Coherent control of light transport in a dense and disordered atomic ensemble

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Light transport in a dense and disordered cold atomic ensemble, where the cooperation of atomic dipoles essentially modifies their coupling with the radiation modes, offers an alternative approach to light-matter interfacing protocols. Here, we show how the cooperativity and quasi-static dipole interaction affect the process of light propagation under the conditions of electromagnetically-induced transparency (EIT). We perform comparative analysis of the self-consistent approach with ab-initio microscopic calculations and emphasize the role of the interatomic interaction in the dipoles’ dynamics. Our results show that in such a dense and strongly disordered system the EIT-based light storage protocol stays relatively insensitive to configuration variations and can be obtained with essentially less atoms than it is normally needed for dilute configurations.

Coherent control of light propagation through a cold and optically deep atomic ensemble has been the basis for a variety of remarkable light-matter interfacing protocols, including single-photon or entanglement storage in Raman- or EIT-based quantum memories with applications to quantum information networks [1–6]. In these experiments, and in most associated theoretical studies, relatively low densities and dilute configurations are considered [7, 8]. Interestingly, with higher density – i.e. up to one atom in a volume of radiation wavelength – the effective interface between light and matter and reliable light storage can be obtained with essentially less atoms. In the diffraction limit, for a given optical depth \( b_0 \), the minimal number of required atoms scales indeed as \( N \approx b_0^2/(n_0 \lambda^3) \), where \( n_0 \) is the density of atoms and \( \lambda = k^{-1} \) is the inverse wave number of the radiation field [9]. This estimate can even lead to a smaller number of atoms if the light is transversally confined into a nanophotonic waveguide, e.g. in tapered nanofibers where the divergence associated with Fraunhofer diffraction vanishes [10, 11]. Such appealing configuration offers a promising platform for a new generation of light-matter interfaces [12, 13].

However in the limit of high density and strong disorder, when \( n_0 \lambda^3 \sim 1 \), the cooperative dynamics and static interatomic interactions modify the scattering process and can lead for instance to localization phenomena in the light transport [8, 14, 16]. These features are also crucially important when the transport is controlled by external coherent fields. In this Letter, we therefore address the coherent Raman control of a signal pulse entering such a medium. We present comparative analysis of the problem for Λ-type atoms. Our investigation is based on the self-consistent macroscopic Maxwell description and on the alternative ab-initio microscopic calculation of the scattering process. Our results, confirmed by both calculation schemes, with addressing to atomic arrays with small number of atoms, indicate the ways for further optimization of the atomic memory protocols.

Self-consistent approach - The energy level diagram and excitation scheme are shown in Fig. 1. We consider a dense ensemble of the simplest Λ-type atoms with the minimal accessible number of quantum states, i.e. with angular momentum \( F_0 = 1 \) in the ground state and \( F = 0 \) in the excited state. Initially, all the atoms populate only one Zeeman sublevel, \( \{ F_0 = 1, M_0 = 1 \} \). Two coherent control modes are applied at the empty transitions and a weak left-handed polarized probe propagates through the sample. In the lowest order, the transfer of atoms into \( \{ F_0 = 1, M_0 = −1, 0 \} \) sublevels is negligible and the steady state dynamics of an atomic dipole in the ensemble, similar as in [15], can be expressed by the following equation for its positive frequency component \( d^{(+)}(\omega) \):

\[
-i\omega d^{(+)}(\omega) = -i \left[ \omega_0 + \frac{\Omega_\xi^2}{2(\omega - \omega_c)} \right] d^{(+)}(\omega) + \frac{i}{\hbar} \frac{d_0^2}{4} \left[ \mathcal{E}^{(+)}(\omega) + \frac{4\pi}{3} \mathcal{P}^{(+)}(\omega) \right] - i \Sigma(\omega) d^{(+)}(\omega), \tag{1}
\]

\( \omega_0 \) denotes the transition frequency, \( \Omega_\xi \) the Rabi frequency for the control modes of frequency \( \omega_c \) and \( d_0 = |(d \cdot e)_{nm}| \) is the modulus of the transition matrix element between \( \{ n \equiv F = 0, M = 0 \} \) and \( \{ m \equiv F_0 = 1, M_0 = 1 \} \). The probe driving amplitude \( \mathcal{E}^{(+)}(\omega) \) is the macroscopic transverse electric field at frequency \( \omega \), considered at the point of the dipole’s location and \( \mathcal{P}^{(+)}(\omega) \) is the local mesoscopically averaged polarization. The presence of this last term in Eq. 1 corresponds to the so called Lorentz-Lorenz or local field correction associated with the longitudinal (quasi-static) interaction of the dipole with its local environment. Since in the self-consistent model the proximate dipoles are indistinguishable in the excitation process, the averaged polarization can be writ-
where \( n \) is the electric susceptibility of the sample.

