Phase memory preserving harmonics from abruptly autofocusing beams

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We demonstrate both theoretically and experimentally that the harmonics from abruptly autofocusing ring-Airy beams present a surprising property, they preserve the phase distribution of the fundamental beam. Consequently, this “phase memory” inherits to the harmonics the abrupt autofocusing behavior, while, under certain conditions, their foci coincide in space with the one of the fundamental. Experiments agree well with our theoretical estimates and detailed numerical calculations. Our findings open the way for the use of such beams and their harmonics in strong field science.

Introduction.---The preservation of wave packet phase, after the action of nonlinear or other phase deteriorating effects, is often referred to as “phase memory” [1,3]. Phase memory in general leads to a coherent wave packet behavior [1] while its physical origin is quite diverse ranging from quantum-mechanical physical system configurations [2,3], to optical wave packet distributions [1]. Optical wave packets that exhibit phase memory present exciting applications in strong field physics, like in higher harmonics and attosecond pulses [1].

Optical harmonics are generated by exploiting strong field interactions in nonlinear optical media. An intense pulsed beam of fundamental frequency $\omega$ with electric field amplitude $E(r,t)$ will modulate the dielectric polarization density $P(r,t)$ of the medium. This physical process can be described as [5]:

$$P(r,t) = \varepsilon_0 \chi^{(1)} E_\omega(r,t) + \varepsilon_0 \chi^{(2)} E_\omega^2(r,t) + \ldots$$

where $\varepsilon_0$ is the vacuum permittivity, and $\chi^{(1)}$, $\chi^{(p)}$, $(p = 2, 3, \ldots)$ are, respectively, the linear and the nonlinear optical susceptibilities of order $p$.

Each one of the nonlinear polarization terms can be correlated to a harmonic field $E_p(r,t)$, of order $p$, with phase and amplitude that are in general different to that of the fundamental. Phase memory in this case would be expressed as a preservation of the spatial phase of the fundamental in the harmonics, and can be extremely beneficial when using engineered optical wave packets [6,8] to tailor the harmonic generation process. For example, since the spatial phase controls the wave packet propagation, phase memory would lead harmonics sharing the same propagation properties as the fundamental.

Here we show that phase memory, during harmonic generation, is an inherent property of a family of optical wave packets whose distribution is described by the Airy function [9]. This family includes the accelerating Airy beams [10,12], as well as the cylindrically symmetric ring-Airy wave packets [13,14]. The spatial phase distribution of Airy beams leads to unique properties, including the propagation along parabolic trajectories [10,15,16], bypassing obstacles and self-healing [17,18]. Also, ring-Airy beams exhibit abrupt autofocusing, while experiencing only a minor nonlinear focus shift as their power is increased [19], making them ideal candidates for high intensity applications with precise deposition of energy in space [8,20].

Beside these exiting properties, we demonstrate that phase memory preserves the abruptly autofocusing behavior in the harmonics of ring-Airy wave packets, while under certain conditions their foci coincide in space with the one of the fundamental. We analytically show that this behavior is further preserved under the action of a converging lens for obtaining tighter focusing and higher intensities. Experiments and detailed numerical simulations of the second harmonic of ring-Airy beams validate our theoretical analysis. Our results open the way for the use of accelerating beams and their harmonics in a plethora of nonlinear optics applications.

Theoretical analysis.---Since the generation of harmonics involves powers of the field distribution we will start our analysis form the case of second harmonic generation. The amplitude of a ring-Airy beam is described as:

$$u_{\omega}(r) = u_0 \text{Ai}(\rho)e^{i\alpha \rho},$$

where $u_0$ is the amplitude, $\text{Ai}(\cdot)$ is the Airy function, $\rho \equiv (r_0 - r)/w$, and $r_0$, $w$, and $\alpha$ are, respectively, the primary ring radius, width and apodization parameters. Without loss of generality the amplitude of the generated 2nd harmonic in a thin BBO crystal is given by:

$$u_{2\omega}(r) \propto \chi^{(2)} u_{\omega}(r)^2 = \chi^{(2)} u_0^2 \text{Ai}(\rho)^2 e^{2i\alpha \rho},$$

where $\chi^{(2)}$ is the nonlinear susceptibility. By using well-known approximations of the Airy function [14] we reach to an analytical expression of the square of the Airy function term that appears in Eq. [2,21]:

$$\text{Ai}(\rho)^2 \simeq 2E(\rho)^2 + E(\rho)\hat{S} \cdot \rho \equiv \text{Ai}_2^2(\rho),$$

where $E(\rho) \equiv 1/[2\sqrt{\pi}f(\rho)^{1/4}]$, $\hat{S}$ is a linear scaling operator $\hat{S} \cdot \rho \equiv 2^{2/3} \rho + \pi/(8 \times 2^{1/3})$ and $f$ is an apodization...
Figure 1 shows a comparison $\text{Ai}(\rho)^2$ to the approximation $\text{Ai}_2^2(\rho)$.

