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Quantized-field description of light in negative-index media

Peter W. Milonni^{a,*}, G. Jordan Maclay^b

^a Theoretical Division (T-DOT), Los Alamos National Laboratory, Los Alamos, NM 87545, USA ^b Quantum Fields LLC, 20876 Wildflower Lane, Richland Center, WI 53581, USA

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Abstract

Using a quantized-field approach, we show how radiative recoil, the Doppler effect, and spontaneous and stimulated radiation rates are affected when the radiator is embedded in a host medium having a negative index of refraction. © 2003 Elsevier B.V. All rights reserved.

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1. Introduction

There is nothing to prevent both the electric permittivity ϵ and the magnetic permeability μ from being negative, although no known, naturally occurring material has this property. For such a material an examination of Maxwell's equations and the boundary conditions implies that the refractive index *n* is also negative [1]; the material may be said to be *left-handed* because the vectors **E**, **H**, and **k** form a left-handed system, i.e., the direction of energy flow, given by $\mathbf{E} \times \mathbf{H}$, is opposite to the direction of the wave vector **k**. Pendry's suggestion [2] that a negative-index (or left-handed) medium could be used to make a "perfect lens", and the first

*Corresponding author. Tel.: +1-505-6677763; fax: +1-505-6654055.

E-mail address: pwm@lanl.gov (P.W. Milonni).

experimental demonstration [3] of negative-index features of a "medium" consisting of a periodic array of copper strips and copper split ring resonators assembled into an interlocking lattice, have sparked great interest in the subject and further experimentation [4].

Negative-index media have various odd properties. The Doppler effect, for instance, is reversed. Other familiar phenomena call for a reexamination in the case n < 0. For example, it is well known (and experimentally verified [5]) that, if local field corrections are negligible, the spontaneous emission rate for an electric-dipole transition of frequency ω_0 is $A' = n(\omega_0)A$, where A is the free-space Einstein A coefficient. How is this formula to be interpreted, or modified, if $n(\omega_0) < 0$?

In this paper we consider such examples using a quantized-field description of the electromagnetic field in a negative-index material, which we assume to form an isotropic continuum and to be

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non-absorptive at frequencies of interest. The approach taken here might eventually prove useful for the description of certain aspects of negativeindex materials, although at this point in the evolution of the field it is not necessary for any practical purposes known to us.

We first review a simple quantization procedure [6] for radiation of frequency far from any absorption resonances of a dielectric medium. Poynting's theorem states that $\nabla \cdot \mathbf{S} + \partial u / \partial t = 0$ in the absence of any currents, where $\mathbf{S} = (c/4\pi)\mathbf{E} \times \mathbf{H}$ and

$$\frac{\partial u}{\partial t} = \frac{1}{4\pi} \left(\mathbf{E} \cdot \frac{\partial \mathbf{D}}{\partial t} + \mathbf{H} \cdot \frac{\partial \mathbf{B}}{\partial t} \right). \tag{1}$$

We are interested in a narrow band of frequencies about a frequency ω , within which absorption is negligible, and write

$$\mathbf{E}(\mathbf{r},t) = \mathbf{E}_{\omega}(\mathbf{r},t) \mathbf{e}^{-i\omega t}$$
$$= \int_{-\infty}^{\infty} \mathrm{d}\Delta \, \mathbf{e}_{\omega}(\mathbf{r},\Delta) \mathbf{e}^{-i(\omega+\Delta)t}, \qquad (2)$$

where \mathbf{E}_{ω} is slowly varying in time compared with $\exp(-i\omega t)$. Thus

$$\mathbf{D}(\mathbf{r},t) = \int_{-\infty}^{\infty} \mathrm{d}\Delta \,\epsilon(\omega + \Delta) \mathbf{e}_{\omega}(\mathbf{r},\Delta) \mathrm{e}^{-\mathrm{i}(\omega + \Delta)t}$$
$$\approx \int_{-\infty}^{\infty} \mathrm{d}\Delta \left[\epsilon(\omega) + \Delta \frac{\mathrm{d}\epsilon}{\mathrm{d}\omega}\right] \mathbf{e}_{\omega}(\mathbf{r},\Delta) \mathrm{e}^{-\mathrm{i}(\omega + \Delta)t}$$
$$\approx \epsilon(\omega) \mathbf{E}(\mathbf{r},t) + \mathrm{i} \frac{\mathrm{d}\epsilon}{\mathrm{d}\omega} \mathrm{e}^{-\mathrm{i}\omega t} \frac{\partial \mathbf{E}_{\omega}}{\partial t}, \qquad (3)$$

