Peer Review File

Manuscript Title: Free-space dissemination of time and frequency with 10^-19 instability over 113 km

Reviewer Comments & Author Rebuttals

Reviewer Reports on the Initial Version:

Referees' comments:

Referee #1 (Remarks to the Author):

The manuscript describes comb-based two-way frequency transfer over a 113-km free space link . This is the longest distance yet realized for transmission of stable frequency comb light over the air by a significant factor of 113km/14km=8x in distance, comparing this work to Ref. 21. Because the link loss increases at least quadratically with link loss, the system required much higher comb powers and the authors have used 1-W frequency combs combined with a large aperture telescope. The construction of such high power, but stable combs, is a significant challenge and achievement. The performance shown here is comparable to similar comb-based transfer – the short time stability is degraded but the tight temperature control allows for floor at a few 10^-19 level. This work significantly extends the distance and demonstrates comb-based frequency transfer is compatible with high-power frequency combs. However, several of the other statements of the paper are not as strongly supported and I think there is a fundamental issue with the configuration that impacts the interpretation of the instability and accuracy results.

The paper makes a strong distinction with previous work including the statement that all previous work used mirror-folded links, rather than the two independent links used here. These statements are problematic for two reasons. First, ref. 21 did not use a mirror-folded link, nor did the paper pair of Beloy et al., Nature, 591, 564(2021), and Bodine, et al, PR Research, 2, 033395 (2020). Ref. 21 used two fully independent parallel links (i.e. four terminals and displaced free-space links) over truly independent 14 km paths.

The second problem is more fundamental. The two links here are identical and not independent. The 1540/1570 light is wavelength-multiplexed to travel through the same terminals and air path, i.e. across the exact same spatial single mode. Therefore, the two propagating comb pulses at 1540 nm and 1570 nm suffer almost the exact same time-of-flight variations (to within the slight difference in air index). Therefore, any non-reciprocal path length fluctuations will drop out of their final difference comparison. As configured, the system cannot test if there are penalties associated with comb-based time transfer over the long 113-km distances. In other words, a simple direct subtraction between the one-way timing on the two systems likely yields similar Allan deviations. (In contrast, a mirror-folded link will double the turbulence and is a actually more stringent test.) The instability and 10^-19 accuracy numbers verify the relative frequency locks between the four combs, rather than the two-way suppression of time-of-flight noise. It is impressive that the residual noise on the high power combs reached these levels. However, an independent test of two time-transfer systems would require independent pairs of telescopes and offset free-space links, as in Ref. 21. This would copy, for example, the fiber transfer work of Ref. 18 that used two different fiber paths.

This common link also complicates the interpretation of the performance at low valid data rates. At high link loss, the signals drop below threshold for varying amounts of time. Here, these signal fluctuations will be completely correlated on both links so any systematics or sensitivity to low data rates (or am to pm) is suppressed. With such correlated links, one might expect the two systems to agree down to a few cps since they sample at almost the same time. On the other hand, with independent links, one would need to maintain a certain data rate just to insure overlapping data in time to evaluate the relative clock frequencies.

Regarding novelty, the abstract and introduction could clarify that the innovation here is the incorporation of the high power combs – which is a significant challenge- but the concept of the two-way approach, linear optical sampling, implementation, operational parameters, and processing directly follow earlier work, including refs. 8,9. Additionally, more recent work from the group of Ref. 8,9 is omitted including the paired Beloy et al and Bodine et al, mentioned above, which describe comb-based frequency comparison between full optical atomic clocks over point-to-point link as well as a demonstration of carrier-phase operation (PRL, 120. 050801 (2018)) and velocity-compatible operation (Nat. Comm., 10, 1819 (2019)), which used a similar polarization multiplexed link but off a retroreflector. Some of this more recent work used sophisticated real-time processing, synchronization, and velocity-correction techniques that are not incorporated here. It would be interesting for the authors to comment on whether these would be also supportable with the amplified combs and at what link loss.

Additional technical questions:

The paper states that the use of orthogonal polarization can mimic the point ahead angle encountered in satellite missions in lines 116-118 and elsewhere. I do not understand this connection? I note that this issue has been explored in both theory and experiment in work unreferenced by this manuscript (see for instance the initial theoretical work of Robert et al, PRA 93 2016), and shown to not be problematic.

Can they provide more information on how the data are processed and combined, including time traces? The assumption is the Adevs are from the "difference" between the two systems, which is relevant to discussions of precision and accuracy. Given the above comment on the common-mode path noise, all pairwise combinations of the time traces would be of interest. How are gaps in the data treated at low valid data rates? Given the comment above on the commonality of signal fluctuations, can they provide a more complete analysis of the expected ADevs at high link loss (low data rates)?

What is the detection power threshold (only mean values are given)?

The explanation of the 1-s Allan deviation, which is higher than some other comb-based transfer, could be clarified. Is it the combs themselves (from Figure 13) or the link? ? The sentence gives 40/sqrt(tau x Sn) - Based on a parenthetical comment elsewhere, Sn might be the data rate but the scaling with "tau" does not appear correct for the MDEV? In addition to comparing to other microwave systems, could they compare to other comb-based transfer demonstrations? If there is a small “penalty” from the use of amplified combs, that is a reasonable price for long distance operation.

How is the 4-5 nanosecond ambiguity in timing removed if the GPS is only providing 30 ns relative synchronicity?

It is impressive the system work with a total loss of 89 dB. Where is the 1W is measured and what is the actual comb transmit power out of the aperture? Could the main text mention between which points the 89 dB link loss is measured and what was the aperture-to-aperture loss?

Regarding future ground-to-satellite links, the size/weight and power of this system is large due to the amplified combs and large aperture telescope. Can they comment on what is needed for future space payloads?

Could they elaborate on the derivation of the 10-as number and the assumptions there? Is there a typo in low-pass vs high-pass filtering here?

With regard to Figure 3, the text states there is no obvious correlation with received power since both curves start at 40 fs, but they start at different averaging times – is that not a consequence of lower power and less valid data?

In Figure 11, is 1.414 ps the transform-limited pulse width limit for the spectra?

Figures 1c was challenging to decipher. Figure 9 helps but could benefit from a caption. It might help to define the wavelengths/purpose of the dichroics? Should the “COMS” labels be CMOS and is this meant to indicate a focal plane array? What is the purpose of the APD in Figure 1c? There seem to be other unused laser wavelengths coupling to the terminals?

Could more details be provided on the adaptive optics system, e.g. operation, beacon power, performance, benefit over tip/tilt etc?

The analysis of the link loss is very complete, showing good agreement at a specific cn2. What range of CN2 were observed? The link is fairly close to the ground and presumably highly turbulent, which make the 113-km demonstration all that much more impressive.

Referee #2 (Remarks to the Author):

Review of the manuscript entitled ‘113 km Free-Space Time-Frequency Dissemination at the 19th Decimal Instability’,
by Qi Shen, Jian-Yu Guan, Ji-Gang Ren, Ting Zeng, Lei Hou, Min Li, Yuan, Cao, Jin-Jian Han, Meng-Zhe Lian, Yan-Wei Chen, Xin-Xin Peng, Shao-Mao Wang, Dan-Yang Zhu, Xi-Ping Shi, Zheng-Guo Wang, Ye Li, Wei-Yue Liu, Ge-Sheng Pan, Yong Wang, Zhao-Hui Li, Jin-Cai Wu, Yan-Yan Zhang, Fa-Xi Chen,

Chao-Yang Lu, Sheng-Kai Liao, Juan Yin, Jian-Jun Jia, Cheng-Zhi Peng, Hai-Feng Jiang, Qiang Zhang, Jian-Wei Pan.

The manuscript describes a major breakthrough into the realization of free-space satellite-based long range optical time and frequency dissemination, with impacts for fundamental physics (search for dark matter, test of fundamental constant, test of relativity…). The major achievement of the authors as compare to previous work in the field is the operation of two 1W-power Optical Frequency Comb, that allows the authors to demonstrate world-record in range for free-space optical fiber links, while keeping state of the art in terms of precision. The experimental results are impressive, and the manuscript is very likely to be highly impacting, what desserves publication in Nature.

The manuscript describes other several improvements as compare to their previous paper published in 2021: authors described an improved and original electronics system and an improved interferometer system (length mismatch minimisation, better temperature stabilization) that allows them to reach lower noise floor stabilities, and larger link loss acceptance from 72 dB up to 90 dB, what is another impressive achievement. The authors reports for about 235 hours of operation acquired in 21 days, what is also an impressive achievement given the high loss acceptance of their system.

The manuscript is well written, expose clear explanations. Data and methodology are valid, the authors show high quality data, and appropriate treatment of uncertainties. The manuscript shows rigourous reasoning and robust results, certainly reliable and valid.

Figures are well presented generally speaking, aside of small mistakes I detailed later on in this review. The supplementary materials are technically sound and are very complete. Supplementary materials should provide expert readers with the necessary technical informations they might need, while the main text indeed address a broad audience.

The manuscript has however several weaknesses that required a revision before acceptance.

Major comments, questions and suggestions to the authors:
- The authors claim line 31 (abstract) and line 81 that they established two independent two-way time and frequency links. May be the wording should be slighty softened or put better in context. In my view, the authors are partly right in their statements as they used two independent opto-electronics systems, but the telescopes are still in common mode. So the two links are not totally independent. The authors do not show resiliency with respect to different paths or demonstrate the immunity versus mechanical noise and thermal noise affecting the telescope. To the best of my understanding, in a free space configuration, there should be three telescopes used to realize a ‘lambda-shape’ or a ‘V-shape’, connecting one ground to two satellites, or one satellite to two ground stations.

Line 46-48: The authors state ‘So far, optical carrier phase transfer through fiber links with two-way transfer operation has reached over 1,000 km with a stability of 10−19 at 100 s [17, 18]. However, such a method is incompatible with the existing telecommunication fibers

The sentence is partly true according to citation, but, since 2012-2013, major advances happen with fiber links. One should comment first that these fiber links experiments are using active compensation, and indeed are incompatible with existing telecommunication fibers. But nowadays, fiber links reach several thousand of km with uncertainties <1e-19, and integration time over several months, over telecommunication network. See for instance Cantin et al., , « An accurate and robust metrological network for coherent optical frequency dissemination », New J. Phys., vol. 23, n° 5, p. 053027 (2021), doi: 10.1088/1367-2630/abe79e.
Second, most of the work accomplished with long haul fiber links are using active compensation and not a two-way schemes, and therefore I believe the sentence give a wrong picture to non expert readers.
I suggest to rephrased these sentences, and provide also there a citation to [34], that describe also the fiber connection from NPL to PTB through the REFIMEVE network, that spans 2200 km in total, and shows days of data.

This being said, it does not weaken the second part of the argument, about mountains, marine, intercontinental and space links, that I believe to be true.

Line 121-123 : It seems to me that the adaptative optic is one of the key ingredient of the system. I did not find in the manuscript the bandwidth of the correction applied by the adaptative optics ? It would be helpful too to document the dynamic of the correction.

The authors reach higher power optical frequency comb using two stages EDFAs. Is there any information on the additional phase noise due to the amplification ?

Line 98 : 'the length of mismatch is optimized to be below 5 cm': how is the path imbalance measured ? Is the temperature fluctuations of 7 mK arise from an in-loop measurement or out-of-loop ?

Figure 2c : x-label says Time in (s), but displayed ticks are 17:09 to 20:09. According to the corresponding temperature variations, I have severe doubts that this is seconds. Please correct the x-label.
It would be also much more convenient to show the y-axis in ns, instead of a scaling factor of 1e5 with units in fs.

Line 145-147: On this same plot, authors claims that the free link delay fluctuations are due to temperature, but the correlation does not seem that high. Can the authors provide the level of correlation ? Can wind turbulences and/or humidity variations can alter the propagation delay ? Furthermore, it seems that there is a delay between the temperature variation and the propagation delay variation ? Is it due to the thermal conductivity between the air and the experimental system laying in the laboratories ? Are the temperature recorded at the two laboratories sites ? Is there any weather station data available ?

One main claim of the paper, and related to its potential impact, is that this 113-km link mimics the attenuation of much longer links, to LEO, MEO and GEO orbits. However the two laboratories are static, and a satellite is moving… So I missed a discussion on a moving target. It will be an improvement of the manuscript if the authors can describe how movements of satellites will or will not alter their results, in stability and accuracy (see also comment on line 116-118).

In the same line of idea, I believe the manuscript can be improved if authors draws some perspective on how to solve issues that may arise from a higher probability of multi path when extending their link to several thousands of kilometers.

Minor comments:
Line 52: the sentence is not that much explicit. It would be helpful for non expert readers to explicit why these other experiments are not matching the requirements.

Line 57-58: The sentence is not that clear, as it compares CW in fiber with a situation in air, where there is drop-outs. I suggest to rephrase.

Line 64 : 'Further, we employ linear optical sampling that offers femtosecond accuracy over the whole ambiguous range'. Citation to [22] there would be helpful.

Line 116-118 : Authors states 'it can also simulate the nonreciprocal situation in satellite ground links due to the relative angular motion between a satellite and its ground station'. I did not understand this point.

Fig.3 : It would be better to extend the frame of figures 3A and 3b, so that the last point of the curves can be easily seen.

Bibliography:
Line 44: there was search for dark matter with optical fiber network of optical clocks. I suggest to add the following reference
B. M. Roberts et al., « Search for transient variations of the fine structure constant and dark matter using fiber-linked optical atomic clocks », New J. Phys., vol. 22, nº 9, p. 093010, sept. 2020, doi: 10.1088/1367-2630/abaace.

Typos:
line 81: free-spece > free-space
line 94: fluctuaitons > fluctuations
line 134: tolarence > tolerance
line 144: flucatuation > flutcuations
line 202: apmlifiers > amplifiers
line 237: avarage >average
line 241: distritbution >distribution
line 245: 'and the local OFC power at the LOS is kept same' > and the local OFC power at the LOS is kept the same line 246: obtianed > obtained Line 355-356: The sentence looks incomplete or incorrect grammatically.

