leaves the first integral to evaluate. Now from (6.4) and (6.18), we see that

$$\mathcal{L}_{10}=\mathbf{U}\cdot\frac{\partial}{\partial\mathbf{R}}+\frac{1}{m}\sum_{j=1}^{N}\frac{\partial\theta(\mathbf{r}_j-\mathbf{R})}{\partial\mathbf{r}_j}\cdot\frac{\partial}{\partial\mathbf{U}}, \tag{A4.8}$$

and a short calculation yields

$$(1-\pi_{00})\mathcal{L}_{10}P_{00}f'(t-t')=\beta P_{00}\mathbf{F}\cdot\left(\mathbf{U}+\frac{\partial}{\partial\mathbf{U}}\right)f'(t-t'), \tag{A4.9}$$

where we have set

$$\mathbf{F}=-\frac{1}{m\beta}\frac{\partial}{\partial\mathbf{R}}\sum_{j=1}^{N}\theta(\mathbf{r}_j-\mathbf{R}). \tag{A4.10}$$

Finally, we see from (6.18) and (A4.10) that we can write

$$\Theta_{10}=\beta\mathbf{F}\cdot\frac{\partial}{\partial\mathbf{U}}. \tag{A4.11}$$

Therefore, we get the result that

$$K_{20}(t',N)f'(t-t')=\beta^2\int P_{00}F_\mu e^{-t'\mathcal{L}_{00}}F_\nu\prod_{j=1}^{N}d\mathbf{r}_jd\mathbf{v}_j\frac{\partial}{\partial U_\mu}\left(U_\nu+\frac{\partial}{\partial U_\nu}\right)f'(t-t'). \tag{A4.12}$$

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Quantum Noise. IV. Quantum Theory of Noise Sources

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For a quantum system describable in a Markoffian way, via a set of variables $\mathbf{a}=\{a_1,a_2,\cdots\}$, we show that the Langevin noise sources F_μ in the operator equations of motion $da_\mu/dt=A_\mu(\mathbf{a})+F_\mu$ possess second moments $\langle F_\mu(t)F_\nu(u)\rangle=2\langle D_{\mu\nu}(\mathbf{a},t)\rangle\delta(t-u)$. The diffusion coefficients $D_{\mu\nu}$ can be determined from a knowledge of the mean equations of motion via the (exact) time-dependent Einstein relation $2\langle D_{\mu\nu}\rangle=-\langle A_\mu a_\nu\rangle-\langle a_\mu A_\nu\rangle+d\langle a_\mu(t)a_\nu(t)\rangle/dt$, where $\langle\ \rangle$ represents a reservoir average. The sources F_μ,F_ν do not commute with one another, and as a result the commutation rules of the a_μ are shown to be preserved in time. The mean motion and diffusion coefficients are calculated for a harmonic oscillator, and for a set of atomic levels. We prove that two dynamically coupled systems (e.g., field and atoms) have uncorrelated Langevin forces if they are coupled to independent reservoirs. Radiation-field–atom coupling adds no new noise sources. We thus obtain simply the maser model including noise sources used in Quantum Noise V. *Direct* calculations of the mean motion and fluctuations in a system coupled to a reservoir yield relationships in agreement with the Einstein relation. For reservoirs violating time reversal, anomalous frequency shifts are found possible that violate the Ritz combination principle since $\Delta\omega_{12}+\Delta\omega_{23}+\Delta\omega_{31}$ need not vanish.

1. INTRODUCTION AND SUMMARY

OUR treatment of quantum noise in this paper and the preceding papers in this series[1] closely parallels a corresponding discussion of noise in classical systems.[2] The first paper (I) in our classical series provides a quasilinear approach to stationary Markoffian random processes. In the quasilinear case, it was easy to obtain the corresponding Langevin theory of noise sources. This work was generalized in (III) and (IV) to include classical nonstationary nonlinear Markoffian processes treated first from a Markoffian point of view and second from a Langevin noise-source point of view. In Paper IV, we emphasized the advantages and flexibility associated with the Langevin noise-source approach.

The present paper extends the noise-source technique to quantum systems. Quantum (and classical) systems experience dissipation and fluctuations through interaction with a reservoir. Our philosophy is that the reservoir can be *completely eliminated* provided that the frequency shifts and dissipation induced by the reser-

[1] A reference to QV is a reference to the author's fifth paper on quantum noise and relaxation. QI: Phys. Rev. **109**, 1921 (1958); QII: Phys. Rev. **129**, 2342 (1963); QIII: J. Phys. Chem. Solids **25**, 487 (1964); QIV: present paper; QV: in *Physics of Quantum Electronics*, edited by P. L. Kelley, B. Lax, and P. E. Tannenwald (McGraw-Hill Book Company, Inc., New York, 1966); QVI: "Moment Treatment of Maser Noise" (unpublished); QVII: "The Rate Equations and Amplitude Fluctuations" (unpublished).

[2] A reference to IV is a reference to the author's fourth paper on classical noise. I: Rev. Mod. Phys. **32**, 25 (1960); II: J. Phys. Chem. Solids **14**, 248 (1960); III: Rev. Mod. Phys. **38**, 359 (1966); IV: Rev. Mod. Phys. (to be published); V: Bull. Am. Phys. Soc. **11**, 111 (1966); VI: *ibid.*

voir are incorporated into the mean equations of motion, and provided that a suitable operator noise source with the correct moments are added.

Thus, if $\mathbf{a}=\{a_1,a_2,\cdots\}$ is some set of system operators, and

$$d\langle a_\mu\rangle/dt=\langle A_\mu(\mathbf{a})\rangle \tag{1.1}$$

are the correct mean[3] equations of motion including frequency shifts and damping, then we propose that

$$da_\mu/dt=A_\mu(\mathbf{a})+F_\mu(\mathbf{a},t) \tag{1.2}$$

is a valid set of *operator* equations provided that the operators F_μ are endowed with the correct statistical properties. Thus the reservoir has been replaced by the familiar electrical engineer's black box describable by an impedance (the dissipative resistive terms and frequency-shift–producing reactive terms incorporated into A_μ) and an associated noise source F_μ.

The mean equations of motion in more fundamental recent papers on noise in masers are obtained by eliminating the reservoir to second order in perturbation theory. If one calculates, for example, the time rate of change of the occupancy of some state, one obtains the usual sort of transport equation [see Eq. (1.13) with $i=j$] whose coefficients are transition probabilities calculated to second order. One then adopts the *form* of the resulting equations of motion as a model for a maser—but regards the coefficients as experimentally determined, i.e., as the correct, not the second-order transition probabilities. Moreover, the models usually chosen are Markoffian in the sense that the future of all operators (or equivalently of the density matrix) is determined by the present without requiring an integration over past histories.

Although the F's are operators, for most problems we only need to know the reservoir averages over low-order moments and commutators of these operators. We regard our task then, as the determination of the moments of the F_μ, in terms of the *experimental dissipation coefficients, within a Markoffian description.*

The most obvious method of attack is to calculate the reservoir contribution to A_μ to second order in the system-reservoir interaction V [see Appendices A and B]. Then one must calculate the mean moment $\langle F_\mu(t)F_\nu(u)\rangle$ to second order in V (see Appendix B). By comparing the coefficients in these two calculations we arrive at a fluctuation-dissipation relation valid for nonequilibrium situations. We shall show in Sec. 2, that the relation so obtained is indeed *exact.*

To see how to implement this program, we note that Eqs. (1.1) and (1.2) have been so chosen that the first moment of the Langevin forces vanish:

$$\langle F_\mu\rangle=0\,,\quad A_\mu(\mathbf{a},t)=\langle a_\mu(t+\Delta t)-a_\mu(t)\rangle/\Delta t\,. \tag{1.3}$$

Thus, by computing the change in a_μ over some suitable short time interval[4] Δt, due to the interaction V, we can determine the reservoir contribution to A_μ.

Next we note, that if the reservoir forces possess a finite correlation time, i.e., $\langle F_\mu(t)F_\nu(u)\rangle\neq 0$ for $|t-u|\sim\tau_c$, the system will acquire a memory of the past and become non-Markoffian. Thus we shall take our moments of the random forces F_μ in the form

$$\langle F_\mu(t)F_\nu(u)\rangle=2\langle D_{\mu\nu}(\mathbf{a},t)\rangle\delta(t-u)\,. \tag{1.4}$$

[A direct proof that $\langle F_\mu(t)F_\nu(u)\rangle=0$ for $t\neq u$ is given in IV (8.12) for the classical case, and in (2.19) for the quantum case.]

Setting $t=s$, $u=s'$ and integrating (1.4) from t to $t+\Delta t$ on both these variables, we obtain[5]

$$2\langle D_{\mu\nu}\rangle=\frac{1}{\Delta t}\int_t^{t+\Delta t}ds\int_t^{t+\Delta t}ds'\langle F_\mu(s)F_\nu(s')\rangle\,. \tag{1.5}$$

Integrating Eq. (1.2) over an interval Δt, and inserting the results into (1.5), we obtain

$$2\langle D_{\mu\nu}\rangle=\langle[a_\mu(t+\Delta t)-a_\mu(t)][a_\nu(t+\Delta t)-a_\nu(t)]\rangle/\Delta t \tag{1.6}$$

after discarding terms $(A_\mu\Delta t)(A_\nu\Delta t)/\Delta t$ that disappear as $\Delta t\rightarrow 0$. Equation (1.6) is reminiscent of the traditional definition of a spatial diffusion coefficient:

$$2D=\langle[x(t)-x(0)]^2\rangle/t\,.$$

Equation (1.6) is also the basis of our direct perturbation calculation of the diffusion coefficients in Appendix B. We calculate the change Δa_μ in a_μ over the time interval Δt induced by the interaction V, and average $\Delta a_\mu\,\Delta a_\nu$ over the reservoir variables. Since this average depends on the order of the factors, $D_{\mu\nu}$ is not symmetric. Thus F_μ and F_ν *do not commute.*

The *exact* method of Sec. 2 consists in noticing that the equation of motion (2.7) for $\langle a_\mu a_\nu\rangle$ involves $\langle D_{\mu\nu}\rangle$ and emphasizing that this equation can be inverted to solve for $\langle D_{\mu\nu}\rangle$:

$$2\langle D_{\mu\nu}\rangle=-\langle A_\mu a_\nu\rangle-\langle a_\mu A_\nu\rangle+d\langle a_\mu(t)a_\nu(t)\rangle/dt\,. \tag{1.7}$$

This equation is well known to us as I (5.12). What is new is that we notice that a knowledge of the *mean motion* of all operators provides us with the *fluctuation* moments $D_{\mu\nu}$. In the steady state, the last term of (1.7)

[3] We use single brackets, $\langle\ \rangle$, to denote an average over an ensemble of reservoirs. However, we are dealing with a single system. Thus $\langle a_\mu\rangle$ is still a system operator. A subsequent average over an ensemble of systems will be denoted by double brackets. Thus $\langle\langle a_\mu(t)\rangle\rangle$ is now a number, but it may be time-dependent, if the system is started off in a nonsteady state at time t_0. We will use $\langle\langle a_\mu\rangle\rangle_{ss}$ or $\bar{a}_\mu$ to denote the steady-state system average.

[4] We shall choose this time Δt to be long compared to the reciprocal natural frequency $(\omega_s)^{-1}$ of the system, and short compared to the system relaxation time Γ^{-1}. More precise restrictions will be given in footnote 5 below.

[5] We actually assume that the correlation time τ_c of the Langevin forces is short compared to all system *relaxation* times, Γ^{-1}, but not zero. This correlation time is usually *long* compared to the reciprocals of the natural frequencies $(\omega_s)^{-1}$ of the system. In such a case, the system behaves in an essentially Markoffian way when changes are observed over time intervals Δt that obey $(\omega_s)^{-1}<\tau_c<\Delta t\ll\Gamma^{-1}$. Thus the diffusion coefficients (1.5) or (1.6) are calculated in Appendix B by the use of such time intervals. The motion of $\mathbf{a}(t)$ in $F_\mu(\mathbf{a}(t),t)$ during such time intervals is important as shown in Appendices A and B and in IV, Secs. 5, 6.

can be omitted and (1.7) reduces to the Einstein relation between diffusion coefficients and mobilities [see I (5.18), I (6.14)]. The generalized time-dependent Einstein relation (1.7) is the basis of our *exact* calculations of $D_{\mu\nu}$ for harmonic oscillators in Sec. 3 and nonuniformly spaced multilevel systems in Sec. 4. The resulting exact "fluctuation-dissipation" relations between $D_{\mu\nu}$ and the reservoir contributions to A_μ are in precise agreement with those found by direct use of perturbation theory. The Einstein method, however, guarantees that products of operators propagate properly so that commutation rules are *necessarily preserved in time.*

Note that $\langle D_{\mu\nu}\rangle$ as calculated by (1.7) is not only a system operator,[3] it is, in general, time-dependent. Thus, the way in which the noise sources change during the turn-on of a laser is simply described within the present scheme.

With the noise sources included, our *Eqs. (1.2) are valid operator equations.* In particular, if variables $a_{s+1}, \cdots, a_n$ vary more rapidly than $a_1,a_2,\cdots,a_s$, we can solve for the fast variables in terms of the slow variables, and obtain a set of equations for the usually *much smaller set of slow variables.* The price paid for this is that the slow variable equations contain integrals over history and nonwhite noise sources. If, however, one examines the solutions only at frequencies small compared to the decay constants of the fast variables, then *it is adequate to treat the slow variables as Markoffian and the corresponding noise sources as white.* When this is done, an *enormous practical simplification* has been achieved in the solution of complicated problems such as laser noise.

Since our equations are operator equations, the equations for db/dt and $db^\dagger/dt$ (where b and $b^\dagger$ are destruction and creation operators of a photon field) determine the equation for $db^\dagger b/dt$, the rate of change of the number of photons. By applying the technique just discussed to eliminate all other variables but the upper state population N_2 in a maser and $b^\dagger b$, we show[6] that the *Markoffian* equations for $b^\dagger b$ and N_2 are the familiar rate equations. Moreover, these equations contain just the shot noise sources discussed by McCumber,[7] plus some thermal noise sources important in masers but not lasers. In summary, we have established the rate equations (and their noise sources) on *firm theoretical grounds*, as valid when $b^\dagger b$ and N_2 are the slowest changing variables in the complete set required to describe a maser or laser.[6] This adiabatic approximation can be avoided, however, permitting the extension of the noise calculation to higher frequencies, or to systems with other slowly varying populations or polarizations.[8]

[6] See paper QVII, Ref. 1 (to be published).
[7] D. E. McCumber, Phys. Rev. **141**, 306 (1966).
[8] The nonadiabatic treatment (to be published) represents work done jointly with D. E. McCumber.

Summary

Before discussing the details of our computations, it may be worthwhile to *summarize* our principal results:

If b and $b^\dagger$ are the destruction and creation operators associated with a harmonic oscillator, whose commutator is unity, then we find that the appropriate equations including dissipation and fluctuations are

$$db/dt=-(i\omega_0+\tfrac{1}{2}\gamma)b+f(t)\,;\quad \langle f(t)\rangle=0\,,\quad (1.8)$$

$$db^\dagger/dt=(i\omega_0-\tfrac{1}{2}\gamma)b^\dagger+f^\dagger(t)\,;\quad \langle f^\dagger(t)\rangle=0\,.\quad (1.9)$$

The parameter γ is the decay constant of this harmonic oscillator. The nonvanishing moments of the Langevin forces are provided by

$$\langle f^\dagger(t)f(u)\rangle=\gamma\bar{n}\delta(t-u)\,,$$
$$\langle f(u)f^\dagger(t)\rangle=\gamma(\bar{n}+1)\delta(t-u)\,.\quad (1.10)$$

The parameter $\bar{n}$ can be regarded as defined by the second-moment equation

$$d\langle b^\dagger b\rangle/dt=\gamma\bar{n}-\gamma\langle b^\dagger b\rangle\,;\quad \langle b^\dagger b\rangle\to\bar{n}\,,\quad (1.11)$$

i.e., the reservoir drives the system occupation number $b^\dagger b$ toward a mean value of $\bar{n}$. A harmonic reservoir at equilibrium would do this if its temperature T_R were given by

$$\bar{n}=[\exp(\hbar\omega_0/kT_R)-1]^{-1}\,,\quad (1.12)$$

as shown in Eq. (C11).

