

# Activity Coefficients of Electrons and Holes in Semiconductors with Nonuniform Composition

## I. Nondegenerate

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### ABSTRACT

A simple formalism for the calculation of the equilibrium activity coefficients of electrons and holes in a nondegenerate semiconductor with nonuniform composition is presented. These activity coefficients are functions of bandgap, electron affinity, and the density of states which vary with position. The calculation of carrier activity coefficients requires the selection of chemical potential and electrostatic potential references. The choice of these reference states is addressed. The relations between purely thermodynamic quantities and parameters of the band theory are also presented. It is shown that the intrinsic level is a purely thermodynamic property of the intrinsic bulk semiconductor. The approach presented here allows convenient treatment of nonuniform semiconductors in a manner that is both thermodynamically consistent and consistent with the Poisson-Boltzmann equation for the electrostatic potential.

The activity coefficient is a useful quantity for describing a system (e.g., a liquid mixture, semiconductor, or other system) behavior with deviations from its ideality. It is usually introduced through a relation between the chemical potential and the concentration and may take different forms depending on the way in which concentrations for a given system are expressed, i.e., as molarity, molality, or mole fraction. For instance, the chemical potential ( $\mu_i^\alpha$ ) of a species  $i$  in phase  $\alpha$  may be expressed in the form [1]

$$\mu_i^\alpha = \mu_i^{*\alpha} + RT \ln a_i^\alpha = \mu_i^{*\alpha} + RT \ln \gamma_i^\alpha c_i^\alpha \quad [1]$$

where  $\mu_i^{*\alpha}$  is the reference state chemical potential of the species  $i$  in the phase  $\alpha$ , and is a function only of temperature, pressure, and choice of reference state. The activity coefficient of the species  $i$  in phase  $\alpha$  is  $\gamma_i^\alpha$ , and is a function of temperature, pressure, and concentration. The molar concentration is  $c_i^\alpha$ . The activity of the species  $i$  in phase  $\alpha$  is  $a_i^\alpha$ , and is the product of concentration and activity coefficient.  $R$  is the gas constant, and  $T$  is the temperature in K. Equation [1] can be viewed as the defining equation for the activity coefficient ( $\gamma_i^\alpha$ ) of species  $i$  in phase  $\alpha$ . Note that the chemical potential is generally split into two terms: a composition-independent term ( $\mu_i^{*\alpha}$ ) and a term which is composition-dependent and accounts for the difference between the actual chemical potential of interest and the reference state chemical potential. The concentration defined in Eq. [1] is consistent with the carrier densities ( $n$  or  $p$ ) used in the energy band model for a semiconductor when Eq. [1] is divided by  $N_A$  (Avogadro's number) and  $R/N_A$  is replaced by  $k$  (Boltzmann constant).

Several different techniques have been employed to investigate the activity coefficients for electrons and holes in semiconductors with a uniform energy band structure (2-8). However, a systematic study (including the choice of the reference states) of the activity coefficients of electrons and holes in nonuniform semiconductors does not seem to exist in the literature. For a nondegenerate electron or hole in a uniform semiconductor, the activity coefficients are commonly taken as unity. When the energy bandgap for a solid is nonuniform, such as in a graded bandgap structure or in a doped semiconductor with a nonconstant doping profile under conditions where bandgap narrowing occurs, the working equations for the currents and for the carrier densities contain extra terms over the conventional results (9). It will be shown later that this extra term can be related to the carrier activity coefficients. The activity coefficients no longer equal unity in this case, even though the semiconductor is nondegenerate.

Activity coefficients derived here are obtained by straightforward substitution of the carrier densities into the irreversible thermodynamic definition of the electrochemical potential. These coefficients are functions of electron affinity, bandgap, and the density of states that

vary with position. The results presented here are both thermodynamically consistent and consistent with the Poisson-Boltzmann equation for the electrostatic potential. Thermal equilibrium will be assumed throughout the discussion, and effects of pressure will not be considered.

