Resonant cooperative energy exchange

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New mechanisms of energy exchange are considered for a system of two-level particles behaving cooperatively in the Dicke model. It is shown for the case of dipole-dipole exchange of nonresonant radiation quanta, or the exchange of resonance quanta in the presence of a resonator, that processes involving the relaxation of excited impurity particles are significantly accelerated in a cooperatively behaving medium.

Of the many known collisional processes in atomic and molecular physics,[1,2] there is particular interest in radiative collisions occurring in the field of a strong electromagnetic wave (see, for example, Ref. 3). Such phenomena are usually described in terms of the binary collision approximation, whose parameters (relative velocity and impact parameter) are usually assumed to have a Boltzmann distribution. At the same time, other, coherent, ensembles can also evolve in the field of a strong resonant wave. The Dicke ensemble[4] is an example of this. The cooperative behavior of particles in a coherent ensemble gives rise to new effects such as directional superradiance,[5] superscattering,[6] and so on. In the coherent ensemble, collisional phenomena that are essentially exchanges of radiation quanta can proceed in a different, more complicated manner as compared with the Boltzmann ensemble. In this paper, we examine the influence of these processes on a subsystem of impurity atoms in a Dicke ensemble. The essence of these phenomena is that, when one of the particles of the coherent ensemble exchanges a photon with another, the process can occur synchronously with the exchange of quanta by another pair of particles. We then have an induced rather than a spontaneous radiative process because the presence of two identical photons accelerates each of the exchange processes by a factor of two.

We shall examine this phenomenon within the framework of the simplest model, namely, N identical two-level particles in a radiation field. Collisions of resonantly excited particles with buffer-gas particles and with the walls of the thermostat will be ignored, and we shall suppose that energy exchange occurs mostly via interactions between the resonant particles, induced by the radiation field. The coherent ensemble will be described by the Dicke model.[4] The first step in an analysis of energy exchange between particles in the cooperative ensemble is to determine the distribution function for the populations of the Dicke states in the presence of a laser pump and cooperative spontaneous emission. The second stage is to determine how this ensemble of essentially nonthermal particles facilitates energy exchange between its individual two-level atoms or molecules. We shall split the Dicke system[4] into a main filler gas with a resonant working transition, and an impurity gas with much lower partial density and a working transition that is close enough to resonance. Because of the large number of filler-gas molecules for each impurity gas molecule, we may suppose that the main process of energy exchange is that between the impurity molecules and the filler molecules.

We shall consider two energy exchange mechanisms, namely, the dipole-dipole interaction due to the exchange of nonresonant virtual hard quanta with characteristic wavelengths of the order of the particle separation,[7] and the exchange of resonant quanta when the presence of the resonator ensures that the total energy of these quanta is sufficient.

For simplicity, we shall suppose that the energy levels involved in the working transition are nondegenerate, the linear dimensions of the system are much smaller than the wavelength of the resonant radiation, and the field has a single mode; and the system is located in the resonator. According to the Dicke model, the gas behaves as a single quantum-mechanical system, and spontaneous emission corresponds to transitions between the energy levels of a certain cooperative macroparticle. It is well-known that this Dicke ensemble can be described by the total spin L, and the energy levels are specified by the component L_3 of the total spin along a chosen direction in energy space. The state vector of the Dicke ensemble has the form $|l,m\rangle$, where l is the Dicke cooperative number and m is the quantum number associated with the component L_3 of the total spin.

We can now use the step-up and step-down operators, L^+ and L^-, to express the intensity of spontaneous emission by the Dicke ensemble from the state $|l,m\rangle$ in the form

$$I_{sp}=I_0\langle l,\ m|L^+L^-|l,\ m\rangle=I_0(l+m)(l-m+1), \tag{1}$$

$$I_0={}^4/_3\omega_0{}^3\mu^2/\hbar c^3, \tag{2}$$

where I_0 is the spontaneous transition rate of an individual particle,[3] μ is the matrix element of the operator representing the dipole magnetic moment of the transition, and ω_0 is the resonant frequency.

When spontaneous emission and stimulated absorption and emission are taken into account, the transport equation for the distribution function ρ_m over the states $|l,m\rangle$ of the Dicke ensemble takes the form

$$\frac{\partial\rho_m}{\partial t}=I_0(l+m+1)(l-m)\rho_{m+1}+\frac{\sigma I_p}{\hbar\omega_0}\rho_{m+1}+\frac{\sigma I_p}{\hbar\omega_0}\rho_{m-1}-I_0(l+m)(l-m+1)\rho_m-2\frac{\sigma I_p}{\hbar\omega_0}\rho_m, \tag{3}$$

where I_p is the laser pump intensity in the resonance mode and σ is the photoabsorption cross section.

