SPONTANEOUS DECAY IN LEFT-HANDED MATERIAL

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A quantization scheme for the electromagnetic field interacting with atomic systems in the presence of dispersing and absorbing magnetodielectrics is presented, with special emphasis on left-handed materials. The theory is applied to the spontaneous decay of a two-level atom at the center of a spherical free-space cavity surrounded by magnetodielectric matter, and the problem of local-field corrections within the real-cavity model is addressed.

1 Introduction

Left-handed materials (LHMs) are magnetodielectric media which, in a certain frequency range, simultaneously exhibit negative permittivity and permeability; as a consequence the electric field, magnetic field and wave vector of a plane wave propagating through such a medium form a left-handed triad. This phenomenon, together with a number of additional unusual properties of LHMs such as negative refraction, reverse light pressure, reverse Doppler shift and reverse Cherenkov radiation, was first pointed out by Veselago.1 Since LHMs do not exist naturally, they have remained a merely academic curiosity until recent reports on the fabrication of metamaterials which act as LHMs in the microwave range, followed by experimental verifications of negative refraction, being the most prominent feature of LHMs.2

The analysis of nonclassical properties of radiation in the presence of realistic LHMs requires the quantization of the (macroscopic) electromagnetic field in the presence of dispersing and absorbing magnetodielectrics. We perform this task in the first part of the current work by means of a source-quantity representation based on the classical Green tensor in a similar way as for purely dielectric media.3,4 As a simple application of the quantization scheme, we then study the spontaneous decay of an excited two-level atom in a dispersing and absorbing magnetodielectric environment, with special emphasis on an atom in a spherical cavity. The way, in which a homogeneous magnetodielectric with real and positive permittivity and permeability influences the spontaneous decay of an embedded atom (disregarding local field

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corrections) can immediately be deduced from Fermi’s golden rule. While the electric field in the medium, and thus the dipole interaction responsible for the decay, corresponds to the electric field in free space times $1/\sqrt{\varepsilon}$, the mode density in the medium is proportional to $n^3$ (where $n = \sqrt{\varepsilon\mu}$ is the refractive index). We can thus conclude that the decay rate of the atom in the medium reads

$$
\Gamma = \left( \frac{1}{\sqrt{\varepsilon}} \right)^2 \times n^3 \times \Gamma_0 = \mu n \Gamma_0,
$$

with $\Gamma_0$ being the decay rate in free space. We will show how this formula can be generalized to the realistic case of dispersing and absorbing magnetodielectric matter, including local-field effects.

The article is organized as follows. In Section 2, a quantization scheme for the electromagnetic field in the presence of dispersing and absorbing magnetodielectrics is presented, while the interaction of the medium-assisted field with additional charged particles is considered in Sec. 3. In Sec. 4, the theory is applied to the spontaneous decay of an excited two-level atom. A summary and some concluding remarks are given in Sec. 5.

2 The quantized medium-assisted electromagnetic field

The quantization of the electromagnetic field in a causal linear magnetodielectric medium characterized by both complex $\varepsilon(\mathbf{r}, \omega)$ and complex $\mu(\mathbf{r}, \omega)$ can be performed by generalizing the theory \cite{3,4} for dielectric media. The operator-valued Maxwell equations in frequency space read

$$
\begin{align*}
\nabla \mathbf{D}(\mathbf{r}, \omega) &= 0, \quad \nabla \times \mathbf{E}(\mathbf{r}, \omega) = i\omega \mathbf{B}(\mathbf{r}, \omega), \\
\nabla \mathbf{B}(\mathbf{r}, \omega) &= 0, \quad \nabla \times \mathbf{H}(\mathbf{r}, \omega) = -i\omega \mathbf{D}(\mathbf{r}, \omega),
\end{align*}
$$

where

$$
\mathbf{D}(\mathbf{r}, \omega) = \varepsilon_0 \mathbf{E}(\mathbf{r}, \omega) + \mathbf{P}(\mathbf{r}, \omega), \quad \mathbf{H}(\mathbf{r}, \omega) = \mu_0 \mathbf{B}(\mathbf{r}, \omega) - \mathbf{M}(\mathbf{r}, \omega)
$$

\[\varepsilon_0 = \mu_0^{-1}].\] We have the constitutive relation for polarization,

$$
\mathbf{P}(\mathbf{r}, \omega) = \varepsilon_0 \varepsilon(\mathbf{r}, \omega) - \varepsilon_0 |\mathbf{E}(\mathbf{r}, \omega)|^2 + \mathbf{P}_N(\mathbf{r}, \omega),
$$

with $\mathbf{P}_N(\mathbf{r}, \omega)$ being the noise polarization associated with the electric losses due to material absorption, and similarly we introduce the constitutive relation for magnetization

$$
\mathbf{M}(\mathbf{r}, \omega) = \mu_0 [1 - \kappa(\mathbf{r}, \omega)] \mathbf{B}(\mathbf{r}, \omega) + \mathbf{M}_N(\mathbf{r}, \omega),
$$

where $\kappa(\mathbf{r}, \omega) = \mu_0^{-1}(\mathbf{r}, \omega)$, and $\mathbf{M}_N(\mathbf{r}, \omega)$ is the noise magnetization unavoidably associated with magnetic losses.

