Optical Pumping of Orbital Angular Momentum of Light in Cold Cesium Atoms

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We present experimental results on the transfer of the orbital angular momentum of light to a system of cold cesium atoms. A nondegenerate four-wave mixing process was used as an indirect tool to observe this transfer. Our experiments show, in particular, that the orbital angular momentum of light can be transferred, via optical pumping in the cold atomic sample, from one beam to another oscillating at a different frequency.

PACS numbers: 32.80.Pj, 42.50.Vk, 42.65.Hw

The Laguerre-Gaussian (LG) laser modes possess welldefined angular momentum that can be decomposed into an orbital component and a spin component associated with its polarization [1]. Optical beams with orbital angular momentum (OAM) include screw topological wave front dislocations or vortices. Vortices appear as spiral phase ramps around a singularity where the phase of the wave is undefined; thus its amplitude must vanish. The order of the singularity multiplied by its sign is referred to as the topological charge of the dislocation.

There has been a great deal of interest in the mechanical and optical effects that light beams with OAM can exert on material systems. This angular momentum has been transferred to macroscopic particles trapped in an optical tweezer causing them to rotate [2,3]. An atom moving in a beam with OAM experiences a torque and an azimuthal shift in its resonant frequency in addition to the usual axial Doppler shift and recoil shift [4]. Also the LG modes are important in laser cooling and trapping experiments. For example, in Ref. [5] such beams were used for highresolution spectroscopy in a magneto-optical trap, and a novel trapping scheme using LG modes was proposed and experimentally demonstrated in Ref. [6].

The linear propagation of beams with phase front dislocations and also their single-frequency propagation in cubic and photorefractive nonlinear bulk media in single pass and cavity configurations have been studied in $[7-10]$. The up-conversion by second harmonic generation of light in quadratic nonlinear media has been investigated experimentally in Refs. [11–14]. However, only very limited experimental work has been done with these beams in atomic systems, and has mainly dealt with nonlinear propagation effects [15].

A system of cold atoms as a medium for studying optical processes with beams carrying OAM possesses a number of interesting properties. First, nonlinear effects arising by individual optical transitions in the system of atomic energy levels constitute by itself basic steps that exist in any nonlinear processes in an arbitrary medium.

The OAM of the input beams can be transferred to the atomic system and, hence, modify these nonlinear optical processes. Furthermore, the low temperature of laser cooled atoms, besides allowing to diminish the Doppler broadening of the atomic levels, can also give access to the direct observation of mechanical effects induced by the atom-field interaction [16].

In this Letter, we present experimental results on the nondegenerate four-wave mixing process (NDFWM) performed in cold cesium atoms with optical beams carrying OAM. We use this process as an indirect tool in order to demonstrate that the OAM can be transferred from the optical beam to the system of cold cesium atoms. This effect is similar to the phenomenon of optical pumping induced orientation and alignment in atomic systems due to the spin angular momentum of the photon [17], and to our knowledge corresponds to the first observation of the optical pumping of OAM of light.

We have employed a sample of cold atoms obtained from a vapor cell magneto-optical trap. The trapping beams are provided by a stabilized Ti-sapphire laser, and are red detuned by about 12 MHz from the resonance frequency of the cesium cycling transition $6S_{1/2}$, $F = 4 - 6P_{3/2}, F' = 5$ at $\lambda = 852$ nm, as indicated in Fig. 1(a). The necessary repumping laser is a long external cavity diode laser, which is tuned into resonance with the $6S_{1/2}$, $F = 3 - 6P_{3/2}$, $F' = 3$ transition. Typically the number of trapped atoms, estimated by measuring the absorption of a weak probe beam by the atomic cloud is of the order of $10⁷$ atoms. Recently, four-wave mixing experiments in this atomic system were done with beams without OAM [18].

The experimental setup is shown in Fig. 1(b). The forward pump beams *F* and *P* with linear polarizations have the same frequency ω_1 , and are incident in the trap forming an angle of $2\theta \approx 2^{\circ}$. These beams are provided by a grating-stabilized diode laser (LASER1), and are locked in resonance with the noncycling transition $6S_{1/2}$, $F = 4 - 6P_{3/2}$, $F' = 4$, using an auxiliary saturated

(b)

FIG. 1. (a) Hyperfine energy levels of cesium D_2 line. (b) Experimental setup.

absorption signal. Single-charge and double-charge topological screw dislocations were nested in the beam *P* using two different computer-generated spiral zone plates MASK [19]. A lens *L* allowed us to separate the beam with a nested vortex from the undiffracted light and high-order Fresnel images.

