Divergent Nonlinear Response from Quasiparticle Interactions

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We demonstrate that nonlinear response functions in many-body systems carry a sharp signature of interactions between gapped low-energy quasiparticles. Such interactions are challenging to deduce from linear response measurements. The signature takes the form of a divergent-in-time contribution to the response—linear in time in the case when quasiparticles propagate ballistically—that is absent for free bosonic excitations. We give a physically transparent semiclassical picture of this singular behavior. While the semiclassical picture applies to a broad class of systems we benchmark it in two simple models: in the Ising chain using a form-factor expansion, and in a nonintegrable model—the spin-1 Affleck-Kennedy-Lieb-Tasaki chain—using time-dependent density matrix renormalization group simulations. We comment on extensions of these results to finite temperatures.

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Introduction.—The response of a quantum many-body system to external perturbations is central to experimentally extracting information about its properties. In typical settings, the system is weakly perturbed out of its equilibrium state, and the leading linear response [1,2] contribution can be related to a two-point equilibrium correlator. Consequently, linear response functions are often straightforward to interpret. For instance, in cases where the external perturbation can create single quasiparticle (QP) excitations on top of the ground state, their dispersion can be read off directly from momentum-resolved linear-response data. However, the simplicity that lends power to linear response often limits its utility in more complex situations. For example, various distinct physical mechanisms can give rise to a broad frequency spectrum in linear response functions: e.g., selection rules requiring probe fields to excite multiple QPs, inhomogeneous broadening due to quenched disorder, and homogeneous broadening from QP decay. Differentiating among these using linear response data alone is a challenge. Often, nonlinear response functions give more direct insight into the nature of low-energy excitations [3,4]. Intuitively, this is because they involve multitime correlation functions that, suitably analyzed, can disentangle different sources of broad spectra [4]. Pump-probe experiments [5,6] and two-dimensional coherent spectroscopy [7–12] both extract nonlinear response functions. They have long been used in magnetic resonance and in optical experiments on chemical systems, usually in regimes where the response reduces to that of individual atoms, averaged over a suitable statistical ensemble. The extended many-body systems usually encountered in solid-state materials or ultracold atomic gases do not always admit a similarly simplified description. Developing techniques to compute nonlinear response functions in such systems is thus an important goal [13–29], made more pressing as experiments begin to probe such regimes. Apart from situations that reduce to an ensemble of few-body systems [4,30,31], most controlled results have been obtained for free theories [4,32–35], or exactly solvable models [36–41]. There is thus a need for qualitative insights into universal aspects of nonlinear response outside these settings.

Here, we offer such a perspective by means of a semiclassical theory. We focus on the simplest nontrivial systems, whose low-energy spectrum consists of gapped QPs and where the perturbing field can excite single QPs. For ballistic QPs in one dimension ($d = 1$) and at $T = 0$ we find that the $q = 0$ third-order response functions diverge linearly in the time interval between distinct applications of the external field, with a strength set by the inter-QP scattering phase shifts. This richness is to be contrasted with $q = 0$ linear response for such systems: a delta-function peak at the gap frequency, related to the stability of QPs, and independent of the interactions between QPs.

This qualitative picture applies to a broad class of systems and, importantly, the details of the long-time divergence give access to detailed information about QP interactions such as the two-QP phase shifts.

