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Semiclassical theory of stability in a laser using a generalized entropy*

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The stability criteria in the pumping and lasing states in a cw solid-state laser are investigated using the semiclassical theory of the multimode laser. The model admits of one (trivial) steady state corresponding to thermal equilibrium, for which an N-body principle of minimum-entropy production may be obtained the entropy production serving as a Lyapounov function in this case. The other (multiple) steady states are far from thermal equilibrium. It is possible to construct an N-body Lyapounov function in the general, multimode case for these states within the framework of the rate approximation. Near equilibrium, the linear thermodynamics of the multimode laser are developed with the entropy production.

I. INTRODUCTION

The theory of the generalized entropy developed by Prigogine *et al.*[1] in Brussels and Austin[1-3] suggests a form for an N-body Lyapounov function. The starting point is the Liouville-von Neumann equation for the density operator ρ,

$$i\frac{\partial\rho}{\partial t}=L\rho\,, \tag{1.1}$$

where $L\equiv[H,\ ]$ is the Hermitian Liouville operator. For isolated systems, a "causal representation" may be introduced wherein the density operator $\rho^{(p)}$ is obtained from ρ, through a nonunitary transformation

$$\rho^{(p)}(t)=\Lambda^{-1}(L)\rho(t)\,. \tag{1.2}$$

For weakly coupled systems (as discussed in this paper), $\Lambda=1$ and $\rho^{(p)}$ reduces to the ρ of Eq. (1.1). In the new representation, one may construct a generalized entropy functional,

$$S=-\tfrac{1}{2}\ln\Omega^{(p)}\,, \tag{1.3}$$

where

$$\Omega^{(p)}=\mathrm{Tr}\rho^{(p)\dagger}\rho^{(p)}\,. \tag{1.4}$$

This entropy includes the diagonal and off-diagonal elements of the density operator. The transformation theory mentioned above has been used to study damping and decay in the Friedrichs model.[1] Further, this form of the entropy has been used by Hubert[4] to construct the entropy for a dense hard-sphere gas obeying the Enskog equation and by Henin[5] to study the approach to equilibrium in the stochastic models of Kac and McKean, using a Markovian description. However, little is yet known concerning the formal extension to open systems. A beginning in this direction has been made by McAdory and Schieve,[6] who construct an entropy functional for a stochastic model of a system in weak interaction with a reservoir and obeying Markovian kinetic equations.

In this paper, we demonstrate how the generalized Lyapounov function mentioned above may be applied to investigate the stability of yet another open system. Our model consists of N identical two-level atoms in interaction with a multimode electric field in a cavity. This is a realistic description of a cw laser, which we shall discuss using the semiclassical theory.[7] In the next section, we briefly review the semiclassical theory to introduce the notation and develop the rate approximation. This is used to construct the Lyapounov function for the nonequilibrium steady states. Finally, we calculate the entropy production near equilibrium and prove it to be a minimum, thus obtaining linear irreversible thermodynamics. It should be noted that the only other attempt to construct a Lyapounov function for such a model was made by Hofelich-Abate and Hofelich,[8] who constructed a functional $V(x,y)$ which vanished only at the origin and was positive everywhere else, thereby showing that the equilibrium branch was asymptotically stable in the entire physical region of phase space (corresponding to positive photon numbers).[9] Further, Walgraef[10] has used a Fokker-Planck description to derive a Gibbs entropy for the equilibrium branch and to discuss its stability using the Glansdorff-Prigogine theory.[11] We show here that we may obtain Walgraef's results as well as the stability of the nonequilibrium branch using a simple semiclassical approach.

II. SEMICLASSICAL THEORY AND THE RATE EQUATIONS

The assumption of the semiclassical approach[7] is to treat the electric field in the cavity as being purely classical, describable by Maxwell's equations, the atoms being treated quantum mechanically. We will consider a model of a cw solid-state laser, consisting of N homogeneously-broadened identical two-level atoms interacting with a transverse radiation field. The complete set of semiclassical equations then has the following form[12]:

$$\dot{\rho}_{12,\mu}(t) = (i\omega_{12,\mu} - \Gamma_{12,\mu})\rho_{12,\mu} - i\sum_{\lambda} g_{\lambda,\mu} b_\lambda^* d_\mu , \quad (2.1)$$

