# Quantum signatures in the dynamics of two dipole-dipole interacting soft dimers

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**Abstract.** The quantum covariances of physically transparent pairs of observables relative to two dimers hosted in a solid matrix are exactly investigated in the temporal domain. Both dimers possess fermionic and bosonic degrees of freedom and are dipolarly coupled. We find out and describe clear signatures traceable back to the presence and persistence of quantum coherence in the time evolution of the system. Manifestations of a competition between intramolecular and intermolecular energy migration mechanisms are brought to light. The experimental relevance of our results is briefly commented.

**PACS.** 03.65.Ud Entanglement and quantum nonlocality -61.72.Yx Interaction between different crystal defects; gettering effect -34.30.+h Intramolecular energy transfer; intramolecular dynamics; dynamics of van der Waals molecules

## **1** Introduction

Over the last twenty years the ambitious goal of performing decoherence-protected experiments aimed at investigating the dynamics of a single or few atoms or at manipulating the statistics of a single, material or not, quantum harmonic oscillator has been successfully achieved [1–3]. The first conceptually seminal experiments effectively based on the strong interaction of a single atom with a high-Q cavity single photon [4], have paved the way to the Cavity Quantum Electrodynamics (CQED) research area [3]. Since then other physical scenarios wherein physics at such a basic level could be realized have been searched. Here we quote for instance the area of trapped ions [1,2] where fundamental matterradiation interaction models have been successfully implemented, the role of the electromagnetic field being played by the oscillatory degrees of freedom of the confined ion center of mass. Very recently, it has been moreover experimentally demonstrated that the strong coupling limit of CQED may be reached and also overcome in a circuit consisting of a fully electrically controllable superconducting quantum two-level system, the Cooper pair box, coupled to a single mode of the quantized radiation field in an onchip cavity formed by a superconductive transmissive line resonator [5,6].

A common aspect of all these investigations is that they provide the opportunity of testing quantum mechanics at a very basic level. In this paper we wish to propose another possible scenario of the same type in the context of solid state physics. The natural or artificial presence of defects in a solid host might in fact be exploited to investigate the radiationless dynamics of a microscopic system composed by a pair of coupled defects [7–9]. Our scope is to draw attention on such a solid state laboratory to study simple bipartite systems (that is composed of two subsystems) to bring to light the occurrence of quantum signatures in the behaviour of appropriately chosen observables having a clear physical meaning.

It is worth to underline that this kind of investigations are in the grasp of the experimental current setup potentialities thanks to so called site selective laser techniques today allowing to store induced excitations in a defect placed at a prefixed lattice site at t = 0 and to resolve in time its dynamical behaviour [10]. Moreover, in suitably low-doped crystals wherein the probability of pair formation is of practical interest and different couples of defects are effectively isolated from each other [11,12], the individual study of the dynamics of a pair is in the current experimental reach.

Thus we might have at our disposal the possibility of realizing a system of only two coupled (possibly two-level) defects, within which to fix an initial condition and to read out the subsequent temporal evolution in search of convincing hallmarks of quantum coherence manifestations.

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With this motivations we propose in this paper a simple fully quantistic model to describe radiationless energy migration processes between equal impurity centres closely located in a solid possessing a low dopant concentration. In our model each impurity center is thought of as a *molecule* possessing coupled internal fermionic and bosonic degrees of freedom. Moreover the two molecules are assumed to interact by a dipole-like term. We prove that the Hamiltonian model representing such a composite system may be exactly treated starting from appropriate initial conditions. Our main scope is to study the time evolution of quantum correlations get established in the molecule-molecule system highlighting the role of the intermolecular coupling strength against the intramolecular one in giving rise to unexpected quantum signatures. It is of relevance that investigating the microscopic origin of macroscopic physical properties exhibited by novel materials incorporating optically active ions, not only provides a piece of basic research but at the same time offers conceptual tools to understand the underlying mechanisms on which various applications rely [13].

This article is organized as follows. In the next section, after presenting our Hamiltonian model and showing that it can be exactly solved in its one-excitation subspace, we compare the time evolutions of the superposition of two appropriate states of the bipartite system, considering both the fully quantistic linear combination and the fully classical one. A detailed investigation of the temporal behaviour of the quantum covariances of suitable couples of physically transparent observables pertaining to the two molecules is reported in Section 3, while the experimental relevance of these results is discussed in the subsequent Section 4. Some conclusive remarks are formulated in the final section.

