

Imaging Labeling: A Graphical Interface to Correlation in Multiparticle Ejection Dynamics

K. Zhao, G. Zhang* and W. T. Hill, III

Institute for Physical Science and Technology

and

Laboratory for Atomic, Molecular and Optical Science and Engineering

University of Maryland, College Park, Maryland 20742

Abstract

A novel graphical approach for probing multicomponent collective ejection processes is described. *Image Labeling* provides a visual means for identifying ejection partners and their relative momenta, isolating specific decay channels for detailed study and determining initial electronic and/or molecular geometries prior to ejection. The power of the technique is demonstrated by looking at strong-field multiphoton-induced 3-atom Coulomb explosion spectra.

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*Permanent Address: Tianjin University, PR China

I. INTRODUCTION

There has been growing interest, recently, in multiphoton nonsequential ionization and dissociative-ionization processes. This interest is due to a desire to understand the physics associated with collective phenomena driven by manybody wavefunctions leading to results that cannot be explained by single-particle wavefunctions. Imaging, combined with correlation techniques, have proven to be extremely valuable for such studies [1], [2].

In a previous paper [1], we introduced an image spectrometer that is capable of collecting and analyzing charges ejected over 4π sr with high energy and angular resolution. We demonstrated that spatial imaging makes it possible to identify ejection partners unambiguously and determine if a specific charge is ejected by itself or in conjunction with another charge. This ability allows us to distinguish collective and sequential processes. The focus of our earlier work was on two-atoms systems, H_2 and H_2^+ in particular, and one-electron ionization in which only two entities needed to be correlated. Our current focus is on more complicated systems – three-atom systems and two-electron atoms – in which three or more entities must be correlated. To study such systems, we extended our approach developed for two-component correlation. One of these extensions involves a new way to visualize correlation in the ejection dynamics; we call this technique *image labeling*. Image labeling not only allows correlated partners to be view graphically, it allows the degree of correlation to be measured and the initial geometry prior to the multiphoton-induced atomic or molecular fragmentation to be determined.

II. IMAGE EJECTION DYNAMICS

We will demonstrate the utility of image labeling on the Coulomb explosion [3] spectrum of 3-atom systems. Linearly polarized, diffraction limited 100 fs pulses at 800 nm from a mode-locked Ti:Sapphire laser were used to induce the Coulomb explosions. The pulses were focused into the 4π image detector with a 150 mm radius of curvature, 38 mm diameter

spherical mirror to intensities that could be varied between 10^{14} and 10^{16} W/cm².

The detector's design and principle of operation have been described in detail elsewhere [1] [4]. Briefly, it consists of an image quality microchannel plate (MCP) with a phosphor screen at the back end. Laser pulses are brought to a focus a distance l from the center of the MCP with their polarization axis oriented parallel to the MCP. Charges are swept toward the MCP with a uniform static electric field (~ 250 V/cm) as they are ejected from the focal point. The center of mass of the ejection dynamics coincides with the lab frame with its origin at the center of the image. The momentum distribution of the ejected charges is proportional to the spatial intensity distribution of the light emitted by the phosphor. The light is digitized by a digital ccd camera and recorded to disk in real time at up to 730 Hz. The laser repetition rate is set to match the camera frequency so that each frame corresponds to one and only one laser pulse. The camera exposure time and the MCP/phosphor gain are adjusted to produce a zero background while allowing near single-charge detection.

Measurements are made at gas pressures between 1×10^{-9} and 3×10^{-8} Torr to minimize space charge effects. We exploited the different arrival times for ions of different mass and charge state to isolate specific charges for detailed study by gating the MCP. A temporal resolution of 100 ns was sufficient to separate low mass charges (H, C, N, O, etc.) and their various charge states. Figure 1 shows a typical 3-atom Coulomb explosion image. The figure displays the momentum distribution of doubly charged N and O ions originating from the explosion of NO₂. This total momentum distribution image is a composite of 500,000 camera frames (i.e., 500,000 laser shots).

