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Renewal Theory and ac Conductivity in Random Structures

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A proper treatment of the continuous-time random-walk problem leads to a frequency-dependent conductivity in agreement with our earlier work.

The problem to be solved is the ac conductivity $\sigma(\omega)$ induced by a carrier hopping among a spatially random distribution of localized sites with the jump rate varying rapidly (exponentially) with the separation between pairs of sites.

The present authors[1] (SL) have replaced this intractable problem by a regular array of sites with a probability $\psi(t)dt$ for a hop between successive sites in the time interval $(t, t+dt)$. To insure that this continuous-time random-walk (CTRW) representation of the original problem is reasonable, the density $\psi(t)$ is chosen to reflect the distribution of jump rates associated with the distribution of site separations in the original problem. Successive jumps are treated as independent with the same available-jump-time distribution. The success of our procedure in correlating with the experimental frequency dependence of $\omega^{0.8}$ is related to the ergodic nature of the conductivity process. In traversing the crystal a carrier samples a wide variety of environments. The distribution of hopping times over the entire crystal is, in our model, folded into the hopping-time distribution of a single site. In a discussion of our model Tunaley[2] predicted a frequency-independent conductivity.

Tunaley[2] has made a *literal* interpretation of our CTRW procedure as an ongoing *renewal process*. A renewal process is a stochastic sequence of events in which the time interval between successive events (steps, or hops) is governed by a common normalized probability density $\psi(t)$. He then invokes a theorem of Feller which states that for such a process the *first* step starting from a *random* time of observation has a density $h(t)$ which differs from the density $\psi(t)$ which describes the time intervals between all subsequent steps. An unfortuante consequence is that the conductivity obtained by convoluting $h(t)$ for the first step with an arbitrary number of $\psi(t)$ steps leads to a frequency-independent conductivity, in complete disagreement with experiment and with calculations in SL.

Our point is that identification of the hopping problem with a renewal process (ongoing for a long time) prevents an investigation of the frequency dependence of the conductivity. Feller *constructs* the $h(t)$ so that a uniform occurrence rate for the events (hops) is insured at a random time. This uniform occurrence rate is equivalent to a time-independent flux and, therefore, a frequency-independent conductivity. Thus we do not suggest that Feller's theorem is incorrect, but that the hopping problem cannot be directly

reduced to an ongoing renewal process (as Tunaley did) without a careful examination of the underlying physical problem.

We shall show later that, because of the nature of Gibbsian ensembles, the bias introduced by Tunaley into the first step should not be applied to an impurity hopping problem. To understand the nature of this bias we first shall supply a new, succinct proof of Feller's theorem. We shall introduce an equivalent but more informative expression for the first-step density $h(t)$. This expression depends on the conditional waiting-time density, a quantity not considered or evaluated by Feller. We now define

$$\Phi(\tau) \equiv \int_{\tau}^{\infty} \psi(t)\, dt \tag{1}$$

as the probability of no event occurring in time τ after the last event, and use

$$\psi(t|\tau) = \psi(t+\tau)/\Phi(\tau) \tag{2}$$

as the conditional density for an event at a time t knowing that a time τ has already elapsed since the last event. Thus $\psi(t) = \psi(t|0)$. To obtain $h(t)$ we must average $\psi(t|\tau)$ over the density $\Phi(\tau)$ of lifetimes τ:

$$h(t) = \int_0^{\infty} \psi(t|\tau)\Phi(\tau)\, d\tau \Big/ \int_0^{\infty} \Phi(\tau)\, d\tau. \tag{3}$$

This instructive expression for $h(t)$ is equivalent to the one presented in Feller's theorem:

$$h(t) = [1 - \textstyle\int_0^t \psi(\tau) d\tau]/\bar{t}, \tag{4}$$

where

$$\bar{t} \equiv \int_0^{\infty} t\, \psi(t)\, dt = \int_0^{\infty} \Phi(t)\, dt. \tag{5}$$

For some examples $\psi(t)$ can be expressed as a superposition of jump rates so that

$$\psi(t) = \int_0^{\infty} w \exp(-wt) p(w)\, dw; \tag{6}$$

then from Eq. (4) for $h(t)$, we get

$$h(t) = \left[\int_0^{\infty} w \exp(-wt) \frac{p(w)}{w}\, dw\right] \left(\int_0^{\infty} \frac{p(w)}{w}\, dw\right)^{-1}. \tag{7}$$

If one compares Eq. (7) with Eq. (6) we see that the bias on the first hop is a weighting by the lifetime factor $1/w$.

