Spontaneous Emission in Semiclassical Radiation Theory

R. K. Nesbet
IBM Research Laboratory, Monterey and Cottle Roads, San Jose, California 95114
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It has recently been shown by Jaynes and collaborators that semiclassical radiation theory contains a description of spontaneous emission of radiation and of radiative level shifts. The present paper gives a gauge-invariant derivation of the radiant energy production rate, using only Maxwell's equations and the usual definitions of electric current and charge density for a many-particle material system described by Schrödinger's equation. A complementary derivation, using the time-dependent Schrödinger equation, verifies the instantaneous conservation of energy and probability during the radiation process. The semiclassical rate of spontaneous emission differs from the usual formula (Einstein's A coefficient) because it depends on the occupancy of both initial and final states of the material system. The implications of this with regard to thermal equilibrium and Planck's law are examined. If a new hypothesis is introduced, postulating the decomposition of the equilibrium radiation into incoherent components, each interacting with a specific pair of energy levels of the material system, then Planck's law is shown to hold for the total intensity of radiation. If this hypothesis is not introduced, the equilibrium conditions for different transitions are incompatible, and the semiclassical radiation theory is incapable of describing thermal equilibrium.

I. INTRODUCTION

In semiclassical radiation theory, the electromagnetic field is treated as a classical field, governed by Maxwell's equations, interacting with a material system governed by Schrödinger's equation. The Schrödinger wave function can be considered to be a classical field. The structure of the theory is essentially that of a classical field theory, in which the field equations couple the Maxwell field to the Schrödinger field.

It is commonly assumed that semiclassical radiation theory cannot describe the phenomenon of spontaneous emission—radiation from a material system that is not induced by an incident electromagnetic field. In contrast, stimulated emission and absorption of radiation are described correctly by the theory.

In a recent series of papers,[1] Jaynes and collaborators have reopened the question of the limit of validity of semiclassical theory. They have shown that the coupled semiclassical field equations imply not only spontaneous emission, but also radiative energy-level shifts. Both of these phenomena appear as dynamical effects depending on the initial state of the material system in a way that differs from the prediction of quantum electrodynamics. Stroud and Jaynes[1] point out that definitive experiments testing these dynamical effects have not yet been carried out. They conclude that semiclassical radiation theory cannot yet be ruled out on existing experimental evidence.

If a quantum mechanical system is in a pure stationary state of energy E, the time-dependent phase factor $e^{Et/i\hbar}$ cancels out of all quantities determined by the density function $\psi\psi^*$. Hence physical properties of a stationary state are time independent and do not act as source terms for a radiation field. However, a mixed state has time-dependent cross terms $e^{(E_\beta - E_\alpha)t/i\hbar}$ in $\psi\psi^*$, which provides oscillatory source charge and current densities. Through Maxwell's equations, these source terms produce a radiation field, describing spontaneous emission from any material system in a mixed state. Emission of radiant energy continues until only the lowest initial energy level is populated, reducing the oscillatory cross terms to zero.

One purpose of the present paper is to give a detailed formal derivation of the general result described qualitatively in the preceding paragraph. This derivation, given in Sec. II, is based entirely on Maxwell's equations, leading to a general formula for the rate of radiant energy production by an oscillatory electric current source density. This provides a generalization, as free as possible of simplifying assumptions, of arguments given by Jaynes and co-workers.[1] Incidentally, it provides a complete theory of radiation reaction, since the full dynamics of the coupled classical Maxwell and Schrödinger fields is taken into account.

For completeness, the spontaneous emission rate is derived from the dynamics of the Schrödinger equation in Sec. III, using the usual formalism of time-dependent (Dirac) perturbation theory. This derivation is similar to that given by Crisp and Jaynes.[1] The rate of energy loss by the Schrödinger field is shown to be equal to the rate of energy increase in the Maxwell field derived in Sec. II.