Responses to the Reviewer Comments

Referee #1 (Remarks to the Author):

Comment 0:

The manuscript describes comb-based two-way frequency transfer over a 113-km free space link. This is the longest distance yet realized for transmission of stable frequency comb light over the air by a significant factor of 113km/14km=8x in distance, comparing this work to Ref. 21. Because the link loss increases at least quadratically with link loss, the system required much higher comb powers and the authors have used 1-W frequency combs combined with a large aperture telescope. The construction of such high power, but stable combs, is a significant challenge and achievement. The performance shown here is comparable to similar comb-based transfer – the short time stability is degraded but the tight temperature control allows for floor at a few 10^-19 level. This work significantly extends the distance and demonstrates comb-based frequency transfer is compatible with high-power frequency combs. However, several of the other statements of the paper are not as strongly supported and I think there is a fundamental issue with the configuration that impacts the interpretation of the instability and accuracy results.

Reply:

We thank the referee for his/her time and efforts to review our manuscript. We are glad to see that the referee comment our work as “the longest distance yet realized for transmission of stable frequency comb light over the air by a significant factor of 113km/14km=8x in distance”.

We have addressed the referee’s concerns by implementing new experiments and substantially revising our manuscript. We hope that the referee will be satisfied with the revised version. Below we replied to the comments point by point.

Comment 1:

The paper makes a strong distinction with previous work including the statement that all previous work used mirror-folded links, rather than the two independent links used here. These statements are problematic for two reasons. First, ref. 21 did not use a mirror-folded link, nor did the paper pair of Beloy et al., Nature, 591, 564(2021), and Bodine, et al, PR Research, 2, 033395 (2020). Ref. 21 used two fully independent parallel links (i.e. four terminals and displaced free-space links) over truly independent 14 km paths.

Reply:

We appreciate the referee for reminding us of recent progresses. Indeed, the work pointed out by the referee used two fully independent parallel links over 14 km paths. We have revised our manuscript accordingly.

Revisions made in manuscript:
In the main text,

1. In line 26 of Page 2, this sentence in Abstract, "However, such attempts were limited to dozens of kilometers in mirror-folded configuration." has been modified to: "However, such attempts were limited to dozens of kilometers."
2. In line 52 of Page 3, the sentence of "...previous work have achieved a distance up to a dozen kilometers in a mirror-folded configuration,..." has been changed to:
   "...Previous work, however can only reach a distance up to a dozen kilometers, which limits its applications in many long distance scenario..."
3. The sentence of "Instead of keeping two terminals close and folding links with a remote flat mirror in the previous experiments, we physically separate the two terminals to a distance of 113 km." has been deleted.
4. The paper pair of Beloy et al., Nature, 591, 564(2021), and Bodine, et al, PR Research, 2, 033395 (2020) have been added in the cited list as Ref. 26 and 25.

Comment 2:

The second problem is more fundamental. The two links here are identical and not independent. The 1540/1570 light is wavelength-multiplexed to travel through the same terminals and air path, i.e. across the exact same spatial single mode. Therefore, the two propagating comb pulses at 1540 nm and 1570 nm suffer almost the exact same time-of-flight variations (to within the slight difference in air index). Therefore, any non-reciprocal path length fluctuations will drop out of their final difference comparison. As configured, the system cannot test if there are penalties associated with comb-based time transfer over the long 113-km distances. In other words, a simple direct subtraction between the one-way timing on the two systems likely yields similar Allan deviations. (In contrast, a mirror-folded link will double the turbulence and is a actually more stringent test.) The instability and 10^-19 accuracy numbers verify the relative frequency locks between the four combs, rather than the two-way suppression of time-of-flight noise. It is impressive that the residual noise on the high power combs reached these levels. However, an independent test of two time-transfer systems would require independent pairs of telescopes and offset free-space links, as in Ref. 21. This would copy, for example, the fiber transfer work of Ref. 18 that used two different fiber paths.

Reply:

We appreciate the referee for this valuable comment which definitely help us to improve our work. We fully agree with the referee that the two links, 1545/1563 share the same terminals and air path, which might result in a smaller Allan deviation. Actually, we became aware of this issue soon after the submission of this manuscript. So we have established an independent fiber link connecting two remote locations, and utilized the fiber link to evaluate the two 113 km free space links. The MDEV is below

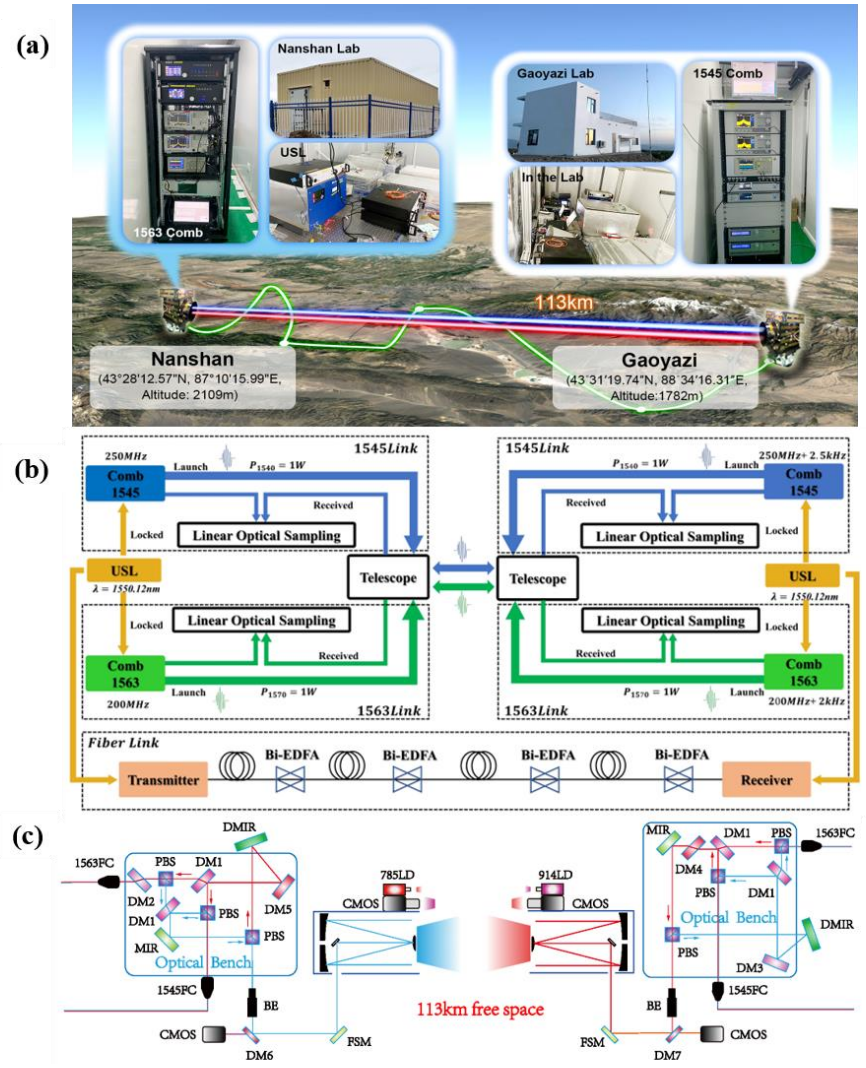
4E-19 at 10,000 seconds at such configurations. The result is slightly higher than our previous result, which might be attributable to the common noise. Nevertheless, it doesn’t affect our claim that our system can distribute time-frequency signal over 113 km free-space link at the 19th decimal instability, which firmly constitutes a critical step towards future satellite time-frequency dissemination. We have added the new experimental results in the revised version.

Revisions made in manuscript:

In the main text,

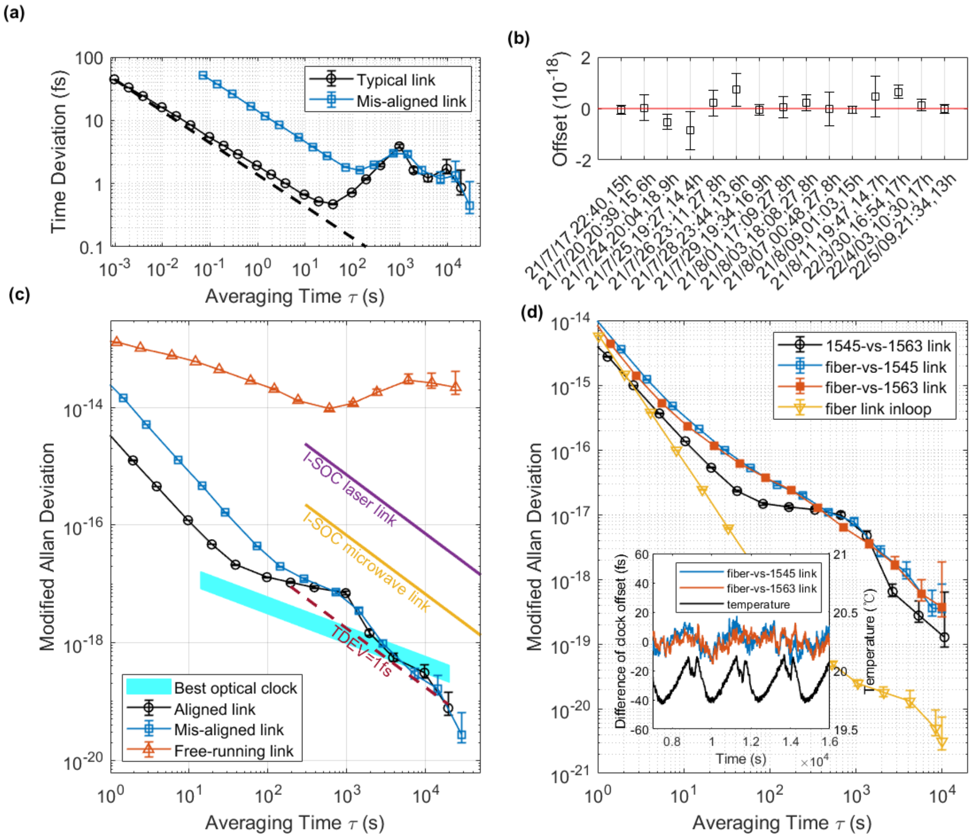
1. In Page 34, the Fig.1a and Fig.1b have been redrawn to add the fiber time-frequency transfer link.

   The new fig.1 (here is R-Fig. 1) is as follows:



R-Fig. 1 The experimental setup. (a). Overview of the 113 km free-space time-frequency dissemination experiment. (b). The main diagram of the system, which contains a 1545 nm free-space link and a 1563 nm free-space link and a fiber link. USL: ultra stable laser. (c). The optical layout of the optical transceiver telescope, where different colors of optical paths indicate different polarizations. PBS: polarizing beam splitter; 1545FC (1563FC): fiber collimator for two-way 1545 (1563) nm comb transceiver; DM: dichroic beam splitter; MIR: mirror; DMIR: deformable mirror; WDM: wavelength division multiplexer; BE: beam expander; FSM: fast steering mirror; CMOS: complementary metal–oxide semiconductor.

2. In Page 36, the Fig.3d have been added about comparisons among the three links of the fiber link and two free space links. The Fig.3b has been redrawn to add three more data sets acquired in 2022.
   The new Fig.3 (here is R-Fig. 2) is as follows:



R-Fig. 2 Experimental results of time-frequency transfer. Captions for (d) is: “The comparison among fiber link and two free-space links. The black circle line: two free-space links; The blue square line: fiber and 1545 comb link; The orange solid square line: fiber and 1563 comb link; The yellow triangle line: in-loop stability of fiber link. The inset shows the trend for the difference between fiber link and free-space links with temperature.”

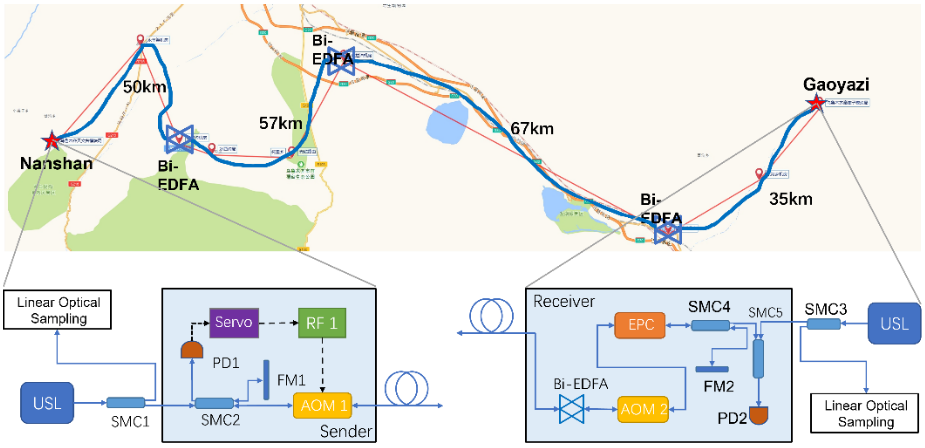
3. In lines 77-80 of Page 4, the sentences have been added to descript the motivation and setup of the fiber link as below:
   “Since the two multiplexing channels share the same free-space link, there exits common mode noise. In order to better evaluate our system, we also established an

independent fiber link connecting the two terminals with a distance of 209 km. All links (1545 nm free-space, 1563 nm free-space and the fiber link) share the same USL at each terminal."

4. In lines 186-194 of Page 8 in the Main text, the paragraph has been added:
   "Relative instability among two free-space links and the fiber link are in the range of 1-4E-19 at 10,000 s as shown in Fig. 3d. Thermal drifts effect (see the inset) limits the link performance for long terms. Although in-loop instability of the fiber link is negligible, there is 0.5 m out-of-loop fiber used to connect the fiber link at both terminals respectively. Instability of two space link is slightly better because equipment location of two free-space links are closer than that of the fiber link. In fact, the phase noise of atmosphere is much less than that of the fiber link and has been efficiently suppressed by two-way operation (See Fig.2 d). So, the thermal drift effect should be dedicated to the fiber parts."
5. In line 29-31 of Page 2 in the abstract, the sentence of
   "we demonstrate free-space time-frequency dissemination over two independent links with femtosecond time deviation, 3E−19 at 10,000 s residual instability and 1.6E−20 ±4.3E−19 offset."
   has been modified to
   "we demonstrate 113-km free-space time-frequency dissemination with 6.5E-20±3.5E-19 offset, and an instability of below 4E-19 at 10,000 s measured by comparison of two wavelength division multiplexing links and an independent fiber link"
6. In lines 153-155 of Page 7, the sentence of
   "…,we carried out a time-frequency transfer experiment over approximately 3 weeks. We obtained a total of 12 data sets, and the continuous acquisition duration of each data set was between 13.6 and 27.8 hours."
   is modified to:
   "…,we carried out a time-frequency transfer experiment, and obtained a total of 15 data sets. And the continuous acquisition duration of each data set was between 13 and 27.8 hours."
7. The sentence of
   "…, according to 12 times experimental results." has been deleted.