The conductivity $\sigma(\omega)$ was shown in SL, by linear-response theory, to be reducible to

$$\sigma(\omega) = (ne^2/kT) D(\omega), \tag{8}$$

where the complex frequency-dependent diffusion constant $D(\omega)$ is given by

$$D(\omega) = -\tfrac{1}{6}\omega^2 \int_0^{\infty} e^{-i\omega t} \mathrm{tr}\{[\vec{r}(t) - \vec{r}(0)]^2 \rho\}\, dt \tag{9}$$

and ρ is the density operator. The ac conductivity in Eqs. (8) and (9) is proportional to a Fourier component of the mean-square displacement in the field-free ensemble. In the hopping-conductivity problem, Eq. (9) can be reduced[1] to a sum over discrete sites so that

$$D(\omega) = -\tfrac{1}{6}\omega^2 \sum_{\vec{s}, \vec{s}_0} (\vec{s} - \vec{s}_0)^2 \int_0^{\infty} \langle P(\vec{s}, t; \vec{s}_0, 0; W)\rangle e^{-i\omega t} dt, \tag{10}$$

where $P(\vec{s}, t; \vec{s}_0, 0; W)$ is the thermal-averaged two-time probability distribution for finding a carrier at $\vec{s}_0$ at time 0 and $\vec{s}$ at time t for a given configuration described by a set of transition probabilities $W_{\vec{s}_1, \vec{s}_2}$ summarized briefly by the symbol W. The angular brackets represent an average over configurations.

If $f(\vec{s}, W)$ is the steady-state distribution for a particular configuration of impurities characterized by W and $P(\vec{s}, t, W|\vec{s}_0, 0)$ is the conditional probability for arriving at $\vec{s}$ at time t given a start at $\vec{s}_0$ at time 0 associated with the same configuration, then the ensemble-averaged two-time distribution is given by

$$\langle P(\vec{s}, t; \vec{s}_0, 0; W)\rangle = \langle P(\vec{s}, t, W|\vec{s}_0, 0) f(\vec{s}_0, W)\rangle. \tag{11}$$

If a particular site $\vec{s}_0$ in a particular configuration has few near neighbors so that the rate of leaving the site is low, we expect the occupancy probability $f(\vec{s}_0, W)$ to be proportional to the reciprocal

$$\left(\textstyle\sum_{\vec{s}'} W_{\vec{s}', \vec{s}_0}\right)^{-1} \tag{12}$$

of the total rate of leaving site $\vec{s}_0$. This lifetime bias factor, which is shown in Eq. (7) to modify the first-hopping probability, thus manifests itself in the dependence on W of the steady-state occupancy

$f(\vec{s}_0, W)$. The effect desired by Tunaley is then produced by the correlation between the conditional probability and the occupancy factor in Eq. (11) induced by the dependence of both these factors on W.

It must be recognized, however, that in a Gibbsian ensemble, site $\vec{s}_0$ with a given energy E_0 has an occupancy probability determined by E_0 and the temperature T and independent of the transition rates:

$$f(\vec{s}_0, W) = f(\vec{s}_0). \tag{13}$$

(This result follows from the fact that detailed balance is maintained in each member of the ensemble, so that if the jump rate is reduced from site 1 to 2 it must be reduced proportionately from 2 to 1, thus maintaining the occupancy probabilities independent of the transition rates.) Without approximation, therefore, we can use

$$\langle P(\vec{s}, t; \vec{s}_0, 0; W)\rangle = \langle P(\vec{s}, t, W|\vec{s}_0, 0)\rangle f(\vec{s}_0). \tag{14}$$

Thus only the conditional probability remains to be computed. The CTRW model is therefore *applied only to the conditional probability* and not to the two-time probability in order to maintain the exact relation, Eq. (14). Furthermore, Eq. (14) tells us to start our carrier *definitely* at position $\vec{s}_0$, thus providing an initial condition for the CTRW problem. Only at the final stage of evaluating the diffusion coefficient by means of Eqs. (10), (11), and (14) does one average over the density $f(\vec{s}_0)$ of initial positions.