The beam *P* is focused so that its waist in the trap is much smaller than the trap size (about 2 mm), while the beam *F* is collimated to a diameter of the order of 3 mm. To reveal the topological phase structure of the beam *P*, we used a part of the beam with the frequency ω_1 as a reference beam. The reference beam is reflected from the mirrors *M*1, *M*2, and *M*3, and after a beam splitter BS3 provided an interference pattern with the incident beam *P*, back reflected from the mirror *M*4. This mirror is removed during the four-wave mixing experiments. The intensity pattern was analyzed by a charge coupled device camera.

Another independent, grating-stabilized diode laser (LASER2) generated the backward pump beam *B*. The frequency ω_2 of the beam *B* is set, using other auxiliary reference absorption signal, around the frequency of the cycling transition $6S_{1/2}$, $F = 3-6P_{3/2}$, $F^{\dagger} = 2$. This beam, also linearly polarized, has a diameter of the order of 3 mm and is incident in the trap satisfying the phase-matching condition, i.e., $\sin\beta = (\omega_1/\omega_2)\sin\theta$. The powers of beams *P*, *F*, and *B* are $P_P = 5 \mu W$, $P_F = 10 \mu W$, and $P_B = 200 \mu W$, respectively. The

generated beam, PC, is therefore nearly counterpropagating to the beam *P*. Its phase structure was revealed using an interference pattern after a beam splitter BS4 obtained with the reference beam of frequency ω_2 . To measure the phase structure of the incident beam *P* we stop the beam from the LASER2, while to measure the phase structure of the beam PC we stop the reflection from the mirrors *M*2 and *M*4. The efficiency of the NDFWM process, measured by the ratio P_{PC}/P_P , was of the order of 0.1%.

Figures 2(a) and 2(b) show, respectively, the intensity profile and interferogram of the pump beam *P*, when the MASK generates into the beam *P* a single charge phase dislocation. Figures 2(c) and 2(d) show the corresponding distributions for the generated beam PC. As seen, the beam PC includes also a phase front dislocation and hence, an OAM. Owing to the reversed sense of rotation of the interference spiral due to the reflection in mirror *M*4, the topological charge of the beams *P* and PC are the same. Therefore, as these beams are nearly counterpropagating, they carry opposite OAM.

If the topological charge of the pump beam *P* is equal to 2, the corresponding intensity profile and interferogram are shown in Figs. $3(a)-3(d)$. We again observe the generation of the PC beam with the same topological charge as the one of the beam *P*.

We have modeled our system by considering the incident FWM beams to be of the form $\vec{E}_{\mu} = \frac{1}{2} \vec{A}$ are \vec{E} (*n*, *t*, \vec{L} = 3)] \vec{E} as weight \vec{E} = \vec{E} \vec{B} \vec{B} . $\frac{1}{2} \vec{A}_{\mu} \exp[i(\omega_{\mu}t - \vec{k}_{\mu} \cdot \vec{r})] + \text{c.c.,}$ with $\mu = F, P, B;$ $\omega_F = \omega_P = \omega_1$; $\omega_B = \omega_2$, and \vec{A}_μ being the corresponding complex field amplitudes. The fields \vec{E}_F and \vec{E}_B are taken as plane waves, so that \vec{A}_F and \vec{A}_B do

FIG. 2. Observed light distributions and interferograms of the pump beam *P* [(a) and (b)] and the diffracted beam PC [(c) and (d)]. The topological charge of phase dislocation in the pump beam *P* is equal to 1 and is the same as that of the beam PC since mirror *M*4 reverses the sense of rotation of the spiral as discussed in the text.

FIG. 3. Same as in Fig. 2 but with the double topological charge of phase dislocation in the pump beam *P*.

not depend on the coordinates, while for \vec{E}_P , the field amplitude of the LG mode at the beam waist, located at $z = 0$ and coincident with the trap position, is given by $\vec{A}_P = \vec{A}_0 \left(\frac{\sqrt{2}r}{w} \right)^l \exp(-il\phi) \exp[-\left(\frac{r^2}{w^2}\right)] L_p^l (2r^2/w^2).$ Here *w* is the half-beam width, \vec{A}_0 is the beam amplitude, L_p^l is the associated Laguerre polynomial, and *r* and ϕ are, respectively, the radial and angular coordinates in cylindrical polar coordinate system with its *z* axis being along the beam propagation direction. The beam *P* employed in the experiments corresponds to $l = 1$; $p = 0$ (charge 1), and $l = 2$; $p = 0$ (charge 2).

