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BASIC SPIN TENSORS FOR NMR OF MANY-SPIN 1/2 SYSTEM

(Presented by E. Lippmaa)

From algebraic point of view Hamiltonian H and density operator P of a *n*-level nuclear spin system are vectors defined in the n^2 -dimensional euclidian space H of hermitian operators of the system. Lie algebra $\mathfrak{su}(n)$ of the space H forms the mathematical framework which governs NMR phenomena. In particular, adiabatic spin dynamics can be described in terms of representations of Lie group SU(n) and Lie algebra $\mathfrak{su}(n)$ [¹]. In order to describe H and P in terms of sets of real numbers, a suitable basis is to be introduced into H, and commutation relations between basic operators are to be set up. With the help of these commutation relations, any Lie algebraic relationship between hermetian operators can be studied.

Two different kinds of orthogonal sets of basic operators have been introduced in [1] — the A-basis and the I-basis. The A-basis originates from matrix representation of hermitian operators. Quantum dynamics described in terms of A-basis does not depend upon the nature of the system and of the interactions included in H. So, general relationships of quantum dynamics can be formulated in terms of this basis. For a recent demonstration of the use of A-basis in NMR, we refer to [2]. However, in NMR the original form of H and of observables are given in terms of I-basis. Thus knowledge of algebraic properties of both bases is needed. In some situations (weakly coupled spin systems), it is rational to solve the dynamical problem in terms of I-basis only (the direct method).

The *I*-basis is composed of direct products of single spin operators. The basic operators (basic spin tensors) are defined with the reference to a system of space coordinates and they transform like ordinary tensors when this system rotates. The full set of commutation relations for *A*-basis is given in [³]. This paper presents the commutation relations for *I*-basis. The properties of basic spin tensors are reviewed and some demonstrations of the use of these operators in NMR are given.

1. Basic Spin Tensors

1.1. Definition of basic spin tensors. Consider a system of N nuclear spins (labelled by spin numbers $\lambda = 1, 2, ..., N$), each with the spin quantum number $\frac{1}{2}$.

The unitary space $C(\lambda)$ of pure quantum states of a single spin λ is 2-dimensional. Hermitian operators acting in $C(\lambda)$ constitute a 4-dimensional euclidian vector space $H(\lambda)$. This space is spanned by single spin operators

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$$I_{j\lambda}(\lambda) \in \mathbf{H}(\lambda) \quad (j_{\lambda} = 0, x, y, z).$$
 (1)

In Eq. (1) single spin operators with $j_{\lambda} \neq 0$ have the usual meaning, but $I_0(\lambda) = (1/2)E(\lambda)$, where $E(\lambda)$ denotes the unit operator acting in $C(\lambda)$. The scalar product of single spin operators (1) is given by Eq. (2)

$$(I_{j\lambda}(\lambda), I_{k\lambda}(\lambda)) = \operatorname{tr} I_{j\lambda}(\lambda) I_{k\lambda}(\lambda) = (1/2) \,\delta_{j\lambda,k\lambda}, \tag{2}$$

where $\delta_{j\lambda,k\lambda}$ is the Kronecker symbol. Thus, the basis (1) is orthogonal, and the basic operators possess a common norm.

Single spin operators (1) can be considered as generators of a representation of the Lie ring u(2) in space $H(\lambda)$, provided the Lie product is defined as follows:

 $-i[I_{i\lambda}(\lambda), I_{j\lambda}(\lambda)] = I_{k\lambda}(\lambda), \qquad (3)$

with $i_{\lambda}j_{\lambda}k_{\lambda}=xyz$ and cyclic transpositions [1]. The three single spin operators with $j_{\lambda}\neq 0$ are generators of the Lie algebra su(2), which is isomorphic with ordinary vector algebra. This isomorphism is established by relating each of the three single spin operators to the corresponding unit vectors of a cartesian space coordinate system.

