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The nature of plaser-powdered laser

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Abstract

A lasing effect in powdered luminophores (plasers) was demonstrated in 1986. It was shown that the width of the spectral line narrows, and the emission signal from polycrystalline neodymium or praseodymium doped samples of a powdered luminophore becomes a series of short, intense pulses when resonant optical pulsed pumping exceeds a threshold level; that a narrow emission line of plasers with shapeless particles is situated at the center of the luminescence band, while the emission of plasers with shaped crystalline seeds can have several narrow components across the line of luminescence; and that in mixtures of powders that have different plaser frequencies, the generation frequency depends on the weighted overlap of the spectra. Lasing mechanisms in powders, such as the amplification of spontaneous emission (ASE) and the effect of the distributed feedback due to scattering are discussed herein. Possible ceramic zeolite-like and cavernous plasers, plasers with nonlinear optical effects, lasing foams and emulsions are considered as devices where the effects of photonic crystals on the generation processes in plasers could be observed. Plaser cathode-ray screens with a narrow band (~0.1 nm) and short duration (~1 ns) of luminescence are suggested.

Keywords: Powdered laser; Plaser; Lasing powder; Lanthanide spectroscopy; Neodymium; Light scattering

1. Introduction

To discuss the nature of plaser or powdered laser [1–4]; let us first remember its history and characteristics in hope that experimental data will throw some light on the mechanisms of generation. Then we'll analyze the most probable mechanisms and discuss possible developments and applications.

2. The history and properties

It was years ago when Varsanyi, looking at tiny luminescent crystals of praseodymium chlorides and bromides, guessed about the possibility of obtaining lasing in powdered luminophores [5]. Three years earlier, in 1968, Letokhov published a theory of lasing excitation in scattering active media [6]. For me the history of the lasing effect in powdered luminophores (plasers) began 14 years ago, in March of 1985, when in the course of some routine experiments – obtaining the excitation spectra of a powdered infrared luminophore, lanthanum oxide doped with neodymium – I observed a strange instability of the luminescence signal. My colleague, Dr. V.M. Markushev, looked into the cause of this instability and observed some irregular, intense pulses of infrared radiation at a high intensity of resonant optical pumping. Three hours later he presented a report on the properties and a simple theory of the first powdered laser. Later it was shown [1–4,7,8] that:

1. the width of the spectral line narrows and the emission signal from polycrystalline neodymium doped samples of a powdered luminophore becomes a series of short, intense pulses when resonant optical pulsed pumping exceeds a threshold level [1–4];
2. a narrow emission line of plasers with shapeless particles is situated at the center of the luminescence band, while the emission of plasers with shaped crystalline seeds can have several narrow components across the line of luminescence [7,8];
3. in mixtures of powders that have different plaser frequencies, the generation frequency depends on the pumping frequency [7,9].

The same dynamic and spectral characteristics of the generation signal were observed [10] in the case of lanthanum oxisulfide doped with praseodymium in the region of the $^3P_0$–$^3F_4$ transition in the spectrum of Pr^{3+}, in conditions of optical pumping in the region of the $^3H_4$–$^3P_1$ transition in the absorption spectrum of the same ions.

The active substance of a plaser could be synthesized in

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the form of a powder or obtained by grinding crystals. The seeds of compounds such as lanthanum oxisulfide synthesized from a melt tend to have a more or less regular form. Seeds of ground powders are as a rule more irregular. When seeds are charged or polarized by friction, they tend to unite in fractal-like clusters. In electron-microscope photograms, shapeless pentasodium-lanthanum-tetramolybdate seeds with sizes in the region of microns look like mountain ranges.

In the excitation spectra of generation, in samples with high concentrations of neodymium, saturation and inversion of the most intense lines, in comparison to the same lines of the spectra of luminescence excitation, were observed [1–3,7]. All this could be attributed to filtering of the pumping radiation and also to a nonlinear dependence of the mean thickness of the generating layer of powder on the doping concentration and on the intensity of the pumping. Use of the same optical waveguide in channels of pumping and indication of the generation signal (the channels are separated by interference filters and mirrors) makes it possible to lower the effects of pumping filtration on the saturation and inversion of lines in the excitation of luminescence spectra, but not in the spectra of generation excitation, where the dependence of the dimensions of the generating volume on the intensity of pumping underlines this inversion, especially at the level of pumping slightly above the generation threshold.

