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THE QUANTIZATION OF ELECTRODYNAMICS IN NONLINEAR DIELECTRIC MEDIA[1]

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The study of the propagation of quantized fields in nonlinear dielectric media is an area of increasing theoretical and expeimental activity. As a result of their interaction with the medium the fields can acquire nonclassical properties such as squeezing. In order to describe these phenomena it is first necessary to quantize electrodynamics in the presence of a nonlinear medium. We explore three approaches to this problem: the phenomenological, the macroscopic, and the microscopic. In the macroscopic approach the medium is characterized only by its susceptibilies while in the microscopic a model of the medium is required.

1. Introduction

The interaction of light with a nonlinear dielectric medium is responsible for a number of the nonclassical effects which have been studied in quantum optics in recent years. Squeezing and sub-Poissonian statistics can be produced in $\chi^{(2)}$ media and quantum phase diffusion occurs in $\chi^{(3)}$ media [1-4]. More recently studies of the propagation of quantized fields in nonlinear media have been undertaken. Squeezing in quantum solitons has been predicted and observed [5,6], as has phase diffusion [6-8]. Collisions of quantum solitons can be used to perform a quantum nondemolition measurement of photon number [9-11].

The first step which is necessary for the description of these phenomena is the quantization of the electromagnetic field in the presence of a nonlinear dielectric medium. This has been done in three ways. The first is the phenomenological approach in which one starts with the classical field equations and simply substitutes the usual expressions for field operators for the classical fields. This method has its risks and can lead to a Hamiltonian which does not reproduce Maxwell's equations [12]. A more methodical way of proceeding, the macroscopic approach, is to describe the medium by means of its

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linear and nonlinear susceptibilities and to apply the canonical quantization procedure to this theory. That is, one finds a Lagrangian which gives the macroscopic Maxwell equations as equations of motion, from the Lagrangian the canonical momenta and Hamiltonian are found, and, finally, the canonical commutation relations are imposed [12]. The third method, the microscopic approach, involves constructing a microscopic model for the medium and retaining the medium degrees of freedom in the theory [13,14].

Here we would like to discuss the macroscopic and microscopic approaches. They each have advantages and disadvantages. In the macroscopic approach the description of dispersion is more complicated and it is not possible to address questions of operator ordering. On the other hand, the microscopic approach is limited by the model of the medium which has been chosen while the macroscopic theory needs only a set of numbers, the susceptibilities, to characterize the medium. The macroscopic theory has been developed by P. Drummond and S. Carter into a useful tool for the study of the propagation of quantized fields in nonlinear media [15].

2. Macroscopic Approach

Maxwell's equations inside a dielectric medium are given by (in Heaviside-Lorentz units)

$$\nabla \cdot \mathbf{D} = 0 \qquad \nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t}$$
$$\nabla \cdot \mathbf{B} = 0 \qquad \nabla \times \mathbf{B} = \frac{\partial \mathbf{D}}{\partial t}, \tag{1}$$

in the absence of external charges and currents. Here $\mathbf{D} = \mathbf{E} + \mathbf{P}$ is the displacement field and the polarization $\mathbf{P}$ is given by

$$\mathbf{P} = \chi^{(1)} : \mathbf{E} + \chi^{(2)} : \mathbf{EE} + \chi^{(3)} : \mathbf{EEE} + \ldots. \tag{2}$$

The quantities $\chi^{(j)}$ are the $(j+1)$-rank susceptibility tensors. We shall assume that the medium is uniform, lossless, and nondispersive. We want to find a Langrangian which has Eqs. (1) as its equations of motion. Before doing so we need to choose a particular field which is to be the basic dynamical variable in the problem. There are two possibilities. The first is the usual vector potential $A = (A_0, \mathbf{A})$ where

$$\mathbf{E} = -\frac{\partial \mathbf{A}}{\partial t} - \nabla A_0 \qquad \mathbf{B} = \nabla \times \mathbf{A}, \tag{3}$$

and the second is the dual potential $\Lambda = (\Lambda_0, \mathbf{\Lambda})$ where

$$\mathbf{B} = \frac{\partial \mathbf{\Lambda}}{\partial t} + \nabla \Lambda_0 \qquad \mathbf{D} = \nabla \times \mathbf{\Lambda}. \tag{4}$$

This potential can only be used if external charges and currents are absent. We shall discuss both approaches [12].