The rate of spontaneous emission derived from semiclassical theory differs from that assumed by Einstein in his classical derivation of Planck's

radiation law.[2] This issue, which was not treated in detail by Jaynes and co-workers, is considered in Sec. IV. The essential result is either that thermal equilibrium is not possible in the semiclassical theory, or that a new hypothesis must be made, which is proposed here as a way of reconciling the theory with physical reality. This hypothesis restores Planck's law, but requires a novel interpretation of the nature of equilibrium thermal radiation.

II. ENERGY PRODUCTION IN MAXWELL FIELD

Any vector field $\vec{v}$ can be expressed as a sum of transverse and longitudinal component fields,

$$\vec{v} = \vec{v}^{tr} + \vec{v}^{long}, \tag{1}$$

where

$$\mathrm{div}\vec{v}^{tr} = 0, \quad \mathrm{curl}\vec{v}^{long} = 0 . \tag{2}$$

This decomposition is unique if boundary conditions are specified for $\vec{v}^{long}$.

When separated into transverse and longitudinal components, Maxwell's equations, valid at the subatomic level of detail, become

$$\begin{aligned} &\mathrm{div}\vec{E}^{long} = 4\pi\rho , \\ &\frac{1}{c}\frac{\partial}{\partial t}\vec{E}^{long} = -\frac{4\pi}{c}\vec{j}^{long}, \\ &\mathrm{curl}\vec{E}^{tr} = -\frac{1}{c}\frac{\partial}{\partial t}\vec{B}^{tr}, \\ &\mathrm{curl}\vec{B}^{tr} = \frac{1}{c}\frac{\partial}{\partial t}\vec{E}^{tr} + \frac{4\pi}{c}\vec{j}^{tr} . \end{aligned} \tag{3}$$

If $\vec{E}^{long}$ is required to vanish at points remote from the source charge density ρ and current density $\vec{j}$, the equation for $\partial\vec{E}^{long}/\partial t$ follows from the first equation here and from the equation of continuity

$$\mathrm{div}\vec{j} + \frac{\partial}{\partial t}\rho = 0 . \tag{4}$$

This boundary condition implies that

$$\vec{E}^{long} = -\mathrm{grad}\phi, \tag{5}$$

where

$$\phi(P) = \int R_{PQ}^{-1}\,\rho(Q)\,d\tau_Q . \tag{6}$$

A radiation field is defined physically as one that can remain finite at points remote from the field's sources. Because $\vec{E}^{long}$ vanishes in this limit, the electromagnetic radiation field consists solely of $\vec{E}^{tr}$ and $\vec{B}^{tr}$. Equations (3) provide a gauge-invariant definition of the radiation field. If potential functions are introduced, it is convenient to use the Coulomb gauge condition

$$\mathrm{div}\vec{A} = 0 , \tag{7}$$

so that the vector potential is purely transverse. Then the electrostatic potential ϕ is defined by Eq. (6). The radiation field intensities are

$$\vec{B}^{tr} = \mathrm{curl}\vec{A}^{tr}, \quad \vec{E}^{tr} = -\frac{1}{c}\frac{\partial}{\partial t}\vec{A}^{tr}. \tag{8}$$

From Eqs. (3), the Cartesian components of $\vec{A}^{tr}$ satisfy the inhomogeneous wave equation

$$\left\{\nabla^2 - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right\}A^{tr} = -\frac{4\pi}{c}\vec{j}^{tr} . \tag{9}$$

Because the last two of Eqs. (3), for the transverse field components, are of the same form as the original Maxwell equations, an energy conservation law of the usual form holds for the radiation field $\{\vec{E}^{tr}, \vec{B}^{tr}\}$ independently of $\vec{E}^{long}$ and of its source densities ρ and $\vec{j}^{long}$. This conservation law for the radiation field is

$$\frac{\partial}{\partial t}u + \mathrm{div}\vec{\pi} = -\vec{E}^{tr}\cdot\vec{j}^{tr}, \tag{10}$$

where the energy density is

$$u = (1/8\pi)(\vec{B}^{tr}\cdot\vec{B}^{tr} + \vec{E}^{tr}\cdot\vec{E}^{tr}) , \tag{11}$$

and the Poynting vector or energy flux is

$$\vec{\pi} = (c/4\pi)(\vec{E}^{tr}\times\vec{B}^{tr}) . \tag{12}$$

The right-hand member of Eq. (10) gives the rate of energy transfer from a material system whose transverse electric current density is $\vec{j}^{tr}$. If the material system is an atom or molecule, it can be enclosed in a volume of finite extent. The total rate of radiative energy emission is

$$dW/dt = -\int \vec{E}^{tr}\cdot\vec{j}^{tr}\,d\tau, \tag{13}$$

integrated over all space.

