Manipulating Light with Quantum Coherence in Solid Hydrogen

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Abstract

We discuss how the light propagation can be manipulated in solid hydrogen using Raman scheme based on electromagnetically induced transparency. We describe for two cases, far-off-resonance Raman scattering scheme and resonant Raman scheme. For the far-off resonance case, we demonstrate the existence of normal mode which governs the propagation in the far-off resonance scheme. We show that a strong driving field allows us to slow down the pulse-peak velocity of the Stokes and anti-Stokes fields to order of $c/10,000$ with the splitting of the temporal profile of the sideband-field pulses into a two-peak structure originating from the normal modes. For the resonant Raman medium, we propose a novel optical fiber scheme in which one can slow down the light propagation via evanescent field interaction using a tapered optical fiber whose thin region is embedded in solid hydrogen.

Introduction

Coherent control of dispersion and absorption properties of atoms and molecules has led to many spectacular quantum optical phenomena. In recent years, it has been demonstrated that the steep dispersion associated with a transparency window in dynamically controllable quantum interference; electro-magnetically induced transparency (EIT) [1], in resonant atomic or molecular gas mediums can be used as an excellent tool for controlling the group velocity of a light pulse in the medium. Examples are drastic reduction of the group velocity of light, freezing of light pulse, storage of quantum information of light, superluminal group velocity propagation, and control of the group velocity between subluminal to superluminal propagation [2-6].

In the present work, we discuss the slow light propagation in solid medium, especially on solid hydrogen. We describe two schemes. Firstly, we discuss the propagation normal mode for a far-off resonance Raman scheme. We show that regarding the slow normal mode the group velocity can be down to the order of $c/10,000$ even for a transparent medium using a pure
vibrational Raman transition of parahydrogen crystal [7]. Secondly, we propose a scheme to manipulate the pulse propagation in an optical fiber. The scheme is to use a resonant Raman transition via a vibrational transition of orthohydrogen molecule doped in parahydrogen crystal matrix. We use nuclear spin sublevels of orthohydrogen molecule. We show numerically that the light pulse can be slowed down via evanescent field interaction in an optical fiber using a tapered fiber, whose thin region is embedded in solid hydrogen crystal [8].

Before we proceed, we briefly summarize some main features of solid hydrogen. Solid hydrogen is the simplest molecular crystal consisting of H₂ molecules [9]. In this work, we use almost pure parahydrogen crystal. Regarding the experimental procedures how to grow the crystal, one can find the details in Ref. [10]. The H₂ molecules in the solid have well-defined vibrational and rotational states like free molecules in the gas phase. The characteristic frequencies of these quantum states are almost the same as the gas phase values. Moreover, the spectral widths of the transitions are extremely narrow, sometimes in the order of MHz, that is much narrower than the widths for gas phase molecules [11,12]. Regarding ortho-H₂ molecules in the crystal, they behave as impurities, and the ortho-H₂ concentration can be controlled from 40 ppm to 5 %. One remarkable feature of orthohydrogen molecule in the solid is that the vibrational transition becomes allowed transition due to the crystal field effect. We use this special nature for the resonant scheme.

Far-Off Resonance Scheme

The energy-level diagram of the Raman transition is illustrated in Fig. 1, which is for para-H₂ system. States a and b correspond to the vibrational states ν=0, J=0 and ν=1, J=0, respectively. The coupling and probe lasers are far from resonance with the intermediate states j which are located more than 90,000 cm⁻¹ above the ground state. Under a strong coupling field at frequency ω₀, we observe the propagation of a probe field at a Raman sideband ω₁(+1) and a new field at the opposite Raman sideband ω₁(-1). The latter is generated by the parametric interaction. One can readily suspect that the two sideband pulse waveforms do not propagate independently; They should be coupled each other. Theoretically, one can find two normal modes for the pulse envelope propagation, dark-state mode and bright-state mode.

![Energy level diagram for the slow light propagation in a far-off resonance system](attachment:image.png)

**Fig. 1.** Energy level diagram for the slow light propagation in a far-off resonance system
Dark-state mode is decoupled from the field-medium interaction and propagates with a vacuum speed of light. Bright-state mode propagates with a reduced group velocity [13].