If the vector potential A is used as the basic field the Lagrangian density is

$$L(A, \dot{A}) = \frac{1}{2}(\mathbf{E}^2 - \mathbf{B}^2) + \frac{1}{2}\chi^{(1)}_{ij} E_i E_j + \frac{1}{3}\chi^{(2)}_{ijk} E_i E_j E_k + \frac{1}{4}\chi^{(3)}_{ijkl} E_i E_j E_k E_l. \tag{5}$$

From this we find that the canonical momentum corresponding to A, which we designate by $\Pi = (\Pi_0, \mathbf{\Pi})$, is

$$\Pi_0 = \frac{\partial L}{\partial(\partial_0 A_0)} = 0 \qquad \Pi_i = \frac{\partial L}{\partial(\partial_0 A_i)} = -D_i. \tag{6}$$

Here we note two things. The first is that the canonical momentum is different from that in the noninteracting theory where $\Pi_i = -E_i$. This is a consequence of the fact that the interaction depends on $\dot{\mathbf{A}}$. The second is that the vanishing of Π_0 implies that A_0 is not an independent field. If we impose the Coulomb gauge condition, $\nabla \cdot \mathbf{A} = 0$, we find that it can, in fact, be expressed in terms of the canonical momenta Π_i.

For the Hamiltonian we have [12]

$$H(\mathbf{A}, \mathbf{\Pi}) = \int d^3r [\frac{1}{2}(\mathbf{E}^2 + \mathbf{B}^2 + \chi^{(1)}_{ij} E_i E_j) + \frac{2}{3}\chi^{(2)}_{ijk} E_i E_j E_k + \frac{3}{4}\chi^{(3)}_{ijkl} E_i E_j E_k E_l]. \tag{7}$$

It is useful to express this directly in terms of the canonical momenta, D_i. To this end we define the tensors $\beta^{(i)}$ by

$$E_i = \beta^{(1)}_{ij} D_j + \beta^{(2)}_{ijk} D_j D_k + \ldots. \tag{8}$$

These tensors can be expressed in terms of the susceptibility tensors

$$\beta^{(1)} = (1 + \chi^{(1)})^{-1}$$
$$\beta^{(2)}_{imn} = -\beta^{(1)}_{ij} \beta^{(1)}_{km} \beta^{(1)}_{ln} \chi^{(2)}_{jkl}. \tag{9}$$

Making use of Eq. (8) we find for the Hamiltonian

$$H(\mathbf{A}, \mathbf{\Pi}) = \int d^3r [\frac{1}{2}(\mathbf{B}^2 + \beta^{(1)}_{ij} D_i D_j) + \frac{1}{3}\beta^{(2)}_{ijk} D_i D_j D_k + \frac{1}{4}\beta^{(3)}_{ijkl} D_i D_j D_k D_l]. \tag{10}$$

The theory is quantized by imposing the equal-time commutation relations

$$[A_j(\mathbf{r}, t), \Pi_k(\mathbf{r}', t)] = i\delta^{(tr)}_{jk}(\mathbf{r} - \mathbf{r}'). \tag{11}$$

Here, as in standard QED, we use the transverse delta function in order to be consistent with both the Coulomb gauge condition, $\nabla \cdot \mathbf{A} = 0$, and Gauss' law, $\nabla \cdot \mathbf{D} = 0$. As in the case of free QED it is possible to perform a mode expansion for the field and to define creation and annihilation operators. In particular, for the mode with momentum $\mathbf{k}$ and polarization $\hat{\epsilon}_\alpha(\mathbf{k})$ we have the annihilation operator

$$\begin{aligned} a_{\mathbf{k},\alpha}(t) &= \frac{1}{\sqrt{V}}\int d^3r e^{-i\mathbf{k}\cdot\mathbf{r}}\hat{\epsilon}_\alpha(\mathbf{k})\cdot[\sqrt{\frac{\omega_k}{2}}\mathbf{A}(\mathbf{r},t) \\ &\quad -\frac{i}{\sqrt{2\omega_k}}\mathbf{D}(\mathbf{r},t)], \end{aligned} \tag{12}$$

where $\omega_k = |\mathbf{k}|$. Note that because $a_{\mathbf{k},\alpha}$ depends on $\mathbf{D}$, and consequently contains matter degrees of freedom, it is not a pure photon operator. It represents a collective matter-field mode.