According to principles of quantum mechanics [4], the space C of pure quantum states of N spin system is the direct product of single spin spaces:

$$\mathbf{C} = \mathbf{C}(1) \times \mathbf{C}(2) \times \dots \mathbf{C}(N), \tag{4}$$

$$\dim \mathbf{C} = n = 2^N. \tag{5}$$

The n^2 -dimensional euclidian space **H** of all hermitian operators acting in **C** is a similar direct product:

$$\mathbf{H} = \mathbf{H}(1) \times \mathbf{H}(2) \times \dots \mathbf{H}(N). \tag{6}$$

Eq. (6) indicates that a possible orthogonal basis of \mathbf{H} can be set up performing direct products of single spin operators (1):

$$I_{j1j2...,jN} = I_{j1}(1) \times I_{j2}(2) \times ... I_{jN}(N), \qquad (7)$$

where

$$j_1, j_2, \ldots, j_N = 0, x, y, z.$$
 (8)

The basis of H composed of basic spin tensors (7) is called *I*-basis. It is related to the space coordinate system as established above.

The set of single spin coordinate labels (8), ordered by rising spin numbers, specifies a particular basic spin tensor. The number of labels with $j_{\lambda} \neq 0$ in this set equals to the tensorial rank of this basic operator (see Sec. 1.2).

In order to diminish the number of coordinate labels, we also adopt a simplified notation of basic spin tensors (7). The following example clarifies the principle of this notation

$$I_{0z0x} = I_{zx}(2,4). \tag{9}$$

Therefore, zero coordinate labels are dropped in the simplified version of notation, but spin numbers of not zero coordinate labels are added in brackets.

Eq. (10) shows that basic spin tensors are orthogonal and possess a common norm

$$(I_{j1j2\dots jN}, I_{j1j2\dots jN}) = (1/2)^N.$$
 (10)

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Existence of a common norm of basic spin tensors (7) is useful in writing down relationships between basic tensors of different rank. This is the reason we use basic spin tensors (7) throughout this work. However, it should be noted that another kind of spin tensors which differ from basic spin tensors (7) by a scaling factor, are usually used in Hamiltonians and in other physical quantities. So, instead of the basic spin tensor (9) the spin tensor

$$I_{zx}^{(2)}(2,4) = E(1) \times I_z(2) \times E(3) \times I_x(4)$$
(11)

is used in standard texts.

1.2. Transformational properties. If the standard basis of $C(\lambda)$ is defined by Eq. (12)

$$I_{z}(\lambda) | m_{\lambda} \rangle = m_{\lambda} | m_{\lambda} \rangle, \quad m_{\lambda} = \pm 1/2$$
(12)

and by some additional phase conventions given in [5], the standard basis of C is given by the direct product

$$|m_1\rangle|m_2\rangle\dots|m_N\rangle. \tag{13}$$

Unitary operator $D(\lambda) \in SU(2)$ transforms the standard basis $|m_{\lambda}\rangle \in C(\lambda)$ to another orthonormal basis $D(\lambda) |m_{\lambda}\rangle \in C(\lambda)$, whereas the single spin operators rotate according to Eq. (14) [4,1]

$$\Re(\lambda) I_{j\lambda}(\lambda) = D(\lambda) I_{j\lambda}(\lambda) D(\lambda)^{-1}.$$
(14)

Note that the transformed operators (14) share its eigenvalues but possess another space direction than the original operators. In order to give $I_z(\lambda)$ the space direction with spherical coordinates ψ_{λ} , ϑ_{λ} , it is sufficient to take

$$D(\lambda) = \exp\left(-\mathrm{i}\psi_{\lambda}I_{z}(\lambda)\right)\exp\left(-\mathrm{i}\vartheta_{\lambda}I_{y}(\lambda)\right).$$
(15)

The orthogonal superoperator $\Re(\lambda)$ belongs to the adjoint representation [6] of $D(\lambda)$, symbolically:

$$D(\lambda) \to \Re(\lambda)$$
. (16)

The notation

$$D = D(1) \times D(2) \times \dots D(N) \to \Re = \Re(1) \times \Re(2) \times \dots \Re(N)$$
(17)

means: unitary transformation $D \in SU(2)_N$ of space C induces a corresponding orthogonal transformation \Re of the space H. By these transformations different single spin spaces transform independently. In particular, if $D(\lambda)$ is given by Eq. (15), the N pairs of angles ψ_{λ} , ϑ_{λ} describe N different space directions of single spins.