Apart from enjoying the simplifying features of stable QPs, the primary example we consider below—the transverse-field Ising chain (TFIC)—is also integrable. As is well known, it maps to a theory of free fermionic QPs, whose statistics enforce a scattering phase shift of -1 leading to singular nonlinear response. We benchmark semiclassical analysis for the TFIC against exact results using form-factor expansions, detailed in upcoming work [42]. While the form-factor approach is applicable to a subset of integrable models, the semiclassical approach applies to a broad class of (integrable or nonintegrable) systems, characterized by the existence of stable, gapped QPs in some range of momentum. To confirm the validity of the semiclassical approach in this context we perform time-dependent density matrix renormalization group (tDMRG) simulations in the (nonintegrable) Affleck-Kennedy-Lieb-Tasaki (AKLT) spin-1 chain [43,44]. Furthermore, with only slightly more work, the semiclassical approach can be generalized to treat [low] finite T. Relaxing the restriction to $q = 0$ to allow momentum resolution permits direct extraction of the QP scattering matrix by combining linear and nonlinear response data. Our work thus promises an intuitive way to compute and understand nonlinear responses in a variety of quantum many-body systems.

Setup.—We initially focus on the TFIC, with Hamiltonian $H = -J\sum_{j=0}^{L-1}(\sigma_j^z\sigma_{j+1}^z + g\sigma_j^x)$. We work in the paramagnetic phase ($g > 1$) and consider $q = 0$ perturbations coupling to the order parameter $M = \sum_j \sigma_j^z$ (recall that only such "Ising-odd" operators can excite single QPs above the ground state). Extensions to $q \neq 0$ are straightforward and will be reported elsewhere [42].

The model is exactly solvable by means of a Jordan-Wigner transformation [45] that maps H to a quadratic fermion problem. This yields a QP dispersion relation $\epsilon(k) = 2J\sqrt{1 + g^2 - 2g\cos(k)}$, with a gap $\Delta = \epsilon(0)$. However, since σ^z maps to a nonlocal (stringlike) operator, response functions involving M cannot be easily computed using Wick's theorem. We therefore resort to techniques developed in Refs. [45–53]. Similarly, any local spin operator that can create single fermionic QPs must be nonlocal in the fermion basis. Consequently, their nonlinear response is sharply distinct from that of fermion-local spin observables [4], that only excite even numbers of QPs. We first study the pump-probe signal,

$$\Xi_{\mathrm{PP}} = -\frac{i}{L}\langle 0|e^{i\mu M(0)}[M(t_1+t_2), M(t_1)]e^{-i\mu M(0)}|0\rangle + \frac{i}{L}\langle 0|[M(t_2), M(0)]|0\rangle, \tag{1}$$

which can be viewed as the difference in the linear response (measured between times $t_1, t_1 + t_2$) computed in two states: the original QP vacuum $|0\rangle$, and a "pumped" state $e^{-i\mu M(0)}|0\rangle$ obtained by perturbing the QP vacuum at $t = 0$ with a "kick" of strength μ coupling to M [54].

In the regime where the pump only weakly perturbs the system away from equilibrium, we can expand Ξ_{PP} in μ and evaluate the resulting correlators in equilibrium: odd powers vanish by symmetry, so $\Xi_{\mathrm{PP}} = \mu^2\chi_{\mathrm{PP}}^{(3)} + O(\mu^4)$. The superscript denotes a third-order response, which we split into pieces divergent and convergent in time, $\chi_{\mathrm{PP}}^{(3)} = \chi_{\mathrm{PP};d}^{(3)} + \chi_{\mathrm{PP};c}^{(3)}$, where the former

$$\chi_{\mathrm{PP};d}^{(3)} = \frac{2}{L}\Im\underbrace{\langle 0|Me^{iH(t_1+t_2)}}_{\text{bra}}\underbrace{Me^{-iHt_2}Me^{-iHt_1}M|0\rangle_C}_{\text{ket}} \tag{2}$$

is our focus. Only the connected correlator (denoted by the subscript C) contributes to the response, as required by causality. Both from the Heisenberg picture in (1) and the Schrödinger one (2), it is evident the correlators are not time ordered. It is convenient to also distinguish the "ket" and "bra" sides of (2). Formally these correspond to forward and backward branches of the Keldysh time contour, which runs from $t = 0$ to $t = t_1 + t_2$ and back. At $t = 0$, the operator M acts on bra and ket sides, whereas at time $t = t_1$ it acts solely on the ket side. Both sides are then evolved up to time $t = t_1 + t_2$, whereupon M is measured. We adopt the standard nomenclature where nth order response functions involve n external perturbations and $n + 1$ operators; the extra operator corresponds to the measured observable. [Note that $\chi_{\mathrm{PP}}^{(3)} \propto \mu^2$, but appears at third order when expanding in terms of *all* external fields, as an extra perturbation is required to extract the linear response function in Eq. (1), cf. [54].]