FIG. 1: (Color online) Excitation scheme of a dense and disordered atomic ensemble in the presence of control modes. The atoms have a total spin angular momentum \( F_0 = 1 \) in the ground state and \( F = 0 \) in the excited state. A weak probe beam with \( \sigma_+ \) polarization propagates along the sample. The two control modes with equal Rabi frequencies \( \Omega_c \) address the empty adjacent transitions with orthogonal polarizations \( \sigma_+ \) and \( \pi \).

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\[
P^{(+)}(\omega) = n_0 d^{(+)}(\omega) = \chi(\omega) \mathcal{E}^{(+)}(\omega) \tag{2}
\]

where \( n_0 \) is the local density and \( \chi \) the macroscopic dielectric susceptibility of the sample.

The radiation losses, given by the last term in Eq. 1, can be expressed as follows

\[
\Sigma(\omega) = \frac{\omega^2 d_0^2}{\hbar^2 c^2} \int \frac{d^3 k}{(2\pi)^3} D^{(\perp)}_{ll}(k, \omega) = -i \left[ \frac{\gamma_o + \gamma_c(\omega)}{4} \right] + \Delta_{\text{Lamb}} \tag{3}
\]

It corresponds to the radiation damping created by the transverse field emitted in the scattering process and resulting into losses of the probe escaping the sample incoherently, i.e. out of its original propagation direction. The last term in the right hand side, \( \Delta_{\text{Lamb}} \), selects a diverging contribution to the vacuum Lamb shift, which should be renormalized and incorporated into a physical energy of the atomic transition. The integral evaluated over wave vector together with the sum over the tensor indices \( l = x, y, z \) recover the spatial components of the field emitted by the dipole and overlapping with its own location. For an infinite medium with anisotropic dielectric susceptibility, responding only on a left-handed polarized mode, the trace (sum over \( l = x, y, z \)) of the transverse electric field Green’s function, contributing in Eq. 3, is given by

\[
D^{(\perp)}_{ll}(k, \omega) = -\frac{4\pi \hbar}{k^2} \left\{ 2 + \frac{\omega^2}{c^2 k^2 - \omega^2} + \frac{\omega^2 [1 + 4\pi \chi(\omega)]}{c^2 (k^2 - k^2) 2\pi \chi(\omega)} \right\} \tag{4}
\]

Evaluation of the integral leads to \( \gamma_o = \gamma \) and

\[
\gamma_c(\omega) = \frac{4 \hbar^2 (\omega)^3}{\pi c^2} \sqrt{1 + 4\pi \chi(\omega) \over 2\pi \chi(\omega)} \arcsin \sqrt{\frac{2\pi \chi(\omega)}{1 + 2\pi \chi(\omega)}}. \tag{5}
\]

We associate \( \gamma_o \) with radiation emission into the ordinary mode (first denominator’s pole in Eq. 3) and \( \gamma_c \) with emission into the extraordinary mode (second denominator’s pole in Eq. 4). The last value contains not only density correction of the decay rate, but also cooperative correction to the radiation Lamb shift. The key feature of this result is the existence of two different modes in anisotropic medium, namely ordinary and extraordinary, with decay rates different for the emission into each of them.

The above formulas enable to obtain a closed algebraic equation for the local dielectric susceptibility \( \chi(\omega) \). This equation can be solved numerically and further applied for the description of the signal pulse transport through the atomic sample. In particular, for a slab sample of length \( L \) filled in by the medium with dielectric constant \( \epsilon(\omega) = 1 + 4\pi \chi(\omega) \) the transmission amplitude at frequency \( \omega \) is given by

\[
T_\omega = \frac{2 \sqrt{\epsilon(\omega)}}{2 \sqrt{\epsilon(\omega) \cos \psi(\omega) - i(1 + \epsilon(\omega)) \sin \psi(\omega)}} \tag{6}
\]

where \( \psi(\omega) = L \sqrt{\epsilon(\omega)} \omega / c \). The transmission coefficient \( |T_\omega|^2 \) can be calculated and compared with the counterparting result of the microscopic calculations presented later in the paper.

