Polarizability fluctuations in dielectric materials with quenched disorder

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(Received 2 May 2000)

We study a model of dielectric response for spatially disordered materials. In this model the local polarizability $\alpha_{\mathbf{r}}$ is a quenched random variable. From a one-loop level renormalization-group analysis, we predict that with increasing length scale L, the dimensionless fluctuation strength $\overline{\alpha}\sigma$, where $1/\overline{\alpha}$ and σ^2 are the average and the variance of the distribution for $1/\alpha_{\mathbf{r}}$, decays as $1/L^2$ universally at large length scales. The interplay of the random polarizability and the long-range dipole-dipole interaction is discussed.

PACS number(s): 05.50.+q, 77.22.−d, 05.40.−a

Polarization fluctuations significantly influence electron transfer, energy transfer, and solvation dynamics in dielectric materials. For simple polar solvents, both static and dynamic manifestations of these fluctuations can be reasonably treated in terms of simple generalizations of dielectric continuum theory. In these approaches, the macroscopic frequency-dependent dielectric constant is related to a *homogeneous* local polarizability by the Clausius-Mossotti equation [1–3]. For the spatially disordered or inhomogeneous systems such as proteins and zeolites, however, this approach is no longer valid at length scales comparable to the characteristic length of the inhomogeneity. For example, a particular protein environment contains specific regions of high polarity and low polarity. Thus, the dielectric response varies significantly from one region to another in such systems. On the other hand, when considering length scales much larger than the inhomogeneity length, the effects of disorder are negligible, and the homogeneous or continuum model is valid. This paper is concerned with the approach to this continuum limit, analyzing how the effective inhomogeneity changes as a function of length scale.

The particular model we consider is the disordered dielectric described in Ref. [4]. The local polarizability is random and the randomness is ''quenched'' in the sense that its relaxation is ignored. Inhomogeneity is thus characterized by the disorder length scale and strength. The former characterizes the density of disorder, and the latter is essentially the variance of the local polarizability distribution. In Ref. [4] we treated the dielectric response of this model with a plausible but approximate real-space renormalization procedure. While the procedure seems to have general applicability, its accuracy remains to be established. This paper takes a step in that direction. An important result of Ref. [4] is the prediction of nontrivial scaling in the approach to the continuum limit. Here, we consider this scaling again, but with the aid of field theoretic renormalization-group techniques, specifically with replicas [5] and the $\epsilon=4-d$ expansion in d dimensions ($d=3$ in our case) [6]. We obtain the recursion equations of the polarizability fluctuation for general dipole-dipole interaction strength and disorder strength. The dimensionless fluctuation strength $\overline{\alpha}\sigma$ decays as $L^{-\nu}$ when increasing the length scale L, where $1/\overline{\alpha}$ and σ^2 are the average and the variance of the $1/\alpha_{\mathbf{r}}$ distribution, and $\alpha_{\mathbf{r}}$ is the polarizability at position $\mathbf{r}$. At the one-loop level in perturbation theory for the self-energy [7], we show that ν varies from $\nu=3/2$, valid at small L, to $\nu=2$, valid at large L. The former, $\nu=3/2$, is the ''randomness dominated'' result. It follows from uncorrelated polarizability statistics. The latter, $\nu=2$, is the ''interaction-dominated'' result, which is determined by the new fixed point due to the dipole-dipole interaction.

We begin with the discrete model, which is defined on a cubic lattice [4]. The unit cell with $L=1$ has size a. The Hamiltonian reads

$$H=\frac{1}{2}\sum_{\mathbf{r}}\frac{\mathbf{m}_{\mathbf{r}}^2}{\alpha_{\mathbf{r}}}-\frac{1}{2}\sum_{\mathbf{r}\neq\mathbf{r}'}\mathbf{m}_{\mathbf{r}}\cdot\mathbf{T}_{\mathbf{r}\mathbf{r}'}\cdot\mathbf{m}_{\mathbf{r}'}, \qquad (1)$$

where $\mathbf{m}_{\mathbf{r}}$ is the polarizable dipole moment in the cell at point $\mathbf{r}$, $\alpha_{\mathbf{r}}$ is the polarizability at that cell, and $\mathbf{T}_{\mathbf{r}\mathbf{r}'}$ is the 3×3 dipole-dipole tensor. In the continuum limit, $a\to0^+$,

$$\mathbf{T}_{\mathbf{r}\mathbf{r}'}\to3\frac{(\mathbf{r}-\mathbf{r}')(\mathbf{r}-\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|^5}-\frac{\mathbf{I}}{|\mathbf{r}-\mathbf{r}'|^3}.$$