$$\frac{\partial \mathbf{D}}{\partial t} \approx \epsilon \frac{\partial \mathbf{E}}{\partial t} + \omega \frac{\mathrm{d}\epsilon}{\mathrm{d}\omega} \frac{\partial \mathbf{E}_{\omega}}{\partial t} \mathrm{e}^{-\mathrm{i}\omega t}$$

$$= \left[\epsilon \frac{\partial \mathbf{E}_{\omega}}{\partial t} - \mathrm{i}\omega\epsilon \mathbf{E}_{\omega} + \omega \frac{\mathrm{d}\epsilon}{\mathrm{d}\omega} \frac{\partial \mathbf{E}_{\omega}}{\partial t}\right] \mathrm{e}^{-\mathrm{i}\omega t}$$

$$= \left[\frac{\mathrm{d}}{\mathrm{d}\omega}(\epsilon\omega) \frac{\partial \mathbf{E}_{\omega}}{\partial t} - \mathrm{i}\omega\epsilon \mathbf{E}_{\omega}\right] \mathrm{e}^{-\mathrm{i}\omega t}, \qquad (4)$$

where we use the assumption that only a narrow band of frequencies is significant. Thus

$$\mathbf{E} \cdot \frac{\partial \mathbf{D}}{\partial t} \approx \frac{1}{4} \frac{\mathrm{d}}{\mathrm{d}\omega} (\epsilon \omega) \frac{\partial}{\partial t} |\mathbf{E}_{\omega}|^{2}.$$
 (5)

A similar calculation for $\mathbf{H} \cdot \partial \mathbf{B} / \partial t$ then yields [7]

$$u_{\omega} = \frac{1}{16\pi} \left[\frac{\mathrm{d}}{\mathrm{d}\omega} (\epsilon \omega) |\mathbf{E}_{\omega}|^2 + \frac{\mathrm{d}}{\mathrm{d}\omega} (\mu \omega) |\mathbf{H}_{\omega}|^2 \right]$$
(6)

for the field energy density at frequency ω .

Now write

$$\mathbf{E}(\mathbf{r},t) = C\alpha(t)\mathbf{F}(\mathbf{r}),\tag{7}$$

where $\alpha(t) = \alpha(0) \exp(-i\omega t)$, **F**(**r**) is a (normalized) mode function, and C is a constant to be chosen. Maxwell's equations imply

$$\mathbf{B}(\mathbf{r},t) = -\mathrm{i}\frac{c}{\omega}C\alpha(t)\nabla\times\mathbf{F}(\mathbf{r}),\tag{8}$$

$$\mathbf{D}(\mathbf{r},t) = \epsilon C \alpha(t) \mathbf{F}(\mathbf{r}), \tag{9}$$

$$\mathbf{H}(\mathbf{r},t) = -\frac{\mathrm{i}}{\mu} \frac{c}{\omega} C \alpha(t) \nabla \times \mathbf{F}(\mathbf{r}).$$
(10)

It then follows that the field energy associated with frequency ω is

$$U_{\omega} = \int d^{3}r u_{\omega}$$

= $\frac{1}{16\pi\mu} |C|^{2} |\alpha(t)|^{2} \left[\mu \frac{d}{d\omega} (\epsilon\omega) + \epsilon \frac{d}{d\omega} (\mu\omega) \right]$
= $\frac{n}{8\pi\mu} |C|^{2} |\alpha(t)|^{2} \frac{d}{d\omega} (n\omega),$ (11)