Our atomic equations in the absence of any regularities of energy spacing are given as

$$d(a_i^\dagger a_j)/dt=(i\omega_{ij}-\Gamma_{ij})a_i^\dagger a_j+\delta_{ij}\sum_k w_{ik}a_k^\dagger a_k+F_{ij}\,,\quad (1.13)$$
$$\Gamma_{ij}=\Gamma_{ji}\,;\quad \omega_{ij}=-\omega_{ji}\,;\quad F_{ji}=(F_{ij})^\dagger\,,$$

where the w_{ik} is a transition probability and the Γ_{ij} and ω_{ij} are defined in

$$\Gamma_{ij}=\tfrac{1}{2}(\Gamma_i+\Gamma_j)+\Gamma_{ij}^{\mathrm{ph}}\,;\quad \Gamma_{ii}^{\mathrm{ph}}=0\,;$$
$$\omega_{ij}=\omega_i-\omega_j+\Delta\omega_{ij}\,.\quad (1.14)$$

The superscript ph denotes a contribution to the results associated with phase fluctuations. We define

$$\Gamma_j=\sum_{k\neq j}w_{kj}\,;\quad w_{jj}\equiv0\,,\quad (1.15)$$

so that Γ_j represents the total transition rate out of state j. Our random forces F_{ij} have a vanishing first moment and second moments defined in

$$\langle F_{ij}\rangle=0\,;\quad \langle F_{ij}(t)F_{kl}(u)\rangle=2\langle D_{ijkl}\rangle\delta(t-u)\,,\quad (1.16)$$

$$D_{ijkl}=(D_{lkji})^*\,.\quad (1.17)$$

An explicit and nontrivial expression for the general diffusion constant $\langle D_{ijkl}\rangle$ (in the absence of regularities in energy spacing) is[3]

$$2\langle D_{ijkl}\rangle=\delta_{jk}[\Gamma_{ij}+\Gamma_{kl}-\Gamma_{il}]\langle a_i^\dagger a_l\rangle +\delta_{il}\delta_{jk}\sum w_{iq}\langle a_q^\dagger a_q\rangle -\delta_{ij}w_{ik}\langle a_k^\dagger a_l\rangle-\delta_{kl}w_{kj}\langle a_i^\dagger a_j\rangle -i\delta_{jk}(\omega_{ij}+\omega_{kl}-\omega_{il})\langle a_i^\dagger a_l\rangle. \quad (1.18)$$

The form of Eq. (1.13) was obtained by a perturbation treatment in Appendix A. However, once we grant this form, the diffusion constants of Eq. (1.18) follow without approximation. The parameters Γ_{ij}, w_{ij} can be regarded as experimentally determined. The last term in Eq. (1.18) vanishes in all cases in which ω_{ij} can be decomposed into two parts, one associated with ω_i and another associated with ω_j in the usual subtractive fashion. Whether or not ω_{ij} can be written in such a subtractive form is not a formal but rather a physical question. If the interaction between our system and our reservoir takes the form of

$$V=\hbar\sum a_i^\dagger a_j f_{ij}, \quad (1.19)$$

where f_{ij} represents a set of reservoir operators, then the transition probabilities above are given in

$$w_{mj}=\int_{-\infty}^{\infty} dt\,\exp(-i\omega_{mj}t)\langle f_{jm}(t)f_{mj}(0)\rangle. \quad (1.20)$$

The phase contribution to the damping constant is shown in Appendix A to be given in the form of

$$\Gamma_{ij}^{\text{ph}}=\frac{1}{2}\int_0^{\infty} dt\langle[f_{ii}(0)-f_{jj}(0), f_{ii}(t)-f_{jj}(t)]_+\rangle, \quad (1.21)$$

and the change in ω_{ij} is given in

$$\Delta\omega_{ij}=\Delta\omega_i-\Delta\omega_j+\Delta\omega_{ij}^{\text{ex}}, \quad (1.22)$$

$$\Delta\omega_j=\sum_{m\neq j}\text{Im}\int_0^{\infty} dt\,\exp(-i\omega_{mj}t)\langle f_{jm}(t)f_{mj}(0)\rangle +\tfrac{1}{2}i\int_0^{\infty} dt\langle[f_{jj}(0),f_{jj}(t)]\rangle, \quad (1.23)$$

$$\Delta\omega_{ij}^{\text{ex}}=\tfrac{1}{2}i\int_0^{\infty} dt\langle[f_{jj}(0),f_{ii}(t)]\rangle. \quad (1.24)$$

The "extra" term shown in Eq. (1.24) is not ordinarily decomposable in a subtractive way. We shall show in Appendix A that this anomalous frequency-shift term vanisbes in cases in which the reservoir obeys time reversal symmetry and the levels i and j are nondegenerate levels with respect to time reversal. Moreover, we show that this extra term in Eq. (1.24) vanishes whenever the reservoir forces can be decomposed into independent excitations such as phonons or spin waves, provided that these excitation operators belonging to different modes commute with one another at all times. Thus to see an anomalous frequency shift it is desirable to use a reservoir violating time reversal, for example, a ferromagnet or an antiferromagnet. In this case, the extra frequency shift will involve the interactions between the spin waves.

The general formula (1.18) for the diffusion constant D_{ijkl} is not particularly illuminating and it is worthwhile to display explicit results for a number of special cases. From now on if the subscripts of D are indicated by different letters then they are understood to be necessarily different unless otherwise indicated. Our first results are those appropriate to shot noise:

$$2\langle D_{iiii}\rangle=\sum_{q\neq i} w_{iq}\langle a_q^\dagger a_q\rangle+\Gamma_i\langle a_i^\dagger a_i\rangle =\text{atomic (rate in}+\text{rate out)}, \quad (1.25)$$

$$2\langle D_{iijj}\rangle=-w_{ji}\langle a_i^\dagger a_i\rangle-w_{ij}\langle a_j^\dagger a_j\rangle =-(\text{transfer rate}). \quad (1.26)$$

Equation (1.25) describes the typical rate-in plus rate-out contribution to the shot noise source associated with population of level i and Eq. (1.26) contains the sum of the transfer rates from levels i to j. These results have previously been obtained for classical systems in I, Sec. 12 and IV, Sec. 9. The diffusion constant most relevant for off-diagonal elements of the atomic density matrix is given in

$$2\langle D_{ijji}\rangle=(\Gamma_j+2\Gamma_{ij}^{\text{ph}})\langle a_i^\dagger a_i\rangle+\sum w_{iq}\langle a_q^\dagger a_q\rangle, \quad (1.27)$$

which has no simple classical analog. This moment D_{ijji} is valid even in the presence of coupling to a radiation field that induces transitions between levels i and j. In this case the transport equation for the population of level i is given by

$$d\langle a_i^\dagger a_i\rangle/dt=-\Gamma_i\langle a_i^\dagger a_i\rangle+\sum w_{iq}\langle a_q^\dagger a_q\rangle+\langle B_i\rangle, \quad (1.28)$$

$$B_i=\text{radiative rate into } i. \quad (1.29)$$

Assuming that we are in the steady state, in other words setting Eq. (1.28) equal to 0, we can simplify the right-hand side of Eq. (1.27) to obtain the steady-state second moment[3]

$$2\langle\langle D_{ijji}\rangle\rangle_{\text{steady state}}=2\Gamma_{ij}\langle\langle a_i^\dagger a_i\rangle\rangle_{\text{ss}}-\langle\langle B_i\rangle\rangle_{\text{ss}}. \quad (1.30)$$

The subscript ss is to remind us that the steady-state-system ensemble average is understood here. This result is stated without proof in QV (2.7) using $\bar{\sigma}_{ii}$ and $\bar{B}_i$ as abbreviated notations for the ss averages. A more complicated diffusion coefficient is given in

$$2\langle D_{ijjl}\rangle=(\Gamma_j+\Gamma_{ij}^{\text{ph}}+\Gamma_{jl}^{\text{ph}}-\Gamma_{il}^{\text{ph}})\langle a_i^\dagger a_l\rangle -i(\omega_{ij}+\omega_{jl}-\omega_{il})\langle a_i^\dagger a_l\rangle, \quad (1.31)$$

which appears to depend on the anomalous frequency shifts. Some diffusion coefficients descriptive of correlated population and phase fluctuation are given by

$$2\langle D_{ijkk}\rangle=-w_{kj}\langle a_i^\dagger a_j\rangle, \quad (k \text{ can equal } i) \quad (1.32)$$

$$2\langle D_{iikl}\rangle=-w_{ik}\langle a_k^\dagger a_l\rangle, \quad (l \text{ can equal } i) \quad (1.33)$$

$$2\langle D_{iiij}\rangle=\Gamma_i\langle a_i^\dagger a_j\rangle, \quad (1.34)$$

$$2\langle D_{ijjj}\rangle=\Gamma_j\langle a_i^\dagger a_j\rangle. \quad (1.35)$$

In Sec. 5, we establish that if two systems that interact with independent reservoirs are coupled together dynamically, no new noise sources are introduced, and no correlations occur between the noise sources associated with different reservoirs. The original noise sources are shown to be slightly modified by the dynamic interaction.

In Sec. 6, we use the results to construct the model of a maser used in QV. In Sec. 7, we obtain a "commutation rule Einstein relationship" and use it to show how the moments must be modified if the population difference in a maser is treated as a number rather than an operator.

Relation to Previous Work

There is, of course, an extensive literature on dissipation in quantum mechanics which we cannot hope to review properly here. This literature can be divided roughly into five categories:

(A) The consideration of a system in interaction with a reservoir, and the (approximate) elimination of the reservoir to obtain effective equations of motion for system operators, or the system density matrix.[9] The disadvantage of this procedure, including our own QIII, is that it provides information only about operators, or fluctuations at one time. To obtain two-time correlations one must use the equilibrium fluctuation-dissipation theorem, as in Sec. 7 of QIII, or one must use our generalization of this theorem in QII to nonequilibrium systems. Only in this way, can Scully, Lamb, and Stephen[9] argue that the decay constant they find is indeed the maser spectral linewidth.

(B) Green's function and moment methods[10] attack two-time correlation functions directly, but generally can be solved only by applying a truncation procedure or a linearization.

(C) A number of papers have been written introducing classical noise sources in a heuristic way.[11]

(D) Quantum noise sources have also been introduced into maser calculations in a heuristic way, or by methods similar to the perturbation techniques adopted here.[12]

(E) Detailed consideration has been given to the harmonic-oscillator[13] and two-level systems.[14] Louisell and Walker[13] have provided an exact solution for the system harmonic oscillator interacting linearly with a bath of harmonic oscillators, for "thermal" initial conditions.[15] Although the Louisell-Walker calculation is exact, it suffers from the usual objections to method (A) above. Schwinger and Senitzky have actually written down harmonic-oscillator equations with quantum noise sources. Senitzky,[13] in our notation,[16] uses the equations

$$\dot{Q}=P,\quad \dot{P}=-\gamma P-\omega_0^2Q+F(t),\tag{1.36}$$

$$\langle F(t)F(u)\rangle=2\gamma\hbar\omega_0\left[(\bar{n}+\tfrac{1}{2})\delta(t-u)-\frac{i}{2\pi}\mathcal{P}\frac{1}{t-u}\right],\tag{1.37}$$

where we have corrected a sign in the last term involving the principal-valued reciprocal. We show, in Appendix C, that Senitzky's commutation rule

$$\langle[F(t),F(u)]\rangle=-\frac{2i\gamma\hbar\omega_0}{\pi}\mathcal{P}\frac{1}{t-u}\tag{1.38}$$

leads to a commutator

$$\langle[Q,P]\rangle=i\hbar\left[1-\frac{1}{\pi}\left(\frac{\gamma}{\omega_0}\right)+O\left(\frac{\gamma}{\omega_0}\right)^2\right]\tag{1.39}$$

that is close to the desired value when $\gamma\ll\omega_0$, whereas the correct commutation rule [see Eq. (C33) when γ is not frequency-dependent] for the Langevin forces

$$\langle[F(t),F(u)]\rangle=2i\hbar\gamma\delta'(t-u)\tag{1.40}$$

leads to precisely the correct commutation rule for Q and P, as shown in (C37).

In any case, the commutator is odd in $t-u$. Hence its Fourier transform is odd in frequency ω and the spectrum of noise for a *quantum* oscillator can then not take rigorouly the white (flat) form needed to make the treatment Markoffian.

Of course, a noise spectrum need not be exactly white for the Markoffian approximation to be a good

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[15] An extension of Louisell and Walker's work to the initial condition of a definite excitation state has been made by H. Cheng and M. Lax, in *Quantum Theory of the Solid State*, edited by Per-Olav Löwdin (Academic Press Inc., New York, to be published). See also A. E. Glassgold and D. Holliday, Phys. Rev. **139**, A1717 (1965).

[16] Since Senitzky (Ref. 13) uses a harmonic oscillator that couples to a reservoir via its momentum (whereas we use the coordinate), comparison with our notation can be made by the transformations: $Q\to-P$, $P\to Q$, $\beta=\gamma$, $D(t)=F(t)$, and by setting his $4\pi c^2=\omega_0^2$.

one. It need only change little over the width of the resonance. Our method of calculation of the strength of the noise sources seems to (automatically) replace nonwhite sources by white sources whose strengths agree at the resonance frequency. For the harmonic oscillator, a special difficulty arises because there are *two* resonant frequencies ω_0 and $-\omega_0$, and the noise does not agree at these frequencies. However, these frequencies must be well separated $\omega_0 \gg \gamma$, in order that[4,5]

$$\omega_0^{-1} \ll \Delta t \ll \gamma^{-1}, \tag{1.41}$$

which is required for a Markoffian description to exist. If $\gamma \ll \omega_0$ the rotating-wave approximation[17] (RWA) is valid, and the relation $P \propto (b^\dagger + b)$ is essentially a split of P into its positive and negative frequency parts, and $F \propto f^\dagger + f$ is a similar split. Thus $f^\dagger$ should have a white spectrum that corresponds to the spectrum of F at ω_0, and f is similarly determined by the spectrum of F at $-\omega_0$. When we treat b and $b^\dagger$ as our fundamental variables, in the RWA in Sec. 3, these results appear automatically. If, however, we had insisted on using Q, P as variables, without the RWA, we could not have achieved a consistent Markoffian description that preserved the commutation rules.

The two-level analysis of Senitzky[14] does not seem to express the moments of the Langevin forces in terms of transition probabilities and populations so that it is difficult to make a comparison. The work of the Haken school[12] has so far not indicated any population dependence of their noise sources. Their quantum-noise-source treatment also seems to be special to a two-level system.

Our general formula (1.18) for the noise sources in a multilevel system seems therefore to be completely new. The phase or zero-phonon contributions to the decay and frequency shifts in (1.13) are most closely related to the work of McCumber.[18] The general expressions for the phase contributions to the damping and frequency shift (especially the anomalous shift) in a multilevel system seem to be new.