### Energy Bands in Nonuniform Semiconductors

The energy band diagram (shown with the underlying assumption of the following analysis) of a semiconductor with nonuniform composition is depicted in Fig. 1. The validity of this energy band model has been discussed by Marshak and van Vliet (9). From Fig. 1, we have

$$E_L(x) = E_0 - qV(x) \quad [2]$$

$$\chi(x) = E_L(x) - E_c(x) = E_0 - qV(x) - E_c(x) \quad [3]$$

and

$$E_G(x) = E_c(x) - E_v(x) \quad [4]$$

where  $E_0$  is the field-free vacuum level,  $E_L$  is the local vacuum level,  $V$  is the electrostatic potential,  $\chi$  is the electron affinity,  $E_G$  is the bandgap,  $E_c$  is the bottom of the conduction band,  $E_v$  is the top of the valence band, and  $q$  is the magnitude of the electronic charge.

If we assume Boltzmann statistics and write the equilibrium carrier densities as

$$n_o(x) = N_c(x)e^{(E_F - E_c)/kT} = n_i(x)e^{(E_F - E_i)/kT} \quad [5]$$

and

$$p_o(x) = N_v(x)e^{(E_v - E_F)/kT} = n_i(x)e^{(E_i - E_F)/kT} \quad [6]$$

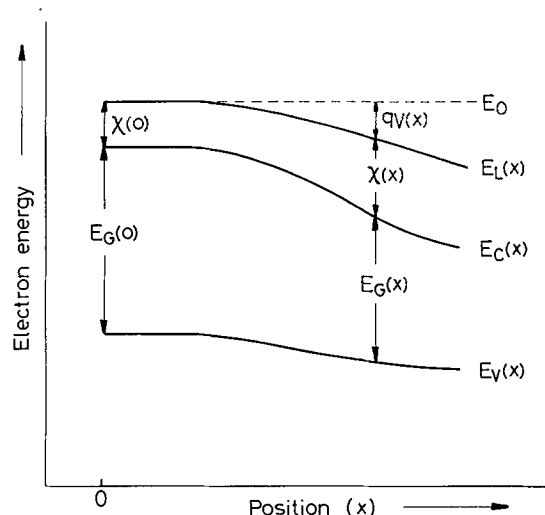


Fig. 1. Energy bands for inhomogeneous material

where

$$n_i(x) = [N_v(x)N_c(x)]^{1/2} e^{-E_G(x)/2kT} \quad [7]$$

is the position-dependent intrinsic carrier concentration,  $N_c(x)$  and  $N_v(x)$  are effective densities of states in the conduction band and in the valence band, respectively. Using Eq. [3]-[7], we obtain the intrinsic energy level

$$E_i(x) = E_o - \chi(x) - 1/2 E_G(x) + \frac{kT}{2} \ln \left[ \frac{N_v(x)}{N_c(x)} \right] - qV(x) \quad [8]$$

It is obvious from Eq. [8] that  $E_i(x)$  is not, in general, parallel to  $V(x)$  as it is in a uniform semiconductor. It will be shown later that the intrinsic level  $E_i$  is a purely thermodynamic property of the intrinsic bulk semiconductors. Consequently, for a heterojunction, one cannot identify  $(-E_i/q)$  with electrostatic potential as it is a common practice for homojunctions (10).

Algebraic manipulation of Eq. [5]-[8], with the neutrality condition at  $x = 0$  and the reference electrostatic potential  $V(0) = 0$ , gives the following carrier densities under thermal equilibrium (11)

$$n_o(x) = n_o(0) \exp \left\{ \frac{qV(x) + A\Delta E_g}{kT} \right\} \quad [9]$$

and

$$p_o(x) = p_o(0) \exp \left\{ \frac{-qV(x) + (1 - A)\Delta E_g}{kT} \right\} \quad [10]$$

where

$$\Delta E_g = -[E_G(x) - E_G(0)] + kT \ln \left[ \frac{N_v(x)N_c(x)}{N_v(0)N_c(0)} \right] \quad [11]$$

and

$$A = \frac{[\chi(x) - \chi(0)] + kT \ln \left[ \frac{N_c(x)}{N_c(0)} \right]}{\Delta E_g} \quad [12]$$

