Spectroscopic Observation of the Rotational Doppler Effect

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We report on the first spectroscopic observation of the rotational Doppler shift associated with light beams carrying orbital angular momentum. The effect is evidenced as the broadening of a Hanle electromagnetically induced transparency coherence resonance on Rb vapor when the two incident Laguerre-Gaussian laser beams have opposite topological charges. The observations closely agree with theoretical predictions.

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Laguerre-Gaussian (LG) modes of the electromagnetic field, with nonvanishing azimuthal phase variation, are familiar examples for light beams with twisted wave fronts. Such beams carry orbital angular momentum (OAM) along their propagation direction. The properties of LG modes have attracted considerable attention in recent years and a wide range of applications were suggested [1,2]. LG beams were used for atom trapping and cooling [3-5] and special attention was put on the application of LG beams for the exchange of orbital angular momentum between light and Bose-Einstein condensates [6-9]. Some of us have demonstrated that OAM can be recorded in the position dependent population and coherence of a cold atom sample [10], and transferred between internal atomic states. In addition, the generation of new fields with OAM via nonlinear wave mixing in coherently prepared cold atoms was observed [11]. Quite recently, the use of photons in LG modes was suggested for quantum information processing. The state of such a photon lies in a multidimensional Hilbert space, describing the total (intrinsic plus orbital) angular momentum, in which quantum computation with improved efficiency should be possible [12]. Entanglement between pairs of photons in modes with OAM was recently reported [13].

The interaction of a moving atom with a LG field raises the fundamental question of the Doppler effect [14]. As an atom moves across the helicoidal wave fronts of the LG mode, it experiences, in addition to the usual Doppler shift related to the velocity in the light propagation direction (and a small shift associated to radial motion in a curved wave front), a most intriguing frequency shift, the so-called rotational Doppler effect (RDE) associated with the azimuthal velocity. To date, the RDE has only been observed interferometrically. The RDE results in frequency shift when the light beam is rotated around its propagation axis [15]. Such shift was observed in Refs. [16,17] using millimeter waves. In the optical domain, a frequency shift in the field generated by a rotating plate was observed in Ref. [18]. A different approach, used in Ref. [19], relates the RDE to the asymmetric interferometric spatial pattern occurring in the superposition of a Gaussian and a LG modes. In this work we present the first experimental demonstration of the RDE arising directly from the interaction of LG light beams with an atomic sample.

Laguerre-Gaussian modes are usually identified with two integer numbers: l and p [20]. The topological charge l corresponds to the phase variation (in units of 2π) of the field along a loop encircling the optical axis. The integer p + 1 corresponds to the number of maxima of the field intensity along a radius. In this study we are concerned with modes with p = 0. The Doppler shift experienced by a LG field in a moving frame is given by [14]

$$\delta_{\rm LG} = -\left[k + \frac{kr^2}{2(z^2 + z_R^2)} \left(\frac{2z^2}{z^2 + z_R^2} - 1\right) - \frac{(2p + |l| + 1)z_R}{z^2 + z_R^2}\right] V_z - \left(\frac{krz}{z^2 + z_R^2}\right) V_R - \left(\frac{l}{r}\right) V_\phi$$
(1)

where (r, z, ϕ) and (V_R, V_z, V_{ϕ}) represent in cylindrical coordinates a position in space and corresponding velocity of the moving frame. $z_R \equiv \pi w_0^2 / \lambda$ is the Rayleigh range and w_0 the beam waist.

The first term on the right hand side of Eq. (1) is the usual Doppler shift associated with motion along the propagation axis direction, dominated by the leading term $-kV_z$ analogous to the Doppler shift of a plane wave with wave vector **k** directed along *z*. The second term represents the contribution to the Doppler of the radial velocity due to wave front curvature. This term is smaller with respect to the first by a factor of the order of the beam divergence angle θ (typically $\theta \sim 10^{-4}$ for well collimated beams). The last term in Eq. (1), represents the RDE which is proportional to the topological charge *l* of the mode. This term is smaller than the axial Doppler shift by a factor $l\lambda/2\pi r$ which is of the order of 10^{-4} under standard experimental conditions ($r \sim 1$ mm, $\lambda \sim 1$ µm).