$$\dot{b}_\lambda^*(t) = (i\omega_\lambda - \kappa_\lambda) b_\lambda^* + i\sum_{\mu} g^*_{\lambda,\mu}\rho_{12,\mu} , \quad (2.2)$$

$$\dot{d}_\mu = \frac{d_\mu^0 - d_\mu}{T_1} + 2i\sum_{\lambda} (g_{\lambda,\mu} b_\lambda^* \rho_{21,\mu} - \text{c.c.}) , \quad (2.3)$$

together with the conjugate equations for $\rho_{21,\mu}$ and b_λ. Here, the indices λ and μ refer to the cavity mode and the atom respectively, and the symbols used above have the following meanings: ω_λ is the cavity mode frequency, ω_{12} the frequency separation of atomic levels, Γ_{12} the atomic half width, κ_λ the cavity half width, T_1 the atomic relaxation time, and g the coupling constant. ρ_{12} and ρ_{21} are the correlation components of the atomic-density matrix, ρ_{11} and ρ_{22} being the diagonal components or probabilities. We have further defined $d_\mu \equiv (\rho_{22} - \rho_{11})_\mu \equiv (N_2 - N_1)_\mu$, the population difference of the levels, $d_\mu^0 \equiv d_\mu$ at equilibrium. Further,

$$\rho_{12} = \rho_{21}^* \quad (2.4)$$

and

$$\Gamma_{12} = \Gamma_{21} , \quad (2.5)$$

to this order. Also,

$$\rho_{11} + \rho_{22} = 1 , \quad (2.6)$$

because of probability conservation. We readily find from the above,

$$\rho_{22,\mu} = \tfrac{1}{2}(1 + d_\mu) \ \text{ and } \ \rho_{11,\mu} = \tfrac{1}{2}(1 - d_\mu) . \quad (2.7)$$

Let us now briefly introduce the rate approximation. We assume that the atom follows the field motion adiabatically and write (2.1) in the form

$$\dot{\rho}_{12,\mu} = i\omega_{12}\rho_{12,\mu} - \frac{1}{T_{2,\mu}}\rho_{12,\mu} - i\sum_{i} g_{\lambda,\mu} b_\lambda^* d_\mu , \quad (2.8)$$

with $T_{2,\mu}^{-1} \equiv \Gamma_{12,\mu}$. We will consider the case[13]

$$T_2^{-1} \gg T_1^{-1} . \quad (2.9)$$

The atom is then in a quasi-steady-state near threshold where the correlations decay rapidly compared to the probabilities. Hence, $\dot{\rho}_{12} \approx 0$ and we may set $\rho_{12} \equiv \rho_{12}^s$, its steady-state value in the Eqs. (2.2)–(3). From (2.1) we find

$$\rho_{12,\mu}^s = \frac{i\sum_\lambda g_{\lambda,\mu} b_\lambda^* d_\mu}{i\omega_\mu - T_2^{-1}} = \rho_{21,\mu}^{s*} . \quad (2.10)$$

Substituting (2.10) into (2.2–3) we find the following rate equations:

$$\dot{d}_\mu = \frac{d_{0,\mu} - d_\mu}{T_1} - \frac{4\sum_\lambda T_2^{-1} |g_{\lambda\mu}|^2 n_\lambda d_\mu}{(\Omega_\lambda - \omega_\mu)^2 + T_2^{-2}} , \quad (2.11)$$

$$\dot{n}_\lambda = -2\kappa_\lambda n_\lambda + \frac{2\sum_\mu \Gamma |g_{\lambda\mu}|^2 n_\lambda d_\mu}{(\Omega_\lambda - \omega_\mu)^2 + T_2^{-2}} , \quad (2.12)$$

where we set $n_\lambda = b_\lambda^* b_\lambda$ and assume that

$$b_\lambda \sim B_\lambda \exp(i\Omega_\lambda t) , \quad (2.13)$$

because of the lasing process.