From Eqs. (8) and (9),

$$\left\{\nabla^2 - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right\}\vec{E}^{tr} = \frac{4\pi}{c^2}\frac{\partial}{\partial t}\vec{j}^{tr} . \tag{14}$$

A particular solution of this equation is

$$\vec{E}^{tr}(P) = -\frac{1}{c^2}\int R_{PQ}^{-1}\left[\frac{\partial}{\partial t}\vec{j}^{tr}(Q)\right]_{ret} d\tau_Q, \tag{15}$$

where the source term is evaluated at a retarded time interval

$$\Delta t = -R/c \tag{16}$$

prior to the time coordinate of the field point P. In the absence of an incident radiation field, Eq. (15) describes the field produced spontaneously by any time-varying transverse electric current density distribution. If this field is substituted into Eq. (13), it gives a general formula for the classical rate of emission of radiant energy,

$$\frac{dW}{dt} = \frac{1}{c^2}\iint \vec{j}^{tr}(P,t)\cdot R^{-1}\frac{\partial}{\partial t}\vec{j}^{tr}\left(Q, t - \frac{R}{c}\right) d\tau_P\, d\tau_Q . \tag{17}$$

A time-varying real current density can be expanded in Fourier components,

$$\vec{j}^{tr} = \sum_{\pm\omega}\vec{j}_\omega^{tr} e^{-i\omega t}, \tag{18}$$

where

$$\vec{j}_{-\omega} = \vec{j}_{\omega}^{*}. \tag{19}$$

If this expansion is substituted into Eq. (17), the time-averaged emission rate, obtained by retaining only nonoscillatory terms, is

$$\frac{dW}{dt} = \sum_{\omega} \frac{1}{c^2} \iint [i\omega \vec{j}_{\omega}^{\,tr}(P) \cdot \vec{j}_{-\omega}^{\,tr}(Q) e^{-i\omega R/c} - i\omega \vec{j}_{-\omega}^{\,tr}(P) \cdot \vec{j}_{\omega}^{\,tr}(Q) e^{i\omega R/c}] R^{-1} d\tau_P d\tau_Q \tag{20}$$

$$= \sum_{\omega} \frac{2\omega^2}{c^3} \iint \mathrm{Re}[\vec{j}_{\omega}^{\,tr}(P) \cdot \vec{j}_{-\omega}^{\,tr}(Q)] \times \left(\frac{\sin(\omega R/c)}{\omega R/c}\right) d\tau_P d\tau_Q. \tag{21}$$

This expression involves no approximations other than time averaging to remove high-frequency components.

For frequencies of spectroscopic interest, $\omega R/c$ is small if R is confined to the spatial extent of an atom or small molecule. The expansion

$$\frac{\sin(\omega R/c)}{\omega R/c} = 1 - \frac{1}{6} \frac{\omega^2 R^2}{c^2} + \cdots \tag{22}$$

separates Eq. (21) into a sum of multipole-multipole interactions.