The set of local polarizabilities, $\{\alpha_{\mathbf{r}}\}$, is chosen at random for each realization of the system. The distribution of $\alpha_{\mathbf{r}}$ is quenched or ''frozen.'' In general, this distribution should be chosen to imitate the actual physical system of interest. In the simplest case considered here, we assume that the distribution is isotropic, we ignore spatial correlations, and for the purpose of being concrete we assume each $\alpha_{\mathbf{r}}$ can have only one of two values. As such, the distribution is characterized by the average $\langle 1/\alpha_{\mathbf{r}}\rangle\equiv1/\overline{\alpha}$ and the variance $\langle(1/\alpha_{\mathbf{r}})^2\rangle-\langle1/\alpha_{\mathbf{r}}\rangle^2\equiv\sigma^2$. The characteristic dimensionless fluctuation strength is $\chi\equiv\overline{\alpha}\sigma$. For this model, we address the following question: when we increase the length scale to make every unit cell larger and larger, if we still use Hamiltonian (1) to describe the dielectric response of the system with a new length scale, how does the distribution of $\alpha_{\mathbf{r}}$ evolve? Specifically, how does the dimensionless variance of this distribution χ change as increasing the length scale?

It is instructive to first answer this question in the trivial case where the dipole-dipole tensor in Eq. (1) is set to zero. Let H_{t} denote the resulting Hamiltonian. It describes noninteracting dipoles with random polarizability. Since each random polarizability is independent of each other, $\chi(L)$ will scale as $L^{-3/2}$. Thus, in this case, $\chi(2)=\chi(1)2^{-3/2}$. We can derive this expected result by studying how the effective Hamiltonian evolves with increasing the length scale. We define $\tilde{\mathbf{m}}_{\mathbf{r}}=\Sigma_{i=1}^{N}\mathbf{m}_{i\mathbf{r}}$ as the coarse-grained dipole moment for a cell with lattice size $L=2$. Here, there are $N=2^3$ of the original unit cells in this larger cell centered at $\mathbf{r}$, and $\mathbf{m}_{i\mathbf{r}}$ is the dipole of the ith unit cell in this larger cell. The partition function at $L=2$, $Z_2=\int\mathcal{D}\tilde{\mathbf{m}}_{\mathbf{r}}\exp(-\tilde{H}_t/k_BT)$, should be the same as that at $L=1$, $Z_1=\int\mathcal{D}\mathbf{m}_{\mathbf{r}}\exp(-H_t/k_BT)$. Both can be calculated assuming σ^2 is a small and keeping only the leading term in σ^2. Comparing both estimates thereby relates $\overline{\alpha}(2)$ and $\sigma^2(2)$ to $\overline{\alpha}(1)$ and $\sigma^2(1)$. The result of this straightforward calculation is $\overline{\alpha}(2)=2^3\overline{\alpha}(1)$ and $\sigma^2(2)=\sigma^2(1)/2^9$, giving the expected result $\chi(2)=\chi(1)2^{-3/2}$.

More generally, when the dipole-dipole interactions are not omitted from the Hamiltonian (1), we can analyze the scaling with a well-known renormalization-group approach. This approach requires that we work in a general d-dimensional space. The scaling of $\overline{\alpha}$ follows from $\overline{\alpha}=\Sigma_{\mathbf{r},\mathbf{r}'}\langle\tilde{\mathbf{m}}_{\mathbf{r}}\cdot\tilde{\mathbf{m}}_{\mathbf{r}'}\rangle/\mathcal{N}$ and the observation that spatial self-similarity requires $\langle\tilde{\mathbf{m}}_{\mathbf{r}}\cdot\tilde{\mathbf{m}}_{\mathbf{r}'}\rangle\sim|\mathbf{r}-\mathbf{r}'|^{-(d-2+\eta)}$. Here, $\langle\cdots\rangle$ denotes a thermal average, $\mathcal{N}$ is the number of the unit cells in the whole system, i.e., $\mathcal{N}(L)=\mathcal{N}(1)L^{-d}$, and η is the anomalous dimension [11]. Dimensional analysis thus shows that $\overline{\alpha}(1)=L^{2-\eta}\overline{\alpha}(L)$. As such,

$$\chi(L)=L^{-2+\eta}\overline{\alpha}(1)\sigma(L). \tag{2}$$

Once we find the recursion equation for σ, the flow behavior of $\chi(L)$ can be obtained.