where $n^2(\omega) = \epsilon(\omega)\mu(\omega)$. Setting $C = (4\pi\mu/n\gamma)^{1/2}$, where $\gamma = d(n\omega)/d\omega$, we have $U_{\omega} = \frac{1}{2} |\alpha(t)|^2$. Let us furthermore write $\alpha(t) = \alpha(0) \exp(-i\omega t) = p(t) - i\omega q(t)$, which implies that $\dot{q} = p$, $\dot{p} = -\omega^2 q$, i.e., we have the Hamilton equations of motion for a simple harmonic oscillator. To quantize the field mode of frequency ω we quantize this harmonic oscillator, replacing q and p by operators \hat{q} and \hat{p} satisfying $[\hat{q}, \hat{p}] = i\hbar$. The photon annihilation and creation operators are then $\hat{a} = (1/\sqrt{2\hbar\omega})(\hat{p} - i\omega\hat{q})$ and $\hat{a}^{\dagger} = (1/\sqrt{2\hbar\omega})(\hat{p} + \mathrm{i}\omega\hat{q}), \ [\hat{a}, \hat{a}^{\dagger}] = 1.$

The electric field $\frac{1}{2} [C\alpha(t)\mathbf{F}(\mathbf{r}) + C^*\alpha^*(t)\mathbf{F}(\mathbf{r})^*]$ is similarly replaced by the operator

$$\hat{\mathbf{E}}(\mathbf{r},t) = \left(\frac{2\pi\hbar\omega\mu}{n\gamma}\right)^{1/2} [\hat{a}(t)\mathbf{F}(\mathbf{r}) + \hat{a}^{\dagger}(t)\mathbf{F}(\mathbf{r})^{*}] \quad (12)$$

when we quantize. For our purposes it suffices to work with the plane-wave modes $\mathbf{F}(\mathbf{r}) =$ $(i/\sqrt{V})\mathbf{e}_{\mathbf{k}}\exp(i\mathbf{k}\cdot\mathbf{r})$, where $k=n(\omega)\omega/c$ and $\mathbf{e}_{\mathbf{k}}$ is a unit polarization vector ($\mathbf{k} \cdot \mathbf{e}_{\mathbf{k}} = 0$):

$$\hat{\mathbf{E}}(\mathbf{r},t) = i \left(\frac{2\pi\hbar\omega\mu}{n\gamma V}\right)^{1/2} \left[\hat{a}(t)e^{i\mathbf{k}\cdot\mathbf{r}} - \hat{a}^{\dagger}(t)e^{-i\mathbf{k}\cdot\mathbf{r}}\right] \mathbf{e}_{\mathbf{k}},$$
(13)

where we have taken $\mathbf{e}_{\mathbf{k}}$ to be real. Similarly, from Eqs. (8)–(10), we write the quantized fields

$$\hat{\mathbf{B}}(\mathbf{r},t) = i \left(\frac{2\pi\hbar\mu c^2}{\omega n\gamma V}\right)^{1/2} \left[\hat{a}(t)e^{i\mathbf{k}\cdot\mathbf{r}} - \hat{a}^{\dagger}(t)e^{-i\mathbf{k}\cdot\mathbf{r}}\right]\mathbf{k} \times \mathbf{e}_{\mathbf{k}},$$
(14)

$$\hat{\mathbf{D}}(\mathbf{r},t) = i \left(\frac{2\pi\hbar\omega n\epsilon}{\gamma V}\right)^{1/2} \left[\hat{a}(t)e^{i\mathbf{k}\cdot\mathbf{r}} - \hat{a}^{\dagger}(t)e^{-i\mathbf{k}\cdot\mathbf{r}}\right] \mathbf{e}_{\mathbf{k}},$$
(15)

$$\hat{\mathbf{H}}(\mathbf{r},t) = \mathrm{i} \left(\frac{2\pi\hbar c^2}{\omega n\mu\gamma V}\right)^{1/2} \left[\hat{a}(t)\mathrm{e}^{\mathrm{i}\mathbf{k}\cdot\mathbf{r}} - \hat{a}^{\dagger}(t)\mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{r}}\right] \mathbf{k} \times \mathbf{e}_{\mathbf{k}}.$$
(16)

These expressions apply to a single-mode field in a dispersive dielectric, provided the mode frequency is far from any absorption resonance. In particular, they apply to the case of a negativeindex material, where $\epsilon(\omega)$, $\mu(\omega)$, and $n(\omega)$ are all negative. Note that $\gamma > 0$ for a negative-index medium (as well as for a positive-index medium) under the assumptions we have made. To see this, write $n = -\sqrt{|\epsilon||\mu|}$ ($\epsilon, \mu < 0$), in which case

