Quantum interferences and the question of thermodynamic equilibrium

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We derive from first principles the dynamical equations for the interaction between a heat bath and a multilevel atom with some near degenerate states. Such dynamical equations exhibit atomic coherence terms which arise from the interference of transition amplitudes. We address the question whether such equations lead to a steady state that is consistent with the thermodynamic equilibrium. We show that coherence affects the dynamics of the system, but the equilibrium conditions are still characterized by Boltzmann factors. We also show how an asymmetric treatment of spontaneous and stimulated processes could lead to a steady state which is at variance with the principles of thermodynamic equilibrium. We show that such a steady state can be realized by pumping with broadband laser fields. Finally, we show that coherences in the dynamical equations can be probed via the spectrum of fluorescence.

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I. INTRODUCTION

It is well known that quantum coherences can be produced by pumping a system with coherent fields-an outstanding example being the phenomenon of coherent population trapping [1]. It is also well understood how quantum coherence can be created in interactions involving a common bath with a set of closely lying states [2-12]. These types of coherences have led to very remarkable phenomena like lasing without population inversion [4], etc. One would like to understand the role of coherences if the bath is at a finite temperature. At the outset one would not expect any coherences if the system is in thermodynamic equilibrium as the density matrix has the form $\exp(-\beta H)$, which is clearly diagonal in a basis in which H is diagonal. However, a microscopic derivation of the master equation for a system interacting with a heat bath does show the appearance of coherence terms in dynamical equations. Clearly, one needs to demonstrate the consistency of the dynamical equation with thermodynamic equilibrium. This then raises a very interesting question: what could then be the observational consequence of such coherence terms in the master equation? The present paper deals with such aspects. We derive from first principles the dynamical equation, which exhibits coherences, and which we show to be consistent with thermodynamic equilibrium. We give several examples of physical quantities that can be used to study the effect of coherences in the dynamical equations.

The organization of the paper is as follows. In Sec. II we derive the basic equations of motion for our model and show the possibility of atomic coherence due to interaction with a bath. In Sec. III we show how *thermodynamic equilibrium is achieved* in a steady state even in the presence of such coherence terms in the master equation. In Sec. IV we show how the coherence terms in the master equation can be probed through the emission spectrum. We also demonstrate how an *asymmetric* treatment of spontaneous vs stimulated emission can lead to a steady state which is at variance with thermodynamic equilibrium. In Sec. V we demonstrate how

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such situations can be realized by pumping with a broadband laser.

II. EQUATIONS OF MOTION

We consider a collection of three-level atoms, the excited levels $|1\rangle$, $|2\rangle$, and ground level $|3\rangle$ (V system) in a bath of thermal field (Fig. 1). The Hamiltonian for this system will be

$$H = H_0 + H_{AR}, \tag{1}$$

where

$$H_0 = \hbar \omega_{13} A_{11} + \hbar \omega_{23} A_{22} + \sum_{ks} \hbar \omega_{ks} a_{ks}^{\dagger} a_{ks},$$
$$H_{AR} = -\sum_{ks} \{ (g_{ks} A_{13} + f_{ks} A_{23}) (a_{ks} + a_{ks}^{\dagger}) + \text{H.c.} \}.$$

Here $A_{lm} = |l\rangle\langle m|$ and $\hbar \omega_{lm}$ is the energy separation between the levels $|l\rangle$ and $|m\rangle$. The annihilation (creation) operator corresponding to the radiation field in the mode *ks* is



FIG. 1. Schematic of a V system in a thermal bath. The γ 's denote the spontaneous emission rates and the excited levels are assumed to be coupled via the vacuum field.