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Partial dephasing in interacting many-particle systems and current echo

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Current echos as a response to a sequence of two ultrashort voltage pulses and their delay dependence are studied for interacting particles. The echo amplitude is reduced compared to its noninteracting counterpart, and decays even without an external bath coupling. However, it ends up with a *nonzero* value in the large-delay limit. This partial dephasing can be traced back to correlated energy differences in the many-body system. It is expected for all echo phenomena in small isolated systems. The time-dependent Hartree-Fock approximation fails to describe the current echo's dephasing even qualitatively. [S0163-1829(98)02307-8]

I. INTRODUCTION

The ultrafast kinetics in interacting systems became experimentally accessible in the last few years. This stimulates theoretical efforts to go beyond simple concepts like dephasing rates. Generally dephasing, i.e., the decay of a signal as function of time elapsed between two events (excitations, measurements, etc.), is attributed to one or more of the following aspects: (i) bath coupling, (ii) thermodynamic limit for the system size, and (iii) complex internal structure of the system.

Paradigmatic for the measurement of ''real'' dephasing, as opposed to signal cancelation due to inhomogeneous broadening, are echo experiments performed with spin[1] or optical[2] excitation. A conceptually particularly simple and astonishing echo is the recently predicted current echo[3] (also see Ref. 4, which discusses current echos for classical particles in inhomogeneous magnetic fields): A finite current can appear spontaneously at time 2τ in a disordered system as a response to a sequence of two short voltage pulses[5] separated by a time delay τ. The authors of Ref. 3 based their prediction on properties of ensembles of independent particles. Only results up to third order in the driving fields have been presented ($\chi^{(3)}$ level). An experimental verification of this echo effect is still missing. In fact, it is not clear so far whether the current echo still exists for large exciting pulses which might be required in order to obtain a recordable echo signal. Such pulses can no longer be treated within $\chi^{(3)}$. It is even more questionable whether the echo survives in the presence of Coulomb interaction, as investigated for the photon echo in Refs. 6 and 7.

We present results that rest on an exact treatment of the interaction in terms of system eigenfunctions of small interacting many-particle systems. It is shown that, at least in one spatial dimension, a current echo exists for small or well-localized systems even in the presence of the Coulomb interaction. The signal is, however, reduced compared to the noninteracting case. Furthermore, and in contrast to the noninteracting situation, the echo amplitude decreases with time starting from the noninteracting value down to a *finite* fraction thereof.[8] This suggests that in general particle-particle interaction in isolated and finite systems leads to dephasing which, however, is not complete. For comparison, we also present results within the time-dependent Hartree-Fock (TDHF) theory.

II. THEORY

As a model system, we consider a one-dimensional Pariser-Parr-Pople[9] Hamiltonian with disorder or, equivalently, Anderson's disorder Hamiltonian with additional Coulomb interaction. It is written as

$$H=\sum_{i,s} T(c^{\dagger}_{i+1,s}c_{i,s}+\text{H.c.})+\sum_{i,s} u_i c^{\dagger}_{i,s}c_{i,s} + \frac{1}{2}\sum_{i,j,s,s'} v_{ij}c^{\dagger}_{i,s}c^{\dagger}_{j,s'}c_{j,s'}c_{i,s} \tag{1}$$

in terms of electron creation operators $c^{\dagger}_{i,s}$ for sites i ($i=1,\dots,N$) and spin directions $s=\uparrow,\downarrow$. The on-site energies u_i are Gaussian distributed uncorrelated random numbers with variance $\langle u_i^2\rangle=W^2$. For the interaction v_{ij} a modified Coulomb form[10] with a finite on-site value is used: $v_{ij}=e^2/(4\pi\epsilon_0\epsilon_s\sqrt{|i-j|^2a^2+b^2})$, with dielectric constant ϵ_s and lattice spacing a. A constant positive background charge is added to ensure charge neutrality on average. The strength of the interaction can be controlled by assigning to ϵ_s values between those of order of unity up to very large values. Our numerically exact calculations for interacting systems involve *all* eigenstates, and therefore are restricted to small systems. Thus we can not disentangle the effects of the short- and long-range parts of Eq. (1). This issue can, however, be addressed in the TDHF scheme, which allows the treatment of much larger systems, and would also allow the inclusion of interchain coupling and local-field effects.