To find this equation, we use the replica technique to transform the Hamiltonian with quenched random variables into a translational invariant one with no random variables [5]. In particular, averaging the logarithm of the partition function Z over the probability distribution of $\alpha_{\mathbf{r}}$, together with the identity $\log Z=\lim_{n\to 0}(Z^n-1)/n$, leads us to consider an effective Hamiltonian,

$$H_{\mathrm{eff}}=\frac{1}{2}\sum_{\mathbf{r},\mu}\frac{\mathbf{m}_{\mathbf{r}}^{(\mu)}\cdot\mathbf{m}_{\mathbf{r}}^{(\mu)}}{\overline{\alpha}}-\frac{1}{2}\sum_{\mathbf{r}\neq\mathbf{r}',\mu}\mathbf{m}_{\mathbf{r}}^{(\mu)}\cdot\mathbf{T}'_{\mathbf{rr}}\cdot\mathbf{m}_{\mathbf{r}}^{(\mu)}$$
$$-\frac{1}{8k_BT}\sum_{\mathbf{r},\mu,\nu}\sigma^2\mathbf{m}_{\mathbf{r}}^{(\mu)}\cdot\mathbf{m}_{\mathbf{r}}^{(\mu)}\mathbf{m}_{\mathbf{r}}^{(\nu)}\cdot\mathbf{m}_{\mathbf{r}}^{(\nu)}+\ \ldots\ . \tag{3}$$

Here μ and ν are replica indices summed from 1 to n, k_{B} is the Boltzmann constant, T is temperature. The replica dipole moment variable $\mathbf{m}_{\mathbf{r}}^{(\mu)}$ has $3n$ components. Subsequent terms not exhibited in Eq. (3) involve higher orders of $\mathbf{m}_{\mathbf{r}}^{(\mu)}$ than fourth order, which are irrelevant operators since, according to the dimensional analysis, the coupling constants in these terms have negative momentum dimensions, and have no effect on the renormalization of the system [7]. To study properties of disordered systems, the calculation for a general number of replicas, n, is carried out, and then the limit $n\to 0$ is evaluated [5,8–10]. This replica representation can be applied at different length scales L, in which case the coefficients $1/\overline{\alpha}$ and σ in Hamiltonian (3) are functions of L.

With Fourier transforms, we can rewrite and generalize the Hamiltonian (3) as [10,12,13].

$$H_{\mathrm{eff}}=H_0+H_{\mathrm{int}}, \tag{4}$$

$$\frac{H_0}{k_BT}=-\frac{1}{2}\int_q\left[(r_0+q^2)\delta^{ij}+g_0\frac{q^iq^j}{q^2}\right]\phi_\alpha^i(q)\phi_\alpha^j(-q), \tag{5}$$

$$\frac{H_{\mathrm{int}}}{k_BT}=-\frac{\kappa^\epsilon}{4!}\int_{q_1}\int_{q_2}\int_{q_3}\int_{q_4}(v_0F_{\alpha\beta\gamma\delta}^{ijkl}+u_0S_{\alpha\beta\gamma\delta}^{ijkl}+2w_0T_{\alpha\beta\gamma\delta}^{ijkl})$$
$$\times\phi_\alpha^i(q_1)\phi_\beta^j(q_2)\phi_\gamma^k(q_3)\phi_\delta^l(q_4)\delta\left(\sum_i^4 q_i\right), \tag{6}$$