In steady state, (3) leads to the equation of detailed balance for the population fluxes produced in the upward direction by the pump field and in the downward direction

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by spontaneous emission to all the field modes and stimulated transitions due to the pump:

$$\alpha(l+m+1)(l-m)\rho_{m+1}+\rho_{m+1}=\rho_m, \tag{4}$$

where we have introduced the dimensionless parameter

$$\alpha=I_0\hbar\omega_0/\sigma I_p=(I_0\hbar/\mu E_н)^2=2\omega_0{}^6\mu^2/9\pi c^5 I_p. \tag{5}$$

For a smooth distribution function, i.e., when $|(\rho_{m+1}-\rho_m)/\rho_{m+1}|\sim\alpha l^2\ll 1$, we have

$$\rho_s=B\exp\left[-\alpha s^2\left(l-\frac{s}{3}\right)\right],$$

$$B=\left\{\int_0^{2l}\exp\left[-\alpha s^2\left(l-\frac{s}{3}\right)\right]ds\right\}^{-1}, \tag{6}$$

where $s\equiv l+m$ varies between 0 and $2l$ and B is a normalizing factor (when $\beta\equiv\alpha l^3\gg 1$, we have $B\approx 2(\alpha l/\pi)^{1/2}$.

Let us now investigate energy exchange between a particular two-level particle and the Dicke ensemble. In the near zone of the chosen particle, the main energy exchange mechanism is the dipole-dipole interaction. The corresponding interaction operator is

$$\hat{V}=-\sum_{k=1}^{N}\frac{\mu^2}{R_k^3}(\sigma_k{}^+\sigma^-+\sigma_k{}^-\sigma^+), \tag{7}$$

where R_k is the distance between the centers of mass of the k th particle and the chosen particle, σ^+ describes the transition from the lower to the upper level, and σ^- the transition from the upper to the lower level. The dipole-dipole interaction disturbs the phases of the dipole moments of the particles. The analysis given here is therefore valid only for the initial stage of the dipole-dipole exchange between the chosen particle and the Dicke ensemble, during which the dipole-dipole interactions between the filler-gas particles do not as yet significantly disturb the coherence of the Dicke enemble.

We shall suppose that the particles of the medium, which form a single Dicke macroensemble by their mutual interaction via the radiation field, have parallel dipole moments and are therefore responsible for the macroscopic polarization of the medium, whereas the macroscopic polarization of the Dicke particle aligns the dipole moments of the individual particles in the same direction. This enables us to reduce $\hat{V}$ to a form that corresponds to the interaction between the chosen particle and the Dicke macroensemble in the dipole-dipole approximation, i.e.,

$$\hat{V}=-\hbar\tilde{\omega}(L^+\sigma^-+L^-\sigma^+), \quad \tilde{\omega}=\frac{1}{\hbar N}\sum_{k=1}^{N}\frac{\mu^2}{R_k^3}, \tag{8}$$

where $\tilde{\omega}$ is the effective interaction frequency. Let us suppose that the chosen particle is initially ($t=0$) in an excited state. We can then use the method of slowly-varying amplitudes to derive the expression for the population of the upper state of the chosen particle on the assumption that a stationary distribution has been established in the system over the energy levels of the Dicke particle (8):

$$\rho_{22}(t)=\mathrm{Sp}\{\rho_m\cos^2[{}^1/_2\tilde{\omega}t(\langle l,m|L^-L^+|l,m\rangle)^{1/2}]\}. \tag{9}$$

For a weakly excited system ($\beta\gg 1$), we find, using (2) and (8), that

$$\rho_{22}(t)\approx\frac{1}{2}+\int_0^{2l}\left(\frac{\alpha l}{\pi}\right)^{1/2}\exp(-\alpha l s^2)\cos\{\tilde{\omega}t[2l(s+1)]^{1/2}\}ds. \tag{10}$$