The current echo can only be observed on time scales smaller than that of external interactions (bath phonons), i.e., in the ultrashort-time regime. Further, the current echo is visible only in the total response from an *ensemble* of many

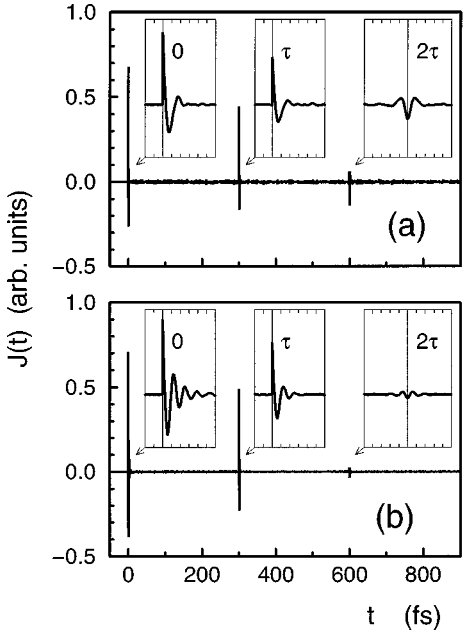

FIG. 1. Ensemble-averaged current as function of time for six-site chains (a) without and (b) with interaction and parameters appropriate for polyacetylene ($a=0.122$ nm, $b=0.158$ nm, and $\epsilon_s=1.5$, and $W=T=2.4$ eV corresponding to $\hbar/T\approx 0.3$ fs). The insets show on a 1-fs scale the direct current responses near $t=0$ and $t=\tau=300$ fs, and the echo near $t=2\tau=600$ fs. Note the sign rule, Eq. (10), to be fulfilled.

chains with different disorder realizations and thus different oscillations. Figure 1 shows that the directly induced currents near $t=0$ and $t=\tau$ decay rapidly to zero due to the ensemble average. However, at $t=2\tau$, a clear echo signal with symmetric shape is seen.

For a theoretical understanding, the current echo is best discussed in the basis of *many-particle* eigenstates $H|\Psi_m\rangle=E_m|\Psi_m\rangle$. A short pulse with a duration shorter than the inverse transition energies centered around t_1 with integrated strength $\phi_1=(a/\hbar)\int_{t_1-0}^{t_1+0}e\mathcal{E}_1(t)dt$ acts on the single- and many-particle wave functions in the real-space representation mainly as a phase factor. For δ pulses, as considered below, this is exact. For M particles the wave function right after and right before the pulse are related by

$$\Psi(x_1,\ldots,x_M,t_1^+)=e^{i\phi_1\Sigma_i x_i/a}\Psi(x_1,\ldots,x_M,t_1^-). \quad (2)$$

This mixes the eigenfunctions with transition amplitudes

$$S_{nm}^{(1)}=\langle\Psi_n|e^{i\phi_1\Sigma_i\hat{x}_i/a}|\Psi_m\rangle. \quad (3)$$

Starting with the ground state Ψ_0 at $t=0^-$, we have after the first pulse at $t_1=0$ ($\hbar=1$, henceforth),

$$|\Psi(t)\rangle=\sum_m|\Psi_m\rangle e^{-iE_m t}S_{m0}^{(1)}. \quad (4)$$

After a second pulse ϕ_2 at $t_2=\tau$, for $t>\tau$,

$$|\Psi(t)\rangle=\sum_{n,m}|\Psi_n\rangle e^{-iE_n(t-\tau)}S_{nm}^{(2)}e^{-iE_m\tau}S_{m0}^{(1)}, \quad (5)$$

yielding, for the expectation value of the displacement current operator, $\langle\hat{J}\rangle=-e\partial_t\langle\hat{X}\rangle$,

$$J(t)=\sum_{n,n',m,m'}e^{-i(E_n-E_{n'})(t-\tau)}e^{-i(E_m-E_{m'})\tau}\cdot S_{m'0}^{(1)*}S_{n'm'}^{(2)*}J_{n'n}S_{nm}^{(2)}S_{m0}^{(1)}, \quad (6)$$

with

$$J_{n'n}=-ie(E_{n'}-E_n)\left\langle\Psi_{n'}\left|\sum_i\hat{x}_i\right|\Psi_n\right\rangle. \quad (7)$$

This expression is the basis for the numerical results presented in this paper.

