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### Tunable phonon resonances and thermal conductance in weakly nonlinear disordered systems with short-range correlation

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Abstract – We theoretically investigate the resonant transmission of phonons and step-like thermal conductance in a weakly nonlinear generalized random n-mer (NGRN) system, where the impurity cluster with short-range correlation is randomly distributed in the host monatomic chain, and the atoms are connected by anharmonic potentials. The weakly anharmonic potential can be reduced to the quasi-harmonic form, which is tuned by external stretching. Due to the delocalization of phonons, resonant transmission is observed and thermal conductance presents a "quantized" feature. By applying external stretching on the NGRN system, the number of resonant modes and their locations are tuned. As a consequence, the "quantized" thermal conductance becomes tunable in this system. This finding may achieve potential applications in designing novel thermal-conducting materials.

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Nowadays much attention has been paid to thermal conductance and thermal-conducting materials [1–3]. On the one hand, in the integrated circuits (IC), power dissipation becomes one of the limits to the miniaturization of electronic devices [4]. It is necessary to develop some materials with high thermal conductance in order to cool the working devices on IC. On the other hand, minimum thermal conductivity is pursued in thermo-electric systems in order to improve the efficiency of energy conversion [5,6]. Therefore, high or low thermal-conducting materials are achieving much attracting applications. One may naturally try to develop a tunable thermal conductance system. Tunable nanoscale thermal links have immediate implications for nano- to macroscale thermal management, biosystems, and phononic information processing [7]. In this letter, we demonstrate theoretically a tunable phonon resonance and thermal conductance in the weakly nonlinear generalized random *n*-mer systems (NGRN). Based on the elastic wave equation, we study phononic transmission and thermal conductivity in the NGRN system, where the impurity cluster with short-range correlation is randomly distributed in the host monatomic chain, and the atoms in the chain are connected by anharmonic potentials. The weakly anharmonic potential can be

reduced to the quasi-harmonic form, which is tuned by external stretching. Multiple resonant transmissions are observed and thermal conductance presents a "quantized" feature. By stretching or compressing the NGRN system, the number of resonant modes and their locations are tuned, thereafter, the "quantized" thermal conductance becomes tunable in this system.

First, we construct a NGRN system, which contains a host atom A and a cluster of impurity atoms  $\mathbf{B} = \{B_1 B_2 \cdots B_n\}$ . The host atom A and the impurity cluster  $\mathbf{B} = \{B_1 B_2 \cdots B_n\}$  are randomly arranged in the chain, as

$$\underbrace{A\dots A}_{X_1}\underbrace{B\dots B}_{Y_1}\underbrace{A\dots A}_{X_2}\underbrace{B\dots B}_{Y_2}\underbrace{\dots A\dots A}_{X_i}\underbrace{B\dots B}_{Y_i}\underbrace{\dots A\dots A}_{X_m}\underbrace{B\dots B}_{Y_m},$$
(1)

where  $X_j$  is the number of atom A in the *j*-th cluster of A, which is random. And  $Y_j$  is the number of cluster **B** in the *j*-th cluster of **B**. All the atoms in the chain are connected by the anharmonic FPU- $\beta$  potential [8], which is well-known in nonlinear systems. Besides, the NGRN chain is a generalized model based on the random-*dimer* system [9].

Now we consider the transport of phonons in the NGRN chain, which is stretched or compressed. The equation of atomic motion in the NGRN chain under stretching can

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be expressed as

$$-m_{i}\frac{\mathrm{d}^{2}\mu_{i}}{\mathrm{d}t^{2}} = \alpha_{i-1,i}(\mu_{i-1}-\mu_{i}-l_{i}) + \beta_{i-1,i}(\mu_{i-1}-\mu_{i}-l_{i})^{3} + \alpha_{i,i+1}(\mu_{i+1}-\mu_{i}+l_{i+1}) + \beta_{i,i+1}(\mu_{i+1}-\mu_{i}+l_{i+1})^{3}, \quad (2)$$