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$$\begin{split} \gamma &= n + \omega \frac{\mathrm{d}n}{\mathrm{d}\omega} \\ &= -\sqrt{|\epsilon||\mu|} - \frac{\omega}{2} \sqrt{\frac{|\epsilon|}{|\mu|}} \frac{\mathrm{d}|\mu|}{\mathrm{d}\omega} - \frac{\omega}{2} \sqrt{\frac{|\mu|}{|\epsilon|}} \frac{\mathrm{d}|\epsilon|}{\mathrm{d}\omega} \\ &= -\frac{1}{2} \sqrt{\frac{|\epsilon|}{|\mu|}} \left(|\mu| + \omega \frac{\mathrm{d}|\mu|}{\mathrm{d}\omega} \right) - \frac{1}{2} \sqrt{\frac{|\mu|}{|\epsilon|}} \left(|\epsilon| + \omega \frac{\mathrm{d}|\epsilon|}{\mathrm{d}\omega} \right) \\ &= \frac{1}{2} \sqrt{\frac{|\epsilon|}{|\mu|}} \frac{\mathrm{d}}{\mathrm{d}\omega} (\mu\omega) + \frac{1}{2} \sqrt{\frac{|\mu|}{|\epsilon|}} \frac{\mathrm{d}}{\mathrm{d}\omega} (\epsilon\omega), \end{split}$$
(17)

which is positive because $d(\mu\omega)/d\omega$ and $d(\epsilon\omega)/d\omega$ are positive. The latter conditions, which ensure that the classical field energy U_{ω} is positive, follow from general dispersion relations based on causality [7].

The field energy (11) becomes, on quantization,

$$\hat{H}_{\text{field}} = \frac{n}{8\pi\mu} \left(\frac{4\pi\mu}{n\gamma}\right) (\hat{p}^2 + \omega^2 \hat{q}^2) \gamma$$
$$= \hbar\omega (\hat{a}^{\dagger} \hat{a} + 1/2). \tag{18}$$

The operator corresponding to the (cycle-averaged) Poynting vector, similarly, is

$$\hat{\mathbf{S}} = \frac{\hbar c^2}{n\gamma V} (\hat{a}^{\dagger} \hat{a} + 1/2) \mathbf{e}_{\mathbf{k}} \times (\mathbf{k} \times \mathbf{e}_{\mathbf{k}}).$$
(19)

Writing

$$\mathbf{k} = \frac{n\omega}{c} \mathbf{z},\tag{20}$$

where \mathbf{z} is the unit vector pointing in the *z* direction, we have

$$\hat{\mathbf{S}} = \frac{\hbar c \omega}{\gamma V} (\hat{a}^{\dagger} \hat{a} + 1/2) \mathbf{z} = \frac{\hbar \omega v_g}{V} (\hat{a}^{\dagger} \hat{a} + 1/2) \mathbf{z}$$
$$= \mathbf{z} v_g (\hat{H}_{\text{field}} / V), \qquad (21)$$

where the (scalar) group velocity $v_g = c/\gamma$. Eqs. (20) and (21) show that, in a negative-index medium, the Poynting vector and the **k** vector point in opposite directions; **E**, **H**, and **k** define a lefthanded triad.

Consider an excited atom of mass *m* moving with velocity **v** in a negative-index medium. For the initial state we take a product of a wave function $\propto \exp(\operatorname{im} \mathbf{v} \cdot \mathbf{r}/\hbar)$ describing the center-ofmass motion, the state $|\phi_i\rangle$ of energy E_i of the atom, and the vacuum state $|0\rangle$ of the field. For the final state we take a product of the center-of-mass wave function $\propto \exp(\operatorname{im} \mathbf{v}' \cdot \mathbf{r}/\hbar)$, the ground state $|\phi_f\rangle$ of energy E_f for the atom, and the field state $|\mathbf{1}_k\rangle$ in which there is a single photon with wave vector **k** and frequency ω . The probability amplitude for the transition $|\Psi_i\rangle \rightarrow |\Psi_f\rangle$ involves a matrix element proportional to

