Using molecules as a quantum interface to store ultrashort optical vortices.

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Abstract. We exploit gas-phase molecules as light-matter interface to store an orbital angular momentum (OAM) or a superposition of OAM states (OAM-based photonic qubits) carried by ultrashort laser pulses. The interplay between spin angular momentum and OAM is exploited to encode the amplitude and spatial phase information of light beams into rotational coherences of molecules. This last is restored on-demand over tens of picoseconds with a reading beam by taking advantage of field-free molecular alignment. The underlying mechanism at the origin of the storage can be interpreted by the spatial structuring of the molecular sample induced by the field. The excitation indeed produces an inhomogeneous spatial distribution of molecular alignment (amplitude & orientation) whose periodical revivals associated with the quantum beatings of the rotational wavepacket enables to restore the spatial beam structure on-demand. The strategy is successfully demonstrated in CO_2 molecules at room temperature. Besides applicability as storage medium with THz bandwidth application, the use of molecules as light-matter interface opens new functionalities in terms of optical processing and versatile control of OAM fields.

1 Introduction

Beams of transverse phase distributions $e^{i\ell\varphi}$ (with φ the azimuthal coordinate in the beam section) are known to carry an orbital angular momentum (OAM) of $\ell\hbar$ per photon, where ℓ=0, ±1, ±2… is the so-called topological charge. Such phase-structured light beams have become these last years a subject of widespread interest with unprecedented applications for optical communication, super-resolution, optical tweezers, chirality or quantum processing [1]. Here, we experimentally demonstrate that gas-phase molecules can be used as a light-matter interface to store and manipulate the OAM carried by ultrashort laser pulses. The experiment is conducted in CO_2 molecules at room temperature in which pure OAM states ℓ=±1, 2, as well as a superposition of ℓ=1,-1 have been encoded [2]. The overall writing and reading processes rely on a pump-probe scheme with circular polarization, exploiting the interplay between OAM and spin angular momentum (SAM).

2 Principle and main results

The experiment is conducted in CO_2 at room temperature. The coupling scheme relies on a non-resonant Λ excitation scheme depicted in Fig. 1, well-suited for the storage of spatial information carried by broadband laser pulses. As shown, an ultrashort pump pulse consisting of a Stokes field carrying the OAM state (or the OAM superposition) to be encoded and of an auxiliary anti-Stokes field (ℓ = 0) interact with the molecular sample.

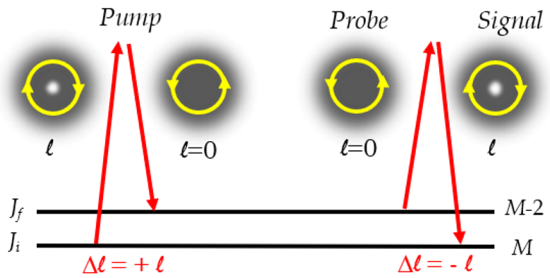

Fig. 1. Coupling scheme for storing an OAM (or a superposition of OAM states) into rotational states of molecules.

This interaction stores the spatial information (phase and amplitude) of the Stokes beam, analogously to holography, by producing a rotational wavepacket in the vibronic ground state of the molecules. The encoded spatial structure can be restored later on with a time-delayed probe pulse by exploiting the quantum beating of the wavepacket through the occurrence of "laser-induced field-free molecular alignment". The Stokes and anti-Stokes fields in such a non-resonant and broadband excitation cannot be distinguished [1] resulting in the

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occurrence of unwanted quantum excitation channels as for instance one channel only driven by the auxiliary beam (storing ℓ=0) or another channel only driven by the structured field. The relevant information to be stored can be preserved from this uncontrolled superposition by using circular polarizations of opposite helicity. Such a strategy enables the selection of the desired quantum channel by spin constraint.

A typical experimental result is depicted in Fig. 2 for the storage of ℓ =1. The signal intensity (a) and phase (b) pattern, measured at a pump-probe delay τ=21.18 ps (corresponding to a peak of molecular alignment), confirm that molecules have restored the helical phase structure of the OAM state encoded by the pump.

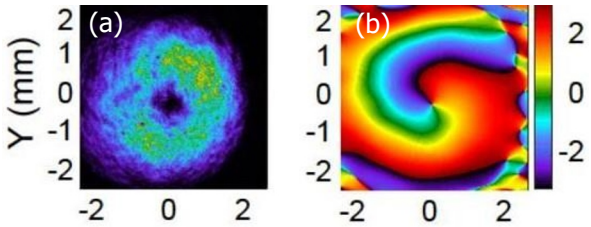

Fig. 2. (a) Signal intensity pattern measured in CO_2 on a revival of alignment for the case ℓ =1 (b) Retrieved spectral phase

The underlying mechanism at the origin of this storage can be interpreted by the spatial structuring of the molecular sample induced by the pump field. For the storage of ℓ =1, the global pump excitation exhibits a polarization vector distribution (see Fig. 3) that follows the azimuthal pattern of neutral axes of a standard q-plate (q=1/2).

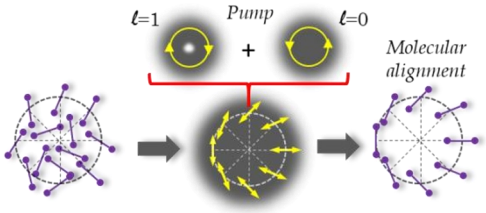

Fig. 3 Illustration of the mechanism for storing ℓ =1 OAM state. The global pump field exhibits an azimuthal distribution of polarization inducing a molecular alignment along the same pattern (equivalent to a q-plate with q = 1⁄2).

A q-plate is a birefringent material whose slow (or fast) axis orientation α varies with the azimuthal angle according to a pattern defined by α =qφ [3]. It is used to generate OAM by SAM to OAM conversion (a q-plate generating OAM numbers ℓ =±2q). For instance, when a left-circularly polarized field interacts with a q-plate q=1/2, it produces a right-circularly polarized field carrying an OAM ℓ=1. The field vector distribution of the pump field in Fig.3 follows the pattern of a q-plate q=1/2. When interacting with molecules, such a field will induce an inhomogeneous spatial distribution of molecular axes following the same azimuthal pattern and resulting in the formation of a "molecular q-plate". The periodical revival of this "molecular q-plate" associated to the quantum beatings of the wavepacket enables to restore the encoded helical phase structure on-demand.

The experimental result corresponding to the storage of pure OAM states ℓ=±1, 2 and of a coherent superposition ℓ=1,-1 will be presented. Prospects and functionalities in terms of optical processing and versatile control of OAM will be discussed.

References

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