Interacting Green’s Function and Lehmann’s Representation in Photoemission Experiments and Interaction Effects

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Abstract

The phenomenon of photoelectric effect was discovered by W. Hertz in 1887 experimentally long ago, and as time passed theoretical explanation was given, the important work of Albert Einstein in 1905 that earned him Nobel Prize in 1921. Then experiments were done to measure Planck’s constant h and the measurement of electron charge, and the award of Nobel Prize to R.A. Millikan in 1923. As Quantum mechanics and quantum field theory was developed, more refined and complex theories to explain photoelectric effect were developed. Especially the theory of Green’s functions, and Greens function Lehmann representation were developed to explain the phenomena of photoemission. Some significant details of the phenomena of photoemission and its theoretical understanding are presented in this article.

Keywords: Green’s Function, Lehmann’s Representation, Photoemission, Photoelectric Effect

Introduction

Photoelectric effect was first discovered (observed) by Heinrich Hertz during experiments with a spark gap generator in 1887. In these experiments, sparks generated between two small metal spheres in a transmitter induced sparks that jump between two different metal spheres in a receiver. Hertz found that he could increase the resistivity of the spark gap device by illuminating it with visible or ultraviolet light. Later, studies by J.J Thomson showed that this increased resistivity was the result of light pushing an electron— a particle that he discovered in 1897. Freeing tiny particles of negative charge from the surface of a metal when electromagnetic radiation falls on it is called photoelectric effect. Subsequent investigations on the photoelectric effects led to the results that the classical theory of electromagnetic radiation could not explain photoelectric effect. When the electromagnetic radiation or light interacted with electrons, it just did not behave like it was supposed to. It meant rebuilding a large portion of Physics from the ground up. In photoelectric effect, the time required for the photo-electron to be emitted from the surface of the metal is ~10⁻¹⁰ s. Whereas using Poynting (theorem) vector to calculate the energy required to release the photo-electron, it will require some 10⁴s to collect enough energy. The E-M theory failed to explain photoelectric effect.

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This article will have the following contents:

1. Interacting Green’s Function
2. Interacting Green’s Function and Lehmann’s Representation
3. The role of 1 and 2 in photoemission experiment and interaction effects.

Energy conservation

Before absorption of photons: hν + E(N,0)

After absorption of emission: E_\text{kin} + E(N-1,i)

Hence, hν + E(N,0) = E_\text{kin} + E(N-1,i)

or h - E_\text{kin} = E(N-1,i) – E(N,0) = -\epsilon_i

\( h = 6.63 \times 10^{-34} \text{ Js}^{-1} \\
= 6.63 \times 10^{-27} \text{ ergs} \\
= 4.14 \times 10^{-15} \text{ evs} \\
hc = 1.99 \times 10^{-23} \text{ Jm} \\
= 1240 \text{ ev nm} \\
h - E_\text{kin} = E(N-1,i) - E(N,0) = -\epsilon_i \\

Figure 1: A sample absorbs a photon and emits an electron
Addition/Removal Energies and Photoemission Experiments

Figure 2: Addition/removal energies and photoemission experiments

Now, if $h\nu$ is the energy of the incident light radiation, and $E^N_0$ is the energy of the N-particle (electrons) before the light radiation falls on it, and $E_{\text{kin}}$ is the kinetic energy of the emitted electron when the light radiation impact on it knocks it out from the system, and $E^{N-1}_n$ is the energy of the system (that may not be in the ground state) after electron emission, then according to the law of conservation of energy,

$$h\nu + E^N_0 = E_{\text{kin}} + E^{N-1}_n$$

or

$$E^{N-1}_n = E^N_0 - E^{N-1}_n = E_{\text{kin}} - h\nu \leq \mu$$

(chemical potential)

For the process to take place, $h\nu > E_{\text{kin}}$, or $E_{\text{kin}} < h\nu$ is negative or it must be $<\mu$. The threshold result will lead to $=\mu$.