Next we proceed to identify the contributions in Eq. (6) which produce the echo. The crucial step in the analysis is to keep only certain terms which in the ensemble average are not subject to destructive interference. The underlying rational is that of a random-phase approximation (RPA) in the original meaning.[11]

Besides contributions which are strong at $t\approx\tau$, we find constructive interference at $t\approx 2\tau$ from the "cross terms" $n'=m$, $m'=n$, yielding

$$J^{cross}(t)=-\sum_{n,m}e^{-i(E_n-E_m)(t-2\tau)}|S_{nm}^{(2)}|^2S_{n0}^{(1)*}J_{nm}S_{m0}^{(1)}. \quad (8)$$

We used $J_{mn}=-J_{nm}$, which is true for finite systems in one dimension, to exclude, e.g., "diagonal" terms with $n'=n$ and $m'=m$. As a function of real time, Eq. (8) gives a symmetric peak around $t=2\tau$, with the width determined mainly by the energy-level distance distribution. The peak height is, in this approximation, independent of delay τ.

Additional information on the sign of the echo current can be gained by comparison of Eq. (8) and the current right after the first pulse [see Eq. (4)],

$$J(t=0^+)=\sum_{n,m}S_{n0}^{(1)*}J_{nm}S_{m0}^{(1)}. \quad (9)$$

Each term in sum (9) also occurs in Eq. (8) multiplied by the negative factor $-|S_{nm}^{(2)}|^2$. This leads to the *heuristic sign rule*

$$J(t\approx 2\tau)\sim -J(t=0^+). \quad (10)$$

This heuristic derivation can be developed into a rigorous proof only for the $\chi^{(3)}$ limit, keeping terms $\sim\mathcal{E}_1\mathcal{E}_1^2$, but under extreme conditions the sign rule is expected to fail.

III. NUMERICAL RESULTS

For *non*interacting systems, chains with up to a few hundred sites, and ensembles of many thousands of chains, can easily be handled with desktop workstations. Thus the current echo in a noninteracting system has been studied extensively in detail.[12] However, in the figures of the present work, we only show results for small, half-filled systems with $N=6$ particles, to allow a direct comparison to the results for interacting particles. However, for their interpretation, we will benefit from Refs. 12 and 3.

The time-dependent current of an ensemble of noninter-

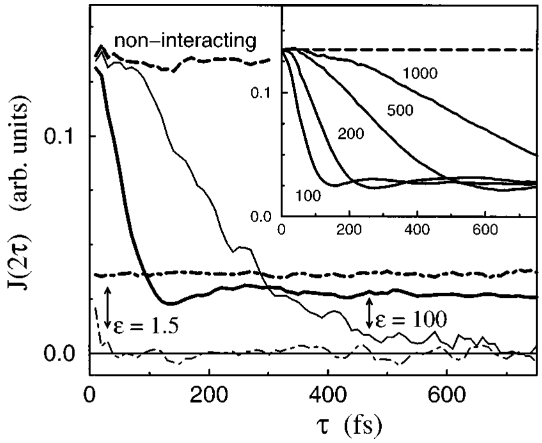

FIG. 2. Current echo peak as function of the delay time for different effective interaction strengths, $\epsilon_s = 1.5$ and 100; other parameters are as in Fig. 1. Thick lines: exact results, thin lines: TDHF approximation. Below 10 fs, the echo cannot clearly be separated from the direct current response. The inset shows exact results for $\epsilon_s = 100$, 200, 500, and 1000. Note that the decay time needed to reach the lower echo level is proportional to ϵ_s.

