nature portfolio

Peer Review File

Ultrafast Floquet engineering of Fermi-polaron resonances in charge-tunable monolayer WSe_2 devices

Corresponding Author: Professor Hyunyong Choi

This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Attachments originally included by the reviewers as part of their assessment can be found at the end of this file.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)
In this paper, Hyojin Choi et. Al., explored the dynamics of out-of-equilibrium attractive and repulsive Fermi polarons in gate-tunable monolayer WSe2 using below-gap ultrafast photoexcitation. They examined both steady-state and transient optical spectroscopy, focusing on the resonance shift changes of both polarons as a function of Fermi level, EF,h. Notable shifts were observed for both types, with repulsive polarons showing a decrease and attractive polarons increasing resonance shift with higher Fermi-sea density, challenging the conventional dressed atom-photon model. The study concludes with a demonstration using the Chevy ansatz Hamiltonian model that circularly polarized light oppositely affects the Fermi sea in different valleys, reducing the binding energy of attractive polarons.
I believe this work is certainly of interest to the community and of potential high impact. However, I found a few issues that need to be addressed before the paper can be further considered for publication in Nature Communications. My concerns are listed below:

1. A key feature of Floquet engineering involves the band hybridization between the original Bloch states and photon-dressed ladder-like Floquet sidebands, which are spaced by integer multiples of the photon energy (should be 1.55 eV here). It is quite hard to understand how the optical modulated polaron peaks shifts are elated to Floquet engineering. The authors are recommended to present an illustration on how the periodic light field modulates the energy band structure of WSe2. Moreover, I believe it is necessary for the authors to clarify how the polaron peak shift phenomena observed in their experiment is attributed to Floquet physics and present a proper illustration on this.
2. In Fig. 3, the resonance shift of Fermi polarons is attributed to the change in binding energy. However, in many cases, the peak shift in TMDCs system is dominated by the energy band. How can the authors rule out the possible influence to the energy band structure induced by the Floquet engineering?
3. In many literature on Floquet engineering, Mid-Infrared (MIR) pump pulses are utilized to mitigate alternative dissipation channels and heating effects, ensuring that the spacing between energy levels is sufficiently small to be observable. However, in this work with “Floquet engineering” as its title, the authors need to clarify why these effects are minor with a high photon energy of 1.55 eV?
4. In many cases, the Floquet engineering is considered transient, namely in the case that the energy of the pump light is below the bandgap, when the pump light pulse ends, the transient Floquet engineering to band structure should be void. Consequently, one would expect a noticeable peak shift in the reflectance spectrum immediately following the termination of the pump pulses in Fig. 2 and Supplementary Fig. 3a. However, in Supplementary Figure 3a, the signal shows a long lifetime for 1.07 to 3.57 ps? In this case, how to understand the lifetime? It is imperative for the authors to engage in a more comprehensive discussion on the dynamic evolution of Fermi polarons with delay time, which I think is a key point of the paper.
5. In this work, the author only discussed the co-circular response to the pump light. My curiosity extends to the cross-circular scenario: in principle, should the opposite valley (σ-) also respond to the pump pulse (σ+) under Floquet Engineering? Incorporating a discussion on this aspect would undoubtedly enrich the understanding of the broader community.

Reviewer #2

(Remarks to the Author)
This study investigates the nonequilibrium dynamics of attractive and repulsive Fermi polarons in monolayer WSe2 devices using ultrafast Floquet engineering. The study demonstrates that the resonance shifts of Fermi polarons are valley-selective and reveals that the resonance shifts of attractive polarons increase with Fermi-sea density, while those of repulsive polarons decrease. The proposed interacting Hamiltonian based on Chevy ansatz effectively explains the observed experimental results. Overall, the research provides insights into the complex interactions of polarons in solid-state systems and suggests an interesting Hamiltonian to describe these phenomena. This paper provides a relatively substantial argument. It may be publishable in this Journal after a few responses to the questions below.

1 What are the potential limitations of using Floquet engineering in practical applications?

2 Are there other methods besides Floquet engineering that can be used to manipulate fermi polarons? What are the differences and advantages and disadvantages between them?

3 How do the results compare with other TMD materials or other 2D materials? Are there any material-specific effects that need to be considered?

4 How does temperature affect Fermi polaron dynamics? Do the results differ at higher or lower temperatures?

5 The theoretical model presented in the paper is based on the Chevy ansatz. Could you please discuss the limitations of this approximation and how it may affect the accuracy of the results?

Reviewer #3

(Remarks to the Author)
See attachment

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)
In the point-by-point responses, the author provided detailed explanations for the comments I raised and partly supplemented the relevant content in the main text. However, I still have one question. Regarding Comment 4, the author agrees that Floquet engineering induces the coherent signal, which means the signal only exists during the duration of the pump pulse. In the methods section of the manuscript, I noticed that the pulse width of the pump femtosecond pulses at 1.55 eV is about 40 fs, while in the point-by-point response, the author mentioned observing Stark light signals lasting about 300 fs. Why is the duration of this signal longer than the pulse width? I would appreciate further clarification on this point.

In a word, the revised version of the manuscript presents a systematic study on the ultrafast floquet engineering of Fermi-polaron resonances in charge-tunable monolayer WSe2 devices. I will be glad to recommend this paper for publication in Nature Communications after the issues I mentioned above are well addressed.

Reviewer #2

(Remarks to the Author)
After reading the reply, I think my concerns about this manuscript are well addressed and the manuscript has been revised accordingly. Hence, I recommend its publication.

Reviewer #3

(Remarks to the Author)
The authors have well addressed my comments and concerns. The manuscript is now in a much improved shape. I would like to recommend publication of this manuscript.

Point-by-point responses to the issues raised by the reviewers

General remarks and comments of Reviewer 1:

In this paper, Hyojin Choi et. Al., explored the dynamics of out-of-equilibrium attractive and repulsive Fermi polarons in gate-tunable monolayer WSe2 using below-gap ultrafast photoexcitation. They examined both steady-state and transient optical spectroscopy, focusing on the resonance shift changes of both polarons as a function of Fermi level, EF,h. Notable shifts were observed for both types, with repulsive polarons showing a decrease and attractive polarons increasing resonance shift with higher Fermi-sea density, challenging the conventional dressed atom-photon model. The study concludes with a demonstration using the Chevy ansatz Hamiltonian model that circularly polarized light oppositely affects the Fermi sea in different valleys, reducing the binding energy of attractive polarons.

I believe this work is certainly of interest to the community and of potential high impact. However, I found a few issues that need to be addressed before the paper can be further considered for publication in Nature Communications. My concerns are listed below:

Response 1:

We thank Reviewer 1 for taking the time to review our manuscript. We appreciate the Reviewer 1's agreement on the potential impact of our research. We also agree that providing more details on the Floquet engineering of Fermi polarons as well as the cross-circular scenario would be highly valuable to the 2D materials community.

While preparing our responses to the Reviewer 1's comments, we have carefully considered the points raised and have thoroughly revised our manuscript. We have added additional discussions regarding the Floquet engineering on the Fermi polarons to further support our main idea. We would also like to clarify that, in order to measure the large shift of the Fermi polarons through Floquet engineering, we select the pump pulse with a photon energy of 1.55 eV.

Below we present our point-by-point responses to the comments raised by Reviewer 1.

Comments 1-1: A key feature of Floquet engineering involves the band hybridization between the original Bloch states and photon-dressed ladder-like Floquet sidebands, which are spaced by integer multiples of the photon energy (should be 1.55 eV here). It is quite hard to understand how the optical modulated polaron peaks shifts are elated to Floquet engineering. The authors are recommended to present an illustration on how the periodic light field modulates the energy band structure of WSe2. Moreover, I believe it is necessary for the authors to clarify how the polaron peak shift phenomena observed in their experiment is attributed to Floquet physics and present a proper illustration on this.

Response 1-1: We appreciate the Reviewer 1's attention to the optical modulated polaron shifts due to the Floquet engineering. We fully agree with the key feature of Floquet engineering mentioned in the **Comments 1-1**, and we acknowledge that it would be instructive to provide appropriate illustrations on the energy band structure of WSe_2. For clearer explanation, we have included a proper illustration (Fig. R1) that shows the shifts of the conduction and the valence bands under off-resonant pump excitation (photon energy of 1.55 eV), highlighting the hybridization between the first Floquet formed by the pump and the original Bloch state.

As Reviewer 1 has correctly mentioned, the Floquet engineering refers to the band hybridization between the original (unperturbed) Bloch states and the photon-dressed ladder-like sidebands. In our work, we mainly consider the hybridization between the first Floquet quasistate generated by the pump and the original Bloch state. The reason for this consideration is two fold. First, the original Bloch state is hybridized with the first Floquet quasistate due to the valley-selective optical Stark effect. In monolayer TMDs, the difference of magnetic quantum number between the conduction and valence bands is ±1 [Science **346**, 6214 (2014)]. Following the optical selection rule (±1 changes of magnetic quantum number), the σ^--polarized (left-circularly) or the σ^+-polarized (right-circularly) pulse selectively generates excitons in the K valley or in the -K valley. When the σ^--polarized pump excitation occurs in the K valley, the first Floquet quasistate of valence band is hybridized with the conduction band of the same magnetic quantum number [Nat. Mater. **14**, 290-294 (2015)]. Second, the optical Stark shift is larger when the difference between the energy level of the Floquet quasistate and the original Bloch state is smaller [Nat. Mater. **14**, 290-294 (2015)]. Therefore, it is appropriate to consider the first Floquet quasistate of the valence band, which is the closest state with the conduction band among the other ladder-like Floquet sidebands. In our case, the energy level of the exciton state is 1.72 eV, while the pump-

photon energy is 1.55 eV. This choice leads us to consider only the first Floquet quasistate, rather than the entire photon-dressed ladder-like Floquet sidebands.

We would like to clarify that the shift of repulsive polarons follows the excitonic Stark effect, which is the representative effect induced by the Floquet engineering. As for the attractive polarons, the shift occurs due to the following two reasons: the excitonic Stark shift and the shift due to the reduced binding energy. In other words, the shift observed in the attractive polarons is a sum of these two effects. In fact, this distinguishes ours from the reported prior Stark effects [Nat. Mater. **14**, 290-294 (2015); Nat. Phys. **14**, 1092-1096 (2018); Nat. Mater. **18**, 1065-1070 (2019); Nat. Commun. **10**, 5539 (2019); Nat. Commun. **12**, 4530 (2021); Phys. Rev. Lett. **132**, 056901 (2024)].

The above points are discussed from line 102, page 5 in the revised manuscript. For the resonance shift of the repulsive polarons, it follows the properties of excitons both in the steady state and under the off-resonant pump excitation. The resonance shift of the repulsive polarons exhibits a blueshift with increasing $E_{\mathrm{F,h}}$ in the steady state (Supplementary Fig. 3e). These repulsive polarons, which primarily reflect the exciton characteristics, experience the Pauli blocking effect and the Coulomb screening by free charges. The increased hole density screens the electron-hole attractive Coulomb interactions in excitons, leading to a decrease in exciton binding energy. This reduction in binding energy results in the blueshift. Similar phenomena have been reported by Chernikov *et al*. [Phys. Rev. Lett. **115**, 126802 (2015)] and Lin *et al*. [npj 2D Mater. Appl. **3**, 46 (2019)]. Now, for the resonance shift of the repulsive polarons, it also follows the Stark shift of excitons under the off-resonant pump excitation. Similar to the excitonic Stark effect, the shift of the repulsive polarons arises when the Floquet quasistate of the valence band (Fig. R1a) induces the repulsive interaction with the conduction band (left-hand side of Fig. R1b). The same shift of the valence band induced by the Floquet quasistate of the conduction band occurs.

We wish to note that for the attractive polarons, however, the resonance shift differs from that of the repulsive polarons. The attractive polarons are regarded as bound states between excitons and carriers. There is an "additional" blueshift in the attractive polarons, which results in a larger overall shift compared to the repulsive polarons. We attribute this additional blueshift to the decreased binding energy of the attractive polarons induced by the below-gap pump excitation. Therefore, the shift in the attractive polarons can be interpreted as the sum of the shift from the conventional Stark model and the shift due to the reduced binding energy.

We illustrate the above processes in the right-hand side of Fig. R1b. Detailed explanations are as followings. (i) the Stark shift of excitons arises with the generation of Floquet quasistates, and (ii) the attractive polarons form through the reduced attractive interaction between the Stark-

shifted excitons and the surrounding carriers. The energy level of the Stark-shifted excitons is given by $E_X + \Delta_X$, where E_X is the energy level of excitons and Δ_X is the shift of the excitons. Next, these excitons experience the attractive interaction with the surrounding carriers, forming the attractive polarons with a reduced binding energy ΔE_{AP}; here "reduced" means that it is small compared to the steady-state binding energy. Consequently, with the pump excitation, the energy level of the attractive polaron becomes $E_X + \Delta_X - \Delta E_{AP}$, and the shift of attractive polaron is $E_X + \Delta_X - \Delta E_{AP} - E_{AP}$, where E_{AP} is the energy level of the attractive polaron in the steady state.

Therefore, the excitonic Stark effect contributes to the shifts in both the attractive and repulsive polarons, indicating that Fermi polaron shifts under the off-resonant pump excitation are influenced by Floquet engineering. We hope this response and the illustration resolve Reviewer 1's concerns.