# 2 The Hamiltonian model and one excitation exact dynamics

The system on which we focus consists of two interacting identical molecules closely located at fixed points in a solid. Each molecule possesses internal degrees of freedom, behaving like a Pauli pseudo-spin  $\frac{1}{2}$  as well as vibrational dynamical variables. For the sake of simplicity we consider only one vibrational normal mode and for this reason sometimes we refer to each molecular subsystem as to a dimer. Let's indicate by  $\sigma_k^{(i)}$  (k = x, y, z) and  $a_{(i)}$  $(a_{(i)}^{\dagger})$  the Pauli operators describing the electronic and vibrational degrees of freedom, respectively, relative to the *i*th molecule. Assume that our physical system is governed by the following Hamiltonian model:

$$H = H_0 + H_{DD} + H_1 + H_2 \tag{1}$$

where

$$H_0 = \frac{1}{2}\omega_0 \left(\sigma_z^{(1)} + \sigma_z^{(2)}\right) + \omega \left(a_1^{\dagger}a_1 + a_2^{\dagger}a_2\right)$$
(2)

is the free energy and

$$H_{DD} = \lambda \left( \sigma_x^{(1)} \sigma_x^{(2)} + \sigma_y^{(1)} \sigma_y^{(2)} \right) \tag{3}$$

describes a dipole-dipole like interaction between the two molecules deprived of its diagonal term  $\sigma_z^{(1)} \sigma_z^{(2)}$ , being mainly interested in contributions describing excitation energy back and forth inter-passages. Finally, in order to take into account intramolecular nonradiative energy transfers, we introduce a direct coupling between the bosonic and fermionic dynamical variables relative to each molecule in the form

$$H_i = \epsilon \left( \sigma_+^{(i)} a_i + \sigma_-^{(i)} a_i^{\dagger} \right) \quad (i = 1, 2) \tag{4}$$

with  $\sigma_{\pm}^{(i)} = \frac{1}{2}(\sigma_x^{(i)} \pm i\sigma_y^{(i)})$ . In (2)  $\omega_0$  is the Bohr frequency associated to the electronic levels of each single dimer whereas  $\omega$  is the frequency relative to its vibrational normal mode. The form of  $H_i$  aims at describing the interaction between vibrational modes and electronic levels of the molecule. The scope we pursue in this paper is not a detailed representation of a specific physical situation. Rather we wish to catch a reasonable ingredient playing an important role in energy exchange mechanism between different degrees of freedom within a single molecule. Neglecting the counter rotating terms,  $\sigma_+^{(i)} a_i^{\dagger}$  and  $\sigma_-^{(i)} a_i$ , in equation (4) confines the applicability of the ideas of the present paper to not too strong regimes ( $\epsilon \ll \omega_0$ ) where in addition  $\omega_0 \sim \omega$ .

It is easy to prove that the total excitation pair number operator defined as

$$N = N_1 + N_2 \tag{5}$$

with

$$N_i = a_i^{\dagger} a_i + \frac{1}{2} \sigma_z^{(i)} + \frac{1}{2} \tag{6}$$

is a constant of motion. This circumstance, as we shown later in this section, gives us the possibility of exactly decoupling the two dipole-dipole interacting dimers provided that the initial total excitation pair number is less than 2. In order to do this let's firstly introduce the unitary operator U given by

$$U = \exp\left\{\frac{\pi}{4} \left(\sigma_{+}^{(1)} \sigma_{-}^{(2)} - \sigma_{-}^{(1)} \sigma_{+}^{(2)}\right)\right\}.$$
 (7)

It is easily demonstrable that

$$U^{\dagger}\sigma_{+}^{(i)}U = \frac{1}{\sqrt{2}} \left(\sigma_{+}^{(i)} + (-1)^{i}\sigma_{+}^{(j)}\right) \quad i = j = 1, 2 \quad i \neq j$$
(8)

and  $[H_0, U] = 0$ . Thus the canonical transformation accomplished by U leaves unchanged the free Hamiltonian  $H_0$  and in particular  $\sigma_z^{(1)} + \sigma_z^{(2)}$  being  $U^{\dagger}(\sigma_z^{(1)} + \sigma_z^{(2)})U \equiv \sigma_z^{(1)} + \sigma_z^{(2)}$ . Moreover, appropriately rewriting the intermolecular coupling given by (3) as function of  $\sigma_{\pm}^{(i)}$ , it is not difficult to prove that

$$U^{\dagger}H_{DD}U = \lambda \left(\sigma_z^{(2)} - \sigma_z^{(1)}\right).$$
(9)

