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Influence of the photon orbital angular momentum on electric dipole transitions: negative experimental evidence

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We describe an experiment of atomic spectroscopy devoted to ascertain whether the orbital angular momentum (OAM) of photons has the same property of interacting with atoms or molecules as it occurs for the spin angular momentum (SAM). In our experiment, Rubidium vapors are excited by means of a laser radiation with different combinations of OAM and SAM, particularly selected to inhibit or enhance the fluorescence according to the selection rules for the electric dipole transitions between the fundamental state and the first excited doublet. Our results clearly show that an electric-dipole-type transition is insensitive to the OAM value. They provide an original validation of a problem long-debated in theoretical works.

OCIS codes: *(050.4865) Optical vortices; (260.6042) Singular optics;(260.5430) Polarization; (300.1030) Absorption.*

Much effort has been made to achieve the purpose of unambiguously defining the orbital angular momentum (OAM) of the radiation through the investigation of its fundamental properties. On the theoretical front, it has been shown that the OAM part of the total angular momentum of a beam with a cylindrical symmetry can be explicitly separated from its spin (SAM) counterpart in the paraxial optics approximation, which allows for the association of the SAM and the OAM of a light beam with its polarization and helical phase fronts respectively [1]. Further studies have been focused on a similar physically meaningful definition of the OAM in the nonparaxial case, where such a clear division is not straightforward anymore [2–8] (see also [9, 10] and references therein). The quantum nature of OAM, as stated by many authors (see [11, 12] and the overview in [13]), would open the way to many applications in telecommunications, in cryptography and quantum information. Several experiments investigated the angular momentum exchange between light and matter in the attempt to disentangle the relative role of OAM and SAM ([14–17], for a comprehensive overview [10]).

For a beam of circularly polarized light collimated along the z-axis, the SAM per photon admits the values $S_z = \pm\hbar$, where $\hbar$ is the reduced Planck constant, while the OAM per photon can be expressed by any positive or negative integer l as $L_z = l\hbar$. The relative sign of the SAM and OAM terms is expected to cause different behaviors in the motion of particles hit by the radiation [18–20]. Recent spectroscopic experiments explored the interaction of matter with light beams with the aim of detecting significant dichroic effects due to the OAM of the incoming radiation [21, 22], also in the presence of magnetic fields (see [23] and references therein) and for the purpose of studying the applications of laser beams with OAM for quantum information processing with single, trapped ions [24]. In parallel, other theoretical and experimental works aimed at determining to what extent the OAM could be entitled of the same 'intrinsic' character as the SAM, which is independent of any choice of the axis about which it is measured [25–27]. Results led to the definition of a 'quasi-intrinsic' character in the case of the OAM, which is related to the dependence of its density on the rotation axis [28].

In this framework, we describe here an experiment of optical pumping among atomic levels connected by electric-dipole-type transitions, by using a circularly polarized laser beam carrying OAM. In this case, results should lead to an unambiguous interpretation of the OAM and SAM roles. As atomic sample, we selected a natural mixture of isotopes of Rubidium, ^{85}Rb and ^{87}Rb, whose doublet D_1 and D_2 includes the most intense optical transitions, from the ground state $5^2S_{1/2}$ to the $5^2P_{1/2}$ and the $5^2P_{3/2}$ excited states, respectively. The selection rules for the electric dipole type transitions are $\Delta L = \pm 1$ and $\Delta m = \pm 1$. If the OAM of the circularly polarized radiation is 1, the values of the total angular momentum expressed as **SAM+OAM** will therefore become $S_T = 0$ or $S_T = 2$. In fact, we take the spin parallel to the wave vector of the radiation so that their respective absolute values are summed with their relative helicity. $S_T = 2$ violates the selection rule and hence the transition is forbidden

while $S_T = 0$, which can be represented as two opposite $\pm\hbar$ spin states, as the linear polarization, would increase the fluorescence compared with circularly polarized radiation and OAM=0. On the contrary, the absence of effects on the fluorescence emission induced by the interaction between the beam and the Rb atoms would prove that the OAM does not affect the selection rules of the atomic transitions, at least in the electric dipole approximation.

