# Devices for generation and detection of subnanosecond IR and FIR radiation pulses

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### ABSTRACT

Mechanisms of saturation absorption of IR-FIR radiation in semiconductors with degenerate band structure have been investigated. As a result of this with degenerate band structure have been investigated. As a result of this investigation a mode locking device for pulsed IR gas lasers has been developed and investigated. Using p-Ge filter cooled to 78K as saturation absorber inside the laser resonator we obtained subnanosecond IR laser pulses from a tunable high pressure  $CO_2$  laser. These pulses were transferred into FIR from a tunable high pressure  $CO_2$  laser. These pulses were transferred into FIR region via stimulated Raman scattering of  $CO_2$  radiation and as result tunable subnanosecond FIR laser pulses ( $\lambda \sim 300~\mu\text{m}$ ) has been obtained for the first subnanosecond FIR laser can be subnanosecond train of short pulses duration 1 ns and accordingly from FIR laser can semiconductor photoelectric detectors for subnanosecond IR-FIR radiation pulses are also described.

# 2. SATURATION ABSORPTION OF IR-FIR RADIATION IN SEMICONDUCTORS WITH COMPLETE BAND STRUCTURE.

Semiconductors with degenerate band structure change their optical properties with the laser radiation intensity increasing. Nonlinear absorption (saturation) of these materials leads to the nonlinearity of the detectors, influences operation of the FIR semiconductor lasers and gives an opportunity of the nonlinear optic elements construction.

The linear absorption of IR-FIR radiation (  $\hbar_{\omega} \ll_{g}$ ,  $\hbar_{\omega}$  is a photon energy, E<sub>g</sub> is a band gap ) in semiconductors with degenerate band structure is due to the following mechanisms: direct (heavy-to-light hole) intraband transitions, the following mechanisms: direct (heavy-to-light hole) intraband transitions and impurity absorption.

We investigated the mechanisms of the saturation absorption in the case of direct intraband (heavy-to-light subband) transitions. Experimental situation when the direct intraband transitions dominate in absorptions can be realized by choosing a material temperature, density and radiation wavelength. In IR region, where absorption on the lattice oscillation and absorption at nondirect intraband transitions are small, it is so at comparable high temperatures, when impurity levels are ionized. In FIR region the probability of absorption at nondirect intraband transitions rapidly increases with wavelength and begin to play essential role. However, absorption at direct and wavelength and begin to play essential role. However, absorption at therefore the nondirect transitions have opposite temperature dependencies and therefore the first one can be emphasize by choosing a temperature. The absorption on the lattice oscillation in FIR region also can be essential. But their also can be split, as it has not dependence on nonequilibrium carrier density  $\rho$  and intraband absorption has linear dependence on  $\rho$ . The scheme of the direct

intraband transitions gives at Fig.1.

There are two mechanisms of saturation absorption in such case. One of them is related to the dipole moment of the system and it becomes significant when the probability of the absorption of a photon is comparable with the probability of the loss of the dipole moment phase 1,2. This mechanism identifies to the saturation absorption in two levels system, which is well known in the gases as Raby oscillations. In our case, the role of two levels plays the initial  $\epsilon_0$  and final  $\epsilon_f$ states of a hole excited by light. The process of the losses of dipole moment phase in semiconductors can determined by any kind of momentum scattering or influence of the external field.

Another mechanism is concerning with changing of the initial and final states population as a result of the finite energy relaxation rate of the photoexcited carriers. There are two possibilities of it. The

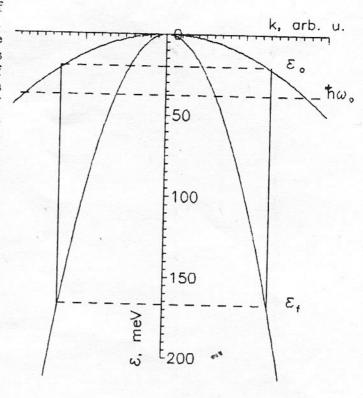


Fig.1

first is "burning" of a dip in the energy population function  $d\rho(E)/dE$  ( $\rho(E)$  is the number of holes with the energy E in the volume unit ) near the energy of the initial state  $E_0$ . It occurs because of the rate of holes excitation to the light absorption exceeds the rate of energy relaxation in the holes system 3-7. To the same result leads shifting of the all population function due to the light heating of holes  $E_0$  (Fig.2). The heating of holes reduces the population of the initial state and, accordingly, absorption coefficient, when  $E_0$  lies in the region of the rising part of the population function ( $E_0$  < 3/2 kT, k is the Boltzmann constant, T is a bulk temperature).