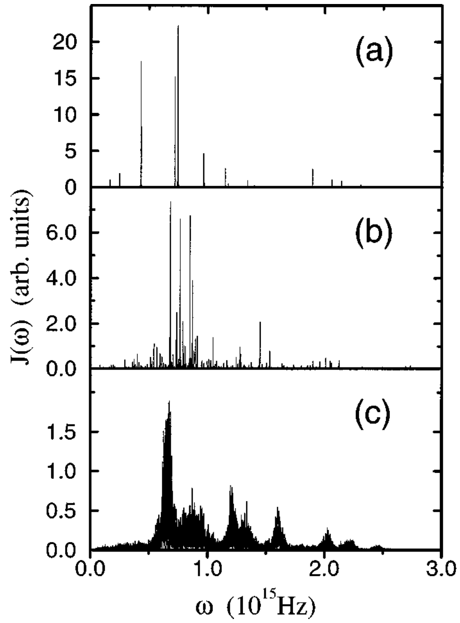

FIG. 3. Fourier transform of the polarization current of a single chain for the (a) noninteracting system, (b) exact solution including interaction, and (c) solution within the TDHF approximation; parameters as in Fig. 1.

acting chains is shown as Fig. 1(a). The echo peak in Fig. 1(a) is considerably smaller than the $\chi^{(3)}$ estimate (not shown), but coincides within a few percent or less with the RPA value [Eq. (8)]. It is found to be independent of τ. The strongest echos per unit length occur when the wave functions are localized within a few lattice constants, calling for short chain length or sizable disorder. This is fortunate, as for interacting particles we are restricted to such short chains in any case.

Now we turn to the interacting case. Again, rather strong disorder ($W = T$) and a chain length of $N = 6$ are used. Only $S_z = 0$ basis states contribute, and a 400×400 matrix has to be diagonalized. Figure 1(b) shows that current echos are possible in interacting systems, too.

However, the magnitude of the current echo peak is substantially reduced (Fig. 2). It depends on the delay time, the interaction strength, and pulse strengths. As a function of the delay time, a surprising behavior is seen: At very short delays the response is close to the corresponding ensemble of noninteracting chains. After falling rapidly to a considerably lower level, it stays almost constant at a value depending only weakly on ϵ_s. The delay time needed to reach the lower echo level, however, scales directly with ϵ_s (the inverse interaction strength).

Qualitatively, these results can be described as a partial dephasing, in contrast to noninteracting systems, which show no dephasing at all. (Note that any bath coupling is neglected, which, in general, would lead to *external* dephasing.) This result is of importance for our general understanding of dephasing in finite and isolated systems.

The echo in interacting systems is expected to be reduced due to the larger number of different energy eigenvalues. For weak interaction, the obvious delay dependence of the echo can be explained as follows: The many-particle spectrum is highly structured in the sense that *energy differences* are correlated. A large number of transitions between many-particle eigenstates correspond to a particular single-particle transition.[13] Their energies are only slightly modified by the Coulomb interaction with the other occupied single-particle states (Coulomb splitting δ_C). The energy differences within such a Coulomb bundle are approximately the same, and for times below $\hbar/\delta_C$ not only the cross terms survive in the sum (6), but nearly resonant terms $E_n - E_{n'} \approx E_{m'} - E_m$ as well. Processes involving the simultaneous excitation of two spin-degenerate single-particle states already contribute in order $\mathcal{E}_2^2 \mathcal{E}_1$. In the photon echo, these terms include the biexciton (n: biexciton, n' and m': exciton, m: ground state).

We find that the total echo scales like

$$J(t = 2\tau) \sim A + B \exp(-\tau^2 \delta_C^2/\hbar^2), \qquad (11)$$

where A/B decreases rapidly with the number of transitions induced by the second pulse. A very good fit to *all* the data in the inset of Fig. 2 is obtained with $\hbar/\delta_C = \epsilon_s$ 1.7 fs.

It should be stressed that form (11) is based on the assumption of two separate energy scales. One, given by the distribution of the noninteracting single-particle levels, i.e., by T and W, guarantees that the echo is a well-defined feature in real time; the other, much smaller one, given by the Coulomb splitting δ_C, determines the echo peak decay (dephasing) as a function of delay time.

This scenario is strongly supported by the Fourier decomposition of the actual current from a single chain, which is shown in Fig. 3. Clearly the single-particle excitation energies [peaks in Fig. 3(a)] split into groups of many-particle excitation energies [peaks in Fig. 3(b)].