As shown in Fig. 1, we choose  $x = 0$  as the reference position for the electrostatic potential at which the material is uniform. So, both  $\Delta E_g$  and  $A$  are positive quantities.  $\Delta E_g$  is called the effective bandgap shrinkage, and  $A$ , called the effective asymmetry factor (12), measures the change in the conduction band edge ( $\Delta\chi$ ) and density of states,  $0 \leq A \leq 1$ . It is noted that a different choice of the reference state for the electrostatic potential leads to different expressions for carrier densities. For example, if we choose  $qV(0) = -kT \ln(-D + \sqrt{D^2 + 1})$  ( $D \equiv (N_D^+(0) - N_A^-(0))/2n_i(0)$ ,  $N_D^+(0) - N_A^-(0) \equiv$  net doping density at  $x = 0$ ), then the carrier densities become (11)

$$n_o(x) = n_i(0) \exp \left\{ \frac{qV(x) + A\Delta E_g}{kT} \right\} \quad [13]$$

and

$$p_o(x) = n_i(0) \exp \left\{ \frac{-qV(x) + (1 - A)\Delta E_g}{kT} \right\} \quad [14]$$

where  $\Delta E_g$  and  $A$  are the same expressions defined by Eq. [11] and [12], respectively. Note that the choice of these references is quite arbitrary. The appropriate choice, however, of these reference states can lead to simple working equations for the problems (11). For  $\Delta E_g = 0$ , Eq. [13] and [14] reduce to the conventional (uniform) results. The extra terms in addition to the conventional results are due to the nonideal behavior of the carriers and will be related to the activity coefficients of the carriers as shown below.

### Reference States

The electrochemical potential ( $\bar{\mu}_i$ ), or Gibbs energy per charged particle, of a charged species  $i$  in a phase is de-

defined as the sum of its chemical potential and its electric potential energy (1, 13)

$$\bar{\mu}_i = \mu_i + Z_i qV \quad [15]$$

where  $Z_i$  is the elemental charge of species  $i$ , and  $Z_i q$  is the electric charge carried by a particle of  $i$ . The potential  $V$  discussed here is the electrostatic potential which is obtained through integration of Poisson's equation ( $\nabla^2 V = -\rho/\epsilon$ ). In thermodynamic equilibrium, it is the electrochemical potential that must be constant throughout (equilibrium condition), and this is another name for the Fermi level ( $E_F$ ) used in the energy band model.

For electrons ( $i = n$ ),  $Z_n = -1$ , substituting Eq. [1] into Eq. [15] and equating  $\bar{\mu}_n = E_F$ , we get

$$E_F = \bar{\mu}_n = \mu_n^* + kT \ln(n_o \gamma_n) - qV \quad [16]$$

It is clear that we have two reference states in Eq. [16], i.e.,  $\mu_n^*$  and  $qV(0)$ . This implies that we have two unknowns and only one equation. To get consistent results, these two reference states cannot be chosen independently. When one is specified, the other must be fixed. In the case of  $qV(0) = 0$ , substituting Eq. [5] ( $x = 0$ ) into Eq. [16] ( $x = 0$ ) followed by some algebraic manipulation results in the following expression for the reference chemical potential in equilibrium

$$\mu_n^* = E_F - kT \ln[n_o(0)] = E_i(0) - kT \ln[n_i(0)] \quad [17]$$

Here we use the fact that  $\gamma_{n_o}(0) = 1$ . Furthermore, by using Eq. [3], [7], and [8], we obtain

$$\mu_n^* = E_o - \chi(0) - kT \ln(N_c(0)) = E_c(0) - kT \ln[N_c(0)] \quad [18]$$

In a similar manner, for  $qV(0) = -kT \ln(-D + \sqrt{D^2 + 1})$ , using Eq. [5], [13], and [16] at  $x = 0$  gives

$$\mu_n^* = E_i(0) - kT \ln[n_i(0)] + qV(0) \quad [19]$$

Again, using Eq. [3], [7], and [8] at  $x = 0$ , we have

$$\mu_n^* = E_c(0) + qV(0) - kT \ln[N_c(0)] \quad [20]$$

which resembles the expression given by Harvey (3). Equation [20] is also similar to the result of Bonham and Orazem (14) if the secondary reference state  $\mu^*$  ( $\mu^* \equiv \mu_n - E_F$ ) as defined by Ref. (14) equals  $qV(0)$ . It is apparent from the above equations that  $\mu_n^*$  depends on temperature and on the choice of reference electrostatic potential. For holes ( $i = p$ ),  $Z_p = +1$ , substituting Eq. [1] into Eq. [15] and equating  $\bar{\mu}_p = -E_F$  gives