Removal of Photon (electron addition)

Figure 3: Removal of Photon (electron addition)

Now in this process, an electron with kinetic energy $E_{\text{kin}}$ enters the system of N-particles, and a photon with energy $h\nu$ is emitted, and the state of excitation is denoted by $E_n^N$, and the number of particles (electrons) of the system increases to $N+1$. Hence according to the law of conservation of energy we can write,

$$E_{\text{kin}} + E^N_0 = h\nu + E^{N+1}_n$$

(2)

$$E^{N+1}_n - E^N_0 = E_{\text{kin}} - h\nu (> m)$$

(3)

Since $E_{\text{kin}} - h\nu$ is positive.

Now if we look at the poles of the time-ordered Green’s Function in the complex plane, we get,

(i) Electron removal energies as,

$$h\nu = (E^N_0 - E^{N-1}_n) + i\eta$$

(4)

(ii) Electron addition energies as,

$$h\nu = (E^{N+1}_n - E^N_0) - i\eta$$

(5)

Case (i) corresponds to electron removal or ionization, and hence the ionization potential (IP) is,

$$IP = E^{N-1}_0 - E^N_0$$

(6)

Case (ii) corresponds to the electron addition or the electron affinity (EA), and hence

$$EA = E^{N+1}_0 - E^N_0$$

(7)

Hence the retarded single-particle Green’s Function leads to the following results.

(1) Electron removal energies; $h\nu = (E^N_0 - E^{N-1}_n) + i\eta$

(2) Electron addition energies; $h\nu = (E^{N+1}_n - E^N_0) - i\eta$

ARPES (Angle-Resolved Photoemission)

Figure 4: Some Experimental Results from the Angle Resolved Photoemission

Some simple experimental facts (results) from the angle-resolved photoemission are given here. It is well known that the incident photon energy is 21.2eV, while the variation of $w$ is given on a scale of 200meV so that for all practical purposes the momentum vector is a fixed length vector. Hence, the angle of emission of the electron with respect to the incident photon suffices to define the value of $k_1$. This means that for a given angle measured from the direction of incidence of the photon, the value of $k_1$ will change. Electrons emitted with zero energy are extracted from the Fermi level. Electrons with a smaller kinetic energy come from states with larger binding energy. The electrons inside the system are not free particles, they are rather quasi-particles whose energy disperses with wave vector and that they have a finite lifetime.
How to calculate the Green's Function of the Problem

To calculate the Green's Function of the system, we have to work out the time dependence of the fermions/boson operators (depending upon whether the assembly is composed of fermions or bosons).

We first consider non-interaction fermions (electrons), and so calculate the K-space Green's function for non-interacting electrons, the Hamiltonian, \( H \), is given by,

\[
H = \sum_{k,\sigma} \lambda_k C_{k\sigma}^+ C_{k\sigma} \tag{8}
\]

where

\[
\lambda_k = \varepsilon_k - \mu \tag{9}
\]

Such that \( \mu \) is the chemical potential, and \( \varepsilon_k \) is the k-th energy state, \( \sigma \) is the spin. The Hamiltonian, \( H \), is diagonal in \( k \) and \( \sigma \) since the system is translationally invariant in space, and as there is no external potential in the Hamiltonian. Hence the Green's Function will also be diagonal in \( k \) and \( \sigma \). Keeping this in mind, we consider the retarded Green's Function,

\[
G_0^R(k, \sigma; t - t') = -i\theta(t - t') \{ C_{k\sigma}(t), C_{k\sigma}^+(t') \} \tag{10}
\]

where \( \theta(t-t') \) is the Heaviside step function, i.e,

\[
\theta(t-t') = \begin{cases} 1, (t - t') > 0 \\ 0, (t - t') < 0 \end{cases} \tag{11}
\]

In the case of retarded Green's function, \( t > t' \), i.e, the cause comes before the effect.

Now to calculate the Green's Function, we have to work out the time dependence of the fermion operators, \( C_{k\sigma} \). Thus we write,

\[
C_{k\sigma}(t) = e^{iHt} C_{k\sigma} e^{-iHt} \quad \text{(h = 1)} \tag{12}
\]

\[
C_{k\sigma} = C_{k\sigma}(t = 0)
\]

and also,

\[
\frac{dC_{k\sigma}(t)}{dt} = i[H, C_{k\sigma}(t)] \tag{13}
\]

If we include \( 1/h \), then we write,

\[
C_{k\sigma}(t) = e^{i\hbar^{-1}Ht} C_{k\sigma} e^{-i\hbar^{-1}Ht} \tag{14}
\]

and

\[
\frac{dC_{k\sigma}(t)}{dt} = i\hbar^{-1}[H, C_{k\sigma}(t)] \tag{15}
\]

