Second-harmonic generation and the orbital angular momentum of light

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Second-harmonic generation has been obtained by the use of Laguerre-Gaussian beams in a variety of mode orders. Each mode becomes doubled in frequency and transformed to a higher order. We show this to be a consequence of the phase-matching conditions. The experiment is consistent with the interpretation that the orbital angular momentum of the Laguerre-Gaussian mode is directly proportional to the azimuthal mode index l. [S1050-2947(96)50711-0]

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The first experiment in nonlinear optics was the generation in 1961 of the second harmonic of a ruby laser beam that had been focused on a quartz crystal [1]. We report the observation of second-harmonic generation using non-zero-order Laguerre-Gaussian light beams and show that the phase-matching conditions dictate that the modes become converted to a higher order in the process. Such mode conversion is shown to be consistent with the fact that the modes possess orbital angular momentum proportional to the mode index l and that orbital angular momentum is conserved within the light fields. In 1993, Basistiy *et al.* [2] made brief mention of the frequency doubling of a laser beam possessing a phase dislocation centered on the axis of the beam; apparently two dislocations resulted. But they offered no explanation as to the physical basis of the observation; the orbital angular-momentum content of the light was not mentioned and the experiment appears only to have been performed for one, evidently non-Laguerre-Gaussian, beam.

A simple description of second-harmonic generation considers the role of plane waves and subsequently defers to the fact that the modes actually used are the lowest order of the commonly occurring family of Hermite-Gaussian modes emitted by real lasers. This family of modes is characterized by Hermite polynomials with indices n and m, the Rayleigh range, and a simple Gaussian amplitude profile. Recent work has shown that the related set of Laguerre-Gaussian modes may be produced from Hermite-Gaussian mode by means of a simple modes converter [3,4] consisting of a pair of cylindrical lenses. Such modes are characterized by the indices p and l where $p=m$ and $l=n-m$ for $n>m$. Again the Rayleigh range is one of their characterizing features. The lowest-order (0,0) mode is common to both sets of modes.

Laguerre-Gaussian modes have been shown theoretically to possess orbital angular momentum of $l\hbar$ per photon [5] in addition to any spin angular momentum due to their state of polarization. The existence of orbital angular momentum has been confirmed by He *et al.* [6] by the transfer of angular momentum from the Laguerre-Gaussian light beam to micrometer-sized metallic particles held in an optical trap. The experiment clearly distinguished between orbital and spin angular momentum, but did not confirm the predicted $l\hbar$ per photon. The interaction of atoms with light possessing orbital angular momentum has already been the subject of detailed investigation [7], particularly with regard to trapping and cooling; new effects have been predicted.

We show that a Laguerre-Gaussian mode may undergo second-harmonic generation and that the resulting frequency-doubled light is of a higher-order mode. A simplistic interpretation of what we see is possible by the use of a naive theory of second-harmonic generation, which assumes that there is no absorption and that the depletion of the input wave is negligible. In this case the amplitude of the second harmonic at frequency 2ω is proportional to the square of the input amplitude at frequency ω; that is, $E^{(2\omega)} \propto (E^{(\omega)})^2$. It is easy to see that when the lowest-order Gaussian mode is used, such that $E^{\omega}=E_1\exp(-r^2/w^2)$, the second harmonic will have an amplitude $E^{2\omega}=E_1^2\exp[-2r^2/(2w)^2] = E_2\exp(-2r^2/2w^2)$. This is again a simple Gaussian beam with the spot size reduced by a factor of $\sqrt{2}$ compared with the fundamental.

The same approximate approach may be applied to our experiment. The general expression for the Laguerre-Gaussian mode amplitude is given by

$$E^{(\omega)}=E_0\exp\left[\frac{-ikr^2z}{2({z_R}^2+z^2)}\right]\exp\left[\frac{-r^2}{w^2}\right] \times\exp\left[-i(2p+l+1)\arctan\left(\frac{z}{z_R}\right)\right] \times\exp[-il\phi](-1)^p\left(\frac{r\sqrt{2}}{w}\right)^l L_p^l\left(\frac{2r^2}{w^2}\right), \tag{1}$$

where k is the wave number, z_R the Rayleigh range, and w the beam width at position z.