$$-E_F = \bar{\mu}_p = \mu_p^* + kT \ln(p_o \gamma_p) + qV \quad [21]$$

A similar reference chemical potential can be obtained for holes as

$$\mu_p^* = -E_i(0) - kT \ln[n_i(0)] \quad [22]$$

or

$$\mu_p^* = -E_v(0) - kT \ln[N_v(0)] \quad [23]$$

for  $qV(0) = 0$ , and

$$\mu_p^* = -E_i(0) - qV(0) - kT \ln[n_i(0)] \quad [24]$$

or

$$\mu_p^* = -E_v(0) - qV(0) - kT \ln[N_v(0)] \quad [25]$$

for  $qV(0) = -kT \ln(-D + \sqrt{D^2 + 1})$ .

Note that negative Fermi energy is used in Eq. [21]. This is because a hole has a charge opposite that of an electron, and the energy diagram (including  $E_F$ ) shown in Fig. 1 indicates electron energies. Again,  $\mu_p^*$  depends on the temperature and on the choice of the reference electrostatic potential. Equation [25] is similar to the result given by Harvey (3).

### Activity Coefficients for Electrons and Holes

The activity coefficients defined above can be obtained by straightforward substitution of the carrier densities into the definition of the electrochemical potential. For  $V(0) = 0$ , substituting Eq. [9] into Eq. [16] gives

$$kT \ln \gamma_{n_0} = -A\Delta E_g - \mu_n^* + kT \ln [n_0(0)] \quad [26]$$

We may eliminate  $\mu_n^*$  by substituting Eq. [17] into Eq. [26]. After some algebra, we find the activity coefficient of conduction electrons as

$$\gamma_{n_0} = \exp\left(\frac{-A\Delta E_g}{kT}\right) \quad [27]$$

It can be seen if  $\Delta E_g = 0$ , then the result reduces to the ideal case ( $\gamma_{n_0} = 1$ ). In a similar manner, the activity coefficient can be obtained for the holes as

$$\gamma_{p_0} = \exp\left[\frac{-(1-A)\Delta E_g}{kT}\right] \quad [28]$$

It is clear that if  $\Delta E_g \neq 0$ , the activity coefficients no longer equal unity as in the case of a homogeneously nondegenerate semiconductor. Note that if  $A = 0.5$  (i.e., symmetric band narrowing), then Eq. [27] and [28] yield  $\gamma_{n_0} = \gamma_{p_0}$ . Similar results can be obtained for  $qV(0) = -kT \ln(-D + \sqrt{D^2 + 1})$ . The results presented here are written in a simple form in which the nonideal effects associated with the nonuniform band structure are described by two quantities, the effective bandgap shrinkage,  $\Delta E_g$ , and the effective asymmetry factor,  $A$ .

The activity coefficients given by Eq. [27] and [28] can be related to their carrier concentrations by [13] and [14] as

$$n_0(x) = \frac{n_i(0)}{\gamma_{n_0}(x)} \exp\left[\frac{qV(x)}{kT}\right] \quad [29]$$

and

$$p_0(x) = \frac{n_i(0)}{\gamma_{p_0}(x)} \exp\left[\frac{-qV(x)}{kT}\right] \quad [30]$$

We note that the usual expressions of  $n_0(x)$  and  $p_0(x)$  (10, 15),  $\gamma_{n_0}(x) = \gamma_{p_0}(x) = 1$ , yield an underestimation of the carrier densities. It is also noted that the pre-exponential factor in Eq. [29] and [30] has the same value only if the bandgap narrowing is symmetric (i.e.,  $A = 1/2$  or  $\gamma_{n_0} = \gamma_{p_0}$ ), which rarely occurs (12).

### Equilibrium $p_0 n_0$ Product and Equilibrium Constant

The equilibrium product of carrier densities  $p_0 n_0$  is of interest because it can be inferred from electrical measurements (16) and can be related to the band structure theoretically. From Eq. [5], [6], [9], and [10], or Eq. [13] and [14], we obtain the equilibrium  $p_0 n_0$  product as

$$p_0(x)n_0(x) = n_i^2(x) = n_i^2(0)e^{\Delta E_g(x)/kT} \quad [31]$$

For a lightly doped semiconductor ( $\Delta E_g = 0$ ), the equilibrium product is a constant at a given temperature, equal to the square of the intrinsic carrier concentration  $n_i(0)$ , but for high doping densities ( $\Delta E_g \neq 0$ ), the product becomes doping dependent. Note that Eq. [31] is obtained from the band model of a semiconductor. It is shown below that Eq. [31] is consistent with the result derived from chemical thermodynamics.