The dependence of the generation frequency of the lasing powder on time was observed in samples of lithium neodymium tetraphosphate and neodymium pentaphosphate [8]. In the case of pentasodium-lanthanum-tetramolybdate powder with small (a few micrometers), shapeless seeds, the generation frequency of the sequence of pulses is constant [7], but in the case of neodymium pentaphosphate [8] powder with relatively robust (30–80 μm) seeds, the dependence of the generation frequency on time in a sequence of pulses can change with the pumping intensity, from constant (‘‘ships sailing in a wake’’) to jumping from pulse to pulse (‘‘a staircase’’) or a linear shift to low frequencies (‘‘ships sailing in a wake under a starboard tack’’). Further, at high pumping power one can observe a few series with different mean frequencies simultaneously. The most probable cause of the time dependence of the generation frequency lies in the change of temperature due to the dissipation of the pumping energy. Several series at the same time could give witness to the excitation of different resonant modes of the seeds.

The next feature of the plasers that should be mentioned here is the dependency of the frequency of generation, in a blend of fractions of the same material with different doping concentrations, on the relative concentration of the components and on the pumping frequency in the region of inversion ‘‘bites’’ from the intense lines of the excitation spectra of generation [7,9]. A shift of the excitation (pumping) frequency from regions of maximal overlap of the excitation spectra of the components increases the contribution of the better pumped component to the frequency shift of the blended plaser. Frequency shifts observed in the spectra of blended plasers witness the fact that many seeds take part in the process of the rise of generation.

3. The present state of art

A group of French scientists who suggested an application for the plaser in a set of laser matches for the ignition of a nuclear fusion reaction obtained the first plaser working at room temperature [11]. Later they evaluated the coherence of the plaser generated radiation by the contrast of the speckle structure, and demonstrated that it was low [12]. We also tried to evaluate the coherence of plaser generated radiation by the same method [13]. Intensity histograms for the speckle pictures of the coherent neodymium-YAG laser, for its second harmonics, and for a dye-laser were obtained, as well as histograms for different subsequent realizations of the generation of powdered lasers. It was demonstrated that there could be different cases. For all cases of generation of a tri-aluminium-lanthanum tetraborate plaser and for most of the cases of generation of a neodymium-pentaphosphate plaser, the radiation has low coherence. For some cases of the neodymium-pentaphosphate plaser the realizations were similar to the histograms of the partly coherent dye laser. Study of the mean contrast of the speckle picture for the neodymium-pentaphosphate plaser gives a relative occurrence of partially coherent radiation, dependent on the pumping power. Those realizations occur at relatively low levels of pumping, where there are one or a few generating aggregates or seeds.

Placing the powdered sample under a horizontal glass slide and using pumping focused at the surface of the sample, we saw in the infrared region irregular ‘‘bursting star’’ pictures of the generation region. In the side view of the same powdered sample pumped through a glass optical waveguide, a ‘‘nebulous’’ distribution of the intensity of the generated light was observed. The absence of homogeneity in these pictures also could indicate some coherence and at the same time could witness non-uniform pumping or dependence on the distribution of seeds.

Lawandy’s group separated the active and scattering roles of the doped seeds of powder in a plaser turning to liquid active media (dye solutions), contributing a high gain and containing tiny passive seeds of titanium dioxide supplying the scattering [14]. That separation gave an opportunity to apply to plasers the theory of photon localization [15] in disordered photonic crystals [16]. Another group, working with passive scattering elements in active media, could even obtain generation from partly ordered nanometer-range scattering fibrous structures – dyed biological tissues [17].

N.E. and M.A. Noginov obtained a convenient low-threshold plaser made of tri-aluminum-lanthanum-tetraborate powder working at room temperature [18]. They also excited lasing in materials doped with titanium or with F-centers [19]. They studied the second harmonic generation of a plaser in a blend of tetraborate with 2-methyl-4-nitro-aniline that has high nonlinear susceptibility [20]. As the efficiency of the generation of the second harmonics was very low, it seems that most of the generated radiation is concentrated within the seeds of the powder. If so, it would be interesting to observe the process of summation of frequency of some short and powerful bearing pulse of infrared radiation with the frequency of the signal of a plaser made of some non-centersymmetric substance such as tri-aluminum-lanthanum-tetraborate. In this case some appreciable signals in the sum-frequency region could be observed and the time-dependence of the generation pulses could be studied by varying the time delay of the bearing pulse.

In a series of pumping pulses, the generation spot in a plaser such as the plaser made of $Al_3Nd(BO_3)_4$ is modified due to the displacement of the particles of powder under the thermosonic action of optical pumping. These displacements could lead to dramatic changes and frustration of the generation. The mechanical and composition variability of plaser frequency could be used in devices with optical memory.