At the beam waist where $z=0$, and for a $p=0$ mode, this becomes simply

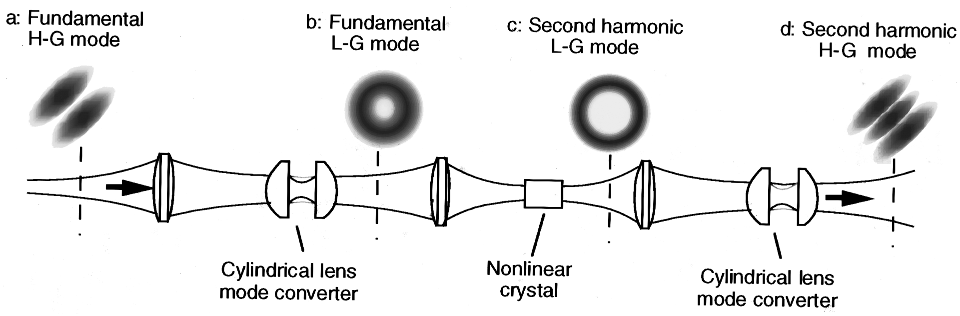

FIG. 1. The experimental configuration showing the mode transformation that occurs when a Laguerre-Gaussian mode is frequency doubled.

$$E^{(\omega)}=E_0\exp\left[\frac{-r^2}{w^2}\right]\exp[-il\phi]\left(\frac{r\sqrt{2}}{w}\right)^l. \qquad (2)$$

If this amplitude is squared we find that the result is again a Laguerre-Gaussian mode with $p=0$, but this time the l index has become $2l$ and w is again reduced by $\sqrt{2}$.

It follows that not only has the frequency been doubled, so has the orbital angular momentum per photon. Two photons might be said to have combined their energy, $\hbar\omega$, to yield one with twice the energy, $2\hbar\omega$, and at the same time two units of $l\hbar$ orbital angular momentum have combined to give $2l\hbar$. This, of course, is not possible for circularly polarized light; a single second-harmonic beam cannot carry spin angular momentum of $2\hbar$.

The experimental arrangement is shown in Fig. 1. An intracavity cross wire was used to generate a variety of Hermite-Gaussian modes from a diode-pumped, Nd:YAG (neodymium-doped yttrium aluminum garnet) laser operating at 1064 nm with a linearly polarized output power of 100 mW. Each mode could be converted into a Laguerre-Gaussian mode by means of a mode converter [4]. The corresponding Laguerre-Gaussian modes were then frequency doubled using a 20-mm-long crystal of lithium triborate [8], temperature tuned to give noncritical, type-I phase matching for the second harmonic at 532 nm. The input Laguerre-Gaussian modes were optimally focused into the nonlinear crystal to maximize the power in the second-harmonic mode, typically a few microwatts. The experiment was also performed with a crystal of KTP which was angle-tuned to give critical, type-II phase matching.

A second mode converter was used to convert both the transmitted input and second-harmonic Laguerre-Gaussian modes back into their corresponding Hermite-Gaussian modes. The modes were recorded using a charge-coupled-device array; the input or second-harmonic light was selected by the use of appropriate filters. The indices of both the fundamental and second-harmonic Laguerre-Gaussian modes were then inferred from the observed n,m indices of the respective Hermite-Gaussian modes.

For a Hermite-Gaussian input mode with indices n,m a Laguerre-Gaussian mode leaving the mode converter has $p=m$ and $l=(n-m)$. This mode produces the second harmonic at frequency 2ω, which, if our argument is correct for $p=0$, will have indices $p=m=0$ and $l=2(n-m)=2n$. The mode converter works equally well either way: just as a Hermite-Gaussian mode can be converted into a Laguerre-Gaussian mode, so a Laguerre-Gaussian mode may be converted into a Hermite-Gaussian using the same or an identical mode converter [4]. Thus if the second-harmonic light is passed into a mode converter, a Hermite-Gaussian mode should result at frequency 2ω with modes indices $2n$ and 0.