Along the polarization axis in Fig. 1, energetic ions appear in the image at a distance from the center of the image given by

$$r = \sqrt{2l\mathcal{E}/qF} \quad (1)$$

where \mathcal{E} is the energy of the ion, q is the charge on the ion, F is the static electric field pushing the ions toward the detector and l is the distance between the focal point and the detector. Since the angular distribution is independent of the azimuthal angle about the

polarization axis, ϕ , Eq. 1 holds absolutely only for polar angles $\theta = 0$ and π . It is possible to deconvolve the images so that Eq. 1 holds for all θ (see Ref. [4]). Although deconvolved images can be used to determine energy and angular distributions, they cannot be used to identify decay partners uniquely when three or more charges are ejected together. In this case, a unique identification is only possible with correlation techniques, such as the one we describe next.

III. IMAGE LABELLING

Image labeling is the 2-dimensional analog of *covariance mapping* that has been employed so successfully to analyze 2-atom Coulomb explosions [5]. The technique allows all the correlated partners to be visualized with a family of momentum images similar to the total momentum distribution image but with only the correlated partners highlighted. We call each of these images a *correlation image* and the family of images a *correlation map*.

To produce a correlation image, we divide the momentum distribution image in Fig. 1 into sectors, $\mathcal{S}(r,\theta)$. The size of each sector is chosen such that $\Delta\mathcal{E}$ ($\propto r\Delta r$, see Eq. 1) and $\Delta\theta$ are the same for all sectors. We show this in Fig. 2 for the image in Fig. 1. Next, we calculate the statistical correlation, C_{ij} , between sectors i and j , which is given by

$$C_{ij} = \frac{\langle \mathcal{S}_i \cdot \mathcal{S}_j \rangle - \langle \mathcal{S}_i \rangle \cdot \langle \mathcal{S}_j \rangle}{\sqrt{\sigma_{\mathcal{S}_i} \sigma_{\mathcal{S}_j}}}, \quad (2)$$

where $\langle \rangle$ means we average over all laser shots and $\sigma_{\mathcal{S}_i}$ is the square root of the variance in the counts for sector i with laser shots. For C_{ij} to be meaningful, it is critical that the average number of ions generated per sector per laser shot be less than one and that the momentum distribution for each laser shot be stored separately. The i^{th} correlation image, which labels the i^{th} sector, is assembled by replacing the ion counts for the j^{th} sector in the composite image (i.e., the upper left image in Fig. 2) with the correlation value C_{ij} . The right image in Fig. 2 shows one correlation image of a 3-atom Coulomb explosion in NO_2 . There are as many correlation images as there are sectors. Typically, we employ about 500 sectors.

The correlation images not only tell us which ions are ejected simultaneously, they give us a clearer picture of the explosion dynamics. Specifically, they make it possible to extract the final momentum of each of the correlated charges and the total energy for the event. The length and direction of the arrows in the right image in Fig. 2 indicated the momentum of each charge relative to the center of mass of the dynamics, the center of the image. Imposing energy and momentum conservation allows us to determine the initial configuration (e.g., internuclear separation and bond angle in the case of molecular Coulomb explosion) of the system just prior to the explosion. For example, the correlation image in Fig. 2 corresponds to the case where six electrons are removed from NO_2 (six-electron channel) leading to N^{2+} ions moving downward correlated with O^{2+} ions moving toward the upper left and right corners of the image. In this particular case, the explosion occurred from a geometry where the bonds between the N and O nuclei are stretched about a factor of 2.5 compared with the equilibrium bond lengths ($\simeq 0.12$ nm) while the bond angle is approximately the same as the equilibrium bond angle (134°). The correlation map, the family of correlation images, provides a way to study the ejection dynamics as a function of the momentum of a particular partner. Each correlation image corresponds to labeling a different regions of the momentum distribution image.

We are using image labeling to explore the details of strong-field dynamics of linear and bent systems. Figure 3 shows how we can isolate linear and bent channels for Coulomb explosions of CO_2 . In distinction with NO_2 , the equilibrium geometry of CO_2 is linear. We can study the linear channel by labeling ions along the polarization axis as done in the left image. In this case we label an O ion on the righthand side of the system and the correlation image highlights the companion O ion on the opposite side of the systems as well as the C ion in the center. The bent channels are studied by labeling the C ions that are ejected along an axis perpendicular to the polarization axis.

Given the energy and momentum of each partner, it is possible to test models of the Coulomb explosion quantitatively. Consider CO_2 again. From Fig. 3, we can extract an initial potential energy, the classical electrostatic potential, prior to explosion of about 58 eV.