If one takes the dual potential, Λ, as the basic field the resulting theory is somewhat simpler. First, it is no longer convenient to express the polarization in terms of the electric field, but we instead write

$$\mathbf{P} = \eta^{(1)} : \mathbf{D} + \eta^{(2)} : \mathbf{DD} + \eta^{(3)} : \mathbf{DDD}. \tag{13}$$

The tensors $\eta^{(i)}$ are closely related to the tensors $\beta^{(i)}$

$$\eta^{(1)} = 1 - \beta^{(1)} \qquad \eta^{(j)} = -\beta^{(j)} \qquad j = 2,3,\ldots \tag{14}$$

The Langrangian density is now

$$L = \frac{1}{2}(\mathbf{B}^2 - \mathbf{D}^2) + \frac{1}{2}\mathbf{D}\cdot\eta^{(1)} : \mathbf{D} + \frac{1}{3}\mathbf{D}\cdot\eta^{(2)} : \mathbf{DD} + \ldots \tag{15}$$

From this one finds for the canonical momenta

$$\Pi_0 = 0 \qquad \Pi_j = -B_j. \tag{16}$$

In this approach the canonical momenta do not depend on the interaction; they are the same in the free and interacting theories. This is what makes the dual potential theory simpler. When the theory is quantized the equal-time commutation relations are the same as in the free theory. As we saw, this was not the case when the usual vector potential was used. Because of this property it is also straightforward to describe inhomogeneous media using the dual potential. This is considerably more complicated if A is taken to be the basic field, because then the inhomogeneity of the medium appears in the fundamental field commutation relations [12].

The fact that $\Pi = 0$ again means that Λ_0 is not an independent field. In this case, however, if we impose the Coulomb gauge, $\nabla\cdot\mathbf{\Lambda} = 0$, we can choose $\Pi_0 = 0$ [12]. This represents another simplification over the standard vector potential approach.

The Hamiltonian is now

$$\begin{aligned} H &= \int d^3r[\frac{1}{2}(\mathbf{B}^2 + \mathbf{D}^2 - \mathbf{D}\cdot\eta^{(1)} : \mathbf{D}) - \frac{1}{3}\mathbf{D}\cdot\eta^{(2)} : \mathbf{DD} \\ &\quad -\frac{1}{4}\mathbf{D}\cdot\eta^{(3)} : \mathbf{DDD}]. \end{aligned} \tag{17}$$

This together with the canonical equal-time commutation relations

$$[\dot{\Lambda}_j(\mathbf{r},t), \Lambda_k(\mathbf{r}',t)] = i\delta^{(tr)}_{jk}(\mathbf{r}-\mathbf{r}'), \tag{18}$$

define the fully quantized theory.

So far nothing has been said about dispersion. It has been assumed that the response of the medium to the fields is instantaneous. In a dispersive medium the polarization at time t depends on the field not only at t but at previous times as well. That is, the polarization is nonlocal in time. This presents serious problems for the Hamiltonian formulation, and subsequent quantization, of a theory containing dispersion.

One approach to overcoming this difficulty was pioneered by Drummond [16]. He formulated the theory using the dual potential, which he then broke up into narrow band, slowly varying parts. These become the basic fields of the theory. Because of dispersion the linear polarizability is now a function of frequency, ω (dispersion in the nonlinear susceptibilities represents a small effect and is ignored). If in each frequency band the linear polarizability is expanded in $(\omega - \omega_\nu)$, where ω_ν is the central frequency of the band, up to second order, the result is a local theory for the narrow-band fields. This theory can then be quantized.