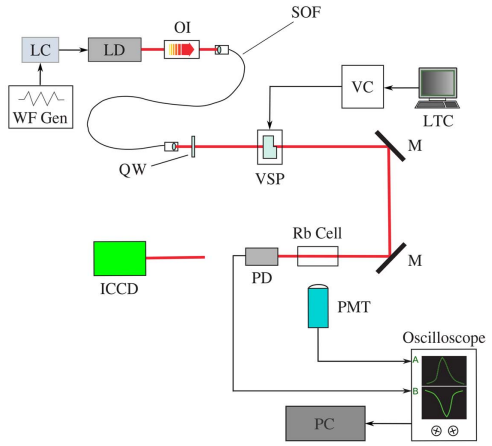

Fig. 1. Experimental apparatus. LC: Laser diode control; LD: Laser diode; ICCD: Intensified CCD; LTC: Laptop computer; M: Mirror; OI: Optical isolator; PD: Photodiode; PMT: Photomultiplier tube; QW: Quarter Wave plate; PC: Desktop computer; SOF: Single mode polarization maintaining optical fiber; VC: Voltage Control; VSP: Variable spiral plate; WF Gen: Wave form generator.

The experimental apparatus is sketched in Fig. 1. A free-running Fabry-Perot laser diode (Thorlabs Sharp LT024MD) emitted electromagnetic radiation resonant with the Rb atoms D_2 transition at 780 nm, which was then fed into a single mode polarization-maintaining optical fiber in order to ensure the filtering of a TEM_{00} mode only. An optical isolator was placed in front of the diode in order to prevent any unwanted feedback into the laser cavity. The laser diode was driven by a low noise current controller. Its current was modulated by a triangular signal coming from a wave form generator; this allowed to sweep the whole D_2 Rb resonance profile. Its temperature control was guaranteed by a bipolar temperature controller ($\Delta T = 0.01$ K within 1h) through a Peltier junction. A quarter-wave plate circularly polarized the output beam (~ 5 mm diameter), which was finally converted into a helical-wavefront beam with a phase singularity at its axis. This conversion was performed by means of a variable spiral plate (VSP) for vortex beam generation (manufactured by ARCoptics). As explained in Refs. [29, 30], such a device, provided with a topological charge of ± 0.5, is able to transform a planar wavefront with circular (left or right) polarization into a beam with an optical vortex, carrying an OAM $= \pm 1$. The VSP was driven by a USB computer controlled electrical power supply. The retardation of the VSP was controlled by an AC bias and could be adjusted to any wanted value between 50-1500 nm. Moreover the orbital momentum could be switched on and off (within 100 ms) simply by changing the bias on the VSP. The output beam was then sent through a cylindrical Pyrex cell (1 cm diameter, 4 cm length) containing a natural mixture of ^{85}Rb (72%) and ^{87}Rb (28%) atoms at a temperature of 42°C to guarantee an adequate vapour pressure ($2 \cdot 10^{-6}$ Torr), still working in an optical thin regime [31, 32]. The absorption and the fluorescence were measured through a photodiode and a photomultiplier placed after the Rb cell and at 90° to the incident light beam, respectively, both connected to an oscilloscope, whose digital signal was elaborated by a desktop computer. Finally, an Intensified CCD (ICCD) Camera (Stanford 4QuickE) was placed on the beam axis for the recording of the far-field intensity distribution.

