimental context, namely elementary scattering processes. We will particularly point out the "energetic costs" of quantumness of correlations and their experimental measurability, which then establishes additional physical insight into the phenomenon. We then proceed by employing the presented concepts to interpret experimental results obtained from neutron scattering (qualitatively, for the time being), which contradict conventional expectations. We believe that these results illustrate a new operational meaning (or interpretation) of quantum correlations in the context of a real experiment.

In this regard it should be mentioned that the importance of the phenomena described above has thus far not been recognized in the context of scattering experiments, e.g. neutron scattering [24, 25], electron scattering [26], or inelastic x-ray scattering from condensed matter [27]. Although it is, for instance, well known that neutron scattering from H_2 molecules may reveal quantum correlations between the two protons of the H_2 (such as the different scattering properties of ortho-H_2 and para-H_2 [23]), possible quantum correlations (or even entanglement) between the neutron and the struck nucleus, which may be created by the scattering interaction, are absent in conventional theory; see section 4.

The paper is organized as follows.

(A) Section 2 is devoted to certain aspects of the aforementioned quantum correlations. First,

in sect. 2.1 we quickly consider the well established concepts of entanglement and non-locality. Second, in sect. 2.2, we present in more detail the new concept or quantumness of correlations beyond entanglement like discord. Needless to say, this section is not meant to provide a complete account of the topic under consideration.

(B) Section 3 considers some thermodynamic and energetic consequences of quantum correlations, starting with two examples about "quantum thermal engines" (sect. 3.1), and then presenting two specific theoretical examples of direct energetic consequences of decaying quantum correlations, i.e. decoherence. The unexpected common feature of both these theoretical results is that, surprisingly, decoherence and erasure of quantum correlations can lead to work production (instead to dissipate work).

(C) An experimental observation of, or related with, this phenomenon is described in section 4, which also presents a short description of a conceptually simple scattering experiment and related experimental results obtained with neutron Compton scattering from liquid ^{4}He. The findings illustrate the significance of quantum correlations and their dynamics in the framework of neutron scattering. This extends the physical insights provided in the context of QIP by offering an additional operational meaning of quantumness of correlations.

2. Quantumness of Correlations
2.1. On Non-Locality and Quantum Entanglement
Quantum entanglement (QE) has been recognized as the most emblematic feature, or even the essence, of standard (non-relativistic) quantum theory [2, 4, 5], raising widespread interest in various branches of natural science as well as quantum information processing (QIP) and the related emerging quantum technologies; cf. [9].

It was Einstein, Podolsky, Rosen [2] and Schrödinger [3] who first recognized a "spooky action at a distance" feature of the formalism of quantum mechanics which has absolutely no classical analogue or any intuitive interpretation. This novel phenomenon, known as quantum entanglement (or simply entanglement, originally called by Schrödinger "Verschränkung"), lies at the center of interest of physics and quantum technology of the 21st century. Entanglement implies the existence of global states of a composite system which cannot be written as a product of the states of individual subsystems. As a consequence, there exist quantum statistical relations between subsystems of a compound quantum system which are by far much stronger than any conceivable classical correlations between the subsystems. Thus it may be said that entanglement represents the most nonclassical manifestation of the quantum formalism. Entanglement also contradicts the so-called Einstein's locality principle. Based on this, Einstein et al. concluded that the quantum description of physical reality is not complete [2].

About 30 years later, this conclusion, and related suggestions for introducing so-called "hidden variables" in order to make quantum mechanics a "complete theory", led Bell to the discovery of the famous Bell inequalities [4] which are experimentally testable. With his work, Bell proved that quantum mechanics is an inherently nonlocal theory that is incompatible with any physical theory in which the principle of locality holds. For instance, two spatially well separated observers measuring states of two entangled systems observe strong correlations between the results of their measurements—correlations that can be so strong that they violate Bell's inequalities. This phenomenon, called quantum non-locality, has been repeatedly observed and confirmed experimentally—which also invalidates the conclusions of [2]. For insightful presentations, one may consult the textbook by Peres [6] or the recent review by Horodecki et al. [7].

In this context, it may be noted that entanglement and non-locality are often considered as two facets of the same physical phenomenon. But now they are recognized as two different physical resources, as Gisin and collaborators have demonstrated [8]. However, the relation between them is yet to be fully understood. While entanglement is necessary for quantum non-

locality, it is not always sufficient. Indeed, entangled states exist which are local, i.e. which do not violate any Bell's inequality.

Despite its fundamental physical significance, however, quantum entanglement was considered by many as a rather "philosophical issue" having no concrete impact in "real physical fields" (like e.g. solid state physics) and/or concrete technological applications (like electronic devices and telecommunication). So it has had to wait about 70 years to enter laboratories as a novel resource as real as e.g. energy. For compound quantum systems, entanglement involves nonclassical correlations between subsystems and has been recognized to have potential for novel quantum processes even of technological relevance, like e.g. quantum cryptography, quantum dense coding and quantum teleportation [7, 9].

2.2. Quantumness of Correlations Beyond Entanglement—Discord
In this subsection, we mainly follow the presentation in Ref. [21] by Modi et al.

Coherent interactions that generate negligible entanglement can still exhibit unique quantum behaviour. This observation has motivated a search beyond entanglement for a complete description of all quantum correlations [17, 18]. Classicality and quantumness of correlations belong to the realm of information theory.

Quantum correlations, seminally quantified by the quantum discord and related measures of quantumness [21], are general manifestations of non-classicality in composite systems. They can be revealed in the process of locally measuring a subsystem, even in states where entanglement or non-locality are absent. Despite a massive surge in recent studies investigating interpretation, quantification, and applications of discord and related quantifiers of quantum correlations, cf. [21], it is a fact that these newly discovered quantities remain far less understood than entanglement. Therefore, going beyond the beautiful mathematical results achieved in this fast developing field, every result about the concrete physical meaning and/or "operational meaning", of discord (and the other measures) is of highest importance.

In simple terms one can say that two systems are correlated if together they contain more information than taken separately. If we measure the lack of information by entropy, this definition of correlations is captured by the mutual information

$$I(A:B) \equiv S(A) + S(B) - S(AB), \tag{1}$$

where $S(X)$ is the Shannon entropy

$$S(X) = -\sum_x p_x \log p_x \tag{2}$$

if X is a classical variable with values x occurring with probability p_x, or $S(X)$ is the von Neumann entropy

$$S(X) = -\mathrm{Tr}(\rho_X \log \rho_X) \tag{3}$$

if ρ_X is a quantum state of system X (all logarithms are base two). For classical variables, Bayes' rule defines a conditional probability as $p_{x|y} = p_{xy}/p_y$. This implies an equivalent form for the classical mutual information

$$J_{\mathrm{cl}}(B|A) = S(B) - S(B|A), \tag{4}$$

where the conditional entropy

$$S(B|A) = \sum_a p_a S(B|a) \tag{5}$$

is the average of entropies

$$S(B|a) = -\sum_b p_{b|a} \log p_{b|a}. \tag{6}$$

The classical correlations can therefore be interpreted as information gain about one subsystem resulting from a measurement on the other.