Using eq. (12) for writing any operator, we can write, the operator \([H, C_{k\sigma}(t)]\) as,

\[
[H, C_{k\sigma}(t)] = e^{iHt}[H, C_{k\sigma}] e^{-iHt} \tag{16}
\]

Hence eq. (13) can be written as,

\[
\frac{dC_{k\sigma}(t)}{dt} = i e^{iHt} [H, C_{k\sigma}] e^{-iHt} \tag{17}
\]

where

\[
[H, C_{k\sigma}] = -\lambda_k C_{k\sigma} \tag{18}
\]

Eq. (17) can now be written as,

\[
\frac{dC_{k\sigma}(t)}{dt} = i e^{iHt} [-\lambda_k, C_{k\sigma}] e^{-iHt} = -i\lambda_k, C_{k\sigma}(t) \tag{19}
\]

Integrating the differential equation in eqn (19), we get,

\[
\frac{dC_{k\sigma}(t)}{C_{k\sigma}(t)} = -i\lambda_k dt
\]

or

\[
\log C_{k\sigma}(t) = -i\lambda_k t
\]

or

\[
\log \frac{C_{k\sigma}(t)}{C_{k\sigma}(0)} = -i\lambda_k t \quad \text{or} \quad \frac{C_{k\sigma}(t)}{C_{k\sigma}(0)} = e^{-i\lambda_k t}
\]

or

\[
C_{k\sigma}(t) = C_{k\sigma}(0) e^{-i\lambda_k t} \tag{20}
\]

and

\[
C_{k\sigma}^+(t) = C_{k\sigma}^+(0) e^{i\lambda_k t} \tag{21}
\]

Eqns. (20) and (21) give the time dependence of the operators for no-interacting electrons.

We can now write the greater Green’s Function \((t > t' \text{ in } t-t')\) as,

\[
G_0^>(k, \sigma, t-t') = -i \{ C_{k\sigma}(t) C_{k\sigma}^+(t') \}
\]

\[
= -i e^{-i\lambda_k t} e^{i\lambda_k t'} \{ C_{k\sigma}(0) C_{k\sigma}^+(0) \}
\]

\[
= -i e^{-i\lambda_k t} t - t' (1 - C_{k\sigma}(0) C_{k\sigma}(0))
\]

\[
= -i e^{-i\lambda_k t} t - t' (1 - \langle C_{k\sigma}(0) C_{k\sigma}(0) \rangle)
\]

\[
= -i e^{-i\lambda_k t} t - t' (1 - n_{k\sigma}) \tag{22}
\]

\[
= -i e^{-i\lambda_k t} t - t' (1 - n_F(\lambda)) \tag{23}
\]
where \( n_r(\lambda) \) or \( n_r(\omega) \) is the Fermi-Dirac distribution function and is given by,

\[
n_r(\omega) = \frac{1}{e^{\beta \omega} + 1} = \frac{1}{e^{\beta \omega} + 1}(\beta = \frac{1}{kT})
\]

At zero temperature \( T = 0, \beta = \frac{1}{kT} = \infty \),

we get, \( n_r(\omega) = \theta(-\omega) \), and hence a state \( k \) is occupied if \( \epsilon_k < \mu \), and empty if \( \epsilon_k > \mu \).

Similarly we can write the lesser Green’s Function

\[
\hat{G}_\omega^\text{L}(k\sigma, t - t'), \text{ here } t < t' \text{ in } (t - t'); \text{ i.e.,}
\]

\[
\hat{G}_\omega^\text{L}(k\sigma, t - t') = +i (C_{k\sigma}(t')C_{k\sigma}(t)) = i e^{i\lambda_k t'} e^{-i\lambda_k t} \ln G_{k\sigma}(t = t = 0) \]

\[
\hat{G}_\omega^\text{L}(k\sigma, t - t') = -t e^{-i\lambda_k t'} n_k(\lambda_k)
\]

