Disorder-induced zero-energy spectral singularity for random matrices with correlations

S. N. Taraskin and S. R. Elliott
Department of Chemistry, University of Cambridge, Lensfield Road, Cambridge CB2 1EW, United Kingdom
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A zero-energy midband singularity has been found in the energy spectrum of random matrices with correlations between diagonal and off-diagonal elements typical of vibrational problems. Two representative classes of matrices, characterizing the instantaneous configurations in liquids and mechanically unstable lattices (which mimic the former), have been analyzed. At least for disordered lattice models, the singularity is universal and its origin can be explained within a mean-field treatment.

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Many physical phenomena can be described by the Anderson Hamiltonian with off-diagonal disorder (see, e.g., Ref. 1 and references therein). Stochastic transport,[2] atomic vibrations in disordered structures (see, e.g., Ref. 3 and references therein), and instantaneous normal-mode (INM) analysis in liquids and glasses[4] (called below ''vibrational'' problems) are among these. However, there is one key point which distinguishes vibrational problems from the standard electron problems: there exist strong sum-rule correlations between the off-diagonal and diagonal elements (the sum of all the elements for a particular row is zero) of the relevant random matrices for the vibrational problem.

In different dimensions ($D=1$–3), the energy spectrum of the electron Anderson Hamiltonian with pure off-diagonal disorder defined on the simple (hyper) cubic lattice (but not, as we have checked, on the fcc lattice) exhibits a peculiar feature: a disorder-induced, midband (zero-energy) singularity occurs.[1,5] Normally, disorder smears out sharp features in a spectrum (e.g., van Hove singularities), but in this case it creates a singularity, the origin of which in different dimensions is still controversial. However, can disorder induce a similar (or different) singularity for vibrational problems and, if so, can its origin be revealed? In this paper, we demonstrate the existence of a midband zero-energy singularity in the spectrum of dynamical matrices involved in an INM analysis of a model of a monatomic liquid, and investigate analytically and numerically the nature of this zero-energy singularity in the spectrum of random matrices with sum-rule correlations, appearing in disordered lattices which mimic topologically disordered liquids.

We have observed the occurrence of a singularity by numerical experiment, in which structural models of a liquid with predominantly icosahedral order (similar to those discussed in Ref. 6) and of glassy and liquid silica (similar to those discussed in Ref. 17) have been constructed at different temperatures T by slow molecular dynamics quenches (by constant volume and temperature steps). The INM spectra of the dynamical matrix are shown in Fig. 1. A zero-energy singularity is clearly present in the energy spectrum $g(\varepsilon)$ for the icosahedral liquid and possibly in silica [at least, the tendency of the change of the density of states (DOS) around zero is consistent with the model presented below]. Similar zero-energy peaks have been found *but not explained* in liquid theories[7,8] and in simulations of a binary Lennard-Jones liquid.[9] Normally, the DOS in the frequency domain, $g_\omega = 2\sqrt{|\varepsilon|}g(\varepsilon)$, is of interest in the INM analysis, but the singularity is masked there by the linear factor 2ω, and that is, probably, why it has not been carefully studied before.

Therefore, there exists a peculiarity in the spectrum of random matrices with sum-rule correlations (instantaneous dynamical matrices) and its origin can be revealed analytically as follows. Consider a Hamiltonian describing both the electron and vibrational problems:

$$\hat{\mathbf{H}}=\sum_i \left(\varepsilon_i-\gamma\sum_{j\neq i} t_{ij}\right)|i\rangle\langle i|+\sum_{i,j\neq i} t_{ij}|i\rangle\langle j|, \qquad (1)$$

where ε_i and t_{ij} stand for the random on-site energies and the random transfer integrals between sites i and j, respectively. The parameter γ controls the correlations between diagonal and off-diagonal matrix elements. The standard electron Anderson Hamiltonian with pure off-diagonal disorder corresponds to $\gamma=0$ and $\varepsilon_i=0$, with t_{ij} being random variables taken, for example, from a uniform (box) distribution of width 2Δ centered around $t_0=-1$, $t_{ij}\in[t_0-\Delta,t_0+\Delta]$. The vibrational (scalar) problem corresponds to $\gamma=1$ with the other parameters being the same. For atomic vibra-