Long-time divergences and nonperturbative effects.—Our main result is that $\chi_{\mathrm{PP};d}^{(3)}$ diverges at late times,

$$\chi_{\mathrm{PP};d}^{(3)} \simeq 2\sin(\Delta t_2)(t_1 + t_2)\mathcal{C}_{\mathrm{PP}}\left(\frac{t_2}{t_1 + t_2}\right) \tag{3}$$

with a nonuniversal scaling function $\mathcal{C}_{\mathrm{PP}}$ whose numerical behavior is shown in Fig. 2 for the TFIC. This divergence has a simple semiclassical interpretation, involving ballistic propagation of quasiparticles and their scattering (Fig. 1).

Before detailing the semiclassical analysis, we argue that we expect $\chi_{\mathrm{PP}}^{(3)}$ to diverge on general grounds. Even for arbitrarily small perturbation strength μ, Ξ_{PP} will saturate to an $O(1)$ value independent of μ at late times. This is most easily seen for sufficiently large t_1, such that the system effectively thermalizes after the initial kick. The perturbed state $e^{-i\mu M(0)}|0\rangle$ is then effectively at a finite (but small) T. (In the integrable case, it can be approximated by a generalized Gibbs ensemble, but this is not essential to the analysis.) The first line of (1) is then approximately a

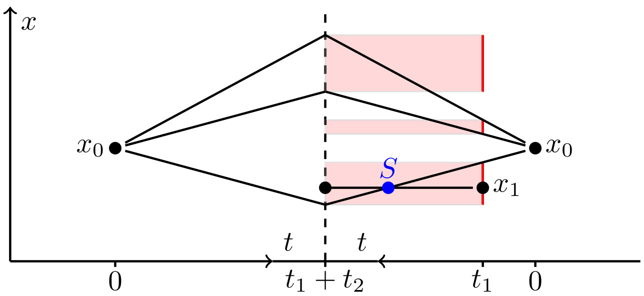

FIG. 1. Illustration of processes contributing to the late-time divergence of $\chi^{(3)}_{PP;d} \sim \langle M(0)M(t_1+t_2)M(t_1)M(0)\rangle$. A dashed line, corresponding to time $t = t_1 + t_2$ separates the bra (left) and ket (right) sides of the time evolution; time increases toward this line. Solid lines denote QP worldlines, and circles denote the action of the operator M. When two lines cross, the amplitude for the diagram is multiplied by $S = -1$. For a fixed x_0, the red segments indicate the set of x_1 giving rise to a scattering-connected contribution. The length of the red set is proportional to the overall timescale, leading to the linear divergence in (3).

linear-response function in a finite-T state, which is expected to decay exponentially in time

$$\langle e^{i\mu M(0)}[M(t_1+t_2), M(t_1)]e^{-i\mu M(0)}\rangle \sim e^{-\gamma_\mu t_2}, \qquad (4)$$

intuitively due to stochastic scattering events with the QPs created by $e^{-i\mu M(0)}$ [55]. Therefore, at long times, the effect of the perturbation becomes $O(\mu^0)$. A natural possibility is that $\gamma_\mu \propto \mu^2$, suggesting that $\chi^{(3)}_{\mathrm{PP}}$ diverges linearly in t_2 whenever we have a stable QP excitation at zero momentum. The full pump-probe response Ξ_{PP} thus initially shows a linear increase, probed in the response limit, which eventually saturates at late times $t \gg 1/\mu^2$ to an $O(\mu^0)$