The meaning of (9) is that, as a result of the unitary transformation U, the presence of  $H_{DD}$  in the original microscopic model H renormalizes the electronic frequency of the two identical molecules in a different way. As far as the intramolecular coupling terms we immediately have

$$H_{12} \equiv U^{\dagger}(H_1 + H_2)U = \frac{\epsilon}{\sqrt{2}} \left[ (a_1 + a_2\sigma_z^{(2)})\sigma_+^{(1)} + (a_2 - a_1\sigma_z^{(1)})\sigma_+^{(2)} + h.c. \right].$$
(10)

The existence of the constant of motion N above introduced, implies that, starting from a state with a well defined total number of pair excitations n, the dynamics of the physical system here considered remains confined in the finite-dimensional Hilbert subspace singled out by this eigenvalue of the operator N. If we consider the case n < 2only, it is not difficult to convince oneself that the interaction operator  $\tilde{H}_{12}$  expressed by (10), may be, indeed, effectively represented in the form

$$\tilde{H}_{12} = \frac{\epsilon}{\sqrt{2}} \left[ (a_1 - a_2)\sigma_+^{(1)} + (a_1 + a_2)\sigma_+^{(2)} + h.c. \right].$$
(11)

We stress that  $\tilde{H}_{12}$  as given by (11) is not coincident with the operator defined by equation (10) the latter one being the restriction of the former in the Hilbert subspace under scrutiny.

From now on we are, by hypothesis, within this prefixed subspace.

Looking at (11) we may appreciate that  $\tilde{H}_{12}$ , when compared with the original not transformed operator  $H_1 + H_2$ , describes a new situation wherein the fermionic dynamical variables of each molecule get coupled with both vibrational modes through the two different linear combinations  $a_1 - a_2$  and  $a_1 + a_2$ . This circumstance suggests that, accordingly combining the vibrational dynamical variables of the two dimers, we might succeed in decoupling the two dipole-dipole interacting molecules. If, indeed, we introduce the unitary operator

$$T = \exp\left\{\frac{\pi}{4}\left\{a_1^{\dagger}a_2 - a_1a_2^{\dagger}\right\}\right\}$$
(12)

satisfying the following transformation properties

$$T^{\dagger}a_1T = \frac{1}{\sqrt{2}}(a_1 + a_2) \tag{13}$$

$$T^{\dagger}a_2T = \frac{1}{\sqrt{2}}(a_1 - a_2) \tag{14}$$

we immediately obtain

$$T^{\dagger}H_0T = H_0 \tag{15}$$

and

$$T^{\dagger}\tilde{H}_{12}T = \epsilon \left[ \left( a_1 \sigma_+^{(1)} + h.c. \right) + \left( a_2 \sigma_+^{(2)} + h.c. \right) \right].$$
(16)

Summarizing we can thus conclude that, confining our attention on the Hilbert subspace correspondent to total excitation pair number less than 2, the action of the unitary operator UT is to transform the microscopic Hamiltonian model (1) into the following one

$$\tilde{H} \equiv T^{\dagger} U^{\dagger} H U T \equiv H^{(1)} + H^{(2)} \tag{17}$$

where

$$H^{(i)} = \frac{1}{2} (\omega_0 + (-1)^i \lambda) \sigma_z^{(i)} + \omega a_i^{\dagger} a_i + \epsilon \left[ (a_i \sigma_+^{(i)} + h.c. \right].$$
(18)

