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Dear Dr. Jaydev Upponi and Dr. Benjamin Werth

Editors of the Journal of Visualized Experiments

Thank you for the reply and editorial comments on our manuscript,

JoVE54917 "Direct Imaging of Laser-driven Ultrafast Molecular Rotation".

Following Editorial and reviewer’s comments, we have modified the manuscript.

The followings are the replies to the comments.

Because both reviewers recognize the novelty of the present method, and the Review 1 agree to the submission to JoVE after the minor revision, we hope the present version satisfy the standard of JoVE publication.

**Editorial comments:**

•Many errors in English remain in the manuscript. Additional editing is required prior to acceptance, should be performed by a Native English speaker. Some specific errors are noted below:

-Line 60 – “of motions of interests”

-Line 77 – “motionusing”

-Line 78 – “have led” – should be “has”

-Line 103 – “seriously low” – “Seriously” is the wrong word

-Line 124, 213, 295, 740 – “equipments” or “equipment’s”

-1.5.7 – “a standard mounting procedures”

-2.2.2 – “passthrough”

-2.4.2 – “polarizedsecond”

-2.5 – “each pulses”

We are sorry for the typo and some errors due to the unexpected work of Word program. We have carefully checked the manuscript, and all the above mentioned errors have been fixed.

•Additional detail is required:

-2.3.1 – How are the pulses obtained? Is there a particular readout to watch? Is a laser turned on? Please describe for filming.

New NOTE was added to clarify the origin of the pulse.

-2.3.2 –How are they aligned parallel (are some parts of the apparatus adjusted to achieve this)? Please describe what actions are being performed for filming.

A sentence and NOTE which describe the detail were inserted in the 2.3.2.

-2.5.1 note – Please clarify “the purpose of two pumps”.

•Formatting: 2.4.3/2.5 – There is a heading for 2.5 at the end of this step, plus an additional 2.5 heading below it.

Both of them were errors to be deleted. We fixed them.

•Visualization: The protocol is discontinuous. Please highlight 4.2.2 and 4.5.3.2, which appear to be important for success of the protocol. Make sure that all steps required for success of the protocol are highlighted for filming (excepting computational steps).

We highlighted the steps you suggested.

•Results: Please provide scale bars in Figure 5.

The scale bar is in the top-right of the figure. The second column is also scaled in the polar plot. The third cannot be scaled, because they are schematics as indicated in the caption.

•JoVE reference format requires that the DOIs are included, when available, for all references listed in the article. This is helpful for readers to locate the included references and obtain more information. Please note that often DOIs are not listed with PubMed abstracts and as such, may not be properly included when citing directly from PubMed. In these cases, please manually include DOIs in reference information.   
 DOI info are now included.

Please include a line-by-line response letter to the editorial and reviewer comments along with the resubmission.

Reviewer 1:

>> I especially like the expanded discussion on how the "time zero" of this experiment is achieved

We added detailed NOTE in 2.5.1 to clarify this.

Major Concerns:

1. Other 3D techniques should be cited such as Vallance and co-workers and Li and co-workers in the introduction.

We added all the important reference as recommended.

Minor Concerns:

2. Section 2 - Construction of the optical set-up; it would be helpful to describe the types of optics used (such as are the beamline mirrors all metallic or dichroic high reflectors). Nothing more than a sentence is needed on this.

3. More information regarding the optical set-up could be provided. For example, the NLC (Non-linear Crystal) material + thickness + cut and importantly the conversion efficiency should be added. Additionally, some samples of the laser power along different parts of the beam would also be instructive. Many of these could be simple additions added to Figure 3, or a few extra lines of text.

In section 2, the types of optics and NLC are indicated and some of them are listed in the materials list.xls. Laser pulse energy is shown through the section 2.

4. Do they use any additional pulse control to remove any chirp introduced from the beam line? Or is the pulse pre-chirped for highest power or temporal pulse duration at the experiment?

5. The authors should state their cross correlation of their three beams as this limits the ultimate temporal resolution

The new sentences and NOTE have been added in the 2.5.6 to address the above two comments.

6. What are the rotational levels of N2 are present in the ensemble in the molecular beam? And what is its rotational period?

We added descriptions for this in 1.8, 4.2.3, and 4.5.4.

7. What causes their laser stability issues? Is it vibrations through the table or thermal effects changing the laser pointing? It is worth mentioning for readers who are interested in building a similar set-up.

Some details on the laser stability are given in the Discussion section.

Reviewer 2:

>>(1) their protocol contains many very detailed instructions, some indeed very important for proper implementation of the technique, but many others completely meaningless,…

>>(2) their protocol provides a detailed description of the methods that they use in the laboratory, but more of than not, fails to describe the reason for a certain action. That makes the protocol more difficult to follow and less useful than it should be. For example, in 1.2 the authors write "Attach the pulsed valve to the chamber partition using four threaded posts (15 cm, diameter 12 mm),…

Most of the suggested points relate to the NOTE added to follow the Editorial comment for previous revisions. We agree with the reviewers comment “but meaningless (and more than likely, wrong) for similar setups that other researchers would put together, unless their setup becomes an exact clone of the setup used by the authors”, but we also have to describe details on apparatus in order to clarify what we actually built up. Therefore, we added many comments and NOTEs throughout the manuscript to notify the essence of the procedures and meaning of details.

>>In order for the protocol to possibly become such a useful resource, I think it is essential that the authors alter the scope of the protocol, limiting the protocol to the novel aspects of the method that they have developed

Following the editorial policy of JoVE, we think limiting the procedure description is not suitable and then, we just added comment on the specificity of the present manuscript and the notification of the readers who would like to use the present method.

 We hope this modified manuscript is satisfactory for the editor and reviewers.

Sincerely,

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