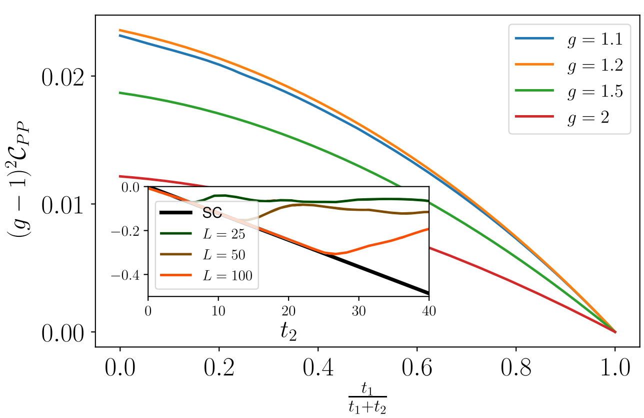

FIG. 2. Scaling function $\mathcal{C}_{\mathrm{PP}}[t_1/(t_1+t_2)]$ as defined in (3) for various values of the transverse field g and $J = 1$. For graphical convenience, $\mathcal{C}_{\mathrm{PP}}$ has been rescaled by $(g-1)^2$. Inset: $\chi^{(3),[3,4,3]}_{PP}/[2\sin(\Delta t_2)]$ for $t_1 = 0$, $g = 2$, and $J = 1$. Various colored curves denote the exact results obtained by numerically summing the form factors in an L-sites chain. The black line gives the linear scaling due to $\mathcal{C}_{PP,3}$ computed semiclassically, cf. (5).

value—much larger than the perturbatively weak response of few-body systems. From here on we focus on $\chi^{(3)}_{\mathrm{PP}}$ and refer to it as nonlinear response. We will show the existence of late-time divergences in $\chi^{(3)}_{\mathrm{PP}}$, which are to be understood as describing the behavior of the full Ξ_{PP} in the parametrically large time window $t \lesssim \mu^{-2}$. We note that the semiclassical approach can be used to establish late-time divergences in higher-order nonlinear response functions $\chi^{(n>3)}_{\mathrm{PP}}$ as well.

Furthermore, the presence of such long-time divergences in the nonlinear response of gapped systems is in stark contrast to their linear response, which is always finite. The situation in gapless systems is quite different: as we demonstrate by studying the XXZ chain, here linear response functions can already exhibit long-time divergences, which are further enhanced in nonlinear response [25,54].

Semiclassical picture.—The scaling form (3) and the scaling function $\mathcal{C}_{\mathrm{PP}}$ can be quantitatively understood from a simple semiclassical picture inspired by the seminal approach of Refs. [55,59] (see also Refs. [60–63]). Our basic objects are wave-packet (WP) states $|r,k\rangle$ of QPs, which we will think of as having approximately well-defined positions r and momenta k in the sense that the effects of the dispersion of the wave packets will be subleading. Multi-WP states $|\boldsymbol{r};\boldsymbol{k}\rangle$ are obtained as tensor products of single-WP states and by locality of the Hamiltonian (approximately) have simple dynamics as long as the WPs are spatially well separated. By construction n-WP states are in one-to-one correspondence with scattering states of n QPs. On the ket side, the action of the operator $M(0)$ will, after a sufficiently long time, result in "intermediate" n-WP states (with n odd), where the individual WPs approximately originate from the same point x_0 (which is integrated over) and whose momenta sum to 0. Each WP (approximately) propagates ballistically with its group velocity $v(k) = \epsilon'(k)$, i.e., $e^{-iHt}M(0)|0\rangle$ is approximately a superposition of states of the form $\int dx_0 |\boldsymbol{r};\boldsymbol{k}\rangle$ with $\boldsymbol{r}$ specified by $r_j = x_0 + v(k_j)t$ and $\boldsymbol{k}$ such that $\sum_j k_j \approx 0$, but otherwise arbitrary. We start by considering processes where all WPs proceed undisturbed to time $t_1 + t_2$, whereupon they annihilate with the n WPs produced by $M(0)$ on the bra side. Note that, in order to have a non-negligible overlap between the bra and the ket, the momenta $\boldsymbol{k}$ on the two sides must approximately coincide, as well as the position x_0 at which the WP shower is created (see Fig. 1). In the TFIC the amplitude associated with creating and annihilating a shower of n WPs with a given set of momenta $\boldsymbol{k}$ is $|F_n(\boldsymbol{k})|^2(d^n\boldsymbol{k}/(2\pi)^{n-1})\delta(\sum_{j=1}^n k_j)$ for every initial position x_0. Here $F_n(\boldsymbol{k}) = \langle \boldsymbol{k}|\sigma^z_0|0\rangle$ is the n-QP form factor on top of the ground state, whose precise value is unnecessary to proceed with the semiclassical calculation. [Note that in principle $F_n(\boldsymbol{k})$ and $v(k)$ can be numerically computed for nonintegrable systems using MPS [64]].