In the Methods text，

1. In Page 22-24, the details about the fiber time-frequency transfer link is added as a new section: " fiber time-frequency transfer link" as follows:
   "
   The total fiber length is 209~km, and the loss is 65~dB. To compensate for the transmitted loss, three bidirectional amplifiers are put on the path and one is put at terminal B. The detailed config is shown in R-Fig. 3.



R-Fig. 3 Setup for fiber time-frequency transfer. USL: Ultra-stable laser. SMC: Single-mode coupler. PD: Photon diode. RF: Radio frequency source. FM: Faraday mirror. AOM: Acoustic optical modulator. EPC: electric polarization controller. Bi-EDFA: Bidirectional erbium doped fiber amplification.

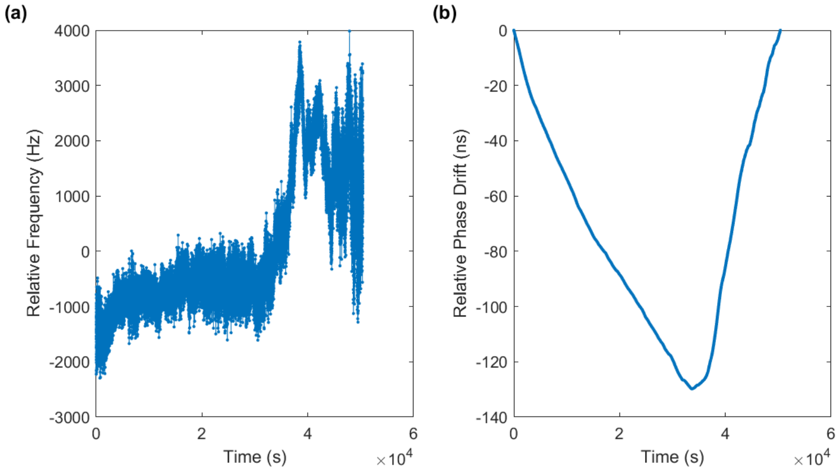
At terminal A, we split the USL into two parts. One is used to lock the combs, the other is going through an optical module for fiber time-frequency transfer. The module is actually a highly unbalanced Michelson interferometer, with the short arm in the local optical module and the long arm up to terminal B. The AOM at terminal A is used to compensate for the noise in the fiber link, and the AOM at terminal B is used to distinguish the reflected laser from terminal B with the backward scattering signal during the link. The reference for locking electrics is synchronized with the repetition rate of the comb, to ensure that the fiber time-frequency transfer system shared the same reference frequency with the free-space system. At terminal B, the local USL beats with the signal laser. The beating signal is filtered with a tracking oscillator and measured with a K-K phase recorder. A Direct Digital Synthesis (DDS) converts the repetition rate of 1563 nm OFC to 10~MHz to drive the phase recorder.

In the free-space time-frequency transfer link, the clock offset is calculated with the time base that came from the 1545 OFC. The phase difference of the fiber link should be converted to this time base too before we compare it with the free-space link. At terminal A, the ultra-stable laser and the locking electrics are all synced with the time base, so the frequency of the signal transmitted to terminal B can be calculated. At terminal B, we also record the ratio between the frequency of the ultra-stable laser and the 10~MHz as k. When the K-K phase recorder reads the frequency as $\mathrm{f_{beat}}$, the real frequency at the time base of the terminal A should be $\frac{f_{\mathrm{cw,B}}}{k} * \frac{\mathrm{f_{beat}}}{10\ MHz}$. We then have

$$f_{\mathrm{cw,B}} + \frac{f_{\mathrm{cw,B}}}{k} * \frac{\mathrm{f_{beat}}}{10\ MHz} = f_{\mathrm{signal}}.$$

The value of $f_{\mathrm{cw,A}} - f_{\mathrm{cw,B}}$ can be easily calculate from above formula. To convert the frequency to phase/time, one needs to be aware that reference frequencies of K-K phase recorder at different terminal are not same. Another ratio is needed to correct the gate time. The ratio can also be figured out from $f_{\mathrm{cw,B}}$ and $f_{\mathrm{cw,A}}$. Finally, the offset between the start recording time of the free-space link and the fiber link can be determined by minimizing the phase drift of the comparison result. Most of our fiber links are aerial optical fiber. There are several breakpoints in fiber link data while optical cycle slips happen. For each breakpoint, we remove 50 seconds of data before and after this point.

The comparisons among the fiber link and two free-space links are measured. The stability of the fiber-vs-1545 link is 3.5E-19@10000s and the stability of the fiber-vs-1563 link is 3.7E-19@10000s. The stability of the 1545-vs-1563 link is 1.3E-19@10000s. There is some common-mode noise effect between the 1563 and 1545 link and its value should be below 4E-19 at 10,000 s, which is mainly attributed to the thermal drift of fibers since the out-of-loop connecting fiber link is located at outside of the original thermal shield box. This result shows the stability of difference of fully independent links can reach the 4E-19 level. In fact, since the phase noise of the atmosphere caused by the piston effect and temperature drift mainly concentrates on low frequencies below 10 Hz, it has been efficiently suppressed by two-way operation. Indeed, the noise of the aerial optical fiber (shown in R-Fig. 4.) is much larger than the noise of the atmosphere; while the residual noise contribution is negligible (see fiber link in-loop).



R-Fig. 4 (a) The frequency of driver signal for AOM~1 relative to its center value. The AOM~1 is used to compensate for the Doppler noise in the fiber link. So the changing of the frequency implies the changing of doppler noise. (b) The accumulated phase change in the link. This is integral for the frequency data at the subfigure (a).
”

Comment 3:

This common link also complicates the interpretation of the performance at low valid data rates. At high link loss, the signals drop below threshold for varying amounts of time. Here, these signal fluctuations will be completely correlated on both links so any systematics or sensitivity to low data rates (or am to pm) is suppressed. With such correlated links, one might expect the two systems to agree down to a few cps since they sample at almost the same time. On the other hand, with independent links, one would need to maintain a certain data rate just to insure overlapping data in time to evaluate the relative clock frequencies.

Reply:

We agree with the referee's statement that we should address the measured link stabilities is reliable at high-loss condition, because the obtained data existing time coherence.

The extremely high loss experiment is to show what data rate we can obtain, which it is useful for guiding followers to design parameters of the OFC power, detection sensitivity and channel loss.

It is reasonable to suppose that the high loss link degrades instability only for short terms. Recently, we have compared the free-space link with different attenuation with the fiber link several times, and there is no additional asymmetry observed. We actually measured the noise level of the free space link, and we know the transfer function of two way transfer mechanism. Thus, we can expect that the attributed instability should be negligible for long terms. Indeed, all link are based on assumption of two way transfer. Noise of the fiber link is roughly two order in magnitude higher than that of free space, and the link instability characterized with MDEV is still very small. Of course, it is interesting to know if there are additional asymmetries while the link loss is high. It could become limitation for better links or other remote sensing applications, we may investigate it later.

Comment 4:

Regarding novelty, the abstract and introduction could clarify that the innovation here is the incorporation of the high power combs – which is a significant challenge- but the concept of the two-way approach, linear optical sampling, implementation, operational parameters, and processing directly follow earlier work, including refs. 8,9. Additionally, more recent work from the group of Ref. 8,9 is omitted including the paired Beloy et al and Bodine et al, mentioned above, which describe comb-based frequency comparison between full optical atomic clocks over point-to-point link as well as a demonstration of carrier-phase operation (PRL, 120. 050801 (2018)) and velocity-compatible operation (Nat. Comm., 10, 1819 (2019)), which used a similar polarization multiplexed link but off a retroreflector. Some of this more recent work used sophisticated real-time processing, synchronization, and velocity-correction techniques that are not incorporated here. It would be interesting for the authors to comment on whether these would be also supportable with the amplified combs and at what link loss.

Reply:

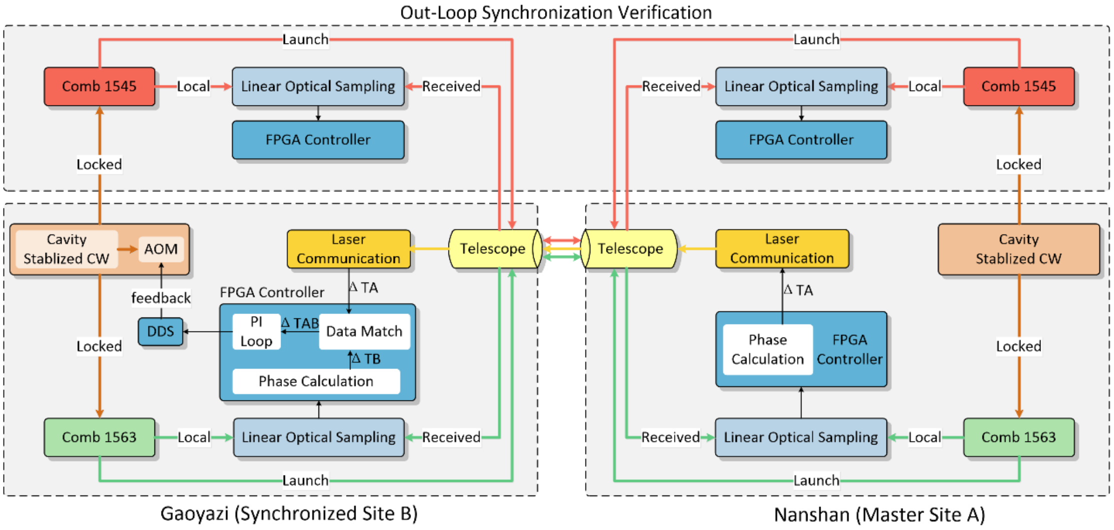
We thank the referee for the comments. Indeed, our work is based and triggered by previous pioneering work, mentioned by the referee. We take our work as a step towards the ultimate goal of global disseminating time and frequency with high stability. According to the referee’s suggestion, we have added additional experiments and comments in this revised version.

We agree with the referee that for future actual application scenarios, the sophisticated real-time processing, synchronization, and velocity-correction techniques should be also considered. In fact, the real-time processing, synchronization, and velocity-correction techniques can also work together with the high power comb and large link loss.

We add an real-time synchronization experiment result in the method part, including the real-time processing、real-time feedback、high power comb and large link loss simultaneously.

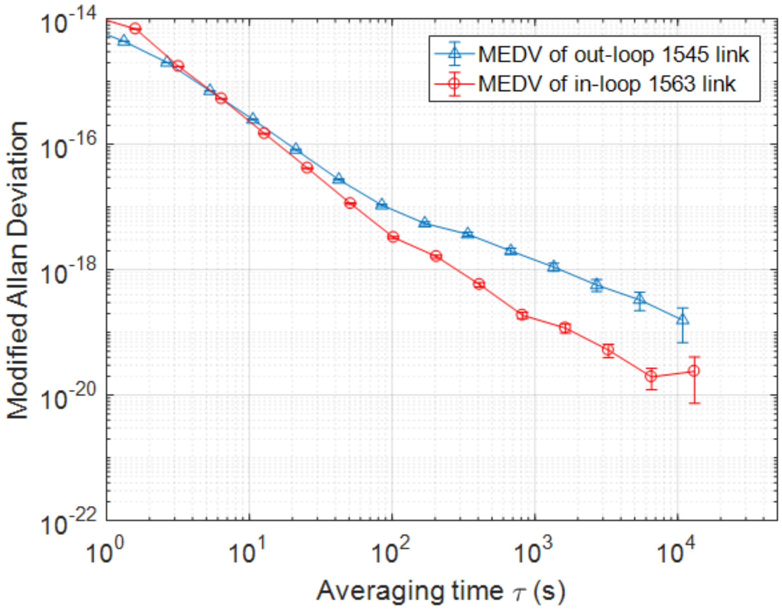
“

The synchronization experimental setup is shown in R-Fig. 5 as bellow.



R-Fig. 5 Synchronization experimental setup.

At both terminals (Nanshan and Gaoyazi) the OFCs with wavelengths of 1563nm and 1545nm are locked to USLs, which are the reference clock sources. The interference pulses between local and received comb are detected by the LOS module, and then the detected interferograms are processed by a FPGA controller. To synchronize the time at two remote terminals, three conditions should be satisfied, i.e. real-time measurement of time offset, real-time data communication and real-time clock offset adjustment. Therefore, the phase calculation algorithm is implemented in the FPGA and a laser communication link is utilized. The calculated phase information of terminal A, ΔTA, is transferred to terminal B over the 113km free-space laser communication link after coding and electro-optical conversion. Once obtained at terminal B, the ΔTA is combined with local calculated phase information of site B, ΔTB, to calculate the clock offset ΔTAB. Then through proportional integral (PI) loop filter and direct digital synthesizer (DDS), a feedback signal which represents clock offset is generated. An acoustic optical modulator (AOM) is driven by this feedback signal, which is applied to adjust the local reference clock offset relative to the master clock of terminal A. With a 5-Hz feedback bandwidth, the two terminals are synchronized via the 1563 comb link. Besides, the synchronization verification is also carried out via an out-of-loop measurement of the time offset in the 1545 comb link.



R-Fig. 6 Modified Allan Deviation for the synchronized time-frequency link.

R-Fig. 6, as show above, shows the results of the synchronized 113-km free-space link with an average link loss of 80 dB. The fractional frequency stability of in-loop link and out-loop link reach 2.4E-20 and 1.6E-19 at 10,000 s, respectively.”

