

## SUPERRADIATION OF A BOSON AVALANCHE

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A theory of spontaneous development of coherent states in a system of  $N$  particles is developed. It is shown that such a system emits a coherent boson field with an intensity proportional to  $N^2$ . The relations derived are used to interpret the experimental data pertaining to development of a phonon avalanche in paramagnetic systems.

AT the present time, in interpreting processes of longitudinal or energy relaxation (for example, paramagnetic spin lattice or nonradiative optical relaxation), one starts from the concepts of (a) isolated and (b) non-interacting impurity centers. In addition, in the existing theories of relaxation no account is taken of the fact that (c) when the system is excited by a coherent external generator the relaxation processes can also proceed partially in a coherent fashion. Thus, if the time of longitudinal relaxation  $\tau_{\parallel}$  of one isolated particle is larger than the time of the transverse or phase relaxation  $\tau_{\perp}$  due to two-particle interactions, then actually the longitudinal relaxation proceeds via scattering of collective excitations (of the exciton type) by the vibrations of the thermal reservoir, and consequently the approximation (b) is not applicable. Certain aspects of the case (c) were considered in [1,2].

However, even if the conditions (b) and (c) are satisfied but (1) the average distance  $r_1$  between particles is of the order of the wavelength  $\lambda$  of the field generated during the relaxation, or (2) the coherence length of the quanta of this field is larger than  $r_1$ , then the particles are coupled via the generated field and condition (a) is violated. In this case the time of longitudinal relaxation of the system of particles can differ by many orders of magnitude from the relaxation time of the particles that are isolated in the sense of conditions (1) and (2).

Let us consider the evolution of a system of  $N$  particles with an equidistant discrete spectrum, which at the initial instant of time  $t = 0$  was at a negative absolute temperature and, consequently, its state was incoherent [3]. We assume that the particles do not interact with one another in the sense of (b). When the system of particles relaxes, a boson field is generated. As a result of the interaction with the boson field, the system relaxes at the instant  $t = 0$  and emits bosons of energy  $n\hbar\omega_0$  ( $\hbar\omega_0$ —distance between energy levels). This can produce bosons with different values of the wave vector  $k$ . But conditions can exist when the bosons with definite  $k_0$  have a maximum value of the product of the production probability and the damping time. This allows us to investigate the mechanism of the occurrence of the boson avalanche only under the influence of bosons with  $k = k_0$ , in analogy with the separation of the modes at which lasers operate.

As a result of the appearance of these bosons, the particles begin to interact with one another via this boson field and go over into a superposition state,

which is described by certain parameter  $\varphi$ . For particles with two energy levels  $\sin^2(\varphi/2)$  is the probability that the particle will go over during the time of its interaction with the boson from the excited state to the ground state. When  $\varphi \neq 0$  sets in, the system begins to generate, besides the spontaneous incoherent boson radiation, also a coherent boson field of power  $\sim \sin^2\varphi$ . This generation leads to further increase of the angle  $\varphi$ , and consequently of the generation power. Obviously, this process increases in avalanche fashion and the radiation power reaches a maximum at  $\varphi = \pi/2$ . We emphasize that the occurrence of  $\varphi \neq 0$  is a spontaneous transition of the system into a superradiating state with respect to the boson field, and the process of boson-avalanche formation is spontaneous.

Apparently the formation of the phonon avalanche which was observed in experiments [4,5] proceeded via the described mechanism. In the investigation of the process of formation of the boson avalanche, we confine ourselves for simplicity to the case of an effective spin  $1/2$ . This approximation makes it possible to obtain the main results in the simplest and most convenient form for interpretation, without loss of generality. The collective states of such a system of particles are best described with the aid of the cooperation quantum number  $r$  and its projection on the quantization axis  $m$ . At a given number  $N$ , the number  $r$  can take on the values  $N, N-1, \dots, 0$ , and for a given  $r$  we have  $|m| \leq r$ ,  $m = (\frac{1}{2})(n_+ - n_-)$ , where  $n_+ - n_-$  is the difference between the populations of the excited ( $n_+$ ) and ground ( $n_-$ ) states of the particles.