In the processes of interest, $M(t_1)$ creates a single $q=0$ WP at x_1, which is spatially well separated from the position at time t_1 of the n WPs produced by the action of $M(0)$ on $|0\rangle$. The WP created by $M(t_1)$ is then annihilated by $M(t_1+t_2)$, giving rise to an amplitude $|F_1(0)|^2 e^{-i\Delta t_2}$. Naively, integrating over the arbitrary positions x_0 and x_1 generates a contribution to $\chi^{(3)}_{\mathrm{PP}}$ that diverges proportionally to L in the thermodynamic limit. However, the contribution of processes in which the WP trajectories do not cross is simply equal to the product of two-point functions $L^{-1}\langle M(0)M(0)\rangle\langle M(t_1+t_2)M(t_1)\rangle$, and therefore the extensive spatial divergence cancels when we subtract the disconnected contributions.

In contrast, each time a pair of WP trajectories cross, the amplitude for the process picks up a factor of the scattering matrix S. In the TFIC, this simply encodes the fermionic statistics of QPs, i.e., $S=-1$. Therefore, a process involving an odd number of scattering events (cf. Fig. 1) does not cancel against disconnected contributions; we refer to processes which are connected only because of such events as *scattering connected*. After subtracting the disconnected component we obtain a factor $S-1$, evaluating to -2 in the TFIC. Finally, we must integrate over all possible x_1 that produce a scattering-connected process. (The integration over x_0 will produce a factor L, cancelling the L^{-1} in the definition of $\Xi^{(3)}_{\mathrm{PP}}$.) One can verify that, for a given set of momenta $\boldsymbol{k}$ of the WPs produced by the pump, the range of x_1 leading to scattering-connected processes has length $v_{\mathrm{PP}}(t_1+t_2)$, where v_{PP} is a linear combination of the velocities of the various WPs [54]. In this way we obtain a divergent contribution to $\chi^{(3)}_{\mathrm{PP};d}$ of the form (3) with $\mathcal{C}_{\mathrm{PP}}=4\pi\sum_{n,\mathrm{odd}}\mathcal{C}^{(n)}_{\mathrm{PP}}$, where

$$\mathcal{C}^{(n)}_{\mathrm{PP}}=-\frac{|F_1(0)|^2}{n!}\int\frac{d^n\boldsymbol{k}}{(2\pi)^n}\delta\left(\sum_{j=1}^{n}k_j\right)|F_n(\boldsymbol{k})|^2 v_{\mathrm{PP}}(\boldsymbol{k}). \quad (5)$$

Note that for $g\gtrsim 1.1$, $\mathcal{C}_{\mathrm{PP}}$ is dominated by $\mathcal{C}^{(3)}_{\mathrm{PP}}$, with higher values of n yielding numerically smaller corrections. This allows us to numerically estimate $\mathcal{C}_{\mathrm{PP}}$, reported in Fig. 2 for representative values of g.