Figure 2 shows the intensity distributions obtained when the laser was operated in the Hermite-Gaussian (1,0) mode for second-harmonic generation in LBO. Figures 2(a) and 2(b) show the input Hermite-Gaussian and Laguerre-Gaussian modes, respectively. Figure 2(c) shows the second-harmonic Laguerre-Gaussian and Fig. 2(d) is the Hermite-Gaussian recorded after the second mode converter. Figures 2(c) and 2(d) were then used to infer the mode indices of the second-harmonic Laguerre-Gaussian modes as outlined in the preceding paragraph. We find that for the frequency-doubled mode the azimuthal mode index l is doubled while p remains zero. We have investigated $p=0$ modes for $l=0,1,2,\ldots,7$ and found for each mode that second-harmonic generation doubles l. Figure 3 shows the same behavior for a laser operating initially in the Hermite-Gaussian (3,0) mode for second-harmonic generation in KTP.

Phase-matching for a nonlinear process depends on the vector addition of the wave vectors of the interacting fields. In a second-order nonlinear process, such as frequency doubling, optimum phase matching requires that

$$\Delta\mathbf{k}=\mathbf{k}_3^{(2\omega)}-\mathbf{k}_2^{(\omega)}-\mathbf{k}_1^{(\omega)}=\mathbf{0}, \qquad (3)$$

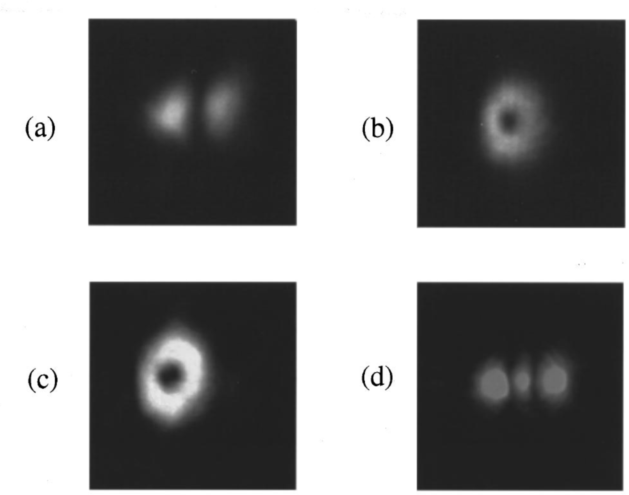

FIG. 2. Second-harmonic generation in LBO: (a) shows the input Hermite-Gaussian ($n=1$, $m=0$); (b) shows the corresponding Laguerre-Gaussian ($l=1$, $p=0$); (c) shows the Laguerre-Gaussian ($l=2$, $p=0$) produced after second-harmonic generation; and (d) shows the second-harmonic mode converted back to a Hermite-Gaussian ($n=2$, $m=0$).

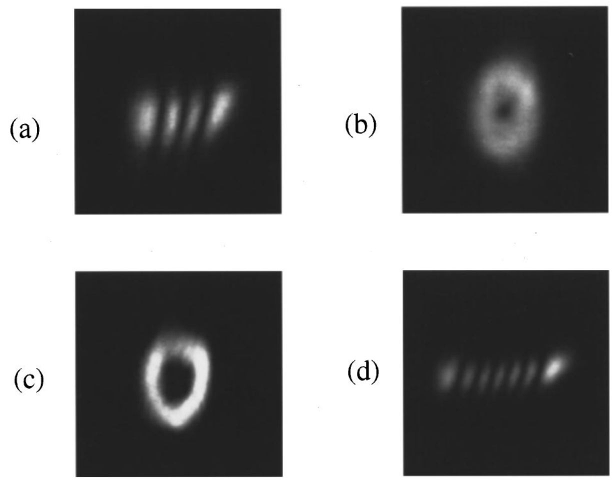

FIG. 3. Second-harmonic generation in KTP: (a) shows the input Hermite-Gaussian ($n=3$, $m=0$); (b) shows the corresponding Laguerre-Gaussian ($l=3$, $p=0$); (c) shows the Laguerre-Gaussian ($l=6$, $p=0$) produced after second-harmonic generation; and (d)shows the second-harmonic mode converted back to a Hermite-Gaussian ($n=6$, $m=0$).

where $\mathbf{k}_i^{(\omega')}$ is the wave vector of the ith field at frequency ω'.