2. EINSTEIN RELATIONS, DIFFUSION COEFFICIENTS, AND EQUIVALENCE TO MOMENT METHODS

If we rewrite our fundamental Langevin equation in the form

$$da_\mu/dt = A_\mu(\mathbf{a},t) + F_\mu(\mathbf{a},t), \tag{2.1}$$

then the assumption that the system has a Markoffian description can be phrased in the form

$$F_\nu(s) \text{ is independent of } a_\mu(t) \quad \text{if} \quad s>t. \tag{2.2}$$

In particular, these operators must commute. If (2.2) were not obeyed, the statistics of the random forces F would depend on past history and the system would acquire memory effects. For present purposes we only need this independent assumption in the much weaker form

$$\langle F_\nu(s) C(\mathbf{a}(t))\rangle = \langle C(\mathbf{a}(t)) F_\nu(s)\rangle = 0 \quad \text{if} \quad s>t, \tag{2.3}$$

which merely describes a lack of correlation between the random force, at time s, and an arbitrary operator $C(\mathbf{a})$ at an earlier time t. We now write down the algebraic identity

$$a_\mu(t+\Delta t) a_\nu(t+\Delta t) - a_\mu(t) a_\nu(t) = \Delta a_\mu \Delta a_\nu + \Delta a_\mu a_\nu + a_\mu \Delta a_\nu, \tag{2.4}$$

where

$$\Delta a_\mu = a_\mu(t+\Delta t) - a_\mu(t), \tag{2.5}$$

$$\Delta a_\mu \approx A_\mu \Delta t + \int_t^{t+\Delta t} F_\mu(s) ds. \tag{2.6}$$

From Eq. (2.4), we can now write down the equation for the mean motion of the product of two operators:

$$d\langle a_\mu(t) a_\nu(t)\rangle/dt = 2\langle D_{\mu\nu}\rangle + \langle A_\mu a_\nu\rangle + \langle a_\mu A_\nu\rangle. \tag{2.7}$$

In the steady state when the second-order operator does not change with time we obtain the standard Einstein equation

$$2\langle D_{\mu\nu}\rangle_{\text{steady state}} = -\langle A_\mu a_\nu\rangle_{\text{ss}} - \langle a_\mu A_\nu\rangle_{\text{ss}}. \tag{2.8}$$

Aside from the preservation of the order of the operators, Eqs. (2.7) and (2.8) are identical to corresponding classical equations. See, for example, I (14.27) or III (5.10). It is convenient to rewrite Eq. (2.7) in the mnemonic form

$$2\langle D_{\mu\nu}\rangle = -\langle \{da_\mu/dt\} a_\nu\rangle - \langle a_\mu \{da_\nu/dt\}\rangle + \langle d[a_\mu(t) a_\nu(t)]/dt\rangle, \tag{2.9}$$

$$\{da_\mu/dt\} \equiv da_\mu/dt - F_\mu \equiv A_\mu. \tag{2.10}$$

We see, therefore, that $D_{\mu\nu}$ is a measure of the extent to which the usual rules for differentiating a product is violated in a Markoffian system. Equation (2.9) is indeed a useful computational formula because it permits the diffusion coefficients to be calculated in terms of the mean motion of certain system operators. This is in contrast to Eq. (1.6) which requires the direct calculation of fluctuations. Indeed, our presentation in Secs. 3 and 4 will be based on Eq. (2.9). The direct determination of the diffusion constants from the fluctuations moments using Eq. (1.6) is done in Appendix B.

Higher order diffusion coefficients may be determined by a simple generalization of (2.9). But higher moments

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[17] For a harmonic oscillator, the RWA consists in neglecting the counter-resonant term $\frac{1}{2}\gamma b^\dagger$ in $db/dt = -i\omega_0 b - \frac{1}{2}\gamma(b+b^\dagger) + f$, and a corresponding term $\frac{1}{2}\gamma b$ in $db^\dagger/dt = i\omega_0 b^\dagger - \frac{1}{2}\gamma(b+b^\dagger) + f^\dagger$. The distinction between the free decay frequency $[\omega_0^2 - \frac{1}{4}\gamma^2]^{1/2}$ and ω_0 is thus neglected. The RWA has been extensively exploited in magnetic resonance problems: I. I. Rabi, N. F. Ramsey, and J. Schwinger, Rev. Mod. Phys. **26**, 107 (1954); A. Abragam, *Principles of Nuclear Magnetism* (Oxford University Press, Oxford, England, 1961), Chap. II; F. Bloch and A. J. Siegert, Phys. Rev. **57**, 522 (1940).

[18] D. E. McCumber, Phys. Rev. **135**, A1676 (1964); J. Math. Phys. **5**, 221, 508 (1964).

of the Langevin forces are related in a more complicated way to the diffusion constants as shown in IV.

Equivalence of Langevin and Moment Procedures

We would now like to establish the equivalence of the Langevin method of this paper with the moment procedures adopted in QII. The principal result of paper QII is summarized in the statement that the solution for the mean motion at one time,

$$\langle a_i^\dagger(t)a_j(t)\rangle=\sum O_{qp}{}^{ji}(t,t')\langle a_p^\dagger(t')a_q(t')\rangle, \quad (2.11)$$

can be used to obtain the moments containing operators at two times

$$\langle a_i^\dagger(t)a_j(t)a_k^\dagger(t')a_l(t')\rangle =\sum O_{qp}{}^{ji}(t,t')\langle a_p^\dagger(t')a_q(t')a_k^\dagger(t')a_l(t')\rangle. \quad (2.12)$$

This two-time result (2.12) possesses the same t dependence as the transient solution (2.11) of the mean motion equations. This is an expression of the Onsager hypothesis concerning the regression of fluctuations, established for a quantum-mechanical system not in equilibrium nor even necessarily in a steady state.

For comparison with the results in this paper, it is necessary to avoid direct use of the Green's function $O_{qp}{}^{ji}$ and replace it by a differential relationship. We note, however, as remarked at the end of paper QII that this principal result (2.12) did not depend on the system being Markoffian. If we assume that the system is Markoffian its future must be predictable from the present, in other words, its time derivative at time t must be expressible in terms of other quantities expressed at time t. In terms of the Green's function the Markoffian requirement takes the form

$$dO_{qp}{}^{ji}(t,t')/dt=\sum B_{ijmn}O_{qp}{}^{nm}(t,t'). \quad (2.13)$$

If we now differentiate Eq. (2.11) and make use of (2.13), we obtain

$$\langle da_i^\dagger(t)a_j(t)/dt-\sum B_{ijmn}a_m^\dagger(t)a_n(t)\rangle=0, \quad (2.14)$$

which expresses the time derivative of $a_i^\dagger a_j$ in terms of similar operators taken at the same time. Applying the same procedure to Eq. (2.12) leads to

$$\langle[da_i^\dagger(t)a_j(t)/dt-\sum B_{ijmn}a_m^\dagger(t)a_n(t)] \times a_k^\dagger(t')a_l(t')\rangle=0. \quad (2.15)$$

Since the quantity in brackets in Eq. (2.15) is the difference between the time derivative of the operator $a_i^\dagger a_j$ and the corresponding mean motion, it represents the fluctuating random force F_{ij}. Thus, we obtain

$$\langle F_{ij}(t)a_k^\dagger(t')a_l(t')\rangle=0, \quad t>t'. \quad (2.16)$$

But Eq. (2.16) is precisely the same as Eq. (2.3), which we took as our expression in the Langevin point of view of the Markoffian nature of our problem. Thus we have established the equivalence of the two procedures. When both are used without further approximation they should yield equivalent results for the mean motion and the noise in a quantum-mechanical system.

Taking the derivative of (2.16) with respect to t', we obtain

$$\langle F_{ij}(t)da_k^\dagger(t')a_l(t')/dt'\rangle=0, \quad t>t'. \quad (2.17)$$

Taking the appropriate linear combinations of (2.16) and (2.17), we establish that

$$\langle F_{ij}(t)F_{pq}(t')\rangle=0, \quad t>t'. \quad (2.18)$$

A similar argument proves the same result for $t<t'$, so that we establish

$$\langle F_{ij}(t)F_{pq}(t')\rangle=0, \quad t\neq t'. \quad (2.19)$$

3. THE DAMPED HARMONIC OSCILLATOR

To obtain the diffusion constants using (2.9), it is necessary to obtain the mean equations of motion for operators linear and operators quadratic in the oscillator displacements. In Appendix A, we modify slightly some results of QIII which analyzes a system in interaction with a reservoir and then determines the effective equations of motion for the system after averaging over the reservoir. These results of QIII are sufficiently general to include memory effects. After eliminating these memory effects when the correlation times are short, we arrive at Eq. (A13) for an arbitrary system operator M. Equation (A13) is valid even if only reservoir averages[3] $\langle\langle\ \rangle$ instead of $\langle\langle\ \rangle\rangle$) are taken—see (B16). Thus, we can write

$$\frac{\partial\langle M\rangle}{\partial t}=\langle(M,H)\rangle -\frac{1}{\hbar^2}\sum_{ij}\int_0^\infty du\{\langle F_i(u)F_j\rangle\langle[M,Q_i]Q_j(-u)\rangle -\langle F_jF_i(u)\rangle\langle Q_j(-u)[M,Q_i]\rangle\}. \quad (3.1)$$

In (3.1), the Q's are system operators and the F's are corresponding reservoir operators that when multiplied together provide the coupling V between the system and reservoir:

$$V=-\textstyle\sum_i Q_iF_i.$$

The operators $F_i(u)$ and $Q_j(-u)$ are in the interaction representation

$$F_i(u)=\exp(iRu/\hbar)F_i\exp(-iRu/\hbar),$$
$$Q_j(-u)=\exp(-iHu/\hbar)Q_j\exp(iHu/\hbar),$$

where H and R are the system and reservoir Hamiltonians, respectively. [The slightly more general equation (A13) must be used if H depends explicitly on the time.]

For the harmonic-oscillator system, we take

$$H=\hbar\omega_c b^\dagger b; \quad V=i\hbar(b^\dagger g-bg^\dagger). \quad (3.2)$$

With this choice an arbitrary system operator M obeys the equation of motion

$$\partial\langle M\rangle/\partial t=\langle(M,H)\rangle+\alpha\langle b^\dagger[M,b]\rangle-\beta\langle[M,b]b^\dagger\rangle -\alpha^*\langle[M,b^\dagger]b\rangle+\beta^*\langle b[M,b^\dagger]\rangle, \quad (3.3)$$

where the coefficients α and β are integrals over averages of reservoir operators:

$$\alpha=\int_0^\infty du\, e^{-i\omega_c u}\langle gg^\dagger(u)\rangle, \qquad \beta=\int_0^\infty du\, e^{-i\omega_c u}\langle g^\dagger(u)g\rangle. \quad (3.4)$$

Introducing the interpretations

$$\tfrac{1}{2}\gamma-i\Delta\omega=\alpha-\beta, \quad \omega_0=\omega_c+\Delta\omega, \quad \gamma\bar{n}=2\,\mathrm{Re}\beta \quad (3.5)$$

of the parameters α and β in terms of new real parameters γ, $\bar{n}$, and $\Delta\omega$ which have more direct physical meaning, we obtain the equation of motion for the fairly general operator $(b^\dagger)^r b^s$:

$$\partial\langle(b^\dagger)^r b^s\rangle/\partial t=[i\omega_0(r-s)-\tfrac{1}{2}\gamma(r+s)]\langle(b^\dagger)^r b^s\rangle +rs\gamma\bar{n}\langle(b^\dagger)^{r-1}b^{s-1}\rangle. \quad (3.6)$$

These equations are exact in this Markoffian limit when the reservoir consists of a set of harmonic oscillators. (See Appendix D.) In (3.6) ω_0 is the renormalized frequency of the oscillator after the frequency shift $\Delta\omega$ produced by the reservoir has been absorbed. The most important special cases of Eq. (3.6) are given by

$$\partial\langle b\rangle/\partial t=-(i\omega_0+\tfrac{1}{2}\gamma)\langle b\rangle, \qquad \partial\langle b^\dagger\rangle/\partial t=(i\omega_0-\tfrac{1}{2}\gamma)\langle b^\dagger\rangle, \quad (3.7)$$

$$\partial\langle b^\dagger b\rangle/\partial t=\gamma\bar{n}-\gamma\langle b^\dagger b\rangle. \quad (3.8)$$

We now write out our complete Langevin equations including the noise sources as

$$db/dt=-(i\omega_0+\tfrac{1}{2}\gamma)b+f(t), \qquad db^\dagger/dt=(i\omega_0-\tfrac{1}{2}\gamma)b^\dagger+f^\dagger(t). \quad (3.9)$$

Our second moment has the typical δ-function form

$$\langle f^\dagger(t)f(u)\rangle=2D_{b^\dagger b}\delta(t-u), \quad (3.10)$$

where the diffusion constant is computed by means of Eq. (2.9) in the form

$$2D_{b^\dagger b}=d\langle b^\dagger b\rangle/dt-\langle\{db^\dagger/dt\}b\rangle-\langle b^\dagger\{db/dt\}\rangle=\gamma\bar{n}. \quad (3.11)$$

Repeating this calculation with the opposite order of the factors yields

$$\langle f(u)f^\dagger(t)\rangle=2D_{bb^\dagger}\delta(t-u)=\gamma(\bar{n}+1)\delta(t-u), \quad (3.12)$$

a second moment clearly distinct from that in (3.11). Indeed, this lack of commutation of the reservoir forces at f and $f^\dagger$ is shown in Appendix C to preserve the commutation rules. We also compute the additional second-moment equations

$$d\langle b^2\rangle/dt=-2i\omega_0\langle b^2\rangle-\gamma\langle b^2\rangle, \qquad d\langle(b^\dagger)^2\rangle/dt=+2i\omega_0\langle(b^\dagger)^2\rangle-\gamma\langle(b^\dagger)^2\rangle, \quad (3.13)$$

and learn from them that the remaining second moments

$$\langle f(t)f(u)\rangle=\langle f^\dagger(t)f^\dagger(u)\rangle=0 \quad (3.14)$$

are zero.