IV. TIME-DEPENDENT HARTREE-FOCK APPROXIMATION

One might wonder whether the time-dependent Hartree-Fock approximation yields a current echo. The Hartree-Fock approximation trivially becomes exact for noninteracting systems, and we expect the TDHF approximation to show a current echo in this limit. In fact, the TDHF approximation in the form of the semiconductor Bloch equations (SBE),[14–16] has widely, and quite successfully, been applied to echo-related phenomena in semiconductors. On the other hand, echo phenomena are often discussed in terms of an effective time reversal for at least part of the wave function. The force fields from the HF energies are manifestly not time-reversal invariant, suggesting that the current echo in the TDHF approximation should decay at large delays.

Starting with an iteratively determined HF ground state, the single-particle wave functions or, equivalently, the density matrix, are propagated in time with a Runge-Kutta procedure. The thin lines in Fig. 2 show the ensemble-averaged TDHF current echo peak for $\epsilon_s = 1.5$ and 100. Starting again with the noninteracting value at $\tau = 0$, the echo peak falls to zero, as a signature of a complete dephasing—in clear contrast to the exact calculations. Even the initial decay time is qualitatively wrong (too long), and has been found to depend on the excitation level.

The Fourier decomposition of the single-chain current, Fig. 3(c), helps to understand the dephasing behavior of the TDHF approximation. The single-particle transition energies of Fig. 3(a) broaden into a continuum. In this respect, the TDHF result corresponds to what is expected for a system coupled to a bath. This, again, emphasizes the importance of higher-order correlations beyond the Hartree-Fock approximation, which ensure that for the full problem internal fields do *not* act like random fields.

For a discussion of the validity of the TDHF approximation and the SBE, it is useful to distinguish different uses of these terms. The TDHF approximation, in the form of an equation of motion, defines at any given time a unitary transformation of the occupied single-particle orbitals $p = 1, \ldots, M$. These can be expressed, analogously to Eq. (5), as

$$|\varphi_p(t)\rangle = \sum_{r,q} |\varphi_r^{(2)}\rangle e^{-i\epsilon_r^{(2)}(t-\tau)} s_{rq}^{(2)} e^{-i\epsilon_q^{(1)}\tau} s_{qp}^{(1)}, \tag{12}$$

where p, q, and r refer to eigenstates of the Hartree-Fock operators calculated with the momentary particle distributions at times $t<0$, $0<t<\tau$, and $t>\tau$, respectively.

Again, an echo results from the constructive interference of cross terms ($r' = q$, $q' = r$),

$$\langle \varphi_p(t)|\hat{j}|\varphi_p(t)\rangle \sim \mathcal{E}_1 |\mathcal{E}_2|^2 \sum_{r,q} e^{-i(\epsilon_r^{(2)}(t-\tau) - \epsilon_r^{(1)}\tau)} \times e^{-i(\epsilon_q^{(2)}(t-\tau) - \epsilon_q^{(1)}\tau)}, \tag{13}$$

where the single-particle matrix elements $s_{qp}^{(1,2)}$ of the operator $\exp(i\phi_{1,2}\hat{x}/a)$ have been expanded to the lowest order which contributes to the echo. This expansion of the matrix elements s was only for illustration and similiarity to standard procedures. The following arguments are based only on the form of the exponential factors, and are much more general. Now the cancellation of the exponents at $t = 2\tau$ in Eq. (13) is not complete because the Hartree-Fock potentials before and after the second pulse are different. This difference is, to lowest order, determined by the average interaction strength $\overline{v}$ and the amount of redistribution of occupation done by the second pulse $\overline{\delta n_2}$, which scales like $|\mathcal{E}_2|^2$,

$$\epsilon_q^{(2)} - \epsilon_q^{(1)} \sim \overline{v}\ \overline{\delta n_2}. \tag{14}$$

For short delay times and/or low excitation, the residual exponents in Eq. (13) are small, but for large delay times and/or high excitation density, the right-hand side terms of Eq. (13) do not longer interfere constructively, and the echo disappears on a time scale of

$$\tau_{\mathrm{TDHF}} \approx \frac{2\pi}{\overline{v}\ \overline{\delta n_2}}. \tag{15}$$

Note the density dependence, which is not present in Eq. (11). In passing, we state that this is different from the excitation induced dephasing of Refs. 17–19: A prepulse would be irrelevant here, insofar as only the difference of the phases accumulated during the intervals $[0^+, \tau^-]$ and $[\tau^+, 2\tau]$ matters.