Let us assume that the system was initially at a temperature  $T = 0$ , and was then inverted ( $r = N/2$ ,  $m = N/2$ ). As is well known, at the initial instant of time the radiation intensity of such a system of bosons with wave vector  $k$  in a unit solid angle  $\Omega$  in the direction of  $k$  is

$$I(k) = I_0(k)N, \quad (1)$$

where  $I_0(k)$  is the intensity of the spontaneous radiation of one isolated particle per unit solid angle in the direction of  $k$ . This radiation has an incoherent random character. The system begins to relax with generation of bosons having a great variety of  $k$ , corresponding to a disintegration energy  $\hbar\omega_0$ . However, among the large number of such decay there will be included a decay with  $k = k_0$ , for which  $I_0(k_0)$  has a maximum value. Obviously, as already noted, this type of boson determines the further evolution of the system. We

shall therefore consider henceforth the system development due precisely to this type of boson.

Let the linear dimensions of the system be much larger than the wavelength of the generated bosons. Then at the instant of time  $t > 0$  ( $t = 0$  is the instant of emission of the boson with  $\mathbf{k} = \mathbf{k}_0$ ) the intensity of the spontaneous radiation of the system is calculated from the formula

$$I_0(\mathbf{k}_0, t) = I_0(\mathbf{k}_0) \frac{N}{2} \left\{ 1 + \cos \varphi(t) + \frac{1}{2N} \sin^2 \varphi(t) \sum_{j \neq l} \exp[i(\mathbf{k} - \mathbf{k}_0) \mathbf{r}_{jl}] \right\}, \quad (2)$$

where  $\varphi(t)$  is the angle between  $\mathbf{r}$  and the quantization axis at the instant of time  $t$ ;  $\mathbf{r}_{jl} = \mathbf{r}_j - \mathbf{r}_l$ ;  $\mathbf{r}_\xi$  is the radius vector of the location of the particle  $\xi$ .

Let us consider a sample of rectangular cross section. Let the area of the face perpendicular to  $\mathbf{k}_0$  be  $S$ . To determine the variation of  $\varphi(t)$  we start from the following obvious equality, which expresses the law of energy conservation

$$\int_0^t dt' \int d\Omega I(\mathbf{k}_0, t') + \hbar \omega_0 m(t) = \text{const}, \quad m(t) = \frac{N}{2} \cos \varphi(t). \quad (3)$$

Differentiating this equation with respect to  $t$  and summing over  $j \neq l$  in (2), we obtain from (3) the equation

$$\frac{d \cos \varphi(t)}{(1+c) - \cos \varphi(t)[1+c \cos \varphi(t)]} = -adt, \quad (4)$$

$$\alpha = \tilde{I}_0(\hbar \omega_0)^{-1}, \quad \tilde{I}_0 = \int d\Omega I_0(\mathbf{k}_0), \\ c = N\lambda^2 I_0(\mathbf{k}_0) [S \tilde{I}_0]^{-1}.$$

Neglecting small terms, we write the solution of this equation under the initial condition  $\varphi(t) = \varphi_1$  at  $t = 0$  will in the form

$$\sin \varphi(t) = \frac{2 \operatorname{ctg}(\varphi_1/2)}{e^{\xi t} + \operatorname{ctg}^2(\varphi_1/2) e^{-\xi t}}, \quad \xi = \frac{I_0(\mathbf{k}_0) N \lambda^2}{\hbar \omega_0 S}. \quad (5)$$

From (5) with  $\varphi_1 = \pi/2$  we obtain the well-known result<sup>[6]</sup>:

$$\sin \varphi(t) = \operatorname{sech}(\xi t). \quad (6)$$

Substituting (5) in (2), we find that the boson radiation intensity varies with time in accordance with the law

$$I(t) = \tilde{I}_0 N \frac{\operatorname{ctg}^2(\varphi_1/2) e^{-\xi t}}{e^{\xi t} + \operatorname{ctg}^2(\varphi_1/2) e^{-\xi t}} + I_0(\mathbf{k}_0) N \lambda^2 \frac{\operatorname{ctg}^2(\varphi_1/2)}{S (e^{\xi t} + \operatorname{ctg}^2(\varphi_1/2) e^{-\xi t})^2}. \quad (7)$$

The coherent part of the spontaneous boson radiation is described by a term proportional to  $N^2$ ; the term proportional to  $N$  describes incoherent spontaneous noise.