4. ATOMIC DIFFUSION CONSTANTS

For an arbitrary quantum-mechanical system (which we shall visualize as an atom) whose frequency differences ω_{ij} possess no degeneracies, we shall adopt as our typical equation of motion (A23):

$$d(a_i^\dagger a_j)/dt=(i\omega_{ij}-\Gamma_{ij})a_i^\dagger a_j+\delta_{ij}\sum_{p\neq i} w_{ip}a_p^\dagger a_p+F_{ij} \quad (4.1)$$

obtained in Appendix A by the use of Eq. (3.1). Second moments are defined by[3]

$$\langle F_{ij}(t)F_{kl}(u)\rangle=2\langle D_{ijkl}\rangle\delta(t-u). \quad (4.2)$$

The second moments as usual are calculated by Eq. (2.9) which represents a nonstationary Einstein relationship. For the present case, Eq. (2.9) takes the form

$$2\langle D_{ijkl}\rangle=\frac{d}{dt}\langle a_i^\dagger a_j a_k^\dagger a_l\rangle-\left\langle\left\{\frac{d}{dt}a_i^\dagger a_j\right\}a_k^\dagger a_l\right\rangle -\left\langle a_i^\dagger a_j\left\{\frac{d}{dt}a_k^\dagger a_l\right\}\right\rangle, \quad (4.3)$$

where the bracketed symbols defined by Eq. (4.4)

$$\left\{\frac{d}{dt}a_i^\dagger a_j\right\}\equiv\frac{d}{dt}a_i^\dagger a_j-F_{ij} \quad (4.4)$$

simply describe the mean motion of the operators $a_i^\dagger a_j$. In the space of one atom, $\sum a_j^\dagger a_j=1$, we have the identity

$$a_i^\dagger a_j a_k^\dagger a_l=\delta_{jk}a_i^\dagger a_l \quad (4.5)$$

derived in QII. Inserting the bracketed quantities (4.4) into Eq. (4.3), our results immediately simplify into a bilinear expression of the form

$$2\langle D_{ijkl}\rangle=\delta_{jk}\delta_{il}\sum_{p\neq i} w_{ip}\langle a_p^\dagger a_p\rangle -\delta_{ij}w_{ik}\langle a_k^\dagger a_l\rangle-\delta_{kl}w_{kj}\langle a_i^\dagger a_j\rangle +\langle a_i^\dagger a_l\rangle\delta_{jk}[(\Gamma_{ij}+\Gamma_{kl}-\Gamma_{il})-i(\omega_{ij}+\omega_{kl}-\omega_{il})], \quad (4.6)$$

where the quantities Γ_{ij} and ω_{ij} have the symmetry and antisymmetry properties

$$\Gamma_{ij}=\Gamma_{ji}, \quad \omega_{ij}=-\omega_{ji}. \quad (4.7)$$

We see that our results for the second moments depend on the form of Eq. (4.1) and not how the parameters were obtained. Equations (1.19)–(1.24) display, however, the values of these parameters obtained from the explicit reservoir calculations shown in Appendix A. Equation (4.6) is the general result quoted in Eq. (1.18) and important special cases of the diffusion constants were already presented in Eqs. (1.25)–(1.35).

5. COUPLED SYSTEMS: INDEPENDENT RESERVOIRS

Our systems 1 and 2 are coupled together dynamically via the Hamiltonian H_{12}:

$$H=H_1+H_2+H_{12},\quad V_1=-\sum Q_jF_j,\quad V_2=-\sum q_jf_j. \tag{5.1}$$

Here V_1 and V_2 are the couplings of systems 1 and 2, respectively, to their corresponding independent reservoirs. The reservoir forces F_j and f_j are definitely uncorrelated since they come from quite independent reservoirs. If M and m are arbitrary operators belonging to the first and second systems, respectively, they obey the Heisenberg equations

$$dM/dt=(M,H_1+H_{12})-\sum (M,Q_j)F_j, \tag{5.2}$$

$$dm/dt=(m,H_2+H_{12})-\sum (m,q_i)f_i. \tag{5.3}$$

The direct use of Eq. (1.6) then yields

$$2\langle D_{mM}\rangle=\langle[m(t+\Delta t)-m(t)]\times[M(t+\Delta t)-M(t)]\rangle/\Delta t \tag{5.4}$$

$$\approx\sum(m,q_i)(M,Q_j)\int_t^{t+\Delta t}ds\int_t^{t+\Delta t}ds'\times\langle f_i(s)F_j(s')\rangle/\Delta t, \tag{5.5}$$

where the first terms in (5.2) and (5.3) have been dropped since they lead to terms of higher order in Δt. The assumed δ-function character of the correlations of the random forces permits the system operators to be evaluated at the initial time t and removed from the integration in Eq. (5.5). The result involves the reservoir forces F and f directly and these are by hypothesis uncorrelated. The vanishing of D_{mM} implies that the effective Langevin forces that enter the equations for dM/dt and dm/dt are necessarily uncorrelated.

The proof we have given, however, is unnecessarily restrictive. The reservoir forces are not in general δ correlated.[5] They merely possess a correlation time that is short compared to all of the typical system relaxation times. In general, however, their correlation times are long compared to the reciprocals of the various oscillation frequencies of the system. Under these circumstances it is not permissible to remove the system operators from underneath the integral sign in Eq. (5.5), and a new proof is needed. The methods of QIII were, in fact, designed to deal with such situations in which the correlation time is short but not zero. The net result of that paper reduced to the Markoffian limit is Eq. (3.1). If we apply (3.1) to calculate the equation of motion of the operator mM, we obtain

$$\begin{aligned}\frac{d\langle mM\rangle}{dt}&=\langle(mM,H)\rangle\\&-\frac{1}{\hbar^2}\sum\int_0^\infty du\{\langle F_i(u)F_j\rangle\langle[mM,Q_i]Q_j(-u)\rangle\\&-\langle F_jF_i(u)\rangle\langle Q_j(-u)[mM,Q_i]\rangle\}\\&-\frac{1}{\hbar^2}\sum\int_0^\infty du\{\langle f_i(u)f_j\rangle\langle[mM,q_i]q_j(-u)\rangle\\&-\langle f_jf_i(u)\rangle\langle q_j(-u)[mM,q_i]\rangle\}.\end{aligned} \tag{5.6}$$

In Eq. (5.6), correlations between F and f forces have been omitted. However, the time dependence of the system operators is retained underneath the integral sign. If, however, we expand the commutators in Eq. (5.6) and compare them with the corresponding separate equations for the operators M and m, we find that our result has the structure

$$d\langle mM\rangle/dt=\langle m\{dM/dt\}\rangle+\langle\{dm/dt\}M\rangle+0 \tag{5.7}$$

from which we can deduce that the diffusion constant

$$2D_{mM}=0 \tag{5.8}$$

vanishes. The result (5.8) establishes that the Langevin noise sources that enter the equations of two systems that are coupled to independent reservoirs are uncorrelated. While this result is intuitively very reasonable, it is not, in fact, obvious. The Langevin forces are not identical to the reservoir forces which are automatically uncorrelated. Thus, for example, the Langevin force F_{ij} of Eq. (1.13) is not identical to the reservoir operator f_{ij} of Eq. (1.19). The Langevin forces, in effect, involve products of reservoir forces with system operators. Since the time dependence of these system operators must be taken into account, the lack of correlation of the Langevin forces requires the proof just given.

A similar procedure with m and M both taken from the first system yields the same *formal* expression for D_{mM} as if the second system were not present. The interaction operators, however, now include the effect of the dynamic interaction. Unless this interaction is extremely strong, however, its influence during the short correlation time of the reservoirs will be unimportant. This is equivalent, for example, to neglecting the change of atomic state of an atom due to a laser field during the course of its collision with a second atom. Our procedure permits us to include such effects, but we shall omit them in the maser model of the next section.

6. STOCHASTIC MODEL OF A MASER

Our model of a maser is schematically described in Fig. 1. The electromagnetic field is described in terms of a single-cavity mode, although it is easy enough to generalize to the presence of several modes. There are a set of N atoms labeled by the index M. These two systems are coupled by the radiation coupling

$$H_{\text{RAD-ATOM}}=i\hbar\mu\sum_{M=1}^{N}[b^{\dagger}(a_1^{\dagger}a_2)^M-b(a_2^{\dagger}a_1)^M]. \quad (6.1)$$

As shown in Fig. 1, the set of atoms and the radiation field are each coupled to its own reservoir. Indeed, for practical purposes, we can assume that each atom is coupled to its own private reservoir. This is why we have indicated the atomic Langevin forces by a superscript M. Our Langevin equations of motion now take the form

$$db/dt=-(i\omega_c+\tfrac{1}{2}\gamma)b+\mu\textstyle\sum_M(a_1^{\dagger}a_2)^M+f, \quad (6.2)$$

$$d(a_1^{\dagger}a_2)^M/dt=-(\Gamma_{12}{}^M+i\omega^M)(a_1^{\dagger}a_2)^M+\mu b(a_2^{\dagger}a_2-a_1^{\dagger}a_1)^M+F_{12}{}^M, \quad (6.3)$$

$$d(a_2^{\dagger}a_2)^M/dt=w_{20}(a_0^{\dagger}a_0)^M+w_{21}(a_1^{\dagger}a_1)^M-\Gamma_2(a_2^{\dagger}a_2)^M-B^M+F_{22}{}^M, \quad (6.4)$$

$$d(a_1^{\dagger}a_1)^M/dt=w_{10}(a_0^{\dagger}a_0)^M+w_{12}(a_2^{\dagger}a_2)^M-\Gamma_1(a_1^{\dagger}a_1)^M+B^M+F_{11}{}^M, \quad (6.5)$$

$$B^M=\mu[b^{\dagger}(a_1^{\dagger}a_2)^M+(a_2^{\dagger}a_1)^M b], \quad (6.6)$$

$$(a_0^{\dagger}a_0+a_1^{\dagger}a_1+a_2^{\dagger}a_2)^M=1, \quad (6.7)$$

$$\omega^M=\omega_2{}^M-\omega_1{}^M, \quad (6.8)$$

where w_{20} and w_{10} are pump terms. We have assumed, for simplicity only, that the interaction between the atoms and the field is equally strong for all atoms. In such a case it is appropriate to introduce averages over the various atom operators:

$$\sigma_{jj}\equiv(1/N)\sum_{M=1}^{N}(a_j^{\dagger}a_j)^M, \quad (6.9)$$

$$D\equiv\sigma_{22}-\sigma_{11}, \quad (6.10)$$

$$\sigma\equiv(1/N)\sum_{M=1}^{N}(a_1^{\dagger}a_2)^M\exp(i\omega_0 t). \quad (6.11)$$

Moreover, we shall introduce a new field operator

$$b'=b\exp(i\omega_0 t) \quad (6.12)$$

that has absorbed most of the steady motion in the maser so that b' changes only quite slowly with the time primarily because of the Langevin forces. In the following equations we shall for simplicity drop the prime on b. The average rate of radiation per atom is then given by

$$B=(1/N)\textstyle\sum B^M=\mu(b^{\dagger}\sigma+\sigma^{\dagger}b) \quad (6.13)$$

and the appropriately averaged atomic random force is given by

$$F_{ij}=(1/N)\textstyle\sum F_{ij}{}^M\exp(i\omega_{ji}t). \quad (6.14)$$

Strictly speaking F_{12} contains a factor $\exp(i\omega_0 t)$ rather than $\exp(i\omega_{21}t)$,but the spectrum of F_{12} can be assumed not to change much over the small difference between

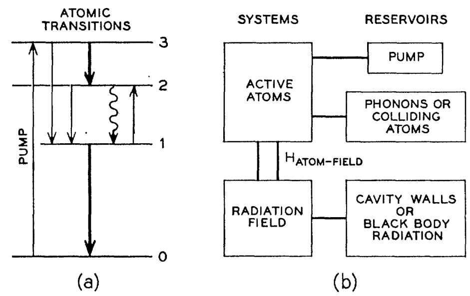

FIG. 1. Model for maser or laser: Radiative transitions (wavy arrow) are induced by the dynamic atom-field coupling. Nonradiative transitions (straight arrows) and quantum noise sources are derivable consequences of the coupling to the reservoirs. We assume in this paper that the transition rate from 3 to 2 is so fast that we are effectively pumping directly into state 2.

these two frequencies. Our coupled atomic and field equations now take the form

$$d\sigma_{22}/dt=w_{20}\sigma_{00}+w_{21}\sigma_{11}-\Gamma_2\sigma_{22}-B+F_{22}, \quad (6.15)$$

$$d\sigma_{11}/dt=w_{10}\sigma_{00}+w_{12}\sigma_{22}-\Gamma_1\sigma_{11}+B+F_{11}, \quad (6.16)$$

$$db/dt=-[\tfrac{1}{2}\gamma+i(\omega_c-\omega_0)]b+N\mu\sigma+F, \quad (6.17)$$

where $F=f\exp(i\omega_0 t)$, as in (C19), and

$$\sigma_{00}=1-\sigma_{11}-\sigma 22.$$

The case of homogeneous broadening can be obtained by making the specialization

$$\omega^M=\omega_a, \quad \Gamma_{12}{}^M=\Gamma. \quad (6.18)$$

In this case Eq. (6.3) reduces to the form

$$d\sigma/dt=-[\Gamma+i(\omega_a-\omega_0)]\sigma+\mu bD+F_{12}. \quad (6.19)$$

The moments of our Langevin forces are now given by

$$\langle F(t)F_{ij}{}^M(u)\rangle=\langle F_{ij}{}^M(u)F(t)\rangle=0, \quad (6.20)$$

$$\langle F_{ij}{}^{M'}(t)F_{kl}{}^M(u)\rangle=0 \quad \text{for} \quad M'\neq M, \quad (6.21)$$

$$\langle F^{\dagger}(t)F(u)\rangle=\gamma\bar{n}\delta(t-u), \quad (6.22)$$

$$\langle F(u)F^{\dagger}(t)\rangle=\gamma(\bar{n}+1)\delta(t-u), \quad (6.23)$$

$$\langle F_{ij}{}^M(t)F_{kl}{}^M(u)\rangle=2\langle D_{ijkl}{}^M\rangle\delta(t-u), \quad (6.24)$$

$$\langle F_{ij}(t)F_{kl}(u)\rangle=(1/N)2\langle D_{ijkl}\rangle\delta(t-u), \quad (6.25)$$

$$\langle D_{ijkl}\rangle=(1/N)\textstyle\sum_M\langle D_{ijkl}{}^M\rangle. \quad (6.26)$$

Here $\bar{n}=\bar{n}(\omega_0)$ as in (C40).

The atomic and field forces are uncoupled as they should be. The factor $1/N$ that appears in Eq. (6.25) is essentially a consequence of the fact that the individual F^M are uncorrelated. The most important diffusion constants for the discussion of phase noise in a maser above threshold are given in

$$2\langle D_{1221}{}^M\rangle=\langle(a_1^{\dagger}a_1)^M\rangle[\Gamma_2+2\Gamma_{12}{}^{\text{ph}}]+\sum_{p\neq 1}w_{1p}\langle(a_p^{\dagger}a_p)^M\rangle, \quad (6.27)$$

$$2\langle D_{1221}\rangle=\{\langle\sigma_{11}\rangle[\Gamma_2+2\Gamma_{12}{}^{\text{ph}}]+\sum_{p\neq 1}w_{1p}\langle\sigma_{pp}\rangle\}. \quad (6.28)$$

In the steady state these results can be simplified as shown in Eq. (1.30) to the form[3]

$$2\langle\langle D_{1221}\rangle\rangle_{ss}=\{\bar{\sigma}_{11}[\Gamma_2+2\Gamma_{12}{}^{ph}]+\Gamma_1\bar{\sigma}_{11}-\bar{B}\} =[2\Gamma\bar{\sigma}_{11}-\bar{B}]. \qquad (6.29)$$

The corresponding diffusion constant for the operators taken in reverse order takes the corresponding form

$$2\langle\langle D_{2112}\rangle\rangle_{ss}=[2\Gamma\bar{\sigma}_{22}+\bar{B}]. \qquad (6.30)$$

The additional diffusion constants

$$2\langle D_{1111}\rangle=\sum_{p\neq1} w_{1p}\langle\sigma_{pp}\rangle+\Gamma_1\langle\sigma_{11}\rangle, \qquad (6.31)$$

$$2\langle D_{2222}\rangle=\sum_{p\neq2} w_{2p}\langle\sigma_{pp}\rangle+\Gamma_2\langle\sigma_{22}\rangle, \qquad (6.32)$$

$$2\langle D_{1122}\rangle=2\langle D_{2211}\rangle=-[w_{12}\langle\sigma_{22}\rangle+w_{21}\langle\sigma_{11}\rangle], \qquad (6.33)$$

$$2\langle D_{1222}\rangle=\Gamma_2\langle\sigma\rangle;\quad 2\langle D_{2212}\rangle=-w_{21}\langle\sigma\rangle, \qquad (6.34)$$

$$2\langle D_{2122}\rangle=-w_{21}\langle\sigma^\dagger\rangle;\quad 2\langle D_{2221}\rangle=\Gamma_2\langle\sigma^\dagger\rangle, \qquad (6.35)$$

$$D_{ijkl}=(D_{lkji})^* \qquad (6.36)$$

can be obtained directly from Eqs. (1.25)–(1.35).

