S.I. COMPUTATIONAL

A. Pure energy state

Gerritsma, et. al. [1] utilized the projection onto the first and the second spinor components, based on the interpretation of the particle being in either the upper or lower state without optical perturbation. Due to the interaction term in the Hamiltonian, the pure upper and lower states are no longer eigenstates. Projection onto the upper and the lower states mixes true eigenstates which evolve differently in time. Even though their use of "component state" is justified, we note that a Gaussian spinor packet is never a pure energy state as seen in eqns. (34) and (42), and Fig. S.1 (a). This is well reflected in how they prepared the pure negative-energy wavepacket. When they prepared a negative-energy only packet, "reverse-engineered eigenstate" in Fig. 3, Ref. 1, they numerically obtained, after spatially separating packets drifting apart, \( \Psi(p, t = 0) = \begin{pmatrix} -0.48e^{-(p-2.26)^2} \\ +0.75e^{-(p-2.14)^2} \end{pmatrix} \) where \( \lambda_C = 1.2\Delta \) and \( p \) is in the unit of \( \hbar/\Delta \). This is equivalent to the different skewedness of \( u_1^\pm \) and \( u_2^\pm \) in eqn. (32), Fig. 1, and Fig. S.1 (a). \( u_1^- \) (and \( u_2^- \)) weighs larger than \( k_0 \) and \( u_2^+ \) (and \( u_1^+ \)) weighs smaller than \( k_0 \), shifting the center of momentum distributions in each components, such that the wavepacket of negative-energy component only becomes as follows:

\[
\Psi(k) = \sqrt{G(k-k_0; \sigma_k)} \begin{pmatrix} u_1^+(k) \\ u_2^+(k) \end{pmatrix} 
\]

\[
= \sqrt{G(k-k_0; \sigma_k)} \begin{pmatrix} u_1^+(k_0) + (k-k_0) \frac{\partial u_1^+}{\partial k} \\ u_2^+(k_0) + (k-k_0) \frac{\partial u_2^+}{\partial k} \end{pmatrix} 
\]

\[
= \sqrt{G(k-k_0; \sigma_k)} \begin{pmatrix} u_1^-(k_0) \exp[\rho_1(k-k_0)] \\ u_2^-(k_0) \exp[\rho_2(k-k_0)] \end{pmatrix} 
\]

where \( \rho_1 = \frac{\partial u_1^-}{\partial k} / u_1^- = \pm \frac{\lambda_C}{2\sigma_k} \sqrt{\frac{k_0+1}{k_0-1}} \) and \( \rho_2 = \frac{\partial u_2^-}{\partial k} / u_2^- = \pm \frac{\lambda_C}{2\sigma_k} \sqrt{\frac{k_0+1}{k_0-1}} \) using eqns. (23) and (29). Then we obtain \( \Psi(p, t = 0) \approx \begin{pmatrix} -0.50e^{-(p-2.24q)^2/(1.00q)^2} \\ +0.73e^{-(p-2.15q)^2/(1.01q)^2} \end{pmatrix} \), where \( \gamma_0 = 2.80 \), \( p_0 = 2.18q \), \( 2\sigma_p = 1.01q \), and \( q = 1.2mc \).

Components in eqn. (S.1) are plotted in Fig. S.1. It can be seen that the spinor cannot be regarded as a constant over the momentum distribution, and the proba-

\[
\begin{align*}
|\mu_1|^2 & = 0.25 \\
|\mu_2|^2 & = 0.75
\end{align*}
\]

\[
\begin{align*}
|\nu_1|^2 & = 0.63 \\
|\nu_2|^2 & = 0.37
\end{align*}
\]

FIG. S.1. (a) Spinor coefficients of a negative-energy planar wave and (b) Gaussian momentum profiles of the total wavefunction (dashed line) and its first (solid line) and second (dotted line) components of the negative-energy wavepacket in Fig. 3, Ref. 1. The first and the second components are shown in black solid and dotted lines, respectively. Vertical dashed lines indicate the center of their profiles.
bility density of the first component is blue shifted (by $+0.06q$), whereas the that of the second component is red shifted (by $-0.03q$), from that of the total wavefunction of negative-energy component only.

### B. Wavepacket Dispersion

Fig. S.2 displays $\sigma_v$ values from an analytical expression and from fitting numerical simulations of the Zitterbewegung behavior. For the analytical expression, we use $\sigma_v = \frac{\sigma_p}{\gamma_0^2}$, which is only valid for a very small $\sigma_p$, and starts to deviate from the fitted value around $2\sigma_p > 0.5mc$. For the numerical simulation with exact Gaussian packets, it plateaus around $1.1c$, which reflects that no velocity can be greater than $c$ in relativity. For the numerical simulation with spinor packets, it starts to decrease around $2\sigma_p > 2mc$, which is because the momentum shifts in spinor packets becomes so severe that the overlapping of two momentum distributions becomes smaller.

### C. Instantaneous angular frequency

Eqn. (50) is not a single frequency oscillation, and if we try to fit the numerical simulation with a single frequency oscillation function, it appears as if the oscillation is initially faster, and then it slows down. In order to illustrate this, we evaluate an instantaneous angular frequency of oscillation by a temporal differentiation of the argument of the sine term in eqn. (50), which is plotted for various momentum spreads at $p_0 = 0$ in Fig. S.3. When the momentum spread is small, the angular frequency is close to $2\omega_0$. As the momentum spread gets larger, the instantaneous angular frequency is substantially larger than $2\omega_0$ and then becomes $2\omega_0$. Furthermore, the larger the momentum spread, the faster the instantaneous angular frequency becomes $2\omega_0$. Fig. S.3 clearly demonstrate that it is not reliable to evaluate the angular frequency of Zitterbewegung behavior by fitting the mean position values with a single frequency oscillation function.