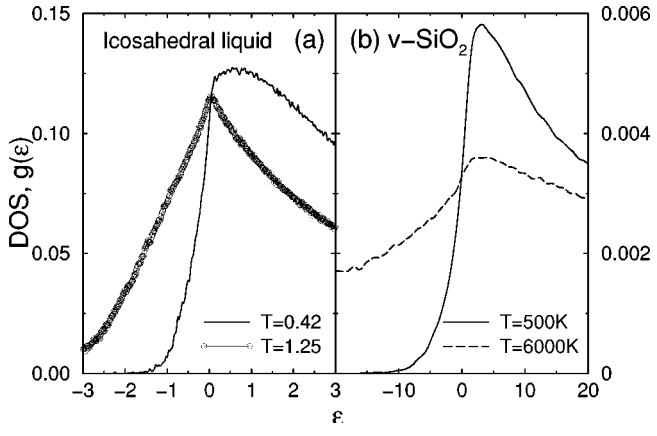

FIG. 1. The energy spectrum $g(\varepsilon)$ for the instantaneous dynamical matrices for an INM analysis of (a) a liquid with predominantly icosahedral order (1620-particle model averaged over 90 configurations) at two temperatures, as marked (the units are the same as in Ref. 6) and of (b) glassy (the solid curve) and liquid (the dashed curve) silica (1650-particle model averaged over 20 configurations). The energy units for silica are THz^2.

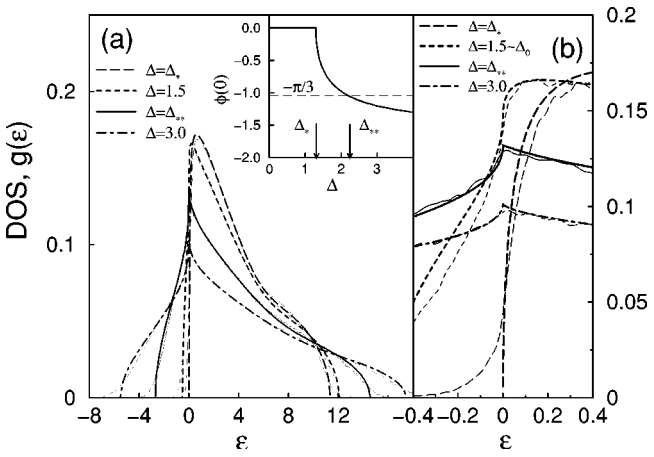

FIG. 2. (a) Evolution of the zero-energy singularity in the DOS for a disordered fcc lattice with various values of disorder Δ ($\Delta_* \simeq 1.3$ and $\Delta_{**} \simeq 2.2$). The thick curves were obtained by the numerical integration of the CPA DOS given by Eq. (2), while the thin ones are the results of precise numerical KPM solution for vector vibrations in a $130\times130\times130$ site fcc lattice. The difference between the CPA and KPM curves around both band edges is due to the known failure of CPA to reproduce the localized band tails. The inset shows the phase $\phi(0)$ of the effective field at zero energy vs disorder Δ. The critical values of disorder are indicated by arrows. (b) An enlargement of the zero-energy region. The CPA results obtained by use of Eqs. (2) and (4) are not distinguishable on this energy scale.

tions, the Hamiltonian corresponds to the dynamical operator, transfer integrals to the force constants and energy to the squared frequency, $\varepsilon=\omega^2$.[3]

We have calculated by the kernel polynomial method[10] (KPM) the spectrum of the ''vibrational'' Hamiltonian (1) (see the thin lines in Fig. 2) for $\gamma=1$ and $\varepsilon_i=0$, defined on the fcc lattice (actually, of its more general version for vector vibrations; see Ref. 3 for more detail). For sufficiently large degrees of disorder, $\Delta>\Delta_0$ ($\Delta_0\simeq1.4$–1.5), the zero-energy singularity is evident (Fig. 2). Moreover, the shape of the singularity is similar to that found for topologically disordered models (Fig. 1), indicating its possible universality (see below). We have also calculated the DOS for the same problem within a mean-field approach [the single-bond coherent potential approximation[2] (CPA)], and found remarkably good agreement with the precise numerical (KPM) results (cf. thick and thin lines in Fig. 2) in the singularity region for $\Delta\gtrsim\Delta_0$ [$(\Delta_0-\Delta_*)/\Delta_*\ll1$; see below for a discussion of Δ_*], i.e., when the localization threshold is far enough below zero energy. This agreement is surprising (at first sight), because it is commonly believed (see, e.g., Ref. 11) that mean-field theories fail to reproduce sharp features in a spectrum. For example, the most successful homomorphic cluster CPA (Refs. 12 and 13) well reproduces the whole spectrum for the electron problem with pure off-diagonal disorder ($\gamma=0$), except for the zero-energy singularity. Below, we explain why the CPA reproduces the zero-energy singularity for vibrational problems but not for the electron one, and we use this insight to reveal the physical origin of the singularity.