The random forces F_{ij} can by the law of large numbers be taken as Gaussian random variables since they are averages over a large set $F_{ij}{}^M$ of identically distributed variables. In the optical region the $F(t)$ are produced by the vacuum fluctuations of the electromagnetic field and are clearly Gaussian. In the microwave region, the source of electromagnetic noise will be the cavity walls. Since many atoms contribute to this "black body" radiation, we can again take F as Gaussian.

7. PRESERVATION OF COMMUTATION RULES (AND SECOND MOMENTS)

Since we have computed our diffusion constants by comparing the mean equations of linear operators with the mean equations of motion of quadratic operators, we have necessarily guaranteed that the correct equation of motion is obtained for the product of any two operators. Since this is true for either order in which the product is taken, commutators obey the correct equations of motion. Thus, if the commutation rules are obeyed at an initial instant of time they will necessarily be preserved in time. In spite of this, it is of some interest to display directly what the commutator of the Langevin forces

$$\langle[F_\mu(t),F_\nu(u)]\rangle=2\langle D_{\mu\nu}-D_{\nu\mu}\rangle\delta(t-u) \qquad (7.1)$$

depends on. Let us rewrite Eq. (2.7) as

$$d\langle a_\mu a_\nu\rangle/dt=2\langle D_{\mu\nu}\rangle+\langle A_\mu a_\nu\rangle+\langle a_\mu A_\nu\rangle. \qquad (7.2)$$

We can next interchange the indices μ and ν and subtract to obtain

$$d\langle[a_\mu,a_\nu]\rangle/dt=2\langle(D_{\mu\nu}-D_{\nu\mu})\rangle+\langle[A_\mu,a_\nu]\rangle +\langle[a_\mu,A_\nu]\rangle. \qquad (7.3)$$

Thus, the commutators of our random forces are expressed by

$$2\langle(D_{\mu\nu}-D_{\nu\mu})\rangle=\langle[a_\nu,A_\mu]+[A_\nu,a_\mu]\rangle +d\langle[a_\mu,a_\nu]\rangle/dt \qquad (7.4)$$

$$=\langle[a_\nu,\{da_\mu/dt\}]+[\{da_\nu/dt\},a_\mu]\rangle +d\langle[a_\mu,a_\nu]\rangle/dt \qquad (7.5)$$

in terms of certain commutators and their time derivatives.

Let us work out one important case for the maser problem as an example. The forces in the population equations commute and therefore we shall not consider them. Instead, let us consider the forces that enter the off-diagonal equations. The appropriate commutator taking account of the factor $1/N$ in (6.25) is

$$2\langle D_{1221}-D_{2112}\rangle/N=\langle[\sigma^\dagger,\{d\sigma/dt\}]+[\{d\sigma^\dagger/dt\},\sigma]\rangle +d\langle[\sigma,\sigma^\dagger]\rangle/dt, \qquad (7.6)$$

where the quantity in brackets is given by

$$\{d\sigma/dt\}=[\Gamma+i(\omega_a-\omega_0)]\sigma+\mu bD. \qquad (7.7)$$

Thus, we obtain

$$2\langle D_{1221}-D_{2112}\rangle/N=-2\Gamma\langle[\sigma^\dagger,\sigma]\rangle-d\langle[\sigma^\dagger,\sigma]\rangle/dt +\mu\langle b[\sigma^\dagger,D]+[D,\sigma]b^\dagger\rangle. \qquad (7.8)$$

The necessary commutators are

$$[\sigma^\dagger,\sigma]=D/N, \qquad (7.9)$$

$$[\sigma^\dagger,D]=-2\sigma^\dagger/N;\quad [D,\sigma]=-2\sigma/N. \qquad (7.10)$$

Thus, our commutators in the steady state take the simple form of

$$2\langle\langle D_{1221}-D_{2112}\rangle\rangle_{ss}/N=-2(\Gamma/N)\bar{D}-2\bar{B}/N =-(2/N)[\Gamma\bar{D}+\bar{B}]. \qquad (7.11)$$

This commutator is precisely what one obtains if one subtracts Eq. (6.30) from Eq. (6.29). Below threshold in paper QV, however, we treat D as a c number. This means that the commutators involving D vanish. Thus if the "dielectric" approximation is made, the last term in Eq. (7.8) must be omitted and the commutator reduces to

$$2\langle\langle D_{1221}-D_{2112}\rangle\rangle_{ss}/N=-2(\Gamma/N)\bar{D} \qquad (7.12)$$

as quoted in QV.

Note added in proof. After the completion of this manuscript (and after the results summarized in Secs. 1 and 6 were presented at the 1965 Puerto Rico conference) we have learned that several members of the Haken school have adopted a Markoffian approach closely related to our own. See H. Haken and W. Weidlich [Z. Physik **189**, 1 (1966)]; C. Schmid and H. Risken [*ibid.* **189**, 365 (1966)]. These papers treat the atomic fluctuations and lead to moments in agreement with ours. For the electromagnetic field, the noise sources are not derived by them but are taken from

Senitzky—see, e.g., H. Sauerman, Z. Physik **189**, 312 (1966). Our procedure obtains the field noise sources by the same method as that used for the atomic noise sources, and moreover derives the independence of field and atomic sources.

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APPENDIX A: MEAN MOTION BY DENSITY-MATRIX METHODS

As discussed in the text, our aim is to calculate the system density matrix accurate to second order in the coupling. We are not concerned with treating the bath to the same accuracy, therefore we shall not follow QIII precisely, since the latter treats the bath on the same footing as the system. The chief difference is that we shall now set the density matrix of system and bath $\rho=\sigma f_0+\Delta\rho$, where $\sigma=\mathrm{tr}_R\rho$ is the system density matrix and $f_0=\exp(-\beta R)/\mathrm{tr}[\exp(-\beta R)]$ is the unperturbed bath density matrix. (Previously, we had set $\rho=\sigma f+\Delta\rho$.) The pattern of calculation is the same as in QIII, and most results look very similar with slightly different meanings for the symbols. To avoid confusion, and establish notation, we shall outline the key steps in the argument.

We start with a total Hamiltonian $H_T=H+R+V$ decomposable into a system part H, a reservoir part R, and an interaction

$$V=-\sum Q_jF_j\,, \tag{A1}$$

where the Q's are system operators, and the F's are reservoir operators whose mean values vanish in the decoupled reservoir:

$$\mathrm{tr}_R(F_jf_0)=0\,,$$

where

$$f_0=\exp(-\beta R)/\mathrm{tr}[\exp(-\beta R)]\,. \tag{A2}$$

The density matrix ρ of system+reservoir obeys

$$\partial\rho/\partial t=(H+R+V,\,\rho)\,, \tag{A3}$$

where

$$(A,B)=[A,B]/i\hbar\,. \tag{A4}$$

The trace of (A3) then yields

$$\partial\sigma/\partial t=(H,\sigma)+\mathrm{tr}_R(V,\Delta\rho)\,. \tag{A5}$$

Subtracting $f_0\partial\sigma/\partial t$ from (A3), $\Delta\rho=\rho-f_0\sigma$ obeys

$$\begin{aligned}\partial\Delta\rho/\partial t&=(H+R,\,\Delta\rho)+(V,\sigma f_0)+C(\Delta\rho)\,,\\ C(\Delta\rho)&=(V,\Delta\rho)-f_0\,\mathrm{tr}_R(V,\Delta\rho)\,.\end{aligned} \tag{A6}$$

A systematic expansion in V can be set up by first ignoring C and the iterating. Since we wish σ to second order in V, i.e., $\Delta\rho$ to first order, we stop at the first term

$$\begin{aligned}\Delta\rho\approx\Delta\rho_1&=\int_{-\infty}^{t}(V(t',t),\sigma(t,t')f_0)dt'\,,\\ \partial\sigma/\partial t&=(H,\sigma)+\int_{-\infty}^{t}dt'\,\mathrm{tr}_R(V,(V(t',t),\sigma(t,t')f_0))\,,\end{aligned} \tag{A7}$$

where

$$\begin{aligned}&V(t',t)\equiv U(t,t')VU(t,t')^{-1}\,,\\ &\sigma(t,t')=U(t,t')\sigma(t')U(t,t')^{-1}=u(t,t')\sigma(t')u(t,t')^{-1}\,,\end{aligned} \tag{A8}$$

where $U(t,t')$ is the operator solution of the unperturbed Schrödinger equation

$$\begin{aligned}i\hbar dU(t,t')/dt&=(H+R)U(t,t')\,,\\ U(t',t')&=1\,,\\ U(t,t')&=u(t,t')\exp[-iR(t-t')/\hbar]\,,\\ i\hbar du(t,t')/dt'&=H(t)u(t,t')\,.\end{aligned} \tag{A9}$$

Equation (A7) is very similar to QIII (2.23) with $f(t,t')$ in the latter replaced by f_0, and V obeys $\mathrm{tr}_RVf_0=0$, but the system average of V need not vanish. We have obtained the same result found by Argyres by projection techniques.[19]

Inserting (A1) into (A7), we obtain the analog of QIII (3.6):

$$\begin{aligned}\frac{\partial\sigma}{\partial t}&=(H,\sigma)\\ &+\sum_{i,j}\int_{-\infty}^{t}dt'\{\langle\tfrac{1}{2}[F_i(t-t'),F_j]_+\rangle(Q_i,(Q_j(t',t),\sigma(t,t')))\\ &\quad+\langle(F_i(t-t'),F_j)\rangle(Q_i,\tfrac{1}{2}[Q_j(t',t),\sigma(t,t')]_+)\}\,.\end{aligned} \tag{A10}$$

The trace of (A10) against an arbitrary system operator M yields

$$\begin{aligned}\partial\langle\langle M\rangle\rangle/\partial t&=\langle\langle(M,H)\rangle\rangle\\ &-\hbar^{-2}\sum_{i,j}\int_{-\infty}^{t}dt'\{\langle\tfrac{1}{2}[F_i(t-t'),F_j]_+\rangle\\ &\quad\times\langle\langle[[M(t,t'),Q_i(t,t')],Q_j]\rangle\rangle_{t'}\\ &+\langle[F_i(t-t'),F_j]\rangle\langle\langle\tfrac{1}{2}[[M(t,t'),Q_i(t,t')],Q_j]_+\rangle\rangle_{t'}\}\,,\end{aligned} \tag{A11}$$

where $\langle L\rangle_{t'}=\mathrm{tr}_SL\sigma(t')$. If the reservoir correlation

[19] P. N. Argyres, in *Magnetic and Electric Resonance and Relaxation*, edited by J. Smidt (North-Holland Publishing Company, Inc., Amsterdam, 1963), p. 555.

times are short, the important (t') are close to t. In this small time interval, the system may change rapidly due to its unperturbed motion, but dissipative effects can be assumed small. Thus we can set $\sigma(t,t')$ [which, by (A8) is the density matrix at t obtained from $\sigma(t')$ by propagating with neglect of interactions] approximately equal to $\sigma(t)$:

$$\sigma(t,t')\equiv u(t,t')\sigma(t')u(t,t')^{-1}\approx\sigma(t). \quad (A12)$$

This makes our density matrix equation (A10) Markoffian. Equation (A11) then reduces to the Markoffian form

$$d\langle\langle M\rangle\rangle/dt=\langle\langle(M,H)\rangle\rangle$$
$$-\hbar^{-2}\sum_{i,j}\int_0^\infty du\{\langle F_i(u)F_j\rangle\langle\langle[M,Q_i]Q_j(t-u,\,t)\rangle\rangle$$
$$-\langle F_jF_i(u)\rangle\langle\langle Q_j(t-u,\,t)[M,Q_i]\rangle\rangle\}. \quad (A13)$$

If the system has a time-independent Hamiltonian, $H(t)=H$, then

$$Q_j(t-u,\,t)=\exp(-iHu/\hbar)Q_j\exp(iHu/\hbar)$$
$$\equiv Q_j(-u), \quad (A14)$$

and (A13) reduces to the result (3.1) quoted in the text.

We now wish to apply Eq. (3.1) to the case of an atomic system. The interaction Hamiltonian takes the form

$$V=\hbar\sum a_m^\dagger a_n f_{mn}, \quad (A15)$$

and Hermiticity guarantees

$$V^\dagger=V;\quad f_{mn}^\dagger=f_{nm}. \quad (A16)$$

Making use of the correspondences

$$\begin{aligned}&Q_i\to a_k^\dagger a_l,\quad F_i\to\hbar f_{kl},\\&Q_j(-u)\to a_m^\dagger a_n\exp(-i\omega_{mn}u),\\&F_j\to\hbar f_{mn},\quad M=a_i^\dagger a_j,\end{aligned} \quad (A17)$$

we obtain our equation for the mean motion of an atomic operator $a_i^\dagger a_j$ in the form[20]

$$d\langle a_i^\dagger a_j\rangle/dt=i\omega_{ij}\langle a_i^\dagger a_j\rangle$$
$$-\sum w_{klmn}^+\langle[a_i^\dagger a_j,a_k^\dagger a_l]a_m^\dagger a_n\rangle$$
$$+\sum w_{mnkl}^-\langle a_m^\dagger a_n[a_i^\dagger a_j,a_k^\dagger a_l]\rangle, \quad (A18)$$

where the coefficients defined by

$$w_{klmn}^+=\int_0^\infty du\exp(-i\omega_{mn}u)\langle f_{kl}(u)f_{mn}(0)\rangle,$$
$$w_{mnkl}^-=\int_0^\infty du\exp(-i\omega_{mn}u)\langle f_{mn}(0)f_{kl}(u)\rangle, \quad (A19)$$

[20] For simplicity of notation we change double brackets to single brackets. This is permissible in view of (B16). In this appendix, $\omega_{ij}=\omega_i-\omega_j$ is an unperturbed frequency difference. In the body of the paper, the perturbed frequency $\omega_i-\omega_j+\Delta\omega_{ij}$ is represented by ω_{ij}, for the sake of brevity.

obey the Hermiticity property

$$(w_{mnkl}^-)^*=w_{lknm}^+. \quad (A20)$$

Making use of the identity (4.5), Eq. (A18) can be simplified to the form

$$d\langle a_i^\dagger a_j\rangle/dt=i\omega_{ij}\langle a_i^\dagger a_j\rangle$$
$$+\sum[-\langle a_i^\dagger a_n\rangle w_{jmmn}^++\langle a_k^\dagger a_n\rangle w_{kijn}^+$$
$$+\langle a_m^\dagger a_l\rangle w_{mijl}^--\langle a_m^\dagger a_j\rangle w_{mnni}^-]. \quad (A21)$$