The spectral density of the relaxation process (line shape) of the chosen particle as it "collides" with the Dicke ensemble is

$$J(\omega)=\frac{1}{2\pi}\int_{-\infty}^{\infty}\exp[i(\omega-\omega_0)t]\rho_{22}(t)dt\approx\frac{1}{2}\delta(\omega-\omega_0)$$

$$+\left(\frac{\alpha l}{\pi}\right)^{1/2}\frac{|\omega-\omega_0|}{l\tilde{\omega}^2}\exp\left\{-\alpha l\left[1-\frac{(\omega-\omega_0)^2}{2l\tilde{\omega}^2}\right]^2\right\}. \tag{11}$$

The characteristic relaxation time for the dipole-dipole cooperative collisions is

$$t_{dd}=(4\pi/\tilde{\omega})(\alpha/l)^{1/4}. \tag{12}$$

After averaging over the realizations of the Dicke ensemble, we find that

$$\tilde{\omega}=(4\pi\mu^2/\hbar V)\ln(\lambda_0/2\pi a_0),$$

where $V=(\lambda_0/2\pi)^3$ is the volume of the system, λ_0 is the wavelength of the resonance radation, and a_0 is a characteristic dimension of the gas particles. If we suppose that the cooperative Dicke number l is not very different from the total number N of particles, i.e., $N-l\ll N$ (this is justified if we suppose that gas-kinetic collisions do not succeed in disturbing the particle phases or the coherence of the Dicke ensemble throughout the time interval), we obtain the following estimate for the relaxation time:

$$t_{dd}=\frac{\hbar\beta^{1/4}}{n\mu^2}\Big/\ln\frac{\lambda_0}{2\pi a_0}, \tag{13}$$

where $n=N/V$ is the gas density and $\beta\gg 1$.

We must now consider the cooperative exchange of resonant photons between particles of the medium through their common radiation fields. We shall suppose that the external radiation field excites the ensemble of particles that are capable of cooperatively emitting spontaneous photons into a large number of field modes. However, a high-Q open optical resonator is tuned to one of these field modes and has the property that photons entering it cannot escape from the system other than by being again absorbed by the gas particles and emitting into other low-Q modes. Such problems have been solved[8,9] under the conditions of thermodynamic equilibrium.

The complete Hamiltonian for the system is

$$\hat{H}=\hat{H}_f+\hat{H}_p+\hat{H}_i,$$

where $\hat{H}_f=\hbar\omega_0 AA^+$ is the field Hamiltonian, $\hat{H}_p=\hbar\omega_0 L_3$ is the Hamiltonian for the Dicke ensemble, $\hat{H}_i=\hbar g(A^+L^++AL^-)$ is the Hamiltonian for the interaction between the Dicke ensemble and the field in the dipole approximation, g is the constant of this interaction, and A and A^+ are, respectively, the operators describing the creation and annihilation of a resonant photon. The mean field energy density and the mean interaction energy are, respectively, given by

$E^2/8\pi=\hbar\omega_0\langle AA^+\rangle/V$ and $\hbar^2g^2\langle AA^+\rangle=(\mu E)^2$

so that

$$g=8\pi^2(2\pi c/\hbar)^{1/2}\mu/\lambda_0^2. \tag{14}$$

Because cooperative processes in the Dicke model are macroscopic, the field can be described classically, but the particle states can be specified by quantum-mechanical variables. According to the correspondence principle, the semiclassical approach is asymptotic for large field quantum numbers. For small field quantum numbers (m approaching $-l$), the validity of the semiclassical description can be illustrated by the following calculation.

In the quantum-mechanical case, the system is specified by state vectors $|n, m, l\rangle$, where n is the number of photons in the resonant mode of the field and m and l have the same meaning as before. We then have

$$A|n, m, l\rangle=(n+1)^{1/2}|n+1, m, l\rangle,$$

$$A^+|n, m, l\rangle=n^{1/2}|n-1, m, l\rangle \tag{15}$$

and (2) is satisfied.

In the interaction representation, the Schroedinger equation

$$i\hbar\partial\Psi/\partial t=\hat{H}\Psi \tag{16}$$

for the wave function of this *particle + field* system in the basis of the state vectors $|n, m-n, l\rangle$ ($-l\leqslant m\leqslant l$, $0\leqslant n\leqslant m+1$) takes the form

$$\begin{aligned}i\dot{\psi}_n=&g[(n+1)(l-m+n+1)(l+m-n)]^{1/2}\psi_{n+1}\\&+g[n(l+m-n+1)(l-m+n)]^{1/2}\psi_{n-1},\end{aligned} \tag{17}$$

