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SUPERCOHERENT OPTICAL CLUSTER MEDIA

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1. Introduction

The rapid development of fibre-optics communications, various technological applications of laser radiation call for creation of optical media with new properties. In the present work specific artificially created optical media are discussed, in which the weak monochromatic electromagnetic radiation may induce autophasing oscillations of dipole particles. The autophasing of dipole oscillations arising as a result of the positive feedback will lead to a sharp increase in the effectiveness of the field-medium interaction and, as a consequence, to new physical phenomena. Such media are called below "supercoherent" or "SC-media". Taking into account the immediate practical applications and terminology used in the literature the term "high dispersion optical media" will be also employed, although the latter refers only to one of the properties of the SC-media, and not all high dispersion media are necessarily supercoherent (see part 4). The aim of this work is to put forth a hypothesis about possible creation of SC-media as well as to discuss some of their properties. A rigorous theoretical justification for the hypothesis will be given in the framework of comprehensive theoretical and experimental investigations of supercoherent media. Below is presented the motivation for the investigations, physical arguments to substantiate the effect of supercoherence, the current status of research on high dispersion media and heterogeneous optical media, the difficulties associated with their creation, the role of the local field effects, the physical mechanism of supercoherence and an analogy with lasers, suggestions about the creation and studies of the properties of cluster supercoherent media as well as references to some experiments; specific examples of cluster media are also discussed.

2. Motivation for investigations

Practical interest in the studies of the SC-media is connected, first of all, with their *high dispersion*, reduction of the velocity of light in a SC-medium by a factor of 2-10 and more, as well as with *the possibility of regulating the refractive index* of the SC-medium. Let us point out only some of the applications for an optically transparent medium with a high and controlled index of refraction.

This is a simple and cheap means of increasing the resolving power of an optical microscope [1] (Fig.1), increasing the density of information may record on optical discs (Fig.2), precision and ultrafast control of the laser beam (Fig.3); a controlled transfocator lens with the variable focal length (Fig.4). Other more special applications are also possible such as provision of coherent energy transfer from the electron beam into the radiation of a free electron laser at a low electron velocity in the beam, particle acceleration in the laser field [2], construction of highly-sensitive superconducting photodetectors [3], laser diagnostics of biological objects [4].

SC-media are interesting from the point of view of their *new fundamental physical properties,* which are the condition of a broad spectrum of various practical applications. There is a close analogy between the SC-medium and laser, where “authophasing” and, as a consequence, high coherence of photon oscillations in a light wave are realised. Due to the coherence of radiation, lasers are also in wide use in fundamental scientific investigations. Unlike lasers the autophasing of particle (dipole) oscillations and not of the electromagnetic field photons takes place in the supercoherent medium, which leads to a very high dispersion. The sharp increase in the dispersion at the expense of the local field, the Clausius-Mossotti catastrophe, has long been predicted theoretically [5,6]. It is suggested a possible transition to the supercoherent medium and is an analogy to the transition to laser generation. The practical realization of supercoherent optical media might lead to progress in fundamental as well as in applied fields of physics much as it happened after the discovery of the laser.

3. On the possibility of SC-media creation

Before starting comprehensive theoretical investigations, it is useful to consider a simple model of a high dispersion medium and adduce elementary physical arguments in favour of its practical realization. Let us assume that we want to create a medium in which the light velocity is ten and more times less than in the vacuum. In order to evaluate its possibility, let us consider the mechanism of changing of the electromagnetic radiation phase velocity (Fig.5).

An electromagnetic wave coming from the free space induces oscillations of the medium's dipole-particles, which are shown in Fig.5 as oscillators. The field in the medium is due to the re-emission of the dipoles. For the linear case, when the external field is not very large, the re-emitted field has the same frequency as the incident one, but the further it is from the medium's boundary, the more it delays in phase. The phase delay is connected with the finite time of absorption and re-emission of the field by each particle. As is seen from Fig.5, the phase delay causes a reduction in the light speed, which implies an increase in the refractive index of the medium. Thus, the increase of the time of the atom-field causes growing of the refractive index of the medium.

How great can be the atom's time of response to the influence of an electromagnetic field? In the linear case, when an atom interacting with the field plays the role of a harmonic oscillator, the response time can be *infinitely large* if the resonant conditions are fulfilled without any losses:

(i) the frequency of the field coincides with the frequency of the atomic transition,

(ii) energy losses of the field in the medium are negligibly small,

(iii) phase distortions at an atom-field interaction are sufficiently small.

When the conditions (i) – (iii) are met, atoms are excited even by a very weak field, which consequently requires more time. In the ideal case, a reduction in the wavelength as well as an increase in the refractive index are restricted only by non-linear effects arising when atoms are highly excited. The weaker is the exterior field, the greater is the maximum value of the refractive index. It is interesting to note that when (i) – (iii) conditions are fulfilled, *nonlinear optical effects, such as harmonic generation, parametrical generation etc., might arise even in weak fields*. In the case of a very weak field, when separate photons are registered, dispersion in a resonant transparent medium is restricted by quantum fluctuations.

A medium in which all conditions (i) – (iii) are fulfilled for each atom interacting with the field is supercoherent. There is a close analogy between the SC-medium and laser amplifier, the linear theory of which predicts infinite amplification of the field at the generation threshold, which is limited in the next approximation by nonlinear effects.

There is no difficulty in meeting the condition (i), which is fulfilled upon making a choice of the resonant atomic transition (for example, the line D_1 of the atomic Rb^{87}, which is resonant to the radiation of a 794 nm semiconducting laser [7]) as well as the condition (ii), which is fulfilled either by adding active atoms into the medium that radiate at the frequency of the external field or by using three-level Λ-schemes [8,9].

The condition (iii) is of fundamental importance and most difficult to fulfil, as resonant media are very sensitive to phase fluctuations, especially when there is a great concentration of particles (see below). But *a-priori* there is no principal limit to the fulfilment of this condition as well. A strict justification for the fulfilment of the (iii) condition in a number of specific cases is one of the key tasks in theoretical investigations of supercoherent media.

4. The current status of research on the studies of high dispersion media and heterogeneous optical media

Optically transparent homogeneous media with a relatively high dispersion (the refractive index n being of the order of a few unities) are well known. They are, for example, semiconducting compounds with possible $n \sim 2 - 3$ and higher [10]. At the same time, such media cannot be called supercoherent, as the frequency of the optical radiation propagating in them without significant losses is far from the resonant with any energy transition. A contribution to the dispersion in non-resonant media is simultaneously made by a lot of transitions, and a high index of refraction is achieved not due to the high excitation of separate dipoles, as expected in SC-media, but owing to the high concentration of particles. Well-known high dispersion transparent media are solids, as a rule, with concentration $N_0 \sim 10^{23}$ atom/sm^3, or liquids. Development of a detailed theoretical model of dispersion as well as calculation of the dielectrical permeability in condensed media is a very complicated problem (see, for example, [11]). As a rule, it is considered more from the point of view of prediction of the medium's physical properties by its known dielectrical permeability than with the aim of creation of a medium with a high and controllable index of refraction.