where  $m_i$  is the mass of the *i*-th atom,  $\mu_i$  is the vibration displacement of the *i*-th atom from its equilibrium,  $\alpha_{i,i+1}$ and  $\beta_{i,i+1}$  are the strength of the harmonic coupling and of the anharmonic coupling between the *i*-th atom and the (i + 1)-th atom, respectively. The coupling parameters  $\alpha_{i,i+1}$  and  $\beta_{i,i+1}$  are chosen according to the distribution of the impurity cluster in the NGRN chain. The parameters  $l_i$  is the stretching length of the *i*-th atom. Consider a static stretch  $f_i$ , we can obtain  $l_i$  and  $l_{i+1}$  by the equations  $\alpha_{i-1,i}l_i + \beta_{i-1,i}l_i^3 = \alpha_{i,i+1}l_{i+1} + \beta_{i,i+1}l_{i+1}^3 = f_i$ . In the case of  $|\mu_{i-1} - \mu_i| \ll l_i$ , we can realize the quasiharmonic approach. All the terms of  $(\mu_{i-1} - \mu_i)$ , whose power is higher than one, are omitted. Then we can apply a rotating-wave approximation, and eq. (2) is now reduced to the quasi-harmonic form as follows:

$$-m_i\omega^2\mu_i = \alpha^i_{eff}(\mu_{i-1} - \mu_i) + \alpha^{i+1}_{eff}(\mu_{i+1} - \mu_i), \quad (3)$$

where  $\alpha_{eff}^i$  is the strength of effective harmonic potential, *i.e.*,  $\alpha_{eff}^i = \alpha_{i-1,i} + 3\beta_{i-1,i}l_i^2$ , and  $\omega$  is the vibration frequency. Obviously, the effective harmonic potential  $(\alpha_{eff}^i)$  depends on the stretching length  $(l_i)$ , which results from the anharmonic term of the FPU- $\beta$  potential. Although nonlinear excitation and mode interaction are omitted in the quasi-harmonic approach of the NGRN chain, the effective harmonic potential can be tuned by external stretching due to the weakly nonlinear coupling in the NGRN, which cannot be realized in an ordinary harmonic chain. Equation (3) can be rewritten in a matrix form as

$$\begin{pmatrix} \mu_{i+1} \\ \mu_i \end{pmatrix} = \begin{pmatrix} 1 + \frac{\alpha_{eff}^i - m_i \omega^2}{\alpha_{eff}^{i+1}} & -\frac{\alpha_{eff}^i}{\alpha_{eff}^{i+1}} \\ 1 & 0 \end{pmatrix} \begin{pmatrix} \mu_i \\ \mu_{i-1} \end{pmatrix} \equiv M_i \begin{pmatrix} \mu_i \\ \mu_{i-1} \end{pmatrix},$$

$$(4)$$

where  $M_i$  is the transfer matrix that correlates the vibration displacements of adjacent sites  $\mu_i$  and  $\mu_{i+1}$ . Therefore, the atomic vibration in the whole chain is determined by a product matrix, *i.e.*, a global transfer matrix  $M = \prod_{i=N}^{1} M_i \equiv \begin{pmatrix} m_{11} & m_{12} \\ m_{21} & m_{22} \end{pmatrix}$ , where N is the number of atoms in the chain. The transmission coefficient of phonons through the whole chain can be described as

$$t = \frac{4}{2 + m_{11}^2 + m_{12}^2 + m_{21}^2 + m_{22}^2}.$$
 (5)

It has been established that the phononic transmission directly determines the heat transport in the system [3,10]. The thermal conductivity  $\kappa$  is contributed by different phononic modes of energy transport [11], *i.e.*,

$$\kappa = c \sum_{\nu} \left(\xi_{\nu,1}^{-2} + \xi_{\nu,N}^{-2}\right)^{-1},\tag{6}$$

where  $\xi_{\nu,n} = \sqrt{m_n} U_{\nu,n}$ ,  $U_{\nu,1}$  and  $U_{\nu,N}$  are the amplitudes of the  $\nu$ -th mode at both ends of the chain, respectively, and c is a coupling constant.