Since the transformation (14) does not change single spin operators $I_0(\lambda)$, the space **H** decomposes into subspaces which are irreducible with respect to transformation \Re of Eq. (17):

$$\mathbf{H} = \mathbf{H}^{(0)} \div \mathbf{H}^{(1)} \div \mathbf{H}^{(2)} \div \dots \mathbf{H}^{(N)}, \tag{18}$$

where, for example,

$$\mathbf{H}^{(1)} = \sum_{\lambda} \mathbf{H}^{(1)}(\lambda), \tag{19}$$

$$\mathbf{H}^{(2)} = \sum_{\lambda < \mu} \sum_{\mathbf{H}^{(2)}} \mathbf{H}^{(2)}(\lambda, \mu).$$
(20)

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In Eq. Eq. (18)—(20) $\mathbf{H}^{(0)}$ is spanned by $I_{00\dots0}$, the 3-dimensional subspace $\mathbf{H}^{(1)}(\lambda)$ is spanned by basic spin tensors $I_{j\lambda}(\lambda)$ $(j_{\lambda}\neq 0)$ and transforms as ordinary vector space, the 9-dimensional subspace $\mathbf{H}^{(2)}(\lambda, \mu)$ is spanned by $I_{j\lambda j\mu}(\lambda, \mu)$ $(j_{\lambda}, j_{\mu}\neq 0)$ and transforms as second rank tensor space. So, the 3N-dimensional subspace $\mathbf{H}^{(1)}$ is composed of tensorial operators of the first rank, $\mathbf{H}^{(2)}$ is composed of tensorial operators of second rank. The other subspaces of Eq. (18) belong to higher-rank tensorial operators.

A note about terminology. According to [7] a hermitian operator $H \in \mathbf{H}^{(2)}(\lambda, \mu)$ is a tensor of second rank with respect to the set of its components, but the set of nine basic operators $I_{j\lambda j\mu}(\lambda, \mu)$ also form a tensorial set of second rank. In this paper only the former understanding of the term tensorial operator is adopted.

Special transformations (17), (15) with the property $\psi_{\lambda} = \psi_{\mu}, \vartheta_{\lambda} = \vartheta_{\mu}$ ($\lambda, \mu = 1, 2, ..., N$) describe the effect of rotating space coordinate system. The total spin operators

$$J_{k} = 2^{N-1} \sum_{\lambda} I_{k}(\lambda) \qquad (k = x, y, z)$$

$$(21)$$

belong to the Lie ring su(2) of infinitesimal operators [6] of this group of special transformations. The operators of this ring compose a 3dimensional subspace $H_z \subset H^{(1)}$ spanned by total spin operators (21). With respect to operators of the Lie ring, all subspaces listed in Eq. Eq. (18)—(20) are invariant, but (in general) not irreducible. Reduction is achieved with the help of irreducible basic spin tensors [7] built up on the basis of basic spin tensors (7).

1.3. Applications in NMR. Decomposition of hermitian operators in terms of *I*-basis selects components of different tensorial rank and of different collections of spin numbers (of different spin subsystems). So, decomposition of the density operator is given by

$$P = I_{0\dots0} + P^{(1)} + P^{(2)} + \dots P^{(N)} \in \mathbf{H},$$
(22)

where, for example,

$$P^{(1)} = \sum_{\lambda} P^{(1)}(\lambda) = \sum_{\lambda} \sum_{j} \pi_{j}(\lambda) I_{j}(\lambda) \in \mathbf{H}^{(1)}.$$
(23)

The component $P^{(1)}(\lambda)$ is a first rank tensor and belongs to the single spin λ , whereas $P^{(2)}(\lambda, \mu)$ is a second rank tensor of the spin pair λ, μ . In case of a homonuclear spin 1/2, system observable NMR signals are due to the time-dependent component $P^{(1)}(t) \in \mathbf{H}_z$ only. Higher-rank components of P determine the microscopic inhomogeneities of the magnetic field generated by individual nuclear magnetic dipole moments.