Of special interest are the studies of the optical properties of *heterogeneous* non-resonant optical media representing an optically transparent matrix with microparticles of a substance having a dialectric constant different from the dielectric constant of the matrix. Some of the heterogeneous optical media, the so-called *photon crystals* (PhC) [12], have "forbidden optical zones": the optical radiation of a special frequency range doesn't propagate in PhC. Some of PhC are two-dimensional: they have forbidden zones only for two dimensions (that is for the electromagnetic radiation propagating in some plane). An atom, placed in a two-dimensional PhC and having a resonant transition with the frequency within the PhC forbidden zone, will radiate for the most part in the direction perpendicular to the plane "forbidden" for the radiation. In this case, PhC act as radiation collimators. This property of PhC is planned to be used for decreasing the space divergence of light beams in the systems of optical communications.

In heterogeneous non-resonant optical media significant increase is observed of the non-linear optical properties of microparticles in comparison with the non-linear optical properties of the homogeneous material that constitutes microparticles [12].

In some experiments an increase in the refractive index was observed in heterogeneous non-resonant optical media [22] and fabrication of an effective antireflection coating based on the heterogeneous polymer film was reported [23].

Comprehensive literature is available on the studies of the properties of the radiation of a separate dipole (atom) near a conducting or dielectrical particle [13]. The main conclusion is that the velocity of the dipole spontaneous radiation in such a system turns out to be many times greater than that of an isolated dipole.

Of particular value is the discussion of resonant optical media (ROM), in which dispersion is conditioned by one or several optical transitions being in resonant with the radiation, other transitions may be neglected. In fact, the polarizability of a resonant atom is $\sim \lambda^3$, where λ is the wavelength of the incident optical radiation (see formula 3), which is many times greater than the size of the atom. On the other hand, the polarizability of a non-resonant particle of the size a is of the order of its volume a^3. Thus, in the dipole approximation when the size of an elementary particle in the medium is much less than λ, the interaction of a resonant atom with the field is much more effective than that of any non-resonant particle. Our prime interest being in the possibilities of the supercoherent medium construction, ROM will be our chief subject of discussion. At the same time, experiments on heterogeneous non-resonant media might turn the necessary "first step" in the direction of experimental studies of heterogeneous ROM.

In the framework of the ROM analysis it is possible to construct a reliable and clear model of a high dispersion medium which, what counts, allows control of the refractive index with the help, for example, of an additional resonant radiation in schemes [8,9]. *One of the tasks of the theoretical investigation is to show that a high refractive index, n ~ 2 – 4 and higher, is possible in ROM with a small absorption.*

The most known model of high dispersion ROM was suggested in [8,9]. It was based on the employment of coherent effects of the so-called Λ-atoms [9] in a gas medium (Fig.6 b). A strong electromagnetic field resonant to the transition *b'-a* together with the non-coherent pumping at the transition *b-b'* lead to the amplification of the weak "probe" field resonant to the transition *b-a*. If the frequency of the probe field is detuned from the resonant by the quantity δ_0, the medium consisting of Λ-atoms becomes absolutely transparent, as is seen in Fig.6 a, where the refractive index and probe field absorption in the gas of Λ-atoms are illustrated as functions of detuning from the resonant. From Fig.6 a it follows that the refractive index of the probe field at the point of transparency will be highly great, of the order maximally possible in the vicinity of the resonant. The curves in Fig.6 a correspond to the concentration of particles N_0, for which $2\pi\alpha_0\pi_0 N_0 = 1$, where $\alpha_0 = \frac{3}{2}\frac{\lambda^3}{(2\pi)^3}\frac{\gamma_{sp}}{\gamma}$ is the characteristic polarizability of the medium in the vicinity of the resonant, λ is the wave length of the probe field, λ and λ_{sp} are correspondingly total and natural line widths of the transition *a* – *b*.

Taking $\lambda_{.}$ to be ~ 1 μm, γ / γ_{sp} ~ 1 we have: with $2\pi\alpha_0\pi_0 N_0$ = 1, $N_0 \sim 4 \cdot 10^{13}$ sm^{-3}, which corresponds to the resonant gas partial pressure not less than 0.001 Torr. As

$$n = 1 + 2\pi\alpha_0 N_0 \cdot \Delta n_1(\delta), \qquad (1)$$

where $\Delta n_1(\delta)$ is curve 2 in Fig.6 a, $2\pi\alpha_0\pi_0 N_0 \geq 100$ is required to provide $n \geq 10$.

A great number of publications, such as [15] and others, are dedicated to the studies of high dispersion resonant media like those suggested in [8,9]. In fact, the evidence for this effect was given in experimental work [7], but the measured values of the refractive index turned out relatively small: $(n-1) \sim 10^{-4}$. First of all, this was due to the fact, that the experiment was carried out at the concentration of resonant atoms $N_0 \sim 10^{12}$ sm^{-3}, while an increase in the concentration destroyed the coherent interaction of the strong field with the probe field, which led to the disappearance of the effect.

Realization of the so-called “slow light” [16] has been recently reported. It deals with the observation of a very small *group* velocity of the light pulse propagation. It is important to stress that only optical media with a small *phase* velocity of the light are under discussion.

In paper [17] a scheme was proposed for increasing and control of the refractive index using the Rayleigh scattering of the probe field into the resonator. In [17] a mechanism was used analogous to the one of increasing the refractive index at the expense of the local field, which is discussed below. This scheme [17] provides, in fact, a better control of the system’s parameters than in the case when the local field is used. Evaluations performed in [17] showed that to achieve a significant increase in the refractive index in real conditions, media with particles the polarizability of which is approximately 100 times higher than the maximal polarizability of a resonant atom are required. This is just the case for the cluster supercoherent media, which can also make it possible to realize the idea [17]. Conceivably, the optimal conditions of the supercoherent medium realization might come about from the joint action of the mechanisms [17] and local field.

In our opinion, the problem of coherence preserve in ROM at the high concentration of particles $N_0 \geq 10^{12} \div 10^{13}$ sm^{-3} (condition (iii)) is of great importance for the achievement of the high dispersion and supercoherence. Both theorists [18, 19] and experimentators [20, 21] are well aware of this problem. Although in a number of cases it is underestimated in the theoretical studies of coherent effects, such as [8,9]. For high dispersion ROM it reduces to the requirement:

$$2\pi\alpha_0 N_0 >> 1. \qquad (2)$$

The difficulties associated with the fulfilment of requirement (2) are discussed in the next part. *A rigorous justification for condition (2) to be fulfilled for a certain resonant medium is one of the key tasks of theoretical studies of high dispersion ROM.*

5. Difficulties associated with creation of high dispersion resonant optical media

The basic problem necessary to solve for creation of high dispersion ROM is the fulfilment of condition (2). Let us consider the typical polarizability

$$\alpha_0 = \frac{3}{2}\frac{\lambda^3}{(2\pi)^3}\frac{\gamma_{sp}}{\gamma}, \qquad (3)$$

meeting requirement (2). Here γ is the total width of the resonant transition line. For a gas consisting of two-level resonant atoms

$$\gamma = \gamma_{sp} + \gamma_{dd} + \gamma_N, \qquad (4)$$

where γ_{sp} is the natural line width, γ_N is the inhomogeneous line width, γ_{dd} is a contribution to the line width either due to the resonant dipole-dipole interaction of particles or to the "self-broadening". The latter plays a decisive role in suppressing coherent processes in resonant media with the concentration of particles $N_0 \geq 10^{12} \div 10^{13}$ sm^{-3}, that is when the fulfilment of requirement (2) can be expected for the electromagnetic radiation within the optical range $\lambda \sim 1$

μm. Now let us discuss the physical mechanism of self-broadening and evaluate the quantity λ_{dd} for gases.

