Reviewers' comments:

Reviewer #1 (Remarks to the Author):

In the present manuscript, Moller and coauthors presents a new stroboscopic ultrafast measurement scheme using Lorentz TEM and systemically engineered external current excitation. Authors present clock-wise and counter-clock-wise rotation of magnetic vortex core in Permalloy, excited by sinusoidal spin transfer torque injection along electrodes. I believe the manuscript is well-written in general, and appropriate credits are given to prior works.

Ultrafast imaging of magnetic configuration has been particularly highlighted these days, as recent studies on the dynamics of exotic magnetic textures (e.g. skyrmions, vortices) have exhibited appealing properties for potential low-power spintronic applications. Although there exist other techniques such as stroboscopic scanning transmission X-ray microscopy (STXM) [Nat. Commum. 8, 15573 (2017)] or magnetic holography [Nat. Phys. 11, 225–228 (2015)], these techniques are only sensitive to perpendicular magnetization and thus cannot fully resolve the 2D in plane magnetization directions, which is crucial to determine the chirality of given magnetic textures. Although time-resolved PEEM measurement may resolve in-plane magnetization configurations with time resolution as studied in [Science 304 (5669), 420-422], PEEM is surface-sensitive measurement scheme and therefore volume-averaged magnetic textures cannot be observed. (Also note that surface magnetic configuration may be different from inside main textures due to existing stray fields, as studied in Sci. Adv. 4, 7 (2018))

In this sense, ultrafast stroboscopic Lorentz TEM measurements presented in this manuscript take a significant advantage, as this method can fully resolve in-plane component of volume-averaged magntizations with superior spatial resolution. Moreover, recent experiments [Nat. Commun., 6, 8957 (2015); Sci. Rep. 8, 5703 (2018).] also revealed that off-focus or tilting-measurements using LTEM can observe Neel-type magnetic configurations, which hasn’t been readily observed at this Frensel-mode LTEM scheme. Therefore, I believe this work provides an important new magnetization dynamics measurement scheme using LTEM, which could be used in wider range of studies on magnetization configuration dynamics in MHz-GHz scale. Therefore, I support the publication of this work in Communications Physics with minor comments attached below.

1. Authors need to provide any magnetic information of their material (Hc, Ms, Hk etc). Although Permalloy is quite common in-plane magnetic material and all properties are well known, authors still need to provide their measured informations (on companion SiOx substrates maybe?) to confirm the magnetic properties. In particular, as authors present largely enhanced magnetization damping compared to known values, it may be beneficial to understand the observed phenomena.

2. Discussion on joule-heating need to be included. Considering that authors apply fairly large amount of current (~10^11 A/m2) for a long exposure time, the current-induced joule heating could be significant, and possibly play a role in magnetization dynamics. Although it would be desired to estimate the effective temperature rise using their materials, any relevant should be included to correctly understand the observed M dynamics.

Reviewer #2 (Remarks to the Author):

The manuscript “Few-nm tracking of magnetic vortex orbits and their decay with ultrafast Lorentz microscopy” deals with the application of Lorentz microscopy to study current-driven LLG type magnetization dynamics. The quality of the data is convincing and especially the aspect of tracking the decay of the oscillation after switching off the drive current is fascinating, as it reveals new information on the otherwise hard to access vortex pinning, which is made possible by the very high spatial resolution.

The manuscript is mostly well written and images are mostly clear, however there are a number of things that should be fixed.

Title: I would recommend including the term current-driven dynamics in some form, as just tracking an orbit seems to be too much centered on the method. The investigated physics is resolved on the time scale of 300 ps (which is fully appropriate for the system), so the term ultrafast is rather misleading, even though the method certainly is capable of ultrafast investigations.

Abstract: "temporal hardening of the free oscillation frequency" appears to be ill-phrased. I could not find meaningful application for "hardening of a frequency", nor can I make physical sense of it. A medium can be hardened, of course. In the manuscript this is repeated as "temporary" instead of "temporal", which is at least not giving the impression of "hardening in the time domain" which is even more confusing. Temporary (or transient) increase of frequency might do?

Page 2: I am aware that in previous literature c was sometimes called "chirality". However, this is very misleading. The chirality of a vortex is obviously given by the product (c p), and there are physical effects that directly depend on this chirality. c alone should be rather called "curl" or (sense of) rotation.

Figure 1: The ground symbol in the sketch should be drawn horizontally, following a bend, to be clearly identified. Defocus is misspelled.

Figure 2: a) What is the origin of the black areas to the right and left of the structure? Is this dark-field imaging, or is there some kind of mask applied on the image? In Ref 42. the membrane is brighter than the structure.