From Eqs. (2) - (5), we obtain by successive substitutions

$$
\nabla \times \kappa(\mathbf{r}, \omega) \nabla \times \mathbf{E}(\mathbf{r}, \omega) - \frac{i\omega^2}{\varepsilon(\mathbf{r}, \omega)} |\mathbf{E}(\mathbf{r}, \omega)|^2 \mathbf{E}(\mathbf{r}, \omega) = i\omega \mu_0 |\mathbf{M}_N(\mathbf{r}, \omega)|^2.
$$
where
\[ \hat{\mathbf{j}}_N(r, \omega) = -i \omega \mathbf{P}_N(r, \omega) + \nabla \times \mathbf{M}_N(r, \omega) \]  
(7)
is the noise current. The solution of Eq. (6) can be given by
\[ \mathbf{E}(r, \omega) = i \omega \mu_0 \int d^3r' \mathbf{G}(r, r', \omega) \hat{\mathbf{j}}_N(r', \omega), \]
(8)
where \( \mathbf{G}(r, r', \omega) \) is the (classical) Green tensor satisfying the equation
\[ \left[ \nabla \times \kappa(r, \omega) \nabla \times -\frac{\omega^2}{c^2} \varepsilon(r, \omega) \right] \mathbf{G}(r, r', \omega) = \delta(r-r') \]
(9)
together with the boundary condition at infinity. The Green tensor fulfills the useful relations\(^{5}\)
\[ \mathbf{G}^*(r, r', \omega) = \mathbf{G}(r, r', -\omega^*), \quad \mathbf{G}(r, r', \omega) = \mathbf{G}^\dagger(r', r, \omega), \]
\[ \int d^3s \left\{ -\text{Im} \kappa(s, \omega) \left[ \mathbf{G}(r, s, \omega) \times \nabla_s \right] \left[ \nabla_s \times \mathbf{G}^*(s, r', \omega) \right] \right. \]
\[ + \frac{\omega^2}{c^2} \text{Im} \varepsilon(s, \omega) \mathbf{G}(r, s, \omega) \mathbf{G}^*(s, r', \omega) \} = \text{Im} \mathbf{G}(r, r', \omega). \]
(10)
Analogously, noise polarization can be related to a bosonic vector field \( \mathbf{f}_p(r, \omega) \) via
\[ \mathbf{P}_N(r, \omega) = i \sqrt{\frac{\hbar \varepsilon_0}{\pi}} \text{Im} \varepsilon(r, \omega) \mathbf{f}_p(r, \omega), \]
(11)
and noise magnetization can be related to a bosonic vector field \( \mathbf{f}_m(r, \omega) \) via
\[ \mathbf{M}_N(r, \omega) = \sqrt{\frac{\hbar \varepsilon_0}{\pi}} \text{Im} \kappa(r, \omega) \mathbf{f}_m(r, \omega), \]
(12)
with commutation relations (\( \lambda, \lambda' = e, m \))
\[ [\mathbf{f}_{\lambda}(r, \omega), \mathbf{f}_{\lambda'}^\dagger(r', \omega')] = \delta_{\lambda\lambda'} \delta_{ij} \delta(r-r') \delta(\omega-\omega'), \]
(13)
\[ [\mathbf{f}_{\lambda}(r, \omega), \mathbf{f}_{\lambda'}(r', \omega')] = 0. \]
(14)
In this approach, the medium-assisted electromagnetic field is fully expressed in terms of the fundamental fields \( \mathbf{f}_\lambda(r, \omega) \) and \( \mathbf{f}_{\lambda'}^\dagger(r, \omega) \). In particular, the electric-field operator (in the Schrödinger picture) reads
\[ \mathbf{E}(r) = \int_0^\infty d\omega \mathbf{E}(r, \omega) + \text{H.c.}, \]
(15)
where \( \mathbf{E}(r, \omega) \) is given by Eq. (8) together with Eqs. (7), (11), and (12). Similarly, the other fields can be expressed in terms of \( \mathbf{f}_\lambda(r, \omega) \) and \( \mathbf{f}_{\lambda'}^\dagger(r, \omega) \), by making use of Eqs. (2) - (5), (11), and (12).
The Hamiltonian of the system composed of the electromagnetic field and the medium including the dissipative system can be given by

\[ H = \sum_{\lambda=e,m} \int d^3r \int_0^\infty d\omega \, \hbar \omega \mathcal{F}_\lambda(r,\omega) f_\lambda(r,\omega). \quad (16) \]