Usually, NMR spectra are analyzed in terms of (static) Hamiltonians

$$H_0 = \sum_{\lambda} H^{(1)}(\lambda) + \sum_{\lambda < \mu} H^{(2)}(\lambda, \mu) \in \mathbf{H}^{(1)} + \mathbf{H}^{(2)}.$$
(24)

In Eq. (24) $H^{(1)}(\lambda)$ is the (anisotropic) chemical shift interaction term for spin λ , but $H^{(2)}(\lambda, \mu)$ include spin-spin interactions of the pair λ, μ . 'Higher-rank terms could describe indirect spin-spin couplings, but are found to be too small. Only symmetric tensors are usually included in $H^{(2)}(\lambda, \mu)$. So, the indirect scalar coupling is given by Eq. (25)

$$H_J^{(2)}(\lambda,\mu) = 2^{N-2} J_{\lambda\mu} J(\lambda,\mu), \qquad (25)$$

where $J_{\lambda\mu}$ denotes the conventional spin-spin coupling constant and

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$$J(\lambda,\mu) = \sum_{j} I_{jj}(\lambda,\mu).$$
(26)

Another spin-spin coupling term, needed for NMR of solids and anisotropic liquids, is the dipole-dipole interaction:

$$H_D^{(2)}(\lambda,\mu) = -2^{N-2} D_{\lambda\mu} (3I_{nn}(\lambda,\mu) - J(\lambda,\mu))$$
(27)

with

$$D_{\lambda\mu} = \gamma_{\lambda} \gamma_{\mu} \hbar / R_{\lambda\mu}^{3}$$
(28)

the conventional dipole-dipole coupling constant for a pair of nucleus

 λ, μ the space locations of which are bound by ordinary vector $R_{\lambda\mu} = R_{\lambda\mu}n$.

For homonuclear systems the subspace H_z has a special meaning in that:

- 1) interaction with time-dependent external magnetic field is described by $H_E(t) \in \mathbf{H}_z$;
- 2) observable NMR signals are due to

$$\langle J_k(t) \rangle = (P(t), J_k), \quad J_k \in \mathbf{H}_z;$$
⁽²⁹⁾

3) the usual initial state P(0), the equilibrium state P^0 , is (in good approximation) given by $P^0 \in \mathbf{H}_z$.

So, the manifold of possible P(t) and $\langle J_k(t) \rangle$ is much more limited as it would be in case of a general $H_E(t) \in \mathbf{H}$ and $P(0) \in \mathbf{H}$. Rational incorporation of these limits into the theory of NMR is up to the day an unresolved problem.

1.4. Commutative subspaces of **H**. Hermitian operators with common set of eigenvectors form a *n*-dimensional commutative subspace of **H**. The commutative subspace \mathbf{H}_A related to the standard basis (13) is called standard. One associates the standard *A*-basis of **H** [¹] with the basis (13) and with the subspace.

The *I*-basis of \mathbf{H}_A is composed of basic spin tensors $I_{0...0}$; $I_z(\lambda)$ ($\lambda = 1, 2, ..., N$); $I_{zz}(\lambda, \mu)$ ($\lambda, \mu = 1, 2, ..., N$); ... $I_{zz...}$ (1, 2, ..., N). So, basic spin tensors of all tensorial rank and of all spin subsystems are presented in this list.

Hermitian operators with all possible eigenvalue spectra are represented in a commutative subspace like H_A . If

$$H[m_1m_2\ldots] = \omega_{m_1m_2\ldots}/m_1m_2\ldots\rangle \tag{30}$$

but, on the other hand,

$$H = -2^{N-1} \sum_{\lambda} \omega_{\lambda} I_{z}(\lambda) + 2^{N-2} \sum_{\lambda < \mu} \omega_{\lambda \mu} I_{zz}(\lambda, \mu) + \dots, \qquad (31)$$

then

$$\omega_{m1m2...} = -\sum_{\lambda} m_{\lambda} \omega_{\lambda} + \sum_{\lambda < \mu} m_{\lambda} m_{\mu} \omega_{\lambda \mu} + \dots \qquad (32)$$

Eq. (32) presents transformation of $H \in \mathbf{H}_A$ from *I*-basis to the standard *A*-basis.