Most applications of the TDHF approximation to the photon echo and related phenomena solve for the full time dependence but employ certain approximations. In particular, phenomenological dephasing rates are introduced.[20–22] With respect to four-wave mixing experiments, the incomplete cancellation in the exponent of Eq. (13) implies a signal decay, as long as the density dependence of the energies is kept. This is true, even if one retains only terms linear in $\mathcal{E}_1$ and focuses on the direction $(2\vec{k}_2 - \vec{k}_1)$ of the diffracted signal. Only if *all* the excitation dependence of the Hartree-Fock interaction is neglected,

$$\epsilon_p^{(2)} = \epsilon_p^{(1)} = \epsilon_p^{(0)}, \tag{16}$$

is the result an echo, which is independent of τ. This final simplification toward *strict* $\chi^{(3)}$ relies on the argument that the prefactor $s^{(1)*}s^{(2)*}s^{(2)}s^{(1)}$ is already of order $\mathcal{E}_1|\mathcal{E}_2|^2$. This clearly demonstrates that, even for weak excitation, higher orders in the exciting fields can become relevant for large (delay) times. Closely related to this *strict* interpretation of $\chi^{(3)}$ is the following: If one retains the excitation dependence of the Hartree-Fock single-particle energies but expands the exponent up to second order, one might obtain a quadratically *growing* signal.[6]

In the existing literature on echo phenomena in semiconductors dealing simultaneously with disorder and interaction, long- and short-range disorder are usually treated differently.[23,21] The former is assumed to yield an inhomogeneous broading and is treated as such, but the latter is lumped into a phenomenological dephasing time T_2 which also acounts for other dephasing, e.g., by phonons, and which might well dominate the dephasing seen in experiments. The importance of Coulomb effects outside the TDHF approximation as, e.g., the biexciton contribution, has been stressed recently.[24] The inclusion of disorder beyond the TDHF level, however, remains a challenge.

To summarize, qualitative and quantitative deviations of the TDHF behavior from the exact results exist, and TDHF calculations of echo phenomena have to be looked at critically, although for realistic system sizes no better approach seems to be feasible today.

V. CONCLUSIONS AND OUTLOOK

We conclude with a few ideas on the possible experimental verification of the current echo, even though the phenomenon of a *partial* dephasing is in no ways restricted to current echos.[25] The current echo as presented in Ref. 3 is part of the response to longitudinal homogeneous electric fields such as generated in a condenser. An intrinsic difficulty for the experimental realization is that the echo response cannot be spatially separated from the strong direct excitation, in contrast to the well-studied photon echo, where the signal is detected in the background-free diffracted direction.[2]

In the numerical results presented here, we chose as parameters those of polyacetylene, because this is an experimentally widely studied disordered one-dimensional system. Another, experimentally even more promising, candidate would be a semiconductor superlattice, where the vastly smaller hopping energy, $T<100$ meV, and the larger superlattice period, $a\gtrsim 5$ nm, reduce the necessary field strength considerably and increase the characteristic times involved. The one-dimensional character with disorder, which helps to localize the electronic excitations, is well realized in porous silicon or in semiconductor quantum wires, which are other systems worth looking at.

Finally, we mention that the main conclusions of this paper, (i) the existence of the current echo in interacting systems, (ii) its delay-dependence due to Coulomb splitting of single-particle levels, and (iii) its only partial dephasing, as well as (iv) the qualitatively wrong behavior of the TDHF approximation (strongly excitation-dependent echo decay) and its explanation, are fully supported by the behavior of two drastically simplified but rather different models which can be solved analytically almost completely:[26] an ensemble of $N=2$ chains with internal Coulomb interaction, and an ensemble of many two-level systems with infinite range interaction between all of them.

ACKNOWLEDGMENTS

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