For a type-I geometry both fields at ω have the same polarization, which is orthogonal to the polarization of the field at 2ω. In noncritical phase matching, all fields propagate perpendicular to the optic axis of the nonlinear material. The temperature of the material is such that the fundamental and second-harmonic light experience the same refractive index. Thus $\mathbf{k}_2=\mathbf{k}_1$, and for perfect phase matching,

$$\Delta\mathbf{k}=\mathbf{k}^{(2\omega)}-2\mathbf{k}^{(\omega)}=\mathbf{0}. \tag{4}$$

In isotropic media the wave vector $\mathbf{k}$ is parallel to the Poynting vector $\mathbf{S}$, but in an anisotropic material this is no longer the case. However, in the noncritical geometry, $\mathbf{k}$ is very nearly parallel to $\mathbf{S}$. The Laguerre-Gaussian modes have spiraling Poynting vectors, [5]. It is therefore not unreasonable to assume that Laguerre-Gaussian modes will exhibit phase-matching conditions different from those of a Hermite-Gaussian mode of the same frequency. In fact, we find that the phase-matching temperature for optimum second-harmonic generation is the same for modes of any order. This implies that in all cases, the wave vectors for the fundamental and second-harmonic Laguerre-Gaussian modes are parallel throughout the length of the crystal. It follows that the spiral paths described by the Poynting vectors must be of the same form for each mode.

For a Laguerre-Gaussian mode with $l\neq 0$ the helical wave fronts imply that the Poynting vector is not collinear with the direction of the beam. It has an azimuthal component such that its rate of rotation about the beam axis [9] is given by

$$\frac{\partial\theta}{\partial z}=\frac{l}{k^{(\omega)}r^2}. \tag{5}$$

If this expression is equated for the fundamental and second-harmonic frequencies, we obtain

$$\frac{l^{(\omega)}}{k^{(\omega)}r^2}=\frac{l^{(2\omega)}}{k^{(2\omega)}r^2}. \tag{6}$$

where $l^{(\omega')}$ is the azimuthal mode index of the light at frequency ω'. As $k^{2\omega}=2k^{\omega}$ it follows that $l^{2\omega}=2l^{\omega}$, as we have observed; it follows too that the orbital angular momentum per photon in the second-harmonic mode is twice that in the fundamental. As previously argued, this is entirely consistent with frequency doubling in the photon picture. A similar, if slightly more complicated, argument can be followed for type-II phase matching [10]. We have confirmed experimentally that the phase-matching angle is independent of mode and that the frequency doubling of a $p=0$ Laguerre-Gaussian mode again leads to a doubling of the azimuthal mode index.

We should note that the relationship between the beam widths $w^{(\omega)}$ and $w^{(2\omega)}$ means that the Rayleigh ranges for the fundamental and second-harmonic modes are identical. This is important, as the Rayleigh range of the mode dictates the design of the mode converter. It explains why it was possible to use the same mode converter to convert both the input and second-harmonic Laguerre-Gaussian modes to Hermite-Gaussian, so that the mode indices may be determined.

Laguerre-Gaussian modes where $p\neq 0$ were also observed to frequency double with the same phase-matching conditions as $p=0$ modes. However, in these cases the mode transformation that occurs is more complex. A $p\neq 0$ mode consists of a series of concentric rings. The nonlinear relationship between the field amplitudes of the fundamental and second-harmonic modes means that the amplitude distribution of the rings changes dramatically on frequency doubling. The second-harmonic beam can no longer be expressed in terms of a single Laguerre-Gaussian mode. Second-harmonic generation with these modes is the subject of further detailed study.

We have demonstrated the frequency doubling of a family of Laguerre-Gaussian modes with azimuthal index from 0 to 7 and $p=0$. The index of the mode has been found to double as well as the frequency. The observed doubling of the l index maximizes the overlap between the fundamental and second-harmonic fields and ensures that the wave vectors associated with the two fields are collinear. The experiment of He *et al.* [6] confirmed that linearly polarized Laguerre-Gaussian modes possess angular momentum. As in our work, the effects found in their experiment cannot arise from polarization and spin angular momentum but must be due to orbital angular momentum. Our experiment is thus consistent with the interpretation that the orbital angular momentum of the Laguerre-Gaussian mode is directly proportional to the azimuthal mode index l. This is as expected from the theory of Allen *et al.* [5], which shows that the ratio of the angular momentum to energy in a linearly polarized beam goes as $l\hbar/\hbar\omega$.

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