We shall now retain only the secular terms, i.e., those on the right-hand side of (A18) or (A21) that vary as $e^{i\omega_{ij}t}$. This is equivalent to retaining those $w_{klmn}^\pm$ for which[21]

$$\omega_{kl}+\omega_{mn}=0, \quad (A22a)$$

i.e.,

$$k=n,\quad l=m\quad\text{or}\quad k=l,\quad m=n. \quad (A22b)$$

After removal of the rapid time dependence contained in $a_i^\dagger a_j$ these are the only terms which survive a short time average: average over a time Δt short compared to any of the relaxation times but long compared to the reciprocal natural frequencies of the system. An explicit proof of this point is given in Appendix B. The set of conditions (A22) define the only ways in which energy can be conserved if the levels are irregularly spaced. In this Appendix, we henceforth assume that there are no special degeneracies such as would occur for example, in a harmonic oscillator. For this reason we have given a separate treatment of a harmonic oscillator in Sec. 3. Retaining only the secular terms then, Eq. (A21) reduces to the form[20]

$$d\langle a_i^\dagger a_j\rangle/dt=(i\omega_{ij}-\Gamma_{ij}^c)\langle a_i^\dagger a_j\rangle$$
$$+\delta_{ij}\sum_{m\neq i}w_{im}\langle a_m^\dagger a_m\rangle, \quad (A23)$$

where the transition probability w_{im} is defined by

$$w_{im}=w_{miim}^++w_{miim}^-$$
$$=\int_{-\infty}^\infty du\exp(-i\omega_{im}u)\langle f_{mi}(u)f_{im}(0)\rangle \quad (A24)$$

and the complex parameter Γ_{ij}^c is given by

$$\Gamma_{ij}^c=-[w_{iijj}^++w_{iijj}^-]+\sum_{\text{all }m}(w_{jmmj}^++w_{immi}^-), \quad (A25)$$
$$\Gamma_{ij}^c=\Gamma_{ij}-i\Delta\omega_{ij}, \quad (A26)$$
$$\Gamma_{ij}=\tfrac{1}{2}(\Gamma_i+\Gamma_j)+\Gamma_{ij}^{\text{ph}}, \quad (A27)$$
$$\Delta\omega_i=-\text{Im}[\sum_{m\neq j}w_{jmmj}^++\sum_{m\neq i}w_{immi}^-]+\Delta\omega_{ij}^{\text{ph}}, \quad (A28)$$

where Γ_i, Γ_j are the decay rates (1.15) and the first term in (A28) is the "Lamb shift" (second-order

[21] The terms in (A22b) always satisfy (A22a). If the system possesses special regularities of spacing, as in a harmonic oscillator or spin system, then (A22a) permits more secular terms than those explicitly shown in (A22b).

perturbation theory energy shift due to reservoir interactions). The contribution of phase fluctuations or in a solid what might be called zero-phonon contributions[18] are summarized in

$$(\Gamma_{ij}-i\Delta\omega_{ij})^{\text{ph}}=w_{jjjj}^{+}+w_{iiii}^{-}-w_{iijj}^{+}-w_{iijj}^{-}$$
$$=\int_0^\infty du\langle[f_{ii}(0)][f_{ii}(u)-f_{jj}(u)]$$
$$-[f_{ii}(u)-f_{jj}(u)]f_{jj}(0)\rangle, \quad (A29)$$

$$\Gamma_{ij}^{\text{ph}}=\frac{1}{2}\int_0^\infty du\langle[f_{ii}(0)-f_{jj}(0), f_{ii}(u)-f_{jj}(u)]_+\rangle, \quad (A30)$$

$$-i\Delta\omega_{ij}^{\text{ph}}=\frac{1}{2}\int_0^\infty du\langle[f_{ii}(0)+f_{jj}(0), f_{ii}(u)-f_{jj}(u)]\rangle \quad (A31)$$

$$=-i\Delta\omega_i^{\text{ph}}+i\Delta\omega_j^{\text{ph}}-i\Delta\omega_{ij}^{\text{ex}}, \quad (A32)$$

$$-i\Delta\omega_i^{\text{ph}}=\frac{1}{2}\int_0^\infty du\langle[f_{ii}(0),f_{ii}(u)]\rangle, \quad (A33)$$

$$-i\Delta\omega_{ij}^{\text{ex}}=\frac{1}{2}\int_{-\infty}^\infty du\langle[f_{jj}(0),f_{ii}(u)]\rangle. \quad (A34)$$

The *extra* contribution to the frequency shift described by (A34) is *anomalous* in that it is not expressible as the difference between a frequency shift of level i and a shift of level j. In order to understand these formulas we have applied them to the case where the electronic levels are coupled to lattice vibrations through the interaction (A15) with the reservoir forces defined in terms of the normal phonon coordinates by

$$f_{ii}(u)=(\hbar)^{-1}\sum_\mu A_\mu{}^i q_\mu(u). \quad (A35)$$

Neglecting anharmonic interactions between the phonons, the time dependence of these phonon coordinates is given by

$$q_\mu(u)=q_\mu\cos\omega_\mu u+(p_\mu/M\omega_\mu)\sin\omega_\mu u \quad (A36)$$

and the commutator is given by

$$[q_\mu(0),q_\nu(u)]=\delta_{\mu\nu}(i\hbar/M\omega_\mu)\sin\omega_\mu u. \quad (A37)$$

Expressing the time integral

$$\int_0^\infty du\sin\omega_\mu u=\mathcal{P}(1/\omega_\mu) \quad (A38)$$

in terms of the principal-valued reciprocal, the shift in level i due to zero-phonon contributions is given by

$$\Delta\omega_i^{\text{ph}}=-\tfrac{1}{2}\sum(A_\mu{}^i)^2/(\hbar M\omega_\mu{}^2) \quad (A39)$$

and the extra anomalous frequency shift for this case vanishes:

$$\Delta\omega_{ij}^{\text{ex}}=0. \quad (A40)$$

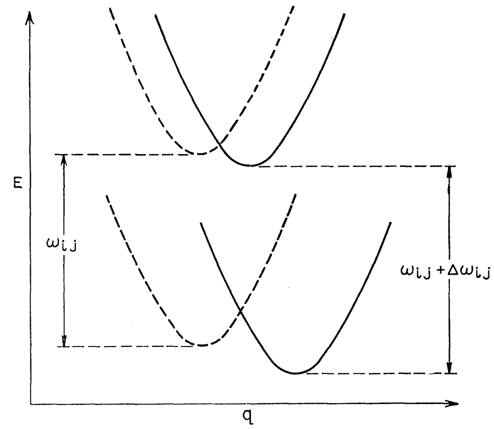

FIG. 2. Configurational coordinate curves of electronic energy E versus normal coordinate q are shown for an electron in two electronic states i and j with $\omega_{ij}=\omega_i-\omega_j>0$. The dashed curves neglect electron-phonon interactions. The solid curves show the shift produced by such interactions. In particular $\Delta\omega_{ij}$ is the change in separation between minima.

The complete energy shift then takes the form

$$\hbar\Delta\omega_{ij}=-\tfrac{1}{2}\sum[(A_\mu{}^i)^2-(A_\mu{}^j)^2]/(M\omega_\mu{}^2). \quad (A41)$$

This result was previously obtained in Eq. (6.7) of our paper on the Franck-Condon principle.[22] The frequency difference ω_{ij} is simply the distance between the minima of the two parabolas shown in Fig. 2 and the level shift $\Delta\omega_{ij}$ is the extent to which this separation has been changed by the linear interactions with the lattice. It is to be emphasized that the perturbative equations on which our mean-motion equation (A23) is based have disregarded the effects of multiphonon transitions. Thus, the frequency ω_{ij} refers to what is customarily called the *zero-phonon line*.[18] This line is indeed a transition from the lowest phonon state in one parabola of Fig. 2 to the lowest phonon state in the other parabola. Since we have neglected changes in the curvatures in these parabolas the zero-point phonon energies cancel and the zero-zero difference is simply the difference between the minima of the parabolas. To see why the anomalous frequency shift vanishes in this case we note that if q is any operator whatever, we can make use of stationarity in the form

$$\langle[q(0),q(t)]\rangle=-\langle[q(t),q(0)]\rangle$$
$$=-\langle[q(0),q(-t)]\rangle. \quad (A42)$$

Thus the integrand of Eq. (A34) is an odd function of the time whereas the integration is taken over an even interval in the time. To get a nonvanishing frequency shift it is necessary therefore to obtain cross terms

$$\langle[q_\mu(u),q_\nu(t)]\rangle\neq 0 \quad \text{for} \quad \Delta\omega_{ij}^{\text{ex}}\neq 0. \quad (A43)$$

We shall now show that even when such cross terms are available if the reservoir obeys time reversal the anomalous frequency shift will vanish.

[22] M. Lax, J. Chem. Phys. 20, 1752 (1952).

The influence of time reversal on our system operators is given by

$$Ka_j^\dagger K^{-1}=a_{Tj}^\dagger\,,\quad Ka_iK^{-1}=a_{Ti}\,, \tag{A44}$$

where Tj and Ti are the time reverses of the states j and i. For any reservoir obeying time reversal, however, we have established[23]

$$\langle L\rangle=\langle\bar{L}\rangle\,, \tag{A45}$$

where the *barring operation* is the combination of Hermitian conjugation and time reversal which obeys

$$V_{\rm bar}\equiv\bar{V}\equiv KV^\dagger K^{-1};\quad (AB)_{\rm bar}=\bar{B}\bar{A}\,;$$
$$B(t)_{\rm bar}=\bar{B}(-t)\,. \tag{A46}$$

Making use of

$$\bar{V}=\hbar\sum(a_i^\dagger a_j)_{\rm bar}\bar{f}_{ij}=\hbar\sum a_{Tj}^\dagger a_{Ti}f_{ij}\,, \tag{A47}$$

we find that the reservoir operators are changed under barring into

$$\bar{f}_{ij}=f_{Tj,Ti}\,. \tag{A48}$$

In this way, we derive

$$\langle[f_{jj}(0),f_{ii}(u)]\rangle=\langle[f_{Ti,Ti}(-u),f_{Tj,Tj}(0)]\rangle$$
$$=-\langle[f_{Tj,Tj}(0),f_{Ti,Ti}(-u)]\rangle\,. \tag{A49}$$

It is clear then that the anomalous frequency shift will vanish unless either Ti differs from i or Tj differs from j or $\langle L\rangle$ does not equal $\langle\bar{L}\rangle$, in other words, if the reservoir violates time reversal, as it would, for example, in any magnetic or antiferromagnetic material.

APPENDIX B: DIRECT CALCULATION OF TRANSPORT EQUATION AND DIFFUSION COEFFICIENTS

An arbitrary-system Heisenberg operator M obeys

$$idM/dt=[M,H_T];\quad H_T=H_0+V;\quad \hbar=1\,, \tag{B1}$$

where $H_0=H+R$ is the sum of system and reservoir Hamiltonians and V is the interaction. We shall assume in this appendix that H_T does not depend explicitly on the time, so that $(H_T)_{\rm Heisenberg}=(H_T)_{\rm Schrödinger}$ and shall hereafter regard H_0 and V as Schrödinger operators. The transformation

$$M(t)=e^{iH_0(t-t_0)}m(t)e^{-iH_0(t-t_0)} \tag{B2}$$

leads to an operator $m(t)$ that varies much more slowly in time [see the similar transformation IV (5.21)]. We can then treat the drift and diffusion of $m(t)$ as if all correlation times are short compared to this motion. After the drift $\langle\Delta m/\Delta t\rangle$ is included in the equation for dm/dt, the reservoir is then eliminated and replaced by Langevin forces with *zero* correlation time that lead to the *same* diffusion. Then, we can transform back to M equations at the time instant t. Thus

$$id\langle M\rangle/dt=[M,H_0]+e^{iH_0(t-t_0)}i\langle\Delta m/\Delta t\rangle e^{-iH_0(t-t_0)}\,, \tag{B3}$$

$$2\langle D_{MN}\rangle=e^{iH_0(t-t_0)}\langle\Delta m\Delta n\rangle/\Delta t e^{-iH_0(t-t_0)}\,, \tag{B4}$$

where

$$\Delta m=m(t+\Delta t)-m(t) \tag{B5}$$

and $\langle\ \rangle$ are reservoir averages. These procedures are not equivalent to calculating $\langle\Delta M/\Delta t\rangle$ and $\langle\Delta M\Delta N\rangle/\Delta t$ (which would unnecessarily smooth the unperturbed motion) but are precisely equivalent to our classical procedures IV (5.24), (5.25). The slow operator m obeys

$$dm/ds=-i[m(s),V(t_0-s)]\,, \tag{B6}$$

where $V(t_0-s)$ is an interaction operator

$$V(t_0-s)\equiv e^{iH_0(t_0-s)}Ve^{-iH_0(t_0-s)}\,. \tag{B7}$$

Equation (B6) can be converted to an integral equation and solved iteratively:

$$\Delta m=-i\int_t^{t+\Delta t}[m(s),V(t_0-s)]ds$$
$$\approx\Delta^{\rm I}m+\Delta^{\rm II}m\,, \tag{B8}$$
$$\Delta^{\rm I}m=-i\int_t^{t+\Delta t}[m(t),V(t_0-s)]ds\,,$$
$$\Delta^{\rm II}m=-\int_t^{t+\Delta t}ds$$
$$\times\int_t^s ds'[[m(t),V(t_0-s')],V(t_0-s)]\,,$$

$$\langle\Delta m/\Delta t\rangle\approx\langle\Delta^{\rm I}m+\Delta^{\rm II}m\rangle/\Delta t=\langle\Delta^{\rm II}m/\Delta t\rangle\,. \tag{B9}$$

Inserting (B9) result into (B3), we obtain

$$\frac{d\langle M\rangle}{dt}=-i[M,H_0]-\frac{1}{\Delta t}\int_t^{t+\Delta t}ds$$
$$\times\int_t^s ds'\langle[[M(t),V(t-s')],V(t-s)]\rangle\,. \tag{B10}$$

Note that t_0, the arbitrary time at which the representations become identical, has disappeared from (B10). This is also true of our expression for the diffusion constant:

$$2D_{MN}=\frac{1}{\Delta t}\int_t^{t+\Delta t}ds\int_t^{t+\Delta t}ds'\langle[M(t),V(t-s')]$$
$$\times[N(t),V(t-s)]\rangle \tag{B11}$$

obtained by inserting $\Delta^{\rm I}m\ \Delta^{\rm I}n$ into (B4).