As discussed above, the negative of the electrochemical potential of the hole at thermal equilibrium is equal to the electrochemical potential of the electron

$$\tilde{\mu}_n = E_F = -\tilde{\mu}_p \quad [32]$$

By using Eq. [15], Eq. [32] may be rewritten in the form

$$0 = \tilde{\mu}_n + \tilde{\mu}_p = \mu_n + \mu_p \quad [33]$$

Note that Eq. [33], expressing the Gibbs condition for thermal equilibrium between electrons and holes, implies the charged particle equilibrium chemical equation

$$0 \rightleftharpoons e^+(p) + e^-(n) \quad [34]$$

The conservation of charge condition is met. There are no reactants, only products. The validity of this chemical equation was demonstrated by Kroger *et al.* (17) and by Thurmond (18). Substituting Eq. [16] and [21] into Eq. [33]

gives

$$\mu_n^* + kT \ln n_0 \gamma_{n_0} = -\mu_p^* - kT \ln p_0 \gamma_{p_0} \quad [35]$$

After rearrangement, one can find

$$a_{p_0} a_{n_0} = p_0 n_0 \gamma_{p_0} \gamma_{n_0} = e^{-(\mu_n^* + \mu_p^*)/kT} = K \quad [36]$$

where  $K$  is a constant called the equilibrium constant. The entire equation can be called the equilibrium law or mass action law for the reaction (Eq. [34]), because when this system is at equilibrium, the value on the left side of the equation must equal the value of  $K$  at a given temperature. If the value on the left side does not equal  $K$ , the system is not in a state of dynamic equilibrium. Substituting Eq. [17] and [22] for  $V(0) = 0$ , or Eq. [19] and [24] for  $qV(0) = -kT \ln(-D + \sqrt{D^2 + 1})$  into Eq. [36] gives

$$a_{p_0} a_{n_0} = p_0 n_0 \gamma_{p_0} \gamma_{n_0} = n_i^2(0) = K \quad [37]$$

It is apparent from Eq. [37] that the equilibrium constant is a function only of temperature because  $n_i(0)$  is a function only of temperature. Furthermore, substituting Eq. [27] and [28] into Eq. [37] yields Eq. [31].

The results presented here imply that the equation  $0 \rightleftharpoons e^-(n) + e^+(p)$  is a legitimate chemical equation. To indicate chemical equilibrium in a reacting system, we use a set of double arrows,  $\rightleftharpoons$ . The use of this notation implies that the forward reaction (the reaction going from left to right) is occurring at the same rate as the reverse reaction. It is noted that the use of a generation-recombination process of the carriers, as discussed by Bar-Lev (15), can lead to a similar result as is presented here.

### Discussion

From Eq. [3] and [4] with constant electrostatic potential, we can obtain

$$\chi(0) - \chi(x) = E_c(x) - E_c(0) \equiv -kT\Delta_e \quad [38]$$

and

$$[E_G(x) - E_G(0)] + [\chi(x) - \chi(0)] = E_v(0) - E_v(x) \equiv -kT\Delta_h \quad [39]$$

Substituting Eq. [38] and [39] into Eq. [27] and [28] gives

$$\gamma_{n_0} = \exp\left\{\frac{[E_c(x) - E_c(0)] + kT \ln \left[\frac{N_c(0)}{N_c(x)}\right]}{kT}\right\} = \exp(\Delta_c - \Delta_e) \quad [40]$$

and

$$\gamma_{p_0} = \exp\left\{\frac{[E_v(0) - E_v(x)] + kT \ln \left[\frac{N_v(0)}{N_v(x)}\right]}{kT}\right\} = \exp(\Delta_v - \Delta_h) \quad [41]$$