Using the approximation of Eq. (3) we can write for the second harmonic:

$$u_{2\omega}(r,0) \propto 2\chi^{(2)}u_0^2 E(\rho)^2 + \chi^{(2)}u_0^2\text{Ai}(\rho')E(\rho)e^{2\rho\rho'}, \quad (4)$$

where $\rho' = (r' - r)/w'$, $r'_0 = r_0 + \pi w/16$, $w' = 2^{-2/3}w$. Equation (4) shows clearly that the amplitude of the generated second harmonic is composed by two terms, a smooth pedestal that does not exhibit any autofocusing behavior and a ring-Airy term that is strongly apodized, carrying $\sim 25\%$ of the total energy. The second term is a clear demonstration of phase memory, which will be resulting in abrupt autofocusing of the second harmonic.

One can calculate the focus position of the fundamental, using the 1D Airy analytical formulas [11, 14, 19]:

$$f^2_{\text{Ai}} = 4\pi {w_0^2\over \lambda} \sqrt{r_0/w + 1} \approx 4\pi {w_0^{3/2}\over \lambda} \sqrt{r_0}, \quad r_0 \gg w. \quad (5)$$

Under the same assumption ($r_0 \gg w$) we can estimate the autofocusing position of the second harmonic autofocusing term by using its primary ring radius $r'_0$ and width $w'$ parameters as estimated by Eq. (4):

$$f^2_{\text{Ai}}(\rho') = 4\pi {w'^2\over \lambda/2} \sqrt{r'_0/w' + 1} \approx 4\pi {w'^{3/2}\over \lambda} \sqrt{r'_0} = f^c_{\text{Ai}}, \quad (6)$$

Interestingly, this result shows that the autofocusing component of the second harmonic will autofocus at the same position as the fundamental. So, the phase memory in the second harmonic acts in a way that its propagation dynamics are identical to the fundamental. As for the pedestal term, it does not interfere with this focus since it diffracts out.

We can now generalize this analysis for the generation of ring-Airy harmonics of any higher order. As we demonstrate in the following, all harmonics exhibit phase memory and inherit the propagation dynamics of the fundamental. In more detail, for the case of even orders $\text{Ai}(\rho)^{2m}$, $(m = 1, 2, \ldots)$ Eq. (3) is generalized to [21, 22]:

$$\text{Ai}(\rho)^{2m} \simeq \text{Ai}_2(\rho)^m = \sum_{n=0}^{m} {2m-n\text{!m}! \over n!(m-n)!} E(\rho)^{2m-n} \text{Ai}(\tilde{S} \cdot \rho)^n. \quad (7)$$

Furthermore, whenever $n = 2^l$, $(l = 0, 1, \ldots)$ the corresponding power term $\text{Ai}(\tilde{S} \cdot \rho)^n$ in Eq. (7) will result to a sum of “net” ring-Airy terms $\sum_{l=1}^{2^m-1} (\cdots) \text{Ai}(\tilde{S}^{l-1} \cdot \rho)$, where $\tilde{S}^q \cdot \rho \equiv \alpha_q \rho + \beta_q$, and $\alpha_q = 2^{q/3}/\beta_q = \pi/(8 \times 2^{q/3})(1 - 2^{q/3}/(1 - 2^{q/3})$. Each of these harmonic ring-Airy terms will autofocus at positions [21]:

$$f_{\text{Ai}}^{2m\rho}(q) = 4\pi {w_0^2\over \lambda} \sqrt{r_{0\rho}/w_0} + 1 \approx m^{2-m} f^c_{\text{Ai}}, \quad (8)$$

where $\lambda(2m \omega) = \lambda/2m$ is the $2m$th harmonic wavelength, $r_{0q} = r_0 + \alpha_q \beta_q w$, $w_q = w/\alpha_q$, and $f^c_{\text{Ai}}$ refers to the autofocus position of the fundamental. Interestingly, in all the even harmonics that are a power of 2 (i.e. $2^{nd}$, $4^{th}$, $\ldots$) there will exist a term that will be autofocusing at $f^c_{\text{Ai}}$. On the other hand, for odd orders $\text{Ai}(\rho)^{2m+1}$, $(m = 1, 2, \ldots)$ Eq. (3) is generalized to:

$$\text{Ai}(\rho)^{2m+1} \simeq \text{Ai}_2(\rho)^m \text{Ai}(\rho) = \sum_{n=0}^{m} {2m-n\text{!m}! \over n!(m-n)!} E(\rho)^{2m-n} \text{Ai}(\tilde{S} \cdot \rho)^n \text{Ai}(\rho). \quad (9)$$

Clearly now there exists only one “net” ring Airy term (for $n = 0$). This harmonic ring-Airy term will be autofocusing at [21]

$$f_{\text{Ai}}^{2(m+1)\omega} = 4\pi {w_0^2\over \lambda(2m+1)\omega} \sqrt{r_{0\rho}/w} + 1 \approx (2m + 1) f^c_{\text{Ai}}. \quad (10)$$

Figure 2 shows an overview of these results up to the $40^{th}$ harmonic. Due to the phase memory, all harmonics exhibit abrupt autofocusing. We can clearly see that even orders exhibit multiple foci, and that when the order is a power of 2 there exists always a focus at $f^c_{\text{Ai}}$.

The next challenge in our analysis is related to reaching higher intensities compared to the ones achievable by autofocusing. A straightforward way of doing this is by using focusing optical elements. As we have previously shown [23], ring-Airy beams can be further focused using a lens, though they behave in a peculiar way, they exhibit double foci, a property characteristic to the family function [21].
of Janus waves [16] where they belong. The main question here is how their harmonics will be affected. Although in the following we are discussing the effect that focusing has on the second harmonic, our analysis can be easily expanded to the case of higher harmonics as well. As shown in Fig. [3] if we assume that a thin lens is located at a distance $\Delta z$ from the focus of the ring-Airy beam we get for the fundamental two foci at positions $z_1^\omega$, $z_2^\omega$ relative to the lens [23]:

$$z_1^\omega = \left( \frac{\Delta z^\omega}{\Delta z^\omega + f^\omega} \right) f, \quad z_2^\omega = \left( \frac{\Delta z^\omega - 2}{\Delta z^\omega - 2 + f^\omega} \right) f, \quad (11)$$

where, $f^\omega \equiv f/f_{\omega}$, $\Delta z^\omega \equiv \Delta z/f_{\omega}$. In the case of the second harmonic the situation is more complex. The second harmonic Airy term will lead to two foci at positions $z_1^{2\omega}$, $z_2^{2\omega}$ while the smooth pedestal term will focus at position $z_3^{2\omega}$:

$$z_1^{2\omega} = \left( \frac{\Delta z^{2\omega}}{\Delta z^{2\omega} + f^{2\omega}} \right) f \simeq z_1^\omega,$$$$

$$z_2^{2\omega} = \left( \frac{\Delta z^{2\omega} - 2}{\Delta z^{2\omega} - 2 + f^{2\omega}} \right) f \simeq z_2^\omega, \quad (12)$$

$$z_3^{2\omega} \simeq f,$$

where $f^{2\omega} \equiv f/f_{2\omega} \simeq f^\omega$, $\Delta z^{2\omega} \equiv \Delta z/f_{2\omega} \simeq \Delta z^\omega$. Interestingly, even after the focusing, the two foci of the second harmonic Airy term overlap with those of the fundamental. So, focusing does not deteriorate its phase memory inherited propagation dynamics.

Note that our analysis applies also to the harmonics and powers of 1D and 2D Airy beams, but in this case the resulting Airy terms in the harmonics do not result in any distinguishable accelerating features, since their propagation is shadowed by the propagation of the non-Airy terms [24][25]. Likewise, the above can be applied in the temporal domain for Airy pulses [12][27][29].