III. LYAPOUNOV FUNCTION FOR THE NONEQUILIBRIUM STEADY STATES

We now define a functional,[1,2]

$$\Omega(t) = \tfrac{1}{2}\,\mathrm{Tr}\,\rho^\dagger(t)\rho(t) \quad (3.1)$$

and

$$\Omega_T = \Omega_S + \Omega_F = \tfrac{1}{2}(\mathrm{Tr}\,|\rho_S|^2 + \mathrm{Tr}\,|\rho_F|^2) , \quad (3.2)$$

the subscripts S and F referring to the system and cavity field respectively. It must be noted here that our Eqs. (2.1)–(2.3) are weakly coupled, so that the density operator ρ appearing in (3.1) is the special case of the more general $\rho^{(p)}$ defined earlier, corresponding to $\Lambda = 1$ and satisfying the Pauli equation. It is apparent from (3.2) that

$$\Omega_T > 0 , \quad (3.3)$$

and

$$\dot{\Omega}_T = \tfrac{1}{2}\,\mathrm{Tr}(\dot{\rho}_S^\dagger \rho_S + \rho_S^\dagger \dot{\rho}_S) + \tfrac{1}{2}\,\mathrm{Tr}(\dot{\rho}_F^\dagger \rho_F + \rho_F^\dagger \dot{\rho}_F) . \quad (3.4)$$

For the multimode case, we have

$$\Omega_S = \tfrac{1}{2}\sum_{\mu\mu'} d_\mu d_{\mu'} \quad (3.5)$$

and

$$\dot{\Omega}_S = \sum_{\mu\mu'} d_\mu \dot{d}_{\mu'} . \quad (3.6)$$

We then obtain, using (2.11),

$$\dot{\Omega}_S = \sum_{\mu\mu'} d_\mu \left(\frac{d_{0,\mu'} - d_{\mu'}}{T_1} \sum_\lambda \frac{4T_2^{-2} |g_{\lambda\mu}|^2 n_\lambda d_{\mu'}}{(\Omega_\lambda - \omega_\mu)^2 + T_2^{-2}} \right) . \quad (3.7)$$

We also find

$$\Omega_F = \tfrac{1}{2}\sum_{\lambda\lambda'} n_\lambda n_{\lambda'} , \quad (3.8)$$

$$\dot{\Omega}_F = \sum_{\lambda\lambda'} n_\lambda \dot{n}_{\lambda'} , \quad (3.9)$$

so that we have from (2.12),

$$\dot{\Omega}_F = \sum_{\lambda\lambda'} n_\lambda n_{\lambda'} \left(-2\kappa_\lambda + \frac{\sum_\mu 2T_2^{-1} |g_{\lambda\mu}|^2 d_\mu}{(\Omega_{\lambda'} - \omega_\mu)^2 + T_2^{-2}} \right) . \quad (3.10)$$

Let us first consider the pumping branch near threshold. We then have[12]

$$d_{0,\mu} - d_\mu < 0\,, \tag{3.11}$$

$$\kappa_\lambda > \frac{T_2^{-1}|g_{\lambda\mu}|^2 d_\mu}{(\Omega_\lambda - \omega_\mu)^2 + T_2^{-2}}\,.$$

Hence we find

$$\dot{\Omega}_S < 0;\quad \dot{\Omega}_F < 0\,, \tag{3.12}$$

so that

$$\dot{\Omega}_T < 0\,. \tag{3.13}$$

The pumping branch is thus stable in the region of validity of the rate equations, in agreement with the results of a linear analysis,[14] Ω_T being a good Lyapounov function for this branch. It may be noted that near equilibrium, $d_0 \sim d$ and the conditions (3.11) reduce to the stability conditions derived elsewhere[14,15] by a linear analysis.

To consider the lasing branches, we recall that in a cw laser, the lasing states may be regarded as a set of multiple nonequilibrium steady states above threshold which persist as long as the cavity is loaded. Using the steady-state solutions to Eqs. (2.1)–(2.3), we readily find

$$\dot{\Omega}_T = 0\,, \tag{3.14}$$

in the lasing states. Hence we finally have

$$\Omega_T > 0 \text{ and } \dot{\Omega}_T \leq 0\,, \tag{3.15}$$

so that Ω_T is a Lyapounov function for both pumping and lasing branches near equilibrium. It should be noted that the inequalities (3.15) could also be obtained by linearizing Ω about the steady state and using a linearized form of the semiclassical Eqs. (2.1)–(2.3). However, this would only demonstrate stability in a very small neighborhood of threshold, whereas the rate approximation has a far greater range of validity. Our results then are more general than the corresponding ones obtained through linearization.