The black lines are explained; however, it is very difficult with all these artifacts to assess the quality of sample fabrication from the TEM images alone. It would be helpful to see an AFM or SEM image of the sample in the methods section that allowed to see possible large scale problems in fabrication and-even more important with respect to pinning-analyze the surface roughness of the Py film.

What is the origin of the observed vortex shift to the right? Is there a residual in-plane field? Reassembling should be resembling.

Page 5: It would be helpful to the reader to briefly comment on the connection between the observed 51 nm profile width and the actual/expected vortex width, even if there were none at all.

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Figure 3: It would increase the information content to see the last oscillation data before t=0 (especially in panel c), if it was recorded.

From the data, one may guess there is a more direct relation between the damping and the vortex velocity (easily calculated from diameter and frequency). Maybe this turns out to be more significant than the shown relation to the diameter alone?

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It would be nice to have some information on the roughness of the Py film in terms of RMS and correlation length, if available, to compare it with the observed density of pinning sites.

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A comment on the further information that could be gained from fitting theta and A should be made. Is it possible to learn something about non-adibaticity. What are the values resulting from the fit?

Reviewer #3 (Remarks to the Author):

Dear authors,

Congratulations to this very intriguing step into resolving magnetization dynamics with ultrafast TEM methods. Your work opens up vast perspectives for studying magnetization dynamics such as spin wave excitations, in particular those with short characteristic length scales, e.g. impacted by large exchange coupling. These studies are currently predominantly carried out with x-rays, which require large facilities and have limits when it comes to nanomagnetic structures. I therefore strongly recommend publication of this work in Communication Physics. Some smaller technical remarks / suggestions for the authors are given below.

Page 4: There could have been some more details on the dedicated RF holder. Although there are some references in the literature, it is not straight forward to built such a holder (e.g., special care must be taken for the cables, plugs, …). This becomes even more important when going for the above mentioned short wave lengths excitations with corresponding higher excitation energies.
Page 4: In the linear defocus regime the contrast linearly depends on the curl of the projected Induction (B-field) and not the gradient of the magnetization.
It could have been useful to also try a more quantitative evaluations (employing a TIE reconstruction of the measured contrast) and tracking this as a function of time. That might have delivered more insight into the vortex dynamics (e.g., amplitude modulations))
Page 6: There could have been a more comprehensive discussion of the magnetic properties revealed through the observed vortex dynamics (e.g., in terms of the shortcomings of the used Thiele equation based model (e.g., what approximations in terms of dipole field / anisotropies have been involved), what are global anharmonicities in the vortex potential?)
Page 11: in the equations describing the dynamics of the vortex core taking the real part is missing in the final step

With best regards, Axel Lubk

Response to Reviewers

We would like to thank all the Reviewers for the time spent with our work, for the helpful comments and questions, and for supporting publication in Communications Physics. The feedback provided to us has been very productive and helped us to revise and improve our manuscript.

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largely enhanced magnetization damping compared to known values, it may be beneficial to understand the observed phenomena.

We thank the Reviewer for these remarks and the comment. The evaporated base material is specified with a saturation magnetization of M_s = 613kA/m. Due to the evaporation, we expect slight change in alloy concentration [A]. Motivated by the Reviewer's comment, we conducted quantitative image simulation for the Fresnel Lorentz contrast of this sample. For the given imaging conditions in the experiments, the image contrast is reproduced for a magnetization around M_s = 600kA/m, in good agreement with the material specifications. We have added the image simulations to a Supplementary Note.
Beyond the present characterization, we plan to conduct an in-depth study of the influence of the structure and properties of the deposited materials on the vortex trajectories and damping in the future.

[A] Carson, K. R., & Rudee, M. L. (1970). Composition Gradients in Ni–Fe Alloy Films Produced by Vapor Deposition from a Tungsten Boat. Journal of Vacuum Science and Technology, 7(6), 573–576. https://doi.org/10.1116/1.1315879

2. Discussion on joule-heating need to be included. Considering that authors apply fairly large amount of current (~10^11 A/m2) for a long exposure time, the current-induced joule heating could be significant, and possibly play a role in magnetization dynamics. Although it would be desired to estimate the effective temperature rise using their materials, any relevant should be included to correctly understand the observed M dynamics.

We thank the Reviewer for this question. We address this point by comparing Lorentz images with and without current excitation. Direct access to current-induced changes of magnetization from Joule heating can be obtained by comparing the intensity of the vortex feature above the background in these Lorentz micrographs. Comparing Fig. 4.a with the micrograph at t=0 in Fig. 2a yields the same contrast, within a 10% margin of error. Therefore, we do not observe a significant change in magnetization. The same holds for the results obtained in Fig. 3, as the average power was below that used for the continuous excitation. We have added these points to the Methods section of the revised manuscript.

