Rigorous Bounds for Time-Dependent Correlation Functions*

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Rigorous upper and lower bounds are determined for time-dependent correlation functions, of the type used in statistical mechanics and spectroscopy. The input data are the values of any finite number of initial time derivatives of the correlation function. As an example, bounds are found for the classical velocity correlation function for a lattice vibration problem. The bounds are found to be much more accurate than the Taylor series based on the same time derivatives.

Time-dependent correlation functions are of fundamental importance in nonequilibrium statistical mechanics, transport theory,[1] scattering theory,[2] and spectroscopy.[3] Therefore it is important to have reliable methods to calculate their time dependence.

The initial time derivatives of a correlation function can often be calculated quite rigorously, since they can be expressed as equilibrium average properties of the system of interest. If a certain finite number of such initial time derivatives are calculated, we may ask to what extent this information determines the time dependence of the correlation function. In this Letter we give a rigorous and optimal answer to this question, for the commonly occurring class of correlation functions which have non-negative Fourier transforms. In particular, we prove that certain upper and lower limits exist, between which limits the correlation function must lie, when these initial time derivatives have specified values. Conversely, any correlation function whose values pass outside these upper and lower limits at any time cannot have the specified initial time derivatives. These upper and lower limits are of course functions of time, which bound the region allowed for the correlation function by the known initial time derivatives. We show how to construct these bounds, and also demonstrate that they are optimal, in the sense that they are the tightest bounds possible on the basis of the known information.

The proof of these results is given in Sect. I. Methods for the numerical construction of the bounds are outlined in Sect. II. An example is given, and these results are shown to be much more accurate than the Taylor series based on the same initial time derivatives.

(I) *Derivation of the bounds.*—The definition of a time autocorrelation function is

$$C(t) = \langle Q(0)Q(t)\rangle , \tag{1}$$

where $Q(t)$ is some dynamical quantity of interest (e.g., velocity of a diffusing particle, dipole moment of a molecule, etc.). The brackets signify an average over the equilibrium states of the system at the initial time (zero).

The real part of a correlation function can be represented as the cosine transform of a spectral density $I(\omega)$:

$$\mathrm{Re}\{C(t)\} = \int_{-\infty}^{\infty}\cos(\omega t)\, I(\omega)\, d\omega. \tag{2}$$

In many cases this spectral density $I(\omega)$ is known to be a *non-negative* function,

$$I(\omega) \geq 0.$$

(For example, the correlation function for the electric moment of a system corresponds to a spectral density proportional to the absorption coefficient for electromagnetic radiation.[3] This absorption is non-negative for systems in equilibrium.) The following derivation of bounds applies to correlation functions with non-negative spectral densities.

For the case of the real part of the correlation function, the relevant known features are the even-order initial time derivatives:

$$(d^{2k}/dt^{2k})\mathrm{Re}\{C(t)\}\,|_{t=0} = (-1)^k \mu_{2k}, \tag{3}$$

where the frequency moments μ_{2k} are defined by

$$\mu_{2k} = \int_{-\infty}^{\infty} \cos(\omega t)\, I(\omega)\, d\omega. \tag{4}$$

Typically, a certain set of these frequency moments can be calculated as equilibrium averages of the initial time derivatives, say for $k = 0, 1, 2, \ldots, n-1$. (Any other finite set of even moments could also be used.) We wish to determine what possible values of $\mathrm{Re}\{C(t)\}$ are consistent with these n constraints. These known values of the moments in Eq. (4) do eliminate many possible spectral densities $I(\omega)$. However, an uncountably infinite class of non-negative functions $I(\omega)$, all of which have the n correct moments, ordi-

narily remain possible for use in computing $\mathrm{Re}\{C(t)\}$ through Eq. (2). Our task is to find the range of values of $\mathrm{Re}\{C(t)\}$ corresponding to this infinite class of allowed $I(\omega)$ functions.

The theory of Tchebychev systems[4] forms the mathematical foundation for finding the allowed values of the correlation function. The factors in the integrands of the frequency moment constraints in Eq. (4),

$$1, \omega^2, \omega^4, \ldots, \omega^{2(n-1)}, \tag{5}$$

are a special case of a "Tchebychev system of functions." Definitions and properties of Tchebychev systems of functions are discussed in detail by Karlin and Studden.[4] Theorem III.6.4[5] can be applied to show that bounds to $\mathrm{Re}\{C(t)\}$ do exist, given the moment constraints, the non-negative character of $I(\omega)$, and the fact that $\cos\omega t$ is a continuous function of ω.