Finally, we may construct an N-body entropy for the nonequilibrium branch by setting[1]

$$S = -\tfrac{1}{2}\ln\Omega_S - \tfrac{1}{2}\ln\Omega_F\,. \tag{3.16}$$

Differentiating with respect to time yields

$$\dot{S} = -(\dot{\Omega}_S/\dot{\Omega}_S + \Omega_F/\Omega_F)\,. \tag{3.17}$$

From (3.12) we find

$$\dot{S} \geq 0\,. \tag{3.18}$$

Thus, the functional S has the properties of an entropy near a steady state far from thermal equilibrium.

IV. THE EQUILIBRIUM BRANCH–ENTROPY PRODUCTION AND THERMODYNAMICS

The results of Sec. III may be applied to the equilibrium branch—using the rate equations one may immediately write down a Lyapounov function of the form (3.1) which satisfies the inequalities (3.15). Let us now consider the thermodynamic description of this branch, close to equilibrium. To do so, we reconsider the semiclassical Eqs. (2.1)–(2.3) and linearize them about equilibrium by setting

$$\rho_{12} = \rho_{12}^e + \epsilon\phi_{12}\,, \tag{4.1}$$

$$b_\lambda = b_\lambda^e + \epsilon b_{\lambda 1}\,, \tag{4.2}$$

$$d_\mu = d_{0,\mu} + \epsilon d_{1,\mu}\,. \tag{4.3}$$

Substituting these in Eqs. (2.1)–(2.3) and retaining order (ϵ) terms only, we arrive at the linearized equations

$$\dot{\phi}_{12,\mu} = (i\omega_\mu - T_2^{-1})\phi_{12,\mu} - i\sum_\lambda g_\lambda b_{\lambda 1}^* d_{0,\mu}\,, \tag{4.4}$$

$$\dot{b}_{\lambda 1}^* = (i\omega_\lambda - \kappa_\lambda)b_{\lambda 1}^* + ig_\lambda^*\phi_{12,\mu}\,, \tag{4.5}$$

$$\dot{d}_{1,\mu} = -d_{1,\mu}/T_1\,, \tag{4.6}$$

where we recall that the atoms are assumed identical and $\rho_{12}^e = 0 = b_\lambda^e$. These equations are microscopic in nature. We now introduce the quantities

$$\rho_{12} = \tilde{\rho}_{12}\, e^{i\omega_{12}t} \text{ and } b_\lambda = \tilde{b}_\lambda\, e^{-i\omega_\lambda t}\,, \tag{4.7}$$

which we substitute into the linearized Eqs. (4.4)–(4.6). Let us consider, for instance, Eq. (4.4) in detail. We find, using (4.7),

$$\dot{\phi}_{12}(t) = -T_2^{-1}\phi_{12} - id_0\sum_\lambda g_\lambda b_{\lambda 1}^*\, e^{i(\omega_\lambda - \omega)t}\,, \tag{4.8}$$

where we have dropped the tilde and the index μ (recalling that all the atoms are identical), setting $\omega_{12} \equiv \omega$. We now time average (coarse grain) both sides of (4.8) using the prescription[16]

$$\langle A(t)\rangle_T \equiv \frac{1}{2T}\int_{-T}^{T} A(t)\,dt\,. \tag{4.9}$$

We then find

$$\langle\dot{\phi}_{12}\rangle_T = -T_2^{-1}\langle\phi_{12}\rangle_T - id_0\frac{1}{2T}\int_{-T}^{T}\sum_\lambda g_\lambda b_{\lambda 1}^*\, e^{i(\omega_\lambda - \omega)t}\,. \tag{4.10}$$