We thank the Reviewer again for the comments.

Reviewer #2 (Remarks to the Author):

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We followed the suggestion of the Referee and altered the title to add the phrase "current-driven". Regarding the relevant timescale and the use of ultrafast, we generally agree that this term should be reserved for studies in the picosecond range. In order to achieve the degree of accuracy demonstrated in Fig. 2, a drift-free synchronization well below 100ps is required. Combined with the fact that the technique in our microscope can readily be equipped with high GHz or even THz driving, we would prefer to keep the term "ultrafast" in the present title.

Abstract: "temporal hardening of the free oscillation frequency" appears to be ill-phrased. I could not find meaningful application for "hardening of a frequency", nor can I make physical sense of it. A medium can be hardened, of course. In the manuscript this is repeated as "temporary" instead of "temporal", which is at least not giving the impression of "hardening in the time domain" which is even more confusing. Temporary (or transient) increase of frequency might do?

We thank the Referee for pointing out that the terminology may lead to misunderstandings. We had adopted this formulation from the context of nonlinear oscillators to describe the transient change towards higher frequencies.
We followed the Referee's suggestion and revised the text accordingly.

Page 2: I am aware that in previous literature c was sometimes called "chirality". However, this is very misleading. The chirality of a vortex is obviously given by the product (c p), and there are physical effects that directly depend on this chirality. c alone should be rather called "curl" or (sense of) rotation.

We agree with the Referee and adopted the proposed terminology.

Figure 1: The ground symbol in the sketch should be drawn horizontally, following a bend, to be clearly identified. Defocus is misspelled.

We changed the position of the ground symbol accordingly and corrected the misspelling.

Figure 2: a) What is the origin of the black areas to the right and left of the structure? Is this dark-field imaging, or is there some kind of mask applied on the image? In Ref 42. the membrane is brighter than the structure.

The dark areas on the left and right of the structure are due to the opaque gold contacts. A sentence was added to the manuscript to clarify that. Please note that the structures in Ref. 42 are not electrically contacted.

The black lines are explained; however, it is very difficult with all these artifacts to assess the quality of sample fabrication from the TEM images alone. It would be helpful to see an AFM or SEM image of the sample in the methods section that allowed to see possible large scale problems in fabrication and-even more important with respect to pinning-analyze the surface roughness of the Py film.

We agree with the Referee that fabrication errors have to be considered in the context of our study. Unfortunately, we do not have an SEM image of the sample used. However, in our experience, Lorentz imaging presents a very reliable and (compared to SEM) superior approach to characterize sample quality. Specifically, we can rule out fabrication errors (e.g., residual magnetic material on top of the permalloy square, after a failed lift-off), as they would easily be spotted as strong phase objects. We have added this comment in the Methods section of the manuscript.

What is the origin of the observed vortex shift to the right? Is there a residual in-plane field? Reassembling should be resembling.

Yes, there is a residual magnetic field from the current-free objective lens, which we believe causes the small shift. This is now clarified in the article.

Page 5: It would be helpful to the reader to briefly comment on the connection between the observed 51 nm profile width and the actual/expected vortex width, even if there were none at all.

Following the Reviewer's suggestion, we added a comment to the manuscript.

Page 6: (and methods) I do not see, why the deflection of a parallel beam by the electrical field between to electrodes should be called “small-angle scattering”. To my understanding the beam is not scattered by the electrodes but homogeneously deflected.

We had used the term according to Ref. 33. However, we agree that deflection is perhaps more suitable and have changed the wording.

Figure 3: It would increase the information content to see the last oscillation data before t=0 (especially in panel c), if it was recorded.

Unfortunately, this data was not recorded.

From the data, one may guess there is a more direct relation between the damping and the vortex velocity (easily calculated from diameter and frequency). Maybe this turns out to be more significant than the shown relation to the diameter alone?

We agree with the Reviewer that the velocity may be a more natural quantity to be related to the damping. In our case, radius and velocity are very close to proportional to each other, such that there is little change to the curve shown. Nonetheless, we have decided to follow the Reviewer's suggestion and have replaced the corresponding panel.

Page 8: temporary hardening (see above)

We changed "temporary hardening" to "transient rise" (see comment above).

It would be nice to have some information on the roughness of the Py film in terms of RMS and correlation length, if available, to compare it with the observed density of pinning sites.

Data on the surface roughness is unfortunately not available. We hope to be able to include this information on related samples in future studies.

Page 9: State the electron energy used in TEM and the strength of the magnetic field in vertical direction at the sample position. The latter will affect the vortex size, depending on p, and thus also the damping.

We thank the Referee for this comment. We used an acceleration voltage of 120kV. The residual out-of-plane magnetic field has a strength of about 10mT. Both values are now stated in the Methods section of the manuscript.