This implies a C-O separation prior to the explosion of about 0.25 nm. Ionization at extended lengths in diatomic systems at 100 fs has been well documented [6] [7] [8] and apparently is active in 3-atom systems as well [9]. We also extract a maximum initial bond angle of about 170° . This maximum angle can be understood in terms of the bending vibrational frequency. For the ground state of CO_2 , this frequency is 667 cm^{-1} ; the corresponding the force constant is about 57 N/m. Classically, these parameters will produce a maximum bend angle in the neighborhood of 168° . Consequently, the 170° angle we extract is understandable.

The family of correlation images provide a quantitative measure of the correlation as well. The degree of correlation, for example, can be determined by comparing the C_{ij} values of the correlated area with that of the labeled area. Although the C_{ij} values for the sectors can be used for this, we find it more accurate to work directly with the pixels, as shown in the lower left image of Fig. 2. This correlation image was generated by taking the difference between the average distribution shown in the upper left in Fig. 2 (all 500,000 frames) and an average distribution composed only of frames that have nonzero counts in the i^{th} sector - a selective average. In this particular example, there is nearly unit correlation between the labeled and correlated areas. This selective average approach provides better resolution and more direct control over the size of the labeled area.

IV. ISOLATING AND CONTROLLING DYNAMICS

In closing we point out that image labeling should be a useful tool for quantum control studies. The 3-atom strong-field dissociative ionization we have been focusing on in this paper serves to illustrate this point as well. The complete breakup of a system XYZ into X , Y and Z ions can proceed through several possible channels. Two possibilities are a sequentially channel, involving an intermediate state such as $XY + Z$ followed by the breakup of XY , and a direct channel where $X + Y + Z$ ions are produced in one step. Figure 4 shows these channels not only exist in NO_2 , they can be distinguished unambiguously. An ability to isolate multiple channels to essentially the same final state (complete decomposition in this

case) provides an opportunity to focus on the *Lagrangian* of the system, which determines which path the system takes not just the final products. Furthermore, since labeling isolates, and hence, simplifies the momentum distribution image, it might possibly provide a clearer picture of the physics undergirding optimal control algorithms.

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- [3] By Coulomb explosion, we mean the energetic dissociation of a multiply ionized molecule into atomic ions. The final kinetic energies of the atomic ions are determined mostly by the classical electric potential associated with the internuclear separation of the ionized nuclei just prior to the explosion.
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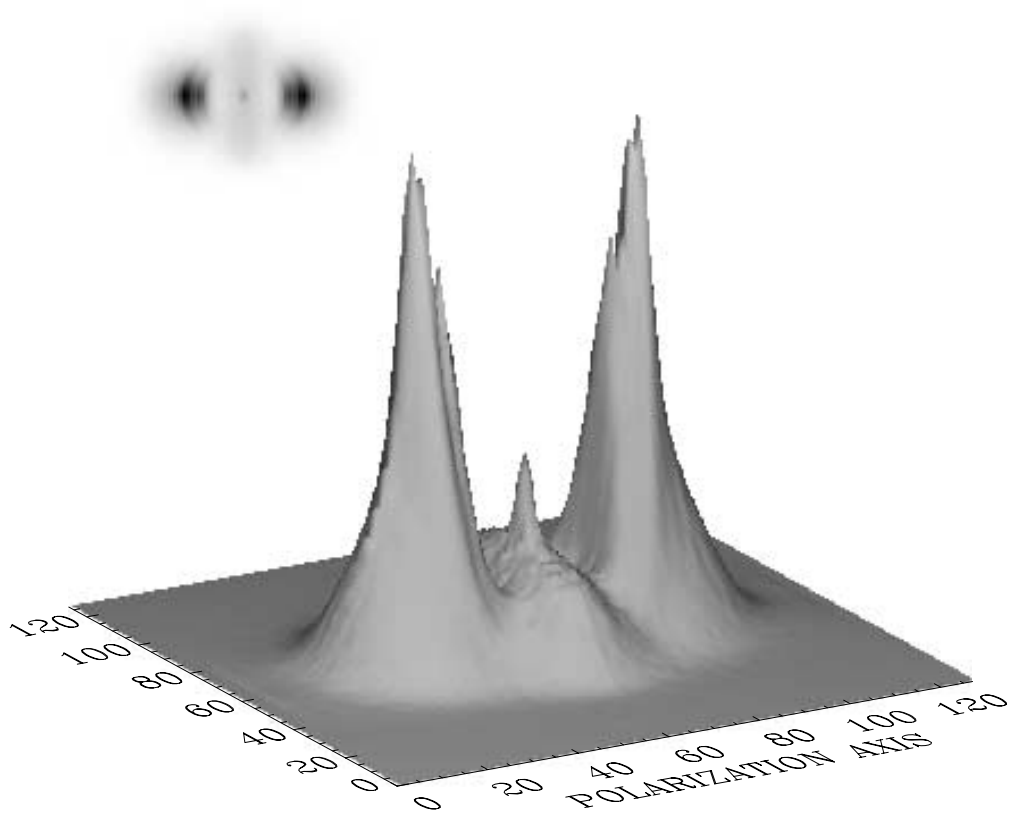
FIGURES