As a first result, figure 2 provides the spectral dependencies of the dielectric susceptibility calculated in the self-consistent approximation for the atomic density \( n_0 k^3 = 1 \). The graphs show how the original single-atom resonance profile is modified by interatomic interactions via static longitudinal and radiation transverse fields. The resonance point is first shifted to the red wing due to the local field correction and the spectral profile also differs from its original Lorentzian shape. With control fields with \( \Omega_c = \gamma \) tuned at the point of resonance, the susceptibility exhibits typical signatures of the electromagnetically induced transparency, i.e. transparency window and reduced group velocity. In the considered case the overall resonance coupling strength, expressed by the transmission amplitude \( |T_\omega| \), can attain the level required for effective pulse delay under EIT protocol, for an atomic array consisting of only one hundred atoms. This is shown in Fig. 2 where the transport of a Gaussian pulse through such a short atomic array practically without losses is displayed. We now turn to the microscopic approach.

**Microscopic approach –** In ab-initio microscopic quantum theory the scattering process is described by the standard \( T \)-matrix formalism [17], which has been adjusted for calculation of light scattering on an ensemble.
atomic state \( q' \) can be any of the accessible ground states of the collective atomic subsystem. The sum expands over all the allowed output scattering channels and the frequency of outgoing photon \( \omega' = \omega \) for either Rayleigh or elastic Raman-type transitions shown in figure 1. The quantization volume \( V \) is internal and finally vanishing parameter of the theory. It should not be confused with the target volume \( V = L^3 \). The second line of Eq. (7), known as the optical theorem, links the total cross section with the elastic scattering amplitude in forward direction. This makes possible calculation of the total cross section even for extremely high number of the output scattering channels \( q' \). The spectral dependence \( Q_0(\omega) \) describes the microscopic and configuration dependent spectral behavior of the scattering process.

For the system of atomic dipoles the \( T \)-matrix can be expressed by the resolvent operator (many particle Green’s function) as follows:

\[
T_{q' e' k', g e k}(E) = \frac{2\pi\hbar\sqrt{\omega'\omega}}{V} \sum_{b,a=1}^{N} \langle \tilde{R}(E) | m_{a} - 1, n, m_{b} + 1 \rangle \langle m_{b} - 1, n, m_{a} + 1 \rangle
\]

where the resolvent \( \tilde{R}(E) = P(H - E)^{-1}P \) is projected by the operator \( P \) onto field vacuum state and onto a set of atomic states with a single optical excitation between the states \( m \) and \( n \). \( H \) is the total system Hamiltonian in the dipole gauge \([15]\). Indices \( a \) and \( b \) numerate the atoms and indicate their locations and occupation states. As in the previous section we assume that the excited atomic level has only one state \( n \), but the atoms can be repopulated among all the ground state Zeeman sublevels \( m \) in the interaction process. The resolvent can be calculated numerically as shown in Ref. \([13, 15]\) and the presence of the control field can be incorporated into its self-energy part via similar term as in the right-hand side of Eq. (1).

The transmission coefficient \( |T_{\omega}|^2 \) defined by Eq. (7) and the spectral profile of the scattering cross section \( Q_0(\omega) \) are different quantities. Nevertheless in classical optics, in accordance with Babinet’s principle, the light scattered by highly absorbing macroscopic sample has its scattering cross section equal to \( 2A \), where \( A \) is geometrical cross-area of the object. In the vicinity of the absorption resonance we can thus expect the approximate relation \( 2A[1 - |T_{\omega}|^2] \sim Q_0(\omega) \), which can be used to test the validity of the self-consistent microscopic description via round of microscopic calculations. In the spectral domain where the sample becomes partially transparent both calculation schemes approach the same limit of weak light scattering on a collection of independent point-like atomic dipoles. In this limit \( Q_0(\omega) = N\sigma(\omega) \), where \( N = n_0V \) is the number of scatterers and \( \sigma(\omega) \)
the cross section of light scattering on a single atom. In
the case of weak interaction the latter quantity also con-
tributes to the susceptibility $4\pi k \text{Im} \chi(\omega) = n_0 \sigma(\omega)$ such that we get asymptotic relation between both the calcu-
lation schemes.