Self-broadening is conditioned by the process of resonant energy exchange between the particles at the expense of their dipole-dipole interaction with the characteristic energy V_{dd} (see Fig.7). This interaction leads to a shift of resonant transition levels [5,6,20] as well as to its self-broadening. The self-broadening is due to the following: the time of interaction for each pair of atoms is different, and the position of the energy levels for each particle changes very rapidly near an average from interaction to interaction. A simple qualitative estimation of the quantity of self-broadening is as follows: $\hbar\gamma_{dd} \sim V_{dd}$, where $V_{dd} \sim \mu^2 / r_0^3$, μ – is the dipole moment of the transition, $r_0 \sim N_0^{-1/3}$ is the mean distance between the particles. Using $\gamma_{sp} = \frac{2}{3}\frac{\mu^2\omega^3}{\hbar c^3}$ the following may be written:

$$\gamma_{dd} = \frac{3}{2}\frac{N_0\lambda^3}{(2\pi)^3}\gamma_{sp} \cdot D, \tag{5}$$

where *D* is the dimensionless constant. In paper [18], where self-broadening was calculated with regard to the fluctuations of the particle density in the gas at the expense of their thermal motion, *D* ~ *10* was obtained, which is in agreement with the experimental data. With no thermal motion, for example, in the case of motionless particles chaotically placed in the matrix, *D* ~ *1* [21] is possible to evaluate, self-broadening, in this case, being connected with the Van Der Waals interaction between the particles through spontaneous radiation. Thus, *D* ~ *1* ÷ *10* is evaluated. Substituting (3)-(5) in formula (1), disregarding the inhomogeneous self-broadening, for the sake of simplicity, it is not difficult to obtain:

$$n = 1 + \cdot\frac{3}{2}\frac{\lambda^3 N_0}{(2\pi)^2}\frac{1}{1+1.5D\lambda^3 N_0/(2\pi)^3}\Delta n_1(\delta). \tag{6}$$

In Fig.8 the curves for the maximal *n* determined for $\delta = \delta_0$ (see Fig.6 a) are given as functions N_0. As is seen, when self-broadening is taken into account, the maximal $n \sim 1.7$ even with the "optimistic" evaluation of *D* = *1*.

Thus, self-broadening is a serious obstacle to creation of high dispersion resonant media. It doesn't allow fulfilment of requirement (2) by simply increasing the number of particles. Therefore, *a search for resonant media with a relatively small self-broadening* is necessary for the creation of SC-media. The idea that self-broadening can be suppressed in the so-called *cluster media* is discussed below, in part 8, its rigorous physical justification will be given separately in the framework of a comprehensive theoretical analysis of SC-media. The problem is simplified, as there is no requirement for the complete suppression of self-broadening for the creation of SC-media. In the next part it will be shown that when factor *D* is decreased up to a critical value, the Clausius-Mossotti catastrophe may arise consisting in a rapid "avalanche-like" increasing of dispersion, which implies a transition to the supercoherent medium.

6. Local field correction

The so-called local field correction may play an important role in creation of "supercoherence" in the medium. It makes allowance for the dissimilarity of the electromagnetic field acting in a medium on the microscopic scale of the order $N_0^{-1/3}$, a mean distance between the particles, from the "mean" field, which is the result of a statistically averaging of the fields due to the sufficiently great number of particles. The "mean" or "Maxwell" field in the medium is a solution of the Maxwell microscopic equations. The local field correction has been calculated in many publications [5,6,20]. It has been shown that in the field in a homogeneous isotropic medium there is

$$\vec{E}_{loc} = \vec{E} + \frac{4\pi}{3}\vec{P}, \tag{7}$$

where $\vec{E}$ is the Maxwell field, $\vec{P}$ is the medium's polarization. The physical meaning of formula (7) is consideration of the energy of a dipole-dipole interaction of particles in the medium polarized by the exterior field. It should be noted that although formula (7) agrees well with the experimental data, it makes allowance for the local field correction averaged over a great number of particles [6]. A more rigorous analysis requires taking into account the fluctuations of the local field which, in fact, are to lead to self-broadening [18]. Thus, both the local field correction and self-broadening have the same physical origin: they arise as a result of a resonant dipole-dipole particle interaction; therefore, in all calculations they must be taken into account simultaneously.

A consequent description of the medium in the electromagnetic field requires the field $\vec{E}$ to be replaced everywhere by $\vec{E}_{loc}$. In doing so, the authors [20] predicted bistability due to the local field.

In order to account for the decisive role of the local field in creation of supercoherence, let us calculate the linear polarizability of the medium taking into account the local field for the case of the Λ-scheme depicted in Fig.6 b. The polarizability induced by the probe field *E* is

$$P = \alpha_0 N_0 \Delta n_1(\delta) E_{loc} \equiv \alpha_0 N_0 \Delta n_1(\delta)[E + (4\pi/3)P], \qquad (8)$$

where the dimensionless function $\Delta n_1(\delta)$ is determined from an analysis of an interaction between Λ-atom and the resonant fields, the function $\Delta n_1(\delta)$ is illustrated in Fig.6 a. Let us chose detuning from the resonant $\delta = \delta_0$ corresponding to the total transparency so that $\Delta n_1(\delta_0)$ is real. Then from equation (8) and the definitions $P = 4\pi\chi E$, $\varepsilon = 1 + 4\pi\chi$, $n = \sqrt{\varepsilon}$ we obtain

$$n = 1 + \frac{2\pi\alpha_0 N_0 \Delta n_1(\delta_0)}{1 - (4\pi/3)\alpha_0 N_0 \Delta n_1(\delta_0)}. \qquad (9)$$

As is seen from equation (9), $n \to \infty$ with

$$N_0 \to N_{cr} = 3/[4\pi\alpha_0 \Delta n_1(\delta_0)] = const \sim 1. \qquad (10)$$

The given peculiarity about the refractive index is specific to any transparent medium where the local field correction is taken into consideration, it has long been known and is called Clausius-Mossotti catastrophe [6]. As far, as the Λ-scheme is concerned, it was pointed out by the authors of [22]. Along with it, a substantial increase in the refractive index predicted in (9) has not been observed in practice for the reasons discussed below. *In our opinion, the peculiarity about expression (9) with $N_0 \to N_{cr}$ testifies to the appearance of the supercoherent medium.* Of course, this process does not lead to an infinite increase in *n*, which is to be conditioned by the nonlinear effects, but it can bring about a substantial increase in *n* as well as supercoherence. Where will the increase in *n* come to a halt, what properties, apart from the high dispersion, will the supercoherent medium have? An unambiguous answer is difficult to provide without an analysis of the nonlinear problem. It is assumed that there may arise domain structure of electric dipoles analogous to that observed for magnetic dipoles.