In contradistinction to the classical case, in the quantum analog there are many different measurements that can be performed on a system, and measurements generally disturb the quantum state. A measurement on subsystem A is described by a positive-operator-valued measure (POVM) [9] with elements $E_a = M_a^\dagger M_a$, where M_a is the measurement operator and a is the classical outcome of the associated measurement. The initial state ρ_{AB} is transformed under the measurement (with unknown result) to

$$\rho_{AB} \rightarrow \rho'_{AB} = \sum_a M_a \rho_{AB} M_a^\dagger, \tag{7}$$

where the subsystem A yields outcome a with probability $p_a = \text{Tr}(E_a \rho_{AB})$ and B has the conditional state $\rho_{B|a} = \text{Tr}_A(E_a \rho_{AB})/p_a$. This allows us to define a so-called classical-quantum version of the conditional entropy,

$$S(B|\{E_a\}) \equiv \sum_a p_a S(\rho_{B|a}), \tag{8}$$

and introduce classical correlations of the state ρ_{AB} in analogy with Eq. 4, [18]:

$$J(B|\{E_a\}) \equiv S(B) - S(B|\{E_a\}). \tag{9}$$

To quantify the classical correlations of the state *independently* of a measurement, $J(B|\{E_a\})$ is maximized over all measurements (on subsystem A),

$$J(B|A) \equiv \max_{\{E_a\}} J(B|\{E_a\}). \tag{10}$$

When the measurement is carried out by a set of rank-one orthogonal projections $\{\Pi_a\}$, the state on the right hand side of Eq. 7 has the form

$$\chi_{aB} = \sum_a p_a \Pi_a \otimes \rho_{B|a}, \tag{11}$$

which involves only fully-distinguishable states for A and some indistinguishable states for B. Such states are often called classical-quantum states. It is important to note that for a classical-quantum state there exists a von Neumann measurement of A which does not perturb the state. By exchanging the roles of A and B, one defines quantum-classical states (QC) by

$$\chi_{Ab} = \sum_b p_b \rho_{A|b} \otimes \Pi_b, \tag{12}$$

The *quantum discord* of a state ρ_{AB} under a measurement $\{E_a\}$ is defined as a difference between total correlations, as given by the quantum mutual information in Eq. (1), and the classical correlations Eq. (9), [17]:

$$\begin{aligned} D(B|A) &\equiv I(A:B) - J(B|A) \\ &= \min_{\{E_a\}} \sum_a p_a S(\rho_{B|a}) + S(A) - S(AB). \end{aligned} \tag{13}$$

Note that the minimization here is equivalent to maximization in Eq. 10. This is just a difference between two classically-equivalent versions of conditional entropy

$$D(B|A) = \min_{\{E_a\}} S(B|\{E_a\}) - S(B|A), \tag{14}$$

where $S(B|A) = S(AB) - S(A)$ is the usual conditional entropy, see e.g. [9]. This equivalence holds for rank-one POVM measurements [9] which, in classical theory, correspond to questions about a value of a classical random variable.

Here some properties of quantum discord may be noted [21, 22, 23]:

(a) It is not symmetric, i.e. in general $D(B|A) \neq D(A|B)$, which may be expected because conditional entropy is not symmetric. Some theorists consider this as a weakness. However, in the context of concrete experiments this asymmetry makes sense, as the following example shows: Knowledge about system A (say, a neutron in the n-He scattering experiment presented below) is *not* equivalent to knowledge about system B (say, the struck He). Or in other terms, it is natural to expect that measurements on A may provide another information content about the composite system AB than measurements on B; cf. Eq. (13).

(b) Discord is nonnegative, $D \geq 0$, which is a direct consequence of the concavity of conditional entropy.

(c) Discord is invariant under local unitary transformations, i.e. it is the same for state ρ_{AB} and state $(U_A \otimes U_B)\rho_{AB}(U_A \otimes U_B)^\dagger$. This follows from the fact that discord is defined via entropies, and the value obtained for measurement $\{E_a\}$ on the state ρ_{AB} can also be achieved with measurement $\{U_A E_a U_A^\dagger\}$ on the transformed state. Note that discord is not contractive under general local operations, and therefore should not be regarded as a strict measure of correlations. However, $J(B|A)$ is contractive under general local operations.

(d) Quantum discord $D(B|A)$ vanishes if and only if the state is classical-quantum [28].

In the last part of this section we point at three particular specializations of the above topics which may be appropriate in context with the experimental scattering measurements presented below (see section 4). These are focusing on the connection of quantum discord (and related measures of quantumness, including entanglement) with the process of quantum measurement. At last, an additional subsection about the recently discovered thermodynamic meaning of negative conditional entropy is presented.

2.2.1. Using Measurement-Induced Disturbance to Characterize Correlations. The above mentioned quantifiers of quantumness of correlations are based on an extremalization procedure involving the set of POVM's, which is a very difficult task to do. Additionally, while in the classical description of nature measurements can be carried out without disturbance, in the quantum description, generic measurements usually disturb the system. Thus classical states are characterized in terms of non-disturbance under a quantum measurement. Formalizing these observations, Luo has proposed a correlation quantifier based on the disturbance that the measurement processes causes in a system [20], called *measurement-induced disturbance* (MID).

This quantity, is defined as the difference between the quantum mutual information of the state, ρ_{AB}, and that one of the completely dephased state, χ_{AB}

$$MID\left(\rho_{AB}\right) = I\left(\rho_{AB}\right) - I\left(\chi_{AB}\right). \tag{15}$$

The dephasing takes place in the marginal basis, leaving the marginal states unchanged. The suitability of MID and its relation to quantum discord and various other correlation measures are discussed in Refs. [22, 21].

It should be mentioned that all quantum correlation quantifiers discussed in this article vanish for the so-called classical-classical (CC) states

$$\chi_{AB} = \sum_{a,b} p_{ab} \Pi_a \otimes \Pi_b. \tag{16}$$

Since all projectors in this decomposition correspond to fully distinguishable states, the probability p_{ab} can be regarded as a classical joint probability of random variables a and b.

It was then proved that that certain separable states, e.g. quantum-quantum states

$$\rho^{AB} = \sum_i p_i \rho_i^A \otimes \rho_i^B, \tag{17}$$

(where ρ_i^A and ρ_i^B are local states pertinent to subsystems A and B) still possess correlations of a quantum nature. He also showed that the specific quantum correlations contained in these mixed quantum-quantum states correspond to a "generalization" of the concept of entanglement [20].

2.2.2. Linking Quantum Discord to Entanglement in a Measurement. The focus of this work, as mentioned earlier, lies on the experimental context of elementary scattering processes in condensed systems and molecules. In this context, the interacting two systems A and B are "disturbed" (or "measured", or "observed") by another system C, e.g. by adjacent particles of the "environment". Thus it is natural to mention here some related works in which the measurement process may have effect on the quantumness of correlations of the composite system $A + B$.

Recently Streltsov et al. [29] have introduced an alternative approach to quantum correlations via an interpretation of a quantum measurement, especially on one of the subsystems of a composite system.

In carrying out a von Neumann measurement on a system S in the quantum state ρ^S, correlations between the system and the measurement apparatus M must be created. E.g., consider a von Neumann measurement in the eigenbasis $|i\rangle$ of the mixed state

$$\rho^S = \sum_i p_i |i^S\rangle\langle i^S| \tag{18}$$

with the eigenvalues p_i. One then expects the final state of the total system to be

$$\rho_{\text{final}} = \sum_i p_i |i^M\rangle\langle i^M| \otimes |i^S\rangle\langle i^S|, \tag{19}$$

where $|i^M\rangle$ are orthogonal states of the measurement apparatus M. This ρ_{final} is a classical-classical state (see above), i.e. the correlations between M and S are purely classical.

The situation changes completely if we consider *partial* von Neumann measurements, i.e. von Neumann measurements which are restricted to a subsystem A of the system AB. (Indeed, these correspond to the usual case in real experiments; cf. section 4.) Streltsov et al. showed that in this case creation of entanglement is usually unavoidable. They also showed the close connection of this approach to the one-way information deficit [30, 22, 21] and the quantum discord. In short, they showed that the one-way information deficit is equal to the minimal distillable entanglement [15, 7] between the measurement apparatus M and the system AB which has to be created in a von Neumann measurement on subsystem A. The quantum discord is then equal to the corresponding minimal partial distillable entanglement. (The minimization is done over all unitaries U which realize a von Neumann measurement on A; see Theorem 2 of [29].) Additionally, it was shown that this approach can be generalized to multipartite von Neumann measurements [29].