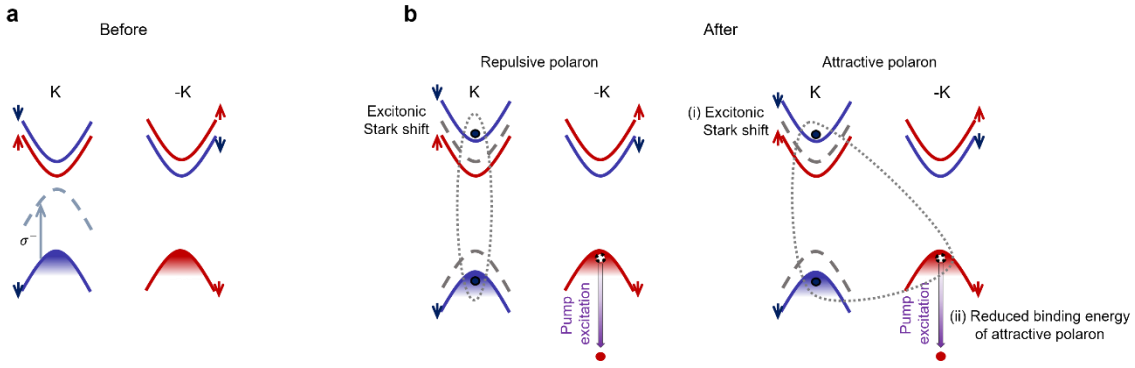

Figure R1 | Energy band structure of Fermi polarons. a Band structure with the pump-induced Floquet bands (blue dashed curves). Spin-up (-down) bands are shown in red (blue) color. Carriers in the spin-up (-down) state are represented by the solid red (blue) circle. **b** Left: The repulsive polaron state follows the excitonic Stark shift. The off-resonant pump excitation at the K valley induces a shift in the upper conduction band and the valence band. Right: Under off-resonant pump excitation, the attractive polaron shift occurs when (i) the Stark-shifted exciton (ii) interacts attractively with the Fermi sea in the opposite valley. The binding energy of the attractive polaron is smaller compared to that in the steady state due to the generation of hot carriers.

Comments 1-2: In Fig. 3, the resonance shift of Fermi polarons is attributed to the change in binding energy. However, in many cases, the peak shift in TMDCs system is dominated by the energy band. How can the authors rule out the possible influence to the energy band structure induced by the Floquet engineering?

Response 1-2: We acknowledge the Reviewer 1's comment that the peak shift in the TMDC system is typically dominated by changes in the energy band structure. Reviewer 1 raises a question regarding how we ruled out the possible influence on the energy band structure induced by the Floquet engineering. In this response, we clarify that the shift due to the reduced binding energy in the attractive polaron can be obtained by considering the Stark shift of the repulsive polaron.

Before addressing **Comments 1-2**, we wish to note that the shift observed in the repulsive polarons follows the excitonic Stark shift, while the shift in the attractive polarons is a sum of the excitonic Stark shift and the shift caused by the reduced binding energy, as detailed in our **Response 1-1**. This distinction allows us to determine the contribution of the reduced binding energy in the attractive polarons by subtracting the repulsive polaron shift from the total shift in the attractive polarons.

As shown in Fig. 3c of the main text, the difference between the shifts of the attractive and repulsive polarons, i.e., the shift due to the reduced binding energy, increases with Fermi energy. The reason is that the higher carrier densities, associated with increasing Fermi energy, result in a larger number of hot carriers generated by the off-resonant pump excitation. Further elaboration on this can be found in **Response 3-5**, where we show that the fractional factor (i.e. how much free carriers contribute to the hot-carrier generation) remains almost constant (approximately 0.3) with varying the carrier densities. Our calculations demonstrate that within the Fermi energy range of 0 meV to 20 meV, the density of states remains relatively constant, and consequently, the proportion of the hot carriers varies only slightly from 0.31 to 0.27 when the Fermi energy is tuned from 0 meV to 100 meV.

Comments 1-3: In many literature on Floquet engineering, Mid-Infrared (MIR) pump pulses are utilized to mitigate alternative dissipation channels and heating effects, ensuring that the spacing between energy levels is sufficiently small to be observable. However, in this work with "Floquet engineering" as its title, the authors need to clarify why these effects are minor with a high photon energy of 1.55 eV?

Response 1-3: We appreciate the Reviewer 1's attention on the photon energy of the off-resonant pump, where the pump-photon energy (1.55 eV) may be too high compared to low-energy (such as mid-IR or terahertz) pump cases. We agree with the Reviewer's insightful comment that the use of MIR pump pulses is to mitigate alternative dissipation channels and heating effects.

When the above-gap pump is employed, it leads to significant dissipation channels and heating effects because the pump directly excites electrons from the valence band to the conduction band. This results in a high density of electron-hole plasma, leading to a significant increase in the carrier population. These carriers can experience various relaxation pathways that introduce the energy dissipation, particularly through radiative recombination processes (such as photon emission) or non-radiative recombination (through heat dissipation via phonon interactions). This increase in the dissipation leads to a corresponding increase in system temperature, which can modify electronic properties, potentially causing bandgap renormalization [Ultrafast Spectroscopy of Semiconductors and Semiconductor Nanostructures (Springer, 1999); Phys. Rev. B **92**, 205415 (2015); Nano Lett. **17**, 4210-4216 (2017)].

As the carrier density increases, additional interactions between free carriers, such as the carrier-carrier scattering, become significant, further contributing to energy dissipation and complicating the photoexcited nonequilibrium phenomena of exciton or polaron dynamics [Phys. Rev. B **77**, 115121 (2008)]. These carriers rapidly lose energy, often through interactions with phonons, leading to lattice heating [ACS Nano **9**, 647-655 (2015)]. The thermalization of the carriers then occurs over picosecond timescales, as the carrier distribution reaches equilibrium with the lattice, further promoting local heating [Appl. Phys. Lett. **66**, 422-423 (1995)].

This increase in temperature leads to effects such as bandgap renormalization, which shrinks the effective bandgap due to the enhanced carrier-carrier interactions [J. Phys. Chem. C **128**, 2271-2290 (2024)]. Such band structure modifications can interfere with the Floquet engineering by introducing unwanted shifts in energy levels [Phys. Rev. B **92**, 205415 (2015)]. The magnitude of

these effects depends on the intensity of the pump pulse, with stronger pulses leading to more pronounced heating and dissipation channels.

One can minimize the alternative dissipation channels and the heating effects by employing the mid-infrared (MIR) pump pulse. The low-energy MIR pump pulses reduce the generating of free carriers or excitons that can lead to significant energy loss through recombination or scattering processes. Readers might have concerns about losing the advantages of using the MIR pump pulses due to the high photon energy of 1.55 eV in the experiments. We would like to clarify the reasons why these effects are minor and why we should use the pump-photon energy of 1.55 eV.

Firstly, the photon energy of the pump pulse (1.55 eV) is sufficiently below the exciton state (1.72 eV), minimizing the probability of generating real electron-hole pairs which are often associated with the energy dissipation. In this case, the exciton state is primarily influenced by virtual interactions, where the virtual excitons are momentarily created and do not lead to long-lived population or the recombination effects. This mitigates the heating effects and the alternative dissipation channels [Nat. Commun. **10**, 5539 (2019); Phys. Rev. Lett. **132**, 056901 (2024)].

Additionally, Fig. 3a in the main text demonstrates that the Stark shift of excitons is inversely proportional to the pump detuning, which is consistent with previous experimental observations of the excitonic Stark effect [Nat. Mater. **14**, 290-294 (2015); Nat. Commun. **12**, 4530 (2021)]. In these cases, the absence of Coulomb repulsion between the virtual excitons confirms not only that real excitons are not formed, but also that the population of the virtual excitons is small. This, in turn, confirms that significant dissipation or heating effects resulting from the virtual excitons are unlikely [Nat. Commun. **10**, 5539 (2019); Phys. Rev. Lett. 132, 056901 (2024)].

Lastly, the photon energy of pump pulse, 1.55 eV, is specifically chosen to induce a sufficiently large Stark effect. The Stark shift is inversely proportional to the energy difference between the photon energy of the pump pulse and the excitonic state, thus using a pump excitation (1.55 eV) near the excitonic state (1.721 eV) is necessary. Additionally, early results of ultrafast Stark spectroscopy [Nat. Mater. **14**, 290-294 (2015)] in monolayer WS_2 also have used a high-energy pump in the range of 1.71 eV to 1.91 eV, and have measured the Stark effect without the effects of alternative dissipation channels or the heating effects.

Comments 1-4: In many cases, the Floquet engineering is considered transient, namely in the case that the energy of the pump light is below the bandgap, when the pump light pulse ends, the transient Floquet engineering to band structure should be void. Consequently, one would expect a noticeable peak shift in the reflectance spectrum immediately following the termination of the pump pulses in Fig. 2 and Supplementary Fig. 3a. However, in Supplementary Figure 3a, the signal shows a long lifetime for 1.07 to 3.57 ps? In this case, how to understand the lifetime? It is imperative for the authors to engage in a more comprehensive discussion on the dynamic evolution of Fermi polarons with delay time, which I think is a key point of the paper.

Response 1-4: We agree that the Floquet engineering induces the coherent signal, which means that the signal should exist only during the duration of the pump pulse. We are grateful for the Reviewer 1's attention regarding the dynamics shown in Fig. 2 and Supplementary Fig. 3a. Regarding the comment on the long-term signal (1.07 to 3.57 ps) shown in Fig. 3a, we acknowledge that we should have provided more detailed explanations. We note that the experimental conditions between Supplementary Figs. 3a and 3b are different. We would like to clarify this point before proceeding.

In Supplementary Fig. 3a, we employ a below-gap pump (1.55 eV) to measure the optical Stark effects. There, we observe optical Stark signals lasting for approximately 300 fs. On the other hand, Supplementary Fig. 3b involves an above-gap pump at 3.1 eV. Because of the photoexcited "real" carriers, a long-term signal is observed. We note that the time scale on the x-axis is different between Supplementary Figs. 3a and 3b, which may cause the confusions.

As shown in Supplementary Fig. 3b, the above-gap (3.1 eV) excitation leads to the generation of highly energetic carriers. The photoexcited carriers experience multiple relaxation pathways. For the probe photon energy at 1.675 eV, which are near the attractive polaron resonance, the relaxation times are 2.68 ps to 2.51 ps at $E_{\mathrm{F,h}}$ of -12.7 meV and -27.9 meV, respectively. Such relaxation time becomes slightly shorter with increasing the hole density. These observations can be explained by considering the interactions between the attractive polarons and the free carriers. Upon initial hole injection, the photoexcitation impulsively enhances the attractive polaron absorption. However, with increasing the hole density (via electrostatic gating), the photo-induced free carriers scatter with the attractive polarons, resulting in a reduced relaxation time [Phys. Rev. Lett. **123**, 027401 (2019)]. For the repulsive polarons, we have measured 3.57 ps and 1.07 ps at $E_{\mathrm{F,h}}$ of 0 meV and -12.7 meV. The relaxation time also decreases because of the same reasons of the attractive polarons.

Comments 1-5: In this work, the author only discussed the co-circular response to the pump light. My curiosity extends to the cross-circular scenario: in principle, should the opposite valley (σ-) also respond to the pump pulse (σ+) under Floquet Engineering? Incorporating a discussion on this aspect would undoubtedly enrich the understanding of the broader community.

Response 1-5: We appreciate the Reviewer 1's insightful comment regarding the response of the σ^--probe (opposite valley) to the σ^+-pump. In the main text, we focus exclusively on the co-circular response to the pump excitation. As shown in Figs. 2c and 2d, signals due to shifts in the co-polarized system are present, while no signals are detected in the cross-circular scenario. Following the Reviewer 1's fruitful advice, we have carefully considered the cross-circular response and provide the reasons for the absence of signals.

When considering the optical Stark effect, the pump-induced probe changes are stronger when the co-circularly polarized pump-probe configuration is employed than the cross-polarized one. This observation is a characteristic feature of the valley-dependent optical selection rule [Nat. Mater. **14**, 290-294 (2015); Science **346**, 6214 (2014); Nat. Commun. **12**, 4530 (2021)]. Because the monolayer TMD exhibits energy degeneracy at K and -K point in the Brillouin zone protected by the time-reversal symmetry, the valley-selective magnetic moment (induced by the circular optical pulses) allows the ac Stark effect to break such valley degeneracy, leading to the effective shifting of the conduction and valence bands in a specific valley.

However, as the Reviewer 1 points out, the opposite valley also responds to the pump pulse. Here we briefly address this issue in the following and present the reasons why we do not need to consider those effects. There are two cases where one can observe the shift in the cross-circularly polarized pump-probe experiment induced by the Floquet engineering: the first is the biexcitonic Stark effect and the other is the Bloch-Siegert effect. For the case of biexcitonic Stark effects, both left and right circularly polarized pulses are necessary for the formation of biexcitons [Nat. Phys. **14**, 1092-1096 (2018)]. For instance, when the pump is right circularly polarized (say, this is coupled with the -K valley), it can lead to the formation of the Floquet quasistate of biexcitons, which then experiences a repulsive interaction with the exciton state in the K valley. This results in shifting the exciton energy level, providing transient Stark-like signals in the left (K) circularly polarized probe. Since this phenomenon was observed when the pump detuning is in the range of ~50 meV, such an effect can be neglected in our case; for our experiment, the pump detuning is 131~191 meV. The other case is the Bloch-Siegert effect. This effect arises when a Floquet quasistate causes an energy shift due to the repulsion interaction with an exciton state in an

opposite valley. [Science **355**, 6329 (2017)] has shown that a counter-rotating pump can generate the Floquet quasistate below the ground state. Due to the matching condition of the photon angular momentum, this repulsion between the Floquet quasistate and the exciton state in the opposite valley leads to the Bloch-Siegert effect. Thus, while the optical Stark effect occurs in the co-polarized configuration, the Bloch-Siegert effect can appear in the cross-polarized configuration. In principle, the optical Stark shift increases when the difference between the energy level of the unperturbed state and that of the pump energy is small. Conversely, the Bloch-Siegert shift becomes more significant when this energy difference is large. Therefore, the Bloch-Siegert shift is less likely to occur in our experiments.

