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Ultrafast optical imaging by molecular wakes

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Ultrafast optical imaging is demonstrated by ultrashort laser pulse induced impulsive molecular alignment, involving optical image storage in the created molecular wakes followed by periodic readout and display. For diatomic molecules in air, both raised and intagliated monochromatic images are demonstrated, which is field-free, periodically achievable, and works as an ultrafast buffer memory for the imprinted optical images. Analogous to the holographic imaging, the phase information of a three-dimensional object can be revealed at various time delays. © *2010 American Institute of Physics*. [doi:10.1063/1.3505138]

Imaging with liquid crystal display[1,2] (LCD) technology has been rapidly developed since its invention and is becoming more and more popular in our daily living for information displaying of text, images, and videos. It basically relies on dynamic alignment control of the liquid crystal molecules with an electric field that facilitates electric-to-optical signal conversion. To date, the LCD technology has already surpassed the conventional cathode-ray tube technology in providing thin and light high-resolution display. It has also stimulated great progress of other display technologies such as organic light-emitting diodes,[3] and recently laser televisions.[4] On the other hand, ultrafast all-optical switches are desired for ultrafast optical imaging, while the switch on/off time of the liquid crystal molecules is limited. Optical image should be better stored in a buffer memory that could be read out later on for further image processing.

We experimentally demonstrate here an all-optical imaging and display technology that supports image storage and periodic readout by using the impulsive molecular wakes[5,6] excited by ultrashort laser pulses. Ultrashort laser pulse induced alignment of gaseous molecules has been extensively studied for molecular orbital reconstruction,[7] high harmonic generation,[8] ultrashort light pulse modulation,[9,10] and so forth. The ultrafast wave plates of the molecular wakes can be further explored for ultrafast optical image processing with multiple functions as the image storage, all-optical display, periodic readout, and time-division phase encoding. Both raised and intagliated monochromatic images are obtained by using various revivals of the molecular wakes, which can also reveal the phase information of a three-dimensional object.

The experiments were performed with an output from an amplified Ti:sapphire laser system (35 fs, 800 nm, 1 kHz), which was split by a beam splitter with one of them as the writing pulse and the other as the reading pulse. As shown in Fig. 1, the writing pulse stored the image information of the mask in the created molecular wakes by impulsively aligning the diatomic molecules in air. The energy of the writing pulse after the mask was generally set to be 0.7 mJ per pulse, and could be tuned by changing a half wave plate before a thin film polarizer in its arm. The reading pulse at 400 nm is obtained by frequency doubling the fundamental-wave pulse at 800 nm with a 100 μm thick beta barium borate (BBO) crystal, which was collinearly combined with the writing pulse by a dichromatic mirror. The polarization of the writing pulse was rotated to be 45° with respect to the reading one, so that an optimal imaging contrast ratio was achieved. The succeeding reading pulse was then used to display the stored images at desired time by tuning its time delay to properly match the molecular wakes with a motorized translation stage. The transmission direction of the polarizer was rotated to be orthogonal to the polarization of the input reading pulse. A broadband high reflective mirror centered at 800 nm and a low-pass filter were used to block the leaked writing pulse before a two-lens imaging system at a 4f configuration. At the output of imaging system, the images were captured with a monochromatic charge coupled device (CCD).

We took molecular N_2 and O_2 in air as example, which were impulsively aligned by femtosecond laser pulses.[11] The field induced dipole moment introduces a net torque to the diatomic molecule when it is exposed to intense laser pulse, which forces the molecule to align along the laser field polarization.[5] Quantum mechanically, by using ultrashort laser pulse with broadband spectrum, a series of rotational molecular wave packets are coherently excited and coupled through the impulsive Raman processes. After the extinction of the ultrashort excitation pulse, the quantum beatings of the pre-excited rotational wave packets lead to the parallel and perpendicular revivals of the molecule with period determined by its rotational constants,[5] referred as molecular wakes. Since the diatomic molecule shows different refractive indexes for the laser field components polarized parallel

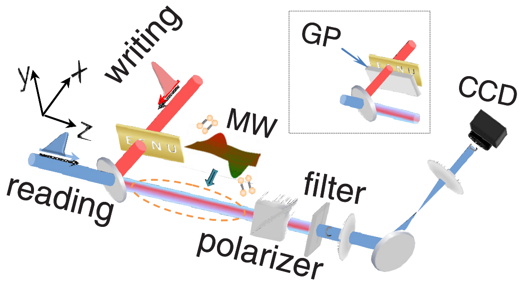

FIG. 1. (Color online) Schematic illustration of the experimental setup. The writing and reading schemes for holographiclike imaging is shown as the inset. MW: molecular wakes, GP: glass plate.