It can be shown that the operators for the electromagnetic field obey the correct (equal-time) commutation relations as well as the time-dependent Maxwell equations (in the Heisenberg picture).5

3 Interaction of the medium-assisted field with charged particles

In order to study the interaction of charged particles with the medium-assisted electromagnetic field, we first introduce the potentials

\[ \varphi(r) = \int_0^\infty d\omega \varphi(r,\omega) + \text{H.c.}, \quad \mathbf{A}(r) = \int_0^\infty d\omega \mathbf{A}(r,\omega) + \text{H.c.}, \quad (17) \]

which, in the Coulomb gauge, are related to the longitudinal and transverse components of \( \mathbf{E}(r,\omega) \), Eq. (8), via

\[ -\nabla \varphi(r,\omega) = \mathbf{E}^\parallel(r,\omega), \quad \mathbf{A}(r,\omega) = (i\omega)^{-1} \mathbf{E}^\perp(r,\omega). \quad (18) \]

For non-relativistic particles of masses \( m_\alpha \) and charges \( q_\alpha \), described by position and canonical momentum operators \( r_\alpha \) and \( p_\alpha \), the total Hamiltonian in the minimal coupling scheme can now be written as

\[ H = \sum_{\lambda=e,m} \int d^3r \int_0^\infty d\omega \, \hbar \omega \mathcal{F}_\lambda(r,\omega) f_\lambda(r,\omega) + \sum_\alpha \frac{1}{2m_\alpha} \left[ p_\alpha - q_\alpha \mathbf{A}(r_\alpha) \right]^2 \]
\[ + \frac{i}{2} \int d^3r \rho_\lambda(r) \varphi_\lambda(r) + \int d^3r \rho_\lambda(r) \varphi(r), \quad (19) \]

where the charge density and the scalar potential of the particles are given by

\[ \rho_\lambda(r) = \sum_\alpha q_\alpha \delta(r - r_\alpha), \quad \varphi_\lambda(r) = \int d^3r' \frac{\rho_\lambda(r')}{4\pi\varepsilon_0 |r - r'|}. \quad (20) \]

Supplementing the medium-assisted electromagnetic field with the longitudinal electric field associated with the charged particles, one can verify that the total electromagnetic field operators in the Heisenberg picture satisfy the appropriate Maxwell equations, and that the motion of the particles is in accordance with the Newton equations of motion.5

4
4 Spontaneous decay of an excited two-level atom

For a two-level atom (position $\mathbf{r}_A$, transition frequency $\omega_A$ between upper state $|u\rangle$ and lower state $|f\rangle$) in electric-dipole and rotating-wave approximations, the Hamiltonian (19) reduces to

$$H = \sum_{\lambda=e,m} \int d^3r \int_0^\infty d\omega \hbar \lambda \mathbf{f}_\lambda^\dagger(\mathbf{r},\omega) \mathbf{f}_\lambda(\mathbf{r},\omega) + \hbar \omega_A |u\rangle\langle u| - \left[ |u\rangle \langle u| \int_0^\infty d\omega \hbar \mathbf{E}(\mathbf{r}_A,\omega) + \text{H.c.} \right],$$  \hspace{1cm} (21)

where $\mathbf{d}_A = \langle u|\mathbf{d}_A|u\rangle$ is the (real) transition dipole moment. To study the spontaneous decay of an initially excited atom, we write the system wave function at time $t$ in the form of

$$\psi(t) = C_u(t) e^{-i\omega_A t} |\{0\}\rangle |u\rangle + \sum_{\lambda=e,m} \int d^3r \int_0^\infty d\omega e^{-i\omega t} \mathbf{C}_\lambda(\mathbf{r},\omega, t) |\mathbf{1}_\lambda(\mathbf{r},\omega)\rangle |f\rangle,$$  \hspace{1cm} (22)