2.2.3. On the Thermodynamic Meaning of Negative Conditional Entropy. In the preceding derivations of quantumness of correlations, the concept of conditional entropy (or conditional infomation) plays a considerable role. Very recently, del Rio et al. [31] showed that the standard formulation and implications of Landauers principle [12, 13] (cf. Introduction) are no longer valid

in the presence of quantum information. Their main result is that the work cost of erasure of information is determined by the entropy of the system, conditioned on the quantum information an observer has about it: the more an observer knows about the system carrying the information, the less it costs to erase the information. Obviously this result gives a direct thermodynamic significance to *conditional entropy*, which plays a dominant role in the theory of quantumness of correlations, see above.

The main result characterizes the work, $W(A|Q)$, that an observer with access to a quantum memory Q correlated with the system A, needs to perform to erase system A. In the "thermodynamic limit", where the observer erases many identical copies of A jointly, the authors have found a specific erasure process [31] whose work cost does not exceed

$$W(A|Q) = S(A|Q) \, kTln(2) \tag{20}$$

per copy of A; (k: Boltzmann constant, T: temperature). Here $S(A|Q)$ is the conditional von Neumann entropy,

$$S(A|Q) = S(AQ) - S(Q). \tag{21}$$

This work cost is shown to be optimal, under the assumption that Landauers principle holds for a classical observer.

In the quantum case, however, novel features may arise. In particular, the last equation implies that the work required for erasure may be negative for an observer with a quantum memory Q: the process may result in a net *gain of work*. For instance, the combined system AQ may be closed and in a pure state, and thus its von Neumann entropy will be zero, $S(AQ) = 0$, whereas the reduced state of the memory Q is mixed and has positive entropy $S(Q) > 0$, which finally yields $S(A|Q) < 0$. This provides a thermodynamic operational meaning for negative conditional entropies, which earlier only had information-theoretical interpretations [31, 21]. Moreover, it was noted that the obtained results suggest that quantum discord can quantify the difference between the respective work costs of erasure using quantum and classical memories.

Furthermore, it was pointed out that the above surprising result does not violate the Second Law of thermodynamics, because the proposed process is not cyclic. That is, the negative work cost is associated with the consumption of entanglement between system and quantum memory, which can only be restored by doing work (i.e., with positive work costs). The overall work costs are then expected to be positive or zero; see addendum of [31].

3. Quantum Correlations and Decoherence—Thermodynamic and Energetic Consequences

Decoherence is the ubiquitous phenomenon that destroys quantum correlations (state superpositions, quantum interference, quantum phases) thus leading to the "appearance of a classical world in quantum theory"; cf. [32]. Decoherence, similarly to many other irreversible mechanisms, is a time-oriented process, i.e. it breaks the time-inversion invariance of the Schrödinger equation. Moreover, it is known to be much faster than energy dissipation (say, due to friction or T_1 relaxation). A considerable number of models of decoherence, based on a variety of physical motivations, have been proposed and investigated in the scientific literature, cf. [32].

3.1. On the Quantum Carnot Engines of Lloyd and of Scully et al.—Thermodynamic Consequences

The quantum heat engine concept has attracted considerable interest since it provides novel insights into the fundamental physics of heat–information–energy conversion. Hence this also concerns our purposes, see section 4 below.

Lloyd [16] analyzed the effects of the quantum Maxwell demon. He found that a quantum device acquiring information in a measurement process and the associated decoherence disturb the system and act as a source of thermodynamic inefficiency. The quantum demon has been realized in the concrete context of nuclear magnetic resonance (NMR) [16, 9]. Of particular interest appears the result that this cost of quantum measurement is realized when decoherence occurs, which destroys the non-diagonal matrix elements of the system's density matrix ρ. (Studying special processes with ρ remaining always diagonal, the quantum demon reaches the well known Carnot efficiency.) In other words, the extra information introduced by quantum measurement and decoherence has been identified as the cause of *decrease* of the efficiency of the Carnot engine [16].

This interesting result pointing out the role of decoherence should be contrasted with the associated result of the well known Lindblad equation presented below in subsection 3.2.1.

Here let us proceed to a second example of a quantum heat engine. Scully et al. [33] proposed and analyzed a new kind of quantum Carnot engine powered by a special quantum heat bath. It was shown to allow extraction of work even from a single thermal reservoir. For this type of heat engine the piston is driven by radiation pressure. The working fluid (say, analogous to steam of a conventional machine) finds its counterpart in the radiation, which is generated by a beam of hot atoms (constituting the thermal reservoir).

In the "regular" case of two-level thermal atoms, the engine's efficiency cannot exceed the well-known Carnot limit. In the case of hot specific three-level atoms constituting a heat bath things look quite different, i.e. when the nearly degenerate lower levels feature a small amount of quantum coherence. The corresponding quantum phase ϕ can be varied as a control parameter to effectively increase the temperature of the radiation field. In this scenario work is obtained even when only one single heat bath is present. Consumption of quantum coherence is shown to produce work.

The deep physics behind the Second Law of thermodynamics, however, is not violated. The assumed atomic coherence itself causes energetic costs, as e.g. it must be generated by the passage of the atoms through a suitable microwave field. An explicit estimation of the necessary microwave field's energy to produce the assumed appreciable coherence between the atomic levels was shown to be greater than the extracted work in the above process. Consequently and in line with the Second Law of thermodynamics, the total entropy of the whole system is constantly increasing [33].

3.2. Pure Decoherence—Energetic Consequences

Pure decoherence (i.e. without dissipation) is usually described with the well-known master equations of Lindblad form, which ensure positivity of the systems reduced density operator.

Quite unexpectedly, it turns out that "pure" decoherence may have a perplexing, peculiar consequence. Notably, as shown below (subsections 3.2.1-2) in the frame of two independent theoretical models, pure decoherence turns out to be intrinsically connected with an increase (!) of mean energy of the system, i.e. with negative energy costs — in the absence of any direct interaction with other systems. This is certainly unexpected, since erasure of quantum phases and/or correlations is not widely acknowledged as a source of energy. The presentations below will show that this theoretical result can be of rather general character, under the restrictive condition that the characteristic time of the process is sufficiently short [50].

In the following two subsections we mainly follow the presentation of Ref. [34].

3.2.1. Lindblad Equation and Spontaneous Energy Increase.

To describe the dynamics of open quantum systems and decoherence, various generalizations of the Schrödinger equation have been proposed; see the textbooks [32]. Among these theoretical approaches, the Born-Markov master equation plays an enormously important role. Master equations of the so-called Lindblad form

refer to a particular (albeit quite general) class of Markovian master equations, which ensure positivity of the reduced density operator $\rho(t)$ describing the system, i.e. $\langle \rho(t) \rangle \geq 0$, for any pure state $|\psi\rangle$ of the system and for all t. The most general mathematical form of such equations was derived by Gorini, Kossakowski and Sudarshan [35] and Lindblad [36].

Consider the simplest Lindblad-type ansatz for the master equation for the statistical operator ρ of an open quantum system, which includes only a single Lindblad operator L; in a real system we would have a multitude of such dynamical variables. We set

$$\frac{\partial \rho}{\partial t} = -\frac{i}{\hbar} [H, \rho] - \Lambda [L, [L, \rho]] \tag{22}$$

where $\Lambda > 0$ is a positive constant and H is the Hamiltonian. The first term on the right-hand side (rhs) describes the usual unitary time evolution of the state. The double commutator term describes decoherence (and/or dephasing). It may be noted that this equation, by suitable choice of L, may describe "pure" decoherence only. Namely, it does not contain any term describing friction explicitly, as is done in Caldeira-Leggett-type equations, cf. [32]. In the following we consider only the case of pure decoherence (i.e. destruction of quantum coherence).