Below we are going to discuss once more the physical origins of supercoherence, which can be now clarified using the local field. But first, we shall discuss why the peculiarity of *n* following from (9) was not observed in the experiment as well as what requirements are to be fulfilled to make it observable. For this purpose, let us consider expression (3) for α_0, in which γ is determined by formulas (4), (5), inhomogeneous self-broadening being disregarded here. Substituting an obvious expression for α_0, into formula (9), it is easy to see that the denominator (9) becomes zero, $n \to \infty$ with the concentration of particles

$$N_0 \to N_{cr} = \left(\frac{\lambda}{2\pi}\right)^3 \frac{1}{2\pi\Delta n_1(\delta_0) - 3D/2}. \tag{10}$$

To provide the peculiarity $n \to \infty$, $N_{cr} > 0$ is necessary, which is possible if

$$D < \frac{4\pi}{3}\Delta n_1(\delta_0). \tag{11}$$

For the case of the Λ-scheme given in Fig.6 b, requirement (11) is equivalent to D < 0.53. Thus, a substantial increase in *n* at the expense of the local field in the gas consisting of Λ-atoms fails to arise even with the "optimistic" evaluations of D~1. Even for the maximally possible for resonant media $\Delta n_1 = 0.5$, which corresponds to a two-level system, $D < 2$ is required, whereas all estimations which are in agreement with the experimental data give $D \sim 10$. Thus, *self-broadening is one of the reasons why the increase in n is not observed experimentally.*

In Fig.9 functions for the maximal $n(N_0)$ are presented, which have been found with the help of expression (9), in which $\Delta n_1(\delta_0)$ is determined from Fig.6 a. When Fig.8 with no local field correction is compared with Fig.9, it becomes clear that the role of the local field increases with the decreasing of self-broadening. A sharp increase in *n* as well as a transition of the gas consisting of Λ-atoms into the «supercoherent» state may be expected as self-broadening decreases approximately in 20 times.

7. Physical mechanism of supercoherence. An analogy with the laser

In order to better comprehend in what way is connected with the local field correction, let us discuss the physical origin of the latter more thoroughly. Let us consider a dipole (a two-level atom) linearly interacting with the resonant electromagnetic field, that is a plane wave $Ee^{i\vec{k}\vec{r}-i\omega t}$. An isolated dipole absorbs an energy $n\hbar\omega$ at the point $\vec{r}=\vec{r}_i$ and starts to oscillate coherently (in phase) with the field *E*. The induced dipole moment of the atom is

$$d_1=\alpha_0\Delta n_1 Ee^{i\vec{k}\vec{r}_i-i\omega t}, \quad (12)$$

where $\alpha_0\Delta n_1$ is the polarizability, α_0 is determined by expression (3), $\Delta n_1 \leq 1$ is a dimensionless function depending on the detuning of δ of the field frequency from the natural frequency of the dipole oscillations and atomic energy levels scheme. For the case of the Λ-scheme, the function $\Delta n_1(\delta)$ is depicted in Fig.6 a. Note that the response (re-emission) of the dipoles to the effect of the plane electromagnetic wave *E* is a field coherent to *E*, not a single plane wave but their superposition (Fig.10 a).

Now let us consider many dipoles in a medium. The energy $\hbar\omega$ absorbed by one of the dipoles leads not only to its coherent oscillations but also to the coherent oscillations of its neighbours exposed to the *"multimode"* radiation of the dipole (Fig.10 b). The difference in the field at each point of the medium with the respect to the average field is connected with the dipole radiation modes that do not coincide with the mode of the incoming field. In averaging over many particles in a volume $>>\lambda^3$, all modes except for the incident one are mutually eliminated. As a result of the difference of the local field from the average field, there arise in a medium consisting of many particles a dipole moment greater as accounted for by one particle than that of an isolated dipole with the same absorbed energy, and the higher is N_0, the greater is this difference (with no regard for self-broadening). According to formula (9), the local field correction becomes relatively big when $\alpha_0 N_0 > 1$. At the same time, according to (3), two or more particles are contained in the volume ~ λ^3 on the average. Thus, for $\alpha_0 N_0 > 1$ an increase in the concentration of particles leads not only to an increase in the polarization of the medium $P \sim N_0$ but also to an increase in the effective dipole momentum accounted for by one particle, which implies *a positive feedback* between P and N_0. Notice that this feedback can be of importance for a gas consisting of resonant atoms the polarizability of which is $\alpha_0 >> r_{at}^3$, where r_{at} is the size of the atom, and therefore, $\alpha_0 N_0$ can be great enough in comparison with unity. For a gas consisting of non-resonant particles consideration of the local field leads only to slight corrections [6]. The polarizability of a non-resonant particle is truly of the order of its volume, therefore, for a gas consisting of non-resonant particles the condition $\alpha_0 N_0 > 1$ is fulfilled only with a very high density of the order of condensed media density whereby gas cannot exist.

There is an analogy between the supercoherent and laser media. In the latter there is a positive feedback between a part of the pumping energy radiated by the active medium into the coherent (laser) mode of the field and the photon number in the laser mode: the greater is the energy of the laser mode, the more effective is the transformation of the pumping energy into the coherent radiation. For the coherent medium it turns out that the greater is the number of particles $N_0 \lambda^3$ in the volume λ^3, the more effectively a separate particle is polarized by the exterior coherent field, which implies a positive feedback between the concentration of particles N_0 and the polarization of the medium P. The positive feedback in the laser gives rise to the macroscopic coherent radiation if the threshold condition is fulfilled, that is if the energy flux into the laser mode exceeds the energy losses in this mode. A positive feedback between P and N_0 might lead to the Clausius-Mossotti catastrophe consisting in an avalanche-like growth of P and a transition of the medium into the supercoherent state. Laser pumping is a mechanism of particle transition from the lower to the upper laser level. For the supercoherent state the pumping consists in an increase of the concentration of particles N_0. A variable undergoing a phase transition in a laser is the photon number, for the supercoherent medium it is the medium polarization. In a laser a phase transition starts from the state when the photon number in the laser mode is relatively small and changes slowly as pumping increases and is completed at the state when the photon number is great and rapid growth of the photon number takes place as pumping increases. In the supercoherent medium a phase transition is carried out by the polarization: it starts from the state conditioned by the linear interaction with the exterior field and is completed at the supercoherent state, the properties of which are as yet unknown.

The threshold requirements for the emergence of supercoherence are formulated by analogy with the laser ones: the energy flux into the coherent mode of the polarization (coinciding with the mode of the weak exterior field) at the expense of an increase in N_0 is to exceed the energy losses in this mode. However, the mechanisms of energy losses for the laser and for the supercoherent medium are radically different. For the laser they consist in a decay of the field from the resonator as well as in the losses through absorption inside the resonator. These losses are comparably easy in control, which is achieved by regulating the transparency of the resonator mirrors and (or) decreasing the inside-resonator losses through absorption. For the supercoherent medium the losses of the macroscopic polarization are conditioned by the phase distortion of the coherent oscillations of the dipoles, which is primarily due to self-broadening. Self-broadening is taken into consideration in an analysis of the Λ-scheme, the results of which are presented in Fig.9. Requirement (10) is a threshold condition, (11) being a necessary requirement for the emergence of supercoherence in a gas consisting of Λ-atoms.

Now it is possible to more specifically formulate one of the key problems of the theory of supercoherent media. This is *a search for a medium in which self-broadening is so small that condition (11) is fulfilled.* In the next part the hypothesis is discussed that requirement (11) is possible to fulfil in the so-called cluster resonant media.

8. Possibility of a high dispersion and supercoherence in cluster resonant media

Let us bring forward some heuristic arguments in favour of the hypothesis formulated in the headline of this section. Further, its rigorous justification will be carried out in the framework of a comprehensive theoretical study of supercoherent media.