The exact Schrödinger wave function of a material system can be expanded in terms of the stationary-state eigenfunctions ϕ_α in the form

$$\psi(t) = \sum_{\alpha} c_\alpha \phi_\alpha e^{E_\alpha t/i\hbar}, \tag{23}$$

where

$$H_0 \phi_\alpha = E_\alpha \phi_\alpha \tag{24}$$

for the unperturbed Hamiltonian operator H_0 of the isolated material system. Unless ψ is a pure stationary state, the charge density ρ and electric current density $\vec{j}$ contain time-dependent terms that arise from cross terms in the density matrix $\psi\psi^*$ of the form

$$c_\alpha^* c_\beta \phi_\alpha^* \phi_\beta e^{(E_\beta - E_\alpha)t/i\hbar}. \tag{25}$$

Consider two stationary states 1 and 2, with $E_2 > E_1$. Then for $\hbar\omega_{12} = E_2 - E_1$, $\vec{j}_{\omega}^{\,tr}$ in Eq. (21) is the transverse component of the vector field

$$\vec{j}_{\omega} = c_1^* c_2 \vec{j}_{12}, \tag{26}$$

where $\vec{j}_{12}$ is the transition current density. Explicitly, at a point P

$$\vec{j}_{12}(P) = \tfrac{1}{2} \sum_a \delta(a, P) \int \cdots \int [\phi_1^* \vec{j}_{op} \phi_2 + (\vec{j}_{op} \phi_1)^* \phi_2] \times [d\tau_1 \cdots d\tau_N]_a', \tag{27}$$

where this notation indicates integration over the coordinates of all but one (the ath) particle (electron or nucleus) followed by summation over all particles.

The current density operator for a single electron (charge $-e$, mass m) is

$$-\frac{e}{m}\left(\vec{p} + \frac{e}{c}\vec{A}\right). \tag{28}$$

There is an additional term in the physical current density of the form $c\,\mathrm{curl}\vec{m}$, where $\vec{m}$ is the magnetization due to electron spin. This magnetization current, which contributes to magnetic dipole radiation, will not be considered here. For a many-particle system $\vec{j}_{op}$ is a sum of terms of the form of Eq. (28), with charge and mass appropriate to each particle. The terms in $\vec{j}_{op}$ that contain the vector potential $\vec{A}$ can be neglected when matrix elements of the remaining operator do not vanish.

If only the constant term is retained in Eq. (22), for a transition between two stationary states Eq. (21) reduces to

$$\frac{dW}{dt} = \frac{2\omega_{12}^2}{c^2} \int \vec{j}_{12}^{\,tr} d\tau \cdot \int \vec{j}_{21}^{\,tr} d\tau \, |c_1|^2 |c_2|^2. \tag{29}$$

This can be expressed in terms of matrix elements of the electric dipole moment operator $\vec{\mu}_{op}$, such that

$$\int \vec{j}_{12} d\tau = (i/\hbar)(1 | [H_0, \vec{\mu}_{op}] | 2) \tag{30}$$

$$= -i\omega_{12} \vec{\mu}_{12}, \tag{31}$$

where

$$\vec{\mu}_{12} = (1 | \vec{\mu}_{op} | 2) \tag{32}$$

is the transition dipole moment. Then Eq. (29) becomes

$$\frac{dW}{dt} = \frac{2\omega_{12}^4}{c^3} \vec{\mu}_{12}^{\,tr} \cdot \vec{\mu}_{21}^{\,tr} |c_1|^2 |c_2|^2. \tag{33}$$

The transverse component of $\vec{\mu}_{12}$ is of magnitude $\mu_{12} \sin\theta$, where θ is the angle between $\vec{\mu}_{12}$ and an assumed direction of emitted radiation. If Eq. (33) is averaged over all directions of emission or, equivalently, averaged over degenerate substates of the initial and final energy levels,

$$\frac{dW}{dt} = \frac{2\omega_{12}^4}{c^3} \mu_{12}^2 \langle \sin^2\theta \rangle |c_1|^2 |c_2|^2 \tag{34}$$

$$= \frac{4\omega_{12}^4}{3c^3} \mu_{12}^2 |c_1|^2 |c_2|^2. \tag{35}$$

The implied rate of transition from state 2 to state 1 is

$$w_{2\to1} = \frac{1}{\hbar\omega_{12}} \frac{dW}{dt} = \frac{4\omega_{12}^3}{3\hbar c^3} \mu_{12}^2 |c_1|^2 |c_2|^2. \tag{36}$$

When $|c_1|^2 \cong 1$, this agrees with the rate of spontaneous emission given by the Einstein A coefficient,

$$w_{2\to1}^{spont} = A(2, 1) |c_2|^2, \tag{37}$$

with

$$A(2, 1) = (4\omega_{12}^3 / 3\hbar c^3) \mu_{12}^2, \tag{38}$$

if states 1 and 2 are considered to be nondegenerate. However, the factor $|c_1|^2$ in Eq. (36) is not present in the usual formula, Eq. (37), derived from quantum electrodynamics.