FIG. 1. The Coulomb explosion image of NO_2 (inset) and surface plot showing the momentum distribution of N^{q+} and O^{q+} ions ($q = 2, 3$). This distribution contains 500,000 laser pulses, each centered at 800 nm, with a pulse width of 100 fs and linearly polarized (horizontal in the inset) with a peak intensity of 10^{15} W/cm². The vertical distribution is composed of N^{q+} ions while the distribution parallel to the polarization axis is a mixture of both N^{q+} and O^{q+} ions.

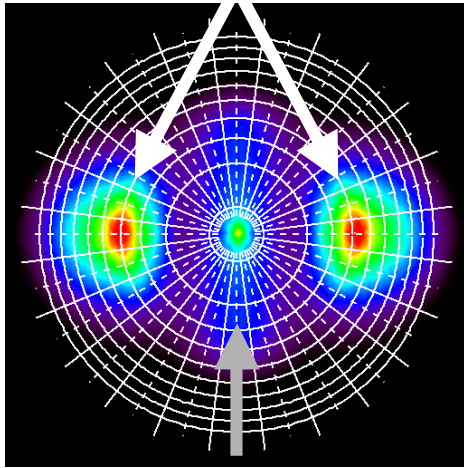
FIG. 2. This figure explains how to read a correlation image. The concentric circles and spokes on the upper left images divide the momentum distribution (the same as that displayed in the inset of Fig. 1) into sectors. The grey arrow indicates *labeled* ions (i.e., a subset of ions with a narrow momentum distribution moving downward, 6 o'clock); the white arrows indicate the correlated sectors (ions moving toward 2 and 10 o'clock). The figure on the right shows the correlation image for the Coulomb explosion where all three atomic ions are ejected simultaneously. This image shows the momenta of the charges ejected simultaneously. The grey (white) arrows indicate the final momenta of the labeled (correlated) charges. The lower left image is the difference between averaging only those frames that have a nonzero count in the labeled sector and the average of all 500,000 frames.

FIG. 3. Two correlation images for the Coulomb explosion of CO_2 taken under the same conditions as Fig. 1. We label O ion moving toward 3 o'clock in the left image and those moving toward 6 o'clock in the right image. We isolate linear explosion events on the left and bent events on the right.

FIG. 4. Two correlation image for NO_2 taken from the same data set as Fig. 1 showing a sequential channel 2-atom channel (left) and a direct channel. The sequential channel is an explosion into $\text{NO} + \text{O}$ followed by the breakup of NO . Pictured on the left is the explosion of NO . The explosion dynamics is asymmetric, the center of the image is not the center of mass, because the NO has nonzero kinetic energy due to the explosion in the first step.