where

$$\Psi=\sum_{n=0}^{m+l}\psi_n|n, m-n, l\rangle \tag{18}$$

is the wave function of the system. In the approximation $l-m+n\simeq l-m$ and $n\ll l-m$ (m assumes values near $-l$), the problem can be solved analytically. If we introduce the dimensionless time

$$\vartheta=gt(l+1-m)^{1/2} \tag{19}$$

and use the method of generating functions, we obtain the solution of this equation in the form

$$\begin{aligned}\Psi(\vartheta)=&\sum_{n=0}^{l+m}\left[\frac{(l+m)!}{n!(l+m-n)!}\right]^{1/2}\\&\cdot(-i\sin\vartheta)^n(\cos\vartheta)^{l+m-n}|n, m-n, l\rangle.\end{aligned} \tag{20}$$

This gives us the required quantum-mechanical expectation value of the field amplitude

$$\begin{aligned}\langle A\rangle=&\sum_{n=0}^{l+m-1}(n+1)^{1/2}\dot{\psi}_{n+1}\psi_n\\\approx&i(l+m)\sin\vartheta\cos\vartheta[\sin^2\vartheta+(l+m)\cos^2\vartheta]^{-1/2}\end{aligned} \tag{21}$$

and the intensity

$$\langle AA^+\rangle=\sum_{n=0}^{l+m}n\dot{\psi}_n\psi_n=(l+m)\sin^2\vartheta. \tag{22}$$

It follows from (21) and (22) that, when the system is weakly excited, it exhibits periodic beats of its own radiation field with small amplitude $(\hbar g/\mu)(l+m)^{1/2}$ and high frequency $g(l-m+1)^{1/2}$, that corresponds to the matched emission and absorption of resonance radiation.

If we suppose that the field is classical, its intensity can be determined by using the solution of Hamilton's equation constructed from the classical Hamiltonian, which has the following form in terms of the action-angle variables:

$$H=2\hbar g[I(l+m-I)(M+I)]^{1/2}\cos\theta, \tag{23}$$

where I is the action variable (classical analog of the component L_3 of the energy spin), θ is the angle variable, $M=l-m+\varepsilon$, and $0<\varepsilon\sim 1$, which gives the necessary square-root dependence of H on I at the turning points $I_1=0$ and $I_2=l+m$. The Hamiltonian (23) is the classical analog of the quantum-mechanical Hamiltonian used in (16) and having the following matrix element in the interaction representation in the basis of the state vectors $|n, m-n, l\rangle$:

$$\begin{aligned}H_{n,n'}=&\hbar g\{[(n+1)(l-m+n+1)(l+m-n)]^{1/2}\delta_{n,n'-1}\\&+[n(l+m-n+1)(l-m+n)]^{1/2}\delta_{n,n'+1}\}.\end{aligned} \tag{24}$$

Of course, this ignores the process of absorption and emission of pump photons during the time of absorption and emission of photons of the resonator modes. It is assumed that the function of the pump radiation is only to form the distribution function for the populations of the Dicke states, which then acts as the initial condition for the generation and absorption of photons in the particular resonator mode. The Hamilton equation then takes the following form (the quantity $\hbar$ is introduced, through a suitable choice of units, for convenience):

$$\hbar\frac{dI}{dt}=-\frac{\partial H}{\partial\theta}=2\hbar g\left[I(l+m-I)(M+I)-\left(\frac{H}{2\hbar g}\right)^2\right]^{1/2}. \tag{25}$$

This gives

$$2gt=\int dI\left[I(l+m-I)(M+I)-\left(\frac{H}{2\hbar g}\right)^2\right]^{-1/2}. \tag{26}$$

In the same approximation as in the quantum-mechanical case, i.e., $M+I\simeq M$, which is valid for values of m close to $-l$, we obtain from (26)

$$I(t)=(l+m)\sin^2(gtM^{1/2})+i(H/2\hbar gM^{1/2})\sin(2gtM^{1/2}). \tag{27}$$

Since $I=\langle AA^+\rangle$, and if we use the correspondence principle, we must put $\varepsilon=1$ and $H=0$ in (27). The semiclassical approach then yields the same values for the intensity of the intrinsic field of the system as the quantum-mechanical calculation. Thus, the semiclassical approach is justified for the description of the weakly excited system. When $H=0$, (26) becomes

$$2gt=\int dI[I(l+m-I)(M+I)]^{-1/2}. \tag{28}$$

Substituting $I=u^2$ and

$$u=[(l+m)M]^{1/2}\sin\psi[M+(l+m)\cos^2\psi]^{-1/2}$$

we can reduce the elliptic intergral (28) to the standard elliptic integral of the second kind:

$$gt=(2l+\varepsilon)^{-1/2}F(\varphi,\ k), \tag{29}$$

where

$$\varphi=\arcsin\left[(2l+\varepsilon)I/(l+m)(l-m+\varepsilon+I)\right]^{1/2},$$

$$k=\left[(l+m)/(2l+\varepsilon)\right]^{1/2},$$

and, hence, we find the expression for

$$I(\tau)=Mk^2\,\mathrm{sn}^2(\tau,\ k)/\mathrm{dn}^2(\tau,\ k) \tag{30}$$

in terms of elliptic functions of the dimensionless time

$$\tau=gt(2l+\varepsilon)^{1/2}. \tag{31}$$

The classical field amplitude

$$E=\frac{\hbar g}{\mu}I^{1/2}=\frac{8\pi^2(2\pi\hbar c)^{1/2}}{\lambda_0^{\,2}}k(1-k^2)^{1/2}(2l+\varepsilon)^{1/2}\frac{\mathrm{sn}(\tau,k)}{\mathrm{dn}(\tau,k)} \tag{32}$$

has the oscillation period

$$T=4K(k)/g(2l+\varepsilon)^{1/2}, \tag{33}$$

where $K(k)$ is the complete elliptic integral of the first kind. The period T increases monotonically from $T=2\pi/g(2l+\varepsilon)^{1/2}$ for $m=-l$, to $T=2\ln[16(2l+\varepsilon)/\varepsilon]/g(2l+\varepsilon)^{1/2}$ for $m=l$, where, for k approaching unity, the period is

$$T(m)\approx 2\ln\left[16\frac{2l+\varepsilon}{l-m+\varepsilon}\right]\Big/\,g(2l+\varepsilon)^{1/2}.$$

The maximum field amplitude $|E|_{max}=\hbar g(l+m)^{1/2}/\mu$ is reached periodically at times $t=T/4,3T/4,5T/4,7T/4,\ldots$, with different periods for different values of m. Each initial state of the system gives rise in the resonator to its own radiation field with its own period. The superposition of radiation fields from states excited by the pump field is therefore a relatively complicated and irregular field. The random character of the phases of the start states (due to the pump and to spontaneous decay) enables us to view this field as random. The chosen particle, which occupies the upper state at $t=0$, is transferred by the resonance field with randomly varying amplitude and phase to a state with mean population of each level equal to 1/2. This is actually a process of coherent damping of the oscillations of the chosen particle in the radiation field of the Dicke ensemble, in which the random parameter is the initial state of the enesemble. Let us examine this process of coherent damping. We shall, of course, neglect the influence of the chosen particle on the field. If we again use the method of slowly-varying amplitudes, we obtain the following expression for the population of the upper state of the chosen particle:

$$\rho_{22}(t)=\mathrm{Sp}\left[\rho_m\cos^2\int_0^t\frac{\mu E(t')}{2\hbar}dt'\right]. \tag{34}$$

Taking the equilibrium distribution function for the energy levels of the Dicke ensemble (6), and using (32) and (31), we obtain

$$\rho_{22}(\tau)=\frac{1}{2}+\frac{1}{2}\int_0^{[2l/(2l+\varepsilon)]^{1/2}}\frac{1-k^2+k^2\,\mathrm{cn}(\tau,k)}{\mathrm{dn}(\tau,k)}$$

$$\cdot 2(2l+\varepsilon)\left(\frac{\alpha l}{\pi}\right)^{1/2}\exp\{-\alpha(2l+\varepsilon)^2k^4[l-{}^1/_3(2l+\varepsilon)k^2]\}dk^2. \tag{35}$$

In the case of a strongly excited Dicke ensemble ($\beta\sim 1$), the characteristic cooperative relaxation time is

$$t_c\approx 1/gl^{1/2}. \tag{36}$$

Suppose that the pump is weak and the system is far from saturation ($\beta\gg 1$). The relaxation of the chosen particle is then slower, i.e., $\tau\gg 1$, and the main contribution to the integral in (35) is provided by the region $k\ll 1$. We can therefore use the approximation

$$\mathrm{cn}(\tau,\ k)\approx\cos\left[\tau(1-k^2/4)\right],$$

$$\mathrm{dn}(\tau,\ k)\approx 1-{}^1/_2k^2\sin^2\left[\tau(1-k^2/4)\right], \tag{37}$$

$$\rho_{22}\approx 1-2\int_0^1\left\{\cos\left[\tau\left(1-\frac{k^2}{4}\right)\right]+\frac{1}{2}\sin^2\left[\tau\left(1-\frac{k^2}{4}\right)\right]-1\right\}$$

$$\cdot\left(\frac{\beta}{\pi}\right)^{1/2}\exp(-4\beta k^4)k^2\,dk^2. \tag{38}$$