Regarding the experiments, to reduce the pulse shape distortion and, consequently, to have a well-defined group velocity, the medium must adiabatically follow the applied fields. Therefore, the experiments were carried out for the Raman detunings which were larger than the Fourier-transformation limited spectral width of the probe laser.

Figures 2(a) and (b) display the temporal profiles of the probe field for the detunings of -50 (±15) MHz (a) and +70 (±15) MHz (b) with the coupling field intensities of 600 MW/cm² and 1 GW/cm². Both experimental and theoretical profiles are displayed in the figure, by solid and dashed lines, respectively. The temporal behavior of the generated new field was essentially the same as that of the probe field. We first discuss the experimental observations. The probe pulses are amplified by a factor of three for both detunings at the coupling intensity of 1 GW/cm², but the temporal profiles reveal very different evolution for each detuning. For the detuning of -50 MHz, see Fig. (a), the pulse-peak delay increases with the coupling laser intensity while the pulse waveform is preserved; the pulse-peak delay reaches about 3 ns at the coupling intensity of 1 GW/cm². This pulse-peak delay is a clear signature of slow group velocity; the group velocity is estimated to be $3 \times 10^4$ m/s ($c/10,000$). On the other hand, we observed a very peculiar temporal behavior at +70 MHz detuning as shown in Fig. (b). As the coupling intensity is increased, the probe pulse evolves to a two-peak structure. The two-peak structure becomes prominent when the coupling intensity is above 800 MW/cm². The splitting between the two peaks is about 10 ns at the coupling intensity of 1 GW/cm². We attribute the observed pulse splitting to the splitting between the two normal modes propagating with different group velocities. The observed splitting readily leads to a group velocity of $1 \times 10^4$ m/s ($c/30,000$) for the
slow normal mode [14].

In order to get insight into the above results, we recall the normal modes of the Raman medium. Based on the normal mode picture, the probe field is understood as a superposition of the two normal modes. At the entrance of the medium, the two normal-mode components of the probe field have comparable amplitudes and opposite phases. Therefore, these components partially cancel each other. The relative phase between the normal modes changes with the propagation distance. The beating between the normal modes becomes less destructive through the propagation, and substantial enhancement of the probe field may be obtained even when the group delay is much less than the probe pulse width, see Fig. 2(a). When the group delay is comparable to or larger than the probe pulse width, the slow coupled mode is separated from the uncoupled mode. Hence, a double-peak structure in the probe field may be observed, with the peak amplitude enhanced as compared to that at the input, see Fig. 2(b). We emphasize the key features of the slow light in our experiment: during the propagation process, the amplitude of the uncoupled mode does not change, the amplitude of the slow coupled mode undergoes two-photon absorption, but the amplitude of the probe field is enhanced. Such enhancement is a result of the slow group velocity and the associated phase shift of the slow coupled mode, and is different from the conventional Raman amplification. The observed amplification can be understood as the result of such enhancement mechanism.

Theoretical temporal profiles of the probe pulse were calculated and are superimposed on the experimental results in Figs. 2(a) and (b). Although some discrepancies are seen for the probe amplitudes and the pulse splitting, the essential features of the experimental observations are reproduced in the theoretical profiles. The theoretical pulse-peak delay at the detuning of -50 MHz reaches 2.5 ns for the coupling intensity of 1 GW/cm². The two-peak structure is clearly observed for the +70 MHz detuning. Regarding the discrepancies, the calculated amplification factor is four at 1 GW/cm², 33 % larger than the experimental value, and the group delay of the second peak is calculated to be 18 ns, which is about twice larger than the experimental value 10 ns. These discrepancies might be attributed to the overestimation in the calculation where the temporal and spatial non-uniformities of the coupling laser beam were not included.

**Slow Light in a Thin Optical Fiber**

In order to extend the slow light physics to real applications, it would be meaningful to develop a technique to manipulate the slow light propagation through an optical fiber. Here, we propose a configuration to tailor the dispersion properties of a fiber mode propagating inside a thin (tapered) optical fiber surrounded by an EIT medium [see Fig. 3], and as a result we show that the group velocity of the fiber mode can be reduced significantly. This method utilizes EIT and hence has the advantage of large control over the dispersion property of the fiber mode. It should be noted that our usage of "thin fiber" should be understood as the case when the diameter
of the fiber $2a > \lambda$, where $\lambda$ is the central wavelength of the field that propagates inside the fiber.

