Fermi's golden rule for N-body systems in a blackbody radiation

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We review the calculation of Fermi's golden rule for a system of N-body dipoles, magnetic or electric, weakly interacting with a blackbody radiation. By using the magnetic or electric field-field correlation function evaluated in the 1960s for blackbody radiation, we deduce a general formula for the transition rates and study its limiting, fully coherent or fully incoherent, regimes.

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The incoherent electromagnetic (EM) radiation within a cavity at thermal equilibrium, namely, the blackbody radiation, has, actually, a certain degree of coherence. This is well evidenced by the analysis of the second-order correlation function between electric or magnetic fields reported more than 50 years ago using techniques analogous to those employed in the theory of isotropic turbulence of an incompressible fluid [1–3]. See also [4] for an experimental result. Quite surprisingly, this result has received little attention in the literature, even in dealing with problems of vast interest (see [5] for an exception). Complex quantum systems, schematized as N-body systems, are usually driven to thermal equilibrium by letting them interact with blackbody radiation. In this equilibration, often described in terms of a quantum optical master equation [6], the transition rates induced by the radiation between two states of the system, as well as the spontaneous emission contribution, describe the core processes. We do not have a formula for these transition rates which covers the whole spectrum of situations, from those in which the coherence properties of the blackbody radiation are important to those in which they are irrelevant. The study of N-body systems, N electric or magnetic dipoles in the simplest case, exchanging photons with blackbody radiation appears to be mandatory for understanding many modern mesoscopic experiments.

In this paper, we review from the very beginning the calculation of Fermi's golden rule for a system of N dipoles, magnetic or electric, weakly interacting with blackbody radiation. Using the magnetic or electric field-field correlation function evaluated in [1–3], we deduce a general formula for the transition rates and study its limiting, fully coherent or fully incoherent, regimes.

Consider an isolated N-body system described by the Hermitian Hamiltonian operator $\hat{H}$ acting on a Hilbert space $\mathscr{H}$ of dimension M. For example, we have $M = 2^N$ in the case of N qubits. We assume that the eigenproblem, $\hat{H}|E_m\rangle = E_m|E_m\rangle$, has discrete, possibly degenerate, eigenvalues and that the eigenstates $\{|E_m\rangle\}$ form an orthonormal system in $\mathscr{H}$. The eigenvalues are thought to be arranged in ascending order $E_1 \leqslant E_2 \leqslant \cdots \leqslant E_M$.

We let the system interact with the EM field of blackbody radiation at thermal equilibrium at temperature T. As usual, we suppose that this interaction is sufficiently weak so that it can be tackled by a first-order perturbative analysis. We specialize the discussion to a system consisting of N chargeless spins $\boldsymbol{\sigma}_i$, $i = 1,\dots,N$, interacting with the radiation as pure magnetic dipoles. Similar considerations apply to spinless charged particles interacting as electric dipoles. The analysis is easily extended to mixed electric and magnetic couplings. We adopt the Gaussian system of units.

Let μ be the magnetic dipole moment associated with each spin and $\boldsymbol{r}_i$ the position vector of the ith spin. The locations of the N spins are considered fixed. Due to the interaction with the radiation inside the cavity, the system can change its quantum state by absorbing or emitting photons. In the semiclassical theory of radiation, these processes are associated with the coupling of the dipoles with, respectively, the real or the imaginary part of the plane-wave magnetic fields $\boldsymbol{B}(\boldsymbol{k})\cos(\boldsymbol{k}\cdot\boldsymbol{r} - kct)$. We thus need to introduce two separate interaction operators for each exchanged photon of wave vector $\boldsymbol{k}$,

$$\hat{V}^{\pm}(\boldsymbol{k},t) = -\sum_{i=1}^{N} \mu\boldsymbol{\sigma}_i \cdot \frac{1}{2}\boldsymbol{B}(\boldsymbol{k})e^{\pm i(\boldsymbol{k}\cdot\boldsymbol{r}_i - kct)}, \tag{1}$$

the operator with the plus sign corresponding to an absorbed EM quantum, and that with the minus sign to an emitted one.