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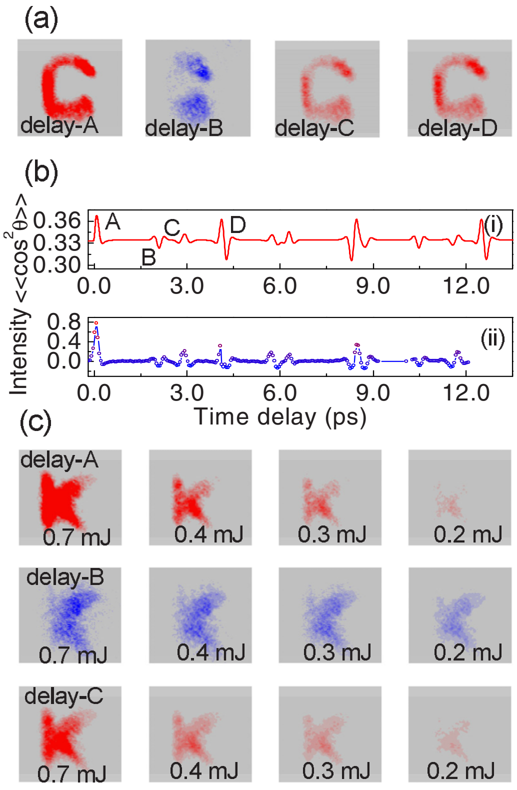

FIG. 2. (Color online) Monochromatic imaging by molecular wakes. (a) The measured monochromatic images of the imprinted ones. (b) The simulated revivals of the molecular wakes of the air molecules (i), and the integrated intensity of the imaged characters vs the time delay of the reading pulse (ii). (c) The measured dependence of the imaging contrast ratio on the pulse energy of the writing pulse when the time delay of the reading pulse is tuned to match various revivals of the molecular wakes.

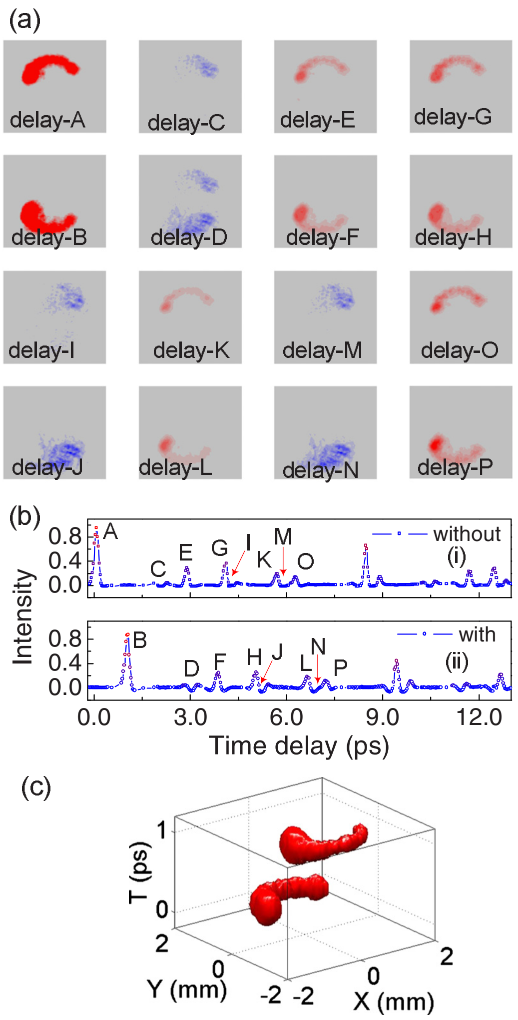

FIG. 3. (Color online) Holographiclike imaging by molecular wakes. (a) The measured images of a three-dimensional object with an glass plate after the mask at various revivals of the molecular wakes. (b) The integrated intensities of the imaged characters vs the time delay of the reading pulse for the object parts without (i) and with (ii) an inserted glass plate. (c) The intuitive presentation of the phase modulated object of "C," which clearly shows a phase delay of $\sim$1 ps induced by the inserted glass plate.

and perpendicular to its axis, the molecular alignment revivals could be treated as ultrafast wave plates with noticeable birefringence to modulate the polarization state of a properly matched ultrashort laser pulse,[12,13] which is the basis of the optical imaging we demonstrated in what follows. The writing pulse created molecular wakes in space according to the imprinted image of the mask, which rotated the polarization state of the reading pulse accordingly, and the imprinted image could hence be recovered by monitoring the transmission pattern of the reading pulse after the polarizer. We could read the stored information at a desired revival time up to a hundred picoseconds for room-temperature air until the substantive decay of the molecular wakes.