During the experiment, the laser diode worked with an injection current of 130 ± 2 mA and at a temperature of 32.3° C. The spectroscopic measurements were performed by circularly polarizing the laser beam left- or rightwise, and then by letting it pass through the VSP first and the Rb atoms cell afterwards. The VSP has the property of maintaining the circular polarization, inverting the sense of rotation. Through the USB driver, it was possible to set the VSP so that it could provide an OAM $= 0$ or an OAM $= 1$ to the incoming radiation. The power of the beam was 3 μW before the VSP, 2.92 μW and 2.85 μW after the VPS when set to OAM $= 0$ and OAM $= 1$, respectively. The beam diameter was about 5 mm.
In Fig. 2 we show the absorption profiles in the case of left-wise and rightwise circular polarization without OAM and with OAM $= 1$. By sweeping the laser frequency in a range of 10 GHz across the D_2 Rb resonance profile, we could observe the four minima in the transmission corresponding to the transitions from the ground state of ^{85}Rb (F=3, F=2, inner minima) and ^{87}Rb (F=2, F=1, outer minima). In fact, the Doppler width at our working temperature is about 529 MHz. This allows us to resolve the iperfine structure of the ground state (3.03 GHz and 6.83 GHz for ^{85}Rb and ^{87}Rb respectively). On the contrary, the structures of the excited levels, whose separations are in the MHz range, remain embedded in the Doppler profile [31, 32]. For simplicity, we took the zero of the sweep at the frequency of the transition starting from F=3 of ^{85}Rb. The same has been done in Fig. 3, where the fluorescence signals measured in the same experimental conditions as Fig. 2 are shown. As expected, the fluorescence exhibits a complementary behavior with respect to the absorption. We could quantify a maximum intensity error of 5% and frequency error of 3% during the sweeping time of the laser (50 ms). The profiles of the transition lines do not exhibit significant variations within the limits of this experimental error and, with both polarizations and OAM $= 1$, no disappearance of the electric dipole transition effects is observed.
In order to obtain a further confirmation of the correct behavior of our apparatus, we captured on the ICCD camera the far-field of the beam, and we compared it, both in the presence and in the absence of the Rb atoms, with the well-known morphology of the far-field of light endowed with OAM [33, 34]. Such a comparison, represented in Fig. 4, shows that the structure of the field distribution is not varied in the two cases, apart a slight difference in the intensity due to the presence of the Rb cell, which demonstrates that no exchange of OAM occurred in the interaction between light and matter. We verified that the VSP specifications are those declared in the data sheet by means of interferometric technique (not shown in the figure). Moreover, the comparison of the patterns produced by the beam, before and after crossing the Rb cell, confirmed that the phase distribution was not affected by the absorption.

The experiment described in this Letter aimed at verifying whether the total angular momentum of a light beam with a

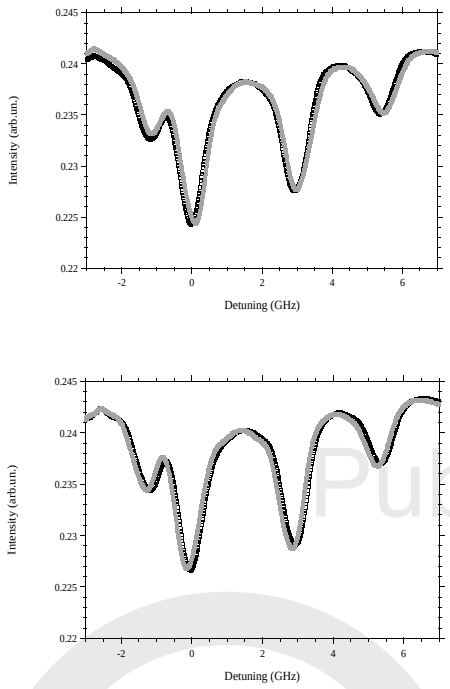

Fig. 2. Transmitted radiation with modes OAM $= 0$ (black open squares) and OAM $= 1$ (gray full squares) for leftwise circular polarization (top) and rightwise circular polarization (bottom) of the laser beam.