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The observation of RDE through atom-field interaction was already suggested in Ref. [14]. However, the smallness of the RDE compared to the axial shift makes the observation of the former quite difficult under conditions where both contributions are present. Our demonstration of RDE through atomic spectroscopy relies on the observation of Doppler line broadening, due to RDE, under conditions where other contributions (axial and radial) to the Doppler broadening exactly cancel. The basic idea is presented in Fig. 1. We consider a transition between two atomic levels, a ground level and an exited level with total angular momentum F and F' = F - 1 respectively. Incident upon this system are two copropagating fields (1 and 2) with identical frequencies and opposite circular polarizations. The two fields are LG modes with the same intensity, equal beam waist w_0 , and topological charges l_1 and l_2 respectively (p = 0). For a moving atom, the resonance condition for a two-photon (Raman) transition between ground state Zeeman sublevels with $\Delta m = \pm 2$, is given by $\delta' - \delta = 0$ where $\delta' = \delta_{LG}^1 - \delta_{LG}^2$ and δ represents the total Zeeman shift ($\delta = 2g\mu_B B$, g is the ground state gyromagnetic factor, and μ_B the Bohr magneton) induced by a static magnetic field B oriented along the propagation axis. Since the axial and radial contributions to the Doppler effect are the same for both fields they exactly cancel and we are left with $\delta' = (\frac{l_1 - l_2}{r})V_{\phi}$. In consequence, pure rotational Doppler line broadening should occur when $l_1 \neq l_2$.

The two-photon resonance results in a reduction of the atomic absorption around B = 0 known as Hanle electromagnetically induced transparency (Hanle/EIT) resonance [21]. At low light levels, the homogeneous line shape *h* of the Hanle/EIT resonance is given by [22]

$$h(\delta' - \delta, r) = AI_1(r)I_2(r)L(\delta' - \delta), \qquad (2a)$$

$$L(x) = \gamma/2\pi [x^2 + (\gamma/2)^2]^{-1}, \qquad (2b)$$

where *A* is a constant, *r* is the atom radial position and $I_j(r)$ [j = 1, 2] the intensity distribution of field *j*. γ is the relaxation rate of the ground state atomic coherence. For an atomic vapor, the total Hanle/EIT signal is given by



FIG. 1. Scheme used for the observation of rotational Doppler effect on Hanle/EIT resonances illustrated on a $F = 1 \rightarrow F' = 0$ transition. The two spatially overlapped fields have identical frequency, opposite circular polarizations, and topological charges l_1 and l_2 .

$$S(\delta) = \int_0^\infty 2\pi r dr \int_{-\infty}^{+\infty} h(\delta' - \delta, r) W(V_\phi) dV_\phi \qquad (3)$$

where $W(V_{\phi})$ is the velocity distribution at thermal equilibrium at temperature T: $W(V_{\phi}) = N(\pi/\alpha)^{-1/2} \times \exp(-\alpha V_{\phi}^2)$ with $\alpha = m/(2k_BT)$ (*m* is the atomic mass, k_B the Boltzmann constant). From Eq. (3) we see that the line shape corresponds to the homogeneous Lorentzian line when $l_1 = l_2$ and presents a distinctive Doppler broadening for $l_1 \neq l_2$. Expressing V_{ϕ} in terms of δ' one gets

$$S(\delta) = A \frac{N}{\sqrt{\pi/\alpha}} \int_{-\infty}^{+\infty} d\delta' \int_{0}^{+\infty} I_1(r) I_2(r) L(\delta' - \delta)$$
$$\times \exp\left[-\frac{\alpha \delta'^2}{(l_1 - l_2)^2}\right] \frac{2\pi r^2}{l_1 - l_2} dr. \tag{4}$$