General remarks and comments of Reviewer 2:

This study investigates the nonequilibrium dynamics of attractive and repulsive Fermi polarons in monolayer WSe2 devices using ultrafast Floquet engineering. The study demonstrates that the resonance shifts of Fermi polarons are valley-selective and reveals that the resonance shifts of attractive polarons increase with Fermi-sea density, while those of repulsive polarons decrease. The proposed interacting Hamiltonian based on Chevy ansatz effectively explains the observed experimental results. Overall, the research provides insights into the complex interactions of polarons in solid-state systems and suggests an interesting Hamiltonian to describe these phenomena. This paper provides a relatively substantial argument. It may be publishable in this Journal after a few responses to the questions below.

Response 2:

We appreciate Reviewer 2's time, consideration, and careful comments regarding our study. We thank Reviewer 2 for recognizing his/her insights into the complex interactions of polarons in monolayer WSe_2 devices. We would also like to express our sincere gratitude for the positive feedback regarding our observation and analysis. The insightful questions raised by the Reviewer 2 have allowed us to engage in deeper discussions, which will enhance the clarity and depth of our manuscript.

Below we present our point-by-point responses to the comments raised by Reviewer 2.

Comments 2-1: What are the potential limitations of using Floquet engineering in practical applications?

Response 2-1: We appreciate Reviewer 2's insightful question regarding the challenges associated with using Floquet engineering in practical applications. In our study, Floquet engineering is employed to dynamically modify the band structure, inducing shifts in both attractive and repulsive Fermi polarons. However, several limitations must be considered for the broader practical use.

Floquet engineering requires a strong external electromagnetic field to drive the material's electronic states. These fields are typically generated by intense laser sources, which can present challenges in practical applications due to the high energy requirements [Nature **600**, 235-239 (2021)]. Maintaining these strong fields over extended periods can be energy-intensive, and thus, scalability is a significant issue for devices requiring continuous or long-term operation.

Strong electromagnetic fields, particularly from high-intensity laser pulses, can lead to heating effects in the material. The absorption of high-energy photons can generate excess heat through phonon interactions [Phys. Rev. B **87**, 205130 (2013)]. Furthermore, laser-fragile materials, which are sensitive to high-intensity laser exposure, may degrade or suffer damage when subjected to such fields, limiting the range of materials that can be used in Floquet-engineered devices.

Floquet engineering relies on external driving fields to induce transient modifications in the material's electronic states. These modifications, typically referred to as Floquet quasistates, persist only as long as the driving field is applied. Once the external field is turned off, the system returns to its original state [Nature **614**, 75-80 (2023)]. Therefore, maintaining the Floquet-engineered states over long durations would require continuous application of the driving field, which may not be feasible for all practical applications.

Additionally, while this is not an inherent limitation of Floquet engineering, our study on Fermi polarons necessitates operation at cryogenic temperatures, which presents a limitation for the practical use. The ultra-low temperatures are essential for resolving the two distinct peaks of the attractive and repulsive polarons, whose small energy difference (21.7-30.6 meV) makes distinguishing them increasingly difficult at higher temperatures. As temperature increases, thermal broadening occurs, where the distribution of energies of the quasiparticles spreads due to interactions with phonons [Nat. Commun. **4**, 1474 (2013); Nanomaterials **13**, 2349 (2023)]. This broadening makes it difficult to resolve spectral features of Fermi polarons.

In conclusion, while Floquet engineering offers a novel way to control Fermi polarons and other quasiparticle dynamics, these limitations—especially the need for strong external fields and the laser-induced heating effects—could limit its practical scalability. Though Floquet engineering itself is not inherently limited by temperature, in our specific case, the ultra-low temperature is necessary to resolve the fine spectral features and to minimize broadening effects in the Fermi polaron spectrum. This requirement is relevant to our study but not a general constraint on Floquet engineering.

Comments 2-2: Are there other methods besides Floquet engineering that can be used to manipulate fermi polarons? What are the differences and advantages and disadvantages between them?

Response 2-2: We appreciate the Reviewer 2's question regarding alternative methods to manipulate Fermi polarons and how these methods can be compared to the Floquet engineering. In our study, we control the shift of Fermi polarons temporarily with the off-resonant pump excitation. In addition to the Floquet engineering, several techniques can influence Fermi polarons, including controlling the doping density, as demonstrated in Fig. 1c of our study, applying magnetic fields or employing multi-dimensional coherent spectroscopy.

First, varying the doping density (i.e. Fermi-sea density) is a well-established method that can modify the polaron spectral behavior. When one alters the carrier concentration in the steady state, various polaronic characteristics, such as energy levels and oscillator strengths of both attractive and repulsive polarons, are modified (see Supplementary Note 2 for more details) [Nat. Phys. **13**, 255 (2017); Phys. Rev. B **95**, 035417 (2017); Phys. Rev. B **103**, 075417 (2021)]. The resonance shifts of the attractive and repulsive polarons exhibit distinct features, such as shifts of Fermi polarons, increasing oscillator strength, and broadening width, with varying the doping density. Increasing the carrier concentration induces several effects, such as bandgap renormalization, Pauli blocking, and carrier-induced screening [Phys. Rev. Lett. **115**, 126802 (2015); J. Chem. Phys. **153**, 071101 (2020); Nat. Commun. **12**, 6131 (2021)]. These effects lead to a reduction in the binding energy of excitons, resulting in the blueshift and the decrease in the oscillator strength of the repulsive polarons. In contrast, the attractive polarons experience the redshift and the increased oscillator strength due to the stronger attractive interaction between the excitons and the carriers. Additionally, as discussed by D. Huang [Phys. Rev. X **13**, 011029 (2023)], two-dimensional coherent electronic spectroscopy reveals that the quantum dephasing of the attractive polarons remains nearly constant with increasing Fermi energy. In contrast, the dephasing rate of the repulsive polarons increases almost quadratically, resulting in a dramatic broadening of their spectral width (see Supplementary Note 2 for more details).

The second method involves applying magnetic fields, which modify the spin and valley degrees of freedom in two-dimensional materials like TMDs, enabling selective control over spin-polarized polarons [Phys. Rev. Lett. **123**, 097403 (2019)]. The magnetic field interacts with the intrinsic magnetic moments associated with the spin and orbital angular momentum of the electrons in the valence and conduction bands, resulting in the splitting of energy levels of excitons

and Fermi polarons. However, while magnetic fields provide a stable method to influence the polaronic behavior, they generally induce smaller band modifications compared to the Floquet engineering. For example, magnetic fields cause shifts of less than 1 meV in excitons of monolayer WSe_2 under a 7 T magnetic field [Nat. Phys. **11**, 148-152 (2015); Nat. Commun. **7**, 10643 (2016)]. Achieving a larger shift, such as 8 meV, in monolayer WS_2 requires much stronger fields, such as 30 T [Nano Lett. **12**, 7899-7904 (2016)].

We would like to highlight that while the Floquet engineering offers dynamic control over polarons by using periodic external fields, other techniques like multi-dimensional coherent spectroscopy have also shown potential in uncovering polaron-polaron interactions. For example, as discussed by Huang *et al*. [Nat. Commun. **13**, 10643 (2022)], the multi-dimensional coherent spectroscopy has been employed to probe interactions between the attractive polarons, revealing the existence of a bipolaron bound state with a large binding energy in electron-doped tungsten-based TMDs. These bound bipolaron states involve excitons from different valleys cooperatively bound to the same electron, indicating strong interactions between opposite-valley polarons, such as singlet polarons in the K valley and triplet polarons in the K' valley. Compared to the Floquet engineering, which modulates band structures, the multi-dimensional coherent spectroscopy offers a deeper understanding of homogeneous linewidths and resolves fine interactions within the system, although it is more complex in both experimental setup and data interpretation.

While doping density, magnetic fields, and multi-dimensional coherent spectroscopy offer alternative approaches to manipulate Fermi polarons, each method has its distinct advantages and limitations. Doping and magnetic fields provide stability and straightforward implementation but are less dynamic compared to Floquet engineering and typically induce smaller energy shifts. Multi-dimensional coherent spectroscopy, though more complex, reveals deeper polaronic interactions that are difficult to detect with other methods. On the other hand, Floquet engineering offers dynamic, real-time control over polaronic states and larger energy shifts but requires strong external electromagnetic fields and presents practical challenges, such as heating effects and spatial limitations. The choice of method ultimately depends on the specific experimental requirements and the desired level of control over the polaronic features.

Comments 2-3: How do the results compare with other TMD materials or other 2D materials? Are there any material-specific effects that need to be considered?

Response 2-3: We sincerely appreciate Reviewer 2's insightful inquiry regarding the comparison with other TMD materials or 2D materials. To address this, we would like to first explain the process of the shifts of Fermi polarons induced by the Floquet engineering and discuss the material-specific effects that influence these shifts.

As mentioned in **Response 1-1**, the shift in the repulsive polarons follows the excitonic Stark effect, while the shift observed in the attractive polarons is the sum of the excitonic Stark shift and the shift due to the reduced binding energy. Both of these effects can vary significantly depending on the material.

One key factor that determines differences in the excitonic Stark shift between materials is the oscillator strength of excitonic transitions (see Supplementary Note 6 for more details). The oscillator strength indicates how strongly excitons couple to light, which directly affects the magnitude of the Stark shift under the two-level approximation [Nat. Mater. **14**, 290-294 (2015)]. Materials with higher oscillator strengths exhibit stronger light-matter interactions, resulting in larger Stark shifts when subjected to an external electric field. For instance, in TMDs such as WSe_2, the strong Coulomb interactions in the electron-hole pairs and high exciton binding energies (e.g., 500 meV in monolayer WSe_2) [Nat. Mater. **13**, 1091-1095 (2014)] significantly enhance the oscillator strength. As a result, TMDs show larger Stark shifts compared to other materials with lower oscillator strength, such as bulk 3D semiconductors.

In contrast, other 2D materials, such as black phosphorus and halide perovskites, exhibit distinct oscillator strength behaviors. Black phosphorus shows smaller binding energy and thus exhibit less efficient light-matter coupling compared to TMDs [Phys. Rev. B **90**, 075434 (2014); Phys. Rev. B **91**, 155311 (2015)]. However, optical spectroscopy using halide perovskites demonstrates high oscillator strengths due to their direct bandgap and efficient light absorption, which can lead to even larger excitonic Stark shifts than those observed in TMDs [Nano Lett. **22**, 808-814 (2022)].

The shift caused by the reduction in the binding energy of the attractive polarons also varies among different materials. As discussed in the main text (line 186, page 9), the binding energy of Fermi polarons in the steady state is expressed as $\Delta E_{\mathrm{P}} = 23\ \mathrm{meV} + 1.3E_{\mathrm{F}}$, and the binding energy under the off-resonant pump excitation is expressed as $\Delta E_{\mathrm{P,pump}} = 23\ \mathrm{meV} + 1.3E_{\mathrm{F}}(1-\eta)$,

where η is a fractional factor accounting for the reduction in the binding energy. The material-specific properties, such as the slope of the binding energy in the steady state (1.3) and the fractional factor η, determine the magnitude of this reduction. As discussed by E. Liu [Nat. Commun. **13**, 6131 (2021)], the splitting energy between the attractive and repulsive polarons and its slope with respect to E_F vary even between TMDs due to differences in the interaction between excitons and the Fermi sea in the steady state. In WSe_2, this slope is larger than in $MoSe_2$, suggesting stronger exciton-Fermi sea interactions in WSe_2 [Nat. Commun. **12**, 6131 (2021)]. This stronger interaction can lead to a larger reduction in the binding energy of Fermi polarons in WSe_2 compared to $MoSe_2$.

For the parameter η, as discussed in the main text (line 162, page 8), the density of hot carriers generated by the pump excitation reduces the binding energy of the Fermi polarons. This hot carrier density is influenced by the fluence of the pump and the absorption coefficient as well as the dielectric response of the materials. For example, in the visible spectrum, MoS_2 has a refractive index of 4.5 to 5.0, while $MoSe_2$ exhibits values between 4.2 to 4.8. Additionally, WS_2 and WSe_2 show refractive indices of 3.8 to 4.8 and 4.0 to 5.0, respectively [Phys. Rev. B **90**, 205422 (2014); Appl. Phys. **105**, 201905 (2014)]. These differences in the refractive indices and the absorption coefficients indicate that the reduced binding energy of Fermi polarons would vary between TMD materials. Consequently, the shift in the Fermi polaron binding energy under the off-resonant pump excitation depends on the material-specific properties.

Comments 2-4: How does temperature affect Fermi polaron dynamics? Do the results differ at higher or lower temperatures?