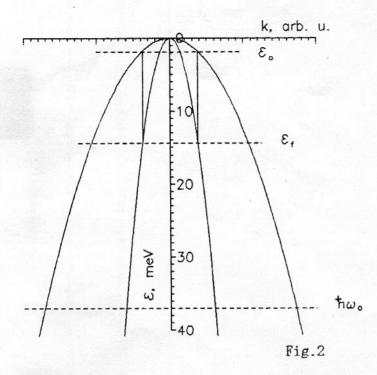
These mechanisms lead to different dependencies of absorption coefficient K on the illumination intensity I, and the net effect is the dependence:

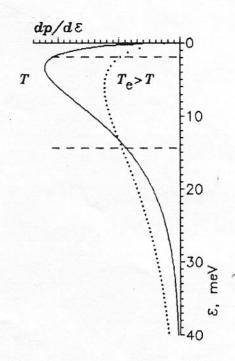
$$K = K_0 / [(1 + I / I_{s2})^{1/2} + I / I_{s1})]$$
 (1)

were  $K_{\mathbf{O}}$  is the absorption coefficient at low intensity,  $I_{\mathbf{S}1}$  =1/ $\tau\sigma_{\mathbf{d}}$  is the saturation parameter associated with the energy relaxation,  $\tau$  is the energy transitions, and  $I_{\mathbf{S}2}$  is the saturation parameter associated with the phase of dipole moment losses.

The saturation absorption of IR-FIR radiation in p-Ge has been investigated in detail in the wide spectral  $\lambda$  (10-340  $\mu\text{m}$ ), temperature (40-340 K) and density  $\rho$  (1012-1017  $_{\text{Cm}}\text{--3}$ ) ranges 4-11

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Our investigations 6-8, 14 show that in IR region ( $\lambda \sim 10 \mu m$ ) the behavior of absorption coefficient describes well by Eq.1 with  $I_{s1} << I_{s2}$ , and, consequently, the second mechanism plays dominate role. It is so because the Ef lies higher than optical phonon energy  $h\omega_0$  (Fig.1.) and dipole momentum phase losses in time shorter than  $10^{-13}$  s. It leads to weak effect deals with Raby oscillations and high value of  $I_{s2}$ . As we mentioned above to saturation

can lead the "burning" of a dip in distribution function and heating of holes gas. As a result of fast several optical phonons emitting main part of the absorbed energy transfers to phonon system and therefore excitation of p-Ge by mid infrared radiation not leads to high electron heating3,6,12. More over lies in the tail of distribution function and temperature influence changing weakly population. So the "burning" of the in distribution function determine the saturation process. Saturation parameter is controlled by most longer energy relaxation time and depend on p and bulk temperature (Fig.3). Curve b (T=78K) shows that there are to parts in this dependence, linear on  $\rho$  at high hole densities, and practically independent on  $\rho$  at low densities. It caused by dependence of energy relaxation time on  $\rho$ . At  $\rho > 10^{14}$  cm<sup>-3</sup> the hole-hole interaction determines energy relaxation

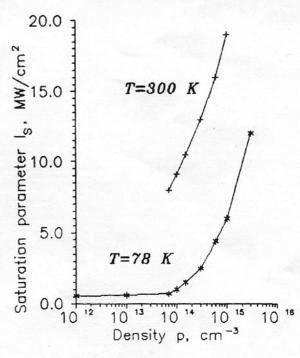


Fig.3

and  $\tau$  linear depends on  $\rho$ . At low densities the energy relaxation is controlled by acoustic phonons and  $\tau$  has not dependence on  $\rho$ . Take note of that as it is shown at Fig.3 changing of bulk temperature and density allows to changing of saturation parameter in a wide range.