$\mathbf{1}_\lambda(\mathbf{r},\omega) \equiv \mathbf{f}_\lambda^\dagger(\mathbf{r},\omega) |\{0\}\rangle$, where $C_u(t)$ and $\mathbf{C}_\lambda(t)$ are slowly varying amplitudes, and $\tilde{\omega}_A = \omega_A + \delta \omega$ is the environment-induced shifted transition frequency. The Schrödinger equation $i\hbar \partial_t \psi(t) = H \psi(t)$ then leads to a set of differential equations for $C_u(t)$ and $\mathbf{C}_\lambda(\mathbf{r},\omega, t)$, which has to be solved under the initial conditions $C_u(0) = 1$, $\mathbf{C}_\lambda(\mathbf{r},\omega, 0) = 0$. Formal integrations of the equation for $\mathbf{C}_\lambda(\mathbf{r},\omega, t)$ and substitution into the equation for $C_u(t)$ leads to

$$\dot{C}_u(t) = i\delta \omega C_u(t) + \int_0^t dt' K(t-t') C_u(t'),$$  \hspace{1cm} (23)

where

$$K(t-t') = -\frac{1}{\hbar \pi \varepsilon_0} \int_0^\infty d\omega \frac{\omega^2}{c^2} e^{-i(\omega - \tilde{\omega}_A)(t-t')} d\mathbf{A} \text{Im} \mathbf{G}(\mathbf{r}_A, \mathbf{r}_A, \omega) d\mathbf{A}. \hspace{1cm} (24)$$

Equations (23) and (24) formally look like those valid for non-magnetic structures. Since the matter properties are fully included in the Green tensor, the results only differ in the actual Green tensor.

In the weak-coupling regime, we may apply the Markov approximation, i.e., replace $C_u(t')$ in Eq. (23) by $C_u(t)$ and approximate the time integral according to

$$\int_0^t dt' e^{-i(\omega - \tilde{\omega}_A)(t-t')} \rightarrow \pi \delta(\omega - \omega_A) + i\mathcal{P} \frac{1}{\omega_A - \omega}. \hspace{1cm} (25)$$

Taking the imaginary parts of the resulting equation and choosing $C_u(t)$ to be real, we arrive at a self-consistent equation for the transition-frequency shift

$$\delta \omega = \frac{1}{\pi \hbar \varepsilon_0 \mathcal{P}} \int_0^\infty d\omega \frac{\omega^2}{c^2} \frac{d\mathbf{A} \text{Im} \mathbf{G}(\mathbf{r}_A, \mathbf{r}_A, \omega) d\mathbf{A}}{\omega_A + \delta \omega - \omega}. \hspace{1cm} (26)$$
Equation (23) then yields $C_u(t) = \exp(-\frac{1}{\tau} \Gamma t)$, where the decay rate $\Gamma$ is given by the well-known formula

$$\Gamma = \frac{2 \bar{\omega}_A^2}{\hbar \varepsilon_0 c^2} \text{d}A \text{Im} \left( G(r_A, r_A, \bar{\omega}_A) \right) \text{d}A. \quad (27)$$

4.1 Non-absorbing bulk material

Let us first consider the limiting case of non-absorbing bulk material, i.e., $\varepsilon(\bar{\omega}_A)$ and $\mu(\bar{\omega}_A)$ are assumed to be real. Using the bulk-material Green tensor, it can easily be seen that

$$\text{Im} \left( G(r_A, r_A, \bar{\omega}_A) \right) = \frac{\bar{\omega}_A}{2 \varepsilon_0 c^2} \text{Re} \left[ \mu(\bar{\omega}_A) n(\bar{\omega}_A) \right] |I| \quad (28)$$

where

$$n(\omega) = \frac{\varepsilon(\omega)}{\mu(\omega)} \left| \frac{\varepsilon(\omega) - i \gamma}{\varepsilon(\omega) + i \gamma} \right| \quad (29)$$

is the refractive index of the material. Substitution into Eq. (27) yields

$$\Gamma = \text{Re} \left[ \mu(\bar{\omega}_A) n(\bar{\omega}_A) \right] \Gamma_0, \quad (30)$$

where

$$\Gamma_0 = \frac{\bar{\omega}_A^3 d_A^2}{3 \hbar \varepsilon_0 c^3} \quad (31)$$

is the free-space decay rate, but taken at the shifted transition frequency. Eq. (30) is a generalization of Eq. (1) to arbitrary dispersive magnetoelectrics. In particular, when $\varepsilon(\bar{\omega}_A)$ and $\mu(\bar{\omega}_A)$ have opposite signs, then the refractive index defined according to Eq. (29) is purely imaginary, thereby leading to $\Gamma = 0$; spontaneous emission is completely inhibited. Note that material absorption always gives rise to a finite value of $\Gamma$.