For the velocity correction, we have discussed this issue in our previous work Optica,8,4(2021), where the Doppler effect of GEO is lower than that of the LEO link. This is to say, the GEO link is more similar as the statistic condition and the limiting case is the synchronous orbit, where there is almost no relative movement between the satellite and the ground station. And a previous study [Nat. Comm., 10, 1819 (2019)] demonstrated that low Doppler velocity can be solved by data processing. While for the GEO link, the major challenges are the high channel loss, which is fixed by the current work.

Higher Doppler velocity in LEO link is also interesting, which will be the next step of this research.

Revisions made in manuscript:

In the main text,

1. In lines 62-63 of Page 3, the sentence of
   "To overcome these challenges, here, we develop a number of novel optical frequency comb-based techniques,..."
   is modified to:
   "To overcome these challenges, here, we develop an optical frequency comb-based link[22],…"
2. The mentioned work above have been cited in the revision text as Ref. 28, 27, including (PRL, 120. 050801 (2018)) , (Nat. Comm., 10, 1819 (2019)).
3. In lines 193-194 of Page 9, the sentence is added:
   "Furthermore, we also implement real time synchronization with our system (see Methods).
   "

In the Methods text,

1. In Page 24-25, the details about the real-time synchronization of the 113 km free-space link is added as a new section: " Real-time synchronization".

Comment 5:

Additional technical questions:
The paper states that the use of orthogonal polarization can mimic the point ahead angle encountered in satellite missions in lines 116-118 and elsewhere. I do not understand this connection? I note that this issue has been explored in both theory and experiment in work unreferenced by this manuscript (see for instance the initial theoretical work of Robert et al, PRA 93 2016), and shown to not be problematic.

Reply:

We thank the referee for the comments.

The point ahead angle in satellite mission will induce additional non-reciprocal optical path both in atmospheric link and optics of the telescope. This issue indeed has been explored in both theory and experiments as pointed out by the referee. Precisely, in the experiment, two separated telescope are used to simulate the receiver and transmitter path. These works are indeed novel and we have cited it in the revised version as reference 33 and 34. But meanwhile, with the actual satellite ground link, the non-reciprocal issue happens in a single telescope. So a simulation of non-reciprocal optical path with a single telescope is also meaningful and interesting. That is the main motivation of our design.

Here, we proposed the orthogonal polarization scheme to sperate the two different directional light and tested that this configuration is available for future ground station with ignorable additional noise on the stability. For the transmitter path, the additional point ahead mirror should be added both in satellite and ground. In the ground, a special deformable mirror is needed in the received path to increase the single mode coupling efficiency. This telescope design, which should be needed for the real scenario, have been simulated here in our setup.

Revisions made in manuscript:

In the main text,

1. In lines 110-113 of Page 5, the sentence of
   "An orthogonal polarization setting is designed to provide large isolation of two-way transfer, and it can also simulate the nonreciprocal situation in satellite ground links due to the relative angular motion between a satellite and its ground station."
   is modified to:
   "The orthogonal polarization setting is designed to provide large isolation of two-way transfer, which is also used to simulate a real satellite ground scenario where the relative angular motion separates the backward and forward transfers[33, 34]."

Comment 6:

Can they provide more information on how the data are processed and combined, including time traces? The assumption is the Adevs are from the "difference" between the two systems, which is relevant to discussions of precision and accuracy. Given the above comment on the common-mode path noise, all pairwise combinations of the time traces would be of interest. How are gaps in the data treated at low valid data rates? Given the comment above on the commonality of signal fluctuations, can they provide a more complete analysis of the expected ADevs at high link loss (low data rates)?

Reply:

We thank the referee for pointing this issue. We would like to note that, we calculate the MDEV instead of the Adev. The detailed postprocessing process has been added in the Methods as shown below:

"

First, we have four groups of data, named "1545 A/B" and "1563 A/B". Each group of data includes one-way time measurement results and corresponding timestamps. Second, we calculate the clock offset for 1545 link by combining 1545 A/B data. In the combining process, we set a gate time and make pairs between the two group of data so that the difference of timestamps in each pair is smaller than the gate time. The data, which couldn't be paired, will be deleted in this step. Based on the paired data, the clock offset and link delay of 1545 link are calculated. The gate time in our program is $\frac{1}{2\Delta f_{r,40}}$, where $\Delta f_{r,40}$ is the difference of repetition rate of the two 1545 combs. The same program is used to process 1563 link, with the gate time changed to $\frac{1}{2\Delta f_{r,70}}$. Finally, we use the same program to process clock offsets of 1545 link and 1563 link. By finding the corresponding pairs between the two link offsets data, the difference of clock offsets is calculated.

Because we filter some data in the post-processing step, the timestamps of the difference of clock offsets are not uniformly distributed. Here, we suppose that the time interval is similar, so that we use the average valid sampling time as the gate time to calculate the MDEV."

We thank the referee for this comment on the commonality of signal fluctuations. This issue may induces the drop events of 1545 Link and 1563 Link has some correlations and leads a higher valid data rate compared to independent two links in high link loss. We have used an independent fiber link to evaluate the commonality with the two free space link loss of 76 dB, and the test results show that the common effect influenced instability at this loss level should be a small part. At higher link loss, we suppose the instability performance is the same, where the limitation for long term stability is the temperature drift. Note that all valid data are obtained when the received power is over the threshold, which means that high loss part are filtered out and do not need to care about.

Revisions made in manuscript:

In the Methods text,

1. In Page 18, the detailed information of data processing and combining have been added in the section "Linear sampling electronics system".

Comment 7:

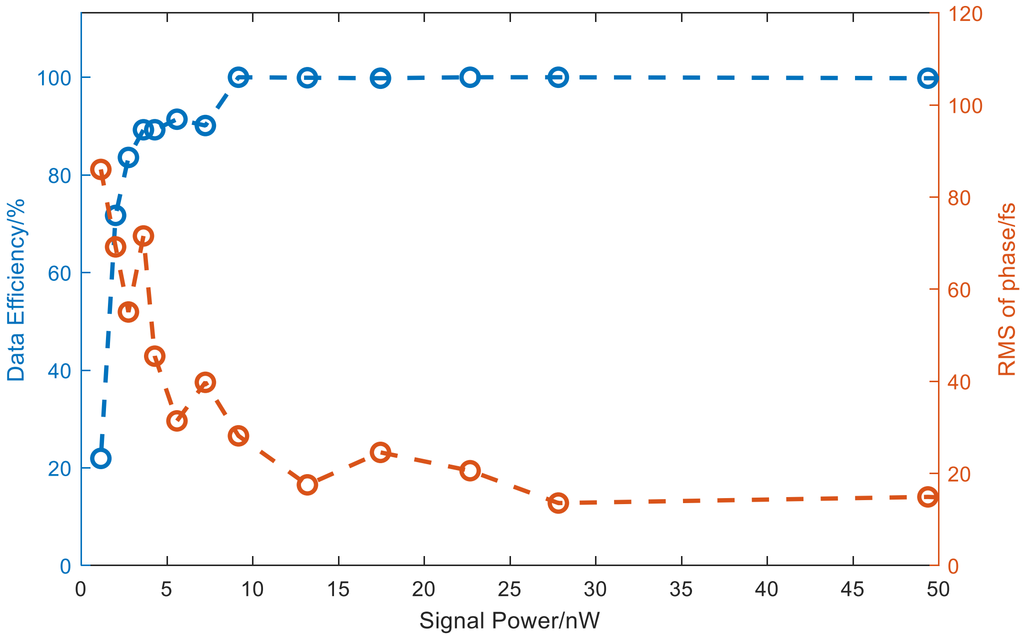
What is the detection power threshold (only mean values are given)?

Reply:

We thank the referee for the comments.

We are sorry for the unclear expression in the former manuscript. The data obtained only if the interference signal is over a threshold voltage, corresponding to a instantaneous threshold power of 2-4 nW, a little different for the four LOS detectors, which is defined by the minimum received comb power at the balanced detectors.

In fact, the valid data ratio is quite sensitive to the signal power at low level. Precisely, when we define a valid data ratio more than 80% and the residuals of linear fit smaller than 60 fs, the minimum received signal power is around 4-6 nW for all the four LOS detectors. Below is the detection power measurement curve.



R-Fig. 7 detection power measurement curve.

Since the comb output power is 1 W and the average link loss is 74 dB and 89 dB for the aligned and mis-aligned link, respectively. So the mean values of the received power is about 40 nW and 1 nW. Note that although the average power is lower than the threshold for the mis-aligned link, however, the atmospheric is fluctuates, so there is still a certain probability that the transient power of a short period is higher than the threshold.

Revisions made in manuscript:

In the main text,

1. The sentence of
   "Here, we set a threshold to select the valid data. 80% valid data can be obtained simultaneously at both terminals when the average received power is approximately 6 nW." is deleted.
2. In lines 160-161 of Page 7, the sentence is added:
   " …the data obtained only if the interference signal is over a threshold voltage, corresponding to an instantaneous power of 2-4 nW for four LOS detectors."

Comment 8:

The explanation of the 1-s Allan deviation, which is higher than some other comb-based transfer, could be clarified. Is it the combs themselves (from Figure 13) or the link? ? The sentence gives 40/sqrt(tau x Sn) - Based on a parenthetical comment elsewhere, Sn might be the data rate but the scaling with "tau" does not appear correct for the MDEV? In addition to comparing to other microwave systems, could they compare to other comb-based transfer demonstrations? If there is a small "penalty" from the use of amplified combs, that is a reasonable price for long distance operation.

Reply:

We thank the referee for the comments. Indeed，the 1-s Allan deviation is about one or half order higher compared to other comb-based transfer demonstrations (Nature Photonics, (2013),7(6); Physical Review X, (2016), 1-15, 6(2); APL Photonics, (2020), 5(7)). It is mainly caused by the use of high power amplified combs. To reduce the nonlinear effects of fiber reference, we reference a large amount of dispersion in the amplifier. The short term stability is mainly limited by the single frame timing jitter and data rate. Here, the single frame timing jitter is around 40-60 fs, which is relatively high, mainly caused by the facts that dual combs dispersion consistency is poor. Compared to low-power comb systems, the comb interference envelope is increased from 3ps to 7ps.

Note that the imperfection of the high power combs can be compensated in principle. However, since it is not the limitation factor for long term stabilities in our system, we haven’t fixed it in this experiment and keep it for the future plan.

We apologize for the wrong description of this sentence gives 40/sqrt(tau x Sn). It should be for the TDEV, not the MDEV, where tau is the sampling time and Sn is the data rate. Accordingly, this sentence has been modified to “The link instability is about $7 \times 10^{-14}/(\tau^{\frac{3}{2}} * Sn^{\frac{1}{2}})$ determined by random noise of the laser system and sampling process for short terms; while it is a few $10^{-15}/\tau$ limited by the thermal effect for long terms.”

Revisions made in manuscript:

In the main text，

1. In lines 170-172 of Page 8, the sentence of

   “The link instability is about $40/\sqrt{\tau * Sn}$ determined by random noise of the laser system and sampling process for short terms; while it is a few $10^{-15}/\tau$ limited by the thermal effect for long terms. ”
   is modified to:

   “ The link instability is about $7 \times 10^{-14}/(\tau^{\frac{3}{2}} * Sn^{\frac{1}{2}})$ determined by random noise of the laser system and sampling process for short terms; while it is a few $10^{-15}/\tau$ limited by the thermal effect for long terms.”
2. In lines 174-178 of Page 8, the paragraph has been added for the explanation of the short-term instability, which is higher than some other comb-based transfer:
   “Short term instabilities are worse than other comb-based transfer demonstrations[8,22,23] mainly due to the use of high power amplified combs. To avoid nonlinear effect, we highly chirped laser pulses, which involves in large dispersion difference. It leads to poor accuracy of time determination. In principle, this effect

can be minimized by carefully dispersion management. ”

In the Methods text,

1. In Page 22, the explanation of the short-term instability, which is higher than some other comb-based transfer, has been further clarified in the Section of “High-power low phase noise optical comb”, as bellow:
   “The short-term instability is relatively high , which is mainly caused by the use of high-power amplified combs. The short-term stability is mainly limited by the time jitter of single interference and valid interference rate. Here, the timing jitter is around 40-60 fs, which is relatively high, mainly caused by the fact that dual combs dispersion consistency is poor, corresponding to a large interference time. Compared to low-power comb systems, the comb interference time is increased from 3 ps to 7 ps. In principle, this effect can be minimized by carefully dispersion management.
   ”

Comment 9:

How is the 4-5 nanosecond ambiguity in timing removed if the GPS is only providing 30 ns relative synchronicity?

Reply:

We thank the referee for the comments. Actually, we did not remove the relative synchronicity (maybe 30 ns) between two sites in this experiment; the 4-5 nanosecond ambiguity is time difference relative to the start point.

Here, in our setup, the GPS PPS signal is used only for the start synchronization of data acquisition of the separated terminals, and it has no relationship with the 4-5 nanosecond ambiguity.

Revisions made in manuscript:

In the main text,

1. In lines 102-103 of Page 5, to clarify this, the sentence of
   “Thanks to GPS receivers at both terminals, the deviation of the time synchronization error between two terminals is approximately 30 ns.”
   is modified to:
   “Thanks to GPS receivers at both terminals, the start time of data collection for both terminals can be synchronized to be below 30 ns.”

Comment 10:

It is impressive the system work with a total loss of 89 dB. Where is the 1W is measured and what is the actual comb transmit power out of the aperture? Could the main text mention between which points the 89 dB link loss is measured and what was the aperture-to-aperture loss?

Reply:

We thank the referee for the comments. The output power of the comb is 1W measured just at the exit of the comb. Then, the comb transmits to the integrated fiber module, connected to the telescope. The transmission loss of the fiber module and telescopes optics is around 2 dB and 3 dB, respectively. So the output power of the telescope aperture is around 320 mW.