where $\phi_\alpha^i(q)$ denotes the ith Cartesian component of the Fourier transform of $\mathbf{m}_{\mathbf{r}}^{(\alpha)}$, $\int_q$ denotes the integration $\int d^dq/(2\pi)^d$ truncated at a large wave-vector cutoff, $|q|\leqslant q_c\sim 1/a$, and κ has the dimension of momentum. The quantity g_0 is the bare strength of the dipole-dipole interaction, r_0 is the bare ''mass,'' and $u_0=-3\sigma^2(1)/(k_BT)^2$. The u_0 term comes directly from the basic model, Eq. (3). We also include the other two terms with coefficients v_0 and w_0, since these terms will be generated by subsequent iterations. Repeated subscript and superscript indices are to be summed, and the tensors $F_{\alpha\beta\gamma\delta}^{ijkl}$, $S_{\alpha\beta\gamma\delta}^{ijkl}$, and $T_{\alpha\beta\gamma\delta}^{ijkl}$ are given in terms of Kronecker deltas $\delta_{\alpha\beta}$ and δ^{ij} as follows:

$$F_{\alpha\beta\gamma\delta}^{ijkl}=\frac{1}{3}(\delta^{ij}\delta^{kl}+\delta^{ik}\delta^{jl}+\delta^{il}\delta^{jk})\delta_{\alpha\beta}\delta_{\beta\gamma}\delta_{\gamma\delta},$$

$$S_{\alpha\beta\gamma\delta}^{ijkl}=\frac{1}{3}(\delta^{ij}\delta^{kl}\delta_{\alpha\beta}\delta_{\gamma\delta}+\delta^{ik}\delta^{jl}\delta_{\alpha\gamma}\delta_{\beta\delta}+\delta^{il}\delta^{jk}\delta_{\alpha\delta}\delta_{\beta\gamma}),$$

$$T_{\alpha\beta\gamma\delta}^{ijkl}=\frac{1}{3}\left(\delta_{\alpha\beta}\delta_{\gamma\delta}\frac{\delta^{ik}\delta^{jl}+\delta^{il}\delta^{jk}}{2}+\delta_{\alpha\gamma}\delta_{\beta\delta}\frac{\delta^{ij}\delta^{kl}+\delta^{il}\delta^{jk}}{2}\right.$$
$$\left.+\delta_{\alpha\delta}\delta_{\beta\gamma}\frac{\delta^{ij}\delta^{kl}+\delta^{ik}\delta^{jl}}{2}\right).$$

In this formulation, the unperturbed or reference Green's function is $G_{\alpha\beta}^{ij}(q)=\langle\phi_\alpha^i(q)\phi_\beta^j(-q)\rangle_0$, where $\langle\cdots\rangle_0$ denotes the average with the weight functional $\propto\exp(-H_0/k_BT)$. Because of the long-range interaction, the Green's function has longitudinal and transverse components,

$$G_{\alpha\beta}^{ij}(q)=[G_0^L(r_0,g_0,q)P_L^{ij}+G_0^T(r_0,q)P_T^{ij}]\delta_{\alpha\beta},$$

where $G_0^L=1/(r_0+g_0+q^2)$, $G_0^T=1/(r_0+q^2)$, and $P_T^{ij}=\delta^{ij}-q^iq^j/q^2$ and $P_L^{ij}=q^iq^j/q^2$ are the projection operators for transverse and longitudinal components, respectively.

The renormalization behavior of this model is controled by the Callan-Symanzik equations [15,7], which can be studied most conveniently by using the dimensional regularization and minimal subtraction procedure of 't Hooft and Veltman [16]. We use this procedure and notation as outlined, for example, in Amit's text [7]. In particular, we use the ϵ expansion to obtain the β functions and renormalization constants up to one-loop level. The singular part of the one-loop diagram, $\int_k G^{ij}_{\alpha\beta}(k) G^{lm}_{\gamma\delta}(k+p)$, at the symmetry point ($p^2 = \kappa^2$) is $\delta_{\alpha\beta}\delta_{\gamma\delta}J^{ijlm}$, where

$$J^{ijlm} = \left[\frac{1}{\epsilon} - \frac{11}{48}\ln(1+g_0/\kappa^2)\right]\delta^{ij}\delta^{lm} + \frac{1}{48}\ln(1+g_0/\kappa^2) \times(\delta^{il}\delta^{jm} + \delta^{im}\delta^{jl}).$$