Response 2-4: We sincerely appreciate Reviewer 2's insightful question regarding the effect of temperature on Fermi polaron dynamics. Temperature can have a significant impact on the system, primarily affecting two key aspects: band structure changes and binding energy reduction in the attractive polaron. Additionally, thermal broadening at higher temperatures makes it difficult to distinguish between the attractive and the repulsive polaron resonances.

At higher temperatures, bandgap shrinkage occurs, leading to a redshift of both the attractive and repulsive polarons, which effectively enhances the excitonic Stark shift. As the temperature increases, the increased number of phonons interacts with the electrons in the conduction and valence bands, causing the bandgap renormalization. This interaction lowers the conduction band energy and increases the valence band, i.e. shrinking the optical bandgap [Phys. Rev. Lett. **110**, 026803 (2013); Phys. Rev. B **93**, 205422 (2016)]. The reduced optical bandgap results in a redshift of the exciton state, decreasing the energy difference between the exciton and Floquet quasistates, thereby increasing the excitonic Stark shift [Nat. Mater. **14**, 290-294 (2015)]. For example, experiments on TMD Stark spectroscopy performed by LaMountain *et al.* [Nat. Commun. **12**, 4530 (2021)] have shown that the pump detuning at different temperatures affects the Stark shift magnitude. At T=10 K, the exciton peak energy is 1.757 eV, while at T=150 K, it shifts to 1.725 eV. Using a pump photon energy of 1.55 eV, the detuning decreases from 207 meV at 10 K to 175 meV at 150 K, resulting in approximately 1.2-fold increase in the Stark shift at higher temperature [Sci. Rep. **6**, 22414 (2016)].

The second effect of the increased temperature is the reduced binding energy of attractive polarons. As temperature increases, the electron-phonon scattering enhances, weakening the attractive Coulomb interactions between excitons and the surrounding carriers [Phys. Rev. B **93**, 075411 (2016); npj 2D Mater. Appl. **2**, 30 (2018); Phys. Rev. Lett **132**, 036903 (2024)]. In TMDs, the bandgap shrinkage dominates the shift of the Fermi polarons with varying the temperature, making it difficult to quantify the reduced binding energy. However, studies on CdSe nanoplatelets show a significant reduction in the attractive polaron binding energy, from 36 meV at low temperatures to 18 meV at higher temperatures [Nanoscale **12**, 14448-14458 (2020)]. This suggests that the binding energy of the attractive polarons decreases with increasing the temperature. Since the shift of the attractive polarons under the off-resonant pump excitation

depends on the changes in binding energy, variations in the temperature can lead to corresponding changes in the shift of the attractive polarons.

Another important temperature-dependent factor is the thermal broadening of the polaron resonances. At higher temperatures, the increased thermal energy leads to more carrier-phonon scattering, causing broadening of both attractive and repulsive polaron resonances [Phys. Rev. B **93**, 045407 (2016); Phys. Rev. Lett. **132**, 036903 (2024)]. As discussed by Ross *et al*., the linewidth of the attractive polarons is 5 meV at T=15 K, but the peaks become indistinguishable above 150 K in monolayer semiconductors [Nat. Commun. **4**, 1474 (2013)].

In summary, the Floquet engineering of Fermi polarons is strongly influenced by the lattice temperature. At higher temperatures, bandgap shrinkage may increase the excitonic Stark shift, but the binding energy of attractive polarons is smaller. Furthermore, thermal broadening makes it difficult to distinguish the polaron resonances. Therefore, conducting experiments at lower temperatures is advantageous for observing sharp and distinct shifts in Fermi polarons.

Comments 2-5: The theoretical model presented in the paper is based on the Chevy ansatz. Could you please discuss the limitations of this approximation and how it may affect the accuracy of the results?

Response 2-5: We thank Reviewer 2's comments regarding the limitations of the Chevy ansatz used in our study. The Chevy ansatz is a valuable approximation method for studying the polaron problem, where a single impurity interacts with a surrounding bath of particles, reducing the complexity by considering only a single particle-hole excitation [Nat. Phys. **13**, 255–261 (2017)]. In the context of the Fermi polaron problem, this approach has been highly successful in the regime of weak to intermediate coupling, providing an accurate description of key physical properties, such as the polaron energy shifts, which is consistent with both theoretical calculations and experimental results [Nat. Phys. **13**, 255–261 (2017)].

However, we agree with the Reviewer 2 that in systems where multiple particle-hole excitations become important, the Chevy ansatz may no longer be adequate for capturing the full many-body physics. This is particularly evident in the context of the temporal dynamics of the Fermi polarons, where the decay of repulsive polarons into attractive polarons and additional particle-hole pairs necessitates a more comprehensive treatment. As shown in [Nature **485**, 615–618 (2012)], the decay rate of repulsive polarons was calculated using numerical methods that account for all orders of particle-hole excitations, leading to a better match with experimental data. Given that below-gap excitations in our experiment may similarly drive the dynamical evolution of the Fermi polarons, we acknowledge that these effects are not fully captured by our current model and would require a higher-order excitation framework to be properly addressed.

Meanwhile, D. K. Efimkin et al., [Phys. Rev. B 103, 075417 (2021)] have reported similar energy shifts of Fermi polarons using two different forms of exciton-electron interactions: the contact interaction and the charge-dipole interaction approximations. In our work, we have employed the contact interaction approximation in calculating the self-energy [Eq. (2) in the main text]. Consequently, the observed energy shifts in our results would not significantly differ if the charge-dipole interaction approximation is used instead.

General remarks and comments of Reviewer 3:

In the manuscript titled "Ultrafast Floquet engineering of Fermi polaron resonances in charge tunable monolayer WSe2 devices", the authors have employed below-gap photoexcitation in the gate-tunable monolayer WSe2. Both attractive and repulsive Fermi polarons exhibit valley-selective resonance shifts in correlation with the pump intensity. The authors report that the dynamical shift of the repulsive polarons (RP) exhibits a decreasing feature, while that of the attractive polarons (AP) is strongly enhanced with increasing Fermi-sea density. While the paper somewhat extends the understanding of Fermi polaron system in condensed matters under below-gap optical field dressing, several key issues remain that the authors should address:

Response 3:

We sincerely appreciate Reviewer 3 for taking the time to carefully review our main text. We are grateful for his/her thoughtful comments and for acknowledging the extension of the understanding of the Fermi polaron system under below-gap optical field dressing in charge-tunable monolayer WSe_2 devices. We appreciate the Reviewer 3's attention to the valley-selective resonance shifts in both attractive and repulsive polarons as a function of the pump intensity, as well as the observed behavior where the repulsive polaron shift decreases and the attractive polaron shift is enhanced with increasing the Fermi-sea density.

We also recognize the important concerns regarding key aspects of our work. His/her insightful questions provide an opportunity to clarify several issues of the study, allowing us to analyze the pump's influence on hole dynamics more thoroughly and advance our theoretical model. This process not only improved the robustness of our approach but also helped to strengthen the overall conclusions of the study.

Below we present our point-by-point responses to the comments raised by Reviewer 3.

Comments 3-1: Mechanism of Pump Dressing: Since polarons are superpositions of excitons and holes (Fermi sea), the exciton components are expected to be affected by external field dressing. How does exciton-photon interaction contribute to the observed Stark shift in RP and the additional Stark shift in AP? The authors should clarify this. In addition, the authors have claimed that the binding energy of the AP is weakened by pump-induced "hot" holes (Line 161), which should be related to the photon-hole interactions. However, a quantitative clarification should be provided to demonstrate that photon-hole interactions are the primary drivers of the additional Stark shift, compared to exciton-photon interactions.

Response 3-1: We thanks Reviewer 3's insightful comments on the pump dressing and the role of exciton-photon/photon-hole interactions. Below we further clarify the associated mechanisms.

As mentioned in **Response 1-1**, the shift in the repulsive polarons follows the exciton-photon interaction, similar to the excitonic Stark effect, while the shift observed in the attractive polarons is more complex, where both the exciton-photon interaction and the photon-hole interaction are involved. Fermi polarons are indeed the superpositions of excitons and holes from the Fermi sea (in our case). Therefore, the exciton contribution is sensitive to the external-field dressing, such as the below-gap pump used in our experiment.

In the case of the Floquet engineering, the exciton-photon interaction induces the Stark shift by perturbing the exciton states, which directly shifts their energy levels. This explains the behavior observed in the repulsive polarons under the off-resonant pump excitation. However, the additional shift in the attractive polarons is primarily influenced by the photon-hole interaction. When the off-resonant pump excitation generates hot carriers, these carriers weaken the binding energy of the attractive polarons. The hot holes promoted to the deeper valence band reduce the Coulomb attraction between the exciton and the Fermi sea, thereby weaken the binding energy of the attractive polarons.

We acknowledge Reviewer 3's concern regarding the quantitative clarification of the photon-hole interaction. It is necessary to calculate the self-energy when the Chevy ansatz is applied in the simplified Hamiltonian; however, this approach does not directly account for the pump excitation within the system. Instead, we explain the effects of the pump by focusing on the reduction of the binding energy of the Fermi polarons, which provides an effective way of capturing the influence of the pump excitation. This simplification allows us to interpret the pump-induced dynamics, especially for the attractive polarons, while maintaining the clarity of our

theoretical model. As described in **Response 3-3**, this approach enables us to align the model with the observed experimental shifts. The reason we attribute the photon-hole interaction as the primary driver for the additional shift in the attractive polarons is due to the distinct nature of the attractive polarons compared to the repulsive polarons, in which the latter follows the excitonic Stark shift. Specifically, the attractive polarons form bound states between excitons and carriers, unlike the repulsive polarons, which primarily exhibit exciton-photon interactions. As seen in the steady-state regime, the attractive polarons experience a redshift with increasing Fermi energy (see Supplementary Note 2 for further details). When subjected to the off-resonant ultrafast pulse, electrons are excited from the deep valence band to the top valence band, as demonstrated in Arsenault *et al.* [Phys. Rev. Lett. **132**, 126501 (2024)]. This excitation primarily affects the attractive polarons by inducing shifts driven by hole dynamics, which are distinct from the exciton-photon interactions observed in the repulsive polaron.

Comments 3-2: The authors attribute the blueshift of AP to pump-induced "hot" holes. However, these photo-induced carriers should exhibit very long lifetimes, as shown in the Supplementary Note 3. This is very confusing.

Response 3-2: We are grateful for Reviewer 3's attention regarding the dynamics of the pump-induced carriers. Upon reviewing the Supplementary Note 3, we acknowledged that a more detailed explanation should have been provided. In this response, we clarify that while photo-induced carriers generated by above-gap pump excitation exhibit a very long lifetime of tens of picoseconds (Supplementary Fig. 3b), the hot carriers generated by the off-resonant pump excitation exhibit a shorter lifetime of a few hundreds of femtoseconds.

In experiments with off-resonant pump excitation (1.55 eV, Supplementary Fig. 3a), we observe that the signals last for approximately 300 fs. On the other hand, Supplementary Fig. 3b involves an above-gap pump of 3.1 eV. Because of the photoexcited "real" carriers, a long-term signal is observed. As shown in Supplementary Fig. 3b, the above-gap (3.1 eV) excitation leads to the generation of electrons in the high energy conduction bands. The photoexcited carriers experience multiple relaxation pathways. For the probe photon energy at 1.675 eV, which are near the attractive polaron resonance, the relaxation times are 2.68 ps to 2.51 ps at $E_{\mathrm{F,h}}$ of -12.7 meV and -27.9 meV, respectively.

Previous studies on above-gap pump excitation have consistently demonstrated that photo-induced carriers generated through this process exhibit relatively long relaxation dynamics. Following photoexcitation by an ultrafast pulse, the conventional semiconductor, e.g. undoped, direct bandgap, such as GaAs, experiences multiple nonequilibrium relaxation processes before returning to thermal equilibrium. After the photoexcitation, the probability redistribution occurs within the carrier system due to carrier-carrier scatterings, causing the initial nonthermal distribution to approach a thermalized Maxwellian distribution. This results in electron and hole temperatures that differ from the lattice temperature and sustains for around 200 fs or so. In the non-thermal regime, electron-hole collisions further thermalize the carrier system, and the electron temperature, representing the plasma temperature, remains higher than the lattice temperature. Depending on materials' specific heat, this phase can last for around 1 ps. The next stage involves the cooling of this plasma to the lattice temperature through interactions with phonons (mostly through optical phonons), occurring over a timescale of 1 ps to tens of ps. Ultimately, the photoexcited carriers reach thermal equilibrium with the lattice temperature and restore the semiconductor to its equilibrium state.

Unlike the free carrier dynamics in conventional semiconductors, the exciton dynamics exhibit a long time to decay, typically in the range of 6 ps to 200 ps, as reported in various studies. For examples, Damen *et al*. [Phys. Rev. B **42**, 7434 (1990)] demonstrate the formation of excitons in 20 ps, and relaxation of nonthermal excitons in 400 ps in GaAs. Similarly, studies by Blom *et al*. [Phys. Rev. Lett. **71**, 3878 (1993)], Eccleston *et al*. [Phys. Rev. B **45**, 11403 (1992)], and Vinattieri *et al*. [Solid State Commun. **88**, 189 (1993)] show the picosecond timescale of relaxation dynamics of photoexcited carriers in various semiconductors. Further investigations by Deveaud *et al*. [Phys. Rev. Lett. **67**, 2355 (1991)] and Vinattieri *et al*. [Phys. Rev. B **50**, 10868 (1994)] reveal that radiative recombination can be enhanced and the relaxation time can be decreased by altering temperature, excitation intensity, and applied electric field. Here because we have used off-resonant pump excitation in our experiments, we do not need to consider the time for formation or recombination of excitons.