III. ENERGY LOSS BY SCHRÖDINGER FIELD

In the presence of an electromagnetic radiation field the Hamiltonian H_0 of the unperturbed material system is augmented by a time-dependent perturbing operator

$$H_1 = \sum_{\pm\omega} V_\omega e^{-i\omega t} , \tag{39}$$

where

$$V_{-\omega} = V_\omega^* \tag{40}$$

and, neglecting terms quadratic in the vector potential,

$$V_\omega = -\frac{1}{c}\sum_a \vec{A}_\omega^{tr}(a)\cdot\vec{j}_{op}^{tr}(a) . \tag{41}$$

The summation is over all particles of the material system. Under the influence of this perturbation, the coefficients c_α in Eq. (23) become time dependent. From the time-dependent Schrödinger equation,

$$i\hbar\dot{c}_\alpha(t) = \sum_{\pm\omega}\sum_\beta (\alpha|V_\omega|\beta)c_\beta(t)e^{(E_\beta - E_\alpha + \hbar\omega)t/i\hbar} . \tag{42}$$

In the absence of an incident field, $\vec{A}_\omega$ is determined entirely by the oscillatory current density of Eq. (26) through the inhomogeneous wave equation (9). This gives

$$\vec{A}^{tr}(P) = (1/c)\int R^{-1}[\vec{j}^{tr}(Q)]_{ret}\, d\tau_Q , \tag{43}$$

with notation as in Eq. (15). Equivalently,

$$\vec{A}_\omega^{tr}(P) = (1/c)\int R^{-1}e^{i\omega R/c}\vec{j}_\omega^{tr}(Q)\, d\tau_Q . \tag{44}$$

When substituted into Eq. (41), this defines the matrix elements $(\alpha|V_\omega|\beta)$ in Eq. (42).

To simplify the derivation, consider only two energy levels, $E_1 < E_2$, with $\hbar\omega = E_2 - E_1$. Then the time dependence of c_2, omitting high-frequency terms from Eq. (42), is given by

$$i\hbar\dot{c}_2(t) = (2|V_\omega|1)c_1(t) \tag{45}$$

$$= -\frac{1}{c}(2|\textstyle\sum_a \vec{A}_\omega^{tr}(a)\cdot\vec{j}_{op}^{tr}(a)|1)c_1(t) \tag{46}$$

$$= -\frac{1}{c}\int \vec{A}_\omega^{tr}(P)\circ\vec{j}_{21}^{tr}(P)\, d\tau_P\, c_1(t) \tag{47}$$

$$= -\frac{1}{c^2}\iint \vec{j}_{21}^{tr}(P)\cdot R^{-1}e^{i\omega R/c}\vec{j}_{12}^{tr}(Q)\, d\tau_P\, d\tau_Q \times c_1^*(t)c_1(t)c_2(t) , \tag{48}$$

where $\vec{j}_{12}$ and $\vec{j}_{21}$ are defined as in Eq. (27). From Eq. (48) and its complex conjugate,

$$\frac{d}{dt}|c_2|^2 = c_2^*\dot{c}_2 + \dot{c}_2^* c_2$$

$$= -\frac{2\omega}{\hbar c^3}\iint \mathrm{Re}[\vec{j}_{21}^{tr}(P)\cdot\vec{j}_{12}^{tr}(Q)] \times\left(\frac{\sin(\omega R/c)}{\omega R/c}\right) d\tau_P\, d\tau_Q |c_1|^2|c_2|^2 . \tag{49}$$