Considering Eq. (22) in the position representation, the double commutator term takes the form

$$-\Lambda(r - r')^2 \langle r|\rho|r' \rangle$$

which, since $\Lambda > 0$, leads to an exponential decay of the non-diagonal elements of the density matrix:

$$\langle r|\rho(t)|r' \rangle = \langle r|\rho(0)|r' \rangle \exp(-\Lambda(r - r')^2 t) \qquad \text{for} \ \ 0 \leq t \tag{23}$$

where $t = 0$ is the initial time [32]. Obviously, this result does not hold for $t \leq 0$. In other words, coherent superpositions of different states $|r\rangle$ and $|r'\rangle$ will be suppressed over time. The most well-known specialization of Eq. (22) is the case of an one-particle system that interacts with an external (thermal) environment, in which case L is taken to be the position operator, q. This corresponds to the Joos-Zeh master equation [37].

It is well known that Eq. (22) preserves the normalization $Tr\rho = 1$ [36], which is satisfactory. However, Ballentine [38] realized that the expectation value of the energy, $\langle H \rangle = Tr(H\rho)$ is in general not conserved. Namely,

$$\frac{d\langle H \rangle}{dt} = \frac{d}{dt} Tr(H\rho) = Tr\left(H \frac{\partial \rho}{\partial t} \right) = -\Lambda Tr([H, L][L, \rho]) \tag{24}$$

(Recall the operator identity $Tr(A[B, C]) = Tr([A, B]C)$.) Obviously, if the Hamiltonian and the Lindblad operator L do not commute, $\langle H \rangle$ is not conserved for every state ρ.

The significance of this result becomes immediately obvious in the special case of a free particle —e.g. represented by a wave packet, *not* by a plane wave with constant wavevector— moving in one dimension with Hamiltonian $H = p^2/2m$. In various well-known theoretical models, the decoherence in position owing to the "environment" is described by taking L to be the particle's position operator q; for details see [37, 38, 32]. Then, since $[p, q] = \hbar/i$ and

$$[p^2, q] = p^2 q - qp^2 = (p^2 q - pqp) + (pqp - qp^2) = p[p, q] + [p, q]p = \frac{2\hbar}{i} p$$

one obtains

$$
\begin{aligned}
\frac{d\langle H \rangle}{dt} &= -\Lambda Tr([p^2/2m, q][q, \rho]) \\
&= -\frac{\Lambda}{2m} \frac{2\hbar}{i} Tr(p[q, \rho]) = -\frac{\Lambda}{2m} \frac{2\hbar}{i} Tr([p, q]\rho) \\
&= +\frac{\Lambda \hbar^2}{m} > 0
\end{aligned}
\tag{25}
$$

Thus the system appears to steadily gain energy at a constant rate [38].

This surprising result holds also in three dimensions, and for inclusion of scalar and/or vector potentials. It also remains valid for more general cases, e.g. even in cases with the decoherence factor Λ becoming a function of the "distance" $|r - r'|$; for details see [38, 39].

Undoubtedly, this result seems quite disturbing as it contradicts every conventional expectation about what the consequences of decoherence should be (besides the expected destruction of coherent superpositions of quantum states). Namely, theorem (25) seems to represent a serious weakness of the theory, because it implies that the mean energy of a free paricle should always increase; or, in other terms, the environment causing decoherence seems to act as an inexhaustible source of kinetic energy. Moreover, this continuous gain of kinetic energy is clearly incompatible with the system's attainment of equilibrium [38]. For a thorough discussion of these paradoxical findings, see [38, 39, 40].

Parenthetically, another surprising consequence of the Lindblad equation and pure decoherence may be shortly noted here. Recently, a "first principles" description of scattering from open quantum systems subject to a Lindblad-type dynamics was provided [41]. It was shown that this time evolution may cause a reduction of the system's transition rate being effectuated by scattering. This is tantamount to a shortfall of scattering intensity [41, 42], or "intensity deficit", which represents a witness of quantumness of correlations. This effect has been observed experimentally [43, 44, 45, 46, 47, 48, 49].

3.2.2. The Theoretical Model by Schulman and Gaveau. Surprisingly, a recent independent and more general theoretical analysis by Schulman and Gaveau [50, 51] appears to lead essentially to the same result. A short description of the general model is as follows.

A quantum system A, e.g. a quantum oscillator, with free Hamiltonian H_A makes an elastic collision with a second system B with free Hamiltonian H_B. Let the interaction potential be V_{AB}. The total Hamiltonian is $H = H_A + H_B + V_{AB}$. Before the collision, the two systems are assumed to be not entangled and so the complete density matrix $\rho(0)$ should be $\rho(0) = \rho_A(0) \otimes \rho_B(0)$. In general, subsequent to their collision they become entangled and the exact density operator

$$\rho(t) = U(t)\,\rho(0)\,U^\dagger(t)$$

($U(t)$: time evolution operator) is not a product state of individual density operators $\rho_A(t) = \text{Tr}_B\rho(t)$ and $\rho_B(t) = \text{Tr}_A\rho(t)$.

However, it is widely believed that once the particles are separated the quantum correlations can be dropped (provided one does not perform an experiment of Einstein-Podolsky-Rosen type), simply because measurements of physical quantities of each of the two particles cannot depend on their correlations. Thus the replacement

$$\rho(t) \rightarrow \rho_A(t) \otimes \rho_B(t) \ , \tag{26}$$

i.e. the erasure of quantum correlations, is usually assumed to be "innocuous" and e.g., not affect the energies of the systems.

The striking result by Schulman and Gaveau [50] contradicts this intuitive expectation. Putting

$$\Delta\rho(t) = \rho_A(t) \otimes \rho_B(t) - \rho(t)$$

and for a particular form of the interaction Hamiltonian, they show that for sufficiently *short* times the following relation holds:

$$\Delta E \equiv Tr(\Delta\rho(t)H) = Tr(\Delta\rho(t)\,V_{AB}) > 0 \ . \tag{27}$$

See Ref. [51] for a detailed derivation of this inequality. In simple words, the replacement of the entangled $\rho(t)$ by the non-entangled state $\rho_A(t) \otimes \rho_B(t)$ necessarily increases the system's

energy [50, 51]. This appears highly paradoxical since, as Schulman and Gaveau put it: "...losing quantum correlations should not heat the gas. You do not burn your finger because of a partial trace over a density matrix" [50].

Moreover, this result was shown to be valid for a large class of potentials, e.g. for two-body interactions, although it does not hold universally [51]. This is an interesting detail, and therefore the following details should be mentioned. The spin-boson model Hamiltonian

$$H = H_A + H_B + V_{SB} = \omega_a a^\dagger a + \omega_b b^\dagger b + g(a^\dagger + a)(b^\dagger + b)$$

was shown to exhibit the considered effect, in clear contrast to the related Jaynes-Cummings model

$$H = H_A + H_B + V_{JC} = \omega_a a^\dagger a + \omega_b b^\dagger b + g(a^\dagger b + b^\dagger a) \ ,$$

in which the effect is absent [50].

In view of the counter-intuitive character of the result (27), one may object that it is unphysical since seems to violate energy conservation. However this is not the case, as the detailed discussions of Ref. [50] explained. It was stressed that, in the situation contemplated, the coupling Hamiltonian must be considered as time dependent, because the physical approach and separation of the particles leads to a time-dependent coupling coefficient. Thus, energy conservation need not apply. Moreover, it was discussed that this "additional" energy $\Delta E > 0$ is supplied by the *translational* degrees of freedom of A and B [50], which do not appear explicitly in the Hamiltonian and thus may be considered to represent an effective "environment".