In doped semiconductors, the relaxation time of free carriers decreases with the doping density. Studies in n-doped bulk GaAs, InP, GaAs/$Al_xGa_{1-x}As$ have shown that hole thermal equilibrium with the lattice is achieved within 400 fs to 700 fs, as reported by Chebira *et al*. [Phys. Rev. B **46**, 4559 (1992)], Zhou *et al*. [Phys. Rev. B **45**, 3886 (1992)], and Tomita *et al*. [Phys. Rev. B **48**, 5708 (1993)]. Additionally, investigations on p-type doped Ge indicate that hole thermalization occurs in approximately 700 fs, as reported by Woerner *et al*. [Phys. Rev. B **49**, 17007 (1994)] and Woerner *et al*. [Phys. Rev. B **51**, 17490 (1995)]. Interestingly, the relaxation dynamics of free carriers in the semi-metallic solids have shown that the excess charges significantly accelerate the relaxation rate. Lui *et al*. [Phys. Rev. Lett **105**, 127404 (2010)] demonstrated that electron thermalization in graphene occurs in a few tens of femtoseconds, and energy transfer from photoexcited electrons to optical phonons occurs within 200 fs. Theoretical work by Song *et al*. [Phys. Rev. B **87**, 155429 (2013)] supports this scenario, indicating that the time for photoexcited electron decay can be reduced to 120 fs due to enhanced intraband electron-electron scattering in the n-doped graphene.

The relaxation dynamics of free carriers in TMDs are reported to occur on a shorter timescale compared to conventional semiconductors as reported by Nie *et al*. [ACS Nano **8,** 10931–10940 (2014)], Wallauer *et al*. [Nano Lett. **21,** 5867-5873 (2021)] and Policht *et al*. [Nat. Commun. **14,** 7273 (2023)]. These studies indicate that the carrier thermalization occurs within 20 fs, and the cooling process occurs within approximately 0.6 ps under resonant pump excitation. Although the dynamics of photoexcited carriers in doped TMDs under off-resonant pump excitation remain unexplored, insights from prior reports (especially under resonant pump excitation in undoped TMDs and enhanced thermalization process in doped semiconductors

compared to undoped semiconductors) suggest that our case, i.e., the rapid relaxation in doped TMDs under off-resonant excitation, is likely to be on the tens of femtosecond scale. Additionally, because the off-resonant pump avoids the formation of excitons, and thus the absence of exciton recombination may result in an even shorter time compared to the resonant pump excitation.

Comments 3-3: Theoretical Model: Regarding the theory model, for the field-dressed Fermi polaron at time zero, the authors should include the pump photon and relavent interactions (at least the type that contributes to the Stark shift of RP), just like the Hamiltonian in Eq. (1) of Ref. [24]. From there, the self-energy averaged over one period of the pump could be derived from the Chevy Ansatz, and the Stark shift of RP and AP could be addressed.

Response 3-3: We thank Reviewer 3's insightful comment regarding the theoretical model. To clarify reviewer's concerns on the pump photon and relevant interactions, we note that the pump photon term in the Hamiltonian presented in Eq. (1) of Ref. [24] specifically accounts for the cavity modes. While the cavity mode, with resonance frequency ω_c, can hybridize with the exciton resonance under certain conditions, this scenario does not apply to our experiment. This is because Eq. (1) of Ref. [24] was used to explain the interaction between the Fermi polarons and the strongly-coupled cavity modes [Nat. Phys. **13**, 255-261 (2017), conducted by Prof. A. Imamoğlu group at ETH], whereas our experiments were performed without a cavity. Therefore, we start our response by explaining the Hamiltonian using the Chevy ansatz.

The Chevy ansatz is introduced to solve the problem of a single minority particle embedded in the Fermi sea of majority atoms. Below we outline the theoretical framework proposed by F. Chevy [Phys. Rev. A **74**, 063628 (2006)]. The Hamiltonian is written as

$$H = \sum_{k,\sigma} \epsilon_k \hat{a}^\dagger_{k,\sigma} \hat{a}_{k,\sigma} + \frac{g_b}{V} \sum_{k,k',q} \hat{a}^\dagger_{k+q,1} \hat{a}^\dagger_{k'-q,2} \hat{a}_{k',2} \hat{a}_{k,1}\,, \tag{S1}$$

where $\epsilon_k = \hbar^2 k^2/2m$, $\hat{a}_{k,\sigma}$ is the annihilation operator of a species σ particle with momentum k, quantization volume V, and g_b is the bare coupling constant characterizing interparticle interactions. To address the Fermi-polaron problems, a variational method inspired from the first-order perturbation theory is used, where the ground state of the system takes the following form:

$$|\psi\rangle = \phi_0 |FS\rangle + \sum_{k,q} \phi_{k,q} |k,q\rangle, \tag{S2}$$

where $|FS\rangle$ is a noninteracting majority Fermi sea plus a minority atom, and $|k,q\rangle$ is the perturbed Fermi sea with a majority atom with momentum q excited to momentum k. Minimizing $\langle H\rangle$ with respect to ϕ_0 and $\phi_{k,q}$ yields the following solutions:

$$E = \sum_{q<k_F} \frac{1}{\sum_{k>k_F}\left(\frac{1}{\epsilon_k + \epsilon_{q-k} - \epsilon_q - E} - \frac{1}{2\epsilon_k}\right) - \sum_{k<k_F}\frac{1}{2\epsilon_k}}. \quad \text{(S3)}$$

This procedure leads to a self-energy term Σ in our case, as demonstrated below:

$$\Sigma(E) = \sum_q \left[\frac{1}{V} - \sum_{k=k_F}^{\Omega} \frac{1}{E + i0^+ - \omega_{\mathrm{X}}(q-k) + \epsilon(k) - \epsilon(q)}\right]^{-1}. \quad \text{(S4)}$$

Here, $\frac{1}{V}$ is $\sum_{k=0}^{k_F} \frac{1}{\Delta E_{\mathrm{P}} - \omega_{\mathrm{X}}(0) + \omega_{\mathrm{X}}(k) + \epsilon(k)}$ and $i0^+$ is the small positive imaginary for the causality of the retarded Green's function. Up to this point, the pump excitation is not incorporated into the system. Instead, all effects of the pump can be effectively captured by introducing a fractional factor η to account for the reduced ΔE_{P}. This strategic choice is aimed at simplifying the problem while maintaining a clear physical interpretation. Specifically, it highlights how the below-gap excitation of the Fermi sea leads to a reduction in the binding energy.

When considering the effects of pump on the Fermi sea within the Hamiltonian, several terms emerge to account for various excitation and thermalization mechanisms, including light-matter, electron-phonon, and electron-electron interactions. While the Chevy ansatz has demonstrated a good accuracy in describing systems of Fermi polarons, the applicability may be limited when dealing with systems perturbed by light excitations. Although this approach requires thorough theoretical investigation, we show that our experimental results are well reproduced by the proposed theory when the pump excitation is phenomenologically considered by a fractional factor η.

Comments 3-4: Wavefunction and Oscillation Strength: The Fermi polaron state $|\Phi\rangle$ considered here, is a superposition of excitons, holes, and the pump photons (if dressed). The observed dynamic changes in reflectivity from the weak probe pulse should be related to the interband dipole transitions like $\langle\Phi|\hat{r}|\Phi\rangle$, where $\hat{r}$ represents the dipole operator. However, not all the wavefunction components of $|\Phi\rangle$ contribute equally to the observations, due to symmetry, particle number, or valley degree of freedom. Does the oscillation strength f_{osc} observed in the experiments plays a role analogous to that in the two-level system? Clarification on this point is crucial for making the argument about the "additional Stark shift of AP" based on f_{osc} differences and blueshifts. On the other hand, the simulated graph in Fig. 4a shows that at low $E_{\mathrm{F,h}}$, the "oscillation strength" of AP is stronger than RP, which conflicts with the results of Fig. 1c. The authors shall discuss this discrepancy, as well as the variance in spectral widths.

Response 3-4: We thank Reviewer 3's careful consideration and insightful questions regarding the role of the wavefunction and oscillation strength (f_{osc}) in the context of Fermi polaron states and the additional Stark shift in attractive polarons.

To address the first point, the Fermi polaron state $|\Phi\rangle$ is indeed a superposition of excitons and holes (see our **Response 3-1, 3-3**). The dynamic changes in reflectivity from the weak probe pulse are related to the interband dipole transitions, represented by $\langle\Phi|\hat{r}|\Phi\rangle$, where $\hat{r}$ is the dipole operator. In this case, not all components of the wavefunction contribute equally to the observable transitions. These factors influence the relative strengths of the dipole transitions that are accessible in the experiment.

In a two-level system, the Stark shift is typically proportional to the oscillator strength, meaning that the stronger light-matter interactions (i.e., a higher f_{osc}) result in the larger Stark shifts. In the case of the attractive polarons, there could be a misunderstanding that the increase in the Stark shift is simply due to the increase in the oscillator strength with increasing the Fermi energy. However, this does not fully explain the additional shift observed in the attractive polarons compared to the repulsive polarons and the excitons, despite their higher oscillator strengths (Supplementary Note 6). The two-level system model does not account for this disparity. Therefore, to explain the additional shift in the attractive polarons, we propose a mechanism involving the exciton-carrier bound state of the attractive polarons. Unlike the repulsive polarons, the attractive polarons involve stronger interactions between excitons and carriers. Under off-resonant pump excitation, the attractive polarons experience additional contributions from photon-carrier

interactions, particularly with hot carriers generated in the Fermi sea, leading to the observed additional shift.

To address the discrepancy between the simulated graph in Fig. 4a and the experimental data in Figs. 1c and 1d regarding the oscillator strength of attractive (AP) and repulsive polarons (RP), the difference stems primarily from the limitations of the model and the simplifications made during simulation.

The main objective of our model is to accurately capture the energy shifts of Fermi polarons rather than to fully resolve the oscillator strengths or spectral widths. The simple Hamiltonian used in this model, including the Chevy ansatz, is effective in predicting the energy levels, which are largely governed by the underlying interaction between excitons and the Fermi sea. However, Chevy ansatz simplifies the interactions, which in turn affects the accuracy in calculating the oscillator strength and the spectral width. In Fig. 1d, the oscillator strengths are derived from the steady-state reflection contrast measurements (Fig. 1c), while Fig. 4a represents a simulated result. In the well-established model, at low doping, the attractive polarons should have a lower oscillator strength than the repulsive polarons, which aligns with the experimental data [Nat. Phys. **13**, 255-261 (2017); Phys. Rev. Lett. **115**, 126802 (2015)]. However, due to the aforementioned simplifications in the simulation model, these oscillator strengths are not reproduced with the same accuracy as the energy shifts.

The discrepancy in oscillator strength and width arises because the simple Hamiltonian does not fully capture the many-body effects, such as carrier-carrier scattering and phonon interactions. As discussed in previous studies [Phys. Rev. B **103**, 075417 (2021); Nat. Commun. **12**, 6131 (2021)], these many-body interactions cause the spectral width to be broadened and modify the oscillator strength as the carrier density increases. Such effects are difficult to accurately simulate using a simple model like ours, which focuses more on energy-level shifts rather than subtle details of linewidth or oscillator strength.

The discrepancy between the oscillator strength and spectral width in Fig. 4a and the experimental data in Fig. 1c can be attributed to the limitations of the simplified model we used. While the model successfully captures the energy shifts of the Fermi polarons, it does not fully account for the many-body effects and scattering processes that influence oscillator strength and spectral broadening. Our primary focus was on reproducing the polaron energy shifts, and we believe that this was achieved with reasonable accuracy.

Comments 3-5: The authors introduced phenomenological shifts of polaron binding energy in the form of "$\Delta E_{P,pump} = 23$ meV + $1.3E_{F,h}$ $(1 - \eta)$ ", the effect of pump dressing is attributed to the η factor. However, this explanation is phenomenological and lacks validation..

Response 3-5: We sincerely apologize for the lack of validation in our previous submission. As mentioned in **Response 3-1**, instead of incorporating quantitative photon- hole interactions, we use a fractional factor η to account for the reduced binding energy of Fermi polarons. We assume that this fractional factor remains constant, independent of Fermi energy, and find that the model with a 30 % reduction in binding energy correlates with the experimental data. In our model, η of 0.3 implies that 70% of the doped holes are effectively bound to form attractive polarons, and this remains constant even when $E_{\mathrm{F,h}}$ is adjusted within the range from 0 meV to 20 meV.

For further clarification, we have calculated the fraction of hot holes, which possess the higher energy than the Fermi level, induced by the below-gap excitation. This phenomenon can be understood through the two-temperature model, e.g. [Sov. Phys. JETP **39**, 375-377 (1974); Heat Transfer **115**, 835-841 (1993); Int. J. Heat Mass Tran. **49**, 307-316]. According to this model, the electron temperature experiences a significant increase when the sample is subjected to a short pulse excitation, even when the pump-photon energy is below the band gap. Subsequently, the increased electron temperature decreases through the electron-phonon coupling, while the lattice temperature rises within a few picoseconds. However, if we focus on the femtosecond time scales, the electron-phonon coupling can be neglected.