Figure 4: a) Would a continuous illumination not rather yield a bright circle as in Ref 32? Or is this recorded also without drive current?

Yes, an image under continuous illumination and excitation would yield a circle. We chose to present an image under continuous illumination without any excitation, allowing the reader to compare the quality of a micrograph acquired with a pulsed and a conventional source. This is now clarified in the text.

b) Is the 100 nm base line real or caused by the resolution of the driven vortex?

The 100 nm baseline is real, similar to the data in Ref. 32. We added a sentence to the manuscript, explaining that the vortex remained stationary beyond the depicted frequency interval.

Page 10: electron scattering, see above. Cold should be gold.

The phrase "small-angle electron scattering" was replaced with "long-camera-length diffraction mode" and the spelling mistake was fixed.

Page 11: Eq. (1) and (2) a squared is missing at the bracket in the denominator of the pre-factor.

We thank the Reviewer for pointing out the error in the equation. It has been fixed in the manuscript.

A comment on the further information that could be gained from fitting theta and A should be made. Is it possible to learn something about non-adibaticity. What are the values resulting from the fit?

Knowing only theta and A is not enough to learn something about the non-adiabaticity. The matrix in Eq. (1) contains three unknown parameters, namely j, H and Xi, yielding an underdetermined system of equations. Beyond that, the fitting results of the free oscillation frequency and the damping parameter already suggest a shortcoming of the model with respect to our measurements. Therefore, we have decided against attempting a further quantitative evaluation - such as the determination of the non-adiabaticity parameter - as it would most likely not yield a robust result.

*We would like to thank the Reviewer again for these very helpful comment*s.

Reviewer #3 (Remarks to the Author):

Dear authors,

Congratulations to this very intriguing step into resolving magnetization dynamics with ultrafast TEM methods. Your work opens up vast perspectives for studying magnetization dynamics such as spin wave excitations, in particular those with short characteristic length scales, e.g. impacted by large exchange coupling. These studies are currently predominantly carried out with x-rays, which require large facilities and have limits when it comes to nanomagnetic structures. I therefore strongly recommend publication of this work in Communication Physics. Some smaller technical remarks / suggestions for the authors are given below.

Dear Dr. Lubk, We would like to thank you for these very positive and encouraging remarks, and for suggesting publication in Comms. Phys..

Page 4: There could have been some more details on the dedicated RF holder. Although there are some references in the literature, it is not straight forward to built such a holder (e.g., special care must be taken for the cables, plugs, …). This becomes even more important when going for the above mentioned short wave lengths excitations with corresponding higher excitation energies.

We thank the Reviewer for this remark. Some further details on the self-constructed sample holder have been added in the methods section.

Page 4: In the linear defocus regime the contrast linearly depends on the curl of the projected Induction (B-field) and not the gradient of the magnetization.

We agree with the Reviewer and have changed the wording.

It could have been useful to also try a more quantitative evaluations (employing a TIE reconstruction of the measured contrast) and tracking this as a function of time. That might have delivered more insight into the vortex dynamics (e.g., amplitude modulations))

We agree with the Reviewer that a more detailed quantitative analysis of the magnetic structures via a reconstruction generally enables additional insight. For the given magnetization state, due to the intensity enhancement at the vortex center, an analysis of the direct image contrast yields superior localization accuracy. Upon further developing the methodology and improving the overall signal-to-noise ratio, we plan to also employ image reconstructions. We have added a mention of the possibility to conduct image reconstructions in the future.

Page 6: There could have been a more comprehensive discussion of the magnetic properties revealed through the observed vortex dynamics (e.g., in terms of the shortcomings of the used Thiele equation based model (e.g., what approximations in terms of dipole field / anisotropies have been involved), *[...]*

Again, we fully agree with the Reviewer. We found relating the observed deviations from predictions of the Thiele equation directly to a specific approximation difficult. As stated later in the manuscript (page 8), it is our opinion that local disorder is amongst the most prominent aspects to be considered.
At this point, we hope to present accurate experimental observations which may stimulate others - including theoretical groups - to revisit the problem.
In addition, we plan to gain better experimental insights into the system in the future by combining a structural analysis with our measurements.

[...] what are global anharmonicities in the vortex potential?)

We thank the Referee for pointing out the ambiguity of the expression “global anharmonicities”. With this term, we intended to refer to overall size- or shape-induced deviations from a parabolic vortex potential, as opposed to local disorder. The given references contain several of these examples. We have added an example from the reference list to the text.

Page 11: in the equations describing the dynamics of the vortex core taking the real part is missing in the final step.

We have added taking the real part.

With best regards, Axel Lubk

Thank you again for the insightful comments and the constructive criticism!