4. Experimental Consequences

4.1. Conventional Neutron Scattering Formalism

Let us briefly consider some basic formulas of conventional neutron scattering theory [24, 52, 53]. The partial differential cross section $d^2\sigma/d\omega d\Omega$ and the associated dynamic structure factor $S(\mathbf{q}, \omega)$ are determined from the experiment. For scattering by a system of N identical atoms they are related according to:

$$\frac{d^2\sigma}{d\omega d\Omega} = Nb^2 \frac{k_1}{k_0} S(\mathbf{q}, \omega) \tag{28}$$

(b: bound scattering length of atom; k_0, k_1: absolute values of wavevectors of incident and scattered neutrons; $d\Omega$: small solid angle subtended by the neutron detector in direction of $\mathbf{k}_1$ at the target). $\hbar \mathbf{q}$ and $\hbar \omega$ are the momentum and energy transfers from the neutron to a scattering nucleus, respectively, i.e, $\hbar \mathbf{q} = \hbar \mathbf{k}_0 - \hbar \mathbf{k}_1$ and $\hbar \omega = E_0 - E_1$. The subscripts "0" and "1" refer to quantities before and after the collision.

It is assumed that the neutron-nucleus interaction is well represented by the Fermi pseudo-potential [52, 24]

$$V(\mathbf{r}) = \frac{2\pi \hbar^2}{m} b \, \delta(\mathbf{r}) \tag{29}$$

(m: neutron mass; $\delta(\mathbf{r})$: delta function) which phenomenologically describes the short-range strong interaction. According to the basic van Hove theory [52] $S(\mathbf{q}, \omega)$ is given by

$$S(\mathbf{q}, \omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \exp(-i\omega t) \, F(\mathbf{q}, t) \, dt \tag{30}$$

where

$$F(\mathbf{q}, t) = \frac{1}{N} \sum_{j,k}^{N} \langle \exp(-i\mathbf{q} \cdot \mathbf{r}_j(0)) \exp(-i\mathbf{q} \cdot \mathbf{r}_k(t)) \rangle. \tag{31}$$

is the so-called intermediate correlation function. $\langle ... \rangle$ stands for the thermodynamic average of the quantum expectation values of the enclosed operators, and the $\mathbf{r}_j(t)$ denotes the position operator of atom j at time t in the *Heisenberg* representation [52, 24].

The derivations of the above formulas are based on Fermi's golden rule, which, for scattering processes, is equivalent to the first Born approximation [52, 24]. Recall that both these approximations are special cases of general first-order perturbation theory, which, however, does not hold for a singular potential like Eq. (29). The justification for the use of Fermi's golden rule in neutron scattering is that, in combination with Eq. (29), it gives the required result of isotropic (s-wave) scattering for a single fixed nucleus; see [24] for further discussions.

In the limit of sufficiently high momentum and energy transfers, $q = |\mathbf{q}| \gg 2\pi/d$ with d being the nearest-neighbor distance of scattering particles, terms with $j \neq k$ are negligible (i.e. the incoherent approximation holds [24, 25]) and one obtains

$$F(\mathbf{q}, t) = \langle \exp(-i\mathbf{q} \cdot \mathbf{r}(0)) \exp(+i\mathbf{q} \cdot \mathbf{r}(t)), \rangle \tag{32}$$

see [24, 25]) for the derivation. Note that $\mathbf{r}(t)$ is in general a N-body operator, despite the (rather misleading) notation of "position operator of a scattering atom". Furthermore, for sufficiently fast scattering, the impulse approximation (IA) [25, 54] applies, and the corresponding expression for the dynamic structure factor simplifies to:

$$S_{IA}(\mathbf{q}, \omega) = \int n(\mathbf{p}) \, \delta(\hbar\omega - \hbar\omega_r - \hbar\mathbf{q} \cdot \mathbf{p}/M) \, d\mathbf{p} \,, \tag{33}$$

Here, $n(\mathbf{p})$ is the (one-body) momentum distribution of the scattering nucleus before the collision,

$$\hbar\omega_r = \hbar^2 q^2 / 2M \tag{34}$$

is the so-called recoil energy of the nucleus, and M is the mass of the scattering nucleus. The delta-function represents energy conservation in the binary neutron-nucleus scattering process.

Eq. (33) is the basic formula describing neutron Compton scattering (NCS) [25, 54], also known as deep inelastic neutron scattering (DINS). It follows that the measured recoil peak is centred at an energy transfer $\hbar\omega_r$, i.e. at the recoil energy. The width of the peak is given by the term $\hbar\mathbf{q} \cdot \mathbf{p}/M$, i.e. the projection of the nuclear momentum before the collision on the measured momentum transfer. This term represents the well-know Doppler effect [25, 54]. Generally, the recoil peak obtained from liquid ^{4}He is broadened due to the momentum of the classical atomic motion and the atomic confinement in the effective potential caused by the atom's environment [54].

From the viewpoint of conventional scattering theory [52, 24, 25], NCS is expected to measure "single particle" quantities, e.g. the mean kinetic energy of an atom. In various cases, an effective Born-Oppenheimer potential for the atom is assumed to exist, which then can be extracted from the measured Compton profile [25].

With respect to electron–atom Compton scattering [47, 48], Bonham et al. provided a thorough analysis in the frame of conventional theory, which (as also NCS) is based on the first Born and Born-Oppenheimer approximations [55].

In this context, it is important to stress that the characteristic scattering time of the processes under consideration is very short; e.g. it is about one femtosecond in the real experiment considered below. This is of the order of the characteristic electronic rearrangement processes in molecules and condensed matter.

The above considerations show that neutron-nucleus entanglement and/or more general quantum correlations do not play any role in conventional neutron scattering theory.
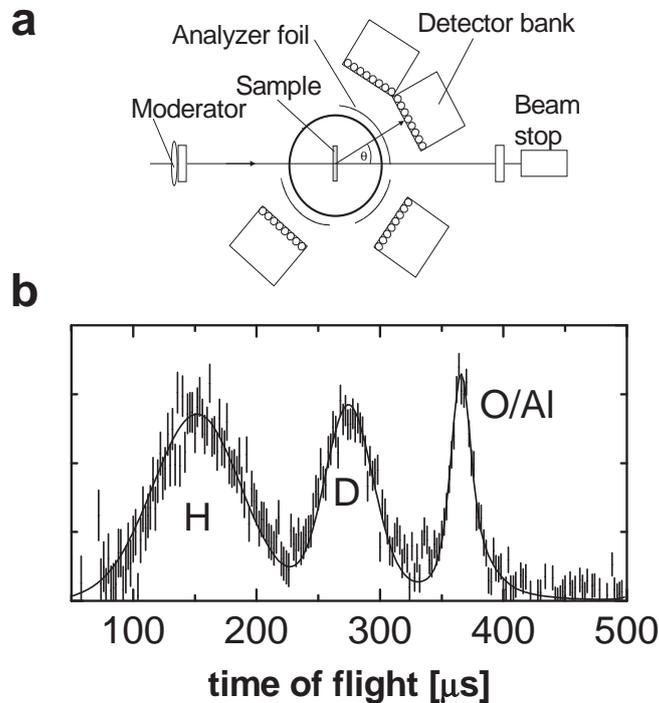
Figure 1. (a) Schematic representation of time-of-flight (TOF) spectrometer eVS/Vesuvio of ISIS. (b) A TOF-spectrum of a 20:80 H_2O-D_2O mixture in an Al cell. The H and D recoil peaks are well separated from each other and from the joint O/Al peak; adapted from [43].