### D. Momentum shifts in spinor wavepackets

Gerritsma et al. [1] varied the effective mass, such that $\lambda_C = 0.6, 1.2, 2.5$, and $5.4$ $\Delta$, respectively, in Fig. 1, Ref. 1. Since $\sigma_z = 1\Delta$ for each case, $2\sigma_p = \frac{\hbar}{\sigma_z} = 0.6, 1.2, 2.5$, and $5.4$ $mc$, respectively. Even though spinor packets are prepared with a zero momentum (with $p_0 = 0$ and (1,1) spinor), the actual positive- and negative-energy components exhibit momentum shifts, magnitude of which depends on the momentum spread, and the mean position of the spinor packets drifts in a positive direction over time.

Figs. S.4 and S.5 (a-d) compares the momentum distributions of the positive- and negative-energy components for (1,0) and (1,1) spinor packets, respectively. When (1,0) spinor is used, it results in contamination by a small negative-energy component, and the distribution of the positive-energy component is centered at $p = 0$. On the other hand, when (1,1) spinor is used, the positive-energy component is blue shifted and the negative-energy component is red-shifted, resulting in both components drifting with a positive velocity. Vertical lines indicate the momentum shifts calculated using eqns. (61) and (62), which are valid for a very small $\sigma_p$ for which spinor coefficients can be linearly approximated. Fig. S.5 (a-d) and (e-h) also compare (1,1) spinors with $p_0 = 0$ and $p_0 = 0.62mc$. For $p_0 \neq 0$, (1,1) spinor results in unequal populations of positive- and negative-energy components ($\Theta = 0.50790$ for $p_0 = 0.62mc$), in which case momentum shifts are also unequal.
E. Position drift in spinor wavepacket

Fig. S.6 (a-d) compares the analytical expression, eqn. (49), with the numerical simulations with the exact wavefunction, eqn. (56), and the spinor wavepacket of equal populations of electron and positron at \( p_0 = 0 \) (\( \Omega = \frac{\pi}{4} \) and \( \Theta = \frac{\pi}{4} \)) as the initial state. The position expectation value for the total wavefunction shows jitter motion when an electron and a positron overlap, and becomes zero. The analytical expression and the exact numerical simulation show a good agreement, whereas the numerical simulation with the spinor wavepacket exhibits a drift. The drift behavior is due to the fact that the spinor wavepacket mixes positive- and negative-energy components, and (1,1) spinor state generates the positive- and the negative-energy components blue- and red-shifted, respectively, in the momentum space, which becomes more severe as the momentum distribution becomes larger (see section IV B). Also note that the apparent Zitterbewegung amplitude becomes smaller for a larger momentum spread (at zero momentum), because the wavepacket spreads faster (see eqn. (B1)), and the first maximum of interference occurs at \( t \neq 0 \). For spinor wavepackets, momentum shifts result in less overlap in momentum space, which leads to a smaller interference.

Fig. S.6 (e-h) also compares the analytical expression and the numerical simulations at \( p_0 = 0.62mc \) with the positive- and the negative-energy component populations determined (\( \Theta = 0.5079 \)) such that it corresponds to (1,1) spinor packet (\( \Omega = \frac{\pi}{4} \)). The analytical expression, which is only a first order solution, deviates from the exact simulation, due to neglecting higher order terms. In particular, the linear approximation employed here (eqn. (38)) implies that the center of the momentum distribution corresponds to the center of the velocity distribution, and that the propagation is determined only by the second terms in eqns. (36) and (37), and that the third terms therein only contributes to dispersion. However, when \( 2\sigma_p \gg mc \) and \( \sigma_z \ll \lambda_c \), higher order terms need to be explicitly considered in eqn. (38), which results in a modified propagation. It is to be noted that the velocity cannot be larger than \( c \) in relativity.

FIG. S.4. Momentum profiles of positive- and negative-energy components of (1,0) spinors with $p_0 = 0$ and $2 \sigma_p = (a) 0.6$, (b) 1.2, (c) 2.5, and (d) 5.4 $mc$. The total probability is shown in a black dashed line, and the positive- and the negative-energy components are shown in red and blue, respectively.
FIG. S.5. Momentum profiles of positive- and negative-energy components of (1,1) spinor with (a-d) \( p_0 = 0 \) and (e-h) \( p_0 = 0.62mc \), and \( 2\sigma_p = 0.6, 1.2, 2.5, 5.4mc \) in Ref. 1. The total probability is shown in a black dashed line, and positive- and the negative-energy components are shown in red and blue, respectively. Gray, red, and blue dashed lines indicate momentum shifts, calculated using eqns. (D5) and (D6).
FIG. S.6. Comparison of position expectation values by numerical simulation for (1,1) spinor (green dotted line) with those by analytical expression (red solid line) and exact numerical simulation (blue dashed line) of corresponding exact states ($\Theta = \frac{\pi}{4}$ and $\Theta = 0.50790$), for $p_0$ and $2\sigma_p$ of (a) 0, 0.6, (b) 0, 1.2, (c) 0, 2.5, (d) 0, 5.4, (e) 0.62, 0.6, (f) 0.62, 1.2, (g) 0.62, 2.5, and (h) 0.62, 5.4 in $mc$. 