Similarly,

$$\frac{d}{dt}|c_1|^2 = \frac{2\omega}{\hbar c^3}\iint \mathrm{Re}[\vec{j}_{12}^{tr}(P)\cdot\vec{j}_{21}^{tr}(Q)] \times\left(\frac{\sin(\omega R/c)}{\omega R/c}\right) d\tau_P\, d\tau_Q |c_1|^2|c_2|^2 . \tag{50}$$

An immediate consequence of these equations is that normalization is preserved:

$$\frac{d}{dt}(|c_1|^2 + |c_2|^2) = 0 . \tag{51}$$

The rate of energy change is

$$\frac{dE}{dt} = \frac{d}{dt}(|c_1|^2 E_1 + |c_2|^2 E_2) \tag{52}$$

$$= \tfrac{1}{2}(E_2 - E_1)\frac{d}{dt}(|c_2|^2 - |c_1|^2) \tag{53}$$

$$= \hbar\omega_{12}\frac{d}{dt}|c_2|^2 = -\hbar\omega_{12}\frac{d}{dt}|c_1|^2 . \tag{54}$$

These equations are identical in form with Eq. (21), applied to a transition between only two energy levels. For dipole radiation they reduce to

$$\frac{dE}{dt} = -\frac{4\omega_{12}^2}{3c^3}\mu_{12}^2|c_1|^2|c_2|^2 , \tag{55}$$

in agreement with Eq. (35), derived from Maxwell's equations, so that

$$\frac{d}{dt}(W + E) = 0 . \tag{56}$$

Thus the total energy of material system plus radiation is conserved.

IV. THERMAL EQUILIBRIUM AND PLANCK'S LAW

In thermal equilibrium, the coefficients of degenerate substates can be assumed to be equal in magnitude. The statistical probability that a material system is in state 1 or 2, of degeneracy $d(1)$ or $d(2)$, respectively, is

$$P(1) = d(1)|c_1|^2, \quad P(2) = d(2)|c_2|^2 . \tag{57}$$

In equilibrium at temperature T, the ratio of probabilities is

$$P(2)/P(1) = [d(2)/d(1)]e^{-\hbar\omega_{12}/kT}, \tag{58}$$

where $\hbar\omega_{12} = E_2 - E_1$.

In Einstein's classical derivation of Planck's radiation law, the total rate of change of $P(1)$ due to transitions between energy levels E_1 and E_2 $(E_2 > E_1)$ is

$$\frac{d}{dt}P(1) = -P(1)w_{1\to 2}^{\text{ind}} + P(2)w_{2\to 1}^{\text{ind}} + P(2)w_{2\to 1}^{\text{spont}} . \quad (59)$$

The three rates of induced and spontaneous transition are

$$w_{1\to 2}^{\text{ind}} = B(1, 2)\rho(\omega_{12}), \quad (60)$$

$$w_{2\to 1}^{\text{ind}} = B(2, 1)\rho(\omega_{12}), \quad (61)$$

$$w_{2\to 1}^{\text{spont}} = A(2, 1), \quad (62)$$

where $\rho(\omega)d\omega$ is the energy density of electromagnetic radiation in the angular frequency interval $d\omega$. The Einstein transition coefficients, for electric dipole radiation, are

$$B(1, 2) = (4\pi^2/3\hbar^2)\mu_{12}^2 d(2), \quad (63)$$

$$B(2, 1) = (4\pi^2/3\hbar^2)\mu_{12}^2 d(1), \quad (64)$$

$$A(2, 1) = (4\omega_{12}^2/3\hbar c^3)\mu_{12}^2 d(1) . \quad (65)$$

For the radiation field to be in equilibrium with the material system, Eq. (59) must vanish when $P(2)/P(1)$ is given by Eq. (58). This determines an equilibrium radiation density,

$$\rho(\omega_{12}, T) = \rho_0(\omega_{12}, T), \quad (66)$$

where ρ_0 is Planck's radiation density,

$$\rho_0(\omega, T) = (\hbar\omega^3/\pi^2c^3)(e^{\hbar\omega/kT} - 1)^{-1} . \quad (67)$$

The Einstein B coefficients can be derived from semiclassical radiation theory or from quantum electrodynamics by well-known arguments. The coefficient A for spontaneous emission is derived in quantum electrodynamics as an immediate consequence of the quantization of the electromagnetic radiation field, verifying Eq. (62) for the spontaneous radiation rate, in agreement with Planck's law.