The distributions $I(\omega)$ which attain the upper and lower bounds are characterized by Theorem III.6.1,[6] as line spectra containing not more than n frequencies:

$$I(\omega) = \sum_{i=1}^{n} \rho_i \delta(\omega - \omega_i). \tag{6}$$

This very simple form of spectrum has non-negative weights ρ_i at frequencies ω_i. [$\delta(x)$ is the Dirac delta function.] The corresponding upper and lower bounds to the correlation function then have the simple form

$$\mathrm{Re}\{C(t)\} = \sum_{i=1}^{n} \rho_i \cos\omega_i t, \tag{7}$$

which is the cosine transform of the spectrum in Eq. (6). This representation theorem for the bounds is really our key result, since now the maximization (or minimization) of the correlation function can be carried out in the *finite*-dimensional space of the $2n$ values of ρ_i and ω_i, rather than in the infinite-dimensional space of all non-negative functions $I(\omega)$. While this maximization in the finite space is difficult, it can be carried out as discussed below, whereas any direct approach to the original infinite-dimensional problem is intractable.

(II) *Numerical evaluation of the bounds.*—The actual construction of the weights ρ_i and frequencies ω_i which optimize the correlation function must require that the n line spectra of Eq. (6) have the correct moments, i.e.,

$$\mu_{2k} = \sum_{i=1}^{n} \rho_i \omega_i^{2k}, \quad k = 0, 1, 2, \ldots, n-1, \tag{8}$$

and we also require $\rho_i \geq 0$. These constraints still leave n degrees of freedom for the $2n$ variables, and we must therefore search only an n-dimensional space to maximize (or minimize) the correlation function.

We have developed two independent numerical methods for solving this constrained optimization problem, a "matrix" method, and a "linear programming" method.

The matrix method is based upon representing the frequencies ω_i^2 as eigenvalues of a tridiagonal matrix, and the weights ρ_i as squares of the first components of the eigenvectors of that matrix. An algorithm has been developed[7] which gives matrix elements for a matrix of order $n/2$, such that the $n/2$ eigenvalues and weights thus defined automatically satisfy the constraint equations (8). Here we note that we can extend this matrix to order n, by adding $n/2$ arbitrary diagonal elements, and $n/2$ arbitrary (symmetric) codiagonal elements. The n eigenvalues and weights determined from this extended matrix will still automatically satisfy the constraint equations (8). The n arbitrary matrix elements required to extend this matrix are now taken to define the n-dimensional space in which the maximization (or minimization) of the correlation function can be carried out. Since this n-dimensional optimization problem is now *unconstrained*, any convenient numerical optimization method can be used. In all the cases we have studied so far, the optima have been found with most of the variable matrix elements zero. Thus the actual space to be searched is of very small dimension (e.g., 1 to 3).

In the linear programming method, the search for the ρ_i and ω_i values is conveniently divided into stages. In the first stage, we choose a grid of possible ω_i values with many (e.g., $\approx 10n$) frequency points spread across the region of frequencies expected to be important in the spectrum $I(\omega)$. The constraints in Eqs. (8) are *linear* in the ρ_i values, and we wish to maximize the correlation function, Eq. (7), which is also a *linear* function of the non-negative ρ_i values. This is a standard linear programming problem, which may be solved by the simplex algorithm.[8] Even though a fine grid of many trial ω_i values is taken initially, only n of the ρ_i values turn out to be nonzero in the solution which maximizes (or minimizes) the correlation function, as expected from the representation theorem.[6] The constraint equations (8) are obeyed exactly by the linear programming solutions, but the ω_i and ρ_i values are

not quite optimal. Nevertheless, the linear programming values for the *bounds* are quite accurate, even at this first stage of approximation.

The linear programming bounds may be refined by a second stage of approximation, in which we add a very finely spaced cluster of ω_i values around each of the n lines which had nonzero weights in the solution from the first approximation. The linear programming algorithm is applied again, to find a new set of n allowed lines, and the correspondingly refined bounds. This whole refinement process can be repeated, until the bounds have converged to their correct values. In practice, convergence of the bounds is quite fast, and even the second stage of refinement has usually not produced a significant change in the bounds. Of course, to obtain such good convergence, some care must be taken that the initial trial ω_i values span the relevant frequency region, and that enough (at least about $10n$) initial ω_i values are used.

The bounds which have been found for the time-dependent correlation function are the *best* that can be obtained from the known time derivatives. This optimal character of the bounds is clear from the method of construction. We have actually found a non-negative spectrum $I(\omega)$ which has the correct known moments. This spectrum produces, on Fourier transformation, a correlation function *equal* to, say, the upper bound. Thus we have a counter example against any other supposed "upper bound" which is lower than the one we obtained. Thus the bounds we obtain are the tightest possible on the basis of the known initial derivatives alone.

As an example of these bounds, we have calculated the classical velocity correlation function for a particle bound by nearest-neighbor harmonic forces in a cubic close-packed lattice. The first forty even time derivatives have been calculated[9] for this model. Upper and lower bounds to the correlation function calculated by the "matrix" and "linear programming" methods agreed, and are plotted in Fig. 1. The upper and lower bounds are indistinguishable over most of the time period plotted, only becoming separated at long times, when the initial time derivatives are no longer sufficient to determine the correlation function.

In order to express the accuracy of the bounds more clearly, the logarithm of the error (upper bound minus lower bound) is plotted in Fig. 2, for various numbers of initial derivatives. It is seen that the uncertainty grows essentially monotonically with time, for any given number of known derivatives. To obtain the correlation function to some specified level of uncertainty, one sees that the number of derivatives required grows roughly linearly with the largest time at which the correlation function is needed.