Under the effect of the time-dependent perturbation given by Eq. (1), in a time t the system evolves from state m to state n according to the first-order transition amplitude [7]

$$a^{\pm}_{n,m}(\boldsymbol{k},t) = -\frac{i}{\hbar}\int_0^t ds\; V^{\pm}_{n,m}(\boldsymbol{k},s)e^{\frac{i}{\hbar}(E_n - E_m)s}, \tag{2}$$

where

$$\begin{aligned} V^{\pm}_{n,m}(\boldsymbol{k},s) &= \langle E_n|\hat{V}^{\pm}(\boldsymbol{k},s)|E_m\rangle \\ &= -\frac{\mu}{2}\sum_{i=1}^{N}\sum_{h=1}^{3}\langle E_n|\sigma_i^h|E_m\rangle B_h(\boldsymbol{k})e^{\pm i(\boldsymbol{k}\cdot\boldsymbol{r}_i - kcs)}. \end{aligned} \tag{3}$$

The squared modulus of Eq. (2) gives the probability of the system's evolving in a time t from state m to state n due to the interaction with the mode $(\boldsymbol{k},\pm)$. However, for m and n fixed, there are several modes $(\boldsymbol{k},\pm)$ contributing to the transition $m \to n$, namely, all those compatible with the energy conservation law $E_n = E_m \pm \hbar kc$. We thus evaluate the effective probability for the transition $m \to n$ in a time t

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by taking the expectation of $|a^{\pm}_{n,m}(\boldsymbol{k},t)|^2$ over all the modes in the cavity:

$$\begin{aligned}\mathsf{E}(|a^{\pm}_{n,m}(\boldsymbol{k},t)|^2)\\ = \frac{\mu^2}{4\hbar^2}\int_0^t ds\, e^{\frac{i}{\hbar}(E_n-E_m)s}\int_0^t du\, e^{-\frac{i}{\hbar}(E_n-E_m)u}\\ \times\sum_{i=1}^N\sum_{j=1}^N\sum_{h=1}^3\sum_{l=1}^3\langle E_n|\sigma_i^h|E_m\rangle\overline{\langle E_n|\sigma_j^l|E_m\rangle}\\ \times\mathsf{E}(B_h(\boldsymbol{k})e^{\pm i(\boldsymbol{k}\cdot\boldsymbol{r}_i-kcs)}B_l(\boldsymbol{k})e^{\mp i(\boldsymbol{k}\cdot\boldsymbol{r}_j-kcu)}).\end{aligned} \quad (4)$$

In the above formula, all the statistical properties of the blackbody radiation are enclosed in the field-field correlation function $\mathsf{E}(B_h(\boldsymbol{k})e^{\pm i(\boldsymbol{k}\cdot\boldsymbol{r}_i-kcs)}B_l(\boldsymbol{k})e^{\mp i(\boldsymbol{k}\cdot\boldsymbol{r}_j-kcu)})$. This correlation function was first evaluated by Bourret [1] in the case of real fields and then extended to the case of complex fields by Kano and Wolf [2] and by Metha and Wolf [3]. The result which applies directly to our case is [3]

$$\begin{aligned}\mathsf{E}(B_h(\boldsymbol{k})e^{\pm i(\boldsymbol{k}\cdot\boldsymbol{r}_i-kcs)}B_l(\boldsymbol{k})e^{\mp i(\boldsymbol{k}\cdot\boldsymbol{r}_j-kcu)})\\ = \int d\boldsymbol{k}\, e^{\pm i(\boldsymbol{k}\cdot(\boldsymbol{r}_i-\boldsymbol{r}_j)-kc(s-u))}\\ \times\frac{1}{\pi^2}\frac{\hbar kc}{e^{\hbar kc/k_BT}-1}\left(\delta_{h,l}-\frac{k_hk_l}{k^2}\right).\end{aligned} \quad (5)$$

Note that an identical expression holds for the electric-field correlation function. On plugging Eq. (5) into Eq. (4), the integrals over the times s and u can be separately performed as follows:

$$\begin{aligned}\int_0^t ds\, e^{\frac{i}{\hbar}(E_n-E_m\mp\hbar kc)s}\int_0^t du\, e^{-\frac{i}{\hbar}(E_n-E_m\mp\hbar kc)u}\\ = \left|\int_0^t ds\, e^{\frac{i}{\hbar}(E_n-E_m\mp\hbar kc)s}\right|^2\\ = 2\pi\hbar t\,\frac{\sin^2\left[(E_n-E_m\mp\hbar kc)\frac{t}{2\hbar}\right]}{\pi(E_n-E_m\mp\hbar kc)^2\frac{t}{2\hbar}}\\ \simeq 2\pi\hbar t\,\delta(E_n-E_m\mp\hbar kc).\end{aligned} \quad (6)$$