The present derivation leads to a spontaneous transition rate that differs from Eq. (62). When degeneracies are taken into account, Eqs. (49) and (50), in the case of electric dipole radiation, become

$$d(1)\frac{d}{dt}|c_1|^2 = -d(2)\frac{d}{dt}|c_2|^2 = \frac{4\omega_{12}^3}{3\hbar c^3}\mu_{12}^2|c_1|^2|c_2|^2 d(1)d(2) \quad (68)$$

or

$$\frac{d}{dt}P(1) = -\frac{d}{dt}P(2) = \frac{A(2, 1)P(1)P(2)}{d(1)} \quad (69)$$

where $A(2, 1)$ is given by the usual formula, Eq. (65). The spontaneous transition rate is

$$w_{2\to 1}^{\text{spont}} = A(2, 1)P(1)/d(1) . \quad (70)$$

This agrees with Eq. (62) only if the material system is in a nearly pure stationary state, with $|c_1|^2 \cong 1$.

With this modified expression for the spontaneous emission rate, the equilibirum requirement that Eq. (59) should vanish subject to Eq. (58) determines an equilibrium radiation density

$$\rho_1(\omega_{12}, T) = \rho_0(\omega_{12}, T)P(1)/d(1), \quad (71)$$

where ρ_0 is Planck's radiation density, Eq. (67).

Equation (71) indicates than an equilibrium radiation density must be associated with each energy level E_i and excitation frequency ω of a material system. Because all effects of matrix elements and selection rules have canceled out of this formula, it can be assumed to hold for any frequency ω, even if transitions between energy levels E_i and $E_i + \hbar\omega$ are immeasurably weak. This implies the general result

$$\rho_i(\omega, T) = \rho_0(\omega, T)P(i)/d(i) \quad (72)$$

for any E_i and ω.

At first sight, this result cannot be reconciled with the stated condition of thermal equilibrium, since *the* equilibrium radiation density should be uniquely defined, the same for all energy levels of any material system. If so, the semiclassical radiation theory must be abandoned at this point.

However, a new hypothesis can be introduced that is not in obvious contradiction with basic physical principles or with the postulates of semiclassical radiation theory. The hypothesis is that the total equilibrium radiation density is the sum of incoherent partial densities associated with each pair of energy levels, or, as in Eq. (72), with each energy level and excitation energy $\hbar\omega$. Then the total radiation density at frequency ω is obtained by summing Eq. (72) over all initial levels E_i, weighted by the degeneracy. The total radiation density is given by Planck's law,

$$\rho(\omega, T) = \sum_i d(i)\rho_i(\omega, T) = \rho_0(\omega, T), \quad (73)$$

since

$$\sum_i P(i) = 1 . \quad (74)$$

V. DISCUSSION

It has been shown here that semiclassical radiation theory contains a description of spontaneous emission that is part of a complete gauge-invariant theory of the radiation reaction. Since all Schrödinger wave functions describe electric source current distributions of finite spatial extent, only finite quantities occur in the theory.

In an attempt to apply this theory to derive the conditions of thermal equilibrium between matter and radiation an apparent inconsistency is encountered. An additional postulate is proposed to resolve this difficulty. The new hypothesis is that the equilibrium radiation field is composed of incoherent components, each of which is associated with a given pair of energy levels of the material system. This postulate replaces the usual pos-

tulate of phase incoherence of components of the Schrödinger wave function made in quantum statistical mechanics. Since the total radiation density obeys Planck's law, there is no conflict with experimental data on blackbody radiation in which total radiant energy is measured.