At the excitation of p-Ge by powerful far infrared radiation the  $\epsilon_f$  lies lower than optical phonon energy  $h\omega_0$  ( Fig. 2 ) and photoexcited electrons can not lose energy by optical phonons emitting. As a result of it strong electron heating (  $T_e > T$  ) take place. It leads to shifting of the Maxwell-Boltzmann population function ( Fig.2 ) and, consequently, to decreasing of the initial state ( $\epsilon_0$ ) population. In this case the saturation parameter  $I_{s1}$  is determined by an energy relaxation time of distribution function in whole<sup>8</sup>.

The dependence of  $I_{\rm S1}$  on density for p-Ge at 78K and excitation by light with  $\lambda$ =90  $\mu$ m gives at Fig.4. We can see that saturation intensities are lower than in the mid IR region and also depend on  $\rho$ .

Our investigations of saturation absorption in p-Ge in conditions mentioned above have shown that the situation when the nonlinear absorption coefficient describes by Eq.1 with  $I_{\rm S1} << I_{\rm S2}$  is more typical . But it is necessary to note that under certain conditions in p-Ge the behavior of summary absorption coefficient can be differ from Eq.1. It is so when at the same time with the process of saturation multiphoton heavy-to-light transitions 13-14 or linear Drude absorption8,11 are essential. influence of these processes leads to more weaker or opposite dependence of absorption coefficient on intensity at high intensities.

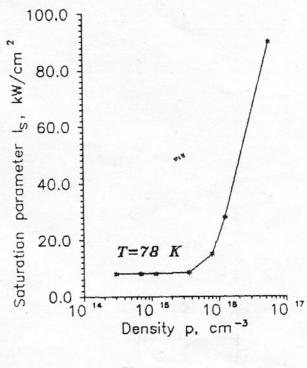


Fig.4

Decreasing of absorption coefficient with intensity increasing can be used to construction of nonlinear optic elements for IR-FIR spectral range. For this purpose it is very important that process of saturation is characterized by low times (  $10^{-9}$ - $10^{-12}$  s. ), take place in a wide spectral range and the saturation parameter  $I_{\rm S}$  can be changed in wide range by means of changing temperature and carrier density. The last one can be changed discrete by  $\rho$  and smoothly by T. On the base of these investigations a reliable mode locking unit for pulse gas IR lasers have been developed and studied. It made possible to achieve stimulated emission of short pulses from a  $\rm CO_2$  laser  $\rm ^{15}$  and generation of subnanosecond FIR pulses.

## 3. LOW-THRESHOLD DEVICE FOR PASSIVE MODE LOCKING OF PULSED INFRARED LASERS.

It is well known that nonlinear absorber placed inside the resonator of pulse laser leads to self mode locking and permits to obtain a train of short pulses. The filter using for this purpose have to satisfy to certain requirements. At first nonlinear absorption have to characterized by short bleaching time. Secondary the values of saturation parameter, linear and nonlinear looses have to correspond to laser gain and lose coefficients. We proposed to use for CO<sub>2</sub> pulse lasers a cooling p-Ge as a nonlinear filter and developed a reliable mode-locking device.

Passive mode locking is achieved by a cooled nonlinear filter (saturable absorber) of p-type germanium, doped with gallium. The broad absorption spectrum permits in principle generation of short pulses in the whole tuning ranges  $\rm CO_2$  laser. At strong excitation this absorption saturates according to the relaxation time of absorption, which lies in the range from  $10^{-10}$ - $10^{-12}$ s and depends on free carriers density and temperature. As it is shown above, decreasing of the filter temperature leads to saturation intensity decreasing Fig.3. As a result of it the mode locking regime can be obtained at lower intensities than at room temperature. Filters having at T=78K the parameters given in Table 1 were used.