Other types of processes only give rise to subleading contributions at late times. Scattering-connected processes where $M(t_1)$ creates more than one QP subsequently annihilated by $M(t_1+t_2)$ give contributions suppressed as $t_2\to\infty$. This follows since QPs created by $M(t_1)$ spread ballistically from one another and therefore cannot be annihilated by a single local operator. For processes that are not simply scattering-connected, the position where all operators act is fixed by the ballistic propagation of the various QPs; therefore, we expect their contributions to remain finite at late times. We stress that the wave-packet analysis presented above in the particular case of the TFIC can be straightforwardly generalized to any system with ballistic QPs, upon replacement of S with the (momentum-dependent) scattering matrix for that system.

Form-factor expansion.—We bolster the semiclassical result with an exact calculation of the late-time asymptotics of $\chi^{(3)}_{\mathrm{PP};d}$ via a form-factor expansion [45–53]. The main steps are as follows. As a result of integrability, the exact energy eigenstates $|\boldsymbol{p}_N\rangle\equiv|p_1,\ldots p_N\rangle$ can be labeled by the momenta of the quasiparticles. Inserting resolutions of the identity in terms of energy eigenstates between each pair of operators yields

$$\chi^{(3)}_{\mathrm{PP};d}=\sum_{l,m,n\geq 0}\chi^{(3),[l,m,n]}_{\mathrm{PP};d}(t;\boldsymbol{K}_{n_a},\boldsymbol{p}_{n_b},\boldsymbol{k}_{n_c}), \quad (6)$$

where $\chi^{(3),[l,m,n]}_{\mathrm{PP};d}\propto\langle 0|M|\boldsymbol{K}\rangle\langle\boldsymbol{K}|M|\boldsymbol{p}\rangle\langle\boldsymbol{p}|M|\boldsymbol{k}\rangle\langle\boldsymbol{k}|M|0\rangle$. A key property of these matrix elements [65–68] is the presence of kinematic poles: as p_i approaches k_j the matrix element becomes singular, $\langle\boldsymbol{p}|M|\boldsymbol{k}\rangle\sim[1/(p_i-k_j)]$. These contributions ultimately give rise to the late-time divergence in $\chi^{(3)}_{\mathrm{PP};d}$ [54].

To benchmark the semiclassical picture, we consider its simplest nonzero contribution, from $\mathcal{C}^{(3)}_{\mathrm{PP}}$ (sketched in Fig. 1). Counting the number of QPs before and after each operator M, we expect that $\mathcal{C}^{(3)}_{\mathrm{PP}}$ is linked to $\chi^{(3);[3,4,3]}_{\mathrm{PP};d}$ in the form-factor expansion. Numerically evaluating $\chi^{(3);[3,4,3]}_{\mathrm{PP};d}$ we see good agreement with the semiclassical expectation (see inset of Fig. 2).

We can also use the form-factor approach to compute the leading correction to (3), which scales as $\sqrt{t_2}$ [69,70]. In an forthcoming work [42] we show that this can be interpreted as due to WP broadening, omitted in the leading semiclassical analysis.

Numerical benchmark in the spin-1 AKLT chain.—To confirm that the semiclassical analysis extends beyond integrable models, we numerically study a nonintegrable Haldane-gap spin-1 chain. This class of systems has been analyzed in great detail both theoretically [71–79] and experimentally [80–86] since the 1990s. All these systems feature stable QP modes in the proximity of momentum $q=\pi$ (and concomitantly two-particle scattering continua near $q=0$). For this reason we study a finite-momentum response function $\chi^{(3)}_{\mathrm{PP};d}(q_1,q_2;t_1,t_2)=(2/L)\Im\langle 0|M(-q_1,0)M(-q_2,t_1+t_2)M(q_2,t_1)M(q_1,0)|0\rangle_C$, with q_1 and q_2 suitably close to $q=\pi$. Here $M(q,t)=\sum_j e^{iqj}S^z_j(t)$ and S^z_j denotes the spin-1 operator along the z axis acting on site j. For numerical convenience we focus on the spin-1 AKLT chain [43,44], but analogous results will hold for other models in this class.