As usual, this approximation is proved to be accurate for t large by using the representation of the Dirac distribution

$$\delta(x) = \lim_{y\to\infty}\frac{\sin^2(xy)}{\pi x^2 y}. \quad (7)$$

We conclude that the effective transition rate from state m to state n is

$$\begin{aligned}P^{\pm}_{n,m} = \frac{1}{t}\mathsf{E}(|a^{\pm}_{n,m}(\boldsymbol{k},t)|^2)\\ = \frac{\mu^2}{2\pi\hbar}\sum_{i=1}^N\sum_{j=1}^N\sum_{h=1}^3\sum_{l=1}^3\langle E_n|\sigma_i^h|E_m\rangle\overline{\langle E_n|\sigma_j^l|E_m\rangle}\\ \times\int d\boldsymbol{k}\, e^{\pm i\boldsymbol{k}\cdot(\boldsymbol{r}_i-\boldsymbol{r}_j)}\frac{\hbar kc}{e^{\hbar kc/k_BT}-1}\\ \times\delta(E_n-E_m\mp\hbar kc)\left(\delta_{h,l}-\frac{k_hk_l}{k^2}\right).\end{aligned} \quad (8)$$

The plus-minus sign in the factor $e^{\pm i\boldsymbol{k}\cdot(\boldsymbol{r}_i-\boldsymbol{r}_j)}$ is irrelevant and is omitted hereafter.

In Eq. (8) we can evaluate the integral over the modulus k of the wave vector by means of the Dirac distribution. We write $\boldsymbol{k} = k\boldsymbol{u}$, with $k = |\boldsymbol{k}|$ and $\boldsymbol{u} = (\sin\theta\cos\phi, \sin\theta\sin\phi, \cos\theta)$ unit vector given in terms of the longitudinal and azimuthal angles θ and ϕ ranging, respectively, in $[0,\pi]$ and $[0,2\pi]$. Using $d\boldsymbol{k} = k^2dk\sin\theta d\theta d\phi$, we get

$$\begin{aligned}P^{\pm}_{n,m} = \frac{\mu^2}{2\pi\hbar c^3}\frac{\omega^3_{n,m}}{e^{\hbar\omega_{n,m}/k_BT}-1}\sum_{i=1}^N\sum_{j=1}^N\sum_{h=1}^3\sum_{l=1}^3 Q^{i,j;h,l}_{n,m}\\ \times\langle E_n|\sigma_i^h|E_m\rangle\overline{\langle E_n|\sigma_j^l|E_m\rangle},\end{aligned} \quad (9)$$

where

$$\omega_{n,m} = |E_n-E_m|/\hbar \quad (10)$$

and

$$Q^{i,j;h,l}_{n,m} = \int_0^\pi \sin\theta d\theta\int_0^{2\pi}d\phi\, e^{i\boldsymbol{u}\cdot(\boldsymbol{r}_i-\boldsymbol{r}_j)\omega_{n,m}/c}(\delta_{h,l}-u_hu_l). \quad (11)$$

The notation in Eq. (9) has been simplified by using $\omega_{n,m} = |E_n-E_m|/\hbar$ instead of two separate angular frequencies $\omega^{\pm}_{n,m}$ for the energy-gaining and the energy-losing transitions. Actually, it results that $\omega^{\pm}_{n,m} = \pm(E_n-E_m)/\hbar = |E_n-E_m|/\hbar$. Note that for $E_n = E_m$ we have $P^{\pm}_{n,m} = 0$, which expresses the fact that there is no zero-mode (constant) EM field, in agreement with the homogeneity and isotropy of the radiation in the cavity. Contributions in which $E_m = E_n$, including also the case $m = n$, may appear only at higher orders of the time-dependent perturbation theory.