TABLE 1

No	p·10-14, I <sub>s</sub> , cm-3 MW/cm <sup>2</sup>		d,	κ,	Kd
			cm	1	
1	1.0	gus 1	3.7	0.036	0.13
2	1.5	1.4	3.7	0.055	0.20
3	2.0	2	3.7	0.075	0.26
4	3.0	2.7	3.7	0.108	0.39
5	8.0	5.2	1.0	0.290	0.29
6	12.0	7.0	1.0	0.420	0.42

For filters of such thickness, it is necessary to take into account interference effects which result in modulation of the transmission as a function of the wavelength with the period  $\Delta \lambda = \lambda^2/2nd$  where n is the refractive index and d is the thickness of the filter. The transmission varies between  $[(1-R)/(1+R)]^2$  and 1 (R is the reflection coefficient of a single face). In order to minimize reflection losses, antireflection coating, comprising a combination of ZnS-Ge layers, were evaporated on faces of the filter. The reflection coefficient of a single face was less than 2%. The coatings permit to stand many times cooling to T=78K at a rate 20 K/min. The optical damage threshold of the coatings for  $CO_2$  laser pulses of 100 ns duration was ~15 MW/cm². We have mounted the germanium plate in an evacuated chamber with ZnSe Brewster windows Fig.5. The chamber permits to cool filter up to liquid nitrogen temperature. This device, which is described in  $^{15}$ , was placed inside the laser resonator close to the output coupler.

Our laser  $^{16}$  (UV preionized, grating tuned, 20 atmospheres  $\mathrm{CO}_2$  laser), with

a setup shown in Fig.5, produces 150 ns pulses with a linewidth of about 5 GHz (FWHM). The laser output coupler is a concave germanium mirror with a radius of 25 m and a reflectivity of 0.7.

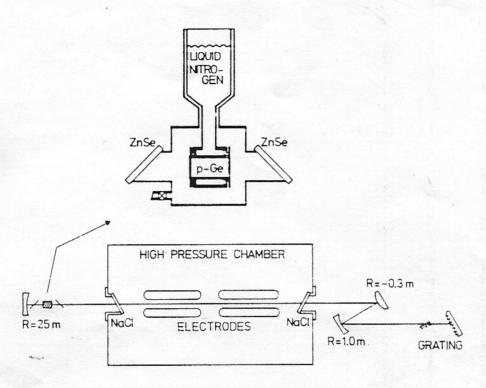


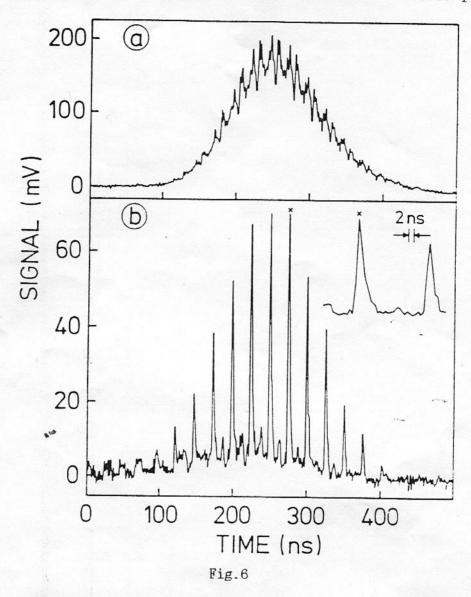
Fig.5

For pulse detection, fast detectors with response times less than 1 ns were used. A small part of the CO<sub>2</sub> pump pulse was reflected by a BaF plate on a infrared photon drag detector (Technoexan PD-10M). The signals obtained from detector was amplified (bandwidth of the amplifiers 1 GHz) and then recorded with a dual channel digital storage oscilloscope (LeCroy 7200, sampling rate 1 GS/s, input bandwidth 400 MHz).