The Fermi-Dirac distribution function, given by

$$f(E,T) = \frac{1}{e^{(E-\mu)/k_B T} + 1}, \tag{S5}$$

which describes the probability that fermions occupy energy states E, where μ is the chemical potential and T is the temperature. The carrier density $n(E)$ can be derived by multiplying the density of states $D(E)$ with the Fermi-Dirac distribution,

$$n(E) = D(E)f(E,T). \tag{S6}$$

In Fig. R2(a), the carrier density as a function of energy is depicted for temperatures $T = 4$ K and 400 K. We estimate η by calculating the proportion of the "thermalized" hot electrons to the total carrier density, obtained as, $\eta = \frac{A_1}{A_1 + A_2}$ (Fig. R2(b)). In Fig. R3, we show our analysis on the

change in η with different Fermi energies. We note that η varies from 0.31 to 0.27 when the Fermi energy is tuned from 0 meV to 100 meV. Within the experimental range of under 20 meV, however, η remains nearly constant. This is attributed to the density of state $D(E)$ being relatively unchanged within 20 meV. Consequently, we can assume that the fractional factor η remains constant within the range of Fermi levels in our experiments.

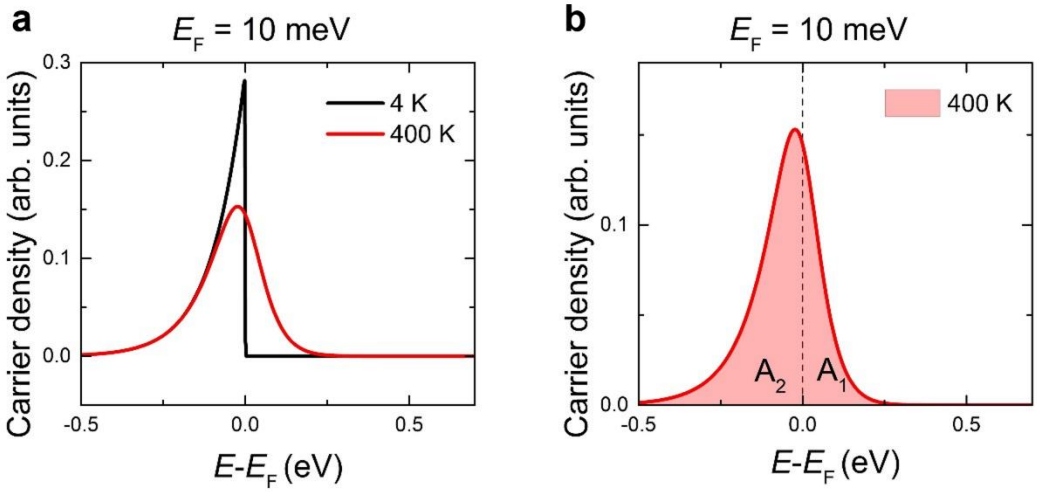

Figure R2 | a, The carrier density distribution at temperatures of 4 K (black) and 400 K (red) with the Fermi energy fixed at 10 meV in both cases. **b,** The carrier density distribution at 400 K, which mimics the thermalized distribution after the short pulse excitation. A vertical black dashed line represents $E = E_{\mathrm{F}}$. $\mathrm{A}_{1(2)}$ indicates the energy integration of carrier density above (below) the Fermi energy E_{F}.

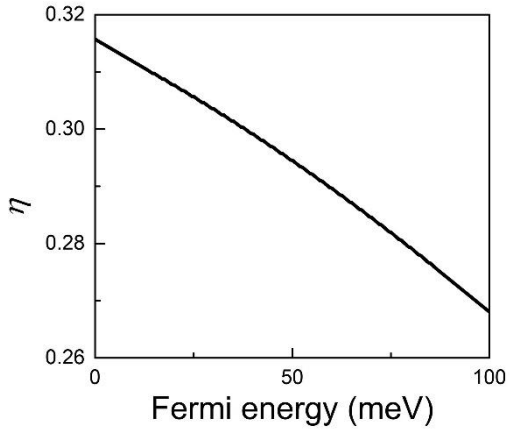

Figure R3 | The calculated fractional factor η, determined by the relation $\eta = \frac{A_1}{A_1+A_2}$, is depicted as a function of Fermi energy. It demonstrates a nearly constant value within the range of Fermi level from 0 meV to 20 meV.

Comments 3-6: The authors have used the phrase "nonequilibrium" In Line 24, 47, 60 and 76. In Floquet engineering, the system is dressed by a periodic field. Even though the system dynamically changes on sub-cycle timescales, it may not be accurate to refer to it as nonequilibrium".

Response 3-6: We sincerely thank the Reviewer 3 for the thoughtful comment regarding the use of the term "nonequilibrium". We understand that readers may be confused the phrase "nonequilibrium" when the Floquet engineering is applied. Following Reviewer 3's advice, we have revised the main text as following.

- Before [line 23, page 2, main text]: Here, we report the nonequilibrium light-driven dynamics of attractive and repulsive Fermi polarons in monolayer WSe_2 devices.
- After [line 24, page 2, revised main text]: Here, we report the light-driven dynamics of attractive and repulsive Fermi polarons in monolayer WSe_2 devices.
- Before [line 45, page 3, main text]: This may be partly because of the limited availability of rigid bosonic candidates in fermion-rich solids; even when a well-characterized solid is prepared, probing the out-of-equilibrium fine structures of the Fermi-polaron branches is experimentally challenging.
- After [line 46, page 3, revised main text]: This may be partly because of the limited availability of rigid bosonic candidates in fermion-rich solids; even when a well-characterized solid is prepared, probing the light-driven fine structures of the Fermi-polaron branches is experimentally challenging.
- Before [line 59, page 4, main text]: In principle, such strong light-matter interactions can dramatically turn the equilibrium properties into dynamical ones when driven into a nonequilibrium regime.
- After [line 60, page 4, revised main text]: In principle, such strong light-matter interactions can dramatically turn the equilibrium properties into dynamical ones.
- Before [line 75, page 4, main text]: Here, we employ the ultrafast Floquet engineering into the gate-tunable monolayer WSe_2 to investigate the nonequilibrium Fermi-polaron problems in presence of a fluctuating Fermi sea.

- After [line 76, page 4, revised main text]: Here, we employ the ultrafast Floquet engineering into the gate-tunable monolayer WSe_2 to investigate the light-driven Fermi-polaron problems in presence of a fluctuating Fermi sea.

We would like to explain our intention for using "nonequilibrium", so as to clarify the effect of below-gap short pulse excitation. First of all, in atomic physics, Floquet engineering is frequently associated with creating new quasi-eigenstates under the influence of a periodic driving field, which do not exist in equilibrium systems. These quasi-eigenstates exhibit phenomena such as dynamical localization or multiphoton absorption, which are impossible to observe under equilibrium conditions due to the lack of external periodic driving. This deviation from equilibrium occurs because the system continuously interacts with an external field that dresses the system's states dynamically [Phys. Rev. Lett. **107**, 25531 (2011); Nature **515**, 237 (2014); Nat. Phys. **12**, 350 (2016)]

Similarly, in solid-state systems, the Floquet engineering has been used to observe unconventional phenomena, including the light-induced topological states and Floquet-Bloch states, which emerge under strong, periodic driving fields. These states can significantly alter the band structure, often by breaking the spatial and time-reversal symmetries; all of which cannot be captured under equilibrium conditions [Nature **530**, 461 (2016); Phys. Rev. Lett. **117**, 227001 (2016); Nat. Phys. **16**, 38 (2020); Phys. Rev. Lett. **131**, 116401 (2023)].

In TMD materials, a representative example of the Floquet engineering is to demonstrate the valley-selective optical Stark shifts. These effects involve driving the system into a regime where the dynamics are dominated by periodic external fields rather than the intrinsic interactions of the system [Nat. Mater. **14**, 290 (2015); Nat. Phys. **14**, 1092-1096 (2018); Nat. Mater. **18**, 1065-1070 (2019); Nat. Commun. **10**, 5539 (2019); Nat. Commun. **12**, 4530 (2021)]. In this context, we have used the term "nonequilibrium" to reflect the fact that the observed Fermi polaron dynamics result from interactions induced by external pump fields, which are not present under equilibrium conditions.

In our study, the Floquet engineering generates resonance shifts of the Fermi polarons that would not occur in an equilibrium condition. The interaction between the excitons and the Fermi sea is modified by the coherently driven pump pulse. By creating non-trivial quasi-steady states, our study differs from the optical Stark effect, where the latter typically assumes the conventional two-level model. This distinction is the reason why we initially use the term "nonequilibrium" in the main text.

We acknowledge that the term “nonequilibrium” might have been too broad. After Reviewer 3’s comments, we agree that a more precise description should have been used. We hope that the above clarification resolves the Reviewer 3’s concerns.

Comments 3-7: The title uses the phrase "Ultrafast Floquet Engineering of XXX." As with the point above, Floquet engineering describes the dressing of a quantum system by a periodic field. If envelope effects are considered, a nonadiabatic Floquet theory should be applied.

Response 3-7: We sincerely appreciate Reviewer 3's comments regarding the use of the term "Ultrafast Floquet Engineering" in our title and the suggestion that nonadiabatic Floquet theory should be applied if envelope effects are considered. To address this point, we would like to clarify the distinction between adiabatic and nonadiabatic Floquet theory and explain our approach.

In a conventional Floquet engineering, a system's evolution depends on how quickly it responds to the external driving fields. Adiabatic Floquet theory assumes that the system evolves slowly enough that it stays close to its instantaneous eigenstate during the external field's driving period [Phys. Rev. A **95**, 023615 (2017)]. This is usually valid when the external driving field is relatively weak, or the detuning from the resonance is large, causing the system to evolve smoothly without significant changes or fast transitions. In such cases, the effects of Floquet engineering are dressing the original quantum states and shifting their energy levels coherently and predictably.

Nonadiabatic Floquet theory, on the other hand, applies when the external driving field is fast or strong enough to induce rapid changes in the system that cannot follow the field smoothly [Phys. Rev. A **81**, 013403 (2010); J. Phys. Chem. C **128**, 27 (2024)]. This is especially relevant when the pulse width is short or when the system's intrinsic timescales are comparable to or longer than the pulse duration. In such scenarios, the system cannot remain in the adiabatic regime, and transitions between different Floquet quasistates may occur, leading to more complex dynamics and non-trivial responses.

In principle, the nonadiabatic Floquet theory should be considered when pulse duration (~300 fs) is comparable to or shorter than the intrinsic timescales of the system. In TMD materials, exciton dynamics typically occur on the picosecond scale [Nat. Commun. **6**, 7372 (2015); Phys. Rev. B **101**, 041405(R) (2020)], which is quite slow compared to the femtosecond timescale of laser pulses used for our experiments. However, in our study, the observed shifts are relatively small (~5.5 meV) compared to the exciton energy level of 1.721 eV, and they occur smoothly over the duration of the pump. This suggests that the system remains relatively close to its original state, allowing for the adiabatic Floquet approximation to hold. For example, Sie *et al.* [Nat. Mater. **14**, 290-294 (2015)] employed an adiabatic Floquet theory to describe shifts in excitonic states, despite the short pump pulse. The weak field strength and the large detuning allowed the system to respond

adiabatically, as observed in their works. Similarly, in LaMountain *et al.* [Nat. Commun. **12**, 4530 (2021)] and Yong *et al.* [Nat. Phys. **14**, 1092-1096 (2018); Nat. Mater. **18**, 1065-1070 (2015)], an adiabatic Floquet engineering was used to explain small Stark shifts in transition metal dichalcogenides (TMDs), even under fast, sub-cycle driving fields.

While nonadiabatic Floquet theory is indeed crucial in many cases [Phys. Rev. X **6**, 021013 (2016); Phys. Rev. B **107**, 184314 (2023)], we believe that the adiabatic assumption remains valid in our study due to the small shifts (~5.5 meV) and the large detuning (~171 meV) from the resonance. Nonetheless, we recognize the importance of nonadiabatic effects and we hope this explanation clarifies our choice for using adiabatic Floquet theory.

Comments 3-8: In line 30, the authors claim that "circularly-polarized optical driving influences the Fermi sea in an opposite valley", but the significance of this effect remains unclear (even in Supplementary Figure 1(g)). The authors should elaborate on its importance.

Response 3-8: We sincerely apologize for the confusion caused by the unclear expression used in our previous submission. This sentence is critical to explaining the significance of our study, and we have revised the sentence as below.

- Before [line 29, page 2, main text]: A model Hamiltonian using Chevy ansatz suggests the circularly-polarized optical driving influences the Fermi sea in an opposite valley, thereby reducing the binding energy of attractive polarons.
- After [line 29, page 2, revised main text]: A model Hamiltonian using Chevy ansatz suggests the off-resonant pump excitation influences the free carriers that interact with excitons in an opposite valley, thereby reducing the binding energy of attractive polarons.