We have computed $\chi^{(3)}_{\mathrm{PP};d}(q_1,q_2;t_1,t_2)$ using tDMRG. The results for $t_1=0$, $q_1=\pi$, and $q_2=2\pi/3$ are reported in Fig. 3. At sufficiently late times $\chi^{(3)}_{\mathrm{PP};d}$ is well fitted by the functional form $At_2\sin[\epsilon(q_2)t_2-\phi]$, which is consistent

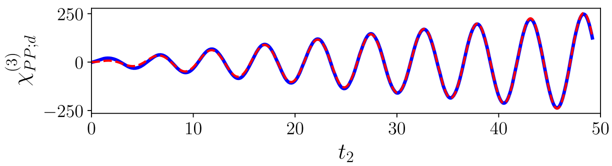

FIG. 3. $\chi^{(3)}_{\mathrm{PP};d}(q_1, q_2)$ (blue line), for $t_1 = 0$, $q_1 = \pi$, and $q_2 = 2\pi/3$. The data is obtained through tDMRG simulation of the spin-1 AKLT chain. A red dashed line indicates a fit of the form $At_2 \sin[\epsilon(q_2)t_2 - \phi]$ consistent with our wave-packet analysis.

with our wave-packet analysis. Here A and ϕ are fitting parameters, while $\epsilon(q_2)$ is independently determined from the numerical computation of the two-point function.

Discussion.—We have shown that in the TFIC and AKLT chain the nonlinear pump-probe response is characterized by a long-time ("infrared") divergence. Furthermore, as we have already argued, this is in fact a general feature of any interacting translationally invariant many-body system that has a stable, gapped single-particle excitation. Such a behavior is already suggested by our analysis where we demonstrated that the late-time pump-probe signal Ξ_{PP} can be reexpressed as the difference of two-point correlation functions at zero and finite temperatures and hence is $O(1)$, independently of the strength of the "pump" perturbation; the divergence in $\chi^{(3)}_{\mathrm{PP}}$ reconciles this result with the perturbative expansion in powers of the applied fields. We therefore expect this long-time growth to apply to a broad class of systems, and be visible on intermediate timescales at low finite temperature.

In cases where QPs propagate ballistically, we have developed a semiclassical picture of WP propagation and scattering that explicitly shows the divergence to be linear in time and identifies the processes that give rise to it. While our discussion has focused on the simple case of the TFIC, the semiclassical arguments generalize to nonintegrable models, and even to finite temperatures [54].

An enticing possibility suggested by our work is the measurement of scattering matrices from third-order response functions. In $\chi^{(3)}_{\mathrm{PP};d}(q_1, q_2; t_1, t_2)$, if the $n = 1$ contribution is the dominant one—as can e.g., be achieved using a frequency-modulated pump with negligible amplitude to excite the system at energies greater than 2Δ—$\chi^{(3)}_{\mathrm{PP}}$ can be expressed in terms of the scattering matrix $S(q_1, q_2)$ and data that can be extracted from linear response, allowing us to read off $S(|q|, 0)$ from the divergent piece of $\chi^{(3)}_{\mathrm{PP}}$ [54]. Even in cases where this is not possible, we expect the measurement of $\mathcal{C}_{\mathrm{PP}}$ to provide important quantitative information on the scattering properties of QPs in materials, that are otherwise not probed directly by linear response. Finally, it would be interesting to explore if similar late-time divergences occur in higher dimensions.

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