Carter and Drummond have applied this theory to describe fields propagating through a fiber with a $\chi^{(3)}$ nonlinearity [15]. The narrow-band field, $\Psi(z,t)$ is assumed to vary only along the fiber (the z direction), and it obeys the commutation relations

$$[\Psi(z,t), \Psi^\dagger(z',t)] = \delta(z-z'). \tag{19}$$

The Hamiltonian is

$$\begin{aligned} H &= \frac{1}{2}\int_0^L dz[iv\left(\frac{\partial\Psi^\dagger}{\partial z}\Psi - \Psi^\dagger\frac{\partial\Psi}{\partial z}\right) + \omega''\frac{\partial\Psi^\dagger}{\partial z}\frac{\partial\Psi}{\partial z} \\ &\quad -v^2\chi^E(\Psi^\dagger)^2\Psi^2], \end{aligned} \tag{20}$$

where v is the group velocity, χ^E is proportional to $\chi^{(3)}$ and ω'' is the second derivative of frequency with respect to wave number evaluated at the center of the frequency band.

3. Microscopic Approach

In order to formulate a microscopic description of nonlinear optics we need a model for the nonlinear medium itself [13,14]. We shall consider one consisting of two-level atoms. The atoms will occupy the entire quantization volume, V, and their density ρ will be such that there are a large number of atoms per cubic optical wavelength. Because we shall only consider optical wavelengths we can partition the medium into small boxes. The size of each box is much less than a wavelength, but it, nonetheless, contains a large number of atoms which we shall assume to be the same for each box and shall call n_0. Because the size of a box is small compared to a wavelength each atom in the box sees the same field. Consequently, the atoms in each box can be described as a spin

$s = n_0/2$ object which interacts with the field. The Hamiltonian describing the boxes interacting with the electromagnetic field is

$$H = \sum_{l=1}^{N_b} E_0 S_l^{(3)} + \sum_{\mathbf{k}} \omega_k a_{\mathbf{k}}^\dagger a_{\mathbf{k}} + \frac{e^2 n_0}{2m} \sum_{l=1}^{N_b} \mathbf{A}(\mathbf{r}_l)^2 + H_{int}, \tag{21}$$

where

$$H_{int} = \sum_{l=1}^{N_b} \sum_{\mathbf{k}} \sqrt{\frac{1}{2\omega_k V}} (a_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}_l} + a_{\mathbf{k}}^\dagger e^{-i\mathbf{k}\cdot\mathbf{r}_l}) i\mu(\mathbf{k})(S_l^{(+)} - S_l^{(-)}). \tag{22}$$

Here we have assumed only one polarization is present, E_0 is the energy difference between the levels, $S_l^{(3)}$, $S_l^{(+)}$, and $S_l^{(-)}$ are the spin operators for the l^{th} box, N_b is the number of boxes, and

$$\mu(\mathbf{k}) = eE_0 \langle a|\mathbf{x}|b\rangle \cdot \hat{\epsilon}(\mathbf{k}). \tag{23}$$

The matrix element $\langle a|\mathbf{x}|b\rangle$ is just the dipole matrix element of the atom and $\hat{\epsilon}(\mathbf{k})$ is the field polarization vector. The wave number k_u is a cutoff imposed to guarantee that the wavelength does not become smaller than the box size.

In order to proceed we want to expand the spin operators. This can be done by using the Holstein-Primakoff representation of the spin operators in terms of boson creation and annihilation operators $\zeta^\dagger$ and ζ. We have [17]

$$S^{(-)} = (2s - \zeta^\dagger\zeta)^{1/2}\zeta \qquad S^{(+)} = \zeta^\dagger(2s - \zeta^\dagger\zeta)^{1/2}$$

$$S^{(3)} = -s + \zeta^\dagger\zeta. \tag{24}$$

The excitation number for the boson operators, i. e. $\zeta^\dagger\zeta$, corresponds to $s_3 + s$, where s_3 is the eigenvalue of $S^{(3)}$. Therefore, the boson vacuum state corresponds to the spin pointing down, i. e. all atoms in their ground states. If we are only considering states whose excitation number is small we can expand the square roots

$$S^{(-)} = \sqrt{2s}(1 - \frac{1}{4s}\zeta^\dagger\zeta)\zeta \qquad S^{(+)} = \sqrt{2s}\zeta^\dagger(1 - \frac{1}{4s}\zeta^\dagger\zeta). \tag{25}$$

In our model of a nonlinear medium the fraction of atoms in each block which is excited is small because we are off resonance. Therefore, the use of this expansion is justified.