It is worthwhile to investigate how the short-range correlation affects the transport of phonons in the NGRN chain. Without loss of generality, we assume that in the NGRN chain, the masses of atoms are  $m_0$  for the atom A, and  $m_1$  for each atom in the cluster  $\mathbf{B} = \{B_1 B_2 \cdots B_n\}$ . To be simplified, the couplings for the connections of A-A and A-B are assumed to be harmonic, whose strength is set as  $\alpha$ . But the atoms in cluster **B** are connected by the FPU- $\beta$ potential, where  $\alpha'$  and  $\beta$  are the strength of the harmonic and of the anharmonic coupling, respectively. Thereafter, the effective potential between the atoms in cluster  $\mathbf{B}$  is  $\alpha_{eff} = \alpha' + 3\beta l^2$ . Here, l is the stretching length between neighbor atoms in the cluster **B**. Obviously, the coupling strength  $\alpha_{eff}$  can be tuned in the NGRN chain by external stretching. Suppose that the atoms in the impurity cluster **B** locate from the first site to the *n*-th sites in the NGRN chain, the vibration displacements of neighbor atoms near the cluster  $\mathbf{B}$  at a specific frequency can be given by

$$\begin{pmatrix} \mu_{n+2} \\ \mu_{n+1} \end{pmatrix} = M_{n+1}M_n \dots M_0 \begin{pmatrix} \mu_0 \\ \mu_{-1} \end{pmatrix} \equiv \begin{pmatrix} a_1 & a_2 \\ a_3 & a_4 \end{pmatrix} \begin{pmatrix} \mu_0 \\ \mu_{-1} \end{pmatrix}.$$
(7)

Suppose there exists the case that at some frequencies, the impurity cluster **B** does not affect the vibration of the neighbor atoms in the NGRN, *i.e.*, the vibration can propagate through the chain without decay. Physically, this situation happens if the behavior of a phonon with a specific frequency in the NGRN chain is similar to that in a homogeneous atom chain of the atom A. Therefore, at those specific frequencies, the phonon dispersion in the NGRN chain should be the same as that in a homogeneous atom chain of the atom A, *i.e.*  $\omega_R = \sqrt{4\alpha/m_0} \sin(qd_A/2)$ , where q is the wave vector and  $d_A$  is the distance between neighboring atoms in a homogeneous atom chain of the atom A. Then we have

$$2a_3\left(\frac{m_0\omega_R^2}{2\alpha}\right)^2 + (a_1 - a_4 - 4a_3)\frac{m_0\omega_R^2}{2\alpha} + a_4 + 2a_3 - a_1 = 0.$$
(8)

Once the frequency of the phonon satisfies eq. (8), the impurity cluster **B** seems "transparent" to the phonon, and the vibration can propagate through the chain without decay. Physically, once the frequency of the phonon satisfies eq. (8), the phonon can propagate through the whole NGRN chain, and from this point of view, the localization-delocalization transition of phonons takes place at the resonant frequency given by eq. (8). The delocalization of phonons in the NGRN chain is similar to the phononic properties in other random systems with short-range correlation [12,13], which can be considered as an analogy to the electronic delocalization in crystals [9,14–17] and also the photonic localization in dielectric microstructures [18].

According to eq. (8), the resonant frequency of phonons can be obtained in the NGRN chain. Interestingly, there are multiple resonant modes of phonons in this system. For example, there is one resonant frequency in the *dimer* chain (*i.e.*, the NGRN system with n = 2), *i.e.*,

$$\omega_R^2 = \frac{2\chi - \gamma}{m_1(m_1 - m_0)},\tag{9}$$

where  $\chi \equiv \beta(m_1 - m_0)$ ,  $\gamma \equiv m_0(\alpha - \alpha_{eff})$ . There are two resonant frequencies in the *trimer* chain (*i.e.*, the NGRN system with n = 3), *i.e.*,

$$\omega_R^2 = \frac{4\chi - \gamma \pm \sqrt{4\chi^2 + \gamma^2}}{2m_1(m_1 - m_0)}.$$
 (10)

And there are three resonant frequencies in the 4-mer chain (*i.e.*, the NGRN system with n = 4), *i.e.*,

$$\omega_R^2 = \frac{6\chi - \gamma + 2\sqrt{6\chi^2 + \gamma^2}\cos\left(\frac{f + 2p\pi}{3}\right)}{3m_1(m_1 - m_0)},\qquad(11)$$

where  $f = \arccos\left(\frac{9\chi^2 - 2\gamma^2}{2\sqrt{(6\chi^2 + \gamma^2)^3}}\right)$ , and p = 0, 1, 2. More resonant modes of phonons are expected as n increases in the NGRN chain. Therefore, both the number of resonant modes of phonons and their locations in the NGRN systems can be tuned by the external stretching and due to the fact that each resonant transmission of phonons will contribute to thermal conductivity in the system, the thermal conductivity will become tunable in the NGRN chain.