4.2. On Time-of-Flight (TOF) Technique

The time-of-flight (TOF) scattering technique (cf. Fig. 1) allows the measurement of the partial differential cross section $d^2\sigma/d\omega d\Omega$ and the associated dynamic structure factor $S(\mathbf{q}, \omega)$ [56]. The TOF t of each detected neutron is determined by

$$t = \frac{L_0}{v_0} + \frac{L_1}{v_1} + t_0 .$$ (35)

Here L_0 is the source–sample distance, L_1 is the sample–detector distance. The detector is positioned at the scattering angle θ. v_0 and v_1 are the velocities of the incident and scattered neutron, respectively. t_0 is a small time offset due largely to electronic delays. We used values for L_0, L_1, θ and t_0 for the individual detectors as provided by the instrument parameter file IP0002 of the ISIS experimental report [57]; see Table 1.

The used spectrometer [57] is a so-called "inverse geometry" instrument meaning that the final velocity v_1 (and thus the wavevector k_1) of the neutrons is fixed, and v_0 varies. The final neutron energy is fixed at $E_1 = 4906$ meV (the resonance energy of ^{197}Au used as analyzer foil [58]), corresponding to a velocity $v_1 = 3.063 \times 10^4$ m/s and wavevector $k_1 = 48.663 \,\text{Å}^{-1}$.

Note that the geometric quantities L_0, L_1 and θ can be determined by appropriate methods; cf. [57]. It follows from Eq. (35) that each t corresponds to an initial velocity v_0 and thus to an energy transfer

$$\hbar\omega = \frac{1}{2}mv_0^2 - \frac{1}{2}mv_1^2 = \frac{(\hbar k_0)^2}{2m} - \frac{(\hbar k_1)^2}{2m}$$ (36)

For the corresponding momentum transfer $\hbar q$ from the neutron to the struck atom, $\hbar\mathbf{q} = \hbar\mathbf{k_0} - \hbar\mathbf{k_1}$, where

$$q = \sqrt{k_0^2 + k_1^2 - 2k_0k_1\cos\theta} \; . \tag{37}$$

It should be emphasized that, for each value of t, the associated momentum ($\hbar\mathbf{q}$) and energy ($\hbar\omega$) transfers from the neutron to the struck particle are uniquely determined. (Note that both q and ω vary over the TOF-range of the recoil peak, which is measured by a detector positioned at a fixed scattering angle θ.)

For scattering from free atoms with zero initial momentum, $p = 0$, conservation of kinetic energy and momentum in an elastic neutron-atom collision yield the kinematic relation

$$\frac{v_1}{v_0} = \frac{k_1}{k_0} = \frac{\cos\theta + \sqrt{(M/m)^2 - \sin^2\theta}}{M/m + 1}. \tag{38}$$

m and M are the masses of the neutron and struck nucleus, respectively. This equation corresponds to neutrons detected at the center of the measured recoil peak; cf. Fig. 1. For a concise account of the above kinematic formulas, see [44].

4.3. Conventional Results in View of Quantumness of Correlations
Let us first "reformulate" the preceding basic formula (33) of NCS by including the neutron state explicitly into the considerations.

First, recall that momentum conservation

$$\mathbf{p}/\hbar + \mathbf{k}_0 = (\mathbf{p}/\hbar + \mathbf{q}) + (\mathbf{k}_0 - \mathbf{q}) = (\mathbf{p}/\hbar + \mathbf{q}) + \mathbf{k}_1^\theta \tag{39}$$

($\mathbf{k}_0 - \mathbf{q} = \mathbf{k}_1^\theta$ by definition; superscript θ indicates the scattering angle at which a specific detector is positioned) and energy conservation

$$\frac{(\hbar k_0)^2}{2m} - \frac{(\hbar k_1)^2}{2m} = \frac{(\mathbf{p} + \hbar\mathbf{q})^2}{2M} - \frac{(\mathbf{p})^2}{2M} = \hbar\omega_r + \hbar\mathbf{q} \cdot \mathbf{p}/M \tag{40}$$

hold in the binary collision. The assumed quantum state of the neutron-nucleus system before collision is factorizable,

$$\rho_{\text{init}} = \left(\int d\mathbf{p}\, n(\mathbf{p})\, |\mathbf{p}\rangle\langle\mathbf{p}| \right) \otimes |\mathbf{k}_0\rangle\langle\mathbf{k}_0| \tag{41}$$

Since conventional theory takes the neutron's final state to be pure (a plane wave with well defined and fixed $\mathbf{k}_1^\theta$), the state of the neutron-scatterer system after scattering is still factorizable (i.e. not entangled),

$$\rho_{\text{final}}^\theta = \left(\iint^\star d\mathbf{q}\, d\mathbf{p}\, n(\mathbf{p})\, |\mathbf{p}/\hbar + \mathbf{q}\rangle\langle\mathbf{p}/\hbar + \mathbf{q}| \right) \otimes |\mathbf{k}_1^\theta\rangle\langle\mathbf{k}_1^\theta| \tag{42}$$

where the $\star$ on the integral denotes restriction to momentum and energy conservation. Recall that $\mathbf{k}_0$ is not fixed, and thus the conservation conditions can be fulfilled by various pairs of values of $\mathbf{p}$ and $\mathbf{q}$. (We use both notations $|\mathbf{p}/\hbar + \mathbf{q}\rangle$ and $|\mathbf{p} + \mathbf{q}\hbar\rangle$ to represent the same physical state.) Clearly, these two-body initial and final states of conventional theory are not entangled and, moreover, classical-classical; see section 2.2. Thus they have zero quantum discord.

4.3.1. Relation to Quantumness of Correlations. We now proceed in reconsidering the derivations and the basic formula (33) of NCS in the light of the modern understanding of the quantumness of correlations; see section 2. Against this background, the basic formulas of the conventional theory of NCS appear to have a serious weakness. Notably, in conventional theory the neutron represents a classical particle, its quantum degrees of freedom are not taken into account. ($\mathbf{k}_0$, $\mathbf{k}_1$ and also $\mathbf{q}$ are not operator quantities but only c-numbers.) Moreover the quantum states of the neutron "disappear", since the average operation $\langle...\rangle$ in the formulas of section 4.1 contains quantum states of the scattering particles only.

According to basic quantum mechanical principles, on the other hand, one generally would expect that a collision of two particles A and B results in entanglement of these particles. E.g., for s-wave scattering, the final state is given by a spherical outgoing wavefunction (in the center-of-mass system) of their relative coordinate $r = |\mathbf{r}_A - \mathbf{r}_B|$,

$$\Psi_{AB} \propto \frac{\exp{(ikr)}}{r} \tag{43}$$

(k: suitable constant) and, due to the strict validity of momentum conservation, A and B will generally become entangled in momentum space after scattering. [This entangling process should not be assumed to be equivalent to the well-known separation of centre-of-mass and relative coordinate motions, which appears also in the two-body problem of classical mechanics.]

Then why the neutron-scatterer entanglement (or more general quantum correlations) does not play a role in the formulas of conventional theory? The final state of the neutron is here *assumed* to be a well defined plane wave $|\mathbf{k}_1\rangle$, which is a pure one-particle state. This assumption is necessary in order to apply the first Born approximation (and the Fermi golden rule) and to derive the scattering probability in the $\mathbf{k}_1$-direction in which the neutron detector stands. Obviously, this is tantamount to tacitly presume a collaps of the entangled (or quantum correlated) neutron-nucleus state and assume a factorizable final state $|\mathbf{k}_1\rangle \otimes |\mathbf{p}/\hbar + \mathbf{q}\rangle$, in which the neutron and the scatterer are in pure final states $|\mathbf{k}_1\rangle$ and $|\mathbf{p}/\hbar + \mathbf{q}\rangle$, with well defined momenta. In other words, the existing entanglement between neutron and scatterer has been "removed by hand". As shown below, this "removal" appears to have measurable energetic consequences which contradict conventional theory.