**Experimental part.**—We have performed second harmonic (SH) generation experiments using autofocusing ring-Airy beams. In our experiments we studied the propagation of the fundamental and SH, both in free propagation and after focusing by a lens. The experimental setup used for the generation of the SH is shown in Fig. [4]. A Ti:Sapphire laser system, delivering Gaussian shaped beams at 800 nm, 35 fs at 50 Hz repetition rate, was used. The ring-Airy beam was generated using a Fourier transform approach [14] in which the phase of the Gaussian laser beam was modulated using a spatial light modulator (SLM, Hamamatsu LCOS-X10468-2). The beam parameters (radius, width) were selected so that the beam abruptly autofocuses at $f_{\omega} = 400$ mm from the generation plane. For generating the second harmonic the autofocusing beam propagated in a type-I $\beta$-barium borate (BBO) crystal of 200 $\mu$m thickness. The conversion efficiency in this case was measured to be 7.3%. The fundamental and its second harmonic were then allowed either to freely propagate or were focused using a focusing lens $f = 100$ mm. The distances between the BBO crystal and that of the focusing lens from the FP were 60 mm and 146 mm respectively. Bandpass interference filters were used to isolate the fundamental and the second harmonic.

The transverse intensity distribution of the beam along its propagation was imaged by a linear CCD camera (14 bit), which was moved along the propagation axis $z$.

In the first experiments we report below we studied the propagation and autofocusing properties of the fundamental and its second harmonic without the presence of any focusing lens. Combining 2D transverse $(x, y)$ images that were captured at various positions along the propagation $z$ axis, the $I(x, y, z)$ intensity profile of the beam was retrieved. An $x$-$z$ cross section of such a profile is shown in Fig. [5] for the above case. Figure [5](a), reveals the autofocusing behavior of the fundamental ring-Airy beam, which focuses at $z = 400$ mm as expected, followed by the characteristic ring-Airy focus intensity distribution. As predicted by our model and shown in Fig. [5](b), the second harmonic exhibits a parabolic trajectory, autofocusing at the same focus position as the fundamental. The non-ring-Airy pedestal term diffracts out and appears as a lower intensity halo in the images.

Our analysis is completed through detailed numerical simulations of the SH generation process and the propa-
gation dynamics of the fundamental and its second harmonic. Our model is based on two coupled equations for monochromatic fields of the fundamental and its second harmonic [5]. In the coupling terms, we assumed perfect phase matching and the second order susceptibility, $\chi^{(2)}$, equal to $4 \times 10^{-12}$ m/V [30]. The corresponding numerical simulations, shown in Figs. 5(c), (d), are in very good agreement with the experimental results, while the analytical predictions of the foci positions nicely agree as well.

Placing a focusing lens after the BBO crystal, results in the peculiar behavior previously discussed. Figures 5(a), (c) show, respectively, the experimental and simulation results for the fundamental beam, presenting two discrete conjugate foci distributions. Again, a very nice agreement is found between the results from our experiments, simulations and analytical predictions, with the two foci located at $z_{1\omega} = 75.3$ mm and $z_{2\omega} = 122.6$ mm. Figures 5(b), (d) show, respectively, the experimental and simulation results for the second harmonic beam. Once more, the agreement between the results from our experiments, simulations and analytical predictions is nice. The central intense focus results from the pedestal term and is located at $z_{2\omega} = 92.7$ mm, practically at the focal plane of the lens. The other two foci are positioned before and after the focal plane of the lens at $z_{1\omega} = 67.4$ mm and $z_{2\omega} = 112.8$ mm, respectively. The combination of the three foci results in an elongated focal volume for the second harmonic, which, as one can clearly see in Fig. 4, overlaps to a great extent with the also elongated focal volume of the fundamental. This extended spatial overlap of the fundamental and second harmonic is of pivotal role in nonlinear wave-mixing experiments, like in the generation of intense THz fields using two-color ring-Airy beams [8].

Conclusions.—In conclusion, we have demonstrated theoretically and experimentally that the harmonics of autofocusing ring-Airy beams preserve the phase of the fundamental. Through an analytic approximation of the powers of the Airy function, we have shown that phase memory during harmonic generation is an inherent property of all optical wave packets described by an Airy distribution. Our findings are thus applicable to accelerating Airy beams (1D and 2D), cylindrically symmetric ring-Airy beams but also in the temporal domain for Airy pulses, and spatiotemporal Airy light bullets. The phase memory in the case of ring-Airy beams results in abruptly autofocusing harmonics, with their focus position coinciding with that of the fundamental for even harmonics of power 2. We have also demonstrated that even after focusing these beams still spatially overlap, surprisingly over elongated focal volumes. Our analytical predictions are in excellent agreement with second harmonic generation experiments and detailed numerical simulations. Our results open the way for using accelerating beams and their harmonics in a plethora of nonlinear optics applications, like for intense THz fields [8, 31], nonlinear wave-mixing [24] and filamentation [10, 19, 32].

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