Brief Reports

Brief Reports are accounts of completed research which, while meeting the usual **Physical Review** *standards of scientific quality, do not warrant regular articles. A Brief Report may be no longer than four printed pages and must be accompanied by an abstract. The same publication schedule as for regular articles is followed, and page proofs are sent to authors.*

Non-Markovian open-system boundary conditions for the time-dependent Schrödinger equation

James R. Hellums* and William R. Frensley
Eric Jonsson School of Engineering and Computer Science, University of Texas at Dallas, Richardson, Texas 75083-0688
(Received 11 June 1993)

The open-system boundary conditions for the one-dimensional Schrödinger equation are derived by dividing the unbounded domain into a finite system and two semi-infinite reservoirs. The resulting boundary conditions on the system are non-Markovian, as they contain a convolution over the history of the system. Thus, time-irreversibility arises in a pure-state problem. The propagator which appears in the boundary condition is derived for a simple discrete model. The correctness of the boundary conditions is verified and the usefulness of the discrete model is demonstrated by a numerical calculation of the time-evolution of a wave packet.

Many quantum systems are open, in the sense that they are coupled to their environment in a way that permits an exchange of particles.[1] Examples include any electron tunneling or wave-guide structures to which conductive leads are attached. To the extent that such systems are describable in terms of pure single-particle states, one would like to use the Schrödinger equation to model their behavior. Also, one typically wishes to expend a minimal effort describing the effects of the environment, and instead concentrate on the description of the system. This is most conveniently done by incorporating the effects of the environment into boundary conditions applied to the equations describing the system.

Open-system boundary conditions are necessarily time-irreversible.[1,2] The appropriate boundary conditions for the steady-state Schrödinger equation are well-established: $\psi + (i/k)d\psi/dx$ is fixed on each boundary.[3] For time-dependent problems, however, only approximate boundary conditions have been developed, and these will be discussed below. We present here a derivation of the exact open-system boundary conditions in a discrete space. These conditions are non-Markovian, in the sense that they contain a convolution over the past history of the system. The problem provides a physically transparent example of the origin of time-irreversibility and non-Markovian behavior. The solution also provides a practical computational scheme.

Our development of the open-system boundary conditions parallels that of Zwanzig's kinetic equation for the density matrix.[4] We begin with a one-dimensional, unbounded domain, and partition it into three regions: a semi-infinite left-hand reservoir, the finite system of interest, and a semi-infinite right-hand reservoir. (The use of the term "reservoir" anticipates the application of this theory to statistical problems, but for the present purposes the reservoirs simply contain that part of the wave function whose time-evolution is not evaluated in detail.) The wave function in these respective regions will be denoted by ψ_{rL}, ψ_s, and ψ_{rR}. Then Schrödinger's equation may be written as

$$i\hbar\frac{\partial}{\partial t}\begin{bmatrix}\psi_{rL}\\ \psi_s\\ \psi_{rR}\end{bmatrix}=\begin{bmatrix}H_{rL} & H_{iL}^{\dagger} & 0\\ H_{iL} & H_s & H_{iR}^{\dagger}\\ 0 & H_{iR} & H_{rR}\end{bmatrix}\begin{bmatrix}\psi_{rL}\\ \psi_s\\ \psi_{rR}\end{bmatrix}. \quad (1)$$

The components of the total Hamiltonian include H_{rL} and H_{rR}, which operate within the respective reservoirs, H_s, which operates within the system, and H_{iL} and H_{iR}, which couple the system to each reservoir. To derive the open-system boundary conditions, we will eliminate ψ_{rL} and ψ_{rR} from (1).