In Fig.6 typical CO<sub>2</sub> laser pulse shapes are shown without (a) and with (b) the mode locking unit. Passive mode locking produces short pulses, consisting of one or several pulse trains with a period of 23 ns, according to the round trip time in the laser. The temporal shape of a single pulse is shown on an expanded time scale in the inset of Fig. 6b. The intensity of the single peaks must have been high compared to the pulse intensities obtained without the saturable absorber (1.8 MW for a 250 mJ pulse). We conclude this from the observation that damage of the NaCl Brewster windows of the high pressure tube occurred for pulse energies above 50 mJ when the saturable absorber was inside the resonator while this damage occurred without saturable absorber only for pulse energies of more than 300 mJ. For filters with lower concentrations than 8·10<sup>14</sup> cm<sup>-3</sup> gallium more single pulses at a period of 23 ns and less contrast between the pulses were obtained. For higher concentrations, laser action vanished.

We also used the radiation of described CO2 laser with passive

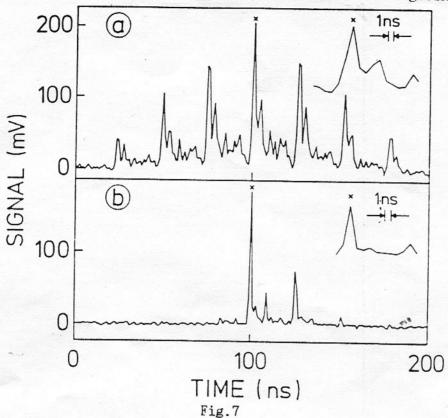
mode-locking unit for generation of short FIR pulses (Fig.7). Some years ago it was demonstrated 17, that tunable picosecond pulses in the far-infrared (FIR) can be generated by parametric interaction of two visible pulses. In



this paper we report for the first time generation of tunable subnanosecond pulses via stimulated Raman scattering of infrared pulses. As Raman laser medium we used CH<sub>3</sub>F gas that is well suitable for tunable generation of FIR radiation by stimulated Raman scattering 16-21; we note that generation of subnanosecond FIR pulses at fixed frequencies has been performed using a mode locked TEA CO<sub>2</sub> laser as pump radiation source<sup>22</sup>. In our paper we show that optical pumping of CH<sub>3</sub>F with a passive mode locked high pressure CO<sub>2</sub> laser leads to tunable FIR pulses.

Infrared pulses were focused into a FIR waveguide laser (Fig.5), length 115 cm, containing 11 mbar  $\text{CH}_3\text{F}$ , with a BaF entrance and a TPX output window 16. To block the remaining  $\text{CO}_2$  laser radiation passing the FIR laser, a plate of crystalline quartz was used.

For generation of the short FIR pulse we made use of Raman transitions in the R(19) tuning branch of  $\text{CH}_3\text{F}$ , which has found to be the most effective one  $^{16}$ . The highest pulse energies (about 100  $\mu\text{J}$ ) were found, as without the saturable absorber, for a pump frequency of 1078 cm $^{-1}$ , causing FIR laser



emission at a frequency of 33 cm-1. With mode locking unit the radiation, emitted from the FIR laser, consists - as the pump radiation - of a train of short pulses. As there is a certain threshold intensity needed for FIR generation and stimulated Raman scattering is a nonlinear process, there are less and shorter pulses in the FIR emission than in the CO2 pump radiation. An example is presented in Fig.7, where both, a pump (a) and the corresponding FIR laser pulse (b) are shown; FIR laser action occurs only for the pump pulses with the highest intensity. For detection of such short FIR pulses the detector (Technoexan PD-3), was used. This detector will be described in section 4. Single CO2 laser and FIR pulses are shown on an expanded time scale as insets of Fig.7. While the CO2 laser pulse has a measured duration of 4 ns (FWHM) the duration of the FIR pulse has a measured duration of 1 ns. Taking into account the bandwidth of the measuring system consisting of detectors and electronics we find that the true width of the FIR pulses must be below 1 ns. Since the frequency of the FIR radiation can be tuned by tuning the emission frequency of the high pressure laser 16-21, frequency tunable short intense pulses can be generated in the FIR region by stimulated Raman scattering. We should note that the pulse generation in both the mode locked CO2 laser and the FIR Raman laser are not reproducible with to pulse sequence and pulse intensity - as it is typical for such highly nonlinear processes 17. A better controlled laser action of the high pressure CO2 laser should improve the reproducibility.