The zero-energy singularity occurs for $\gamma=1$ and disappears if $\gamma\neq1$, i.e., when the exact sum-rule correlations of elements in the dynamical matrix break down. The functional form of the zero-energy singularity observed is universal (i.e. independent of the type of distribution of the force constants, the reference lattice symmetry, scalar or vector type of vibrations, etc.) for the class of Hamiltonians given by Eq. (1) with $\gamma=1$ and $\varepsilon_i=0$, and depends only on the dimensionality of the problem. For sufficiently large disorder, the (parity-independent) singularity occurs in the mid-band region of the spectrum, far from the localized states in the three-dimensional (3D) case (as checked by multifractal analysis) and in the range of prelocalized states for lower dimensions. The analytic mean-field solution shows that its appearance is dictated by the universal nonanalytic behavior of the spectral-density operator in the plane-wave basis and, essentially, is a consequence of the fact that zero-energy (long-wavelength) plane waves contribute anomalously (but not solely) to the disordered eigenstates with energies around zero.

The DOS $g(\varepsilon)$ for a disordered lattice is the trace of the spectral-density operator $\hat{\mathbf{A}}(\varepsilon)=\langle\delta(\varepsilon-\hat{\mathbf{H}})\rangle$ taken in the convenient orthonormal basis of crystalline eigenstates $|\mathbf{k},\beta\rangle$ ($\mathbf{k}$ is the wave vector of a plane wave from the branch β) of the same Hamiltonian, but without disorder ($\Delta=0$) and averaged ($\langle\cdots\rangle$) over the distribution of random variables t_{ij}, $g(\varepsilon)=\int\langle\langle\mathbf{k},\beta|\hat{\mathbf{A}}(\varepsilon)|\mathbf{k},\beta\rangle\rangle g^{\mathrm{cryst}}(\varepsilon_{\mathbf{k}\beta})\mathrm{d}\varepsilon_{\mathbf{k}\beta}$, with $g^{\mathrm{cryst}}(\varepsilon_{\mathbf{k}\beta})$ being the crystalline vibrational DOS. The diagonal matrix element of the spectral-density operator, $A(\varepsilon,\varepsilon_{\mathbf{k}\beta})\equiv\langle\mathbf{k},\beta|\hat{\mathbf{A}}(\varepsilon)|\mathbf{k},\beta\rangle$, can be found within the CPA (Refs. 3 and 14–16) via the complex effective interaction field $z(\varepsilon)=z'(\varepsilon)+\mathrm{i}z''(\varepsilon)$, so that

$$g(\varepsilon)=-\frac{1}{\pi}\int\frac{(\varepsilon_{\mathbf{k}\beta}/\varepsilon)\Gamma(\varepsilon)}{[\varepsilon_{\mathbf{k}\beta}-\overline{\varepsilon}(\varepsilon)]^2+\Gamma^2(\varepsilon)}g^{\mathrm{cryst}}(\varepsilon_{\mathbf{k}\beta})\mathrm{d}\varepsilon_{\mathbf{k}\beta}, \tag{2}$$

with

$$\overline{\varepsilon}(\varepsilon)=\varepsilon z'(\varepsilon)/|z(\varepsilon)|^2,\quad \Gamma(\varepsilon)=-\varepsilon z''(\varepsilon)/|z(\varepsilon)|^2. \tag{3}$$

The effective interaction field $z(\varepsilon)$ is found from the solution of the self-consistent CPA equation (see Ref. 3). The real part $z'(\varepsilon)$ fluctuates around its crystalline value $z'_0=1$, and the imaginary part is smooth and negative in the region of the nonvanishing disordered spectrum, and zero otherwise. The spectral density $\langle A(\varepsilon,\varepsilon_{\mathbf{k}\beta})\rangle$ as a function of $\varepsilon_{\mathbf{k}\beta}$ has the shape of a peak characterized by a width parameter $\Gamma(\varepsilon)$ located at an energy $\varepsilon_{\mathrm{peak}}=|\varepsilon||z(\varepsilon)|^{-1}=[\overline{\varepsilon}(\varepsilon)+\Gamma(\varepsilon)]^{1/2}$ (see Fig. 3).