Now the retarded Green’s Function \( \hat{G}_0^R(k\sigma, t - t') \) is written in terms of \( G \rangle \) and \( G \langle \) as, in general,

\[
\hat{G}_\omega^R(x, t; x', t') = \theta(t - t') [\hat{G}_\omega^\text{R}(x, t; x', t') - \hat{G}_\omega^\text{R}(x, t; x', t')]
\]

Or we can write \( \hat{G}_\omega^R \) by subtracting eqn (26) from eq.(23), such that,

\[
\hat{G}_\omega^R(k, \sigma; t - t') = -i \theta(t - t') e^{-i\lambda_k t'}
\]

Now we have to write the Fourier transform of \( \hat{G}_\omega^R(k\sigma, t - t') \). The general expression for the Fourier transform is

\[
\hat{G}_\omega(\omega) = \int_{-\infty}^{\infty} e^{-i\omega t} G(t) dt
\]

Using eqn (29) we write the Fourier transform of the \( \hat{G}_\omega^R \) in eqn (28), and get,

\[
\hat{G}_\omega^R(k\sigma, \omega) = \int_{-\infty}^{\infty} e^{i\omega t} \theta(t) e^{-i\lambda_k t} dt
\]

Now \( \theta(t) \) will be zero between \( t = -\infty \) to \( t = 0 \); and hence we can write since \( \theta(t) = 1 \) between \( t = 0 \) and \( t = \infty \), \( \hat{G}_\omega^R \) as,

\[
\hat{G}_\omega^R(k\sigma, \omega) = -\int_{-\infty}^{\infty} e^{i(\omega - \lambda_k) t} dt
\]

To make the integral converge at the upper limit, we let \( \omega = \omega' + i\eta \) where \( \eta = 0' \) is a positive infinitesimal. Thus we get,

\[
\hat{G}_\omega^R(k\sigma, \omega) = \frac{1}{\omega - \lambda_k + i\eta}
\]

It should be noted that the Green’s Function in eqn (32), if considered as a function of \( \omega \) for fixed \( k \), has a pole at \( \omega = \lambda_k \), i.e., at the excitation energy \( \lambda_k \) of the system, except that the pole is just shifted infinitesimally off the real axis in the complex \( \omega \)-plane and down into the lower half-plane. Hence the Fourier transform of the retarded Green’s Function has the following properties. It is analytic in the upper half-plane, and the location of its poles (all in the lower half-plane) offer information about the excitation energies of the system. In fact, these are the general features of the Fourier transform of the retarded Green’s Function.

**The Lehmann’s Representation**

Fig. (2) Shows the emission of an electron from an assembly of \( N \) electrons when a photon is incident on the assembly. Whereas fig. 3 shows the emission of a photon when an electron is incident on the assembly of \( N \) electrons. According to fig.(2)the number of electrons reduces by 1 such that the assembly is left with \( N-1 \) electrons. In fig.(3), the number of electrons increases by 1 such that the assembly has \( N+1 \) electrons. We have now to correlate the energies involved \( E_{\omega Nor} = E_{\omega Nor} \) with the corresponding Green’s Functions that are associated with the process. It should also be understood that fig.(2) corresponds to excited states with one more hole, whereas fig.3 corresponds to excited states with one more particle (electron). Photoemission experiments access the place of the weight with reference to fig. 2; whereas Bremsstrahlung Inverse Spectroscopy (BIS) experiments measure what is described in fig.3. Excited particle state contributes to positive frequencies, say \( \omega \), if their excitation energy is larger than the chemical potential, i.e, \( E_m - E_\mu > \mu \), and to negative frequencies otherwise zero frequency means that the excitation energy is equal to the chemical potential \( \mu \). This means that every excited single-particle state or single-hole state corresponds to a delta-function in the spectral weight whose weight depends on the overlap between initial states with one more particle at \( r' \), or one more hole at \( r' \), and the true excited states. In the above lines, in a way, the physical interpretation of the spectral weight is described.

It must be understood that if \( E_m \) is the energy of an eigen state with one more particle than the ground state energy \( E_\mu \), then the minimum energy required to add a particle is the chemical potential energy \( \mu \), and the delta function contributes to positive frequencies. On the other hand, if \( E_m \) is the energy with one less particle, such that \( 0 \leq (E_m - E_\mu) \leq \mu \), since we can remove a particle or create a hole below the Fermi surface. Hence this will lead to negative frequencies.