The mis-aligned 89 dB and aligned 74 dB link loss are both calculated from the comb output to the balanced detector, including the fiber optics module loss of 4 dB (transmitter and receiver), and the telescope link loss. The telescope link loss contains the telescope optical efficiency of the transmitter and receiver, the geometric attenuation, the atmospheric transmittance, the single-mode fiber coupling efficiency, and ATP system related loss.
For the aligned link loss of 74 dB, the telecope link loss is estimated to be 70 dB, including the telescope optical efficiency of 4.8 dB, the geometric attenuation of 21.8 dB, the atmospheric attenuation of ~12.5 dB, the single mode coupling attenuation of 27.9 dB, and the ATP (Aqcuisition, Tracking and Pointing) system related loss of 3 dB. So the aperture-to-aperture loss is around of 34 dB, including the geometric loss, atmospheric loss.
For the mis-aligned link loss of 89 dB, the aperture-to-aperture loss is supposed to be not change, since the additional link loss is added in the single mode coupling efficiency by manually mis aligned the fine tracking point.

The details information about the link loss calculation can be found in the Methods of section of “Link loss simulation of the horizontal free space channel”.

Revisions made in manuscript:
In the main text,

1. In lines 137-139 of Page 6, the sentence is added to made it clearer:
   “The 89 dB average loss of the mis-aligned link is measured from output of the comb to input of the balanced detector, …
   ”

Comment 11:

Regarding future ground-to-satellite links, the size/weight and power of this system is large due to the amplified combs and large aperture telescope. Can they comment on what is needed for future space payloads?

Reply:

We thank the referee for the comments. We have calculated the satellite ground scenario in our previous paper (Optica, (2021), 471, 8(4)). With a 1 m telescope in the satellite of GEO and a 1 m telescope in the ground, the downlink and uplink loss are 55 dB and 71 dB respectively. Considering detection with a sensitivity of 6 nW and 4 dB local optics loss in our experiment and 6 dB additional margin, the power of comb in

the payload is around 50 mW. The ground comb power needs to be as high as 1 W in this scenario.
Note that the 1 m aperture telescope is technically feasible for satellite payloads (https://simple.wikipedia.org/wiki/Hubble_Space_Telescope).

Comment 12:
Could they elaborate on the derivation of the 10-as number and the assumptions there? Is there a typo in low-pass vs high-pass filtering here?

Reply:
We thank the referee for pointing this unclear issue. The phase noise of atmosphere $S_{\varphi,oneway}(f)$ is $486.4f^{-8/3}$ in Fig. 2(d). When we calculate two-way residual phase noise $S_{\varphi,twoway}(f)$, we suppose it is the difference between the one-way phase noise and the one-way phase noise delayed by the link transmitting time $\tau = \frac{L}{c}$. Thus $S_{\varphi,twoway}(f) = S_{\varphi,oneway}(f)|1-\exp(i2\pi f\tau)|^2$.
The $|1-\exp(i2\pi f\tau)|^2$ term is like the one-order high-pass filter. We are sorry for the typo. Given the $S_{\varphi,oneway}(f)$, we can calculate the single-shot RMS jitter by integrating $S_{\varphi,oneway}(f)$ from system measurement bandwidth 2.6 kHz to 10 MHz as

$$RMS\ jitter = \sqrt{\int_{2600}^{10e6} S_{\varphi,twoway}(f)df} = 32as.$$

Revisions made in manuscript:
In the main text，

1. In lines 147-150 of Page 7, to make our statement clearer, we change the sentence "Considering the two-way operation of the link, the transfer function from free link noise to residual link noise is similar to a one-order low pass filter with a bandwidth of light speed over link distance. Thus, the turbulence-induced time jitter is about 10 atto-seconds, which is negligible."
   to :
   "The two-way time transfer further suppresses the link noise to $|1-\exp(i2\pi f\tau)|^2$. Time jitter of the suppressed phase noise from 2.6 kHz (sampling rate) to 10 MHz is only 32 atto-seconds and it is negligible."

Comment 13:
With regard to Figure 3, the text states there is no obvious correlation with received power since both curves start at 40 fs, but they start at different averaging times – is

that not a consequence of lower power and less valid data?

Reply:

We thank the referee for the comments. The start point of 40 fs is just the single frame timing jitter. The different averaging times is determined by the averaging data rate. The start points of the two curves are similar. This is because we set a trigger threshold so that the average power of the signals we collected is similar. Looking closely at the curve with low data rates, the starting point is slightly higher, around 50fs, because statistically the average power is slightly lower at low data rates than that of high data rates.

Revisions made in manuscript:

In the main text,

1. In lines 159-161 of Page 7, to make it clearer, the sentence of
   "The precision of valid data does not exhibit an obvious correlation to the received optical power; the start points of both TDEV curves are around 40 fs."
   is modified to:
   "The starting points of both TDEV are around 40 fs, because the data obtained only if the interference signal is over a threshold voltage, corresponding to an instantaneous power of 2-4 nW.
   "

Comment 14:

In Figure 11, is 1.414 ps the transform-limited pulse width limit for the spectra?

Reply:

No, it isn't. The 1.414 is the calculation coefficient parameter of the measurement processing. The intensity autocorrelation was usually used to measure an ultrashort pulse's intensity vs. time. The intensity autocorrelation of the pulse shown in Figure 11, which is not the pulse intensity. If the pulse shape is assumed to be Gaussian, the FWHM of its intensity autocorrelation trace is $\sqrt{2} \approx 1.414$ times the pulse width.

Comment 15:

Figures 1c was challenging to decipher. Figure 9 helps but could benefit from a caption. It might help to define the wavelengths/purpose of the dichroics? Should the "COMS" labels be CMOS and is this meant to indicate a focal plane array? What is the purpose of the APD in Figure 1c? There seem to be other unused laser wavelengths coupling to the terminals?

Reply:

We thank the referee for the comments. The figures 1C and figure 9 have been modified for clarity according the suggestions. We are sorry for the wrong labels "COMS", it should be "CMOS".

The APD in Figure 1c is used for the adaptive optics system.

There indeed some other unused laser wavelengths coupling to the terminals, there are some wavelengths for laser communication, which is unused in our experiment, which have been removed in the new figures.

Revisions made in manuscript:

In the main text,

1. In Page 34, Figure 1c has been redrawn for clearer.

In the Methods text,

1. In Page 42, Figure 9 has been redrawn for clearer. The caption of Figure 9 has been added for details. The unused wavelengths have been deleted.
2. In Page 19-20, the detail information of the telescope has been rewritten in the section of "Optical transceiver telescope" for clearer, including the purpose of the APD, etc.

Comment 16:

Could more details be provided on the adaptive optics system, e.g. operation, beacon power, performance, benefit over tip/tilt etc?

Reply:

We thank the Referee's suggestion. Indeed, the adaptive optics system is one of the crucial technologies for single mode fiber coupling. However, we couldn't benefit much from the technology in the free space link parallel to the ground of our experiment. We expect that it can improve the single mode fiber coupling efficiency in satellite ground scenario.

The details of the adaptive optics system are as follows:

There are two functions employed to resist atmospheric turbulence and improve the single mode fiber coupling efficiency. One function is the fast-steering-mirror (FSM)-based fine tracking system, which is used for correcting lower-order aberrations. The central wavelengths of the beacon lights are 785 nm and 915 nm, respectively. The power of 785 nm and 914 nm laser is 200 mW and 400 mW, respectively. In the receiver, the fine tracking complementary metal–oxide semiconductor (CMOS) camera detects the tilt-type aberrations via the incoming beacon laser. It then guides the FSM driven by piezoceramics to perform the correction with a closed-loop bandwidth of 100 Hz. The other function is the adaptive optics (AO) system, which is based on a deformable-mirror (DMIR) here. The AO system can correct higher-order aberrations of the wavefront introduced by the atmosphere. The beacon laser is shared in the receiver and divided into two parts, ~30\% for the fine tracking and ~70\% for the AO system. A modal version of the stochastic parallel gradient descent algorithm (M-SPGD) is employed for AO. The coupled power of SMF is the performance metric in the M-SPGD algorithm, which can be found in more detail in our previous work[43]. We utilize a silver-coated DMIR with 97 actuators. In addition, an avalanche photo-detector (APD) is used to detect the coupled power of the reference beacon laser. The AO reference laser before entering the APD is about 2 nW. Note that the fine tracking system is similar as the tip/tilt system used in other works [Appl. Opt. 56, 9406-9413 (2017)].

Revisions made in manuscript:

In the Methods text,

1. In Page 19-20, the detailed information of the adaptive optics system is added in the Methods section of " Optical transceiver telescope".
2. New added citation as Ref. 43: Kui-Xing Yang, *et al.*, Optics Express **28**, 36600 (2020).

Comment 17:

The analysis of the link loss is very complete, showing good agreement at a specific cn2. What range of CN2 were observed? The link is fairly close to the ground and presumably highly turbulent, which make the 113-km demonstration all that much more impressive.

Reply:

We thank the referee for the comments. The range of CN2 were observed between 3e-16 and 5e-16. Note that this value is smaller than that of the NIST work between 1E-15 and 1e-14 [Nature Photonics, (2013), 434-438, 7(6)]. The mainly reason is that the altitude of both terminals are high, with 2000 m and 1800 m, respectively. However, since the long distance atmosphere of 113 km, the integrated turbulence is very strong, which can be reflected by the parameter of R0, which is almost around 1-2 cm.

Referee #2 (Remarks to the Author):

Comment 0:

The manuscript describes a major breakthrough into the realization of free-space satellite-based long range optical time and frequency dissemination, with impacts for fundamental physics (search for dark matter, test of fundamental constant, test of relativity…). The major achievement of the authors as compare to previous work in the field is the operation of two 1W-power Optical Frequency Comb, that allows the authors to demonstrate world-record in range for free-space optical fiber links, while keeping state of the art in terms of precision. The experimental results are impressive, and the manuscript is very likely to be highly impacting, what deserves publication in Nature.

The manuscript describes other several improvements as compare to their previous paper published in 2021: authors described an improved and original electronics system and an improved interferometer system (length mismatch minimisation, better temperature stabilization) that allows them to reach lower noise floor stabilities, and larger link loss acceptance from 72 dB up to 90 dB, what is another impressive achievement. The authors reports for about 235 hours of operation acquired in 21 days, what is also an impressive achievement given the high loss acceptance of their system.

The manuscript is well written, expose clear explanations. Data and methodology are valid, the authors show high quality data, and appropriate treatment of uncertainties. The manuscript shows rigourous reasoning and robust results, certainly reliable and valid.

Figures are well presented generally speaking, aside of small mistakes I detailed later on in this review. The supplementary materials are technically sound and are very complete. Supplementary materials should provide expert readers with the necessary technical informations they might need, while the main text indeed address a broad audience.

The manuscript has however several weaknesses that required a revision before acceptance.

Reply:

We thank the referee for his/her time and efforts to review our work. We are very glad to see that the referee think that our work is impressive and deserves publication of Nature. We have addressed the referee's concerns point by point in below.

Comment 1:

Major comments, questions and suggestions to the authors:

- The authors claim line 31 (abstract) and line 81 that they established two independent two-way time and frequency links. May be the wording should be slighty softened or put better in context. In my view, the authors are partly right in their statements as they used two independent opto-electronics systems, but the telescopes are still in common mode. So the two links are not totally independent. The authors do not show resiliency with respect to different paths or demonstrate the immunity versus mechanical noise and thermal noise affecting the telescope. To the best of my understanding, in a free space configuration, there should be three telescopes used to realize a ‘lambda-shape’ or a ‘V-shape’, connecting one ground to two satellites, or one satellite to two ground stations.

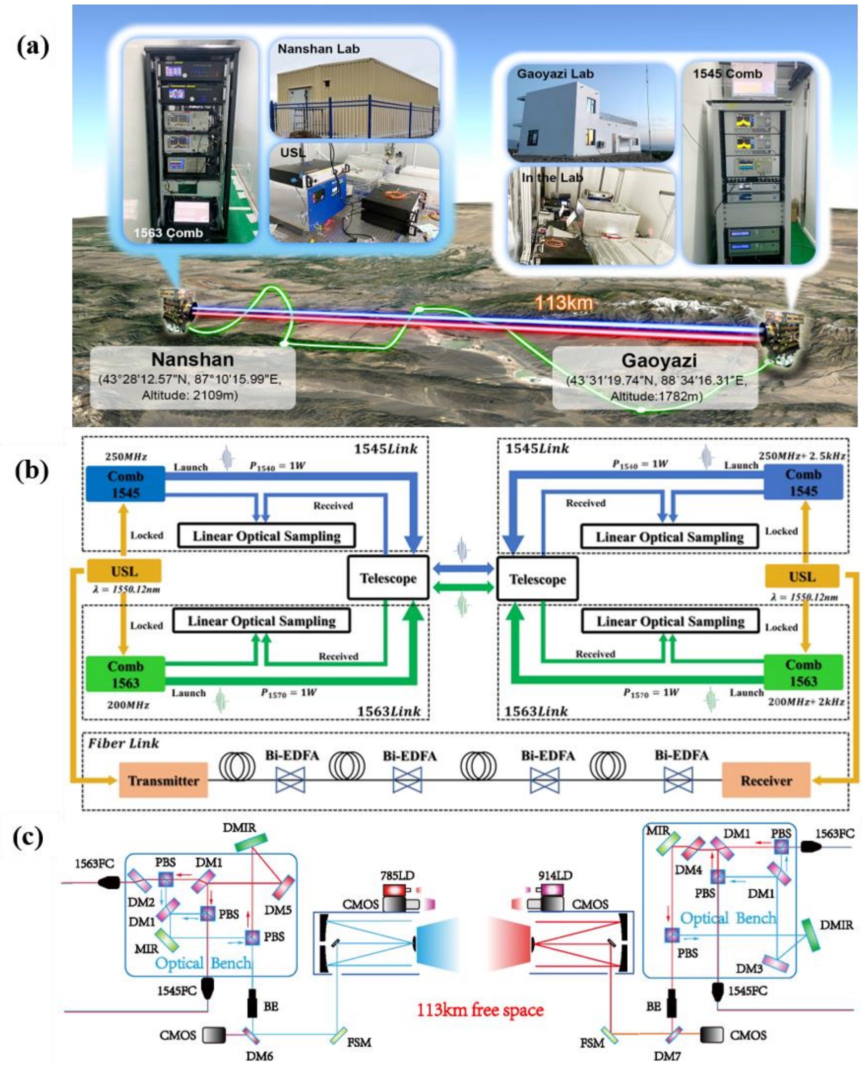
Reply:

We appreciate the referee for this valuable comment which definitely help us to improve our work. We fully agree with the referee that the two links, 1545/1563 share the same terminals and air path, which might result in a smaller deviation. Actually, we became aware of this issue soon after the submission of this manuscript. So we have established an independent fiber link connecting two remote locations, and utilized the fiber link to evaluate the two 113 km free space links. The MDEV is below 4E-19 at 10,000 seconds at such configurations. The result is slightly higher than our previous result, which might be attributable to the common noise. Nevertheless, it doesn’t affect our claim that our system can distribute time-frequency signal over 113 km free-space link at the 19th decimal instability, which firmly constitutes a critical step towards future satellite time-frequency dissemination. We have added the new experimental results in the revised version.