In place of separate resonant atoms interacting with the field, groups consisting of $N_{cl} > 1$ atoms are suggested to be used, which are *clusters* the size $r_{cl} << \lambda / 2\pi$, where λ is the wavelength of the optical radiation. As $\lambda/(2\pi) \sim 2\cdot 10^{-5} sm$ and $r_{am} \sim 10^{-8} sm$ is the characteristic size of the atom, the maximum size of the cluster may be $r_{cl} \sim 2\cdot 10^{-6} sm$, so that a cluster may consist of up to 10^6 particles (with their maximum concentration in the solid state of $\sim 10^{23}\ sm^{-3}$). A cluster may be represented by a metal or semiconductor particulate, as well as consist of a transparent dielectric matrix with impurity resonant atoms. A cluster consisting of one or several resonant atoms near a metal particulate is also possible.

Let us assume that with the characteristic size $r_{cl} \sim 2\cdot 10^{-6} sm$ a cluster contains a not-to-great number of resonant particles, not more than $10^4 \div 10^5$. Correspondingly, their concentration in the cluster is not higher than $N_0^{cl} \sim 10^{19} \div 10^{20} sm^{-3}$, the mean distance between the atoms being $(N_0^{cl})^{-1/3} \sim 10^{-6} \div 10^{-7}\ sm << r_{at}$, so that the atomic energy levels are not overlapped. The characteristic polarizability of one cluster

$$\alpha_{cl} = \alpha_0 N_{cl} (\gamma_{sp} / \Delta\omega) >> \alpha_0, \qquad (13)$$

of the characteristic polarizability of a separate atom, if

$$N_{cl} (\gamma_{sp} / \Delta\omega) >> 1, \qquad (14)$$

where $\Delta\omega \geq \gamma_{sp}$ is the line width of the atomic resonant transition in the cluster conditioned by self-broadening and dielectric matrix inhomogeneities.

Condition (14) is easy to fulfil if the resonant atoms in the cluster form an ordered structure and self-broadening can be disregarded, because of the density fluctuations are absent. As the size of the cluster is much smaller than the wavelength, the cluster can be regarded as a separate dipole with the characteristic polarizability $\alpha_{cl} >> \alpha_0$.

It is possible that the first problem to solve in the experimental studies of cluster resonant media is the creation of a particle the size much smaller than the wavelength the polarizability of which is greater than that of a resonant atom at the allowed dipole transition. Its simple solution is the construction of a cluster consisting of two resonant atoms.

Let us forget, for the time being, that a cluster includes separate resonant atoms and consider it as a structureless particle with the polarizability $\alpha_{cl} >> \alpha_0$. In the same way as in the medium consisting of separate atoms, there may arise in the cluster medium the Clausius-Mossotti catastrophe, a transition to supercoherence with

$$\alpha_{cl} N_0 \to const \sim 1 \tag{15}$$

where N_0 is the concentration of clusters. But if for a medium consisting of separate atoms a transition to supercoherence is not allowed due to self-broadening, self-broadening may turn out sufficiently small in a cluster medium, so that condition (15) will be fulfilled. In fact, if $\alpha_{cl} >> \alpha_0$, condition (15) is fulfilled when the mean distance between the clusters $N_0^{-1/3} >> \lambda$. At the same time, the interaction of the cluster with the dipole radiation of other particles is to provide the local field correction as before, but the fluctuations of the dipole radiation responsible for self-broadening are suppressed as a result of their destructive interference.This hypothesis was used in calculating self-broadening in a gas consisting of atoms in [18], where contribution into the self-broadening of the atom was taken into account made only by the neighbouring particles from the volume $\sim \lambda^3$, the results [18] being in a good agreement with the experiment. Thus, the hypothesis that *the concentration of resonant atoms in clusters the size* $r_{cl} << \lambda / 2\pi$ *reduces density fluctuations on the length scale* $\sim \lambda$ *, and consequently, leads to the decreasing of self-broadening*, seems to be true (see Fig.11). Relatively narrow lines of the resonant absorption observed in semiconductors serve as the circumstantial evidence in support of this hypothesis. Actually, according to formula (5), $\gamma_{dd}/\gamma_{sp} \sim 10^{-16} N_0$ for the optical range when $\lambda/2\pi \sim 10^{-5}\ sm$, and $D \approx 10$. As out of all terms constituting the full line width (see formula (4)) only $\gamma_{dd} \sim N_0$ supposing $\gamma \approx \gamma_{dd}$ with $N_0 >> 10^{16}\ sm^{-3}$. Thus, taking the concentration of resonant particle in a semiconductor to be $\sim 10^{23}\ sm^{-3}$, the characteristic concentration of atoms in a crystal lattice, we obtain that $\gamma/\gamma_{sp} \sim 10^7$, whereas for the real semiconductors $\gamma \geq 10^{-13}\ Hz$, $\gamma_{sp} \sim 10^{-9}\ Hz$, so that $\gamma/\gamma_{sp} \sim 10^4$. This contradiction indicates that self-broadening in semiconductors, where the particles are orderly located in the crystal lattice, must be suppressed. At the same time, as the order in the crystal lattice of semiconductors is not ideal, it is with a high probability disturbed sufficiently far from any predetermined point with the aim of decreasing self-broadening of a sufficiently "short-range" order at the distances, in any case, much less than the size of the crystal.

There have been a number of reports on increasing refractive index of organic multilayers by addition of metal clusters [23], on the production of a high-quality antireflection coating based on porous films with the pores the size much smaller than the wave length [24].

These publications reveal a keen interest shown by the experimenters and engineers in optical cluster media as well as point out their first practical applications.

9. On the experimental determination of the polarizability of the cluster

Theoretical calculations of the polarizability of the cluster must be in agreement with its experimental measurements. Polarizability can be measured by the energy of the radiation re-emitted backwardly by the cluster. Let the radiated power of the monochromatic radiation incident on the cluster be equal to W_{in}. Let us consider a cluster consisting of N two-level atoms resonant to the incident radiation. The number of atoms is such that the mean distance between the atoms $\Delta r >> r_{at}$ than the size of the atom, the size of the cluster $r_{cl} << k^{-1}$ where k is the wave vector of the resonant radiation. For example, if

$\Delta r \sim 10^{-7}\ sm$, so that the concentration of resonant atoms in a cluster $N_0 \sim 2.5 \cdot 10^{20}\ sm^{-3}$ and $r_{cl} \sim 10^{-6}\ sm$, we obtain $N = 10^3$.