$$\int dt \exp\left\{\frac{i}{\hbar} \left(\frac{1}{2}mv^{\prime 2} - \frac{1}{2}mv^{2}\right)t\right\}$$
$$\times \int d^{3}r \exp\left\{-\frac{i}{\hbar}(m\mathbf{v}' - m\mathbf{v}) \cdot \mathbf{r}\right\}$$
$$\times \langle \phi_{\rm f} | \langle \mathbf{1}_{\mathbf{k}} | \hat{\boldsymbol{\sigma}}(t) \hat{\boldsymbol{a}}^{\dagger}_{\mathbf{k}}(t) \mathrm{e}^{i\mathbf{k}\cdot\mathbf{r}} | \mathbf{0} \rangle | \phi_{\rm i} \rangle, \qquad (22)$$

where $\hat{\sigma}$ is the lowering operator for the internal atomic states and $\hat{a}_{\mathbf{k}}^{\dagger}$ is the photon creation operator for the field mode of frequency ω and wave vector **k**. Since the (free) evolution of these operators is $\hat{\sigma}(t) = \hat{\sigma}(0) \exp[-i(E_i - E_f)]$ and $\hat{a}_{\mathbf{k}}^{\dagger}(t) =$ $\hat{a}_{\mathbf{k}}^{\dagger}(0) \exp(i\omega t)$, and $\langle \phi_f | \langle \mathbf{1}_{\mathbf{k}} | \hat{\sigma}(0) \hat{a}_{\mathbf{k}}^{\dagger}(0) | \phi_i \rangle | 0 \rangle = 1$, the transition amplitude is proportional to

$$\int dt \exp\left\{\frac{i}{\hbar} \left(\frac{1}{2}mv^{\prime 2} - \frac{1}{2}mv^{2} + E_{\rm f} - E_{\rm i} + \hbar\omega\right)t\right\}$$
$$\times \int d^{3}r \exp\left\{-\frac{i}{\hbar}(m\mathbf{v}' - m\mathbf{v} + \hbar\mathbf{k})\cdot\mathbf{r}\right\}. \tag{23}$$

The time integral implies energy conservation

$$\frac{1}{2}mv^{2} + \hbar\omega = \frac{1}{2}mv^{2} + \hbar\omega_{0}, \qquad (24)$$

where $\omega_0 = (E_i - E_f)/\hbar$ is the transition frequency of the (stationary) atom. The integral over space implies momentum conservation

$$m\mathbf{v}' = m\mathbf{v} - \hbar\mathbf{k}.\tag{25}$$

Combining (24) and (25), and ignoring terms involving c^{-2} in this nonrelativistic treatment, we obtain

$$\omega = \omega_0 \left(1 + \frac{n}{c} v \cos \theta \right), \tag{26}$$

where θ is the angle between v and z. Thus, as deduced by Veselago from classical considerations, the Doppler effect is reversed in a negative-index medium: if the source is moving towards the detector, the emitted radiation radiation is observed to have a *smaller* frequency. Eq. (25) implies that recoil imparted to the atom upon emission of the photon will be in the *same* direction as the Poynting vector of the emitted field in a negative-index medium.

The multimode generalization of (13), for instance, is

$$\hat{\mathbf{E}}(\mathbf{r},t) = \mathbf{i} \sum_{\mathbf{k}\lambda} \left(\frac{2\pi\hbar\omega\mu_{\omega}}{n_{\omega}\gamma_{\omega}V} \right)^{1/2} \times \left[\hat{a}_{\mathbf{k}\lambda}(t) \mathbf{e}^{\mathbf{i}\mathbf{k}\cdot\mathbf{r}} - \hat{a}_{\mathbf{k}\lambda}^{\dagger}(t) \mathbf{e}^{-\mathbf{i}\mathbf{k}\cdot\mathbf{r}} \right] \mathbf{e}_{\mathbf{k}\lambda}.$$
(27)

 λ labels the polarization of mode \mathbf{k}, λ ($\mathbf{k} \cdot \mathbf{e}_{\mathbf{k}\lambda} = 0$, $\lambda = 1, 2$).