If $H \in \mathbf{H}_A$, then

$$H_{U} = UHU^{-1} \in \mathbf{H}_{U}, \quad U \in SU(n), \tag{33}$$

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where \mathbf{H}_U is a commutative subspace associated with the transformed basis $U|m_1m_2...\rangle \in \mathbf{C}$. The eigenvalue spectrum of H_U coincides with that of H. Both operators, H_U and H, are said to be equivalent. If in Eq. (33) U runs through $\mathbf{SU}(n)$, one gets the class of equivalent hermitian operators. To each $H \in \mathbf{H}_A$ corresponds a class of equivalent operators specified by the eigenvalue spectrum.

2. Commutation relations

2.1. Commutation relations between basic spin tensors. In order to derive these relations, expand the commutator between two arbitrary basic spin tensors in terms of *I*-basis. Then use of scalar products between the commutator and basic spin tensors allows to calculate expansion coefficients. The final result of such calculations is expressed by Eq. (34):

$$-i[I_{i1i2...iN}, I_{j1j2...jN}] = (1/2)^{N-1} \gamma_{k1k2...kN} I_{k1k2...kN}, \qquad (34)$$

where

$$\gamma_{h1h2...hN} = 0, \pm 1.$$
 (35)

In Eq. (34) the sets of coordinate labels $i_1i_2...i_N$ and $j_1j_2...j_N$ specify the set $k_1k_2...k_N$ and the value of the expansion coefficient allowed by Eq. (35). Below, the rules of this specification are clarified.

Let us associate with the commutation relation (34) the following table of coordinate labels

Each column of this table is composed of coordinate labels of a single spin λ .

If $i_{\lambda} \neq j_{\lambda} \neq 0$, then k_{λ} must be: $k_{\lambda} \neq 0$, i_{λ} , j_{λ} . In this case i_{λ} , j_{λ} , $k_{\lambda} = x, y, z$. Such sets of coordinate labels are called proper (single spin) coordinate labels. The order $i_{\lambda}j_{\lambda}k_{\lambda}$ of proper coordinate labels in a column (listed from top to bottom) is essential. In case of $i_{\lambda}j_{\lambda}k_{\lambda} = xyz$ or cyclic transposition, the order is called positive. Otherwise the order is negative.

Three kinds of nonproper sets of coordinate labels listed in Eq. (37), are allowed for columns of the table (36)

The allowed value of the expansion coefficient (35) depends only upon the number of columns with proper coordinate labels as well as upon the sign of these sets. If p denotes the number of proper coordinate labels with positive order, and q — the number of those with negative order, then

$$\gamma_{k1k2\dots kN} = (1/2) \, \mathrm{i}^{2r} [(-1)^q - (-1)^p], \tag{38}$$

where

$$r = (1/2) (p+q-1).$$
 (39)

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Therefore, the value zero (commuting basic spin tensors!) appears if, and only if, both p and q are even numbers (including zero) or both are odd. Otherwise, the sign of the expansion coefficient (35) depends upon the parity of the quantity (39) too.

Transposition of first two rows of the table (36) changes the sign of the expansion coefficient (35). Thus, it is always possible to order the basic spin tensors of the commutator in such a way that the expansion coefficient takes the value ± 1 . For p - q even-odd we get this in case when r is odd. For p - q odd-even this will be in case when r is even. **2.2.** Coupled sets of basic spin tensors. By cyclic transposition of basic spin tensors the commutation relation (34) remains valid, whereas the value of the expansion coefficient (35) does not change. Three basic spin tensors in Eq. (34) are said to form a coupled set. The 3-dimensional subspace of hermitian operators spanned by basic spin tensors of a coupled set possess a closed Lie algebra.

There are commutative and noncommutative coupled sets of basic spin tensors.

In case the table (36) of a commutative set consists only of columns with nonproper coordinate labels (37), the corresponding 3-dimensional subspace is incorporated into a commutative subspace described in Sec. 1.4. This is not valid in case of a general commutative set.