As $r_{cl} << k^{-1}$ the cluster radiates as a dipole. For the sake of simplicity, let us assume that each atom interacts with the radiation independently from the others and denote the polarizability of a separate atom in the cluster as α. Then, the dipole moment of the cluster will be $d = N\alpha E_{in}$ where E_{in} is the incident field strength. According to known formula [25], the power of the dipole radiation into the small solid angle $d\Omega$ is $dW = \frac{\omega^4 |d|^2}{4\pi c^3}\sin^2\theta\, d\Omega$ where θ is the angle between $\vec{d}$ and the direction of the dipole radiation. In considering the backward radiation, so as $\theta \approx \pi/2$, $d\Omega = 2\pi \sin\theta\, d\theta \approx 2\pi\, d\theta$, it is possible to obtain the total power ΔW of the dipole radiation into the small solid angle $\Delta W = \frac{\omega^4 |\alpha|^2 N^2 E_{in}^2}{2c^3} \cdot \frac{D}{L}$, where D is the size of the detector aperture, L is the distance between the cluster and detector, $L >> D$. Let us assume that the line of self-broadening of a separate atom in a cluster is conditioned by the spontaneous radiation, then $\max|\alpha|^2 = 1.5k^{-3}$. Taking into account that $W_{in} = \frac{c}{4\pi} SE_{in}^2$ where S is the area of the cross-section of the incident field , we obtain: $\frac{\Delta W}{W_{in}} = \frac{9\pi}{2}\frac{N^2}{k^2}\frac{D}{LS}$. Assuming that $D = 1\, sm$, $L = 10\, sm$, $S = 0.1\, sm^2$ and $k = 10^5\ sm^{-1}$ we obtain that $\Delta W / W_{in} \sim 10^{-9} N^2$. The present-day techniques allow registration of the scattered laser radiation with $\Delta W / W_{in} \sim 10^{-8}$. Thus, cluster radiation is easy to detect even if a cluster includes only a few resonant atoms. The evaluation will be evidently more optimistic if the dipole radiation of several identical clusters is registered.

As atoms interact with each other due to the dipole radiation, the assumption of independence of the radiation of the cluster's separate atoms must be rejected after a more thorough analysis of the radiation of a separate cluster, which is to be fulfilled in the future.

10. Some types of cluster media and possibilities of their manufacturing

1) Clusters, consisting of a transparent dielectrical matrix, such as "aerogel" SiO_2, or semiconductor matrices with impurity resonant atoms or ions implanted into separate zones of matrices the size $r_{cl} \leq 2\cdot10^{-6} sm << k^{-1}$. One of the possible technologies of clusters manufacturing is *the method of ion implantation* [26]. It allows one to reach the concentration of ions of up to $10^{20}\ sm^{-3}$, this corresponds to $10^2 \div 10^3$ atoms in a cluster, which is, in fact, sufficient for the construction of a supercoherent cluster medium. Clusters are manufactured in the following way: 1. preparation of a macroscopic sample with the maximum possible concentration of ions; 2. destruction of the sample into microparticles; 3. separation of particles the size $r_{cl} \leq 2\cdot10^{-6} sm$, which present clusters; 4. implantation of the clusters into an optically transparent matrix. Construction of clusters is possible by using the ion implantation as such: passing an ion beam through a mask with the holes the size $\sim r_{cl}$. The polarizability of a separate cluster is controlled by changing the number of resonant atoms. The most difficult to realize is the provision of *regular ion distribution in the cluster.* Its realization depends, in particular, on the possibility of controlling the location of a separate ion. *An important problem in the theory of cluster media is studying whether the condition of the atomic regular distribution in the cluster is principally necessary for the achievement of a high polarizability of the cluster.*

2) It is possible that a suitable source material for the formation of clusters will be laser crystals, where resonant particles are impurity metal ions [27,28]. As a resonant transition their absorption bands can be used, which are applied, in the case of a laser, to optical pumping.

Cluster media may be manufactured from laser microcrystals the size $r_{cl} \leq 2\cdot10^{-6} sm$ placed into a transparent matrix, for example SiO_2.

Let us first look closely at some simple consideration in favour of the assumption that the polarizability of a separate cluster must be much greater than the polarizability of a separate ion in a crystal, which is the requirement for the achievement of supercoherence. It is common knowledge that the band width of the ion absorption in laser crystals $\gamma \sim 10^{13}\ Hz$. The line width of an isolated ion in a crystal $\gamma_1 \sim 10^8\ Hz$ and is conditioned by its interaction with phonons. Let us assume that the mechanism of formation of absorption bands is the interaction of an ion with the neibouring impurity ions, that is self-broadening. Inhomogeneous broadening in crystals is negligibly small in comparison with self-broadening. As was mentioned above, the mechanism of self-broadening is a random shift of the resonant level of the ion at its interaction with the neibouring one. Assuming that each neibouring ion makes approximately equal contribution into self-broadening, we obtain that the number of the neibouring atoms forming the band is $N_b \geq \gamma / \gamma_1 \sim 10^5$, and, as was mentioned, these atoms are concentrated in a volume with the linear dimensions $\sim k^{-1}$ (where $k \sim 10^5\ sm^{-1}$ is the wave vector of the resonant radiation), which corresponds to the characteristic concentration of ions $\sim 10^{20}\ sm^{-3}$. As a cluster the size $r_{cl} \leq 2 \cdot 10^{-6} sm$ contains only $N < 10^3$ ions, the absorption line width of the cluster will be $\gamma_{cl} \leq 10^{-2} \gamma$. Its polarizability, correspondingly, will exceed the polarizability of an ion in a macrocrystal by two and more orders of magnitude.

Contraction of the absorption line in a medium consisting of clusters, laser microcrystals, is to be observed regardless of the type of impurity atoms. A corresponding experiment is important for the provision of support for the mechanism of line contraction suggested in the present work. To obtain a supercoherent medium, it is necessary to use impurity ions with the maximum possible dipole moment, such as, for example, Cr-ions. Laser crystals with rare-earths ions are less suitable, as their dipole moments are small; therefore, for the provision of supercoherence cooling of the corresponding crystals will be necessary up to the temperature when the phonon self-broadening will be by one or less order of magnitude less than the natural one.

3) Clusters are semiconducting particles. The electromagnetic field is resonant to the optical transition valence band – conductive band. In this case, a macroscopic semiconducting crystal is destructed into microparticles; particles (clusters) the size $r_{cl} \leq 2 \cdot 10^{-6} sm$ are separated and implanted into a trans-

parent matrix. This is a simpler method than the preceding one, but its weakness is that the polarizability of a separate cluster is limited, as it is fully determined by the polarizability of the source semiconductor sample. *Estimation of the maximally possible polarizability of a semiconducting cluster is one of the tasks of the theoretical investigations of supercoherent media.*

4) Clusters are particles of a material intermediate between the semiconductor and metal, for example, a semiconductor with a high concentration of donors. An electromagnetic field of an optical or infrared range induces in them resonant oscillations of the electron gas at the plasma frequency. They can be manufactured by the method analogous to items 1 and 2. Clusters are placed into a transparent matrix, for example, SiO_2.

5) Clusters are metallic particles implanted into the semiconductor matrix with self-conductivity. Metal implantation is possible, for example, according to technology [3]. In the area of the semiconductor matrix near metal cluster there may arise a p-n transition with a high concentration of active carriers. The truth of this hypothesis is supported by the anomalously high absorption observed in the semiconductor matrix with metal clusters [3]. Determination of the zone structure on the metal cluster – semiconductor boundary is rather a difficult task, which may require much effort and time.

6) Clusters are dielectric particles in a transparent active matrix, probably in a film. Resonant interaction of clusters with the electromagnetic field is not obligatory. This line of investigations has arisen in connection with experiments [29], which will probably start in the nearest future. The appropriate medium has been obtained, the experimental equipment is being mounted. The line of theoretical investigations will be dictated by the results of the experiments.

7) A cluster consisting of a particle of metal with one or several resonant atoms near it. According to [14], the presence of a metallic particle may substantially increase the velocity of the spontaneous radiation γ_{sp} of a resonant atom and, correspondingly, reduce the relative contribution of self-broadening into the full width of the resonant transition (see formula (4)).

Let us make a list of tasks that can be solved in the first stage of theoretical investigations of supercoherent media.