Finally, using the expressions for the far-field intensity of a LG mode, $I_j(r) = I_{0j}r^{2|l_j|}e^{-2r^2/w^2(z)}$ (w(z) is the propagation-distance dependent beam radius [14]) one obtains

$$S(\delta) = C \int_{-\infty}^{+\infty} L(\delta' - \delta) \left[\frac{\alpha \delta'^2}{(l_1 - l_2)^2} + \frac{4}{w^2(z)} \right]^{-q} d\delta'$$
(5)

where $q = |l_1| + |l_2| + 3/2$ and *C* is a coefficient depending on *q*.

A simplified expression for $S(\delta)$ can be obtained in the limit of a very narrow homogeneous resonance $(\gamma \rightarrow 0)$:

$$S(\delta) = C \left[\frac{\alpha \delta^2}{(l_1 - l_2)^2} + \frac{4}{w^2(z)} \right]^{-q},$$
 (6)

and the full width at half maximum of the resonance is given by

$$\Delta = \frac{4|l_1 - l_2|}{\sqrt{\alpha}w(z)}\sqrt{2^{(1/q)} - 1}.$$
(7)

In the general case of a finite homogeneous line width, the convolution integral (5) is evaluated numerically. As indicated by Eqs. (5) and (7), the width of the Doppler broadened resonance grows as $|l_1 - l_2|$ is increased.

We have observed the rotational Doppler broadening of the Hanle/EIT signal on a D1 line transition of ⁸⁷Rb in a room-temperature vapor cell. The experimental setup is presented in Fig. 2. An extended cavity laser diode was used as light source. The laser frequency was kept constant near the center of the well resolved $5S_{1/2}(F = 2) \rightarrow$ $5P_{1/2}(F = 1)$ atomic transition. A spatial filter was used to improve the beam profile. Using a polarizing beam



FIG. 2. Experimental setup. ECDL is the extended cavity diode laser; PBS, polarizing beam splitter; PD, photodiode. M1 and M2 are the Laguerre-Gaussian mode-generating masks.

splitter, the light beam was divided into two equal-intensity arms with orthogonal linear polarizations. Each beam was sent to a LG mode generator [18] consisting on a mask imprinted on a photographic film, a pinhole placed at the first order focus of the mask and a collimating lens with focus on the pinhole. The masks were computer-generated binary spiral zone plates [23] recorded on high contrast photographic film. The two LG mode generators were identical except for the relative orientation of the mask allowing the generation of fields with equal or opposite topological charge l. After propagating equal distances, the two fields were recombined in a second polarizing beam splitter. A quarter-wave plate, placed after the beam splitter, transforms the orthogonal linear polarizations of the two fields into opposite circular polarizations. The overlapping fields are sent through the atomic gas cell placed inside a two-layer μ -metal shield. Inside the shield, a solenoid controls the longitudinal magnetic field. The total light intensity transmitted through the sample is monitored with a photodiode as a function of the applied magnetic field. The light power at the atomic sample was controlled with neutral density filters. During measurements the light power at the sample was approximately 1 μ W (beam radius: 0.5–0.9 mm; see below).

To improve sensitivity, a small ac modulation was added to the dc value of the magnetic field and lock-in amplification was used for the photodiode current. The signal recorded as a function of *B* corresponds to the derivative of the Hanle/EIT signal (provided that the ac modulation is small and slow enough). We measure the linewidth as the peak-to-peak distance on the derivative signal.

To demonstrate the occurrence of Doppler broadening due to RDE we have systematically compared the resonance width obtained under identical experimental conditions when the two fields acting on the atomic sample posses equal or opposite topological charges. In the case $l_1 = l_2$ no RDE is expected and the observed resonance corresponds to the homogeneous line shape. When $l_1 = -l_2$, RDE line broadening should be present. The two situations can be obtained in our experimental setup by using identical masks on the two arms of the light pass. If the two mask are equally oriented we get $l_1 = l_2$. Reversing one of the masks results in $l_1 = -l_2$.