Equation (9) is our general expression of the transition rate $m\to n$ for a system of N magnetic dipoles interacting with blackbody radiation. In the case of N electric dipoles, we have an identical formula with $\mu\boldsymbol{\sigma}_i$ replaced by $\boldsymbol{p}_i$, the moment of the ith electric dipole. In this case, as well as in the case of spatial magnetic dipoles, Eq. (9) still holds if $d_{n,m}\omega_{n,m}/c \ll 1$, where $d_{n,m} = \max_i|\langle E_n|\delta\boldsymbol{r}_i|E_m\rangle|$ and $\delta\boldsymbol{r}_i$ is the vector between the positive and negative charges of the ith dipole. This condition allows for a long-wavelength approximation in Eq. (3), so that the phase factors $e^{\pm i(\boldsymbol{k}\cdot\boldsymbol{r}_i-kcs)}$ can still be considered constant factors in respect of the N-body matrix element.

Depending on the spatial distribution of the N dipoles and the value of $\omega_{n,m}$, two limiting regimes of Eq. (9) can be attained.

Fully coherent limit. If the N dipoles are localized in a region of extension $\ell \ll \lambda_{n,m}$, where $\lambda_{n,m} = 2\pi c/\omega_{n,m}$, we have $|\boldsymbol{r}_i-\boldsymbol{r}_j|\omega_{n,m}/c \ll 2\pi$ for any i,j. This implies that in Eq. (11) we can approximate $e^{i\boldsymbol{u}\cdot(\boldsymbol{r}_i-\boldsymbol{r}_j)\omega_{n,m}/c}\simeq 1$ and straightforwardly perform the integrals over θ and ϕ. The result is

$$Q^{i,j;h,l}_{n,m} = \int_0^\pi\sin\theta d\theta\int_0^{2\pi}d\phi\,(\delta_{h,l}-u_hu_l) = \frac{8\pi}{3}\delta_{h,l}. \quad (12)$$

In this limit, Eq. (9) reduces to

$$P^{\pm}_{n,m} = \frac{4\mu^2}{3\hbar c^3}\frac{\omega^3_{n,m}}{e^{\hbar\omega_{n,m}/k_BT}-1} \times \sum_{h=1}^{3}\left|\langle E_n|\sum_{i=1}^{N}\sigma_i^h|E_m\rangle\right|^2. \tag{13}$$

The total dipole of the system, $\sum_{i=1}^{N}\mu\boldsymbol{\sigma}_i$, couples coherently with an isotropic radiation mode of angular frequency $\omega_{n,m}$ weighed according to the Plank distribution. In particular, in the limit of noninteracting particles we get $\sum_m P^{\pm}_{n,m} = O(N)$.

For $N = 1$, Eq. (13) is the standard textbook formula based on the long-wavelength approximation. Reference [8] suggests that this formula is appropriate for describing many electron atoms in a blackbody radiation upon just replacing the dipole (electric or magnetic) of the single electron with the total dipole of the electrons in the atom.

Fully incoherent limit. A much different result is obtained if the N dipoles are separated from each other by a distance much longer than $\lambda_{n,m}$. Suppose, for simplicity, that the dipoles occupy the sites of a regular linear lattice of spacing a. Choosing the reference frame in such a way that the lattice points are determined by the vectors $\boldsymbol{r}_i = (0,0,ai)$, we have $\boldsymbol{u}\cdot(\boldsymbol{r}_i - \boldsymbol{r}_j) = (i-j)a\cos\theta$, with $i,j = 1,\ldots,N$. In the general expression for the coefficients $Q^{i,j;h,l}_{n,m}$ given by Eq. (11), we can separately evaluate the integral over ϕ and obtain

$$\int_0^{2\pi} d\phi\,(\delta_{h,l} - u_h u_l) = f_h(\theta)\,\delta_{h,l}, \tag{14}$$

where

$$f_h(\theta) = \begin{cases} 2\pi - \pi\sin^2\theta, & h = 1,2,\\ 2\pi - 2\pi\cos^2\theta, & h = 3.\end{cases} \tag{15}$$