First of all, we would like to emphasize that the interaction between the exciton and Fermi sea under Floquet engineering is a key point in our study. The effects induced by Floquet engineering are particularly intriguing. To highlight the novelty of our study, we have extensively studied and looked into the long history of the Stark effects. For the categorization of such a broad aspect, please refer to Table R1 at the end of our response. Starting from the 1970s, the Stark effect was studied in atomic vapors by Liao and Bjorkhole [Phys. Rev. Lett. **34**, 1 (1975)] and Wu *et al*. [Phys. Rev. Lett. **38**, 1077 (1977)]. In 1985, the AC Stark effect was first observed in a semiconductor by Frohlich *et al*. [Phys. Rev. Lett. **55**, 1335 (1985)]. Together with the development of femtosecond laser technologies, this sparked the ultrafast optical studies of the ac Stark effect in quantum-well structures during the 1980s and 1990s, e.g. Mysyrowicz *et al*. [Phys. Rev. Lett. **56**, 2748 (1986)], Frohlich *et al*. [Phys. Rev. Lett. **59**, 1748 (1987)] and Sieh *et al*. [Phys. Rev. Lett. **82**, 3112 (1999)]. Such studies were later extended to TMDs in 2010s. Specifically, the connection of the optical Stark effect to the valley magnetic moment demonstrated the ultrafast lifting valley degeneracy by injecting circularly polarized light, which was shown by Sie *et al* [Nat. Mater. **14**, 290 (2015)] and Kim *et al* [Science **346**, 6214 (2014)].

Period	Journal	Discussion

1970s	Phys. Rev. Lett. **34**, 1 (1975)	Direct observation of optically induced shifts of atomic energy levels in sodium vapor that occur in two-photon absorption
	Phys. Rev. Lett. **38**, 1077 (1977)	The observation of optical amplification in a two-level atomic system driven by a strong, resonant field
1980s, 1990s	Phys. Rev. Lett. **55**, 1335 (1985)	First observation of the AC Stark effect in a semiconductor
	Phys. Rev. Lett. **56**, 2748 (1986)	High-energy shift of exciton resonances in GaAs multiple-quantum-well structures is observed with femtosecond nonresonant radiation
	Phys. Rev. Lett. **82**, 3112 (1999)	A redshift for opposite circularly polarized probe pulses due to memory effects in the Coulomb-induced excitonic correlations for low intensity pump pulses in InGaAs
2000s, 2010s, 2020s	Nat. Mater. **14**, 290 (2015)	First direct evidence of lifted valley degeneracy in the monolayer TMD WS_2 by applying circularly polarized light
	Nat. Phys. **14**, 1092-1096 (2018)	The exciton-biexciton coupling in monolayer $MoSe_2$ breaks the valley selection rules by applying off-resonant pulses
	Nat. Mater. **18**, 1065-1070 (2019)	Observation of the Berry phase-induced splitting of the 2p exciton states using the intraexciton optical Stark spectroscopy
	Nat. Commun. **10**, 5539 (2019)	Excitonic model of the optical Stark effect which includes many-body effects between virtual excitons

	Nat. Commun. **12**, 4530 (2021)	Valley-selective control of polariton energies using the optical Stark effect, extending coherent valley manipulation to the hybrid light-matter regime

Table R1 | Literature researches on the Stark effects.

Since then, further important observations have been reported in TMDs. For example, the Stark effect in exciton-polaritons was explored by Lamountain *et al*. [Nat. Commun. **12**, 4530 (2021)], where the authors have extended the coherent valley manipulation to the hybrid light-matter regime by using cavity structures, confirming that the Stark effect in exciton-polaritons follows the valley manipulation. Additionally, it exhibits a longer lifetime, i.e. the enhanced valley coherence, and the report further has shown that the shift can be controlled across a wider temperature range. On the theoretical side, there exist other interesting studies demonstrating how the conventional two-level model of the Stark effect is deviated when other excitonic complexes are involved. Examples include the effect of biexcitons by Yong *et al*. [Nat. Phys. **14**, 1092-1096 (2018)] and the 2*p* exciton by Yong *et al*. [Nat. Mater. **18**, 1065-1070 (2019)]. Although these studies show the "same" valley selective Stark effect, they reported unexplored phenomena such as exciton redshifts and splitting in cross-polarized pump-probe cases. Cunningham *et al*. [Nat. Commun. **10**, 5539 (2019)] has also challenged the conventional two-level model. The authors examined in details why the Stark shifts cannot be understood by a dressed atom-photon interaction model, especially when the pump-detuning is small. The conclusion of this study is that a quadratic term is involved, where many-body effects among virtually generated excitons play the key role in differentiating the near-resonant Stark effect from the far-detuned case.

In our study, we recognize that the interaction between excitons and the Fermi sea introduces new dynamics. Most previous studies of the Stark effect in TMDs have focused on undoped materials and the shift of the "original" exciton resonance due to the hybridization between the unperturbed Bloch state and the light-driven Floquet quasistate, as reported by Sie *et al.* [Nat. Mater. **14**, 290 (2015)] and Yong at al. [Nat. Phys. **14**, 1092-1096 (2018); Nat. Mater. **18**, 1065-1070 (2019)]. On the contrary, our work investigates the shifts in Fermi polarons in the doped TMD monolayers and reveals the effect of the off-resonant pump excitation, where the primary effect is to modify the interaction between the excitons and the Fermi sea.

We understand that the comment by the Reviewer 3 may intend to reflect this interaction. As noted in Supplementary Note 1, within the hole-doped regime, when an exciton is formed in the K valley, it interacts with the carriers in the opposite valley (e.g., -K valley). This interaction can

be either attractive or repulsive, leading to the formation of attractive or repulsive polarons (as shown in Supplementary Fig. 1g). The sentence “circularly-polarized optical driving influences the Fermi sea in an opposite valley” has been intended to suggest that, the circularly-polarized excitation (e.g., σ^+-polarized pump pulses, which couple with the K valley) influences the free carriers in an opposite valley (e.g., -K valley). The free carriers in -K valley interact with the excitons in K valley. In the main text, we would like to explain that under the off-resonant pump excitation, the generation of hot carriers influences the interaction between the excitons in one valley and the free carriers in another valley. This process leads to the reduction in the binding energy of the attractive polarons. Such an effect results in a larger shift for the attractive polarons, as demonstrated in our data shown in Figs. 3b and 3c.

Comments 3-9: In line 52, the "Fermi-sea density E_F" is used. While Fermi-sea density is related to the Fermi energy, they are not directly interchangeable. Consider using "Fermi-energy E_F" instead?

Response 3-9: We sincerely appreciate Reviewer 3's insightful comment regarding the term "Fermi-sea density E_F." We agree that "Fermi-sea density" and "Fermi energy" are distinct terms and should not be used interchangeably. We recognize that the term "Fermi-sea density" may lead to confusion since it relates to the carrier concentration, while Fermi energy (E_F) refers to the highest energy level occupied by fermions at absorlute zero temperature.

To clarify this distinction and avoid potential misunderstanding, we have revised the manuscript to use "Fermi energy E_F" in place of "Fermi sea density" as below.

- Before [line 51, page 3, main text]: Along with the atom-like bosonic impurity of excitons, the ability to electrostatically tune the Fermi-sea density E_F offers a way to reach a strongly interacting regime of single mobile boson with a fluctuating Fermi sea.
- After [line 52, page 3, revised main text]: Along with the atom-like bosonic impurity of excitons, the ability to electrostatically tune the Fermi energy E_F offers a way to reach a strongly interacting regime of single mobile boson with a fluctuating Fermi sea.
- Before [line 133, page 7, main text]: Based on the above aspects, one can anticipate $\Delta R/R_0$ dynamics to exhibit a strong dependence of the Fermi-sea density.
- After [line 134, page 7, revised main text]: Based on the above aspects, one can anticipate $\Delta R/R_0$ dynamics to exhibit a strong dependence of the Fermi energy.
- Before [line 159, page 8, main text]: Thus, increasing the Fermi-sea density leads to a monotonically decreasing Δ_P feature.
- After [line 161, page 8, revised main text]: Thus, increasing the Fermi energy leads to a monotonically decreasing Δ_P feature.

Comments 3-10: In line 110-112, the authors mention "to prevent any band-to-band population...171 meV ". However, in the experiment data shown in Fig. 2a and 2b, there are still several $\Delta R/R_0$ features when the time delay exceeds 0.2ps (albeit much smaller than at zero delay and different from above-gap excitations). Given the pump field's strength and pulse duration, how many carriers or excitons could be excited? Is their density comparable to that of the holes induced at finite $E_{\mathrm{F},h}$?

Response 3-10: We appreciate Reviewer 3's insightful question regarding the nature of the signal observed in Figs. 2a and 2b, which persists for more than 0.2 ps. We acknowledge Reviewer 3's inquiry that the signal persists for longer than the duration of the pump pulse, which might lead to the interpretation of real carriers or exciton excitation. However, as mentioned in the main text, we employ a below-gap pump excitation with a photon energy of 1.55 eV, resulting in a detuning of 171 meV from the exciton resonance, specifically designed to avoid band-to-band transitions and prevent the real carrier excitation. In this response, we would like to clarify that the large detuning prevents not only band-to-band transitions but also any the Coulomb repulsion between virtual excitons, which typically occurs at smaller detuning values.

To address that no band-to-band transition occurred under the pump excitation we used, we would first like to distinguish between above-gap and below-gap excitation, which directly affects the nature of the carrier generation. In the case of the above-gap excitation, band-to-band transitions occur, generating the real carriers [Nat. Commun. **6**, 7372 (2015); Phys. Rev. B **101**, 041405(R) (2020)]. In Supplementary Note 3, where the pump-pulse energy is 3.1 eV, we observe the generation of the photo-induced carriers with the long relaxation time (1.07 – 3.57 ps), an evidence of real carrier excitation through above-gap pumping. In contrast, below-gap excitation, such as in the main text and in Supplementary Fig. 2a, does not result in the real carrier generation since the photon energy of the pump pulse is insufficient to excite electrons directly from the valence to the conduction band. Instead, the observed effect mainly arises from the optical Stark effect, which is a coherent interaction between the pump field and the exciton states [Nat. Mater. **14**, 290-294 (2015); Nat. Commun. **12**, 4530 (2021); Nat. Phys. **14**, 1092-1096 (2018); Nat. Mater. **18**, 1065-1070 (2015)]. As a result, no excitons or free carriers are generated during the below-gap excitation process.

The long-lasting signal observed in Fig. 2a and 2b is due to the pump pulse duration rather than real carrier generation. We sincerely apologize for any confusion caused by our incomplete description of the pump pulse duration, which may have led readers to believe that the observed

signal persisted beyond the actual pulse duration. While we initially aimed to maintain the short pulse duration of around 40 fs after GVD compensation using prism pairs in the pump path, we employed the bandpass filters (width = 10 nm) to ensure the spectral purity. This filter increased the pulse width to approximately 270 fs. Consequently, the pulse duration of the pump field is extended, and the coherent signal becomes to persist for the longer period, explaining the extended signal beyond 0.2 ps. Similar effects have been observed in various studies, including that of Yong *et al.* [Nat. Phys. **14**, 1092-1096 (2018)], where despite the Ti:sapphire laser oscillator supplying 100 fs pulses, the overall temporal resolution of the measurement was nearly 1 ps. This resulted in the persistence of the Stark signal for over 1 ps.

Therefore, the persistent signal observed is not due to the excitation of the real carriers or excitons but rather the result of the extended pulse duration and the coherent Stark effect. We apologize for the confusion caused by this and appreciate Reviewer 3's attention to this matter.

Comments 3-11: The term “virtual exciton effect” is used without a clear explanation. The authors should provide a more detailed clarification.

Response 3-11: We sincerely appreciate Reviewer 3’s attention to the term "virtual exciton effect" used in the manuscript. To clarify this term, we would like to provide a more detailed explanation.

The "virtual exciton effect" refers to the temporary formation of excitons during off-resonant pump excitation, where the photon energy is insufficient to create real excitons. Instead, virtual excitons arise due to the interaction between the external field (pump pulse) and the material’s electronic structure. In the specific case of a 1.55 eV pump pulse, this energy is below the exciton resonance of 1.721 eV, which means no real exciton or full electron-hole pairs are generated. However, the electric field of the pump momentarily perturbs the system, inducing short-lived excitonic states that can affect the system's energy levels and band structure during the pump's duration. This phenomenon has been discussed in studies such as [arXiv: 2402.16630], where the virtual excitons significantly influence the optical properties of materials, even without generating real excitons. These transient virtual states can cause notable shifts in the system’s spectral features. In fact, these virtual states have also been observed in early studies in III-V quantum wells, where the effects induce nonlinear optical behaviors, particularly under off-resonant excitation conditions [Phys. Rev. Lett. **62**, 1189 (1989)]. The interaction between these virtual excitons and the real excitonic states leads to the phenomenon we refer to as the optical Stark effect. Furthermore, virtual excitons themselves can also exhibit repulsion, contributing to additional resonance shifts in the system.

The optical Stark effect is a well-known manifestation of the interaction between real excitons and virtual excitons with the external field. In this case, the virtual excitons experience repulsion from the real exciton state, leading to a shift in the energy levels. The shift observed due to the optical Stark effect is typically proportional to the intensity of the pump field and inversely proportional to the detuning between the pump photon energy and the exciton resonance. In early days of 1980s, Knox *et al.* [Phys. Rev. Lett. **62**, 1189 (1989)] and Schmitt-Rink and Chemla [Phys. Rev. Lett. **57**, 2752-2755 (1986)] demonstrated similar Stark shifts under strong electric fields. These studies revealed that the magnitude of the Stark effect is stronger dependent on the detuning, with smaller detuning producing larger shifts in energy levels [other references are in J. Shah, “Ultrafast Spectroscopy of Semiconductors and Semiconductor Nanostructures” (Springer. 1999)].