As has been shown by Jaynes and co-workers,[1] the semiclassical theory describes dynamical radiative level shifts as well as radiative lifetimes due to spontaneous emission. These two phenomena both follow from Eq. (48), when applied to the total effect of all levels E_β on a given level E_α. This equation becomes

$$i\hbar\dot{c}_\alpha(t) = (\Delta E_\alpha - \tfrac{1}{2}i\hbar\Gamma_\alpha)c_\alpha(t) \tag{75}$$

$$= \sum_\beta (\Delta E_{\alpha\beta} - \tfrac{1}{2}i\hbar\Gamma_{\alpha\beta})|c_\beta(t)|^2 c_\alpha(t), \tag{76}$$

where

$$\Delta E_{\alpha\beta} - \tfrac{1}{2}i\hbar\Gamma_{\alpha\beta} = -\frac{1}{c^2}\iint \vec{j}^{\,\mathrm{tr}}_{\alpha\beta}(P)\cdot R^{-1} e^{i\omega_{\alpha\beta}R/c}\vec{j}^{\,\mathrm{tr}}_{\beta\alpha}(Q)\,d\tau_P\,d\tau_Q\,. \tag{77}$$

This gives a general formula for the dynamical level shift $\sum_\beta \Delta E_{\alpha\beta}|c_\beta|^2$ and the dynamical width $\sum_\beta \Gamma_{\alpha\beta}|c_\beta|^2$. For two levels, the width formula is equivalent to Eq. (50) for the time rate of change of $|c_1|^2$.

Crisp and Jaynes[1] found that for electric dipole radiation the $1s$-$2p$ level shift obtained from Eq. (77) for nonrelativistic wave functions agrees within experimental error with the experimental value, assuming that $|c_{1s}|^2$ is unity. The $2s$-$2p$ level shift (the Lamb shift) is roughly two-thirds of the experimental value when computed with nonrelativistic wave functions, assuming that $|c_{2s}|^2$ is unity under the conditions of the Lamb-shift experiments. These calculations need to be refined before they could be considered to provide a definitive test of the semiclassical theory.

It is important to note that irreversibility is built into the theory of spontaneous emission given here by assuming retarded solutions of the inhomogeneous wave equation, in Eq. (15). In consequence, the rate of energy production given by Eq. (21) is positive definite, at least for electric dipole radiation. If advanced potentials had been used, the energy production rate would change sign, and it vanishes if half-advanced, half-retarded potentials are used. Thus the choice of retarded potentials establishes an "arrow of time," in that energy is dissipated from an excited material system as time progresses, and an eventually established equilibrium would not be disrupted by reversing the time sense.

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He II-He I Transition in a Heat Current: Model Calculations*

S. M. Bhagat and R. A. Lasken
University of Maryland, College Park, Maryland 20740

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The Ginzburg-Pitaevskii phenomenological theory has been extended to include the effects of vortex lines. The model calculations are compared with some of our recent experimental results on the He II–He I transition in the presence of a heat current.

I. INTRODUCTION

Several years ago Ginzburg and Pitaevskii[1] proposed a phenomenological theory of superfluidity near T_λ. Some of the results of this theory have been discussed in recent papers by Mamaladze,[2] Kramer,[3] and Mikeska[4] and in an unpublished report by Burkhardt and Stauffer.[5] A salient feature of the theories is the prediction that the superfluid density ρ_s should be reduced for finite values of the counter-flow velocity $w = (v_s - v_n)$. Concomitantly, this implies an instability in a driven superfluid current $\rho_s w$. In other words, for any given temperature there should exist a maximum current consistent with superfluidity. In addition, the theory predicts that, barring the instability, the superfluid density should vanish at high enough values of w, i.e., the He II–He I transition temperature should be lowered for finite w. To our knowledge, until recently there has been no unequivocal experimental evidence to support this view. In a recent paper[6] (hereafter referred to as I) we have reported an experiment in which the He II–He I transition was investigated in the presence of a heat current. Figure 1 [cf. Fig. 3(b) of I] shows the thermograms observed at two points along a thermally isolated