In the context of the following considerations it is important to observe that the scattering atom (nucleus) is not free in a real experiment but interacts with its environment. Moreover, the characteristic scattering time τ_{scatt} of NCS is very short but finite (about 1 femtosecond and less [43, 44]), and thus this interaction is expected to cause a partial degradation of the considered entanglement, which may still lead to states containing quantum discord.

The same conclusion is achieved in the general frame of the theoretical work by Streltsov et al. [29] discussed above: Namely, M of [29] corresponds to the neutron, the subsystem A to the single scattering nucleus X, B to the environment $\mathcal{E}$ of the stuck nucleus or atom, and AB to the complete N-body system constituting the sample, $X + \mathcal{E}$. The latter, being an interacting system, may be naturally expected to be quantum correlated with nonzero discord. Then, the neutron scattering process creates entanglement between the neutron and the scattering sample [29].

Now let us consider the concrete physical context of NCS from normal liquid ^{4}He, which we also study in the context of a real experiment (see below). Recalling the fact that NCS transfers momentum $\hbar q$ to one He atom (with, say $q \sim 100$ Å^{-1}), we can reasonably assume the struck atom to strongly interact with adjacent He atoms (i.e. the environment $\mathcal{E}$) a few Ångströms apart, after an ultrashort time (of the order of one femtosecond) following the impulsive neutron-He collision. Note that the scattered neutron is still very near, also a few Ångströms apart. The scattered neutron reaches the detector without further disturbance, at a distance of about 0.6 m away and after some tens of microseconds (see below)—about 9 orders of magnitude later than the previous process.

These estimates suggest the following important insights:

At and shortly after the moment of collision, the neutron and the struck He should become entangled, as explained above, see Eq. (43). Due to the "monogamy" of entanglement [21], one may expect that any entanglement of the struck He atom with adjacent atoms existing previously (i.e. before collision) will be removed.

The first object that interacts with (or "measures") the neutron-He binary system and disturbs and/or reduces its state (say, by a POVM) is not the neutron detector, but the environment $\mathcal{E}$ of the struck He. (Recall that we are considering liquid He.) This happens on timescales of the order of 1 femtosecond after the initial collision, which is a time difference similar to the so-called characteristic scattering time τ_{scatt} of NCS [44, 25]. Due to its many-body characteristics, this He-$\mathcal{E}$ interaction can not reasonably be assumed to lead to a pure "final" state of the He atom. So let ρ_λ^{He} (index λ indicating the states) instead be one of the typical mixed reduced states of He, and correspondingly let ρ_λ^n be the associated mixed reduced states of the scattered neutron. These states may still be partially entangled. Nevertheless one might make the ansatz of a separable state

$$\rho^{n,He}(t \sim \tau_{scatt}) \approx \int d\lambda\, p(\lambda)\, \rho^n(\lambda) \otimes \rho^{He}(\lambda), \tag{44}$$

of the type of Eq. (17), section 2.2.1, which is a quantum-quantum state and thus in general exhibits discord. In other words, the environment $\mathcal{E}$ is continuously "measuring" the "neutron+He" system, thus continuously changing its quantum correlations.

Later, after a characteristic relaxation or equilibration time τ_{relax}, the struck He should be well equilibrated with its environment and again constitute an N-body state $\rho^{He,\mathcal{E}}$ of the same physical nature as that before the collision. The neutron assumes a mixed state ρ^n, which due to the process of detection is further selected and can approximately represented as a pure state $|\mathbf{k}_1^\theta\rangle\langle\mathbf{k}_1^\theta|$ of conventional theory. The final measurement of the neutron by the detector can, due to the principle of causality, not affect the dynamics and/or the correlations of the neutron-He collisional process.

The following presents a *schematic* summary of the physical processes that, according to our point of view, describe the dynamics of a NCS experiment:

$$t = -\infty \quad : \quad \rho^n \otimes \rho^{He,\mathcal{E}} \tag{45}$$

$$\text{collision}: \quad t \approx 0 \quad : \quad \rho^{n,He} \otimes \rho^{\mathcal{E}} \tag{46}$$

$$t \leq \tau_{scatt} \sim 10^{-15}\text{s} \quad : \quad \left(\int d\lambda\, p(\lambda)\, \rho^n(\lambda) \otimes \rho^{He}(\lambda)\right) \otimes \rho^{\mathcal{E}} \tag{47}$$

$$t > \tau_{relax} \quad : \quad \rho^{He,\mathcal{E}} \otimes \rho^n \tag{48}$$

$$t = +\infty \quad : \quad \rho^{He,\mathcal{E}} \otimes |\mathbf{k}_1^\theta\rangle\langle\mathbf{k}_1^\theta| \tag{49}$$

(Note: A symbol appearing in more than one line may refer to varying physical quantities; e.g. $\rho^{He,\mathcal{E}}$ in first line should be significantly different from that in line 4.)

4.4. Experiment

We investigated NCS from normal-state liquid ^{4}He at $T \approx 2$ K and saturated vapor pressure. The original data [59] were kindly provided by ISIS under the "ISIS Data Policy", cf. [60].

Here we present results obtained from TOF-spectra measured with nine detectors (see Table 1) in backscattering, at mean scattering angle $\bar{\theta} = 134.594° \pm 0.2324°$. The mean sample-detector distance is $\overline{L_1} = (0.6392 \pm 0.0038)$ m; the mean time offset is $\overline{t_0} = (-0.3246 \pm 0.0943) \cdot 10^{-6}$ s. The L_0 length is given as $L_0 = 11.0050$ m. These specific detectors were selected because their parameters, and especially their angles, are very similar and thus the TOF spectra can be
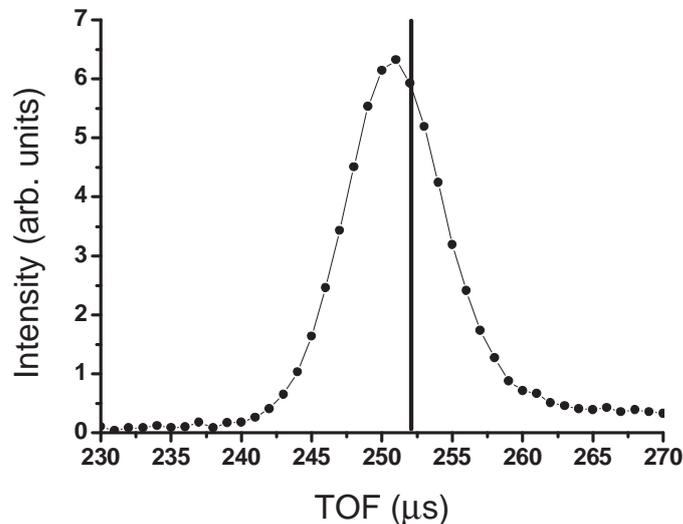
Figure 2. The accumulated TOF spectrum (9 detectors, see Table 1; as measured) of ^{4}He at about 2 K and saturated vapor pressure; see the text. According to the instrument parameters, Table 1, the conventionally expected position of the peak is given by the vertical bar. The observed peak displacement corresponds to a transfer energy increased by ca. 110 meV. This effect has no conventional interpretation; see the text.

Table 1. The instrument parameters of the detectors used in the data analysis; reproduced from file IP0002.DAT [57]

Det. No	θ (degrees)	t_0 (μsec)	L_0 (m)	L_1 (m)
10	134.3881	-0.2896000	11.0050	0.6410817
11	134.8883	-0.3632000	11.0050	0.6379817
12	134.7490	-0.1396000	11.0050	0.6374661
14	134.5436	-0.2448000	11.0050	0.6318235
18	134.9788	-0.3489000	11.0050	0.6433660
54	134.3012	-0.4116000	11.0050	0.6416131
56	134.4089	-0.3321000	11.0050	0.6421713
58	134.5938	-0.4649000	11.0050	0.6356288
62	134.4992	-0.3263000	11.0050	0.6414817

directly accumulated in order to improve signal to noise ratio. The resulting accumulated TOF spectrum (as measured) is shown in Figure 2.