## 4. DEVICES FOR DETECTION OF SUBNANOSECOND IR AND FIR RADIATION PULSES.

Until lately, the main practical requirements to photodetectors were high sensitivity and the limit of radiation detection. Demand for photodetectors of a new type has arose with the development of high-power pulse laser sources and the necessity of measuring and controlling the radiation parameters. Sensitivity for such photodetectors is less important than: high time resolution, wide dynamic range, absence of cooling, etc.

Uncooled photon-drag photodetectors based on p-Ge appear to satisfy the necessary requirements perfectly for IR infrared radiation. This effect  $^{23-24}$  has an extremely low transit time (up to  $10^{-12}\mathrm{s}$ ) and a wide dynamic range of measurements. For detection of FIR radiation pulses p-Ge detectors are not well due to two reasons: one of them is a double sign inversion with wavelength increasing  $^{25-26}$ , which leads to low signals in the ranges around 50  $\mu$ m and 200  $\mu$ m, and another one is inversion of sign of photon-drag effect with FIR radiation intensity increasing due to intraband multiphoton transitions  $^{13}$ , which leads to dynamic range limitation. These phenomenons are due to complete band structures of p-Ge and consequently presence of direct intraband transitions.

We proposed to use for detection of FIR radiation pulses n-Ge which has kept advantages of p-Ge detectors. Since in n-Ge due to simple band structure the disadvantages of p-Ge detectors mentioned above are absent. At the same time the sensitivity, which for such detectors is very small in IR region, rapidly increase with wavelength due to frequency dependence of nondirect transitions absorption coefficient. As a result of it the sensitivity of this detectors in FIR region is comparable or essential higher than the same characteristic of p-Ge photon drag detectors in mid IR region. Oscillogrames show the time resolution of such detectors and p-Ge detectors for IR radiation are given at Fig.7,6 correspondently.

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### 6. REFERENCES

- 1. L.D.Landau, E.M.Lifshitz, Quantum mechanic, Nonrelativistic Theory, Pergamon, 1978.
- 2. D.A.Parshin, A.R.Shabaev, "Theory of Nonlinear IR Absorption by Semiconductor with Degenerate Bands," Sov. Phys. JETP, 65, 827-834, 1987.
- 3. V.L.Komolov, I.D.Yaroshetskii, I.N.Yassievich, "Bleaching Associated with Intraband Transitions in Semiconductors," Sov. Phys. Semicond., 11, pp.48-52, 1977
- 4. A.F.Gibson, C.A.Rosito, C.A.Raffo, M.F.Kimmitt, "Absorption Saturation in Germanium, Silicon and GaAs at 10.6μm," Appl. Phys. Lett., 21, p.356, 1972.
- 5. P.J.Bishop, A.F.Gibson, M.F.Kimmitt, "Absorption Saturation in p-Type Germanium," J. Phys. D: Appl. Phys., 9, pp. L101-L103, 1972.
- 6. E.V.Beregulin, P.M.Valov, I.D.Yaroshetskii, "Experimental Investigation of the Bleaching of a Semiconductor in the Case of Optical

Heating and Cooling of Electrons Involving Intraband Transitions," Sov. Phys. Semicond., 12,pp. 138-140, 1978.

E.V.Beregulin, S.D.Ganichev, I.D. Yaroshetskii, I.N. Yassievich Mechanisms of Energy Relaxation Under Conditions of Nonlinear Absorption of

Light in p-Type Ge," Sov. Phys. Semicond., 16, pp.179-182, 1982.

8. E.V. Beregulin, S.D. Ganichev, K. Yu. Glookh, I.D. Yaroshetskii, "Nonlinear Absorption of Submillimeter Radiation in Germanium due to Optical Heating of Charge Carriers, Sov. Phys. Semicond., 21, pp.615-618, 1987.

9. F.Keilmann, "Infrared Saturation Spectroscopy in p-Type Germanium,"

IEEE J. Quantum. Electr., QE-12, 592-597, 1976.