Revisions made in manuscript:

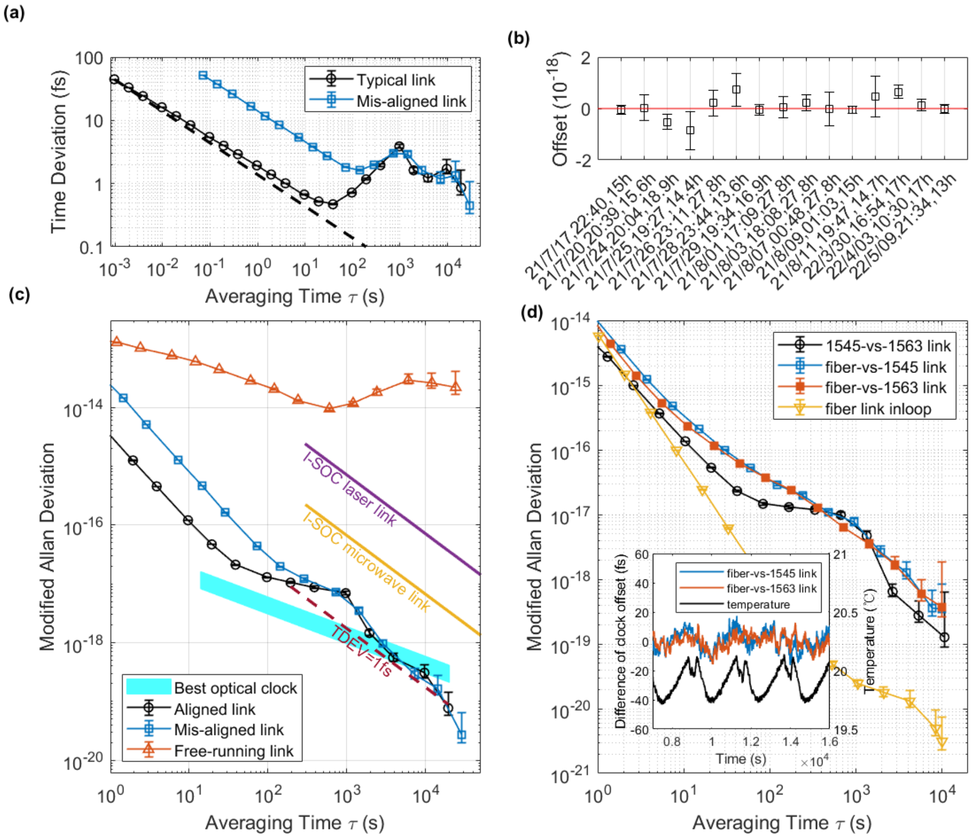
In the main text,

1. In Page 34, the Fig.1a and Fig.1b have been redrawn to add the fiber time-frequency transfer link.
   The new fig.1 (here is R-Fig. 1) is as follows:



R-Fig. 8 The experimental setup. (a). Overview of the 113 km free-space time-frequency dissemination experiment. (b). The main diagram of the system, which contains a 1545 nm free-space link and a 1563 nm free-space link and a fiber link. USL: ultra stable laser. (c). The optical layout of the optical transceiver telescope, where different colors of optical paths indicate different polarizations. PBS: polarizing beam splitter; 1545FC (1563FC): fiber collimator for two-way 1545 (1563) nm comb transceiver; DM: dichroic beam splitter; MIR: mirror; DMIR: deformable mirror; WDM: wavelength division multiplexer; BE: beam expander; FSM: fast steering mirror; CMOS: complementary metal–oxide semiconductor.

2. In Page 36, the Fig.3d have been added about comparisons among the three links of the fiber link and two free space links. The Fig.3b has been redrawn to add three more data sets acquired in 2022.
   The new Fig.3 (here is R-Fig. 2) is as follows:



R-Fig. 9 Experimental results of time-frequency transfer. Captions for (d) is: "The comparison among fiber link and two free-space links. The black circle line: two free-space links; The blue square line: fiber and 1545 comb link; The orange solid square line: fiber and 1563 comb link; The yellow triangle line: in-loop stability of fiber link. The inset shows the trend for the difference between fiber link and free-space links with temperature."

3. In lines 77-80 of Page 4, the sentences have been added to descript the motivation and setup of the fiber link as below:
   "Since the two multiplexing channels share the same free-space link, there exits common mode noise. In order to better evaluate our system, we also established an independent fiber link connecting the two terminals with a distance of 209 km. All links (1545 nm free-space, 1563 nm free-space and the fiber link) share the same USL at each terminal."
4. In lines 186-194 of Page 8 in the Main text, the paragraph has been added:
   "Relative instability among two free-space links and the fiber link are in the range of 1-4E-19 at 10,000 s as shown in Fig. 3d. Thermal drifts effect (see the inset) limits the link performance for long terms. Although in-loop instability of the fiber link is negligible, there is 0.5 m out-of-loop fiber used to connect the fiber link at both terminals respectively. Instability of two space link is slightly better because equipment location of two free-space links are closer than that of the fiber link. In

fact, the phase noise of atmosphere is much less than that of the fiber link and has been efficiently suppressed by two-way operation (See Fig.2 d). So, the thermal drift effect should be dedicated to the fiber parts.”

5. In line 29-31 of Page 2 in the abstract, the sentence of

   “we demonstrate free-space time-frequency dissemination over two independent links with femtosecond time deviation, 3E−19 at 10,000 s residual instability and 1.6E−20 ±4.3E−19 offset.”

   has been modified to

   “we demonstrate 113-km free-space time-frequency dissemination with 6.5E-20±3.5E-19 offset, and an instability of below 4E-19 at 10,000 s measured by comparison of two wavelength division multiplexing links and an independent fiber link”

6. In lines 153-155 of Page 7, the sentence of

   “…,we carried out a time-frequency transfer experiment over approximately 3 weeks. We obtained a total of 12 data sets, and the continuous acquisition duration of each data set was between 13.6 and 27.8 hours.”

   is modified to:

   “…,we carried out a time-frequency transfer experiment, and obtained a total of 15 data sets. And the continuous acquisition duration of each data set was between 13 and 27.8 hours.”

7. The sentence of

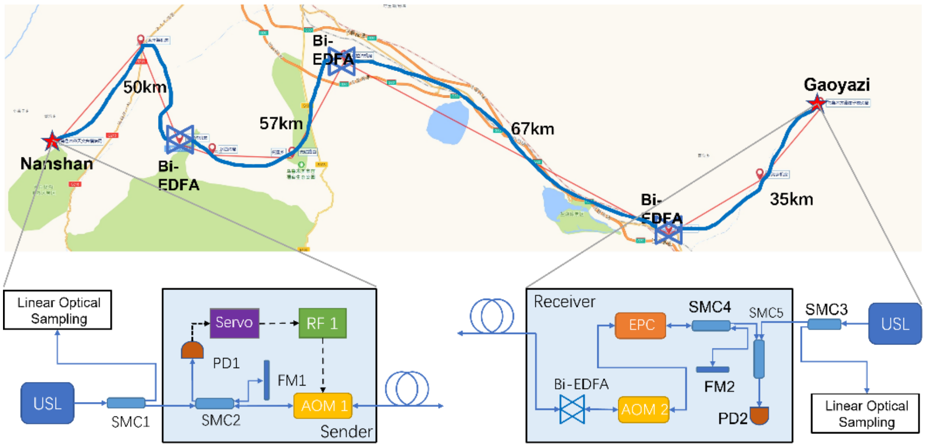
   “…, according to 12 times experimental results.” has been deleted.

In the Methods text,

1. In Page 22-24, the details about the fiber time-frequency transfer link is added as a new section: ” fiber time-frequency transfer link” as follows:

   “

   The total fiber length is 209~km, and the loss is 65~dB. To compensate for the transmitted loss, three bidirectional amplifiers are put on the path and one is put at terminal B. The detailed config is shown in R-Fig. 3.



R-Fig. 10 Setup for fiber time-frequency transfer. USL: Ultra-stable laser. SMC: Single-mode coupler. PD: Photon diode. RF: Radio frequency source. FM: Faraday mirror. AOM: Acoustic optical modulator. EPC: electric polarization controller. Bi-EDFA: Bidirectional erbium doped fiber amplification.

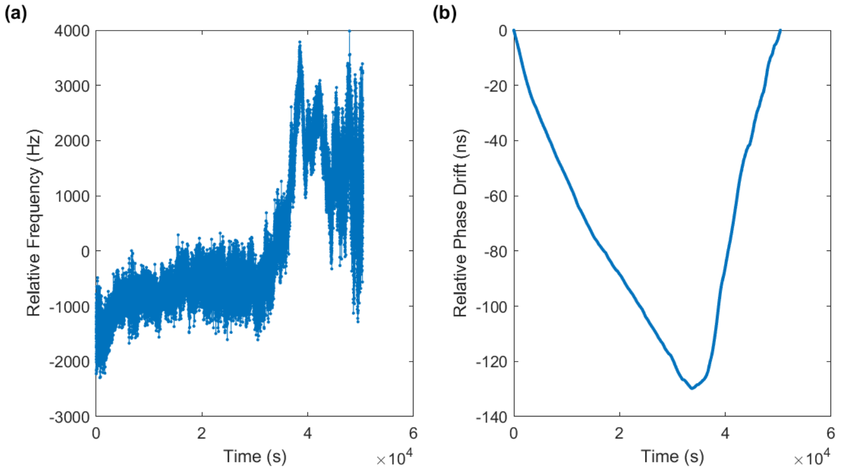
At terminal A, we split the USL into two parts. One is used to lock the combs, the other is going through an optical module for fiber time-frequency transfer. The module is actually a highly unbalanced Michelson interferometer, with the short arm in the local optical module and the long arm up to terminal B. The AOM at terminal A is used to compensate for the noise in the fiber link, and the AOM at terminal B is used to distinguish the reflected laser from terminal B with the backward scattering signal during the link. The reference for locking electrics is synchronized with the repetition rate of the comb, to ensure that the fiber time-frequency transfer system shared the same reference frequency with the free-space system. At terminal B, the local USL beats with the signal laser. The beating signal is filtered with a tracking oscillator and measured with a K-K phase recorder. A Direct Digital Synthesis (DDS) converts the repetition rate of 1563 nm OFC to 10~MHz to drive the phase recorder.

In the free-space time-frequency transfer link, the clock offset is calculated with the time base that came from the 1545 OFC. The phase difference of the fiber link should be converted to this time base too before we compare it with the free-space link. At terminal A, the ultra-stable laser and the locking electrics are all synced with the time base, so the frequency of the signal transmitted to terminal B can be calculated. At terminal B, we also record the ratio between the frequency of the ultra-stable laser and the 10~MHz as k. When the K-K phase recorder reads the frequency as $\mathrm{f_{beat}}$, the real frequency at the time base of the terminal A should be $\frac{f_{\mathrm{cw,B}}}{k} * \frac{\mathrm{f_{beat}}}{10\ MHz}$. We then have

$$f_{\mathrm{cw,B}} + \frac{f_{\mathrm{cw,B}}}{k} * \frac{\mathrm{f_{beat}}}{10\ MHz} = f_{\mathrm{signal}}.$$

The value of $f_{\mathrm{cw,A}} - f_{\mathrm{cw,B}}$ can be easily calculate from above formula. To convert the frequency to phase/time, one needs to be aware that reference frequencies of K-K phase recorder at different terminal are not same. Another ratio is needed to correct the gate time. The ratio can also be figured out from $f_{\mathrm{cw,B}}$ and $f_{\mathrm{cw,A}}$. Finally, the offset between the start recording time of the free-space link and the fiber link can be determined by minimizing the phase drift of the comparison result. Most of our fiber links are aerial optical fiber. There are several breakpoints in fiber link data while optical cycle slips happen. For each breakpoint, we remove 50 seconds of data before and after this point.

The comparisons among the fiber link and two free-space links are measured. The stability of the fiber-vs-1545 link is 3.5E-19@10000s and the stability of the fiber-vs-1563 link is 3.7E-19@10000s. The stability of the 1545-vs-1563 link is 1.3E-19@10000s. There is some common-mode noise effect between the 1563 and 1545 link and its value should be below 4E-19 at 10,000 s, which is mainly attributed to the thermal drift of fibers since the out-of-loop connecting fiber link is located at outside of the original thermal shield box. This result shows the stability of difference of fully independent links can reach the 4E-19 level. In fact, since the phase noise of the atmosphere caused by the piston effect and temperature drift mainly concentrates on low frequencies below 10 Hz, it has been efficiently suppressed by two-way operation. Indeed, the noise of the aerial optical fiber (shown in R-Fig. 11.) is much larger than the noise of the atmosphere; while the residual noise contribution is negligible (see fiber link in-loop).



R-Fig. 11 (a) The frequency of driver signal for AOM~1 relative to its center value. The AOM~1 is used to compensate for the Doppler noise in the fiber link. So the changing of the frequency implies the changing of doppler noise. (b) The accumulated phase change in the link. This is integral for the frequency data at the subfigure (a).
”

<u>Comment 2:</u>

Line 46-48: The authors state ‘So far, optical carrier phase transfer through fiber links with two-way transfer operation has reached over 1,000 km with a stability of 10−19 at 100 s [17, 18]. However, such a method is incompatible with the existing telecommunication fibers.

The sentence is partly true according to citation, but, since 2012-2013, major advances happen with fiber links. One should comment first that these fiber links experiments are using active compensation, and indeed are incompatible with existing telecommunication fibers. But nowadays, fiber links reach several thousand of km with uncertainties <1e-19, and integration time over several months, over telecommunication network. See for instance Cantin et al., , « An accurate and robust metrological network for coherent optical frequency dissemination », New J. Phys., vol. 23, n° 5, p. 053027 (2021), doi: 10.1088/1367-2630/abe79e.