Equation (B11) is consistent with (B10) in the sense that the use of (B10) plus the Einstein relation (2.9) leads, after combining two sets of terms, to (B11). If

[23] M. Lax, *Symmetry Principles in Solid-State Physics* (to be published), Chap. 10.

we insert (A1): $V=-\sum Q_iF_i$, use the stationarity of reservoir averages, and restore $\hbar$'s, we obtain

$$\frac{d\langle M\rangle}{dt}=\langle(M,H_0)\rangle-(\hbar^2\Delta t)^{-1}\int_t^{t+\Delta t}ds\int_t^s ds'$$
$$\times\{\sum_{ij}\langle F_i(s-s')F_j(0)\rangle\langle[M(t),Q_i(t-s')]Q_j(t-s)\rangle$$
$$-\langle F_j(0)F_i(s-s')\rangle\langle Q_j(t-s)[M(t),Q_i(t-s')]\rangle\}\,,\quad\text{(B12)}$$

$$2D_{MN}=-(\hbar^2\Delta t)^{-1}\sum_{ij}\int_t^{t+\Delta t}ds$$
$$\times\int_t^{t+\Delta t}ds'\langle F_i(s-s')F_j(0)\rangle$$
$$\times\langle[M,Q_i(t-s')][N,Q_j(t-s)]\rangle.\quad\text{(B13)}$$

Again these results are consistent. Let us integrate first over s and set $s=s'+u$ to obtain

$$\frac{d}{dt}\langle M\rangle=\langle(M,H)\rangle-(\hbar^2\Delta t)^{-1}\int_t^{t+\Delta t}ds'\int_0^{t+\Delta t-s'}du$$
$$\times\sum_{ij}\{\langle F_i(u)F_j(0)\rangle\langle[M,Q_i(t-s')]Q_j(t-s'-u)\rangle$$
$$-\langle F_j(0)F_i(u)\rangle\langle Q_j(t-s'-u)[M,Q_i(t-s')]\rangle\}.\quad\text{(B14)}$$

The integral over $\langle F_i(u)F_j(0)\rangle$ converges when the upper limit is greater than the correlation time τ_c over which $\langle F_i(u)F_j(0)\rangle$ is appreciable. If we choose $\Delta t\gg\tau_c$, we can replace the upper limit $t+\Delta t-s'$ by ∞. With $v=s'-t$, we get

$$\frac{d\langle M\rangle}{dt}=\langle(M,H)\rangle-(\hbar^2\Delta t)^{-1}\int_0^{\Delta t}dv\int_0^\infty du$$
$$\times\sum_{ij}\{\langle F_i(u)F_j(0)\rangle\langle[M,Q_i(-v)]Q_j(-v-u)\rangle$$
$$-\langle F_j(0)F_i(u)\rangle\langle Q_j(-v-u)[M,Q_i(-v)]\rangle\}.\quad\text{(B15)}$$

The integral over u represents an integral over the duration of the collision between system and reservoir. The average over v (or s') is an average over the *starting* time of the collision. It is permissible in this average to let $\Delta t\to 0$ to obtain

$$\frac{d\langle M\rangle}{dt}=\langle(M,H)\rangle-\hbar^{-2}\int_0^\infty du$$
$$\times\sum_{ij}\{\langle F_i(u)F_j(0)\rangle\langle[M,Q_i]Q_j(-u)\rangle$$
$$-\langle F_j(0)F_i(u)\rangle\langle Q_j(-u)[M,Q_i]\rangle\}\,,\quad\text{(B16)}$$

the result (3.1) derived in (A13). This result, however, retains certain rapidly oscillatory terms whose effect on the long-term motion is of second order. These terms will automatically disappear if Δt is kept large enough. In particular, the system-operator time dependence must be representable as a sum of exponentials

$$Q_i(t)=\sum_\alpha Q_{i\alpha}e^{i\omega_i^\alpha t}.\quad\text{(B17)}$$

The integral over v will involve integrals of the form

$$\frac{1}{\Delta t}\int_0^{\Delta t}dv\,\exp[i\omega_i^\alpha t+i\omega_j^\beta t].\quad\text{(B18)}$$

For $\omega_i^\alpha+\omega_j^\beta\neq 0$, we shall choose Δt large enough so that

$$(\omega_i^\alpha+\omega_j^\beta)\Delta t\gg 1.\quad\text{(B19)}$$

All such terms can then be neglected unless

$$\omega_i^\alpha+\omega_j^\beta=0.\quad\text{(B20)}$$

For the *secular terms* which obey (B20), the integral over v is insensitive to Δt and one can let $\Delta t\to 0$. Thus, the sole effect of the average over v (or s') is to retain only the secular terms when (B15) is expanded using (B17). Our result can be written in the form

$$\frac{d\langle M\rangle}{dt}=\langle(M,H)\rangle-\hbar^{-2}\sum_{i\alpha,j\beta}\delta(\omega_i^\alpha,-\omega_j^\beta)$$
$$\times\left\{\int_0^\infty e^{-i\omega_j^\beta u}\langle F_i(u)F_j(0)\rangle du\langle[M,Q_{i\alpha}]Q_{j\beta}\rangle\right.$$
$$\left.-\int_0^\infty e^{-i\omega_j^\beta u}\langle F_j(0)F_i(u)\rangle du\langle Q_{j\beta}[M,Q_{i\alpha}]\rangle\right\}\,,\quad\text{(B21)}$$

where the Kronecker delta, $\delta(\omega_i^\alpha,-\omega_j^\beta)$, selects the secular terms.

If we apply a similar procedure to (B13), we get

$$2D_{MN}=-\hbar^{-2}\sum_{\alpha i,\beta j}\delta(\omega_i^\alpha,-\omega_j^\beta)\int_{-\infty}^\infty e^{-i\omega_j^\beta u}du$$
$$\times\langle F_i(u)F_j(0)\rangle\langle[M,Q_{i\alpha}][N,Q_{j\beta}]\rangle.\quad\text{(B22)}$$

If the Einstein relation (2.9) is applied to (B21), and the summation indices αi and βj are interchanged in the second term, the same result (B22), is obtained. Thus the Einstein method and the direct method are necessarily in agreement.

The variables Q_j can often be so chosen that

$$Q_j(t)=Q_j\exp(i\omega_jt)\,,\quad\text{(B23)}$$

and the indices α, β are superfluous. For a harmonic oscillator, this suggests the use of b and $b^\dagger$ rather than Q and P. For multilevel "atomic" system the variables $a_i^\dagger a_j$ already obey this requirement. The translation of variables

$$M\to a_i^\dagger a_j\,,\quad Q_i\to a_k^\dagger a_l\,,\quad Q_j\to a_m^\dagger a_n\,,$$
$$F_i\to\hbar f_{kl}\,,\quad F_j\to\hbar f_{mn}\quad\text{(B24)}$$

in (B21) leads to

$$d\langle a_i^\dagger a_j\rangle/dt = i\omega_{ij}\langle a_i^\dagger a_j\rangle - \sum \delta(\omega_{kl}, -\omega_{mn})\{\langle[a_i^\dagger a_j, a_k^\dagger a_l]a_m^\dagger a_n\rangle w_{klmn}^{+} - \langle a_m^\dagger a_n[a_i^\dagger a_j, a_k^\dagger a_l]\rangle w_{klmn}^{-}\}, \quad \text{(B25)}$$

i.e., to Eq. (A18) with the selection of secular terms built in, and the w^+, w^- defined as in (A19).

A similar translation of variables in (B22) leads to

$$2\langle D_{ijkl}\rangle = -\sum \delta(\omega_{mn}, -\omega_{pq})\langle[a_i^\dagger a_j, a_m^\dagger a_n] \times [a_k^\dagger a_l, a_p^\dagger a_q]\rangle w_{mnpq}, \quad \text{(B26)}$$

$$w_{mnpq} = \int_{-\infty}^{\infty} du\, e^{-i\omega_{pq}u}\langle f_{mn}(u) f_{pq}(0)\rangle = w_{mnpq}^{+} + w_{mnpq}^{-}. \quad \text{(B27)}$$

Using (4.5), (B20) can be simplified to

$$2\langle D_{ijkl}\rangle = \sum\langle -a_i^\dagger a_q w_{jklq} + a_i^\dagger a_l w_{jnnk} + a_m^\dagger a_q w_{milq}\delta_{jk} + a_m^\dagger a_l w_{mijk}\rangle. \quad \text{(B28)}$$

In (B28), we assume each factor w_{mnpq} carries with it, the corresponding selection factor $\delta(\omega_{mn}, -\omega_{pq})$. Equations (B25)–(B28) are sufficiently general to *include such regularly spaced systems as harmonic oscillators.* If no regularities occur among the spaces, the only secular terms that remain obey (A22b), i.e., the surviving terms have the form w_{mnnm} and w_{mmnn}.

For the former, one has the identification

$$w_{mnnm} = w_{mnnm}^{+} + (w_{mnnm}^{+})^* = w_{nm} \quad \text{(B29)}$$

(see A20 and A24) in terms of transition probabilities. Retaining only these secular terms, (B28) reduces to

$$2D_{ijkl} = w_{kj}\langle a_i^\dagger a_j\rangle\delta_{kl} - w_{ik}\langle a_k^\dagger a_l\rangle\delta_{ij} + \sum_{m\neq i} w_{im}\langle a_m^\dagger a_m\rangle\delta_{il}\delta_{jk} + \langle a_i^\dagger a_l\rangle \times [\Gamma_j + w_{jjjj} - w_{iijj} + w_{iill} - w_{jjll}]\delta_{jk}. \quad \text{(B30)}$$

Using Eqs. (A20) and (A29), we can write

$$(\Gamma_{ij} - i\Delta\omega_{ij})^{\text{ph}} = \tfrac{1}{2}[w_{jjjj} + w_{iiii} - 2w_{iijj}]. \quad \text{(B31)}$$

Thus, the coefficient of the last term in Eq. (B30) can be rewritten in the form

$$\Gamma_j + w_{jjjj} - w_{iijj} + w_{iill} - w_{jjll} = \Gamma_{ij} + \Gamma_{jl} - \Gamma_{il} - i(\Delta\omega_{ij} + \Delta\omega_{jl} - \Delta\omega_{il}), \quad \text{(B32)}$$

which brings it into agreement with the result (4.6) of the Einstein method.

APPENDIX C: A MARKOFFIAN AND NON-MARKOFFIAN DISCUSSION OF HARMONIC-OSCILLATOR COMMUTATION RULES

Markoffian Commutation Rules

If we subtract Eq. (3.8) from the corresponding equation for $\langle bb^\dagger\rangle$, we find that

$$d\langle[b,b^\dagger]\rangle/dt = \gamma - \gamma\langle[b,b^\dagger]\rangle \quad \text{(C1)}$$

so that, if the commutation rule $\langle[b,b^\dagger]\rangle = 1$ is obeyed at any time, it will be obeyed forever after, even in the Markoffian approximation.

As a further check on our consistency, let us define[24]

$$\langle b_\omega^\dagger b_\omega\rangle = \int_{-\infty}^{\infty} e^{-i\omega t}\langle b^\dagger(t) b(0)\rangle dt,$$
$$\langle b^\dagger(t) b(0)\rangle = \frac{1}{2\pi}\int_{-\infty}^{\infty} e^{i\omega t}\langle b_\omega^\dagger b_\omega\rangle d\omega. \quad \text{(C2)}$$

The transforms of Eqs. (3.9) then yield

$$[\tfrac{1}{2}\gamma + i(\omega_0 - \omega)]b_\omega = f_\omega; \quad [\tfrac{1}{2}\gamma - i(\omega_0 - \omega)]b_\omega^\dagger = f_\omega^\dagger, \quad \text{(C3)}$$

$$\langle b_\omega^\dagger b_\omega\rangle = [(\tfrac{1}{2}\gamma)^2 + (\omega_0 - \omega)^2]^{-1}\langle f_\omega^\dagger f_\omega\rangle, \quad \text{(C4)}$$

$$\langle f_\omega^\dagger f_\omega\rangle = \int_{-\infty}^{\infty} e^{-i\omega t}\langle f^\dagger(t) f(0)\rangle dt = \gamma\bar{n}. \quad \text{(C5)}$$

Set $t=0$ in the second Eq. (C2) and insert (C4) to obtain

$$\langle b^\dagger b\rangle = \frac{1}{2\pi}\int_{-\infty}^{\infty} [(\tfrac{1}{2}\gamma)^2 + (\omega_0 - \omega)^2]^{-1}\gamma\bar{n}\, d\omega = \bar{n}. \quad \text{(C6)}$$

Similarly,

$$\langle bb^\dagger\rangle = \bar{n} + 1 \quad \text{(C7)}$$

and again the commutation rules are preserved.

Relation between Langevin Forces and Reservoir Forces

Because of the linear nature of our system, there is little distinction between the Langevin force f (the extra term on the right-hand side of a dynamic equation), and the reservoir force g that gives rise to the

[24] For any two random operators $A(t)$, $B(t)$, we define

$$A(\omega) = \int_{-\infty}^{\infty} e^{i\omega s} A(s) ds,$$
$$B^\dagger(\omega') = \int_{-\infty}^{\infty} e^{-i\omega' t} B^\dagger(t) dt = [B(\omega')]^\dagger.$$

Assuming stationarity, setting $t = s + u$, and integrating over u, we find for the product average

$$\langle A(\omega) B^\dagger(\omega')\rangle = 2\pi\delta(\omega - \omega')\langle A_\omega B_\omega^\dagger\rangle,$$

where

$$\langle A_\omega B_\omega^\dagger\rangle \equiv \int_{-\infty}^{\infty} du\, e^{-i\omega u}\langle A(0) B^\dagger(u)\rangle.$$

This procedure follows our classical discussion IV, footnote 13, with $\alpha \to B^\dagger$, $\alpha^* \to A$ since in the quantum-mechanical case, it is usually the daggered operators that carry the positive frequencies. Similarly,

$$\langle A^\dagger(\omega) B(\omega')\rangle = 2\pi\delta(\omega - \omega')\langle A_\omega^\dagger B_\omega\rangle,$$
$$\langle A_\omega^\dagger B_\omega\rangle \equiv \int_{-\infty}^{\infty} e^{-i\omega u} du \langle A^\dagger(u) B(0)\rangle.$$

A simple rule to remember is that if we stick to $\exp(-i\omega u)$, it is always the daggered operator that carries the time dependence, and orders of operators are always preserved. One can readily verify, by integration by parts, that it is appropriate to use the rules

$$(dA/dt)_\omega = -i\omega A_\omega; \quad (dA^\dagger/dt)_\omega = i\omega A^\dagger_\omega.$$

Langevin force. The Heisenberg equation

$$db/dt=-i\omega_c b+g(t) \tag{C8}$$

suggests that we set

$$f(t)=g(t)-\langle g(t)\rangle_p; \quad \langle g(t)\rangle_p=-(\tfrac{1}{2}\gamma+i\Delta\omega)b, \tag{C9}$$

where $\langle\ \rangle_p$ is an average over the reservoir as it is perturbed by the system. This interpretation is permissible for non-Markoffian systems. We have, however, made a Markoffian approximation by giving f a flat ("white") frequency spectrum chosen automatically by (3.4) to coincide with the spectrum of g at the frequency ω_c. Thus, for example,

$$\bar{n}=\frac{\mathrm{Re}\beta}{\frac{1}{2}\gamma}=\frac{\mathrm{Re}\int_0^\infty du\, e^{-i\omega_c u}\langle g^\dagger(u)g\rangle}{\mathrm{Re}\int_0^\infty du\, e^{-i\omega_c u}\langle[g,g^\dagger(u)]\rangle} \tag{C10}$$

$$=[\exp(\hbar\omega_c/kT_R)-1]^{-1}. \tag{C11}$$

To obtain the last step, we split $g^\dagger(u)g$ into an anticommutator plus a commutator. We next use the fact that the anticommutator is a real, even function of u, and the commutator is an imaginary, odd function of u to extend the integrals to $-\infty$. If the reservoir is at equilibrium at temperature T_R, we can then use the usual relation QIII (7.7) relating commutators and anticommutators to obtain (C11).

The dissipation coefficient $\frac{1}{2}\gamma$ and frequency shift $\Delta\omega$ regarded as functions of ω_c can be written

$$\tfrac{1}{2}\gamma(\omega_c)=-i\int_0^\infty du\, \sin\omega_c u\langle[g,g^\dagger(u)]\rangle, \tag{C12}$$

$$\Delta\omega(\omega_c)=i\int_0^\infty du\, \cos\omega_c u\langle[g,g^\dagger(u)]\rangle, \tag{C13}$$

so that the Hilbert transform relationship

$$\cos\omega_c u=-\frac{1}{\pi}\mathcal{P}\int_{-\infty}^{\infty}\frac{\sin\omega u}{\omega-\omega_c}d\omega \tag{C14}$$

leads to a Kramers-Kronig relation[25]:

$$\Delta\omega(\omega_c)=-\frac{1}{\pi}\mathcal{P}\int_{-\infty}^{\infty}\frac{d\omega}{\omega-\omega_c}\tfrac{1}{2}\gamma(\omega). \tag{C15}$$

For the case $\gamma(\omega)=\gamma=\mathrm{const}$, $\Delta\omega=0$. In any case, $\Delta\omega$ is usually small compared to the range of frequencies over which $\gamma(\omega)$ varies significantly. Since we must assume that the spectrum of g varies slowly near ω_c, we cannot, with the accuracy of the present discussion distinguish between $\gamma(\omega_c)$ and $\gamma(\omega_0)$. We shall conjecture, that a somewhat more self-consistent analysis would require us to absorb $\Delta\omega$ into a new unperturbed frequency ω_0 and, in our Markoffian analysis, evaluate $\gamma=\gamma(\omega_0)$, $\bar{n}=n(\omega_0)$ at the new frequency ω_0.