Let $\phi(s) = \mathcal{L}[\psi(t)]$, the Laplace transform of $\psi(t)$. Then (1) becomes

$$i\hbar\left\{s\begin{bmatrix}\phi_{rL}(s)\\ \phi_s(s)\\ \phi_{rR}(s)\end{bmatrix}-\begin{bmatrix}\psi_{rL}(0)\\ \psi_s(0)\\ \psi_{rR}(0)\end{bmatrix}\right\}=\begin{bmatrix}H_{rL} & H_{iL}^{\dagger} & 0\\ H_{iL} & H_s & H_{iR}^{\dagger}\\ 0 & H_{iR} & H_{rR}\end{bmatrix}\begin{bmatrix}\phi_{rL}(s)\\ \phi_s(s)\\ \phi_{rR}(s)\end{bmatrix}. \quad (2)$$

Solving for ϕ_{rL} and ϕ_{rR} in terms of ϕ_s and then substituting the results into (2) yields

$$\begin{aligned}i\hbar[s\phi_s-\psi_s(0)] = {} & H_s\phi_s + H_{iL}[(i\hbar s - H_{rL})^{-1}H_{iL}^{\dagger}\phi_s\\ & +(i\hbar s - H_{rL})^{-1}i\hbar\psi_{rL}(0)]\\ & +H_{iR}^{\dagger}[(i\hbar s - H_{rR})^{-1}H_{iR}\phi_s\\ & +(i\hbar s - H_{rR})^{-1}i\hbar\psi_{rR}(0)]. \end{aligned} \quad (3)$$

Let $(i\hbar s - H_r)^{-1} = g(s)$, the Green's function for the reservoir, with the inverse Laplace transform $\mathcal{L}^{-1}[g(s)] = G(t)$, the propagator. Note that $G(t)$ is an operator on the space of all functions ψ_r on the appropriate reservoir. (Also note that the Laplace transform naturally incorporates causality into g; no imaginary infinitesimals are required in the denominator.) Now taking the inverse Laplace transform of Eq. (3) leads to an augmented Schrödinger equation,

$$i\hbar\frac{d\psi_s}{dt} = H_s\psi_s(t) + H_{iL}\left[\int_0^t G_L(t-\tau)H_{iL}^\dagger\psi_s(\tau)\,d\tau + i\hbar G_L(t)\psi_{rL}(0)\right] + H_{iR}^\dagger\left[\int_0^t G_R(t-\tau)H_{iR}\psi_s(\tau)\,d\tau + i\hbar G_R(t)\psi_{rR}(0)\right]. \quad (4)$$

We see that in addition to the Schrödinger equation for the isolated system, we have two new terms associated with each reservoir: a term involving the initial ($t = 0$) state of the *reservoir* and a convolution integral over the history of the *system* from 0 to t. The presence of the convolution makes (4) a non-Markovian equation, and introduces the time-irreversibility required of an open system.

Observe that here non-Markovian irreversibility arises in a pure-state problem, and consequently there is no increase in the thermodynamic entropy, in contrast to the usual notions of irreversibility.[5] There is, however, a loss of information in some sense, as the quantum-mechanical amplitudes propagate into the reservoirs. The irreversibility of the present model is perhaps more mathematical than physical, but we believe that this is the simplest model in which non-Markovian irreversibility occurs.

Let us also point out the differences between the open-system model and the more conventional models of dissipative systems which are known to display non-Markovian irreversibility.[6,7] These models consist of a particle coupled to a heat bath. In this case, the Hilbert-space description of the state of the system plus the heat bath is defined by the direct product of the degrees of freedom of the system with those of the bath. In the open-system case, the space is naturally partitioned into a direct sum of the system and reservoir components as shown in (1). Thus the projection-operator techniques introduced by Zwanzig[4] apply directly to the open-system problem, even in the case of a pure quantum state.

The open-system Schrödinger equation (4) is quite general, but to demonstrate its usefulness we must apply it to a more specific model. We choose a one-dimensional discrete model such as is often derived from a tight-binding approximation, but we will view it as simply a finite-difference approximation to the continuum Schrödinger equation. The wave function is defined only on a discrete set of points $x_j = j\Delta x$, and we denote its value by $\psi_j = \psi(x_j)$. Then the second-order approximation to the Hamiltonian is of a tridiagonal form

$$(H\psi)_j = -c_j\psi_{j-1} + d_j\psi_j - c_{j+1}\psi_{j+1}. \quad (5)$$

We partition the discrete domain so that points $j \le 0$ are the left-hand reservoir, points $1 \le j \le J$ are the system, and points $j > J$ are the right-hand reservoir.