The basic spin tensors of a noncommutative coupled set are generators of the adjoint representation of the Lie algebra su(2) [^{6,1}]. In this case the usual form of commutation relations (3) hold between operators

$$I'_{i1i2...iN} = 2^{N-1} I_{i1i2...iN}. \tag{40}$$

The subspace $\mathbf{H}^{(4)}(\lambda)$ is a particular case of such a Lie algebra $\mathbf{su}(2)$. The commutation relation (34) remains valid if basic spin tensors

experience the transformation (33). Using different unitary operators U, one generates new representations of su(2) in this way.

2.3. Dynamical coupling of tensorial components of density operator. If relaxation is neglected, time-evolution of the density operator (22) is governed by Liouville equation (41)

$$\frac{dP}{dt} = -i[H, P]$$
(41)

in which the Hamiltonian H is given by a tensorial decomposition shown in Eq. (24). Below, the commutation relations given in Sec. 2.1 and Sec. 2.2 are utilized in order to derive a coupled set of equations of motion for tensorial components of the density operator (22).

If l_1 , l_2 , l_3 denote tensorial ranks of basic spin tensors in Eq. (34) (listed from left to right), we express this symbolically as follows

 $[l_1, l_2] = l_3. \tag{42}$

If $l_1 = 1, 2$, only the following relations will hold for noncommutative sets of basic spin tensors:

$$[1, l_2] = l_2,$$
 (43)

$$[2, l_2] = l_2 - 1, \tag{44}$$

$$[2, l_2] = l_2 + 1. \tag{45}$$

In order to demonstrate the relations (43) - (45), we take and write

down a typical table (36) for each of these relations. So, Eq. (43) refers to tables as the following

$$\frac{j_1 j_2 j_3}{k_1 j_2 j_3} - (46)$$

Eq. (44) is demonstrated by the table (47), Eq. (45) — by the table (48)

$$\frac{i_1 i_2 0}{j_1 i_2 j_3} \qquad (47) \quad \frac{j_1 0 j_3}{k_1 0 j_3} \qquad (48)$$

Replacing the tensorial decompositions (22) - (24) in Eq. (41) and utilizing the relations given by Eq. Eq. (43) - (48), we get the following equations for density operator components of first and second rank:

$$\frac{dP^{(1)}(\lambda)}{dt} = -i[H^{(1)}(\lambda), P^{(1)}(\lambda)] - i\sum_{\mu \neq \lambda} [H^{(2)}(\lambda, \mu), P^{(2)}(\lambda, \mu)], \quad (49)$$

$$\frac{dP^{(2)}(\lambda,\mu)}{dt} = -i[(H^{(1)}(\lambda) + H^{(1)}(\mu)), P^{(2)}(\lambda,\mu)] - -i[H^{(2)}(\lambda,\mu), (P^{(1)}(\lambda) + P^{(1)}(\mu))] - -i\sum_{\nu} [(H^{(2)}(\lambda,\nu) + H^{(2)}(\mu,\nu)), P^{(3)}(\lambda,\mu,\nu)].$$
(50)

Eq. (49) shows how spin-spin interaction couples the motions of single spin components $P^{(1)}(\lambda)$ and $P^{(1)}(\mu)$ through $P^{(2)}(\lambda, \mu)$. If $P^{(2)}(\lambda, \mu)$ vanish, one observes a spin decoupled NMR spectrum. If $P^{(3)}(\lambda, \mu, \nu)$ vanish, Eq. Eq. (49), (50) describe the dynamics SU(4) of a spin pair λ, μ . However, in general $P^{(3)}(\lambda, \mu, \nu)$ couples the motions of different spin pairs $\lambda \mu$, $\mu \nu$, $\lambda \nu$. So, the hierarchy of couplings is built up.

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BAASSPINNTENSORID MITME ¹/2-SPINNIGA SÜSTEEMI TUUMARESONANTSIS

Artiklis on esitatud baasspinntensorite kommutatsiooniseosed ning kirjeldatud nende omadusi ja rakendusi tuumaresonantsi teoorias.

В. СИНИВЕЭ

БАЗИСНЫЕ СПИНОВЫЕ ТЕНЗОРЫ ДЛЯ ЯМР СИСТЕМ МНОГИХ СПИНОВ 1/2

В работе приведены коммутационные соотношения для базисных спиновых тензоров, описаны их основные свойства и применения в теории ЯМР.

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