Based on the above analytical analysis, we have carried out the numerical calculations on phononic transport in the NGRN chain. Figure 1 shows the resonant frequency of phonons in several NGRN chains as a function of the effective potential  $(\alpha_{eff})$ , which stands for the quasiharmonic couplings among the atoms in the impurity cluster **B**. We consider two cases: one is the heavy dope corresponding to  $m_0 < m_1$ , and the other is the light dope corresponding to  $m_0 > m_1$ . The response of resonant frequencies on the coupling strength  $\alpha_{eff}$  presents a rich feature. In the generalized random *dimer* chain, the resonant frequency increases monotonously with increasing  $\alpha_{eff}$  in the case of heavy doping (as shown in fig. 1(a)); while the resonant frequency decreases monotonously with increasing  $\alpha_{eff}$  in the case of light doping (as shown in fig. 1(b)). Meanwhile in the generalized random trimer and 4-mer chains, the response of the resonant frequency on the coupling strength  $\alpha_{eff}$  becomes nonlinear (as shown in fig. 1(c)-(f), and the nonlinear response becomes more obvious in light doping than that in heavy doping. It is demonstrated that the resonant frequency of phonons can be adjusted by changing the effective coupling strength of atoms in the NGRN system. Note that the effective potential  $(\alpha_{eff})$  is changed by stretching or compressing the NGRN chain  $(\alpha_{eff} = \alpha' + 3\beta l^2)$ , where l is the stretching length). Thereafter, tunable resonant frequencies of phonons are achieved in the NGRN chain.



Fig. 1: The resonant frequency of phonons  $(\omega_R^2)$  as a function of the effective potential  $(\alpha_{eff})$  among the atoms in the impurity cluster in several NGRN chains. In the *dimer* chain (n = 2): (a) heavy doping, and (b) light doping. In the *trimer* chain (n = 3): (c) heavy doping, and (d) light doping. In the 4-mer chain (n = 4): (e) heavy doping, and (f) light doping. Note that in each light doping, the mass of the impurity atom in **B** is smaller than that of the host atom A, *i.e.*,  $m_0 = 1.5$  and  $m_1 = 1.1$ . While in each heavy doping, the mass of the impurity atom in **B** is heavier than that of the host atom A, *i.e.*,  $m_0 = 1.3$ and  $m_1 = 1.5$ . The strength for the connections A-A and A-B is kept the same as  $\alpha = 1.0$  for each chain.

By using the transfer-matrix method [13], the transmission of phonons in the NGRN chain is calculated based on eqs. (1)–(5). Besides, the type of boundary conditions is crucial to determine the transport of the disordered harmonic chain [11,19]. In our calculation, the fixed boundary condition is used. Figure 2 presents the phononic transmission as a function of frequency through several NGRN chains in the cases of heavy doping and light doping, respectively. It is obvious that some transmission peaks indeed appear in the phononic transmission spectra of the NGRN, and these resonant peaks locate exactly at the resonant frequencies predicted in eqs. (9)-(11) (as shown in fig. 2(a)-(f)). The resonant transmissions originate from the delocalization of phonons in the NGRN chains, which is similar to phononic and electronic behaviors in other correlated random systems [13–15]. Furthermore, both the number and the location of transmission peaks are changed with increasing the effective potential  $\alpha_{eff}$  in the NGRN chain. For instance, by increasing  $\alpha_{eff}$ , one transmission peak can be found and the resonant frequency can be adjusted in the *dimer* chains (as shown in fig. 2(a), (b)). One peak or two peaks can be observed and the resonant frequencies can be adjusted in the *trimer* chains (as shown in fig. 2(c), (d)). One peak, or two peaks, or even three transmission peaks can be found