Additionally, the virtual excitons can also interact with each other, leading to phenomena such as the Coulomb repulsion between virtual excitons. This repulsion arises because even though virtual excitons are transient and not fully formed electron-hole pairs, they still exhibit Coulomb interactions. When two virtual excitons are momentarily induced within close proximity, their Coulomb repulsion can create additional shifts in the excitonic energy levels [Phys. Rev. Lett. **61**, 117-120 (1988); Nat. Commun. **10**, 5539 (2019)]. The magnitude of this repulsion is proportional to the intensity of the external field and inversely proportional to the square of the detuning between the photon energy of the pump and the exciton resonance. This quadratic dependence on detuning makes the effect more pronounced when the detuning is small. [Phys. Rev. Lett. **132**, 056901 (2024); arXiv: 2402.16630].

In our experiment (Fig. 3a), we note that the detuning of the pump is relatively large (151-171 meV), meaning that the effects of virtual exciton-to-exciton repulsion are negligible. This observation supports our discussion that the additional shift shown in the attractive polarons cannot be attributed to the interaction between virtual excitons. Instead, we believe that the additional shift arises from photon-hole interactions, as discussed in detail in **Response 1-1**.

We hope this clarification resolves any confusion regarding the term "virtual exciton effect." Thank you for the opportunity to explain this concept in greater depth, and we have revised the main text to ensure clarity as below.

- Before [line 135, page 7, main text]: If the exciton resonance shift scales inversely with δ_{X}^2, it can be accounted for by the resonant “virtual” excitonic effects, which are known to be dominant when $\delta_X \ll \Delta E_{\mathrm{X}}$.

- After [line 136, page 7, revised main text]: If the exciton resonance shift scales inversely with δ_{X}^2, it can be accounted for by the Coulomb repulsion between virtual excitons, which are known to be dominant when $\delta_X \ll \Delta E_{\mathrm{X}}$.

- Before [line 146, page 7, main text]: Having confirmed that the virtual exciton effect is excluded, we show in Fig. 3b the resonance shifts Δ_{P} of both attractive polarons and repulsive polarons as a function of pump fluence F.

- After [line 147, page 7, revised main text]: Having confirmed that the Coulomb repulsion between the virtual excitons is excluded, we show in Fig. 3b the resonance shifts Δ_{P} of both attractive polarons and repulsive polarons as a function of pump fluence F.

Point-by-point responses to the issues raised by the reviewers

General remarks and comments of Reviewer 1:

In the point-by-point responses, the author provided detailed explanations for the comments I raised and partly supplemented the relevant content in the main text. However, I still have one question. Regarding Comment 4, the author agrees that Floquet engineering induces the coherent signal, which means the signal only exists during the duration of the pump pulse. In the methods section of the manuscript, I noticed that the pulse width of the pump femtosecond pulses at 1.55 eV is about 40 fs, while in the point-by-point response, the author mentioned observing Stark light signals lasting about 300 fs. Why is the duration of this signal longer than the pulse width? I would appreciate further clarification on this point.

In a word, the revised version of the manuscript presents a systematic study on the ultrafast floquet engineering of Fermi-polaron resonances in charge-tunable monolayer WSe2 devices. I will be glad to recommend this paper for publication in Nature Communications after the issues I mentioned above are well addressed.

Response 1:

First of all, we appreciate Reviewer 1's positive feedback and support for our study on ultrafast Floquet engineering in WSe_2 devices. We are very grateful for his/her positive recommendation toward publication in Nature Communications. We also acknowledge that there are remaining issues raised by Reviewer 1: the coherent nature of the Stark effect signal and the pulse width. We would like to provide a detailed explanation of this in the response below.

Regarding the nature of the optical Stark effect signal, Reviewer 1 is absolutely correct that the optical Stark effect signal is coherent, and such an effect exists only within the duration of the pump pulse. We acknowledge Reviewer 1’s inquiry into the discrepancy between the Floquet engineering-induced coherent signal and the observed around 300 fs duration of the optical Stark effect signal. We apologize for the confusion caused by our incomplete description of the pump pulse duration. The pump pulse duration at the sample is, in fact, measured to be around 300 fs (as being outlined below) assuming a Gaussian pulse shape.

Figure R1 shows the measured pump-pulse width using a second-order nonlinear autocorrelator, where the width is initially 48.3 fs (right after the amplifier output, Fig. R1a). As the pulse passes through various optical components, it reaches 103.9 fs after prism compensation (Fig. R1b). We typically obtain the pulse width of 258.9 fs after the 10-nm spectral bandpass filter when measured before the objective lens (Fig. R1c). Group-delay dispersion (GDD) imposed by several optical elements (lenses, linear polarizers, half-wave plates, quarter-wave plates, and etc.) can be readily included when we compare the measured pulse width and the estimated one. Using a pair of two prisms, where a negative GDD of -8763 fs^2 (calculated one) is imposed, the pulse width reaches 103.9 fs (Fig. R1b). For the probe pulse width, we estimate that it is around 120 fs after generating the white-light continuum, which is subsequently recompressed using a two-prism pair. Such a width is obtained while monitoring the stability of white-light continuum.

Here, we wish to note that we have used a band-pass filter (center wavelength of 800 nm with 10 nm bandwidth) to achieve the spectral pump detuning. This filter, while essential for the detuning control, makes spectral narrowing, and thereby increase the pump pulse width to 258.9 fs (Fig. R1c). Subsequently, the pump pulse passed through the objective lens, which has a positive GDD of 1476.6 fs^2, resulting in a calculated pulse width of 266.7 fs at the sample. Thus, the pump pulse entering the sample was close to 270 fs, consistent with the observed Stark effect signal duration. Because the probe pulse is spectrally broader and temporally longer than the pump, the effect of objective GDD should be less than the pump.

Additionally, similar effects have been observed in various studies, including that of Yong *et al.* [Nat. Phys. **14**, 1092-1096 (2018)], where despite the Ti:sapphire laser oscillator supplying 100 fs pulses, the overall temporal resolution of the measurement was nearly 1 ps. This resulted in the persistence of the Stark signal for over 1 ps, highlighting how temporal broadening through optical elements and filters can significantly affect the duration of the coherent signal.

We appreciate the opportunity to clarify these details, and we hope this explanation addresses Reviewer 1's question thoroughly. We thank Reviewer 1 again for his/her critical comments, and we have revised the main text as below.

Before [line 239, page 12, main text]: Pairs of prisms (Thorlabs SF10) are inserted into both pump and probe paths to compensate the dispersion caused by the presence of dispersive optical elements.

After [line 239, page 12, revised main text]: A pair of prisms (Thorlabs SF10) are inserted into both pump and probe paths to compensate the dispersion caused by the presence of dispersive optical elements. The overall temporal resolution of the measurement is around 300 fs.

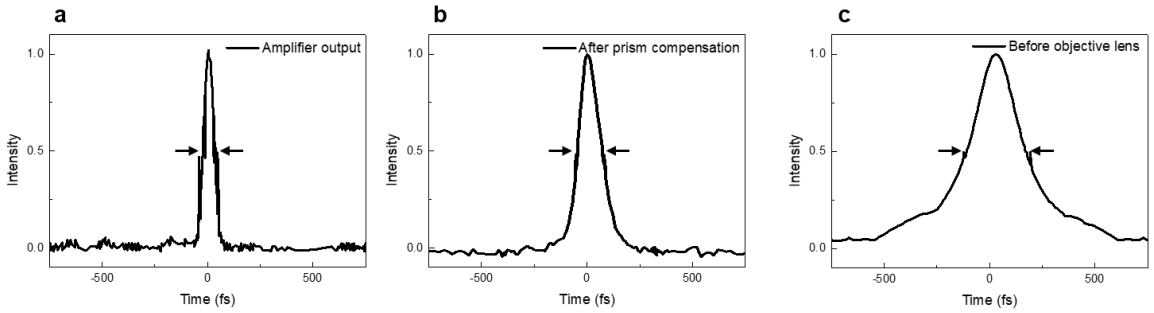

Figure R1 **| Pulse width measurements of the pump pulse at different stages of the setup.** A Gaussian pulse was assumed to fit the data. **a.** Pulse width measured by an autocorrelator right after the amplifier output, showing the width of 48.3 fs (standard error: 0.7 fs). **b.** Measured pulse width after the two-prism pair, resulting in the width of 103.9 fs (standard error: 0.9 fs). **c**. Measured pulse width just before the objective lens, resulting in the width of 258.9 fs (standard error: 4.1 fs).

General remarks and comments of Reviewer 2:

After reading the reply, I think my concerns about this manuscript are well addressed and the manuscript has been revised accordingly. Hence, I recommend its publication.

Response 2: We thank Reviewer 2's time to review our manuscript. We sincerely appreciate his/her positive comment and suggestion regarding the publication of our work. Reviewer 2's support is truly encouraging, and we are grateful for valuable input throughout the review process.

General remarks and comments of Reviewer 3:

The authors have well addressed my comments and concerns. The manuscript is now in a much improved shape. I would like to recommend publication of this manuscript.

Response 3: We sincerely thank Reviewer 3 for his/her time and effort in evaluating our manuscript. We are grateful for the encouraging feedback and recommendation for publication, and we appreciate Reviewer 3's contributions, which have enhanced the quality of our study.

In the manuscript titled “Ultrafast Floquet engineering of Fermi polaron resonances in charge tunable monolayer WSe2 devices”, the authors have employed below-gap photoexcitation in the gate-tunable monolayer WSe2. Both attractive and repulsive Fermi polarons exhibit valley-selective resonance shifts in correlation with the pump intensity. The authors report that the dynamical shift of the repulsive polarons (RP) exhibits a decreasing feature, while that of the attractive polarons (AP) is strongly enhanced with increasing Fermi-sea density. While the paper somewhat extends the understanding of Fermi polaron system in condensed matters under below-gap optical field dressing, several key issues remain that the authors should address:

1. **Mechanism of Pump Dressing:** Since polarons are superpositions of excitons and holes (Fermi sea), the exciton components are expected to be affected by external field dressing. How does exciton-photon interaction contribute to the observed Stark shift in RP and the additional Stark shift in AP? The authors should clarify this.

In addition, the authors have claimed that the binding energy of the AP is weakened by pump-induced “hot” holes (Line 161), which should be related to the photon-hole interactions. However, a quantitative clarification should be provided to demonstrate that photon-hole interactions are the primary drivers of the additional Stark shift, compared to exciton-photon interactions.

2. The authors attribute the blueshift of AP to pump-induced “hot” holes. However, these photo-induced carriers should exhibit very long lifetimes, as shown in the Supplementary Note 3. This is very confusing.

3. **Theoretical Model:** Regarding the theory model, for the field-dressed Fermi polaron at time zero, the authors should include the pump photon and relavent interactions (at least the type that contributes to the Stark shift of RP), just like the Hamiltonian in Eq. (1) of Ref. [24]. From there, the self-energy averaged over one period of the pump could be derived from the Chevy Ansatz, and the Stark shift of RP and AP could be addressed.

4. **Wavefunction and Oscillation Strength:** The Fermi polaron state $|\Phi\rangle$ considered here, is a superposition of excitons, holes, and the pump photons (if dressed). The observed dynamic changes in reflectivity from the weak probe pulse should be

related to the interband dipole transitions like $\langle\Phi|\hat{r}|\Phi\rangle$, where $\hat{r}$ represents the dipole operator. However, not all the wavefunction components of $|\Phi\rangle$ contribute equally to the observations, due to symmetry, particle number, or valley degree of freedom. Does the oscillation strength f_{osc} observed in the experiments plays a role analogous to that in the two-level system? Clarification on this point is crucial for making the argument about the "additional Stark shift of AP" based on f_{osc} differences and blueshifts.

On the other hand, the simulated graph in Fig. 4a shows that at low $E_{F,h}$, the "oscillation strength" of AP is stronger than RP, which conflicts with the results of Fig. 1c. The authors shall discuss this discrepancy, as well as the variance in spectral widths.

5. The authors introduced phenomenological shifts of polaron binding energy in the form of "$\Delta E_{P,\mathrm{pump}} = 23\ \mathrm{meV} + 1.3E_{F,h}(1-\eta)$", the effect of pump dressing is attributed to the η factor. However, this explanation is phenomenological and lacks validation.

Minor questions:

6. The authors have used the phrase "nonequilibrium" In Line 24, 47, 60 and 76. In Floquet engineering, the system is dressed by a periodic field. Even though the system dynamically changes on sub-cycle timescales, it may not be accurate to refer to it as "nonequilibrium".

7. The title uses the phrase "Ultrafast Floquet Engineering of XXX." As with the point above, Floquet engineering describes the dressing of a quantum system by a periodic field. If envelope effects are considered, a nonadiabatic Floquet theory should be applied.

8. In line 30, the authors claim that "circularly-polarized optical driving influences the Fermi sea in an opposite valley", but the significance of this effect remains unclear (even in Supplementary Figure 1(g)). The authors should elaborate on its importance.

9. In line 52, the "Fermi-sea density E_F" is used. While Fermi-sea density is related to the Fermi energy, they are not directly interchangeable. Consider using "Fermi-energy E_F" instead?

10. In line 110-112, the authors mention "to prevent any band-to-band

population…171 meV ". However, in the experiment data shown in Fig. 2a and 2b, there are still several $\Delta R/R_0$ features when the time delay exceeds 0.2ps (albeit much smaller than at zero delay and different from above-gap excitations). Given the pump field's strength and pulse duration, how many carriers or excitons could be excited? Is their density comparable to that of the holes induced at finite $E_{F,h}$?

11. The term "virtual exciton effect" is used without a clear explanation. The authors should provide a more detailed clarification.