An important requirement for our demonstration is to ensure that no spurious line broadening is introduced due to misalignment. Indeed, a small angle ε between the axes of the two LG modes results in Doppler broadening of the order of $\varepsilon \Delta_{\text{Dopp}}$ ($\Delta_{\text{Dopp}} \simeq 500$ MHz). By slightly misaligning the overlapping of the two fields, we have measured a linewidth increase of 110 KHz per milliradian of angular misalignment. Our beam overlapping procedure was accurate to better than 0.2 mrad. In consequence the angular misalignment contribution to the observed width was less than 22 KHz. A confirmation of this is reached by comparison of the linewidth obtained with $l_1 = l_2$ and with a single LG field linearly polarized. In the latter case, the two fields participating in the Hanle/EIT resonance are the circular components of the single field and no broadening due to misalignment is possible. The two situations gave the same linewidth: \sim 52 KHz at low light power. Such lower limit of the homogeneous resonance width is believed to be determined by the effective time of flight of the atoms across the beam and by residual temporal and spatial fluctuations of the magnetic field. Finally, we have checked that no spurious broadening was introduced by the reversing of the orientation of one mask to change the topological charge sign. For this, a pair of masks with $l_1 = l_2 = 0$ was used. No change in the linewidth was observed after reversing the orientation of either of the two masks.

In Fig. 3 we present the recorded signal for different pairs of LG-mode-producing masks with $|l_1| = |l_2| = l = 1$, 2, 3, 4. All four cases produce narrow resonances of similar width for $l_1 = l_2$. A visible broadening due to RDE is observed for $l_1 = -l_2$. A variation of the signal level with l is also observed. Such variation is expected as a



FIG. 3. Observed Hanle/EIT resonances as a function of the total Zeeman shift $\delta = 2g\mu_B B$ for different pairs of LG mode-generating masks. Dashed (solid) lines denote the same (opposite) topological charge on each field.



FIG. 4. Line width dependence on topological charge. Circles experimental results are reflected by solid (hollow) symbols to denote equal (opposite) charge on each field. Triangles show theoretical prediction.

result of the variation of the radial light intensity distribution for LG modes with different *l*. The dependence of the peak-to-peak linewidth with *l* is shown in Fig. 4. Almost constant width is observed for $l_1 = l_2$ with a slight increase for small *l* due to intensity broadening. A monotonic growth of the linewidth with *l* is observed when $l_1 = -l_2$. Also shown in Fig. 4 are the calculated widths obtained using Eq. (5) with a homogeneous width of 52 KHz. In the calculations, we used w(z) = 0.5, 0.65, 0.74, 0.83, 0.89 mm for l = 0, 1, 2, 3, 4, respectively. These values were taken from the fit of the experimental beam cross section (recorded with a CCD camera) to the theoretical intensity profile of a LG mode. They increase with *l* as expected [24].

The results presented in Figs. 3 and 4 are a clear indication of rotational Doppler broadening on the Hanle/ EIT resonances produced by two LG fields with different topological charges. A good agreement is observed between the theoretically predicted and the experimentally observed variation of the resonance width with l. We notice that the experimentally observed width are slightly smaller than the predicted ones. We attribute this fact to small contamination of the actual light field with a Gaussian mode [25] (l = 0).

In summary, we have experimentally observed the rotational Doppler shift associated with light beams carrying OAM via the coherent interaction between these beams and an ensemble of rubidium atoms at room temperature. The effect is clearly evidenced as an inhomogeneous broadening of the Hanle resonance depending on atomic azimuthal velocity. These observations constitute the first demonstration of light-atom interaction depending on azimuthal atomic velocity. They provide experimental support to the suggested use of two-photon coherent resonances for the manipulation of the rotational motion of atomic samples [6,7,9]. This work was supported by Fondo Clemente Estable, CSIC and PEDECIBA (Uruguay). J. W. R. T. acknowledges financial support from CNPq (Brazil).

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