M.A. Noginov observed also a ‘‘sand-lion-pit-for-ants’’ plaser where a pit made in a powdered luminophore by the focused pumping ray lowered the generation threshold [19]. We had observed the same effect on neodymium doped oxo-niobate (La_3NbO_7) ceramics, where pumping light drilled some ‘‘riverbank swallow’s nest’’ caverns that also lowered the generation threshold.

H. Cao demonstrated excitation of a plaser on a broad band-gap semiconductor, ZnO, emitting light in the near ultraviolet region [21].

4. What are the mechanisms of plaser excitation?

At the present time the nature of the mechanism of generation in plasers (superradiance, amplification of spontaneous emission (ASE), effect of distributed feedback due to the scattering) is still unknown for certain, but a mechanism of the generation excitation due to light scattering [6] (photon number multiplication analogous to neutron number multiplication in nuclear reactions or, in other words, ASE in conditions of strong light scattering) can qualitatively explain the properties of plasers. But when one tries to obtain quantitative values of threshold pumping or frequency narrowing of the spectra of plasers, one comes to the conclusion that the results of the calculations using the theory [6] have values too high for both parameters [4,22]. To obtain more plausible values of these parameters, different authors tried to draw in different supplementary mechanisms, such as coupled whispering gallery modes of the same microdroplet with the scatterers [23,24] or the neighboring seeds [4], return of the escaped radiation from the passive to the active scattering medium [22], superradience [25]. In scattering media with quasiperiodic structures, consisting of bulk (balls) or hollow (bubbles) spheroid or cylindric (fibers) elements, randomly distributed feedback [26], or strong Anderson localization of photons in scattering media [15,27] related to photon gap materials [28] (photonic crystals [16]) were suggested.

It seems that the mechanism of plaser excitation depends on the size, form and aggregation of the scattering seeds, varying from the ‘‘normal’’ lasing of microchips (common microlasers) to whispering gallery lasers made of microdroplets or perfectly shaped microcrystals, and to ASE ‘‘lasing’’ in scattering powders consisting of irregular seeds or in microdroplets with the scatterers, and, after all, to partially coherent quasi-photonic crystal lasers due to randomly distributed feedback – randomly distributed quasi-periodic scattering structures in powders. As the powders consisting of seeds united in clusters have some fractal properties [29,30], the conditions for photon localization in them could exist in a wide range of frequencies limited by the inverse sizes of the smallest and the largest of the seeds.

In conditions of a relatively long transversal relaxation time, T_2, (liquid helium temperatures, relatively low concentration of doping ions, perfect crystalline lattice of metal fluorides etc.) superradience was shown to be a main mechanism of plaser generation [25]. In this case the intensity of the generated signal is proportional to the square of the pumping power.

5. Possible developments and applications

Now let us discuss first the most probable future experimental studies of plaser properties and then their possible applications. The photonic crystal properties of scattering media can appreciably influence parameters of plasers such as the threshold values, frequency, linewidth, etc. For the study of these effects, natural (zeolites, layered crystals) or artificial (for example, foams and emulsions that could be ordered or fixed by electric fields or UV) partly ordered media with a low mean density could be used in plasers. Low mean density is needed to obtain resonance at the working frequency of the plaser, between optical Mie modes of the seeds (or bubbles) and the field modes outside them. The demonstration of the influence of photonic crystal properties of scattering elements on the parameters of the plaser can be more successful at the lowest possible working frequency of the plaser. Cavernous plasers could be interesting for studies of this kind.

Many kinds of materials with nonlinear optical properties could be admixed to lasing powders to obtain plasers with nonlinear characteristics, first of all plasers with nonlinear feedback and systems with nonlinear coupling between different elements of a net of plasers. Increasing the size of the optically pumped area, one could study the autowave processes in the active scattering media.

The most obvious applications of plasers are connected with their narrow frequency band and the short (~1 ns) duration of the generated pulses. For example, lasing screens with cathode-ray excitation can appreciably change for the better the characteristics of many contemporary optical systems used for information processing. To make them, one must excite by cathode-ray pumping a luminophore layer, 10–30 microns thick, deposited at the screen. At normal density of the powder the energy of the electron beam must be well above 10 keV, that is too high, raising a problem of heat sink. The development of low density foam luminescent media with photonic crystal properties would make the problem of cathode-ray excitation of photonic laser screen much more hopeful.

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