where  $\Delta_c$  and  $\Delta_v$  are negative values defined as

$$\Delta_i = \ln \left[\frac{N_i(0)}{N_i(x)}\right], \quad i = c, v \quad [42]$$

and represent the shift in the density of states owing to Coulomb and exchange interaction (7). The fraction of the reduction in bandgap that occurs in the conduction band and valence band is represented by  $\Delta_e$  and  $\Delta_h$  (usually positive), respectively. Equations [40] and [41] are consistent with the form of the activity coefficients presented by Harvey (3) and Landsberg and Guy (7). It is noticed that  $\gamma_{n_0}$  and  $\gamma_{p_0}$  are negative deviations ( $\gamma_{n_0}, \gamma_{p_0} < 1$ ) because  $A \Delta E_g$  and  $(1-A)\Delta E_g$  are positive quantities. This implies that the effect of the bandgap shrinkage and the change in density of states would result in a net attraction (e.g., electron-positive impurity ion and hole-negative impurity ion interactions). This further implies that the impurity-band widen-

ing (19) and band tailing (20, 21) effects may play an important role in the attractive interactions.

Using Eq. [5], [7], and [8], and Eq. [6], [7], [8] and [21], by purely algebraic operations, the following relations are obtained between thermodynamic quantities and parameters of the band theory

$$\mu_n^* + kT \ln \gamma_{n_0}(x) = E_c(x) + qV(x) - kT \ln [N_c(x)] \quad [43]$$

and

$$\mu_p^* + kT \ln \gamma_{p_0}(x) = -[E_v(x) + qV(x)] - kT \ln [N_v(x)] \quad [44]$$

By evaluating Eq. [43] and [44] at  $x = 0$ , we obtain the same results of  $\mu_n^*$  and  $\mu_p^*$  given by Eq. [18], [20], [23], and [28]. As discussed above,  $\mu_n^*$  and  $\mu_p^*$  are functions of temperature only, thus,  $E_c(x)$  and  $E_v(x)$  must vary with the electrostatic potential and concentration at a given temperature. Accordingly, apart from the variations in  $N_c(x)$  and  $N_v(x)$ , the energy gap  $E_G(x) = E_c(x) - E_v(x)$  will decrease as a result of attractive interactions leading to negative deviations for  $\gamma_{n_0}(x)$  and  $\gamma_{p_0}(x)$ .

Choosing  $qV(0)$  as the boundary condition for the integration of Poisson's equation (assuming a constant dielectric constant  $\epsilon$ )

$$-\nabla^2 V = \frac{q}{\epsilon} (p - n + N_D^+ - N_A^-) \quad [45]$$

leads to self-consistent results for both the thermodynamic quantities and the Poisson-Boltzmann equation for the electrostatic potential. It is also important to note that if the material at  $x = 0$  is intrinsic, then from Eq. [16], [17], [19], [21], [22], and [24] we find

$$E_F = \mu_n(0) = E_F(0) = -\mu_p(0) \quad [46]$$

Note that this result is independent of the choice of the reference states. It implies that the intrinsic level is a purely thermodynamic property of the intrinsic bulk semiconductor. It thus implies that the intrinsic level is only distantly relative to the potential per se, and not at all to their relative alignment. This is an important result and can be used to explain the problem of the intrinsic level that is not necessarily continuous at the heterojunction interface.

Finally, it is appropriate at this point to place this work in perspective, relative to several related studies mentioned above. In this paper, we have done a consistent thermodynamic analysis with the energy band model, including the choice of the reference states and the explicit equilibrium constant which is a function only of temperature, and some other important results shown above. These results do not exist in the literature. In the future, this work will be extended to degenerate semiconductors with nonuniform composition.

### Conclusions

In this paper, we have presented the activity coefficients expressed in terms of the changes in bandgap, electron affinity, and density of states in the nonuniform semiconductors. These activity coefficients can also be used in the

application of semiconductor junction devices in thermal equilibrium.

Several different techniques for deriving the activity coefficients with uniform composition exist, but the generality and simplicity of our formulation are important advantages. The choice of the reference states and the relations between the thermodynamic quantities and the parameters of the energy band model are discussed. The importance of not using the intrinsic energy level to measure the electrostatic potential within semiconductors has been stressed. The equation  $0 \rightleftharpoons e^+(p) + e^-(n)$  is also shown to be a legitimate equilibrium chemical equation. This approach is restricted, however, by the assumption that the semiconductor is nondegenerate.

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