The effect of the trapping and the repumping beams is taken into account through incoherent optical pumping rates specified by γ_T and γ_R , respectively. We have supposed the same relaxation rate γ for all the excited states. We also have assumed that the residual Doppler shift is much smaller than all the relaxation rates, i.e., $\vec{k}_{\mu} \cdot \vec{v} \ll \gamma, \gamma_R$, where \vec{v} is the atomic velocity. A straightforward calculation using the density matrix in the slowly varying amplitude limit and in the rotatingwave approximation yields for the on-resonance thirdorder induced coherence at the transition $F = 3 - F' =$ 2 the following result [18]:

$$
\rho_{32'}^{(3)} = \frac{iN\mu_{32'}|\mu_{44'}|^2}{\hbar^3\gamma^2} \left(\frac{\gamma_T + \gamma}{2\gamma_T + \gamma}\right) \left(\frac{1}{\gamma} + \frac{1}{\gamma_R}\right) A_B A_F A_P^* \times \exp\{i[\omega_2 t - (\vec{k}_F + \vec{k}_B - \vec{k}_P) \cdot \vec{r}]\}.
$$

In this expression $\mu_{44'}$ and $\mu_{32'}$ are the dipole matrix elements of the transitions involved, and *N* is the total number of atoms in the trap. This induced polarization generates a field oscillating at the frequency ω_2 and propagating along the direction $\vec{k}_{PC} = \vec{k}_F + \vec{k}_B - \vec{k}_P$, under the phase-matching condition. The complex field

amplitude of the generated beam is proportional to the complex conjugated amplitude of the beam *P*. This can lead to the observation of wave-front rectification and especially in this case to the transfer of the OAM from one beam to another beam with different frequency. Furthermore, the complex amplitude of the generated phase conjugated beam PC has the topological phase defect of the same sign as the one in the beam *P*, since the induced polarization is proportional to A_P^* , but the propagation directions of the beams *P* and PC are opposite.

The NDFWM signal can be interpreted in terms of the Bragg diffraction into a transferred population grating. The grating is produced by excitation of the noncycling $6S_{1/2}$, $F = 4 - 6P_{3/2}$, $F' = 4$ transition by the beams *F* and *P*. The excited-state population grating created by these beams is transferred through spontaneous emission into the $6S_{1/2}$, $F = 3$ ground-state hyperfine level. The transferred grating is then monitored by the third laser beam *B*.

In the writing process of the excited-state population grating by absorption of each photon of the beam *P*, the OAM of this beam is transferred to the atomic system. The angular momentum is stored by the atomic system as the rotational movement of the atoms and is conserved during the spontaneous emission into the $6S_{1/2}$, $F = 3$ ground-state level. In the reading process by absorption of each photon from the beam *B* the atomic system generates a photon with OAM in the beam PC. The net OAM transferred to the atomic system depends on the number of photons absorbed in the beam *P* and the number of photons emitted in the beam PC. Since the atoms in the lower ground state $6S_{1/2}$, $F = 3$ do not experience any confining force, there is no external torque acting on the atoms to compensate the corresponding change of OAM due to the interaction with the light. Therefore, the atomic sample as a whole should rotate. By taking into account the measured Bragg diffraction efficiency and the absorption of beam *P*, and the fact that each absorbed and generated photon carries $\pm l\hbar$ of OAM, we can roughly estimate the flux of OAM transferred to the atomic system. For the beam *P* with $l = 1$ this value is of the order of 1×10^{-21} Nm/s. Further investigations to detect directly this atomic motion are currently under way. As we have mentioned before, the transfer of OAM to the cold atomic sample presents a close analogy with the well-known phenomenon of optical pumping induced orientation in atomic systems, a phenomenon which has been used to the production of cold sample of spinpolarized atoms [20].

The present effect could in principle be observed in other nonlinear media, as, for example, in a hot atomic vapor. However, cold atoms with low velocities seem to be essential if one wants to observe the mechanical effects induced on the atomic system due to the OAM exchange with light. Moreover, as light beams carrying

OAM have been suggested to excite vortex states in a Bose-Einstein condensate (BEC) [21], our results, in fact, present a first step towards this goal. Also, as has been demonstrated recently [22], a BEC can be used to perform coherent four-wave mixing of matter waves prepared in well-defined momentum states. The extension of this phenomenon to include the OAM or vortex in the atomic wave packets certainly is of great interest and should provide a better understanding for the coherent matterwave interaction.

In conclusion, we have observed experimentally the interchange of the OAM between two waves of different frequencies as a result of the optical pumping of the OAM into the system of cold cesium atoms. We hope that this demonstration will trigger further investigation on the coupling between the internal and external degrees of freedom of an atomic system interacting with light, where, in addition to the well-known effect of linear momentum transfer, one also will need to consider its angular counterpart for light carrying OAM.

We are grateful to R. Y. Chiao and J. Rios Leite for stimulating discussions. The work of J. W. R. T. was supported by FINEP, CNPq-Pronex, Brazilian Agencies.

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