To apply (4) to the discrete model, we require the reservoir propagator $G(t)$. We will index the reservoir points by $l = 1, 2, 3, \ldots$, with $l = 1$ being the point adjacent to the system. One can derive the exact propagator for an unbounded uniform discrete model by any of a number of techniques, and then use the method of images to obtain $G_{ll'}(t)$ for the semi-infinite reservoir:

$$G_{ll'}(t) = (1/i\hbar)e^{-id_r t/\hbar} \times [i^{l-l'}J_{l-l'}(2c_r t/\hbar) - i^{l+l'}J_{l+l'}(2c_r t/\hbar)], \quad (6)$$

where J_n is a Bessel function of the first kind, and with c_r and d_r as defined in (5). Each interaction Hamiltonian H_i has only one nonzero element. Thus, in (4) only terms involving G_{1l} appear in the initial-value terms and only G_{11} appears in the convolution integral. This can be simplified to

$$G_{11}(t) = e^{-id_r t/\hbar}\frac{J_1(2c_r t/\hbar)}{ic_r t}. \quad (7)$$

One can also derive G_{11} from a recursion relation often used to model electron waveguides.[8]

To demonstrate the effectiveness of the open-system boundary conditions, a computer program for integrating (4) was developed. The Cayley time-discretization scheme[9] was generalized to include the memory integrals in (4). Because much of the motivation for the study of open systems comes from the recent progress of semiconductor technology, the program was tested by calculating the motion of an electron wave packet in the conduction band of GaAs. The domain was taken to be 50 nm wide and the potential was assumed to be constant. The mesh spacing was $\Delta x = 1.0$ nm and a time step of $\Delta t = 0.2$ fs was used. The initial wave packet was Gaussian,

$$\psi(x_j, 0) = e^{ik_0 x_j}\exp[-(x_j - x_c)^2/2\sigma^2], \quad (8)$$

centered at $x_c = 10$ nm, of width $\sigma = 10$ nm, and of average wave vector $k_0 = 0.7$ nm^{-1} (corresponding to an average energy $E_0 = 0.267$ eV). The results of this calculation are shown in Fig. 1, which verifies that reflectionless propagation through the boundary has indeed been achieved. If any components of the wave function were reflected, one would see interference effects in the probability density. Such interference is readily observable if the boundary conditions are not accurately implemented, as in the presence of minor programming errors. The results of Fig. 1 also illustrate the operation of the initial-value term in (4), as a part of the initial wave packet lies in the left-hand reservoir and subsequently propagates into the system.

While the example of Fig. 1 used a constant potential, the open-system boundary conditions still apply when the potential within the system is complicated and even time-dependent. The propagators (6) and (7) are correct so long as the potential within each reservoir is constant.

Prior work on open-system boundary conditions for Schrödinger's equation occurs in the context of wave-packet calculations like that illustrated above. The first such calculations circumvented the open-system problem by using a closed system large enough to model the phenomena of interest without generating reflec-

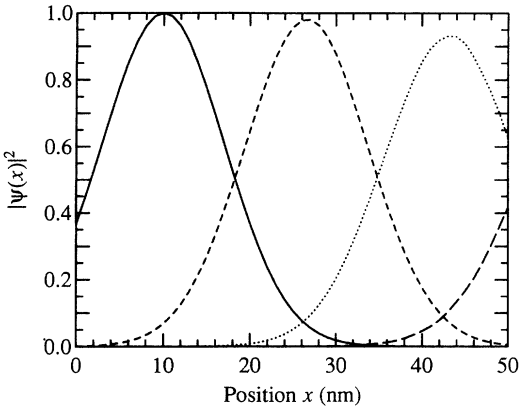

FIG. 1. Time evolution of an electron wave packet in an open system. The open-system boundary conditions are applied at $x = 0$ nm and $x = 50$ nm, and the simulation parameters are described in the text. The probability density density is plotted for the wave packet at the indicated times. Note the propagation of the wave packet into the system from the left-hand boundary, and the absence of interference or reflections as the wave packet passes through the right-hand boundary.