For each spin operator in the Hamiltonian we substitute the corresponding expressions in terms of the creation and annihilation operators $\zeta_l^\dagger$ and ζ_l (each box has its own set of creation and annihilation operators). In addition we go to a continuous representation where ζ_l is replaced by $\zeta(\mathbf{r})$ where

$$[\zeta(\mathbf{r}), \zeta^\dagger(\mathbf{r}')] = \delta^{(3)}(\mathbf{r} - \mathbf{r}'). \tag{26}$$

The resulting Hamiltonian is

$$H = H_0 + H_{int}^{(1)} + H_{int}^{(2)}, \tag{27}$$

where

$$\begin{aligned} H_0 &= -\frac{1}{2}NE_0 + E_0 \int_V d^3r \zeta^\dagger(\mathbf{r})\zeta(\mathbf{r}) + \sum_{|\mathbf{k}|<k_u} \omega_k a_{\mathbf{k}}^\dagger a_{\mathbf{k}} \\ &+ \frac{e^2\rho}{2m} \sum_{|\mathbf{k}|<k_u} \frac{1}{2\omega_k}[a_{\mathbf{k}}a_{-\mathbf{k}} + a_{\mathbf{k}}a_{\mathbf{k}}^\dagger + a_{\mathbf{k}}^\dagger a_{\mathbf{k}} + a_{\mathbf{k}}^\dagger a_{-\mathbf{k}}^\dagger] \\ H_{int}^{(1)} &= \frac{i}{\sqrt{V}} \sum_{|\mathbf{k}|<k_u} \int_V d^3r (a_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}} + a_{\mathbf{k}}^\dagger e^{-i\mathbf{k}\cdot\mathbf{r}}) g_{\mathbf{k}} (\zeta^\dagger(\mathbf{r}) - \zeta(\mathbf{r})) \\ H_{int}^{(2)} &= -\frac{i}{2\rho\sqrt{V}} \sum_{|\mathbf{k}|<k_u} \int_V d^3r (a_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}} + a_{\mathbf{k}}^\dagger e^{-i\mathbf{k}\cdot\mathbf{r}}) g_{\mathbf{k}} (\zeta^\dagger(\mathbf{r})^2\zeta(\mathbf{r}) \\ &-\zeta^\dagger(\mathbf{r})\zeta(\mathbf{r})^2). \end{aligned} \tag{28}$$

Here $g_{\mathbf{k}} = \mu(\mathbf{k})\sqrt{\rho/2\omega_k}$ and N is the total number of atoms.

The Hamiltonian $H_0 + H_{int}^{(1)}$ describes the electromagnetic field interacting with a linear medium. It is closely related to the Hamiltonian considered by Hopfield in his study of fields in a linear dielectric [18]. He found that the dynamics of this system is most easily described in terms of modes which diagonalize the Hamiltonian, which are known as polaritons. They are neither pure field nor pure matter modes, but mixtures of the two.

The operator $H_{int}^{(2)}$ describes the nonlinear interaction betwen the field and the medium. It is not in a form which is familiar from nonlinear optics. In particular, for a single-mode field interacting with a two-level atom medium we would expect a Hamiltonian more like the one which is used to describe self-phase modulation

$$H_{int} = \lambda(a^\dagger)^2 a^2. \tag{29}$$

Is it possible to extract an interaction of this form from $H_{int}^{(2)}$?

The answer to this question is yes, and the key to answering it is polaritons. One first diagonalizes $H_0 + H_{int}^{(1)}$ in terms of new operators $\alpha_{\mathbf{k}}$ and $\beta_{\mathbf{k}}$ which are linear combinations of $a_{\mathbf{k}}$, $\zeta_{\mathbf{k}}$, $a_{-\mathbf{k}}^\dagger$, and $\zeta_{-\mathbf{k}}^\dagger$, and satisfy boson commutation relations. Here

$$\zeta_{\mathbf{k}} = \frac{1}{\sqrt{V}} \int_V d^3r e^{-i\mathbf{k}\cdot\mathbf{r}} \zeta(\mathbf{r}). \tag{30}$$