As follows from Eq. (2), the disordered DOS is generically related to the crystalline DOS for a reference system convolved with the peak-shaped spectral density, $\langle A(\varepsilon,\varepsilon_{\mathbf{k}\beta})\rangle$. For small disorder ($\Delta\to0$), the spectral density is very narrow and close in functional form to $\delta(\varepsilon-\varepsilon_{\mathbf{k}\beta})$, so that the disordered DOS strongly resembles its crystalline counterpart. With increasing disorder but for $\Delta\leqslant\Delta_*$ (where the spectrum of the Hamiltonian is still non-negative and the

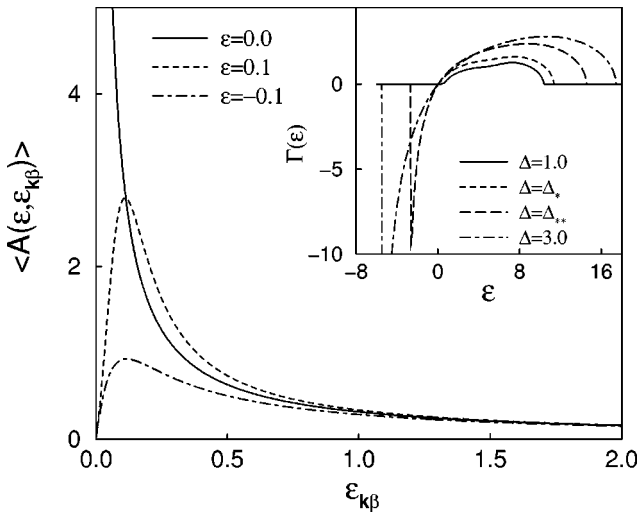

FIG. 3. Dependence of the spectral density $\langle A(\varepsilon,\varepsilon_{\mathbf{k}\beta})\rangle$ on the crystalline energy $\varepsilon_{\mathbf{k}\beta}$ at values of disordered energy ε as marked (for $\Delta=\Delta_{**}$), for vector vibrations in the force-constant disordered fcc lattice. The inset shows the energy dependence of the spectral-density peak width parameter, $\Gamma(\varepsilon)$ [Eq. (3)], for different values of disorder, as marked.

lattice is mechanically stable[3]), the spectral-density peak becomes broader and washes out all the van Hove singularities in the crystalline spectrum, except the boundary singularity around the lowest band edge, $\varepsilon_{\min}=0$.

When the disorder exceeds the critical CPA value, $\Delta>\Delta_*$, the number of negative force constants becomes so large that the system is no longer mechanically stable even in the mean-field limit, and negative eigenvalues appear in the spectrum (as in instantaneous configurations of liquids[4]); i.e., the lower boundary of the CPA spectrum moves below zero energy, $\varepsilon_{\min}<0$. Exactly in this regime, the midband zero-energy singularity in the disordered CPA spectrum evolves. Indeed, if we look at the evolution of the spectral-density width parameter $\Gamma(\varepsilon)$ with increasing disorder (see the inset in Fig. 3), we can clearly see that the peak width is still zero at zero energy, $\Gamma(0)=0$, being a consequence of Eq. (3), even though the value of the effective field becomes finite at this point $[z''(0)\neq 0]$. This means that the spectral density $\langle A(\varepsilon=0,\varepsilon_{\mathbf{k}\beta})\rangle$ as a function of the crystalline energy $\varepsilon_{\mathbf{k}\beta}$ has a singularity at $\varepsilon_{\mathbf{k}\beta}=0$ (see Fig. 3). The zero-energy point, in this regime ($\Delta>\Delta_*$), belongs to the midband region. The finite value of $z''(0)$ immediately gives rise to a different singular shape of the spectral density, $\langle A(0,\varepsilon_{\mathbf{k}\beta})\rangle \simeq -\pi^{-1}[z''(0)/|z(0)|^2]\varepsilon_{\mathbf{k}\beta}^{-1}$ (see Fig. 3), as compared to the δ-functional shape of the spectral density for the $\Delta<\Delta_*$ regime, where both $\Gamma(0)$ and $z''(0)$ are zero. The δ-functional shape of the spectral density reproduces the crystalline van Hove singularity, $g(\varepsilon)\propto\varepsilon^{(D/2)-1}$, at the lower band edge, but the $\varepsilon_{\mathbf{k}\beta}^{-1}$ shape of the spectral density at $\varepsilon=0$, convolved with the crystalline DOS [see Eq. (2)], results in a singularity of a different type in the disordered DOS which is not related to the crystalline van Hove singularity at $\varepsilon=0$ (see below).