We now briefly describe how to understand the spectral representation of the photoemission experiments using what is called the Lehmann’s representation of the Green’s Function. The spectral representation is obtained by introducing a complete set of orthonormal states (without the total restriction on the total number of particles) into the expressions for \( \hat{G}_\omega \) \( (\omega = \omega' > 0) \) and \( \hat{G}_\omega \) \( (\omega' > 0) \). It should be emphasized that we are concerned with the exact eigen states of the complete system, with interactions, and not with any kind of approximate states. By introducing the orthonormal set between the \( a_\sigma(\gamma) \) (annihilation operator) and \( a_\sigma^\dagger(\gamma) \) (creation operator), we can write \( \hat{G}_\omega \) \( (\gamma>0) \) as, [here \( G_k(\gamma>0) \) is the Fourier transform of the Green’s Function \( G(\sigma, \tau, r') \),

\[
G_k(\gamma > 0) = -\langle \phi_\gamma | a_\sigma(k) a_\sigma^\dagger(t) | \phi_\gamma \rangle
\]

\[
= -\sum_{\eta} \langle N; 0 | a_\sigma(t_1) N + 1; 1 | N + 1; 1 | a_\sigma^\dagger(t_2) | N; 0 \rangle
\]
Now if $H$ is the Hamiltonian of the system, we can write $a_k(t)$ and $a_k^*(t)$ as,

$$a_k(t_1) = e^{iHt_1}a_k e^{-iHt_1}$$ \hspace{1cm} (35)

$$a_k^*(t_2) = e^{iHt_2}a_k^* e^{-iHt_2}$$ \hspace{1cm} (36)

Substituting eqn. (35) and (36) in eqn. (34), we get, \(t_1 - t_2 = \tau\)

$$G_k(\tau > 0) = -i \sum_t \langle N; 0 | e^{iHt_1}a_k e^{-iHt_1} | N + 1; 1 \rangle \langle N + 1; 1 | a_k^* e^{-iHt_2} | N; 0 \rangle$$

$$= -i \sum_t \langle N; 0 | a_k^* | N + 1; 1 \rangle e^{i\omega_{L,0}t} e^{-i\omega_{N+1,1}t}$$

$$= -i \sum_t \langle N; 0 | a_k^* | N + 1; 1 \rangle \langle N + 1; 1 | a_k^* | N; 0 \rangle e^{i(\omega_{N,0} - \omega_{N+1,1})} (t_1 - t_2)$$ \hspace{1cm} (37)

where $t_1 - t_2 = \tau$

Similarly, we can prove that,

$$G_k(\tau > 0) = -i \sum_t |\langle a_k^* | N+1; 1 \rangle|^2 e^{i(\omega_{N,0} - \omega_{N+1,1})} \tau$$ \hspace{1cm} (38)

In eqn.(37) and (38), $E_{N+1,1}$ refers to the energy of the state $l$ of (N+1) particle system; and $E_{N+1,1}$ refers to the energy of the state $l$ of (N-1) particle system; and $E_{N+1,1}$ refers to the energy of the state $l$ of (N-1) particle system; and $E_{N+1,1}$ refers to the energy of the ground state of the N-particle system.

Now the energy difference can be written as,

$$E_{N+1,1} - E_{N,0} = \omega_{L,0} + \mu$$ \hspace{1cm} (39)

$$E_{N+1,1} - E_{N,0} = \omega'_{L,0} - \mu'$$ \hspace{1cm} (40)

where

$$\omega_{L,0} = E_{N+1,1} - E_{N+1,0}$$ \hspace{1cm} (41)

$$\omega'_{L,0} = E_{N-1,1} - E_{N-1,0}$$ \hspace{1cm} (42)

$$\mu = E_{N+1,0} - E_{N,0}$$ \hspace{1cm} (43)

$\omega_{L,0}$ = excitation energy of the (N+1) particle system.

$\omega'_{L,0}$ = excitation energy of the (N-1) particle system.

$\mu$ = chemical potential of the (N+1) particle system.

$\mu'$ = chemical potential of the (N-1) particle system.