One then has

$$H_0 + H_{int}^{(1)} = \sum_{|\mathbf{k}|<k_u} [E_1(k)\alpha_{\mathbf{k}}^\dagger\alpha_{\mathbf{k}} + E_2(k)\beta_{\mathbf{k}}^\dagger\beta_{\mathbf{k}}], \tag{31}$$

where

$$\begin{aligned} E_1(k) &= \frac{1}{\sqrt{2}}([E_0^2 + \omega_k(\omega_k + 2C_0)] \\ &+ [[E_0^2 - \omega_k(\omega_k + 2C_0)]^2 + 16E_0\omega_k g_{\mathbf{k}}^2]^{1/2})^{1/2} \end{aligned}$$

$$E_2(k) = \frac{1}{\sqrt{2}}([E_0^2 + \omega_k(\omega_k + 2C_0)] - [[E_0^2 - \omega_k(\omega_k + 2C_0)]^2 + 16E_0\omega_k g_{\mathbf{k}}^2]^{1/2})^{1/2}, \quad (32)$$

with $C_0 = (e^2\rho)/(2m\omega_k)$. The next step is to express $H_{int}^{(2)}$ in terms of $\alpha_{\mathbf{k}}$ and $\beta_{\mathbf{k}}$. This results in a great many terms, but a considerable simplification occurs if only a small number of modes are highly excited, and we keep only terms containing these modes. For example, if the $\alpha_{\mathbf{k}_0}$ mode is the only one which is highly excited we find that

$$H_{int}^{(2)} \to \lambda(\mathbf{k}_0)(\alpha_{\mathbf{k}_0}^\dagger)^2\alpha_{\mathbf{k}_0}^2, \quad (33)$$

where $\lambda(\mathbf{k}_0)$ is a constant which is found from the transformation which relates the polariton to the matter and field operators. By going to the polariton basis we have recovered the self-phase modulation interaction. This tells us, in addition, that the operators in Eq. (29) should not be field operators, but should, in fact, correspond to mixed matter field modes.

Suppose that instead of a single mode we have a pulse. In particular, let us assume that the pulse consists of $\alpha(\mathbf{k})$ modes where $\mathbf{k}$ is in a small neighborhood, S, of $\mathbf{k}_0$. In that case we have

$$H_{int}^{(2)} \to \lambda(\mathbf{k}_0)\sum_{\mathbf{k}\in S}\sum_{\mathbf{k}_1\in S}\sum_{\mathbf{k}_2\in S}\sum_{\mathbf{k}_3\in S}\delta_{\mathbf{k}+\mathbf{k}_1,\mathbf{k}_2+\mathbf{k}_3}\alpha_{\mathbf{k}}^\dagger\alpha_{\mathbf{k}_1}^\dagger\alpha_{\mathbf{k}_2}\alpha_{\mathbf{k}_3}. \quad (34)$$

The equations of motion for the operators $\alpha_{\mathbf{k}}$ which emerge from the theory obtained by combining Eqs. (31) and (34) are similar in form to those obtained for the mode annihilation operators in the Carter- Drummond theory.

Finally, let us note two features of the microscopic theory. First, dispersion is included in a natural way because the basic modes in the theory correspond to polaritons and not photons. The polariton dispersion relation is different from that of photons. The second concerns operator ordering. The polariton operators do not appear in the Hamiltonian in normal order. In deriving Eqs. (33) and (34) we have dropped terms arising from commutators. We expect these terms to be small in the situations considered here, but this will not always be the case. The microscopic theory gives us a way of examining this issue while the macroscopic does not.

4. Conclusion

We have examined three different approaches to the quantization of the electromagnetic field in the presence of a nonlinear dielectric medium: the phenomenological, the macroscopic, and the microscopic. The phenomenological approach has found the greatest use but is not well grounded in the underlying theory. The macroscopic theory is built on firmer foundations and has been applied to the study of the propagation of quantum fields in fibers by Carter and Drummond. The microscopic approach is the most fundamental, but its development is at an early stage.

The study of the propagation of quantized fields in nonlinear media is an area of increasing experimental and theoretical activity. It is essential that we achieve a better understanding of the underlying theory for the field to proceed.

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