It is of relevance to underline at this point that the advantage stemming from the canonical transformation accomplished by UT is that the new Hamiltonian model (17) gets rid of the direct dipolar coupling term, representing indeed a pair of fictitious non interacting dimers. Since, in addition, the mathematical structure of the resulting intramolecular coupling has the form of the well-known exactly treatable Jaynes-Cummings interaction [14], it turns out that the dynamics of  $\tilde{H}$  may be solved without any approximation. In order to make clearer this statement, let's suppose that at t = 0 the system under scrutiny is prepared in a state  $|\psi(0)\rangle$  belonging to the one excitation Hilbert subspace generated by the following basis B = $\{|0_1, 0_2, +, -\rangle, |0_1, 0_2, -, +\rangle, |1_1, 0_2, -, -\rangle, |0_1, 1_2, -, -\rangle\}$ being  $|n_1, n_2, \sigma_1, \sigma_2\rangle$  a common eigenstate of  $a_1^{\dagger}a_1, a_2^{\dagger}a_2,$  $\sigma_z^{(1)}$  and  $\sigma_z^{(2)}$ , such that

$$\langle n_1^{\dagger} a_1 | n_1, n_2, \sigma_1, \sigma_2 \rangle = n_1 | n_1, n_2, \sigma_1, \sigma_2 \rangle,$$
 (19)

$$a_{2}^{\dagger}a_{2}|n_{1}, n_{2}, \sigma_{1}, \sigma_{2}\rangle = n_{2}|n_{1}, n_{2}, \sigma_{1}, \sigma_{2}\rangle$$
(20)

$$\sigma_z^{(1)}|n_1, n_2, \sigma_1, \sigma_2\rangle = u(\sigma_1)|n_1, n_2, \sigma_1, \sigma_2\rangle, \quad (21)$$

$$\sigma_z^{(2)}|n_1, n_2, \sigma_1, \sigma_2\rangle = u(\sigma_2)|n_1, n_2, \sigma_1, \sigma_2\rangle$$
(22)

$$n_i = 0, 1, 2, \dots$$
  $\sigma_i = \pm, \quad u(\pm) = \pm 1.$  (23)

At any time instant t > 0 we can thus write

$$\begin{aligned} |\psi(t)\rangle &\equiv e^{-iHt} |\psi(0)\rangle = UT e^{-iHt} T^{\dagger} U^{\dagger} |\psi(0)\rangle \\ &\equiv UT e^{-iH^{(1)}t} e^{-iH^{(2)}t} T^{\dagger} U^{\dagger} |\psi(0)\rangle. \end{aligned}$$
(24)

Equation (24) immediately suggests that, knowing the action of UT and  $T^{\dagger}U^{\dagger}$  upon the basis B, in order to evaluate the temporal evolution of the system we may take advantage from the already known solutions of the Jaynes-Cummings dynamics [14].

#### 3 Quantum correlation effects

Exploiting the results presented in the previous section, we now investigate on some interesting features characterizing the temporal evolution of the system under scrutiny supposing, in accordance with the underlying assumptions making (1) unitarily equivalent to (17), that at t = 0 only one excitation is injected into the system. In order to bring to light the appearance of quantum effects stemming from the quantum correlations which get established between the two dipole-dipole coupled molecules, we will concentrate our attention on two exemplary cases. In more detail we suppose that the system is initially prepared in a statistical mixture or in a quantum coherent superposition of the two states  $|0_1, 0_2, +, -\rangle$  and  $|0_1, 0_2, -, +\rangle$  both belonging to the one excitation Hilbert subspace. Studying the quantum covariance of the "stretching operator" of the two molecules as well as that of the dipole moments we will put transparently into evidence interesting quantum signatures directly traceable back to the establishment of quantum correlations. First of all let's observe that, taking into account the transformation properties (8) and (13) it is possible to prove that

$$\begin{aligned} |\psi_1(t)\rangle &\equiv e^{-iHt} |0_1, 0_2, +, -\rangle \\ &= a(t)|0_1, 0_2, +, -\rangle + b(t)|0_1, 0_2, -, +\rangle \\ &+ c(t)|1_1, 0_2, -, -\rangle + d(t)|0_1, 1_2, -, -\rangle \end{aligned}$$
(25)

with

$$a(t) = \cos\left(\frac{\lambda}{2}t\right)\cos\left(\frac{R}{2}t\right) - \frac{\lambda}{R}\sin\left(\frac{\lambda}{2}t\right)\sin\left(\frac{R}{2}t\right)$$