Second, most of the work accomplished with long haul fiber links are using active compensation and not a two-way schemes, and therefore I believe the sentence give a wrong picture to non expert readers.

I suggest to rephrased these sentences, and provide also there a citation to [34], that describe also the fiber connection from NPL to PTB through the REFIMEVE network, that spans 2200 km in total, and shows days of data.
This being said, it does not weaken the second part of the argument, about mountains, marine, intercontinental and space links, that I believe to be true.

Reply:
We thank the referee for kindly point out this issue, which definitely help to improve our manuscript to provide an updated picture of the field.

Revisions made in manuscript:
In the main text,

1. In lines 46-48 of Page 3, according to the referee's suggestion, we have revised these sentences,
   "So far, optical carrier phase transfer through fiber links with two-way transfer operation has reached over 1,000 km with a stability of 10−19 at 100 s. However, such a method is incompatible with the existing telecommunication fibers, and can be difficult to reach certain locations such as mountains, marine and intercontinental ranges, and spaces"
   to "So far, optical carrier phase transfer through fiber links has reached over 1,000 km with a stability of 10−19 at 100 s. However, such a method can be difficult to reach certain locations such as mountains, marine and intercontinental ranges, and spaces."
2. The paper (New J. Phys., vol. 23, nº 5, p. 053027 (2021)) has been cited added in the revision as Ref. 20.

Comment 3:
Line 121-123 : It seems to me that the adaptative optic is one of the key ingredient of the system. I did not find in the manuscript the bandwidth of the correction applied by the adaptative optics? It would be helpful too to document the dynamic of the correction.

Reply:
We thank the referee for the comments. Indeed, the adaptive optics system is one of the crucial technologies for single mode fiber coupling. However, we couldn't benefit much from the technology in the free space link parallel to the ground of our experiment. We expect that it can improve the single mode fiber coupling efficiency in satellite ground scenario.

The details of the adaptive optics system is as follows:

There are two functions employed to resist atmospheric turbulence and improve the single mode fiber coupling efficiency. One function is the fast-steering-mirror (FSM)-based fine tracking system, which is used for correcting lower-order aberrations. The central wavelengths of the beacon lights are 785 nm and 915 nm, respectively. The power of 785 nm and 914 nm laser is 200 mW and 400 mW, respectively. In the receiver, the fine tracking complementary metal–oxide semiconductor (CMOS) camera detects the tilt-type aberrations via the incoming beacon laser. It then guides the FSM driven by piezoceramics to perform the correction with a closed-loop bandwidth of 100 Hz. The other function is the adaptive optics (AO) system, which is based on a deformable-mirror (DMIR) here. The AO system can correct higher-order aberrations of the wavefront introduced by the atmosphere. The beacon laser is shared in the receiver and divided into two parts, ~30\% for the fine tracking and ~70\% for the AO system. A modal version of the stochastic parallel gradient descent algorithm (M-SPGD) is employed for AO. The coupled power of SMF is the performance metric in the M-SPGD algorithm, which can be found in more detail in our previous work[43]. We utilize a silver-coated DMIR with 97 actuators. In addition, an avalanche photo-detector (APD) is used to detect the coupled power of the reference beacon laser. The AO reference laser before entering the APD is about 2 nW. Note that the fine tracking system is similar as the tip/tilt system used in other works [Appl. Opt. 56, 9406-9413 (2017)].

Revisions made in manuscript:

In the Methods text，

1. In Page 19-20, the detailed information of the adaptive optics system is added in the Methods section of “ Optical transceiver telescope”.
2. New added citation as Ref. 43: Kui-Xing Yang, *et al.*, Optics Express **28**, 36600 (2020).

Comment 4:

The authors reach higher power optical frequency comb using two stages EDFAs. Is there any information on the additional phase noise due to the amplification ?

Reply:

We thank the referee for the comments. Since the amplifier is inside the comb locking loop, the additional phase noise of certain amplified comb teeth is negligible. However, The deterioration of the spectral and dispersion consistency of the amplifiers leads to poor time measurement accuracy of LOS. Short-term stability deteriorates. However, it does not limit link performance for long-terms.

Revisions made in manuscript:

In the main text，

1. In lines 174-178 of Page 8, the paragraph has been added:

   "Short term instabilities are obviously worse than other comb-based transfer demonstrations mainly due to the use of high power amplified combs. To avoid nonlinear effect, we highly chirped laser pulses, which involves in large dispersion difference. It leads to poor accuracy of time determination. In principle, this effect can be minimized by carefully dispersion management.
   "

Comment 5:

Line 98 : 'the length of mismatch is optimized to be below 5 cm': how is the path imbalance measured ? Is the temperature fluctuations of 7 mK arise from an in-loop measurement or out-of-loop ?

Reply:

We thank the referee for the comments.

The imbalance path requirement of 5 cm is guaranteed by the manufacturer, which is not directly measured.

In our system, we tested the module's temperature coefficient. We tuned the module temperature and test it's influence to the time transfer data. The coefficient is tested to be around 1fs/K， which corresponds to about imbalance path of 300 nm. Giving the Fiber Refractive Index Temperature Coefficient of 1E-5, imbalance path can be calculated to be around 3cm.

The temperature fluctuations of 7 mK is arise from an in-loop measurement.

Revisions made in manuscript:

In the main text，

1. In lines 96-97 of Page 5, to state it clearer, the sentence of:

   "…the temperature is stabilized with a thermoelectric cooler (TEC), exhibiting a standard deviation of 7 mK,…"

   Has been modified to:

   "…, the temperature is stabilized with a thermoelectric cooler (TEC), exhibiting an in-loop deviation of 7 mK …"

Comment 6:

Figure 2c : x-label says Time in (s), but displayed ticks are 17:09 to 20:09. According to the corresponding temperature variations, I have severe doubts that this is seconds. Please correct the x-label.

It would be also much more convenient to show the y-axis in ns, instead of a scaling factor of 1e5 with units in fs.

Reply:

We thank the referee for the comments. The figure has been modified according the suggestion.

Revisions made in manuscript:

In the main text,

1. In Page 35, the x-labeb of Figure 2c “Time (s)” is modified to “Time”. The y-axis is modified to be in ns.

Comment 7:

Line 145-147: On this same plot, authors claims that the free link delay fluctuations are due to temperature, but the correlation does not seem that high. Can the authors provide the level of correlation ? Can wind turbulences and/or humidity variations can alter the propagation delay ? Furthermore, it seems that there is a delay between the temperature variation and the propagation delay variation ? Is it due to the thermal conductivity between the air and the experimental system laying in the laboratories ? Are the temperature recorded at the two laboratories sites ? Is there any weather station data available ?

Reply:

We thank the referee for the well and detailed comments and questions.

The temperature is the air sampled at one terminal site of Gaoyazi. It can not reflect the average temperature of the entire 113 km link, which is difficult to obtain quantitatively. So the correlation does not seem that high. We think the influence of wind speed and humidity should be relatively small.

Unfortunately, we do not have the data of the weather station. We think it’s interesting and will be the direction of future experiments, including the atmospheric spectrum measurements based on our setup [similar work in Optica 4, 7(2017)], even humidity, and wind speed.

Note that the phase noise of atmosphere is much less than that of the fiber link and has been efficiently suppressed by two-way operation (See Fig.2 d in main text and Fig.16 in Methods), which can be ignored in our experiment. However, the influence of the atmosphere weather station to the short term instability should be investigated when better performance is expected.

Comment 8:

One main claim of the paper, and related to its potential impact, is that this 113-km link mimics the attenuation of much longer links, to LEO, MEO and GEO orbits. However the two laboratories are static, and a satellite is moving… So I missed a discussion on a moving target. It will be an improvement of the manuscript if the authors can describe how movements of satellites will or will not alter their results, in stability and accuracy (see also comment on line 116-118).

Reply:

We thank the referee for the comments. We have discussed the moving target issue in our previous work Optica,8,4(2021), where the Doppler effect of GEO is lower than that of the LEO link. This is to say, the GEO link is more similar as the statistic condition and the limiting case is the synchronous orbit, where there is almost no relative movement between the satellite and the ground station. And a previous study [Nat. Comm., 10, 1819 (2019)] demonstrated that low Doppler velocity can be solved by data processing. While for the GEO link, the major challenges are the high channel loss, which is fixed by the current work.

Higher Doppler velocity in LEO link is also interesting, which will be the next step of this research.

Revisions made in manuscript:

In the main text,

1. In lines 202-205 of Page 9, the paragraph is added:
   "The Doppler effect will be a challenge for satellite due to relevant transfer asymmetry in the next step. The previous study[27] has demonstrated \$10^{-19}\$ instability of the link under low Doppler velocity such as 24 m/s; while large Doppler velocity condition still needs to be further investigated.
   "

Comment 9:

In the same line of idea, I believe the manuscript can be improved if authors draws some perspective on how to solve issues that may arise from a higher probability of multi path when extending their link to several thousands of kilometers.

Reply:

We thank the referee for the comments.

To solve issues for links with distance of several thousands of kilometers, it can not be directly seen by free-space in the near ground limited by the radius of curvature of the earth. The future global time-frequency network should be connected by multi paths including the satellite-ground free-space link and the ground long-distance fiber link.

Another concept of multi path effect is that the fluctuation of signal arrival time due to the received signals travel different paths commonly coming from different directions through reflection or scattering with the direct signal. This is a crucial issue in the area of microwave GNSS (See Ref: https://gssc.esa.int/navipedia/index.php/Multipath). Here, for optical laser transmission, since the laser has good collimation, with small exit divergence angle and receiving field angle, there is no the traditional multi path effect reflected by other objects. For example, the receiving field angle is only 9 urad for the single mode coupling in our setup.

However, the strong atmosphere turbulence can cause intensity scintillations and phase discontinuity, which leads to error in wave-front measurement. This effect can also be considered as multi path issue, which induces spatial mode distortion. In our setup, the single mode receive coupling has implemented spatial filtering to greatly reduce the influence of multipath issues. In fact, the R0 of the atmospheric is much smaller than the aperture of the telescope, multi path issue has been exist. For the satellite-ground link of several thousands of kilometers or even longer, the atmosphere turbulence is effectively less than that of the 113 km near ground link. So, the so-called multi path issue will not get more worse.

Revisions made in manuscript:
In the Methods text,

1. In Page 16 of the Section “Link loss simulation of the horizontal free-space channel”, the sentence is added:
   “Note that the r0 of the atmospheric is much smaller than the aperture of the telescope, the multipath issue exists. In our setup, spatial filtering is inserted by the single mode coupling to greatly reduce the influence of the multipath issue. For the satellite-ground link of several thousands of kilometers, the atmospheric turbulence is actually less than that of the 113 km near ground link, so that the multipath issue should not be a problem. ”

Comment 10:

Minor comments:
Line 52: the sentence is not that much explicit. It would be helpful for non expert readers to explicit why these other experiments are not matching the requirements.

Line 57-58: The sentence is not that clear, as it compares CW in fiber with a situation in air, where there is drop-outs. I suggest to rephrase.

Line 64 : ‘Further, we employ linear optical sampling that offers femtosecond accuracy over the whole ambiguous range’. Citation to [22] there would be helpful.

Line 116-118 : Authors states ‘it can also simulate the nonreciprocal situation in satellite ground links due to the relative angular motion between a satellite and its ground station’. I did not understand this point.

Fig.3 : It would be better to extend the frame of figures 3A and 3b, so that the last point of the curves can be easily seen.

Bibliography:
Line 44: there was search for dark matter with optical fiber network of optical clocks. I suggest to add the following reference
B. M. Roberts et al., « Search for transient variations of the fine structure constant and dark matter using fiber-linked optical atomic clocks », New J. Phys., vol. 22, nº 9, p. 093010, sept. 2020, doi: 10.1088/1367-2630/abaace.

Typos:

line 81: free-spece > free-space
line 94: fluctuaitons > fluctuations
line 134: tolarence > tolerance
line 144: flucatuation > flutcuations
line 202: apmlifiers > amplifiers
line 237: avarage >average
line 241: distritbution >distribution
line 245: ‘and the local OFC power at the LOS is kept same’ > and the local OFC power at the LOS is kept the same line 246: obtianed > obtained Line 355-356: The sentence looks incomplete or incorrect grammatically.

Reply:

We thank the referee for the comments.

The corresponding text have been modified according the suggestions.

Revisions made in manuscript:

In the main text,

1. In line 52 of Page 3, the sentence of “On this path, previous work have achieved a distance up to a dozen kilometers[8, 20−22] in a mirror-folded configuration, which cannot meet the high demands required by future satellite-ground time-frequency dissemination.” is modified to:

   “Previous work, however can only reach a distance up to a dozen kilometers [23,24], which limits its applications in many long-distance scenario. Meanwhile, the current technology cannot meet the high demands of link loss required by future satellite-ground time-frequency dissemination. ”
2. In line 58-59 of Page 3, the sentence of “The continuous-wave (CW) laser carrier used in fiber time-frequency links typically has an ambiguous range of a few fs,…” Is modified to

   “The continuous-wave (CW) laser carrier used in time-frequency links[29] typically has an ambiguous range of a few fs, …” Here, a new citation of [Physical Review Letters, (2022), 128(2)] is added to show that this CW method is also being investigated to use in free space.
3. In line 63, the Citation to [22] is added.
4. For Line 116-118 issue, we are sorry for the unclear description.

   The point ahead angle in satellite mission will induce additional non-reciprocal optical path both in atmospheric link and optics of the telescope. This issue indeed has been explored in both theory and experiments as pointed out by the referee #1. Precisely, in the experiment, two separated telescope are used to simulate the receiver and transmitter path. These works are indeed novel and we have cited it in the revised version as reference 33 and 34. But meanwhile, with the actual satellite ground link, the non-reciprocal issue happens in a single telescope. So a simulation of non-reciprocal optical path with a single telescope is also meaningful and interesting. That is the main motivation of our design.