![Configuration to tailor the pulse propagation in an optical fiber.](image)

Under such a condition large part of the energy of the fiber mode lies outside the thin fiber, evanescent region of the thin fiber. We introduce ortho-$\text{H}_2$ doped in a matrix of para-$\text{H}_2$ as an EIT medium surrounding the fiber to demonstrate the above mentioned reduction in the group velocity.

Orthohydrogen molecules (with nuclear spin $I=1$) doped in the solid parahydrogen ($I=0$) molecular crystal is used as an EIT medium. The transition under consideration is a crystal field induced vibrational-rotational transition $Q_1(1)$ ($v=1\leftarrow 0$, $J=1\leftarrow 1$, $M_I = -1\leftarrow 0$, $0\leftarrow 1$). This transition occurs at infrared $\lambda=2.4\mu\text{m}$, and by applying an axial magnetic field, the degeneracy in the hyperfine levels $I=1$ can be removed to obtain a six level scheme as depicted in the Fig. 4. We should note that the choice of this medium as the EIT medium is of particular importance to the present problem, due to the following: (a) Solid hydrogen can grow in a cell in which an optical fiber is set, (b) this solid has very narrow spectral widths compared to other solid EIT medium, (c) since we are working here with the solid medium, we can neglect the transit time broadening.

![Schematic diagram for EIT using nuclear magnetic sublevels of ortho- hydrogen molecule doped in parahydrogen crystal.](image)

Fig. 3. Configuration to tailor the pulse propagation in an optical fiber.

Fig. 4. Schematic diagram for EIT using nuclear magnetic sublevels of ortho- hydrogen molecule doped in parahydrogen crystal.
The magnetic degenerate sub-levels can be coupled selectively by choosing suitable polarizations of the laser fields. We use a strong $\sigma^+$ polarized laser field that couples $|2> \leftrightarrow |4>$ and $|3> \leftrightarrow |5>$ transitions, and a $\sigma^-$ polarized weak probe field that couples the $|1> \leftrightarrow |5>$ and $|2> \leftrightarrow |6>$ transitions.

The propagation of the probe field inside the thin fiber was numerically calculated using the two-dimensional beam propagation method. In Fig. 5, we show the variation of the slope of the refractive index along the transverse direction. It is clear that the slope is positive only in the region close to the fiber, and hence favorable for the slow group velocity. For $|x|>2 \mu m$ the slope decreases and becomes negative for $|x|>4 \mu m$. However, the contribution from the latter part is extremely small and virtually no effects on the propagation. We should note that the group velocity reduction in a fiber is larger than that in a bulk EIT medium (without fiber), which is due to following reason: group velocity of probe pulse is directly proportional to the control field intensity. In case of a fiber, the control field intensity decreases along the transverse direction causing the tail part of the mode to move extremely slowly, which reduces the group velocity of the fiber mode drastically. However in the case of a bulk sample, the intensity is assumed to be constant along the transverse direction. Therefore with the same peak value for control field, the probe field pulse velocity is slower in the fiber compared to that in the bulk medium. We have shown that the slow velocity can be down to the order of 100 m/s.

Regarding the experiments, we have measured the orthohydrogen absorption spectrum at 2.4 $\mu m$ wavelength region with a resolution better than 1 MHz. The spectrum showed extremely narrow spectral widths less than 10 MHz fwhm, revealing the orthohydrogen molecule in solid parahydrogen matrix as a very good solid medium for EIT works.

![Fig. 5. Variation of slope of the real part of the refractive index for a fiber mode in the transverse direction.](image)
Prospect

We have discussed how the light can be manipulated using solid hydrogen in terms of the propagation characteristics. Essential point why the solid hydrogen is so special is that solid hydrogen satisfies almost single molecular quantum characteristics as in the gas phase molecules, and moreover it has high molecular number density of $10^{22}/cm^3$. Using the special nature of solid hydrogen, many fascinating ideas may be brought into the optical processes other than the processes we discussed here. We hope that many researchers can join to this new field which may remove the boundary between two categories of optical physics originating from atomic/molecular physics and solid state physics.

References