If the number of particles of the system is very large, then we can write,

$$\omega_{L,0} \approx \omega'_{L,0}$$ and $\mu \approx \mu'$ \hspace{1cm} (45)

The error introduced in obtaining eq. (45) is of the order of $1/N$, which is very small for large $N$. Substituting eqns. (39), (40) and (45) in eqns. (37) and (38) we get,

$$G_k(\tau > 0) = -i \sum_t |\langle a_k^* | N+1; 1 \rangle|^2 e^{-i(\omega_{L,0} + \mu) \tau}$$ \hspace{1cm} (46)

$$G_k(\tau < 0) = +i \sum_t |\langle a_k^* | N-1; 1 \rangle|^2 e^{i(\omega_{L,0} - \mu) \tau}$$ \hspace{1cm} (47)

For ($\tau > 0$), the intermediate states $|l>$ contain (N+1) particles and have total momentum $k$. Whereas for ($\tau < 0$), these states correspond to (N-1) particles with momentum $-k$.

Assuming that the states $|l>$ are densely packed, we can regroup the different states $|l>$ by defining two functions,

$$A(k, \omega) = \sum_t |\langle a_k^* | N+1; 1 \rangle|^2 \delta(\omega - \omega_{L,0})$$ \hspace{1cm} (48)

$$B(k, \omega) = \sum_t |\langle a_k^* | N-1; 1 \rangle|^2 \delta(\omega - \omega'_{L,0})$$ \hspace{1cm} (49)

$A(k, \omega)$ and $B(k, \omega)$ are real positive functions, whose physical interpretation is quite simple. For instance, let us make a spectral analysis of the state $a_k^* |\phi_0>$. The total norm of the state is distributed among the different frequencies, and the ‘density of the norm’ at the frequency $\omega_{L,0}$ being given by $A(k, \omega)$. Similarly, $B(k, \omega)$ is the density of the norm at the frequency $\omega_{L,0}$. Actually if $a_k^* |\phi_0>$ were an exact eigen state of the system of excitation energy $\omega_{L,0}$, then the function $A(k, \omega)$ would reduce to the Dirac $\delta$-function $\delta(\omega - \omega_{L,0})$. Actually $a_k^* |\phi_0>$ is not an eigen state; the delta function spreads out and gives rise to a finite density $A(k, \omega)$. However, $A(k, \omega)$ can approximately exhibit a sharp peak and this peak corresponds to an approximate elementary excitation.

Multiplying eqn.(48) by $e^{-i(\omega_{L,0} + \mu) \tau}$ and eqn.(49) by $e^{i(\omega_{L,0} - \mu) \tau}$ and integrating from 0 to $\infty$, we get,

$$G_k(\tau > 0) = -i \int_0^\infty A(k, \omega) e^{-i(\omega + \mu) \tau} d\omega$$ \hspace{1cm} (50)

$$G_k(\tau < 0) = i \int_0^\infty B(k, \omega) e^{i(\omega - \mu) \tau} d\omega$$ \hspace{1cm} (51)

Fourier transforming eqns.(50) and (51), we get,

$$G_k(E) = G_k(E) \int_{-\infty}^{+\infty} G_k(\tau) e^{iEt} d\tau$$
where $\eta$ is a real positive infinitesimal quantity. The spectral representation given by eq.(52) was derived by Lehmann in quantum field theory.[Lehmann H. Nuovo Cimento, 11(1954)342]. This is the Lehmann representation of Green’s function. The complex function $G(k, E)$ actually depends on two real functions $A(k, \omega)$ and $B(k, \omega)$ defined in the interval $(0, +\infty)$.

Spectral Weight for Non-Interacting Particles
To discuss the physical meaning of the spectral weight, a formal general expression of the Lehmann representation is written, and this allows us to see its more general meaning.

The general expression for the spectral weight $A_0(k, \omega)$ in terms of the Green’s Function for non-interacting particles is,

$$A_0(k, \omega) = i \left( \frac{1}{\omega - \lambda_k + i\eta} - \frac{1}{\omega - \lambda_k - i\eta} \right)$$

$$= 2\pi \delta(\omega - \lambda_k)$$

Conclusion
In physical terms, this tells us that for non-interacting particles in a translationally invariant system, a simple excited particle or hole of momentum $k$ added to an eigen state is a true excited eigen state located at an energy above or below the Fermi level. In the interacting case, the Lehmann representation will show us clearly that the results obtained in eqn. (52) is the correct representation.

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