tions from the boundaries within the lifetime of those phenomena.[9,10] Another expedient involved adding a negative imaginary potential near the boundary to attenuate the wave function before it had an opportunity to reflect.[11,12] Recent interest in the simulation of tunneling devices has prompted the development of a number of improved, though still approximate, open-system boundary conditions.[13–16] In general, these schemes involve fitting the wave function in the vicinity of the boundary to a simple functional form which can then be extrapolated to find the value of the wave function on the boundary. This approach was introduced by Mains and Haddad,[13,14] and has recently been extended by Register, Ravaioli, and Hess.[15] In a technique developed by Shibata[16] the $\omega(k)$ dispersion relation is approximated over an appropriate region by a linear function, which leads to a boundary condition containing only first-order time and space derivatives. Shibata's work provides a Markovian approximation to the more general solution presented here. All of these methods work to varying degrees, over some energy range, depending upon the accuracy with which the wave function may be fit within their assumptions.

In a more general context, one often encounters spurious effects when one must truncate a set of basis functions in a quantum-mechanical calculation. An example occurs in chemical physics in the study of dissociative systems.[17] The general approach presented here, drawing upon the projection operator techniques of statistical physics, might be applicable to such problems if the appropriate propagators can be conveniently evaluated. Another possible application involves optical propagation within the paraxial approximation.[18] This approximation leads to a form of the Schrödinger equation, and in this case the open-system boundary conditions would represent radiation losses transverse to the direction of propagation.

In summary, we have presented the general solution to the open-system problem for the one-dimensional Schrödinger equation. The general formulation leads to a practical technique for simulating the time-evolution of quantum states within a simple discrete model. The open-system equation (4) also demonstrates, in a simple intuitive context, the origin of irreversible and non-Markovian behavior in a quantum system.

The authors wish to thank C. D. Cantrell, J. C. Englund, and L. Tamil for helpful suggestions and reading the manuscript.

* Permanent address: Semiconductor Group, Texas Instruments Incorporated, Dallas, TX 75265.

[1] W. R. Frensley, Rev. Mod. Phys. **62**, 745 (1990).
[2] R. Landauer, IBM J. Res. Dev. **1**, 223 (1957).
[3] C. S. Lent and D. J. Kirkner, J. Appl. Phys. **67**, 6353 (1990).
[4] R. Zwanzig, J. Chem. Phys. **33**, 1338 (1960).
[5] H. J. Kreuzer, *Nonequilibrium Thermodynamics and its Statistical Foundations* (Clarendon, Oxford, 1981), Chap. 11.
[6] A. O. Caldeira and A. J. Leggett, Physica **A121**, 587 (1983).
[7] V. Hakim and V. Ambegaokar, Phys. Rev. A **32**, 423 (1985).
[8] H. U. Baranger, D. P. DiVincenzo, R. A. Jalabert, and A. D. Stone, Phys. Rev. B **44**, 10 637 (1991).
[9] A. Goldberg, H. M. Schey, and J. L. Schwartz, Am. J. Phys. **35**, 177 (1967).
[10] I. Galbraith, Y. S. Ching, and E. Abraham, Am. J. Phys. **52**, 60 (1984).
[11] R. Kosloff and D. Kosloff, J. Comp. Phys. **63**, 363 (1986).
[12] D. Neuhasuer and M. Baer, J. Chem. Phys. **90**, 4351 (1989).
[13] R. K. Mains and G. I. Haddad, J. Appl. Phys. **64**, 3564 (1988).
[14] R. K. Mains and G. I. Haddad, J. Appl. Phys. **67**, 591 (1990).
[15] L. F. Register, U. Ravaioli, and K. Hess, J. Appl. Phys. **69**, 7153 (1991).
[16] T. Shibata, Phys. Rev. B **43**, 6760 (1991).
[17] M. Founargiotakis and J. C. Light, J. Chem. Phys. **93**, 633 (1990).
[18] B. E. A. Saleh and M. C. Teich, *Fundamentals of Photonics* (Wiley, New York, 1991), pp. 50–51.