From this result and the renormalization condition at the symmetry point, where the four-point vertex function [7] is $\kappa^{\epsilon}(vF^{ijkl}_{\alpha\beta\gamma\delta} + uS^{ijkl}_{\alpha\beta\gamma\delta} + 2wT^{ijkl}_{\alpha\beta\gamma\delta})$, we obtain

$$\beta_v \equiv \left(\kappa\frac{\partial}{\partial\kappa}v\right)\bigg|_0 = -\epsilon v + \left(2 - \frac{7}{12}\frac{1}{1+\kappa^2/g}\right)v^2 + \left(2 - \frac{2}{3}\frac{1}{1+\kappa^2/g}\right)vu + \left(6 - \frac{5}{3}\frac{1}{1+\kappa^2/g}\right)vw, \tag{7}$$

$$\beta_u \equiv \left(\kappa\frac{\partial}{\partial\kappa}u\right)\bigg|_0 = -\epsilon u + \left(\frac{4}{3} - \frac{17}{36}\frac{1}{1+\kappa^2/g}\right)u^2 + \left(2 - \frac{1}{2}\frac{1}{1+\kappa^2/g}\right)vu + \left(\frac{10}{3} - \frac{13}{18}\frac{1}{1+\kappa^2/g}\right)uw + \left(\frac{2}{3} - \frac{1}{9}\frac{1}{1+\kappa^2/g}\right)vw + \left(2 - \frac{7}{12}\frac{1}{1+\kappa^2/g}\right)w^2, \tag{8}$$

$$\beta_w \equiv \left(\kappa\frac{\partial}{\partial\kappa}w\right)\bigg|_0 = -\epsilon w + \left(\frac{8}{3} - \frac{9}{12}\frac{1}{1+\kappa^2/g}\right)w^2 + \left(2 - \frac{13}{18}\frac{1}{1+\kappa^2/g}\right)uw + \left(\frac{2}{3} - \frac{5}{18}\frac{1}{1+\kappa^2/g}\right)vw + \frac{1}{36}\frac{1}{1+\kappa^2/g}u^2. \tag{9}$$

The symbol $|_0$ indicates that all derivatives are to be taken at fixed bare parameters. When g goes to infinity, the model we consider here is essentially the same as that studied by Aharony [10]. In that limit, we reproduce Aharony's results regarding critical exponents in the limit $g\rightarrow\infty$, though the fixed points we find in this limit are different from those in Ref. [10] due to different coefficients for u_0, v_0, and w_0. There are six nontrivial fixed points altogether, which may be divided into two groups of three. The first three all have $v^* = 0$, and only one of them with $u_0^* = -3.055\epsilon$ and $w_0^* = 2.504\epsilon$ is stable and physically reachable [10,14].

The recursion relations for the coupling constants in the model, u, v, and w, are determined by the above β functions. When we change the length scale in real space, the momentum scale changes accordingly. If we use parameter l so that $\kappa(l) = \kappa l$, the functions $v(l)$, $u(l)$, and $w(l)$ will obey the following equations $l[dv(l)/dl] = \beta_v$, $l[du(l)/dl] = \beta_u$, and $l[dw(l)/dl] = \beta_w$.

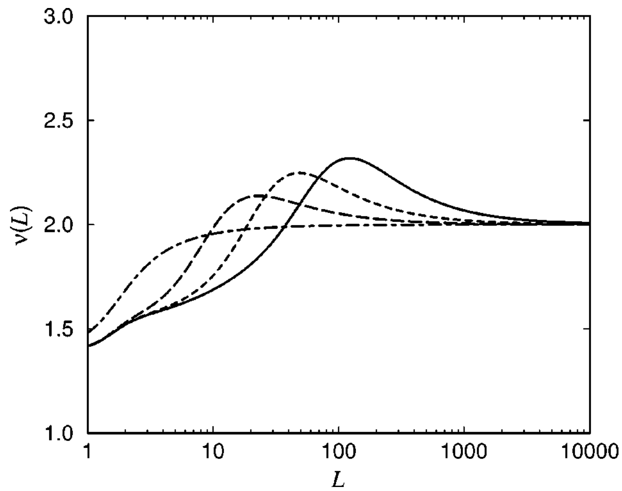

FIG. 1. Exponent ν as a function of length scale. Solid, short-dashed, long-dashed, and dot-dashed lines correspond to $g = 10^{-4}$, 10^{-3}, 10^{-2}, and 10^0, respectively. The initial fluctuation strength is fixed with $u(1) = -1.0$.