1) Calculation of the polarizability of a cluster with a small number of resonant atoms, $N_{cl} \geq 2$. Determination of the dielectrical function of the corresponding cluster medium as well as of the requirements for a transition to supercoherence.

2) Analysis of the mechanisms of broadening of atoms in a cluster and self-broadening in a cluster medium.

3) Calculation of the polarizability of the cluster, the dielectric function of the cluster medium and the requirements for the transition to supercoherence for a great number, $1 << N_{cl} \leq 10^5$, of resonant atoms in a cluster.

4) The same as for the semiconducting and metal clusters in "the aerogel" SiO_2.

Possible lines of experiments on the construction of supercoherent media as well as on investigation of their properties:

1) Creation of a medium consisting of a polymer or any optically transparent "matrix" (including liquids, ice and so on) and dielectric nanoparticles (that is the size much less than the wavelength) dissolved in it. The dielectrical constant of a dielectric is different from the dielectric constant of a matrix. Investigation of the optical characteristics of the medium: its index of refraction, absorption, scattering (Rayleigh scattering) for various frequencies of incident radiation. Investigation of this medium as a photon crystal is a search for the "forbidden zones" in the spectrum of the propagating optical radiation. Comparison with the corresponding characteristics of homogeneous media: media consisting only of the substance of the matrix or only of the substance of particles.

2) Creation of a medium consisting of an optically transparent matrix with metallic nanoparticles as well as investigation of its optical properties.

3) Creation of semiconducting nanoparticles interacting in resonant with the electromagnetic field: the interband transition frequency of the semiconductor coincides with the frequency of the electromagnetic radiation. Investigation of the optical properties of one or several nanoparticles, the resonant transition line width including. Comparison with the characteristics of semiconducting macrocrystals. Analogous experiments on nanoparticles of laser mate-

rials, for example, of the NdYAG crystals, glasses with Nd impurity atoms. Comparison with the optical characteristics of separate atoms. The final aim is the creation of a nanoparticle the polarizability of which exceeds the polarizability of a separate atom.

4) Investigation of the optical properties of resonant nanoparticles near metallic particles.

5) Creation of a medium consisting of an optically transparent matrix with resonant nanoparticles as well as investigation of its optical properties.

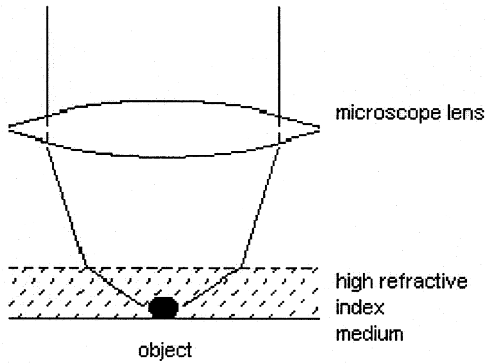

Fig.1 The use of a medium with a high index of refraction for increasing the resolving power of an optical microscope

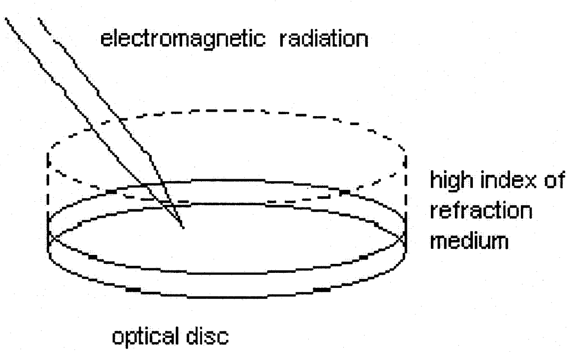

Fig.2 Increasing of the information record density on an optical disc

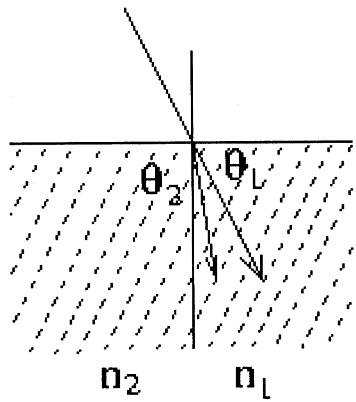

Fig.3 As the refractive index changes from n_1 to n_2 the angle of the electro-magnetic radiation propagation changes from θ_1 to θ_2

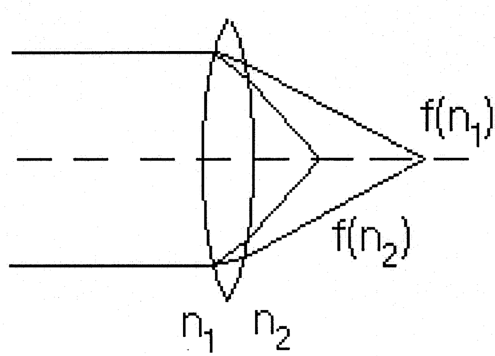

Fig.4. As the refractive index of the lens changes, its focal length also changes

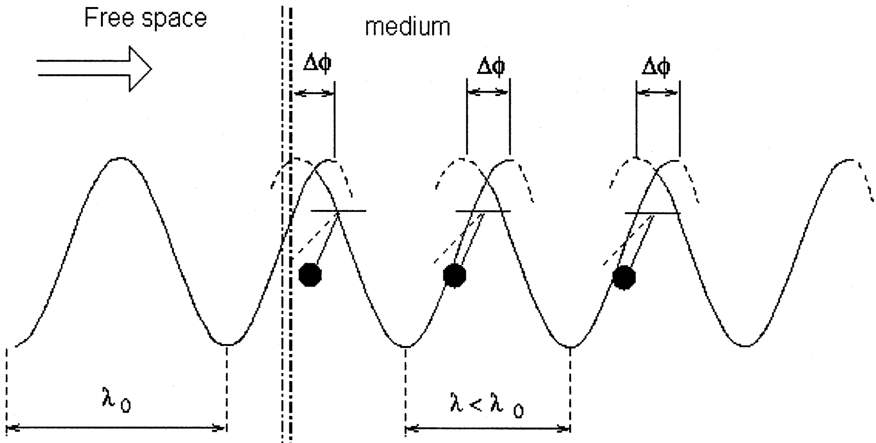

Fig.5 Physical mechanism of emergence of the electromagnetic radiation dispersion in a medium

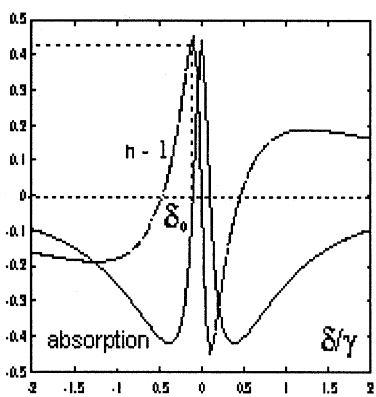

Fig.6 a

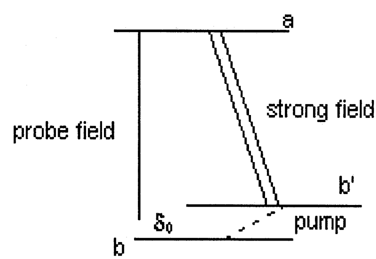

Fig.6 b

(a) Refractive index of a weak resonant probe field and its absorption coefficient as functions of detuning δ from the resonant transition a-b