The shape of the midband zero-energy singularity can be obtained analytically within the CPA by splitting the integration region in Eq. (2) into a low-energy part, $\varepsilon_{\mathbf{k}\beta}<\varepsilon_0$, where $g^{\text{cryst}}(\varepsilon)\simeq\chi_D\varepsilon^{(D/2)-1}$ (the Debye law), and the rest of the band, $\varepsilon_{\mathbf{k}\beta}>\varepsilon_0$ (irrelevant for the shape of the singularity). The final result for the disordered DOS (3D case) in the limit $|\varepsilon|\to 0$ is the following:

$$g(\varepsilon)\simeq -\frac{C\sin\phi(\varepsilon)}{\pi|z(\varepsilon)|}+\frac{\chi_3|\varepsilon|^{1/2}}{|z(\varepsilon)|^{3/2}}\sin\left[\frac{3\phi(\varepsilon)}{2}+\theta(\varepsilon)\frac{\pi}{2}\right], \tag{4}$$

where $C=\int[g^{\text{cryst}}(\varepsilon)/\varepsilon]d\varepsilon$ is a model-dependent constant, $\phi(\varepsilon)\equiv\arg[z(\varepsilon)]$ is the phase of the effective field, and $\theta(\varepsilon)$ is the Heaviside function, $\theta(\varepsilon<0)=0$ and $\theta(\varepsilon>0)=1$. Bearing in mind that the effective field is a smooth function of energy around zero, we can conclude from Eq. (4) that, in the 3D case, the disordered DOS is a continuous function around zero but its derivative shows $\varepsilon^{-1/2}$ singular behavior (see Figs. 1 and 2).

The expression (4) for $g(\varepsilon)$ has been obtained in the limit $|\varepsilon|\to 0$ for any degree of disorder. The evolution of the singularity with disorder (see Fig. 2) is determined by the dependence on Δ of the effective field at zero energy. The phase $\phi(0;\Delta)$ varies in a critical manner with disorder [see the inset in Fig. 2(a)], being zero below the critical disorder, $\Delta\leqslant\Delta_*$, when the zero-energy singularity is just a lower band-edge van Hove singularity. Above the critical disorder, $\Delta>\Delta_*$, the derivative of the DOS becomes singular from both sides about $\varepsilon=0$. If the disorder is not too large, $\Delta_*<\Delta<\Delta_{**}$, the sign of the derivative is positive on both sides of the singularity and the DOS is monotonic in the singularity range [see the upper curve in Fig. 2(b)]. For higher disorder, $\Delta>\Delta_{**}$, the derivative changes sign at the singularity and the disordered DOS exhibits a sharp maximum at zero energy [see the lower curve in Fig. 2(b)]. The characteristic value of disorder, $\Delta=\Delta_{**}$, at which such a transformation of the shape of the singularity occurs (the solid line in Fig. 2) can be found from the solution of the equation $\phi(0;\Delta_{**})=-\pi/3$ [see the inset in Fig. 2(a)], resulting from the condition $g'(\varepsilon\to 0+)=0$.