READING CORRELATION IMAGES



AVERAGE IMAGE



CORRELATION IMAGE

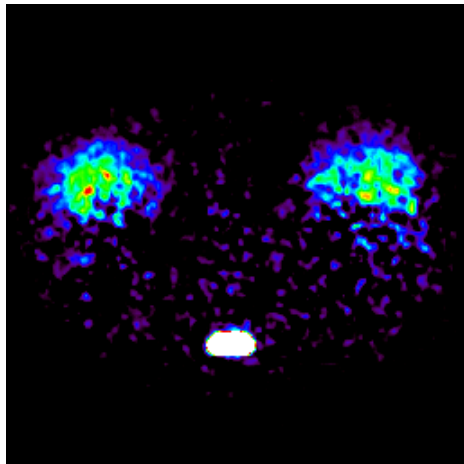


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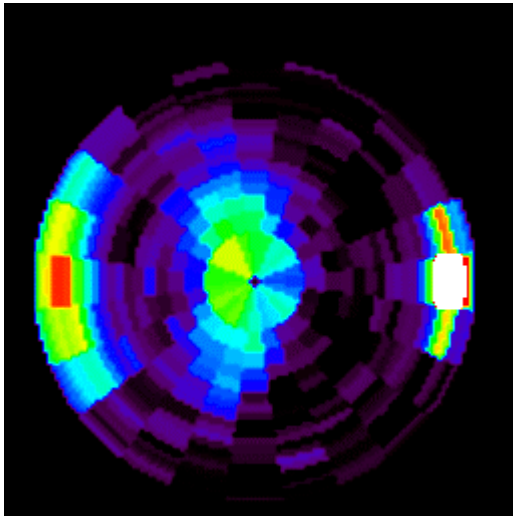


POLARIZATION
AXIS

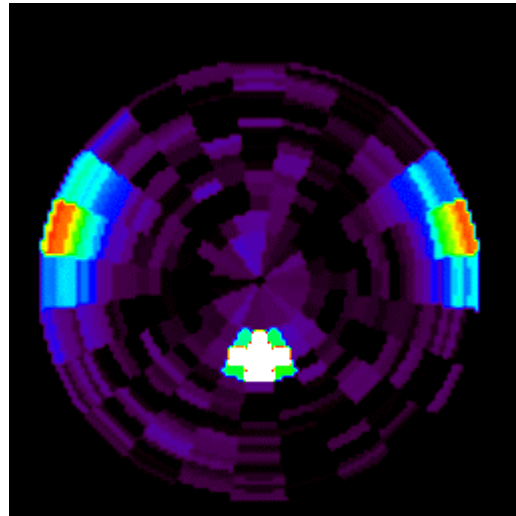


SELECTIVE AVERAGE

CO₂ CORRELATION IMAGES



LINEAR EXPLOSIONS

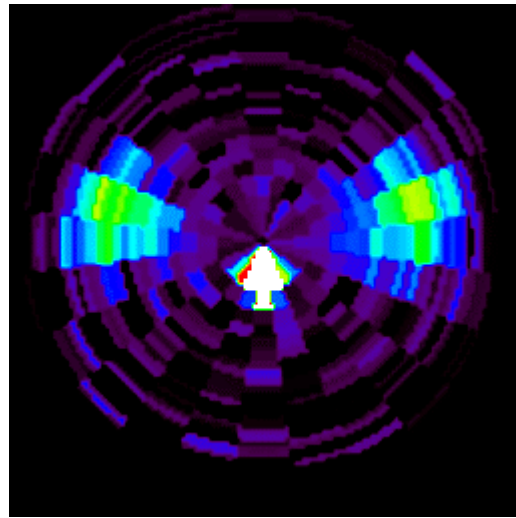
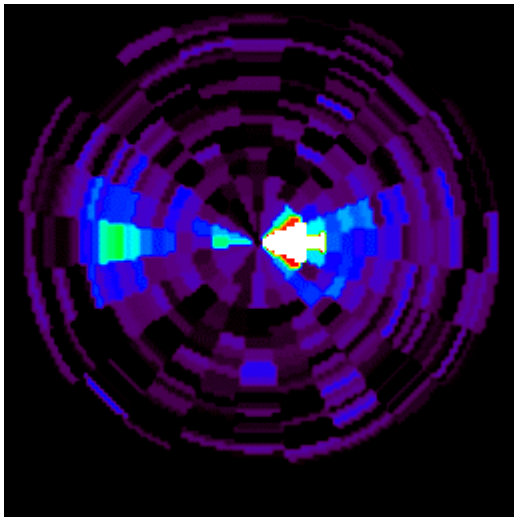


BENT EXPLOSIONS



POLARIZATION AXIS

NO₂ CORRELATION IMAGES



SEQUENTIAL CHANNEL

DIRECT CHANNEL



POLARIZATION AXIS