The conditional probability will be written below as a sum of terms. The nth term is the probability for getting from $\vec{s}_0$ to $\vec{s}$ in n steps. This nth term can be factored into probabilities for each step. The factor for the first step is identical to the factor for later steps. *Since the factor $f(\vec{s}_0)$ has been separated, there is no way to treat the first step differently from the later steps.* Thus, biasing of the first step, proposed in Ref. 2, does not occur.

Let us consider now the random-walk problem on a cubic lattice with jump rate $W_{\vec{s},\vec{s}'}$ from $\vec{s}'$ to $\vec{s}$. The probability density $P(\vec{s}, t, W|\vec{s}_0, 0)$ conditioned on starting at site $\vec{s}_0$ at time 0 obeys

$$\partial P(\vec{s}, t, W)/\partial t = -\Gamma_{\vec{s}} P(\vec{s}, t) + \sum W_{\vec{s},\vec{s}'} P(\vec{s}', t), \tag{15}$$

where

$$\Gamma_{\vec{s}} = \textstyle\sum_{\vec{s}'} W_{\vec{s}',\vec{s}} \tag{16}$$

and for simplicity of notation we have suppressed the initial conditions. Then the Laplace transform

$$\hat{P}(\vec{s}, u, W|\vec{s}_0, 0) \equiv \int_0^\infty e^{-ut} P(\vec{s}, t, W|\vec{s}_0, 0)\, dt \tag{17}$$

obeys

$$(u + \Gamma_{\vec{s}})\hat{P}(\vec{s}, u) = \textstyle\sum_{\vec{s}'} W_{\vec{s},\vec{s}'} \hat{P}(\vec{s}', u) + \delta_{\vec{s},\vec{s}_0}, \tag{18}$$

$$\hat{P}(\vec{s}, u, W|\vec{s}_0, 0) = \hat{\Phi}(\vec{s}, u, W)[\hat{\psi}(\vec{s} - \vec{s}_0, u, W) + \hat{\psi}(\vec{s} - \vec{s}', u, W)\hat{\psi}(\vec{s}' - \vec{s}_0, u, W) + \hat{\psi}(\vec{s} - \vec{s}', u, W)\hat{\psi}(\vec{s}' - \vec{s}'', u, W)\hat{\psi}(\vec{s}'' - \vec{s}_0, u, W) + \ldots], \tag{19}$$

where repeated indices are understood to be summed and

$$\hat{\psi}(\vec{s} - \vec{s}', u, W) \equiv W_{\vec{s},\vec{s}'}/(u + \Gamma_{\vec{s}}), \tag{20}$$

$$\hat{\Phi}(\vec{s}, u, W) \equiv 1/(u + \Gamma_{\vec{s}}). \tag{21}$$

Suppose now that the $W_{\vec{s},\vec{s}'}$ are not fixed but selected from an ensemble of possible values compatible with statistical mechanics. Then an ensemble average must be taken over the right-hand side of Eq. (19). Because the factor $f(\vec{s}_0)$ does not enter this average, as shown in Eq. (14), the $\hat{\psi}$ associated with the first jump [the last factor in any term of Eq. (19)] appears on an equal footing with all other $\hat{\psi}$'s and does not receive the special treatment advocated by Tunaley.