The physical significance of this transition is related to the fact that, at $\Delta\simeq\Delta_{**}$, the peak width of the spectral density around the singularity becomes comparable with the peak position. This means that Δ_{**} corresponds to the Ioffe-Regel crossover for the propagation of plane waves characterized by the energies $\varepsilon_{\mathbf{k}\beta}\to 0$.[17] In other words, for $\Delta\lesssim\Delta_{**}$, there is a finite low-energy interval of the weak-scattering regime for plane-wave propagation, but for $\Delta\gtrsim\Delta_{**}$, all plane waves propagate in the regime of strong scattering.

Similar CPA analyses can be performed for lower dimensions, and they result in zero-energy singularities as well. In 2D (for $\Delta>\Delta_*$), a logarithmic singularity $g(|\varepsilon|\to 0)\simeq\pi^{-1}[z''(\varepsilon)/|z(\varepsilon)|^2]\chi_2\ln|\varepsilon|$ evolves on the background of the van Hove band-edge singularity. In 1D, the divergence of the disordered DOS from both sides of zero energy is even more pronounced: $g(\varepsilon)\simeq-\chi_1|\varepsilon|^{-1/2}|z(\varepsilon)|^{-1/2}\sin[\phi(\varepsilon)/2-\theta(\varepsilon)\pi/2]$.

The link between lattice models and topologically disordered liquids and glasses is, of course, not straightforward. The above mean-field analysis for disordered lattices uses the existence of an orthonormal plane-wave basis, which is absent in topologically disordered systems. Nevertheless, an orthogonalized basis resembling a plane-wave basis, at least in the region of zero energy, can readily be constructed,[17] so that it might be expected that the same universal singular behavior of the DOS around $\varepsilon=0$ should also occur for topologically disordered systems. Indeed, the singularity is evident for a model liquid with predominantly icosahedral order [Fig. 1(a)], and it is remarkable that the evolution of the shape of this singularity with decreasing temperature mirrors that found in lattice models with decreasing force-constant disorder [Fig. 2(b)]. Both regimes of the singular behavior are evident from the numerical experiment [Fig. 1(a)]: (i) the high-temperature liquid state (circles) can be characterized by relatively large disorder ($\Delta \gtrsim \Delta_{**}$); (ii) the state just above the glass-transition temperature (solid curve) can be associated with the intermediate disorder regime ($\Delta_{*} \lesssim \Delta \lesssim \Delta_{**}$). The situation for vitreous silica [Fig. 1(b)] is different. At both temperatures, below and above the glass transition, the models stay in the regime $\Delta \lesssim \Delta_{**}$, which is not surprising because silica is a very strong glass-forming liquid [especially in comparison with the fragile system shown in Fig. 1(a)], and is characterized by well-defined and stable local tetrahedral order even at very high temperatures (this means that the disorder, i.e., Δ, is relatively small). A more detailed analysis (including the glass-transition region) will be presented elsewhere.

In conclusion, we have demonstrated the presence of a midband zero-energy singularity in the spectrum of the instantaneous dynamical matrices of topologically disordered structural models of liquids and of the dynamical matrices of disordered lattices which well mimic the former. The presence of exact sum-rule correlations between the diagonal and off-diagonal elements in the disordered dynamical matrix causes this singularity. The shape of the disorder-induced singularity, at least in lattice models, is universal and depends only on the dimensionality of the model. Such a universality is related to the universal $\varepsilon_{\mathbf{k}\beta}^{-1}$-behavior of the spectral density in the plane-wave basis, which is due to the multiplicative nature of the effective interaction mean field [the mean-field energy of the quasiparticles, $\tilde{\varepsilon}_{\mathbf{k}\beta}$, is the product of the dimensionless effective interaction $z(\varepsilon)$ and the bare crystalline energy $\varepsilon_{\mathbf{k}\beta}$, i.e., $\tilde{\varepsilon}_{\mathbf{k}\beta}=z(\varepsilon)\varepsilon_{\mathbf{k}\beta}$]. This property distinguishes the above class of Hamiltonians from those with pure on-site energy disorder, characterized by an additive effective field,[14] which do not exhibit a zero-energy singularity. Thus, the failure of the homomorphic effective field (containing both additive and multiplicative contributions) in reproducing the zero-energy singularity for the Anderson Hamiltonian with pure off-diagonal disorder without correlations in simple cubic lattices can be explained. A successful effective field for such a problem should not contain the additive part, at least at zero energy, but its construction is still an open question.

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