We will assume further that $b_{\lambda 1}(t)$ is slowly varying compared to the oscillatory terms when $\omega_\lambda \neq \omega$, so that it may be taken outside the integral. Also, since we are near equilibrium, we expect $\langle A\rangle_T$ to be independent of the initial time so that we may let $T \to \infty$ in the second term on the right-hand side of (4.10). This is an extension of Birkoff's theorem which states[17] that

$$\langle A(t)\rangle_T = \langle A(t)\rangle_\infty , \tag{4.11}$$

provided the phase space is metrically indecomposable. Assuming that the laser modes in the cavity form a continuum, we may write

$$\sum_\lambda \to \int_0^\infty d\omega_\lambda D(\omega_\lambda), \tag{4.12}$$

$D(\omega_\lambda)$ being the density of states. Substituting (4.12) into (4.10) and interchanging the order of integration, we arrive at

$$\langle \dot\phi_{12}\rangle = -T_2^{-1}\langle\phi_{12}\rangle - igd_0 D(\omega)\langle b_1^*\rangle . \tag{4.13}$$

Similarly, we obtain for Eqs. (4.5) and (4.6),

$$\langle \dot b_1^*\rangle = -\kappa\langle b_1^*\rangle + ig^*\langle\phi_{12}\rangle , \tag{4.14}$$

$$\langle d_1\rangle = -\langle d_1\rangle / T_1 . \tag{4.15}$$

We stress here that the Eqs. (4.13)–(4.15) involve macroscopic quantities so that we may use them to introduce a thermodynamic description of the system. Further, because of our time-smoothing procedure, the index λ no longer appears—Eqs. (4.13)–(4.15) effectively describe a monomode laser.

We now introduce the generalized entropy as in the previous section,

$$S_S = -\tfrac{1}{2}\ln\Omega_S . \tag{4.16}$$

Close to equilibrium we have[3] (apart from positive multiplicative constants),

$$\dot S_S = -\dot\Omega_S . \tag{4.17}$$

To order ϵ^2 we have

$$\dot S_S = -(\langle\phi_{12}\rangle\langle\dot\phi_{21}\rangle + \langle\dot\phi_{12}\rangle\langle\phi_{21}\rangle + \tfrac{1}{2}\langle d_1\rangle\langle\dot d_1\rangle) . \tag{4.18}$$

Using the linearized Eqs. (4.13)–(4.15), we find

$$\dot S_S = 2T_2^{-1}|\langle\phi_{12}\rangle|^2 + id_0 D(\omega)(g\langle b_1^*\rangle\langle\phi_{21}\rangle - \text{c.c.}) + \langle d_1\rangle^2/2T_1 . \tag{4.19}$$

For the radiation field we find

$$\dot S_F = -\langle b_1\rangle\langle b_1^*\rangle(\langle b_1\rangle\langle b_1^*\rangle) . \tag{4.20}$$

However, the leading term in (4.20) is of order ϵ^4 and does not contribute near equilibrium. Hence, to order ϵ^2, we may write

$$\dot S_S \equiv \sigma_S , \tag{4.21}$$

σ_S being the entropy production, which we may cast in the form

$$\sigma_S = 2T_2^{-1}|\langle\phi_{21}\rangle - (ig^*d_0/T_2^{-1})D(\omega)\langle b_1\rangle|^2 + \langle d_1\rangle^2/2T_1 - g^2 d_0^2[D(\omega)]^2|\langle b_1\rangle|^2 . \tag{4.22}$$

It has been shown[14] that

$$\sigma_S \geq 0 , \tag{4.23}$$

the equality holding at equilibrium. Evidently, the entropy production plays the role of a Lyapounov function near equilibrium.

Let us now define "generalized forces,"

$$X_1 = \langle\phi_{12}\rangle = X_2^*,\quad X_3 = \langle b_1\rangle = X_4^*,\quad X_5 = \langle d_1\rangle . \tag{4.24}$$

Then we have,[18]

$$\sigma_S = \sum_i J_i X_i = \sum_{i,k} L_{ik}X_iX_k . \tag{4.25}$$

The J_i's are the generalized "fluxes" corresponding to the forces X_i, and L is the matrix of Onsager coefficients. We find, from (4.22),

$$J_1 = T_2^{-1}\langle\phi_{21}\rangle - id_0 g^* D(\omega)\langle b_1\rangle = J_2^* , \tag{4.26}$$

$$J_3 = -id_0 g^* D(\omega)\langle\phi_{12}\rangle = J_4^* , \tag{4.27}$$

$$J_5 = \langle d_1\rangle / T_1 . \tag{4.28}$$

Also,

$$L = \begin{bmatrix} 0 & T_2^{-1} & -id_0 g^* D(\omega) & 0 & 0 \\ T_2^{-1} & 0 & 0 & id_0 g D(\omega) & 0 \\ -id_0 g^* D(\omega) & 0 & 0 & 0 & 0 \\ 0 & id_0 g D(\omega) & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1/2T_1 \end{bmatrix} . \tag{4.29}$$

We see that

$$L_{ii} \geq 0 ,$$

and

$$L_{ik} = L_{ki} \quad \text{for every } i \neq k . \tag{4.30}$$

These conditions are sufficient[11] for the near-equilibrium branch to be stable and have a minimum entropy production associated with it.