The CTRW procedure which treats all sites as equivalent and independent corresponds in the present language to a factorized ("Hartree approximation") average of Eq. (19) which leads to

$$\hat{P}(\vec{s}, u|\vec{s}_0, 0) = \hat{\Phi}(\vec{s}, u)[\hat{\psi}(\vec{s} - \vec{s}_0, u) + \hat{\psi}(\vec{s} - \vec{s}', u)\hat{\psi}(\vec{s}' - \vec{s}_0, u) + \hat{\psi}(\vec{s} - \vec{s}', u)\hat{\psi}(\vec{s}' - \vec{s}'', u)\hat{\psi}(\vec{s}'' - \vec{s}_0, u) + \ldots], \tag{22}$$

where

$$\hat{\psi}(\vec{s}-\vec{s}', u) \equiv \langle \hat{\psi}(\vec{s}-\vec{s}', u, W)\rangle, \tag{23}$$

$$\hat{\Phi}(\vec{s}, u) \equiv \langle \hat{\Phi}(\vec{s}, u, W)\rangle. \tag{24}$$

Equation (22) is identical to the result of our CTRW procedure.[4] Since $\hat{\psi}(\vec{s}-\vec{s}', u)$ is the Laplace transform of $\psi(\vec{s}-\vec{s}', t)$, the latter is defined by

$$\psi(\vec{s}-\vec{s}', t) \equiv \langle W_{\vec{s},\vec{s}'} \exp(-\Gamma_{\vec{s}} t)\rangle. \tag{25}$$

The definition, Eq. (25), was in fact used by SL to calculate $\psi(\vec{s}-\vec{s}', t)$.

The factorization procedure is the approximation which makes the resulting problem tractable. A treatment including the effects of correlations between jumps would be desirable, but it too must treat the first jump on an equal footing with the others. The main point of the above discussion is that as a consequence of the general validity of statistical mechanics the occupancy $f(\vec{s}_0)$ associated with the starting point factors out, and all jumps, including the first, are to be treated alike.

[1]H. Scher and M. Lax, Phys. Rev. B 7, 4491 (1973).
[2]J. K. E. Tunaley, Phys. Rev. Lett. 33, 1037 (1974).
[3]W. Feller, *An Introduction to Probability Theory and Its Applications* (Wiley, New York, 1971), Vol. 2, 2nd ed., Chap. XI, Sect. 4.
[4]For a more complete discussion, the reader is referred to Appendix B of Ref. 1, where a comparison is made between the procedure outlined here and the ensemble average of a random walk on a random media.

Limits on the Radiative Decay of Neutrinos

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Astronomical observations at the x-ray, optical, and radio frequencies are used to show that the lifetime of neutrinos for radiative decay divided by the rest mass, τ_0/m_ν, exceeds 10^{17} sec/eV. If one makes the further assumption that $m_\nu > 10^{-3}$ eV, then $\tau_0 \geq 10^{19}$ sec. If there are other competing decays of neutrinos, it is then shown that $\Gamma_{\nu_e}(\nu_e \to x+\gamma)/\Gamma_{\nu_e}(\text{total}) \leq 10^{-15}$ and $\Gamma_{\nu_\mu}(\nu_\mu \to x+\gamma)/\Gamma_{\nu_\mu}(\text{total}) \leq 3\times 10^{-6}$.

In this paper I discuss the limits that can be placed on the radiative instability of the neutrinos from various kinds of observations. In this regard I am motivated by several recent papers[1-5] which have considered the possibility that neutrinos could have finite rest mass and could therefore decay. One particular set of these[3,4] considers the mixing of ν_e and ν_μ and predicts observable widths for the lepton-number–nonconserving decays, such as $\mu \to e+\gamma$ and $\nu_\mu \to \nu_e+\gamma$. Independent of such theoretical considerations it is worthwhile to study the observational limits on such processes.

The astrophysical environment provides excellent possibilities for such a study of very weak processes: Path lengths of $\sim 10^{28}$ cm are available for the decay process to take place, huge in comparison with the $\sim 10^2$ cm available in most laboratory studies. Also there are regions, such as the cores of very hot stars, where the weak processes dominate, as the products of the competing electromagnetic channels are suppressed completely because of the enormous time scales needed for the diffusion of photons to the stellar surface.[6]

Recognizing that both neutral and charged currents are of comparable strength in weak interactions, one finds that there are several locations in nature where copious generation of ν_e, ν_μ, etc., takes place. In this Letter I consider at first that the neutrino decays solely through the channel

$$\nu \to x+\gamma, \tag{1}$$

where x is any particle with a mass smaller than m_ν, and consider later the effects due to compet-