As follows from (7), the intensity of the coherent radiation increases up to a certain instant of time  $t_1$ , and then decreases rapidly. The instant of time  $t_1$  can be readily determined from the extremum condition, which yields

$$t_1 = \left( \ln \operatorname{ctg} \frac{\varphi_1}{2} \right) \xi^{-1}. \quad (8)$$

As already noted above,  $\varphi_1$  is determined from the condition that the system of particles goes over into a superposition state with emission of a boson with  $\mathbf{k} = \mathbf{k}_0$  at the instant of time  $t = 0$ . Since  $\varphi_1$  is very small, (8) can be represented in the form

$$t_1 = \ln \left( \frac{2}{\varphi_1} \right) \xi^{-1} = \frac{1}{2} \frac{(\ln N) \hbar \omega_0 S}{I_0(\mathbf{k}_0) N \lambda^2} \approx \frac{1}{2} \frac{(\ln N) S \tau}{N \lambda^2}, \quad (9)$$

where  $\tau$  is the time of the spontaneous decay of one polarized particle. At instants of time  $t > t_1$  there oc-

curs a rapid damping of the coherent part, with a characteristic time

$$t_2 = \xi^{-1} = S \hbar \omega_0 [I_0(\mathbf{k}_0) N \lambda^2]^{-1} \approx S \tau [N \lambda^2]^{-1}. \quad (10)$$

The ratio of the times is

$$t_2 / t_1 = [\ln(2/\varphi_1)]^{-1} \sim 2 / \ln N. \quad (11)$$

so far, apparently, boson avalanches were observed on phonons.

Brya and Wagner<sup>[4]</sup> investigated the formation of a phonon avalanche in single-crystal  $(\text{La}_{0.998}\text{Ce}_{0.002})_2\text{Mg}_3(\text{NO}_3)_{12} \cdot 24\text{H}_2\text{O}$  on  $\text{Ce}^{3+}$  impurities. The phonon avalanche was revealed by observing the longitudinal magnetic relaxation of  $\text{Ce}^{3+}$ . In the case when the relaxation process was observed at a non-equilibrium positive population difference, no formation of the phonon avalanche was observed and the paramagnetic centers relaxed practically like isolated particles. At sufficiently low temperatures  $T (\text{k}_B T \ll \hbar \omega_0, \text{k}_B - \text{Boltzmann constant})$ , such a process is described by the term proportional to  $N$  in formula (2). At higher temperatures, the relaxation is due to simulated single-phonon and combination processes and also proportional to  $N$ . It follows therefore that the relaxation times observed in this case should correspond to the time of the individual isolated centers. The coherent ( $\sim N^2$ ) part of the radiation should in this case be absent, for the reason that the creation of nonequilibrium populations occurred within a time exceeding the transverse relaxation time, and by the end of the excitation of the system the spins were out of phase ( $\mathbf{r} = 0$ ).

At positive populations, the spontaneous formation of a coherent state is also excluded, for in this case  $\pi > \varphi(t) > \pi/2$  and  $\sin^2 \varphi(t)$  decrease with evolution of the system, and therefore even if an admixture of coherent states is produced spontaneously in the system at a certain instant of time, it attenuates rapidly. On the other hand, when the relaxation process was investigated at negative populations, a sharp shortening of the time of the longitudinal relaxation of the system of  $\text{Ce}^{3+}$  centers was observed. In this case the evolution of the system is described by the relations obtained above.

Similar results were obtained also by Shiren<sup>[5]</sup>, who investigated the formation of a phonon avalanche by using  $\text{Fe}^{2+}$  ions in an  $\text{MgO}$  crystal placed in a strong constant magnetic field. Unlike in<sup>[4]</sup>, in this case the generated phonons were observed directly. The process of formation of the phonons avalanche observed in<sup>[4,5]</sup> has the following distinguishing features, which can all be explained within the framework of the theory developed above:

1. After formation of a negative population, the phonon avalanche is observed not immediately, but after a certain time interval, which can naturally be identified with the time  $t_3$  during which the intensity of the coherent part of the spontaneous radiation reaches the intensity of the spontaneous noise. This time is shorter than the time  $t_1$ . As follows from (7), the intensity of the spontaneous noise in this region  $t \leq t_3$  is practically constant and  $\sim \tilde{I}_0 N$ . In<sup>[4]</sup> this process gives rise to a gently-sloping initial part of the curve describing the variation of the population difference from the instant after the termination of the exciting pulse. In<sup>[5]</sup>, where

the generated hypersound was observed, it is seen directly that during the time  $t_3$  the power of the coherent spontaneous emission increases smoothly.

2. The gently-sloping section of the curve of<sup>[4]</sup> is followed by a sharp decrease of the population difference. In<sup>[5]</sup> this section corresponds to an equally sharp increase of the sound intensity, the power of which at a certain instant of time reaches a maximum value. The time evolution of these sections of the curves, as follows from (7), is described by the formula

$$I_{ac}(t) = -\Delta N(t) = I_0(k_0)N^2 \frac{\lambda^2}{S} \operatorname{ctg}^2 \frac{\varphi_1}{2} e^{-2\frac{t}{\tau_1}}, \quad (12)$$

where  $I_{ac}(c)$  is the intensity of the sound and  $\Delta N(t)$  is the difference of the populations at the instant of time  $t$ .