Fig. 2: (Colour on-line) The transmission coefficient (T)as a function of frequency in several NGRN chains. In the dimer chain (n=2 and N=5819): (a) heavy doping with  $m_0 = 2.0, m_1 = 2.5, \alpha = 1.5, \text{ and } \alpha_{eff} = 1.0, 1.5, 2.0, \text{ respec-}$ tively; (b) light doping with  $m_0 = 2.5$ ,  $m_1 = 2.0$ ,  $\alpha = 1.5$ , and  $\alpha_{eff} = 1.0, 1.6, 2.3$ , respectively. In the trimer chain (n = 3)and N = 8713): (c) heavy doping with  $m_0 = 1.4$ ,  $m_1 = 1.6$ ,  $\alpha = 1.5$ , and  $\alpha_{eff} = 0.6$ , 1.0, 1.4, respectively; (d) light doping with  $m_0 = 1.5$ ,  $m_1 = 1.0$ ,  $\alpha = 1.3$ , and  $\alpha_{eff} = 0.6$ , 1.0, 1.4, respectively. In the 4-mer chain (n = 4 and N = 13923): (e) heavy doping with  $m_0 = 1.3$ ,  $m_1 = 1.5$ ,  $\alpha = 1.0$ , and  $\alpha_{eff} = 0.6, 1.0, 1.4, 1.8$  respectively; (f) light doping with  $m_0 = 1.6, \ m_1 = 1.5, \ \alpha = 1.4, \ \text{and} \ \alpha_{eff} = 0.9, \ 1.3, \ 1.7, \ 2.1,$ respectively. Note that the red dotted curves give the resonant frequency of phonons vs. the effective potential  $(\alpha_{eff})$ , in order to show that the resonant transmission peaks locate exactly at the resonant frequencies predicted in eqs. (9)-(11).

in the 4-mer chains, respectively (as shown in fig. 2(e), (f)). The shifts of transmission peaks with increasing  $\alpha_{eff}$  in fig. 2 are in good agreement with eq. (11). As we discussed above, by stretching or compressing the NGRN chain, the effective potential ( $\alpha_{eff}$ ) is tuned by the stretching length. In some sense, the tunable resonant transmission of phonons is reached in the NGRN chain.

Actually, the delocalization of phonons in NGRN chain can also be characterized by a zero Lyapunov coefficient at the resonant frequency. It is known that the Lyapunov coefficient is an important parameter to characterize the

Fig. 3: (Colour on-line) The Lyapunov coefficient ( $\Gamma$ ) as a function of frequency in several NGRN chains. In the dimer chain (n = 2): (a) heavy doping; (b) light doping. In the trimer chain (n = 3): (c) heavy doping; (d) light doping. In the 4-mer chain (n = 4): (e) heavy doping and (f) light doping. The parameters in each figure are the same as those in fig. 2. The red dotted curves also give the resonant frequency of phonons vs. the effective potential  $(\alpha_{eff})$ , in order to show that the zero Lyapunov coefficients locate exactly at the resonant frequencies predicted in eqs. (9)–(11).

physical nature of random matrices [17]. In the NGRN system, the Lyapunov coefficient can be expressed as [3,13]

$$\Gamma = \frac{1}{N} \ln(m_{11}^2 + m_{12}^2 + m_{21}^2 + m_{22}^2), \qquad (12)$$

where  $m_{ij}(i, j = 1, 2)$  is the element of the global matrix M. According to the Furstenberg theorem [20], the Lyapunov coefficient exists and converges to its mean value for sufficiently long chains. In fact, in a vibration system, the Lyapunov coefficient is inverse to the localization length of phonons. Once the length of the chain is sufficiently long, zero Lyapunov coefficient corresponds to delocalized states with infinite localized length. Therefore, based on the Lyapunov coefficient, we can obtain the overall behavior of the phonons, *i.e.* we can know whether they are localized or delocalized at specific frequencies in the system. Figure 3 shows the Lyapunov coefficient as