$$b(t) = -i\left[\sin\left(\frac{\lambda}{2}t\right)\cos\left(\frac{R}{2}t\right) + \frac{\lambda}{R}\cos\left(\frac{\lambda}{2}t\right)\sin\left(\frac{R}{2}t\right)\right]$$
(26)

$$c(t) = -\frac{2i\epsilon}{R}\cos\left(\frac{\lambda}{2}t\right)\sin\left(\frac{R}{2}t\right)$$
(28)

$$d(t) = -\frac{2\epsilon}{R}\sin\left(\frac{\lambda}{2}t\right)\sin\left(\frac{R}{2}t\right)$$
(29)

where  $R = \sqrt{\lambda^2 + 4\epsilon^2}$  and  $\lambda \gg |\omega_0 - \omega|$ . Starting from (25) and taking into account the Hamiltonian model given by (1), it is easy to convince oneself that

$$\begin{aligned} |\psi_2(t)\rangle &\equiv e^{-iHt} |0_1, 0_2, -, +\rangle \\ &= b(t) |0_1, 0_2, +, -\rangle + a(t) |0_1, 0_2, -, +\rangle \\ &+ d(t) |1_1, 0_2, -, -\rangle + c(t) |0_1, 1_2, -, -\rangle. \end{aligned}$$
(30)

Thus, starting from the initial statistical mixture

$$\rho(0) = \frac{1}{2} (|0_1, 0_2, +, -\rangle \langle 0_1, 0_2, +, -| \\
+ |0_1, 0_2, -, +\rangle \langle 0_1, 0_2, -, +|) \quad (31)$$

at any time instant t the state of the system can be written as

$$\rho_M(t) = \frac{1}{2} (|\psi_1(t)\rangle \langle \psi_1(t)| + |\psi_2(t)\rangle \langle \psi_2(t)|).$$
(32)

The state of the system at a generic time instant t assumes on the contrary the form

$$\rho_S(t) = |\psi(t)\rangle\langle\psi(t)| \tag{33}$$

with

$$|\psi(t)\rangle = \frac{1}{\sqrt{2}}(|\psi_1(t)\rangle + |\psi_2(t)\rangle) \tag{34}$$

if we suppose to prepare the system at t = 0 in the linear quantum superposition

$$|\psi(0)\rangle = \frac{1}{\sqrt{2}}(|0_1, 0_2, +, -\rangle + |0_1, 0_2, -, +\rangle).$$
 (35)

Denoting by  $q_i = \alpha(a_i^{\dagger} + a_i)$  with  $\alpha = 1/2\omega$  the "stretching operator" relative to the *i*th molecule, the quantity

$$C_q \equiv Tr\{\rho(t)q_1q_2\} - Tr\{\rho(t)q_1\}Tr\{\rho(t)q_2\}$$
(36)

expresses, by definition, the quantum covariance of the stretching operators of the two molecules. It is easy to demonstrate that when we start from the statistical mixture given by (31), these observables never correlate, that is  $C_q(t) = 0$  at any t. Vice versa, the stretching covariance  $C_q(t)$  is governed by the temporal law

$$C_q(t) = \alpha^2 \frac{4\epsilon^2}{R^2} \sin^2\left(\frac{R}{2}t\right) \tag{37}$$

when we suppose our system prepared at t = 0 in the linear superposition (35). Thus we find the interesting result that the quantity  $C_q(t)$  is apt to clearly distinguish between the two different initial conditions. In the same sense the covariance between the two dipole moments, defined as

$$C_{\sigma}(t) \equiv Tr\left\{\rho(t)\sigma_x^{(1)}\sigma_x^{(2)}\right\} - Tr\left\{\rho(t)\sigma_x^{(1)}\right\} Tr\left\{\rho(t)\sigma_x^{(2)}\right\}$$
(38)

provides another quantity helping to establish whether the system makes its temporal evolution starting from the statistical mixture (31) or from the pure state (35). It is indeed possible to demonstrate that in the former case we have  $C_{\sigma}(t) = 0$  whereas, in the latter one,

$$C_{\sigma}(t) = |a(t)|^2 + |b(t)|^2.$$
(39)

We wish to emphasize that (38) and (39) provide two examples of dynamical behaviour having genuine undoubt quantum origin. This means that such a behaviour stems directly and manifests the existence and persistence of quantum coherence in the time evolution of the system. It is of relevance to note that, at least in principle, any experiment aimed at resolving in time such a behaviour might be used to monitor the unavoidable progressive washing out of these oscillations traceable back to the influence of the environment. In other words one might hopefully project an experiment providing a direct insight into a process at the heart of quantum mechanics since one might check not only the existence of an initial quantum coherence but also observe its progressive disappearance as example of a decoherence phenomenon transforming the quantum superposition into a statistical mixture.