The sound intensity reaches a maximum value at an instant of time which can be naturally identified with  $t_1$  from formula (9). After attainment of the maximum,  $I_{ac}(t)$  decreases rapidly. If the decrease is not due to the time of the transverse relaxation, then the decrease time is the time  $t_2$  given by formula (10).  $t_2$  is the time during which the intensity of the coherent part of the radiation is of the order of the intensity of the spontaneous noise. The ratio of these times observed in<sup>[5]</sup> is  $t_1/t_2 \sim 10$ . We see that a quantity of the same order is obtained from formula (11) at  $N \sim 10^{19} - 10^{20}$  particles. Values of  $N$  of this order correspond to the experimental conditions. Consequently, measurements of the ratio  $t_1/t_2$  yield a method for measuring the concentration of impurity particles without knowledge of the parameters of interaction between the impurity and the lattice.

Attention was called in<sup>[5]</sup> to the fact that, as follows from experiment, the phonon avalanche is produced in a mode having a minimum damping, and not in all the modes that are possible in the given disintegration and capable of taking part in the ordinary spin lattice relaxation; this contradicts the usual concept whereby heating of the phonon spectrum is due to the "bottleneck" effect<sup>[7]</sup>.

It is obvious that a boson avalanche can be produced by relaxation of other excited centers in gases, liquids, and solids. For example: 1) in quadrupole and dipole longitudinal relaxation of nuclear spins, which, depending on the physical conditions, can proceed with production of either a phonon or a photon avalanche; 2) in the relaxation of nuclear spins via electron spins, spin waves, and a system of conduction electrons; 3) in relaxation of electron spins via interaction with more rapidly relaxing centers of a different nature (for example, via exchange pairs); 4) in relaxation of spin waves, helicons, and plasmons; 5) in radiative and non-radiative optical relaxation. Naturally, the formation of a boson avalanche can proceed also via multiquantum transitions or in nuclear reactions.

The question of the influence of "phonon bottleneck" effects on the electronic and nuclear spin-lattice relaxation and on dynamic polarization of nuclei has been extensively discussed in the literature of late<sup>[4, 5, 8]</sup>. As noted by the authors of these papers, there is still no agreement between the conclusions of the theory of the phonon "bottleneck" and experiment. In particular, when the samples become smaller and the thermal contact with the helium bath is improved, the influence of

the "bottleneck" effect on the longitudinal relaxation should decrease. However, this is not observed experimentally<sup>[8]</sup>. The "bottleneck" effect should vanish also upon increase of the concentration of the paramagnetic centers, which should lead to an increase of the time of longitudinal relaxation. Experiments give the opposite dependence: the spin lattice relaxation time decreases with increasing concentration<sup>[8]</sup>.

We wish to call attention to the fact that these anomalies can be naturally explained within the framework of the theory of coherent relaxation (phonon avalanche) developed above. Thus, if conditions are created for the formation of a phonon avalanche, then, at the same geometry and at the same size of the sample, the spin-lattice relaxation time should decrease with increasing concentration of the paramagnetic centers. When the sample is made thinner, the number of paramagnetic impurities  $N$  decreases, and in accordance with (9) and (10) this should lead to a lengthening of the time of relaxation of the electron spins. This effect was observed in<sup>[4]</sup>. However, if the ratio  $S/N$  remains constant as the geometry of the sample changes, then shortening of the time  $\tau_{\parallel}$  may not be observed<sup>[8]</sup>. The coherent electron relaxation can be investigated also by the method of dynamic polarization of nuclei<sup>[9]</sup>. A decrease of the maximum attainable degree of polarization of nuclei with increasing concentration of the paramagnetic center, as well as an absence of any changes in the processes of polarization with decreasing sample thickness, was observed in<sup>[8]</sup>, in full agreement with the above-developed theory of coherent avalanche spin-lattice relaxation. We emphasize that the phonon avalanche cannot be produced under conditions when the "bottleneck" effect does not take place. Moreover, the formation of the phonon avalanche under "bottleneck" conditions is hindered by the reaction of the radiated phonons. As is well known from laser emission theory, this should lead to oscillations in the power of the emitted phonons and obviously to oscillations of the populations, as is indeed observed in experiment.<sup>[10]</sup>

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