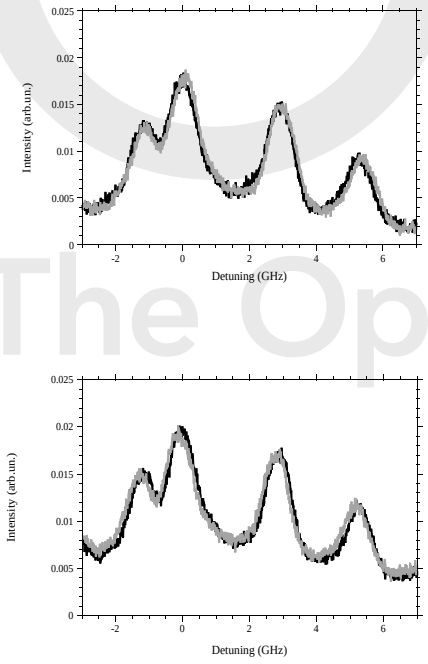

Fig. 3. D_2 Rb fluorescence with modes OAM $= 0$ (black line) and OAM $= 1$ (gray line) for leftwise circular polarization (top) and rightwise circular polarization (bottom) of the laser beam.

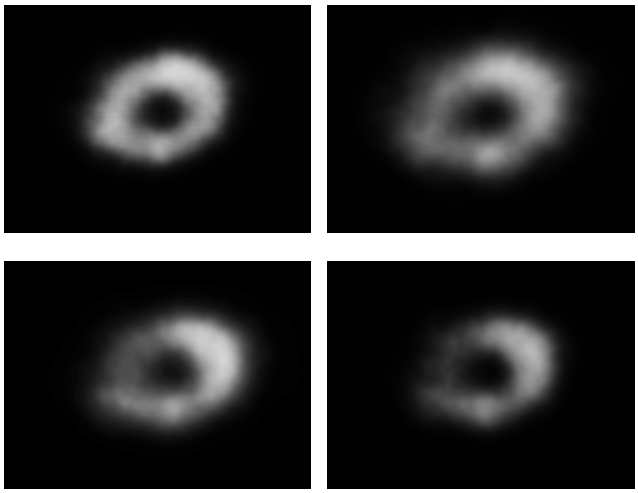

Fig. 4. OAM $= 1$: far-field intensity distribution for the leftwise (top) and rightwise (bottom) polarized radiation in the absence (on the left) and in the presence (on the right) of the Rb cell.

OAM component was such as to induce fluorescence excitation on alkali atoms, or to inhibit it, depending on the values of the OAM and of the SAM of the beam. The idea of assessing whether the OAM can influence the internal electronic degrees of freedom of the atoms is at the core of various theoretical analysis, both in the electric dipole approximation [35] and for higher-order transitions. In Ref. [23], an intuitive argument is given for explaining the absence of magnetic orbital dichroism in an isotropic medium as a function of the sign of the OAM. The authors suggest that this effect cannot be observed in transitions essentially described by the electric dipole approximation, but only when considering (at least) the higher quadrupole order. This explanation is reinforced by the theoretical results obtained in Ref. [36], where it is shown, through a Hamiltonian description of the interaction of light with matter, that in the electric dipole approximation an exchange of OAM only occurs between the light and the center of mass of the atoms (or molecules), while an exchange of OAM also involving the internal electronic motion takes place only when considering the weaker electric quadrupole interaction. Such a conclusion is further confirmed by a study on the effects of twisted light on atoms beyond the paraxial approximation [37], where the probability that the internal state of an atom acquires orbital angular momentum from light is also discussed. Very recently, a first experimental confirmation of this computation was given in Ref. [38], where it was demonstrated that a transfer of OAM from the beam to the internal electronic degrees of freedom could be observed for a quadrupole transition of a single trapped atom.

Our results highlighted the absence of any observable effect on the atomic transitions of the Rb atoms hit by the radiation endowed with OAM. This outcome corroborates the theoretical predictions according to which the orbital angular momentum of a single light beam does not directly influence dipole atomic transitions of an atomic sample [39, 40].

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