Table 1 contains the data of the used detectors, and is reproduced from the file containing instrument parameters, IP0002.DAT, in Ref. [57], pages 39-41.

Using the aforementioned final neutron energy $E_1 = 4906$ meV, one obtains $k_1 = 48.663$ Å^{-1}. For scattering from ^{4}He, and using the formulas of subsection 4.2, the corresponding initial wavevector at $\bar{\theta}$ is $k_0 = 75.517$ Å^{-1}, and the related momentum transfer is $\hbar q$ with $q = 115.027$ Å^{-1}. The associated recoil energy (or energy transfer) is $\hbar\omega_r = 6908.7$ meV.

These numerical values, which are obtained from conventional theory, and the basic formula Eq. (35) predict the TOF position of the ^{4}He recoil (Compton) peak to be at $t_{conv} = 252.067$ μs. This value is shown with a vertical bar in Figure 2. One observes a significant displacement

of the measured recoil peak to shorter TOF's, and equivalently to larger energy transfers (since the instrument is of "inverse geometry"; see above). The observed displacement $\Delta_Q t$ of the peak maximum is about one microsecond, $\Delta_Q t \approx 1.0$ μs. This corresponds to an increased initial velocity (and energy) of the neutron, and thus to an energy transfer increased by

$$\Delta_Q E \approx 110 \text{ meV}. \tag{50}$$

Due to the uncertainty of the TOF-position of the peak, we estimate the error of $\Delta_Q E$ to be about $\pm 10\%$.

It may be noted that the intrinsic width of the recoil peak of ^{4}He is very narrow, and thus the width shown in Figure 2 is mostly due to the instrumental resolution. Hence $\Delta_Q E$ should not be considered as being "too small".

4.4.1. Quantum Maxwell demon and $\Delta_Q E > 0$. Note that the epithermal neutron, the struck ^{4}He atom, and the adjacent ^{4}He atoms (being strongly disturbed by the former) do not represent thermalized systems or baths, due to the smallness of the characteristic time window τ_{scatt}, see above. Thus the experimental result (50) cannot be directly compared with the well know theoretical result

$$\Delta W = W_Q - W_C = kTD(A|B) \tag{51}$$

(k: Boltzmann constant; T: temperature) connecting quantum discord [61], Eq. (13), with the difference between the work-extraction efficiency of the quantum and classical demons, which may extract works W_Q and W_C, respectively. The quantity ΔW is also called work deficit, cf. [21].

However, using the traditional demon's scenario, one may obtain an associated result as follows. Let the neutron correspond to the subsystem A, and the struck ^{4}He atom together with certain adjacent ^{4}He atoms interacting with it to subsystem B. The demon continuously "reads off", or erases, quantum correlations (i.e. information) encoded in $A+B$ during the time window τ_{scatt} of the collision. For a classical demon, this erasure should have an inherent positive work cost [12], e.g. it may dissipate some kinetic energy of A, resulting to a *smaller* energy transfer $\hbar\omega_r - W_C$ for the binary neutron-^{4}He collision than the conventional recoil energy $\hbar\omega_r$. In the case of a quantum demon, however, this process may have "negative costs of erasure" [31], e.g. it may "extract" some additional work W_Q, resulting to a *larger* energy transfer $\hbar\omega_r + W_Q$ for the binary collision than the conventionally expected value $\hbar\omega_r$. Thus we can define the "efficiency" of the quantum demon by the ratio:

$$\frac{W_Q - W_C}{\hbar\omega_r} \geq \frac{\text{measured E-transfer} - \hbar\omega_r}{\hbar\omega_r} = \frac{110 \text{ meV}}{6908.7 \text{ meV}} \approx 1.6 \text{ \%} \tag{52}$$

In other words, the Maxwell demon observing the actual NCS process from ^{4}He is a quantum one—and about 1.6% more efficient than any classical demon.

5. Discussion and Conclusions

At first glance, one obvious objection to the theoretical arguments presented is that the observed shift of the NCS peak might have a more simple explanation. For instance, the following possibility might be proposed: The observed mean energy transfer is slightly higher than the one predicted by standard theory. Could it not be interpreted simply as being caused by a slightly higher effective mass of the scatterer? The assumption of a free scatterer is clearly a crude (although very common) approximation. If the scatterer is partially bound with adjacent particles, one scatters on a seemingly little heavier object, as the environment to which it is bound has a certain mass. This effective increase of mass might be due to some simply classical

binding (e.g., the extreme case would be analogous to the well known Mössbauer effect, where the crystal as a whole acts as a scatterer. Similarly in solid state physics it is common to talk about heavy electrons, which is nothing more than simply a breakdown of the independent particle scatterer approximation due to correlation which causes an apparent heavier mass). However, the proposed consideration demonstrates quite the opposite of what is intended. Namely, according to standard theory of binary collisions, the mentioned higher effective mass of the scattering ^{4}He must necessarily cause a lower (not higher!) energy transfer (e.g. collision with a scatterer of infinite mass causes no energy transfer at all). In other terms, and in the light of the above remarks, it is clear that no classical partial bonding to the environment can lead to a smaller effective mass of the scatterer—which would then provide a conventional interpretation of the experimentally measured increased energy transfer.

The experimental results discussed in the previous section can be understood by noting that erasure of quantum correlations in the system "neutron+^{4}He", which is interacting with the environment, may have negative work costs [31] that affect the quantum dynamics of neutron scattering on ultrafast timescales. First indications of this effect have been recently observed in scattering from molecular H_2 [62] and D_2 [63].

The aforementioned theoretical considerations for the interpretation of the experimental finding (50) in the frame of quantum information theory are qualitative; a quantitative theoretical model for calculating the positive energy-transfer shift of the NCS-recoil peak is presently not available.

The phenomena of entanglement, discord and decoherence are not considered in conventional neutron scattering theory [25, 24, 52, 53]. As mentioned above, the observed positive energy-transfer deviation, $\Delta_Q E > 0$, contradicts all conventional expectations. This becomes even more evident by considering the opposite (and conventionally expected) case of a negative $\Delta_Q E$, which is attributed to so-called final-state-effects [25]. Such effects arise if the energy transfer is not sufficient to validate the impulse approximation [25, 54], in which case the struck particle is not "fully free" and a part of the neutron's kinetic energy must be used to overcome the particle-environment binding forces, resulting in a reduction of the energy transferred to the struck particle.

In contrast to conventional theory, and based on the aforementioned theoretical understanding of quantum correlations and their dynamics accompanying elementary scattering processes, we attribute the result $\Delta_Q E > 0$ to "negative energetic costs" of correlations-erasure discussed in sections 2 and 3. In particular we refer to the qualitatively similar results following from the Lindblad equation [38] (sect. 3.2.1), the result of the Schulman-Gaveau analysis [50] (sect. 3.2.2), and the "negative work costs" of negative conditional entropy by del Rio et al. [31] (sect. 2.2.3). Note that in all these cases the quantumness of correlations leads to higher work-values as compared to the associated processes in the absence of quantum correlations. The same holds for quantum Maxwell demons, which can extract more work from quantum correlations than classical demons; cf. [21, 22, 61].

In view of these remarks and considerations, our scattering effect offers new physical insights into entanglement, discord and other measures of quantumness of correlations, as well as on their operational meaning. Moreover, the general character of its causes suggests that this effect may be observed in other experimental areas involving scattering (e.g. inelastic x-ray scattering, electron-atom Compton scattering, etc.), and also in a lower energy-transfer range (e.g. in vibrational spectroscopy with neutrons [64, 65, 66, 67]). Related work is in progress [68].

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