#### 4 Intra-, inter-molecular competition

The results achieved in the previous section may be put in the following form

$$C_q(t) = \alpha^2 \frac{1}{1 + (\frac{\lambda}{2\epsilon})^2} \sin^2 \left( \sqrt{1 + \left(\frac{\lambda}{2\epsilon}\right)^2} \epsilon t \right)$$
(40)

$$C_{\sigma}(t) = 1 - \frac{1}{1 + \left(\frac{\lambda}{2\epsilon}\right)^2} \sin^2\left(\sqrt{1 + \left(\frac{\lambda}{2\epsilon}\right)^2} \epsilon t\right).$$
(41)

Equations (40) and (41) clearly show that if the system composed by the two interacting defects starts from the pure state (35), its evolution gives rise to the occurrence of quantum correlations periodically varying in a very simple way. The aspect we wish to stress is the dependence of the amplitude of these oscillations on the ratio  $r = \lambda/2\epsilon$  which is a direct measure of the relative strengths between intermolecular and intramolecular energy exchanges. When r = 0, even if the intermolecular energy exchange channel is of course closed, the system indeed develops time dependent quantum correlations between the two elastic degrees of freedom, albeit initially absent. From a physical point of view such a behaviour has a pure quantum origin directly stemming from the initial quantum coherence between the Pauli degrees of freedom. When r increases reaching values of the order of 10, the oscillation amplitudes of both  $C_q(t)$  and  $C_{\sigma}(t)$  tend to vanish. As a consequence the position correlation becomes less and less visible, being initially absent. On the contrary the dipole correlation tends to preserve its initial value. This behaviour clearly evidences the occurrence and the growing up of competition processes between intramolecular energy exchange mechanisms against those ruling non adiabatic intramolecular energy exchanges. Thus, when  $\lambda \gg 2\epsilon$ ,  $C_q(t)$  is practically equal to zero while  $C_{\sigma}(t) \sim 1$  at any time instant t.

### **5** Conclusive remarks

In this paper we have investigated a fully quantistic model describing a simple possible radiationless energy transfer process between two identical dopant centres possessing both electronic and vibrational degrees of freedom. The Hamiltonian term responsible for the excitation energy migration between the two matter subsystems has a dipole-like coupling form.

We have exactly solved the quantum dynamics of our Hamiltonian model in the one-excitation Hilbert subspace investigating the temporal evolution of the correlations get established between the two molecules. To this end we have calculated and analyzed the stretching covariance  $C_q(t)$  and the dipole-dipole covariance  $C_{\sigma}(t)$  between the two molecules. The original results reported in this paper prove to be attractive and intriguing both from a qualitative and quantitative point of view. Studying the evolution of quantum covariances of physically transparent observables relative to the two molecules,  $C_q(t)$  and  $C_{\sigma}(t)$ , we have indeed brought to light the possibility of distinguishing between two different initial conditions, that is a classical statistical mixture or a quantum linear superposition of two appropriate states of the bipartite system. From a quantitative point of view, moreover, it is interesting to point out that the analysis presented in the previous section evidences that measuring  $C_q(t)$  as well as  $C_{\sigma}(t)$  it is possible to have an estimation of the ratio  $r = \lambda/2\epsilon$ . The two covariances are indeed sinusoidal functions whose amplitudes are directly related to r. Thus, measuring the

stretching covariance  $C_q(t)$ , as well as the dipole covariance  $C_{\sigma}(t)$ , we may quantitatively estimate the relative strength of the intermolecular and intramolecular interaction mechanisms characterizing our physical system. We wish finally to stress that, although the presence of environmental effects has been neglected from the very beginning, our analysis has however the merit of having singled out a simple physical scenario, in the reach of the experimentalists, wherein to look for evidences of the transition between quantum and classical behaviour.

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