V. CONCLUSION

We have demonstrated in this paper how the N-body Lyapounov function (3.1) may be used to prove the stability of the cw laser, both near and far from equilibrium, it being possible to define a generalized entropy in each case. For the near-equilibrium branch, the entropy production σ has been shown to be a minimum, consistent with the results of linear irreversible thermodynamics. σ is a Lyapounov function for this branch. It should be stressed here that the results of Sec. IV hold only close to equilibrium (order ϵ^2). To higher orders in ϵ, the contributions from the cavity field must be considered and (4.21) would have to contain terms in $\mathring{S}_F$. Little is known about the entropy production of this system, or for that matter of any open system far from equilibrium. This will be the subject of continuing investigation.

A final remark is in order. The Eqs. (4.26)–(4.28) indicate that we have as many flows as there are levels of description in the system even though the forces (4.24) are macroscopic. This was first discussed by Klein and Meijer,[19] who showed that the minimum entropy theorem held in a microscopic description wherein the entropy production was considered a function of the microscopic probabilities (and correlations) and minimized with respect to these parameters. A general proof has been given by Callen,[20] and our results bear this out.

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[1]I. Prigogine, C. George, F. Henin, and L. Rosenfeld, Chem. Scr. 4, 5 (1973).

[2]W. C. Schieve, Int. J. Quantum Chem. Symp. 10, 375 (1976).

[3]A. R. Bulsara and W. C. Schieve, J. Chem. Phys. 65, 2532 (1976); Int. J. Quantum Chem. S9, 475 (1975).

[4]D. Hubert, J. Stat. Phys. 15, 1 (1976).

[5]F. Henin, Physica (Utr.) 76, 201 (1974); Bull. Cl. Sci. Acad. R. Belg. 60, 721 (1974).

[6]R. McAdory and W. C. Schieve, J. Chem. Phys. 67, 1899 (1977).

[7]M. Sargent, M. Scully, and W. Lamb, *Laser Physics* (Addison-Wesley, Reading, Mass., 1974).

[8]E. Hofelich-Abate and F. Hofelich, Z. Phys. 211, 142 (1968).

[9]The semiclassical theory has further been used by Graham R. Graham in *Fluctuations, Instabilities and Phase Transitions*, edited by T. Riste (Plenum, New York, 1976) in his treatment of the laser threshold as a second-order phase transition using the Landau theory.

[10]D. Walgraef, J. Stat. Phys. 14, 399 (1976).

[11]P. Glansdorff and I. Prigogine, *Structure, Stability and Fluctuations* (Wiley, New York, 1971).

[12]H. Haken, *Handbuch der Physik*, Vol. 25/2C, edited by S. Flugge (Springer-Verlag, Berlin, 1970).

[13]C. Tang, J. Appl. Phys. 34, 2935 (1963).

[14]A. R. Bulsara and W. C. Schieve, in Proceedings of the Fourth Rochester Conference on Coherence and Quantum Optics, 1977 (unpublished).

[15]H. Risken and K. Nummedal, J. Appl. Phys. 39, 4662 (1968).

[16]J. G. Kirkwood, J. Chem. Phys. 14, 180 (1946); 15, 72 (1947).

[17]I. E. Farquhar, *Ergodic Theory in Statistical Mechanics* (Wiley, New York, 1964).

[18]S. deGroot and P. Mazur, *Non-Equilibrium Thermodynamics* (North-Holland, Amsterdam, 1969).

[19]M. Klein and P. Meijer, Phys. Rev. 96, 250 (1954); P. Meijer, in *Transport Processes in Statistical Mechanics* (IUPAP, Brussels, 1956).

[20]H. Callen, in Ref. 19.