4.2 Atom in a spherical cavity

For absorbing bulk material, the imaginary part of the Green tensor shows a singularity at equal positions, which can be avoided by accounting for the atom always being localized in a small free-space region. In order to achieve this, we separate the Green tensor into vacuum $G^V(r, r', \omega)$ and scattering $G^S(r, r', \omega)$ parts,

$$G(r, r', \omega) = G^V(r, r', \omega) + G^S(r, r', \omega), \quad (32)$$

resulting in the following expression for the decay rate given by Eq. (27):

$$\Gamma = \Gamma_0 + \frac{2 \bar{\omega}_A^2}{\hbar \varepsilon_0 c^2} \text{d}A \text{Im} \left( G^S(r_A, r_A, \bar{\omega}_A) \right) \text{d}A. \quad (33)$$
In particular, for a two-level atom placed at the center of a spherical (vacuum) cavity surrounded a homogeneous absorbing and dispersing magnetodielectric, by substituting the appropriate Green tensor into (33), we arrive at

\[
\frac{\Gamma}{\Gamma_0} = 1 + \text{Re} \left\{ \left[ 1 - i(n + 1)z - n(n + 1) \frac{\mu-n}{\mu-n^2} z^2 + i n \frac{\mu-n}{\mu-n^2} z^3 \right] e^{iz} \right. \\
\left. \times \left[ -i \sin z - (n \sin z - i \cos z)z + \left( \cos z - i \frac{1-\mu}{\mu-n} \sin z \right) n z^2 \right. \right. \\
\left. \left. - (n \sin z + i \mu \cos z) \frac{n^2}{\mu-n^2} z^3 \right]^{-1} \right\}
\]  (34)

\[\mu = \mu(\tilde{\omega}_A), \quad n = n(\tilde{\omega}_A), \quad \text{and} \quad z = R\tilde{\omega}_A/c, \text{with } R \text{ being the cavity radius},\] which generalizes the rate formula derived for purely dielectric matter.

4.3 Local-field corrections

Using the result (34), we can now obtain a more accurate expression for the decay rate of an atom in magnetodielectric bulk material, by taking into account local-field corrections. To this end, we perform the limit \( R\tilde{\omega}_A/c \ll 1 \) in Eq. (34), thus arriving at the real cavity model. We derive

\[
\frac{\Gamma}{\Gamma_0} = \text{Re} \left[ \left( \frac{3\varepsilon}{1 + 2\varepsilon} \right)^2 \frac{e^{i\omega_A t}}{1 + 2\varepsilon} \left( \frac{c}{\omega_A R} \right)^3 \right] + \frac{9}{5} \text{Im} \left[ \frac{\varepsilon(1 + 3\varepsilon + 5\mu\varepsilon)}{(1 + 2\varepsilon)^2} \right] \left( \frac{c}{\omega_A R} \right) + O(R).
\]  (35)

For transition frequencies that are sufficiently far away from any medium resonance frequency and the local-mode frequency \( 2\varepsilon(\tilde{\omega}_A) \approx -1 \), so that material absorption can be disregarded, Eq. (35) reduces to

\[
\Gamma \simeq \left[ \frac{3\varepsilon(\tilde{\omega}_A)}{1 + 2\varepsilon(\tilde{\omega}_A)} \right]^2 \text{Re} [\mu(\tilde{\omega}_A)n(\tilde{\omega}_A)] \Gamma_0,
\]  (36)

and hence the local-field correction simply results in multiplying the rate obtained for the case of non-absorbing bulk material [Eq. (30)] by the factor \( \left( 3\varepsilon(\tilde{\omega}_A)/(1 + 2\varepsilon(\tilde{\omega}_A)) \right)^2 \).

5 Summary and conclusions

We have quantized the electromagnetic field in the presence of causal magnetodielectric matter, including LHMs. The quantization scheme is based on a source-quantity representation of the medium-assisted electromagnetic field in terms of the classical Green tensor and two independent infinite sets of appropriately chosen bosonic basis fields. We have further presented the minimal-coupling Hamiltonian governing the interaction of the medium-assisted electromagnetic field with additional charged particles. The theory can be used
to study the influence of magnetodielectric bodies on a wide class of phenomena, such as the generation and propagation of nonclassical radiation, Casimir forces, van-der-Waals forces etc.

As an example, we have treated the spontaneous decay of a two-level atom in the presence of arbitrarily configured, dispersing and absorbing media, deriving general expressions for the decay rate and the frequency shift in terms of the classical Green tensor. To be more specific, we have studied the decay rate of an atom at the center of a spherical cavity surrounded by a magnetodielectric, obtaining the real-cavity model for including local-field corrections in the decay rate of the atom in bulk material, by taking the limit of small cavity radii.

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