Consider the rate of spontaneous emission of an atom in a negative-index medium. We assume the most important case, that of an electric-dipole transition. (The result will apply equally well to the radiation rate of a dipole antenna.) The coupling constant for the field mode \mathbf{k} , λ and the atomic transition with electric dipole matrix element **d** is

$$V(\omega) = -i \left(\frac{2\pi\hbar\omega_k\mu_k}{n_k\gamma_k V}\right)^{1/2} \mathbf{d} \cdot \mathbf{e}_{\mathbf{k}\lambda}.$$
 (28)

Fermi's golden rule then implies the spontaneous emission rate

$$\frac{2\pi}{\hbar} |V(\omega_0)|^2 \rho_e(\omega_0), \tag{29}$$

where ω_0 is the transition frequency and ρ_e is the density (in energy) of final states

$$\rho_e(\omega_0)\hbar d\omega = \frac{V}{(2\pi)^3} d^3k = \frac{V}{(2\pi)^3} k^2 d\Omega_k dk$$
$$= \frac{V}{8\pi^3 c^3} n^2(\omega) \omega^2 \frac{d}{d\omega} [n(\omega)\omega] d\omega d\Omega_k, \quad (30)$$

where $d\Omega_{\mathbf{k}}$ is the differential element of solid angle about **k**. The rate of spontaneous emission into all solid angles and polarizations is then, from (28)–(30),

$$A' = \frac{2\pi}{\hbar} \frac{2\pi\hbar\omega_0\mu(\omega_0)}{n(\omega_0)\gamma(\omega_0)V} \frac{Vn^2(\omega_0)\omega_0^2}{8\pi^3 c^3\hbar}\gamma(\omega_0)$$
$$\times \sum_{\lambda} \int d\Omega_{\mathbf{k}} |\mathbf{d} \cdot \mathbf{e}_{\mathbf{k}\lambda}|^2 = n(\omega_0)\mu(\omega_0)A, \quad (31)$$

where $A = 4|\mathbf{d}|^2 \omega_0^3 / 3\hbar c^3$ is the free-space radiation rate. Eq. (31) differs from the familiar result cited earlier by the factor $\mu(\omega_0)$, which ensures that A' > 0 in a negative-index medium.

Absorption and stimulated emission are likewise affected. The Einstein B coefficient is calculated in the same manner as A' to be

$$B' = \frac{\mu(\omega_0)}{\gamma(\omega_0)n(\omega_0)}B,\tag{32}$$

where *B* is the coefficient for an atom in free space. Obviously B' > 0 for both positive- and negativeindex media. This generalizes the expression $B' = B/n^2(\omega_0)$ that appears frequently in the literature [8]. The latter is seen to be applicable if dispersion is negligible $[\gamma(\omega_0) \rightarrow n(\omega_0)]$ and $\mu(\omega_0) \approx 1$. The expressions given here for *A'* and *B'* also generalize the results obtained in Reference [6], where it was assumed that the host medium is nonmagnetic ($\mu = 1$).

The spectral density of thermal radiation may be obtained in the familiar way by assuming that the absorption and emission rates for a transition of frequency ω are equal in thermal equilibrium; thus

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$$\rho(\omega) = \frac{A'/B'}{e^{\hbar\omega/kT} - 1} = \frac{\gamma(\omega)n^2(\omega)A/B}{e^{\hbar\omega/kT} - 1}$$
$$= \frac{\gamma(\omega)n^2(\omega)\hbar\omega^3/\pi^2c^3}{e^{\hbar\omega/kT} - 1},$$
(33)

as obtained in [6].

We have shown that some of the unusual properties of a negative index of refraction material suggested by a classical analysis are verified when analyzed in terms of the quantized electromagnetic field. In the derivation, we have assumed the presence of a spectral band in which there is no absorption and $\epsilon(\omega)$, $\mu(\omega)$ and $n(\omega)$ are all negative. There are additional interesting phenomena that arise in the quantum case that we did not consider in this brief survey. Some interesting considerations involve the role of evanescent waves, for example, in focusing elements, in internal reflection, and in the propagation of radiation in photonic crystals.

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