Up to one-loop level, $g(l) = g$ [13]. Since initially, $v_0 = 0$ in our model, according to Eq. (7), v will remain zero with changing the length scale. Thus we just need to solve two coupled equations

$$l\frac{du(l)}{dl} = -\epsilon u(l) + \left(\frac{4}{3} - \frac{17}{36}\frac{1}{1+l^2/g}\right)u(l)^2 + \left(\frac{10}{3} - \frac{13}{18}\frac{1}{1+l^2/g}\right)u(l)w(l) + \left(2 - \frac{7}{12}\frac{1}{1+l^2/g}\right)w(l)^2, \tag{10}$$

$$l\frac{dw(l)}{dl} = -\epsilon w(l) + \left(\frac{8}{3} - \frac{9}{12}\frac{1}{1+l^2/g}\right)w(l)^2 + \left(2 - \frac{13}{18}\frac{1}{1+l^2/g}\right)u(l)w(l) + \frac{1}{36}\frac{1}{1+l^2/g}u(l)^2, \tag{11}$$

with initial conditions $u(1) = -\sigma^2$ and $w(1) = u^2(1)/2$. Here, we take κ equal to one.

Having obtained solutions $u(l)$ and $w(l)$ to Eqs. (10) and (11), we can compute the exponent $\nu(L)$ for the scaling of polarizability fluctuation strength $\chi(L) \sim L^{-\nu(L)}$. Noting $l \sim 1/L$, $\nu(L)$ can be expressed as

$$\nu(L) \equiv -\frac{d\ln\chi(L)}{d\ln L} = (2-\eta) + \frac{1}{2}\frac{d\ln u(l)}{d\ln l} = (2-\eta) + \frac{1}{2}u^{-1}(l)\beta_u(l). \tag{12}$$

This expression is valid to all orders of the loop expansion. At one-loop level, the anomalous dimension $\eta=0$ and $\beta_u(l)$ is given by Eq. (10).

Figure 1 depicts how exponent $\nu(L)$ for the dimensionless width of polarizability distribution flows as increasing the length scale. We fix the initial fluctuation strength $u(l=1)=-1.0$ and vary the dipole-dipole interaction strength g. We can see that when the length scale becomes very large, $\nu(L)$ approaches 2. This is because when L is very large (or $l\rightarrow 0$), $u(l)$ will be at its fixed point and $\beta_u=0$, thus $\nu(L)=2-\eta$, and at the one loop level, $\eta=0$.

For the noninteracting model H_t, there is no symmetry breaking and only the u term in the Hamiltonian (6) is nonzero. Moreover, there is no divergence for the diagrams at any order of u. Therefore $\beta_u=-\epsilon u$, showing that the noninteracting model has only the trivial zero fixed point. Because in this case $\eta=0$, from Eq. (12), $\nu(L)=2-\epsilon/2=3/2$. This is the result we noted earlier that follows from the uncorrelated statistics for the independent random variables.

In Fig. 1, when the length scale is not very large, the deviation from this universal exponent may be significant. At small length scales, exponent $\nu(L)$ is around 3/2, indicating that in this regime, the behavior of polarizability fluctuations is close to the result for uncorrelated random variables. In this regime the randomness dominates over the dipole-dipole interactions. At large scales, since the effective strength of dipole-dipole interactions is renormalized as $g(l)/\kappa^2(l) \sim g/l^2 \sim gL^2$, the dipole-dipole interactions become more and more significant as increasing the length scales. In this regime the interaction dominates over the randomness, giving rise to a universal behavior. We see from the figure, when the strength of dipole-dipole interactions is weak, exponent $\nu(L)$ increases slowly as increasing length scales, and at some regions $\nu(L)$ becomes greater than 2 and eventually goes to 2. When the strength of dipole-dipole interactions is strong, $\nu(L)$ increases fast from 3/2 and reaches 2 monotonically. The different behaviors of $\nu(L)$ for different strengths of σ^2 and g reflect competition and cooperation between long-range dipole-dipole interactions and local polarizability fluctuations as changing length scales.

D.C. has been supported by the Office of Basic Energy Sciences, Chemical Sciences Division of the U.S. Department of Energy, Grant No. DE-FG03-87ER13793.

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