Here, we proposed the orthogonal polarization scheme to sperate the two different directional light and tested that this configuration is available for future ground station with ignorable additional noise on the stability. For the transmitter path, the additional point ahead mirror should be added both in satellite and ground. In the ground, a special deformable mirror is needed in the received path to increase the single mode coupling efficiency. This telescope design, which should be needed for the real scenario, have been simulated here in our setup.

In lines 111-113 of Page 5, this sentence has been modified to:
"The orthogonal polarization setting is designed to provide large isolation of two-way transfer, which is also used to simulate a real satellite ground scenario where the relative angular motion separates the backward and forward transfers[33,34]"

5. In Page 36, the fig.3A and fig.3B have been modified according to the suggestion.
6. Bibliography: In line 45 of Page 2, this citation is added as Ref. 16.
7. All the typos are corrected mentioned above.

Reviewer Reports on the First Revision:

Referee #1 (Remarks to the Author):

The authors have substantially revised their manuscript. As noted in their reply, they too realized the issue of common-mode noise shortly after submission and must have immediately embarked on a very significant experimental effort to add the fiber link between the two sites. This effort led to measurement campaigns on 3/30, 4/03 and 5/09. It is very impressive they were able to accomplish this significant experimental effort over the spring and, most importantly, the additional data runs show excellent results (Fig. 3b). By adding the fiber link, they successfully address the fundamental problem with the initial submission regarding “truth”. In addition, they have made other clarifications to the manuscript, as well as including real time synchronization in the final methods section. I think the revised manuscript is definitely suitable for publication. I have two minor revisions they could consider listed below.

1) The only actual data traces appear in a small inset of Figure 3d. Could they add a supplemental figure that gives the full time traces for one example run (perhaps one of the later runs showing all four one-way time data and the fiber data)? This would help the reader understand the experiment as otherwise the figures are limited to the “statistics” (e.g. MDEVs) . Similarly, could they specify which data runs gave the MDEV curves of Figure 3d?

2) The revised manuscript is much clearer on the contributions to the link loss, particularly in the methods section. Some distilled version of this information could appear in the main text. For example, the abstract quotes “channel loss of 89 dB”, which could be read to mean just the free-space link loss but includes the ~8.8 fixed loss of the transmitter/receiver (“LOS optics loss” + telescope optical efficiency). This 8.8 dB total is actually quite low considering the complexity of the system, but they might separate out this contribution either there or in the paragraph of 119-139, e.g. by mentioning the 74 dB/ 89 dB include that 8.8 dB. (The reason is for comparison to existing free-space optical comm systems or design of future free-space systems).

Referee #2 (Remarks to the Author):

Review of the manuscript entitled ‘113 km Free-Space Time-Frequency Dissemination at the 19th Decimal Instability’,
by Qi Shen, Jian-Yu Guan, Ji-Gang Ren, Ting Zeng, Lei Hou, Min Li, Yuan, Cao, Jin-Jian Han, Meng-Zhe Lian, Yan-Wei Chen, Xin-Xin Peng, Shao-Mao Wang, Dan-Yang Zhu, Xi-Ping Shi, Zheng-Guo Wang, Ye Li, Wei-Yue Liu, Ge-Sheng Pan, Yong Wang, Zhao-Hui Li, Jin-Cai Wu, Yan-Yan Zhang, Fa-Xi Chen, Chao-Yang Lu, Sheng-Kai Liao, Juan Yin, Jian-Jun Jia, Cheng-Zhi Peng, Hai-Feng Jiang, Qiang Zhang, Jian-Wei Pan.

The manuscript describes a major breakthrough into the realization of free-space satellite-based long range optical time and frequency dissemination, with impacts for fundamental physics (search for dark matter, test of fundamental constant, test of relativity…). The major achievement of the authors as compare to previous work in the field is the operation of two 1W-power Optical Frequency Comb, that allows the authors to demonstrate world-record in range for free-space optical fiber links, while keeping state of the art in terms of precision. The experimental results are impressive, and the manuscript is very likely to be highly impacting, what desserves publication in Nature.

The manuscript describes other several improvements as compare to their previous paper published in 2021: authors described an improved electronics system and an improved interferometer system (length mismatch minimisation, better temperature stabilization) that allows them to reach lower noise floor stabilities below 1e-19, and larger link loss acceptance from 72 dB up to 90 dB, what is another impressive achievement. The authors reports for about 235 hours of operation acquired in 21 days, what is also an impressive achievement given the high loss acceptance of their system. The authors compare the results with an additional aerial bi-direcitional fiber link and show consistency in their results.

The manuscript is well written, expose clear explanations. Data and methodology are valid, the authors show high quality data, and appropriate treatment of uncertainties. The manuscript shows rigourous reasoning and robust results, certainly reliable and valid.

Figures are well presented generally speaking, aside of small mistakes I detailed later on in this review. The supplementary materials are technically sound and are very complete. Supplementary materials should provide expert readers with the necessary technical informations they might need, while the main text indeed address a broad audience.

Several weaknesses were pointed out by referees. The main one was about the two independent two-way the authors claimed. In this revised manuscript, the authors provide additional data based on an additional bidirectional fiber link, sharing the same ultra-stable laser. The revised figures 1,2 and 3 address the point. The new experimental results in the revised version are convincing.

Technical missing details and appropriate references were added to the revised manuscript.

In conclusion I recommend the manuscript for publication.

-- minor comments:
- line 445 : "electrics" should be 'electronics' ?
- line 448: "The beating signal is filtered with a tracking oscillator and measured with a K-K phase recorder." K-K phase recorder may not be known by non-expert reader. I suggest to replace with "dead-time free phase recorder" with, ideally, the model reference and the brand.

Author Rebuttals to First Revision:

Responses to the Reviewer Comments

Referees' comments:

Referee #1 (Remarks to the Author):

Comment 0:

The authors have substantially revised their manuscript. As noted in their reply, they too realized the issue of common-mode noise shortly after submission and must have immediately embarked on a very significant experimental effort to add the fiber link between the two sites. This effort led to measurement campaigns on 3/30, 4/03 and 5/09. It is very impressive they were able to accomplish this significant experimental effort over the spring and, most importantly, the additional data runs show excellent results (Fig. 3b). By adding the fiber link, they successfully address the fundamental problem with the initial submission regarding “truth”. In addition, they have made other clarifications to the manuscript, as well as including real time synchronization in the final methods section. I think the revised manuscript is definitely suitable for publication. I have two minor revisions they could consider listed below.

Reply:

We thank the referee for his/her time and efforts to review our manuscript. We are glad to see that the referee thinks the revised manuscript is suitable for publication.

Comment 1:

1) The only actual data traces appear in a small inset of Figure 3d. Could they add a supplemental figure that gives the full time traces for one example run (perhaps one of the later runs showing all four one-way time data and the fiber data)? This would help the reader understand the experiment as otherwise the figures are limited to the “statistics” (e.g. MDEVs) . Similarly, could they specify which data runs gave the MDEV curves of Figure 3d?

Reply:

We thank the referee for the comments.

A supplemental figure has been added in the Supplemental Information file, which gives the full time traces of one run, including all four one-way time data and the fiber data. The data run of 22/05/20 gives the insert and MDEV curves of Figure 3d. In addition, the corresponding system offset value is updated to be “$6.3 \times 10^{-20} \pm 3.4 \times 10^{-19}$” instead of the old value “$6.5 \times 10^{-20} \pm 3.5 \times 10^{-19}$”.

Revisions in the Supplemental Information file:

1. The Figure 11 is added, as shown below: “

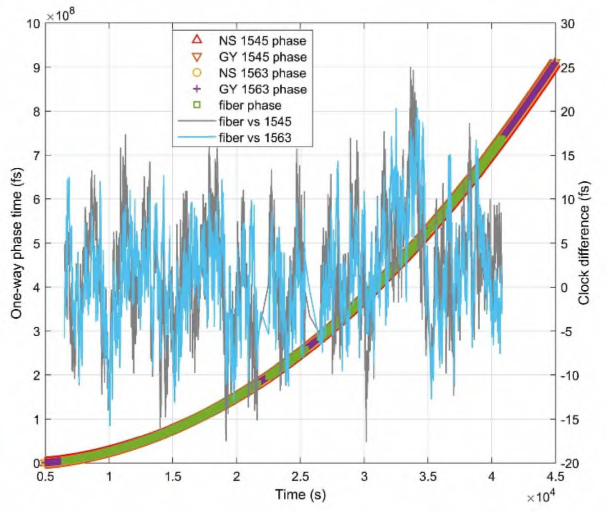


Figure 11. The time variations of one-way free-space links, where the time variation measured by the1545 nm optical combs in NanShan (NS) station is in red (up triangle), 1545 nm optical comb in Gaoyazi (GY) station (orange down triangle), 1563 nm optical comb in NS station (yellow circle) and 1563 nm optical comb in GY station (purple cross), the clock offset measured with the fiber link (green square, left axis). Difference of clock offset (marked with right axis) between 1545 nm optical comb and fiber link (gray line), 1563 nm optical comb and fiber link (light blue line) are shown in the figure as well.”

2. The section of “The original one-way free-space links and fiber link data” is added as below: “The time variations of the phase data for four one-way free-space links and the clock offset of the fiber link are shown in Fig.11, which is the 10~s averaged data of the raw data. The phase data obtained from the optical comb interferogram of the sampling system include the phase difference of two ultra-stable laser and the change of flight time. Because we use two independent ultra-stable lasers, a nearly linear drift can be observed for short terms, resulting in a quadratic curve of the clock offset ΔT. The change of flight time is several nanoseconds, which is much smaller than the accumulative time variation due to ultra-stable laser drift. Therefore, the phase curves of the one-way time-of-flight and the clock offset measured by the fiber link are indistinguishable. There are a few breaks for the fiber link, which is attributed to sudden large noise and poor signal-to-noise (SNR) of the fiber link due to 70% of the link fiber is aerial.”

Comment 2:

2) The revised manuscript is much clearer on the contributions to the link loss, particularly in the methods section. Some distilled version of this information could appear in the main text. For example, the abstract quotes “channel loss of 89 dB”, which could be read to mean just the free-space link loss but includes the ~8.8 fixed loss of the transmitter/receiver (“LOS optics loss” + telescope optical efficiency). This 8.8 dB total is actually quite low considering the complexity of the

system, but they might separate out this contribution either there or in the paragraph of 119-139, e.g. by mentioning the 74 dB/ 89 dB include that 8.8 dB. (The reason is for comparison to existing free-space optical comm systems or design of future free-space systems).

Reply:

We thank the referee for the comments.

The sentence of "The 89 dB average loss of the mis-aligned link is measured from output of the comb to input of the balanced detector, corresponding to loss of a 190 km link according to simulation. " is modified to

"The 89 dB average loss of the mis-aligned link is measured from output of the comb to input of the balanced detector, including ~8.8 dB fixed loss of telescope optical efficiency and LOS optics loss of the transmitter/receiver (Supplementary Information), which corresponds to loss of a 190 km link according to simulation."

Referee #2 (Remarks to the Author):

Comment 0:

Review of the manuscript entitled '113 km Free-Space Time-Frequency Dissemination at the 19th Decimal Instability',
by Qi Shen, Jian-Yu Guan, Ji-Gang Ren, Ting Zeng, Lei Hou, Min Li, Yuan, Cao, Jin-Jian Han, Meng-Zhe Lian, Yan-Wei Chen, Xin-Xin Peng, Shao-Mao Wang, Dan-Yang Zhu, Xi-Ping Shi, Zheng-Guo Wang, Ye Li, Wei-Yue Liu, Ge-Sheng Pan, Yong Wang, Zhao-Hui Li, Jin-Cai Wu, Yan-Yan Zhang, Fa-Xi Chen, Chao-Yang Lu, Sheng-Kai Liao, Juan Yin, Jian-Jun Jia, Cheng-Zhi Peng, Hai-Feng Jiang, Qiang Zhang, Jian-Wei Pan.
The manuscript describes a major breakthrough into the realization of free-space satellite-based long range optical time and frequency dissemination, with impacts for fundamental physics (search for dark matter, test of fundamental constant, test of relativity…). The major achievement of the authors as compare to previous work in the field is the operation of two 1W-power Optical Frequency Comb, that allows the authors to demonstrate world-record in range for free-space optical fiber links, while keeping state of the art in terms of precision. The experimental results are impressive, and the manuscript is very likely to be highly impacting, what desserves publication in Nature.
The manuscript describes other several improvements as compare to their previous paper published in 2021: authors described an improved electronics system and an improved interferometer system (length mismatch minimisation, better temperature stabilization) that allows them to reach lower noise floor stabilities below 1e-19, and larger link loss acceptance from 72 dB up to 90 dB, what is another impressive achievement. The authors reports for about 235 hours of operation acquired in 21 days, what is also an impressive achievement given the high loss acceptance of their system. The

authors compare the results with an additional aerial bi-direcitional fiber link and show consistency in their results.
The manuscript is well written, expose clear explanations. Data and methodology are valid, the authors show high quality data, and appropriate treatment of uncertainties. The manuscript shows rigourous reasoning and robust results, certainly reliable and valid.
Figures are well presented generally speaking, aside of small mistakes I detailed later on in this review. The supplementary materials are technically sound and are very complete. Supplementary materials should provide expert readers with the necessary technical informations they might need, while the main text indeed address a broad audience.
Several weaknesses were pointed out by referees. The main one was about the two independent two-way the authors claimed. In this revised manuscript, the authors provide additional data based on an additional bidirectional fiber link, sharing the same ultra-stable laser. The revised figures 1,2 and 3 address the point. The new experimental results in the revised version are convincing.
Technical missing details and appropriate references were added to the revised manuscript.
In conclusion I recommend the manuscript for publication.

Reply:

We thank the referee for his/her time and efforts to review our manuscript. We are glad to see that the referee recommends the manuscript for publication.

Comment 1:

-- minor comments:
- line 445 : "electrics" should be 'electronics' ?
- line 448: "The beating signal is filtered with a tracking oscillator and measured with a K-K phase recorder." K-K phase recorder may not be known by non-expert reader. I suggest to replace with "dead-time free phase recorder" with, ideally, the model reference and the brand.

Reply:

We thank the referee for the comments. We have revised the manuscript accordingly.