Performing the remaining integral over θ, we get

$$Q^{i,j;h,l}_{n,m} = \int_0^{\pi}\sin\theta d\theta\, e^{ia\cos\theta(i-j)\omega_{n,m}/c}\, f_h(\theta)\,\delta_{h,l} = q^{i,j;h}_{n,m}\,\delta_{h,l}, \tag{16}$$

where

$$q^{i,j;h}_{n,m} = \begin{cases} \frac{4\pi(b^{i,j}_{n,m}\cos b^{i,j}_{n,m} + ((b^{i,j}_{n,m})^2-1)\sin b^{i,j}_{n,m})}{(b^{i,j}_{n,m})^3}, & i\neq j,\ h = 1,2,\\ \frac{8\pi(\sin b^{i,j}_{n,m} - b^{i,j}_{n,m}\cos b^{i,j}_{n,m})}{(b^{i,j}_{n,m})^3}, & i\neq j,\ h = 3,\\ \frac{8\pi}{3}, & i = j,\end{cases}$$

and

$$b^{i,j}_{n,m} = (i-j)a\omega_{n,m}/c. \tag{17}$$

For $a\omega_{n,m}/c \gg 2\pi$, i.e., $a \gg \lambda_{n,m}$, neglecting terms $O(\lambda_{n,m}/a)$, we can approximate

$$Q^{i,j;h,l}_{n,m} = \frac{8\pi}{3}\,\delta_{i,j}\delta_{h,l}. \tag{18}$$

In this limit, Eq. (9) reduces to

$$P^{\pm}_{n,m} = \frac{4\mu^2}{3\hbar c^3}\frac{\omega^3_{n,m}}{e^{\hbar\omega_{n,m}/k_BT}-1} \times \sum_{i=1}^{N}\sum_{h=1}^{3}\left|\langle E_n|\sigma_i^h|E_m\rangle\right|^2. \tag{19}$$

The transition rate $m \to n$ is now the incoherent sum of N contributions from the single dipoles. Note, however, that the matrix elements $\langle E_n|\sigma_i^h|E_m\rangle$ between two eigenstates of $\hat{H}$ still retain their full N-body character. Equation (18) and, therefore, the fully incoherent formula, (19), apply also when the N dipoles are placed at arbitrary positions, provided the minimal distance between two of them is still $a \gg \lambda_{n,m}$. As in the coherent case, for noninteracting particles we have, again, $\sum_m P^{\pm}_{n,m} = O(N)$.

The conditions for the validity of the fully coherent and fully incoherent limits are better expressed in terms of the energies of the levels n and m. We have, respectively,

$$|E_n - E_m| \ll hc/\ell, \quad \ell = \max_{i\neq j}|\boldsymbol{r}_i - \boldsymbol{r}_j|, \tag{20}$$

$$|E_n - E_m| \gg hc/a, \quad a = \min_{i\neq j}|\boldsymbol{r}_i - \boldsymbol{r}_j|. \tag{21}$$

Observing that $hc = 1.23$ eV μm, it is evident that for atomic or molecular systems in which $|E_n - E_m|$ is, at most, a few electron volts and ℓ is not larger than a few tens of angstroms, Eq. (20) is well satisfied and the fully coherent formula, (13), applies. Vice versa, for microscopic systems in which a is 1 μm and the energy-level separations $|E_n - E_m|$ are much larger than the atomic electron volt scale, condition (21) is met and we can apply the fully incoherent formula, (19). However, this may not be true for systems having, in the thermodynamic limit $N\to\infty$, a phase transition which implies the existence of a vanishing gap. For systems of intermediate extension or in particular regions of the energy spectrum in the presence of a phase transitions, the general formula, (9), must be applied.

Equation (9) has been obtained on the basis of the semiclassical theory of radiation. The field-field correlation function, (5), which is its foundation, can be evaluated in the framework of the quantized theory of radiation and provides an identical result [9]. In this case, however, the interaction of the N-body system with the zero-point energy of the quantized EM modes gives rise to spontaneous emission processes which add to the transition rate for stimulated emission $P^{-}_{n,m}$. The total emission rate, stimulated and spontaneous, is still given by our $P^{-}_{n,m}$, with the average number of photons at energy $\hbar\omega_{n,m}$ increased by one unity [8], namely,

$$\frac{1}{e^{\hbar\omega_{n,m}/k_BT}-1} \to \frac{1}{e^{\hbar\omega_{n,m}/k_BT}-1} + 1.$$

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