Fig. 4: Thermal conductance  $(\kappa)$  as a function of frequency in several NGRN chains, where the effective potential  $(\alpha_{eff})$ adjusts the thermal conductance. (a) In the *dimer* chain (n = 2and N = 5819):  $m_0 = 2.0$ ,  $m_1 = 2.5$ ,  $\alpha = 1.5$ , and  $\alpha_{eff} = 1.4$ , 1.6, respectively. (b) In the *trimer* chain (n = 3 and N = 8713):  $m_0 = 1.4$ ,  $m_1 = 1.6$ ,  $\alpha = 1.5$ , and  $\alpha_{eff} = 1.0$ , 1.1, respectively. (c) In the 4-mer chain (n = 4 and N = 13923):  $m_0 = 1.3$ ,  $m_1 =$ 1.5,  $\alpha = 1.0$ , and  $\alpha_{eff} = 1.2$ , 1.3, respectively.

a function of the phononic frequency, in several NGRN chains, in the cases of heavy and light doping. It can be seen that around the resonant frequencies in each chain, the Lyapunov coefficient approaches zero even though fluctuation exists (as shown in figs. 3(a)-(f)). Actually, fluctuation can be eliminated when the length of the chain increases. Because the zero Lyapunov coefficient appears at the resonant frequencies, the localized length of phonons is infinite at these frequencies. At these modes, atomic vibration cannot feel "defects" in the chain, and vibration can propagate through the chain without decay. Therefore the delocalization of phonons occurs at each resonant mode in the NGRN chain.

As we know, the thermal conductivity comes from the contribution of different modes, at which phonons can propagate through the whole NGRN chain. The tunable transmission of phonons may lead to a tunable thermal conductivity in the NGRN system. In order to show the contribution of each vibration mode to the thermal conductivity in the NGRN, we try to calculate the thermal conductivity as a function of frequency, which is contributed by all the modes below the frequency [3]. This treatment is similar to evaluate the integrated density of states. Figure 4 shows the calculated thermal conductivity of several NGRN chains, which is tuned by the effective potential  $(\alpha_{eff})$ . It is shown that the thermal conductivity presents a step-like increase in the NGRN chains (as shown in fig. 4(a)-(c)). Around each resonant frequency, the thermal conductivity in the NGRN chain has a jump; while away from the resonant frequency, the thermal conductivity almost remains unchanged. This feature indicates that the localized states merely contribute to the thermal conductivity. Obviously it agrees with the result in a random system in fixed-boundary condition, where the thermal conductivity contributed by the localized states vanishes in a long system [19]. This phenomenon indicates that only the delocalized phonons

can contribute to the thermal conductance, thereafter, the thermal conductance in the NGRN forms a "quantized" feature as frequency increases [13,21]. There exist multiple resonant modes of phonons, consequently, the thermal conductance has multiple jumps and steps in the NGRN system (as shown in fig. 4(b), (c)). Furthermore, it is found that the thermal conductivity is very sensitive to the strength of  $\alpha_{eff}$ . First, the thermal conductivity increases significantly with increasing  $\alpha_{eff}$  (as shown in fig. 4(a)). Secondly, the height of the jump in the thermal conductivity is changed by increasing  $\alpha_{eff}$  (as shown in fig. 4(b)). For example, when  $\alpha_{eff}$  increases slightly from  $\alpha_{eff} = 1.0$ to  $\alpha_{eff} = 1.1$  in the trimer chain, the thermal conductance increases more than 28% (as shown in fig. 4(b)). In the third, both the number and the width of the steps in thermal conductivity can be increased by increasing  $\alpha_{eff}$ . For instance, two steps in thermal conductivity are found when  $\alpha_{eff} = 1.2$ , and three steps are obtained when  $\alpha_{eff} = 1.3$  in the 4-mer chains (as shown in fig. 4(c)). Now that the effective potential  $(\alpha_{eff})$  can be changed by stretching or compressing the NGRN chain, the NGRN chain provides a possible phononic system with tunable phonon resonance and tunable thermal conductance. It is possible to design tunable thermal-conducting materials based on the NGRN systems.

In summary, we theoretically study the phononic transmission and its influence on the thermal conductance in the NGRN system. The weak anharmonic potential can be reduced to the quasi-harmonic form, which is tuned by external stretching. It is shown that due to the delocalization of phonons, resonant transmission is observed and thermal conductance presents a "quantized" feature. Both the number of resonant modes and their locations are controlled by the external stretching or compressing on the NGRN chain. As a consequence, the "quantized" thermal conductance can be tuned in this nonlinear correlated random system. This finding may achieve potential applications in designing novel thermal-conducting materials.

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