The traditional use for initial time derivatives of correlation functions has been to construct a truncated Taylor series expansion in powers of the time variable, to approximately represent the correlation function. However, this truncated Taylor series lies well *outside* our bounds, ex-

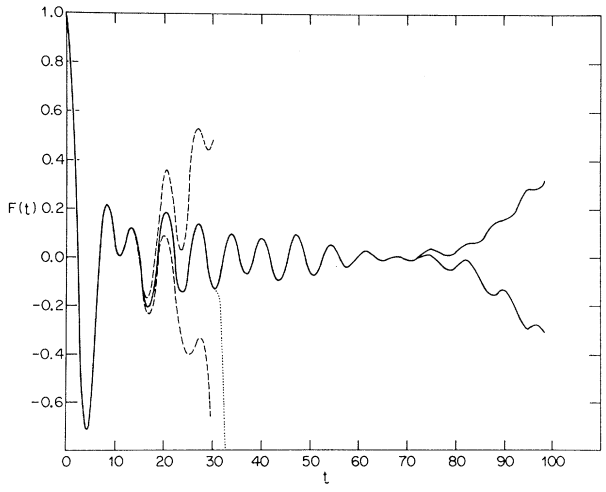

FIG. 1. Time-dependent velocity correlation function. Solid line, upper and lower bounds based on forty initial derivatives; dashed lines, based on ten initial derivatives; dotted line, truncated Taylor series expansion based on forty derivatives.

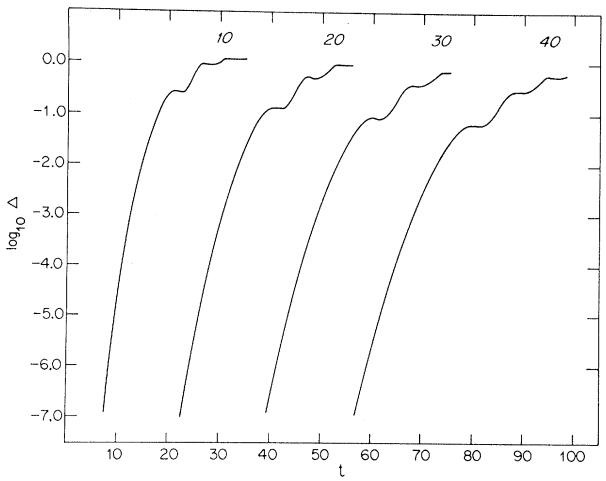

FIG. 2. Logarithm of the differences between the upper and lower bounds to the correlation function in Fig. 1, for the cases of ten, twenty, thirty, and forty known initial derivatives.

cept at very short times where the bounds and series are practically indistinguishable.

Since the bounds and the truncated Taylor series are based on exactly the same information, it may appear surprising that they disagree over most of the time range. The error must lie in the Taylor series result, since the bounds are rigorous. In truncating the Taylor series, one sets the higher (unknown) terms to zero. This implies that all the higher moments of $I(\omega)$ *vanish,* which is clearly inconsistent with $I(\omega)$ having strictly positive lower even moments. This inconsistency of the truncation procedure results in the truncated Taylor series values lying outside our bounds.

We conclude that our bounding technique for time correlation functions represents a major advance, in both accuracy and reliability, over the Taylor series representation based on the same initial time derivatives. Since we have also proven that our bounds are the best possible on the basis of this information, any further increases in accuracy must make use of some new information or other constraints on the correlation function.

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Breaking and Turbulent Transition in Ion Acoustic Waves

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New wave-particle processes determining the evolution of large-amplitude, low-frequency ion waves have been observed in numerical and laboratory experiments.

Previous studies of finite-amplitude ion waves have been concerned with hydrodynamic processes, which lead to steepening and soliton formation,[1] or else with wave-particle interactions,[2] notably "trapping." We describe here a new class of wave-particle processes which occur on *time scales intermediate* between those of hydrodynamic and "trapping" processes. These processes can become dominant, leading to final states which are turbulent.[3] Besides their fundamental interest, the processes may have technical importance in recently proposed schemes[4] for wave heating of plasmas.

It is well known[1] that finite-amplitude, low-frequency waves will distort and form a soliton pulse in the breakdown time $t_{br}=1/\omega\epsilon$, where ω is the angular frequency of the launched wave and $\epsilon=\delta n/n$ is the initial ion density perturbation. Wave-particle interactions, however, become important as the number of resonant ions increases; these are the ions which satisfy the inequality $|v-c|\leq(2e\varphi/m_i)^{1/2}$, where v is the ion velocity and φ is the wave potential. This number increases with $(\varphi)^{1/2}$, and also as $T_e/T_i\to 1$, since then the wave phase velocity, $c\cong v_{th}(T_e/T_i+3)^{1/2}$, approaches the ion thermal velocity, $(T_i/m_i)^{1/2}=v_{th}$.