The spectral density (line shape) for the process is

$$J(\omega)=\frac{1}{2\pi}\int_{-\infty}^{\infty}\exp[i(\omega-\omega_0)t]\rho_{22}(t)\,dt$$

$$\approx\delta(\omega-\omega_0)+\frac{4}{g}\left(\frac{\beta}{2\pi l}\right)^{1/2}\{(1+\Omega)\exp[-4\beta(1+\Omega)^2]$$

$$+(1-\Omega)\exp[-4\beta(1-\Omega)^2]-8(1+2\Omega)\exp[-4\beta(1+2\Omega)^2]$$

$$-8(1-2\Omega)\exp[-4\beta(1-2\Omega)^2]\},\qquad \Omega\equiv\frac{\omega-\omega_0}{2g(2l)^{1/2}}. \tag{39}$$

The characteristic relaxation time in this case is

$$t_{c\beta}\approx t_c\beta^{1/2}=\frac{1}{3}\left(\frac{\hbar}{I_p}\right)^{1/2}\frac{l}{\lambda_0}. \tag{40}$$

Hence, it is clear that the relaxation time is not very dependent on the effect of the pump on the system.

In conclusion, let us consider some numerical estimates of the rates of the above processes, and discuss physical situations in which such phenomena could be observed. In the infrared, in which a stationary Dicke ensemble can be set up by pulsed radiation from a CO_2 laser ($\lambda_0=10.6\,\mu$m) at a pressure of about 1 torr ($n\simeq 3.2\times 10^{16}$ cm^{-3}), and collisional processes do not fully succeed in manifesting themselves during pulse lengths of about 10–100 ns, the following results are valid for a gas of molecules with characteristic dipole moment $\mu\simeq 0.3$ D:

cooperative Dicke number

$$l\approx 1.5\cdot 10^5 p[\text{ torr}];$$

characteristic degree of excitation of Dicke particle

$$s=(\alpha l)^{-1/2}\approx 2.8\cdot 10^{3}\ (I_p[\ \mathrm{W}\ ]/p[\ \mathrm{torr}\ ])^{1/2};$$

characteristic energy exchange time due to the cooperative dipole-dipole interaction

$$t_{dd}=\hbar\beta^{1/4}/n\mu^{2}\ln(\lambda_0/2\pi a_0)\approx 2.9\cdot 10^{-7}\sec(p[\ \mathrm{torr}\ ]I_p[\ \mathrm{W}\ ])^{-1/4};$$

characteristic relaxation time due to the exchange of resonant photons (process occurring in the resonator)

$$t_c=\hbar^{1/3}l/3\lambda_0 I_p^{1/3}\approx 5\cdot 10^{-10}\sec p[\ \mathrm{torr}\ ](I_p[\ \mathrm{W}\ ])^{-1/3}.$$

We particularly draw attention to the short relaxation time t_c in the resonator. It may well be that this is the mechanism capable of explaining the short relaxation time in a gas illuminated by radiation pulses in a waveguide-type cell.[10]

A more detailed study of the process will, of course, require special investigation. In our view, this could be carried out by probing the state of a medium excited by pulsed radiation over time intervals shorter than the collisional times. The optimum probing method can be based on SRS processes that are not sensitive to the degree of saturation (Ref. 6). The degree of excitation of the impurity gas can be monitored by recording the line shape of another transition between one of the working levels and a nonworking level chosen so that the line is absent from the spectrum of the filler gas. The SRS processes should be used for such levels. It is likely that both the near and far zones can be examined in this way. Cooperative energy exchange in the far zone can probably be investigated in special resonators, e.g., elliptic resonators, in which there is strong optical coupling between different points in space. The above process should then ensure efficient energy exchange between individual particles in one of the foci and the Dicke particle produced by laser radiation in the other. We suggest that this could be verified experimentally by using the vibrational-rotational part of the spectrum of molecules containing a small number of atoms.

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