(b) (b) Three-level Λ-scheme

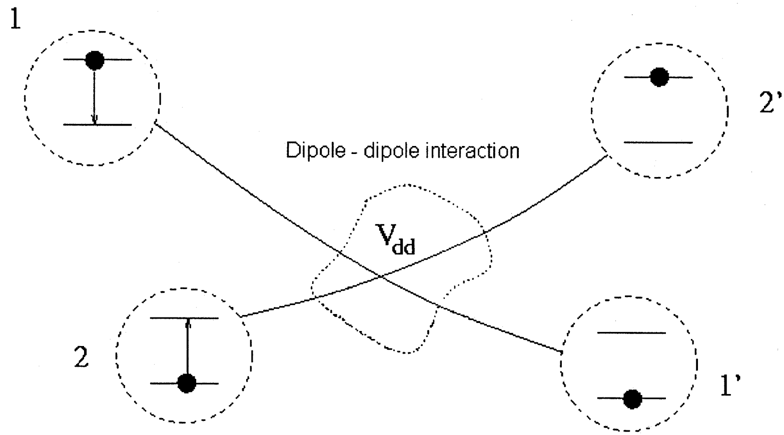

Fig.7 Resonant dipole-dipole interaction of a pair of atoms at the collision leading to self-broadening

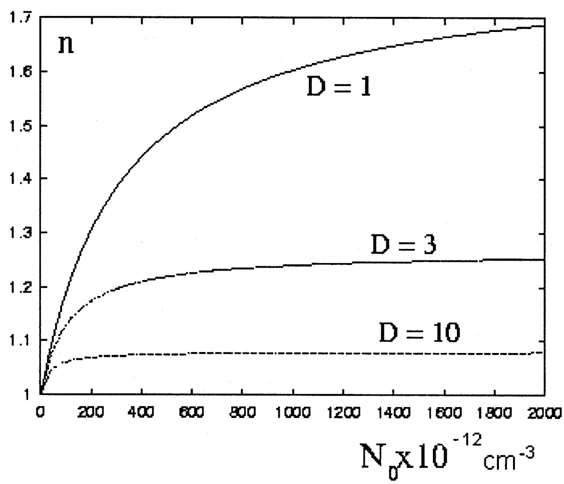

Fig.8 Refractive index of a probe field resonant to the transition a-b of a three-level L-atom (Fig.6.b) as a function of the concentration of atoms at the point of transparency ($\delta = \delta_0$) with consideration of self-broadening but without regard for the local field correction. Self-broadening increases as factor D also increases (see formulas (5), (6)).

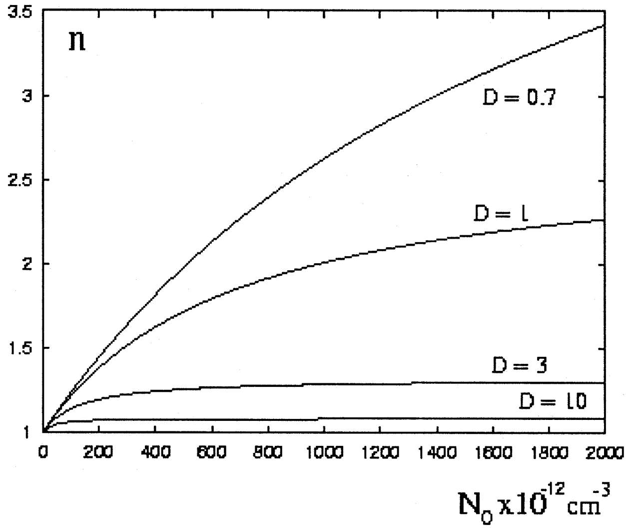

Fig.9 Refractive index of a probe field resonant to the transition a-b of a three-level L-atom as a function of the concentration of atoms at the point of transparency taking into account self-broadening and local field corrections.

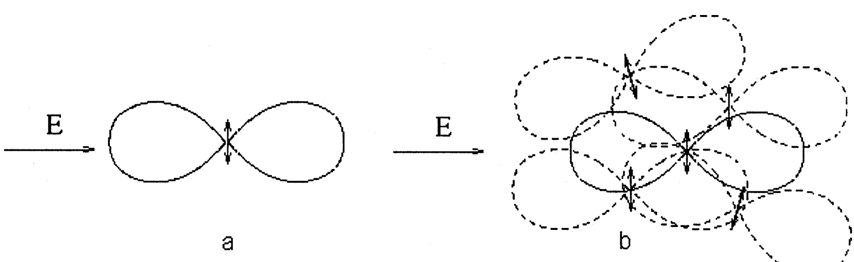

Fig.10. a – radiation of a separate dipole, b – dipole interaction through re-emitted fields

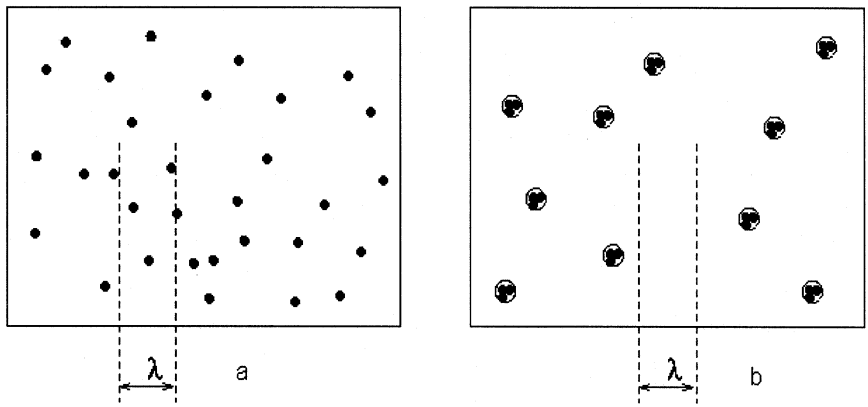

Fig.11 a – basic contribution into self-broadening is made by the atomic interaction at the distance ~ λ.

b – with the concentration of clusters $N_0^{-1/3} >> \lambda$ the probability of the atomic interaction at the distances ~ λ is slight, therefore, self-broadening is less than in Fig.11 a.

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Проценко И.Е., Самойлов В.Н., Займидорога О.А. Д4-2001-90
Суперкогерентные оптические кластерные среды

Обсуждаются возможности создания и свойства принципиально нового типа гетерогенных оптических сред, состоящих из частиц-кластеров размером много меньше длины волны. Каждая из частиц включает большое количество атомов. Предсказывается, что в кластерных средах может возникнуть явление «суперкогерентности» — взаимной фазировки колебаний атомов в резонансном поле. Переход к суперкогерентности — фазовый переход, который может существенно изменить оптические свойства среды. В частности, ее показатель преломления может увеличиться в 10 и более раз.

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Supercoherent Optical Cluster Media

The possibilities of creation as well as the properties of a new type of heterogeneous optical resonant media consisting of cluster particles, the size much smaller than the wave length, are discussed. Each particle includes many resonant atoms. It is predicted that the effect of «supercoherence» might arise in cluster resonant media, which consists of mutual phasing of atomic oscillations in the resonant field. A transition to supercoherence is a phase transition, which can change essentially the optical properties of the medium. Particularly, its refractive index can be increased up to factor 10 and even more.

The investigation has been performed at the Laboratory of Particle Physics, JINR, at the Scientific Center of Applied Researches, JINR, and at the P.N.Lebedev Institute of Physics of RAS, Moscow.

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