10. C.R. Phipps , S.J. Thomas, "Saturation Behavior of p-Type Ge at CO2

Laser Wavelength, Opt.Lett., 1, pp.93-95, 1977.
11. R.Till, F.Keilmann, Saturation of the Far Infrared Intervalence Band Transition in p-Germanium," Fourth Int. Conf. on infrared physics, R.Kesselring and F.K.Kneubuhl, pp., 636-637, Zurich, 1988.

12. I.N. Yassievich I.D. Yaroshetskii, "Energy Relaxation and Carrier Heating and Cooling Processes in the Intraband Absorption of Light in

Semiconductors, Sov. Phys. Semicond., 9, pp. 565-570, 1975.

13. S.D. Ganichev, S.A. Emel yanov, E.L. Ivchenko, E. Yu. Perlin, Ya. V. Terent ev A.V. Fedorov, I.D. Yaroshetskii, "Multiphoton Absorption in Semiconductors at Submillimeter Wavelength, "Sov. Phys. JETP, 64, pp.729-737, 1986.

14. E.V. Beregulin, S.D. Ganichev, I.D. Yaroshetskii, "Nonlinear Absorption

of CO2 Laser Radiation in p-Germanium, Sov. Phys. Semicond., 20, 1986.

E.V.Beregulin, P.M. Valov, S.D. Ganichev, Z.N. Kabakova, I.D.Yaroshetskii, "Low-Threshold Device for Passive Mode Locking of Pulsed Infrared Lasers," Sov. J. Quantum Electron., 12, 175-178, 1982.
16. U.Werling, K.F.Renk, Wan Chong-Yi, "Tuning Characteristics of a High

Pressure CO2 Laser Pumped CH3F Raman Laser," Int. J. Infrared Millimeter

Waves, 7, 881-891, 1986.

17. M.Berg, C.B.Harris, T.W.Kenny, P.L.Richards, "Generation of Intense Tunable Picosecond Pulses in the Far-Infrared, Appl. Phys., 47, p. 206, 1985.

- 18. D.G.Biron, B.G.Danly, R.J.Temkin, B.Lax, "Far Infrared Raman Laser with High Intensity Laser Pumping," IEEE J. Quantum Electron. QE-17, pp. 2146-2152, 1981.
- 19. B.G.Danly, S.G.Evangelides, R.J. Temkin, B.Lax, "Frequency Tuning and Efficiency Enchancement of High-Power Far-Infrared Lasers," Infrared and Millimeter Waves, Ed.K.J.Button, 12, pp. 195-275, 1984.
- 20. B.G.Danly, S.G.Evangelides, R.J.Temkin, B.Lax, "A Tu-Infrared Laser," IEEE J. Quantum Electron., QE-20, pp. 834-837, 1984.
- Far-Infrared Laser," IEEE J. Quantum Electron., QE-20, pp. 834-83 21. P.Mathieu, J.R.Izatt, "Continuously Tunable CH3F Raman Far-Infrared Laser," Opt. Lett., 6, pp. 369-372, 1981.

- 22. S.H.Lee, S.J.Petuchowski, A.T.Rosenberg, T.A.DeTemple, Opt. Lett. 4, p.6, 1979.
- 23. A.M. Danishevskii, A.A. Kastal'skii, S.M. Ryvkin, I.D. Yaroshetskii, Drag of free carriers by photons at direct interband transitions in Semiconductors, "Sov. Phys. JETP, 31, pp.292-297, 1970. 24. A.F.Gibson, M.F.Kimmitt, A.C. Walker, "Photon Drag in Germanium,"

Appl. Phys. Lett., 17,pp.75-77, 1970.

25. A.F.Gibson, M.F.Kimmitt, " Photon Drag Detection," Infrared and

Millimeter Waves, Ed.K.J.Button, 3, pp. 189-193, 1980.

26. S.D.Ganichev, S.A.Emel'yanov, I.D.Yaroshetskii," Spectral Sign Inversion of Photon Drag at FIR Wavelength," Sov. Phys. JETP Lett., 35, pp.368-370, 1982.

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