It is really more accurate to write

$$\langle e^{-i\omega_0 t}f^\dagger(t)e^{i\omega_0 u}f(u)\rangle\approx\gamma\bar{n}\delta(t-u) \tag{C16a}$$

than

$$\langle f^\dagger(t)f(u)\rangle\approx\gamma\bar{n}\delta(t-u), \tag{C16b}$$

since these equations imply

$$\gamma\bar{n}=\int_{-\infty}^{\infty}e^{-i\omega_0 t}\langle f^\dagger(t)f(0)\rangle dt \tag{C17a}$$

or

$$\gamma\bar{n}=\int_{-\infty}^{\infty}\langle f^\dagger(t)f(0)\rangle dt, \tag{C17b}$$

respectively. Within the Markoffian limit of delta-function autocorrelations, there is no distinction between these alternatives, but if the correlation time is long compared to ω_0^{-1}, the first of these alternatives is preferable. We shall therefore define

$$b'=be^{i\omega_0 t}, \quad (b')^\dagger=b^\dagger e^{-i\omega_0 t}, \tag{C18}$$

$$F=fe^{i\omega_0 t}, \quad F^\dagger=f^\dagger e^{-i\omega_0 t}, \tag{C19}$$

so that our new equations of motion are

$$db'/dt=-\tfrac{1}{2}\gamma b'+F(t),$$
$$d(b')^\dagger/dt=-\tfrac{1}{2}\gamma(b')^\dagger+F^\dagger(t), \tag{C20}$$

with

$$\langle F^\dagger(t)F(u)\rangle\approx\gamma\bar{n}\delta(t-u),$$
$$\langle F(u)F^\dagger(t)\rangle\approx\gamma(\bar{n}+1)\delta(t-u). \tag{C21}$$

The $\delta(t-u)$ could then have a width of the order of the correlation time without changing the results appreciably. These points are somewhat beyond the scope of a Markoffian approach.

Non-Markoffian Discussion of Harmonic Oscillators

As mentioned in the Introduction near Eq. (1.41), this consistent fusion of quantum mechanics with a Markoffian description has been achieved by using a rotating-wave approximation (RWA). The RWA was introduced by the choice of the interaction V in (3.2). The full interaction would have been

$$V=-QG; \qquad G=(2\hbar\omega_c)^{1/2}(g+g^\dagger), \tag{C22}$$

$$Q=(\hbar/2\omega_c)^{1/2}i(b-b^\dagger); \quad P=(\tfrac{1}{2}\hbar\omega_c)^{1/2}(b+b^\dagger) \tag{C23}$$

and would have lead to a decay term of the form $\frac{1}{2}\gamma(b+b^\dagger)$ in both the db/dt and $db^\dagger/dt$ equations, or to the equations

$$dQ/dt=P; \quad dP/dt=-\omega_0^2Q-\gamma P+F(t). \tag{C24}$$

[25] N. N. Bogoliubov and D. V. Shirkov, *Introduction to the Theory of Quantized Fields* (Interscience Publishers, Inc., New York, 1959), Sec. 46. Equation (C15) is really an equation for $\Delta\omega(\omega_c)-\Delta\omega(\infty)$, but we can usually assume $\Delta\omega(\infty)=0$.

A proper comparison with the full interaction procedure can only be made outside the Markoffian framework. The Langevin equations (C24) when combined and written in Fourier form lead to[24]

$$[\omega_0^2-\omega^2-i\omega\gamma]Q_\omega=F_\omega \tag{C25}$$

so that the spectrum of F is given by

$$F_\omega^\dagger F_\omega=DQ_\omega^\dagger Q_\omega;\quad D=[(\omega_0^2-\omega^2)^2+(\omega\gamma)^2], \tag{C26}$$

where

$$\langle Q_\omega^\dagger Q_\omega\rangle=\int_{-\infty}^{\infty}e^{-i\omega t}\langle Q(t)Q(0)\rangle dt \tag{C27}$$

can be found from QIII Sec. 7 to be

$$\langle Q_\omega^\dagger Q_\omega\rangle=2\gamma(\omega)\hbar\omega\bar{n}(\omega)/D(\omega), \tag{C28}$$

where the dissipation coefficient γ in the non-Markoffian case is allowed to be frequency-dependent. In this way, we find

$$\langle F_\omega^\dagger F_\omega\rangle=2\gamma(\omega)\hbar\omega\bar{n}(\omega), \tag{C29}$$

$$\langle F_\omega F_\omega^\dagger\rangle=2\gamma(\omega)\hbar\omega[\bar{n}(\omega)+1], \tag{C30}$$

$$\bar{n}(\omega)=[\exp(\hbar\omega/kT)-1]^{-1}, \tag{C31}$$

where T is the temperature of the reservoir with which the harmonic oscillator interacts. If the reservoir consists of a set of harmonic oscillators, these results are, in fact, exact.[26]

The results (C29), (C30) are not independent since

$$F_\omega^\dagger=F_{-\omega};\quad \gamma(-\omega)=\gamma(\omega),\quad \bar{n}(-\omega)=-[\bar{n}(\omega)+1]. \tag{C32}$$

The commutator is given by

$$\langle[F_\omega,F_\omega^\dagger]\rangle=2\hbar\omega\gamma(\omega) \tag{C33}$$

an odd function of ω. *The noise cannot therefore be made white even when $\gamma(\omega)=\gamma=a$ constant.* It is easy to verify in this case that the commutation rules are obeyed by using

$$\langle[Q,P(t)]\rangle=\frac{1}{2\pi}\int_{-\infty}^{\infty}e^{i\omega t}d\omega\langle[Q_\omega,P_\omega^\dagger]\rangle, \tag{C34}$$

$$P_\omega^\dagger=i\omega Q_\omega^\dagger, \tag{C35}$$

$$\langle[Q,P]\rangle=\frac{1}{2\pi}\int d\omega i\omega\langle[F_\omega,F_\omega^\dagger]\rangle/D(\omega). \tag{C36}$$

If we use the commutator (C33) with $\gamma(\omega)=\gamma$; $\omega_0=$ const,

$$\langle[Q,P]\rangle=\frac{i\hbar}{\pi}\int d\omega\frac{\gamma\omega^2}{(\omega^2-\omega_0^2)^2+(\omega\gamma)^2}=i\hbar. \tag{C37}$$

Senitzky[13] has chosen the commutation rule (1.38) in terms of principal-valued reciprocals. Thus,

$$\langle[F_\omega,F_\omega^\dagger]\rangle_{\text{Senitzky}}=2\gamma\hbar\omega_0\omega/|\omega|. \tag{C38}$$

The insertion of (C38) into (C36) then yields

$$\langle[Q,P]\rangle=i\hbar[1-\mu^2]^{-1/2}\times\{1-1/\pi\arctan[2\mu(1-\mu^2)^{1/2}]\}, \tag{C39}$$

where $\mu=(\gamma/2\omega_0)$. This result has errors of first order in γ/ω_0 as shown in (1.39).

If we were to regard (C22) as a split of F into its positive- and negative-frequency parts, we would have

$$\begin{aligned}\langle f_\omega f_\omega^\dagger\rangle&=(\omega/\omega_0)\gamma[\bar{n}(\omega)+1],\quad &\omega>0\\&=0, &\omega<0.\end{aligned} \tag{C40}$$

If, furthermore, we were to set $\omega=\omega_0$ in (C40), this would lead to a spectrum that is white except for a jump at $\omega=0$ from the spectrum at $-\omega_0$ to that at ω_0. This is Senitzky's result. Our procedure is equivalent to the choice

$$\begin{aligned}\langle f_\omega f_\omega^\dagger\rangle&=\gamma[\bar{n}(\omega_0)+1],\quad &\text{all }\omega,\\ \langle f_\omega^\dagger f_\omega\rangle&=\gamma\bar{n}(\omega_0), &\text{all }\omega,\end{aligned} \tag{C41}$$

i.e., two white spectra with no jump. Within the RWA, our procedure leads to exact preservation of the commutation rules as shown in (C1) and (C6).

APPENDIX D: SOLUTION OF THE MARKOFFIAN HARMONIC-OSCILLATOR DENSITY-MATRIX EQUATION

Instead of proving the exactness of (3.6), we shall instead derive an equation for the density matrix σ of a Markoffian harmonic oscillator. We shall then solve the equation for σ and show that the solution for σ is identical to the exact solution found by Louisell and Walker[13] for the case of rotating-wave coupling to a reservoir consisting of a set of harmonic oscillators.

An equation for $\sigma(t)$ can be obtained by combining (A10) and (A12). To avoid having to re-identify the constants, we shall start instead by rewriting (3.3), using the identification (3.5), in the form

$$\partial\langle M\rangle/\partial t=\langle(M,\hbar\omega_0 b^\dagger b)\rangle+\tfrac{1}{2}\gamma\langle b^\dagger[M,b]-[M,b^\dagger]b\rangle+\gamma\bar{n}\langle[b,[M,b^\dagger]]\rangle. \tag{D1}$$

We then note that if

$$\partial\sigma/\partial t=\textstyle\sum_i A_i\sigma B_i, \tag{D2}$$

where A_i and B_i are any operators,

$$\partial\langle M\rangle/\partial t=\textstyle\sum_i\langle B_i M A_i\rangle \tag{D3}$$

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[26] By using the Heisenberg equations of motion and solving for the reservoir oscillators in terms of the known motion of the system oscillator, one can readily show that the mean equation of motion of our system oscillator, e.g., QIII (4.13) (omitting $j\neq i$ terms) is exact. The spectrum (C28) or QIII (7.11) is then exact since it uses only the correct mean motion and the fluctuation-dissipation theorem. Note, however, that if $\gamma(\omega)\neq$constant we must also regard $\omega_0^2\equiv\omega_c^2+\text{Re}b(\omega)$ of QIII (4.13) as frequency-dependent. The moments of the Langevin forces (C29), (C30) are expressible directly in terms of $\gamma(\omega)$, independently of ω_0.

so that the equation for σ can be obtained by inspecting the equation for $\langle M\rangle$. After some rearrangement, our equation for σ takes the form

$$\partial\sigma/\partial t=(\hbar\omega_0 b^\dagger b,\sigma)+\gamma\sigma+\tfrac{1}{2}\gamma\{b^\dagger[b,\sigma]+[\sigma,b^\dagger]b\}+\gamma(\bar{n}+1)[[b,\sigma],b^\dagger], \quad \text{(D4)}$$

$$\partial\sigma/\partial t=(\hbar\omega_0 b^\dagger b,\sigma)+\gamma\sigma+\tfrac{1}{2}\gamma\{b^\dagger\partial\sigma/\partial b^\dagger+\partial\sigma/\partial b b\}+\gamma(\bar{n}+1)\partial^2\sigma/\partial b\partial b^\dagger, \quad \text{(D 5)}$$

where we use

$$\frac{\partial}{\partial b}\frac{\partial\sigma}{\partial b^\dagger}\equiv[[b,\sigma],b^\dagger]=\frac{\partial}{\partial b^\dagger}\frac{\partial\sigma}{\partial b}\equiv[b,[\sigma,b^\dagger]]. \quad \text{(D6)}$$

The Louisell-Walker solution[13] (when no external driving forces are present) is a function only of $b^\dagger b$. For such a σ, the first term in (D4) or (D5) vanishes. The remaining terms in (D5) have, with malice aforethought, been arranged so that if $\sigma=\sigma^{(n)}$ is in normal order[27] (all $b^\dagger$ operators to the left of all b operators) these terms are already in normal order.

To solve (D5) we use the normal ordering operator $\mathfrak{N}$ discussed by Louisell.[27] If $g(\bar{b},\bar{b}^\dagger)$ is any classical function of the c numbers $\bar{b}$ and $\bar{b}^\dagger$, and if $g^n(\bar{b},\bar{b}^\dagger)$ is the same classical function arranged so that all $(\bar{b}^\dagger)$'s appear to the left of all $\bar{b}$'s, then the operator obtained by replacing $\bar{b}$ by b and $\bar{b}^\dagger$ by $b^\dagger$ is written

$$g^n(b,b^\dagger)\equiv\mathfrak{N}\{g^n(\bar{b},\bar{b}^\dagger)\}. \quad \text{(D7)}$$

Thus, the operator $\mathfrak{N}$ converts a c-number function to an operator (that is already in normal order) by a definite rule.

Let us now assume that a solution to (D5) can be written in the form

$$\sigma^{(n)}(b,b^\dagger)=\mathfrak{N}\{S(z)\}\,;\quad z=\bar{b}^\dagger\bar{b}. \quad \text{(D8)}$$

Since differentiations do not disturb normal order, we have

$$\partial\sigma^{(n)}/\partial b b=\mathfrak{N}\{\partial S(z)/\partial\bar{b}\bar{b}\}. \quad \text{(D9)}$$

[27] For a lucid account of normal ordering and the normal ordering operator $\mathfrak{N}$ see W. H. Louisell, *Radiation and Noise in Quantum Electronics* (McGraw-Hill Book Company, Inc., New York, 1964), Chap. 3.

Underneath the $\mathfrak{N}$, we have only c numbers, and can use the usual rules of differentiation, and can write factors in any order. Thus,

$$\partial\sigma^{(n)}/\partial b b=\mathfrak{N}\{\bar{b}^\dagger\partial S/\partial z\bar{b}\}=\mathfrak{N}\{z\partial S/\partial z\}, \quad \text{(D10)}$$

Eq. (D5) can in this manner be rewritten as

$$\mathfrak{N}\left\{\frac{\partial S(z)}{\partial t}\right\}=\mathfrak{N}\left\{\gamma\frac{\partial}{\partial z}(zS)+\gamma(\bar{n}+1)\frac{\partial}{\partial z}\left[z\left(\frac{\partial S}{\partial z}\right)\right]\right\}, \quad \text{(D11)}$$

from which we obtain the c-number equation

$$\frac{\partial S(z,t)}{\partial t}=\gamma\frac{\partial}{\partial z}(zS)+\gamma(\bar{n}+1)\frac{\partial}{\partial z}\left[z\frac{\partial S}{\partial z}\right]. \quad \text{(D12)}$$

By direct substitution, we can verify that

$$S(z,t)=[y(t)]^{-1}\exp[-z/y(t)] \quad \text{(D13)}$$

is a solution of (D12), provided that y obeys

$$dy/dt=\gamma(\bar{n}+1)-\gamma y \quad \text{(D14)}$$

so that the density matrix is given by

$$\sigma=[y(t)]^{-1}\mathfrak{N}\{\exp[-\bar{b}^\dagger\bar{b}/y(t)]\}. \quad \text{(D15)}$$

The relationship Eq. (3.68) of Louisell[27]

$$\mathfrak{N}\{\exp[(e^x-1)\bar{b}^\dagger\bar{b}]\}=\exp(xb^\dagger b) \quad \text{(D16)}$$

permits this result to be rewritten in the form

$$\sigma=\frac{1}{y(t)}\left[1-\frac{1}{y(t)}\right]^{b^\dagger b} \quad \text{(D17)}$$

from which it is evident that

$$\mathrm{tr}\sigma=1. \quad \text{(D18)}$$

The relations (D14), (D15) define the exact Louisell-Walker[13] solution for the case $\gamma=\text{const}$ (independent of frequency), and $y(t)$ has the interpretation

$$y(t)=\langle b(t)b^\dagger(t)\rangle. \quad \text{(D19)}$$