In the following we investigate how the effect of dis-
order, associated with the dense random distribution
of atomic scatterers, can affect the previous result and
make absorption profile potentially sensitive to a spe-
cific atomic configuration. The supplementary details
of the microscopic calculation scheme can be found in
[9, 15]. Generally for a large atomic ensemble consist-
ing of $N$ atoms with degenerate ground state the exact
resolvent operator is defined in an Hilbert subspace of
large dimension, which is exponentially rising up with
$N$ as $d_\text{e} N d_\text{g}^{-1}$. Here $d_\text{g}$ and $d_\text{e}$ are the degeneracies of
the ground and excited states respectively. By evalu-
ating the total cross-section via the optical theorem (7)
we can take into account that in the Feynman diagram ex-
pansion for the diagonal matrix element of the resolvent
operator the off-diagonal components can contribute only
in the inner parts of the diagrams with tracking recurrent
coupling and virtual repopulation among the neighbor-
ing atoms sharing a single excitation shown in Fig. 1
as detailed in [6]. For intermediate densities, $n_0 \chi^3 \sim 1$,
in the self-energy diagrams it is sufficient to keep such
off-diagonal coupling only with the atoms separated by a
distance of few $\lambda$. This can essentially reduce the number
of inelastic scattering channels coupled with the elastic
channel and as a consequence the subspace dimension
for the resolvent operator as well as the number of equa-
tions to be solved, which would scale now as $d_\text{e} N d_\text{g}^{-1}$. Here $n-1$ corresponds to the effective number of the
proximate neighbors, which mainly contributes in the rec-
current diagrams responsible for “dressing” of an excited
state propagator associated with any randomly selected
atomic excitation in ensemble. In practical calculations
this number is varying up such that we can verify that the
entire calculation scheme with increasing $n$ becomes rapidly self-converging and in the limit of $n \gg n_0 \chi^3$ it
approaches the exact result.

This behavior is demonstrated in Fig. 3 for an en-
sample consisting of fifty atoms with density $n_0 \chi^3 \sim 1$
and for a number of proximate neighbors contributing in
the recurrent coupling up to five. The calculations were
done for a particular configuration and reproduce a
randomly created quasi-energy spectral structure of the
resolvent operator for the chosen configuration. As a ref-
ence dependence we have shown here a fragment of the
transmission spectrum calculated in the self-consistent
approximation near the resonance point and applied the
Babinet’s principle for the total cross section estimate at
the resonance. Both the calculation scheme are in
agreement in their general behavior and microscopic re-
sult has a clear signature of the local field correction be-
ing slightly asymmetric to the red wing. The fact that

![Graph of total cross-section vs. energy change](image)

FIG. 3: (Color online) Total cross section for a single photon scattering on an ensemble consisting of fifty atoms randomly
distributed with a density $n_0 \chi^3 \sim 1$. The microscopic cal-
culations have been performed for a particular configuration
and for different numbers $n$ of proximate neighbors involved
in the recurrent coupling, as described in the text. The in-
set shows the microscopic verification of EIT interaction with
control pulses calculated for the same parameters as used in
Fig. 2. The self-consistent (SC, dotted line) estimate of the
cross section spectrum (black dotted) is scaled in accordan
cese with the Babinet’s principle and only the part of maximal
absorption is shown, see text.

the microscopic cross section is a bit larger at the point
of maximum indicates that the macroscopic description
is not so straightforwardly applicable to system of meso-
scopic size. There is no here real contradiction between
micro and macroscopic descriptions because the sample
size $L = \sqrt[5]{50} \lambda$ hits the region $\lambda < L < \lambda$, so it is not
large enough to follow the Fraunhofer diffraction and the
Babinet’s benchmark. Unfortunately it is demanding so
far to extend the applied algorithm here up to macro-
scopic object consisting of a larger number of atoms and
overcoming the conditions $L > \lambda$ as it was done in [12]
for V-type two level atoms. Nevertheless the absorption
profile is realistically reproducible by the self-consistent
calculation scheme and variations in the atomic config-
uration only slightly affects the spectral behavior of the
cross section near the central resonance. The EIT phe-
nomenon can be involved in the microscopic calculation
scheme by adding the self-energy part from Eq.(11)
in the atomic propagators of unoccupied states. The dip
of transparency window, shown in inset and calculated
for the same parameters as in Fig. 2 is also little sensi-
tive to a particular atomic configuration.

In conclusion, we have shown that in a dense and dis-
ordered atomic ensemble the effective slow-light trans-
port and storage can be obtained in atomic array with
a relatively small number of atoms compared with di-
lute systems. The effect of disorder, which could be
expected to randomize the transmission spectrum, has
only slight signature of the configuration dependence.
The scattering spectra demonstrate indeed a tendency to self-averaging and smooth behavior near the main resonance peak, which is in turn reliably reproducible by the self-consistent model. As a consequence this allows effective coherent control of a signal pulse under the regime of EIT with a fixed set of external parameters, such as Rabi frequency and control mode detuning, irrespective of any random realizations of the atomic configuration. Extension of this study to one-dimensional array, such as obtained with atoms trapped along an optical nanofiber where disorder can manifest due to limited filling factor, is work for future.

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