We first demonstrate the monochromatic optical imaging of the imprinted images through the molecular wakes. Figure 2(a) shows the imaging results of the character "C" at various time delays [as labeled in Fig. 2(b)(i)] with respect to the writing pulse, where the background was subtracted from the captured images and a mean intensity level was then restored. Figure 2(b)(i) shows the simulated impulsive molecular alignment of room-temperature air excited by an ultrashort laser pulse. The first molecular wake appeared around $\sim$80 fs after the writing pulse excitation[14] (delay A), and then revived with periods of 8.3 ps and 11.6 ps, respectively, for molecular N_2 and O_2.[5] Therefore, as shown in Fig. 2(a), the stored image information could be read out periodically during the revivals of the molecular wakes even after the extinction of the writing pulse. Different contrast ratios of the images at different delays account to various revival strengths of the molecular wakes as indicated in Fig. 2(b)(i). The molecular wake was strongest around delay A as a combination of both N_2 and O_2 molecules. The strength of the molecular wake was closely dependent on the pump intensity of the writing pulse. Before its saturation, an intense writing pulse created a strong molecular wake. For instance, as shown in Fig. 2(c) for the character "K," the imaging contrast ratio was improved as the writing pulse intensity increased, corresponding to the increased strength of the molecular wakes.

Interestingly, as shown in Fig. 2(a) for the time delay B which accounts for the antiphase revival of the molecular wake, the intensity of the image is less than the background, referred as intagliated image. Here, we use the red and blue colors to represent the intensities are greater and less than the background, respectively. These intagliated images were induced by the cross-defocusing effect[13,15] of the perpendicularly aligned molecules, which not only changed the polarization state but also spatially diffracted the reading pulse. On the other hand, the in-phase revival of the molecular wakes led to raised images based on the cross-focusing effect[13,15] of the parallel aligned molecules. Therefore, for an imprinted image, both raised and intagliated images could be obtained by tuning the time delay of the reading pulse to match the in-phase and antiphase revivals of the molecular wakes, which are clearly shown in Fig. 2(b)(ii) by integrating the intensities of the imaged characters. The values greater (less than) zero stands for the raised (intagliated) images.

Analogous to the holographic imaging,[16,17] the molecular wakes based imaging technology also shows the capacity to extract the phase information of a three-dimensional object. One of the most important aspects of the holographic imaging is that the distance-dependent phase information of the images can be revealed, which is usually recorded by interfering the signal beam with a reference one and then recovered in the reversing approach.[16] Here, the phase information of the object could be similarly extracted by considering the time-delayed response of the molecular wakes. For instance, different transparent parts of a three-dimensional object with different thicknesses introduce different group delays of the writing pulse, which cause the molecular wakes to be created at different time delays. This results in a time-division encoding of the thickness-dependent phase information of a transparent three-dimensional object. It can be further extracted by a reading pulse of a scanning time delay in a way as the time-phase tomography.

As an instant demonstration, as shown in the inset of Fig. 1(a) 500 μm thick glass plate was partly inserted after the mask to introduce a phase difference of the object. It accounts for an effective optical thickness of $\sim 300\ \mu$m for a refractive index of ~ 1.6. As shown in Fig. 3(a), the two different parts of the character "C" with and without glass plate were imaged at different time delays by the reading pulse. Figure 3(b) shows the integrated intensities of the imaged characters for the parts with and without the glass plate by scanning the time delay of the reading pulse, which clearly indicates the corresponding molecular wakes for raised and intagliated images of the object. The shifted time delay of ~ 1 ps for the curve in Fig. 2(b)(ii) with respect to that of Fig. 2(b)(i) is consistent with the effective thickness of the glass plate. This time-delayed imaging indeed reflected the different thicknesses or optical paths of the three-dimensional object, which was analogous to the holographic imaging technology. As more intuitively presented in Fig. 3(c), the object parts with and without glass plate are displayed as the time delay of the reading pulse was increased. An interesting intagliated image of the whole object at delay D as shown in Fig. 3(a) was caused by the apropos overlapping of the molecular wakes of oxygen and nitrogen molecules for these different parts [see Fig. 3(b)].

In summary, we have experimentally demonstrated an all-optical ultrafast imaging technology by using ultrashort laser pulse excited molecular wakes, which shows the capacities of raised and intagliated monochromatic imaging and holographiclike imaging. This technique may serve as a promising extension of the well-developed LCD technology.

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