Electron Beam Sustained Plasma as a Medium for Amplification of Electromagnetic Radiation in Subterahertz Frequency Band

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Abstract: It is demonstrated that low-temperature weakly-ionized nonequilibrium plasma created by a high-energy electron beam in gases (gas mixtures) with Ramsauer minimum in a transport cross section and effective attachment of slow electrons can be used as a medium for amplification and generation of electromagnetic radiation in subterahertz frequency band. Analysis of the electron energy distribution function (EEDF) in Xe – F_2 mixture is performed. Energy interval with growing EEDF is found to exist in such a mixture. Such an interval provides the existence of population inversion of the electron spectrum in continuum and is responsible for positive value of a gain factor of microwave radiation. A gain factor depending on an amplified radiation frequency and plasma parameters is analysed.

1 INTRODUCTION

The sources of terahertz (THz) and subterahertz radiation are of significant interest in a number of practical applications, in particular, in chemistry (Skinner, 2010), molecular biology (Meister et al., 2013), medicine (Titova et al., 2013), materials science (Grady et al., 2013). The interest in terahertz and subterahertz radiation and its possible applications is caused by its ability to penetrate through a lot of materials (Jepsen et al., 2011), which are usually opaque in the infrared and visible ranges. Also, a number of physical and chemical processes can be controlled and governed by such a low frequency radiation.

Recently (Bogatskaya and Popov 2013; Bogatskaya et al., 2014; Bogatskaya et al., 2015) propose to use plasma channels created in gas by powerful femtosecond UV laser pulse for amplification of the electromagnetic radiation in subterahertz frequency band. To obtain the effect of amplification one needs to have a population inversion in the system. Such a population inversion obviously arises in the process of multiphoton ionization of gaseous media by a powerful laser pulse of femtosecond duration. Really, in this case the pulse duration is less than the time interval between the elastic electron – atomic collisions and, hence, photoelectron spectrum consists of a number of peaks corresponding to the absorption of a certain number of photons. Such a spectrum has energy ranges characterized by population inversion. It was demonstrated that the regime of amplification of microwave radiation can be achieved only in a gas with the energy interval of growing with energy transport cross section (Bekefi et al., 1961; Bunkin et al., 1973). In this case positive value of a gain factor appears to exist if the position of energy peak in the photoelectron spectrum is located within the energy range with increasing transport cross section (Bekefi et al., 1961; Bunkin et al., 1973) at time intervals corresponding to the relaxation of the of electron energy distribution function (EEDF). It was found that among different atomic and molecular gases that can be used for amplification xenon has some advantages due to the large value of transport cross section with a pronounced Ramsauer minimum, large atomic mass and also absence of excited levels in the energy interval where the photoelectron peak is formed. For the microwave frequency $\omega = 5\times10^{11}$ s^{-1} a gain factor $k_\omega \approx 0.01$ cm^{-1} can be reached for the durations of tens of

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nanoseconds (Bogatskaya et al., 2015). The possibility to amplify radio-frequency (RF) radiation in nonequilibrium plasma was firstly experimentally proved in (Okada and Sugawara, 2002).

It was mentioned by (Bogatskaya and Popov, 2013) that the effect of amplification of electromagnetic radiation in a plasma channel created by an intense femtosecond laser pulse is close from physical point of view to the effect of negative absolute conductivity in a gas discharge plasma predicted by (Rokhlenko, 1978; Shizgal and McMahon, 1985), experimentally detected by (Warman et al., 1985), and discussed in detail in reviews (Aleksandrov and Napartovich, 1993; Dyatko, 2007). In (Dyatko, 2007) different physical situations leading to the possibility of absolute negative conductivity to appear are analyzed. Among them is the non-self-sustained steady-state discharge supported by high energy electronic beam in a gas mixture with an efficient attachment of slow electrons and pronounced Ramsauer minimum in transport cross section. Such a situation can be realized, for example, in Ar:CCl_4 (Dyatko et al., 1987), Ar:F_2 (Rozenberg et al., 1988) or Xe:F_2 (Golivinskii and Shchedrin, 1989) mixtures.

In this paper we apply the analysis based on the Boltzmann kinetic equation in two-term approximation to study EEDF in a plasma in Xe:F_2 sustained by a high-energy e-beam in the presence of a microwave field. The possibility to use such plasma as a medium for RF field amplification is studied in dependence on mixture parameters (partial concentrations of mixture species, electronic density) as well as frequency and intensity of RF radiation.

2 BOLTZMANN EQUATION FOR THE EEDF IN A PLASMA SUSTAINED BY HIGH-ENERGY E-BEAM

The energy spectrum of electrons in a plasma $f(\varepsilon,t)\sqrt{\varepsilon}$, normalized according to the condition

$$\int f(\varepsilon,t)\sqrt{\varepsilon}\,d\varepsilon=N_e(t) \qquad (1)$$

(N_e is the electron density) was analyzed using the kinetic Boltzmann equation in the two-term approximation (Ginzburg and Gurevich, 1960):

$$\frac{\partial f(\varepsilon,t)}{\partial t}\sqrt{\varepsilon}=\frac{\partial}{\partial\varepsilon}\left(\frac{e^2E_0^2\nu_{tr}(\varepsilon)}{3m(\omega^2+\nu_{tr}^2)}\varepsilon^{3/2}\frac{\partial f}{\partial\varepsilon}\right)+ \sum_i\frac{2m}{M_i}\frac{\partial}{\partial\varepsilon}\left(\nu_{tr}^{(i)}(\varepsilon)\varepsilon^{3/2}\left(f+T_g\frac{\partial f}{\partial\varepsilon}\right)\right)+ Q^*(f)+Q_b(\varepsilon). \qquad (2)$$

Equation (2) has the form of the diffusion equation in an energy space. Here, E_0 and ω are the amplitude and frequency of the amplified RF field, T_g is the gas temperature (below, we take $T_g\approx 0.03$ eV), m is the mass of the electron, M_i ($i=1,2$) are ~~the~~ masses of the xenon atom and fluorine molecule respectively, $\nu_{tr}^{(i)}=N_i\sigma_{tr}^{(i)}(\varepsilon)\sqrt{2\varepsilon/m}$ is the partial transport frequency, where $\sigma_{tr}^{(i)}(\varepsilon)$ is the transport scattering cross section for Xe (i=1) and F_2 (i=2) molecule, $\nu_{tr}=\sum\nu_{tr}^{(i)}$ is the total transport frequency, N_1 and N_2 are-concentrations of Xe and F_2 in a gas mixture respectively, $Q_b(\varepsilon)$ is the rate of slow electron production by a high-energy electron beam, $Q^*(f)$ is the integral of inelastic collisions. These integrals are described in detail in the review (Ginzburg and Gurevich, 1960). Further we will assume that excitation of vibrational and electronic levels of fluorine molecules do not contribute to the EEDF evolution. This is possible if relative concentration of fluorine molecules satisfies ~~to~~ the inequality

$$\alpha=N_2/N_1<<(2m/M_1)\frac{\sigma_{tr}^{(1)}}{\sigma^*}\frac{\langle\varepsilon\rangle}{I^*} \qquad (3)$$

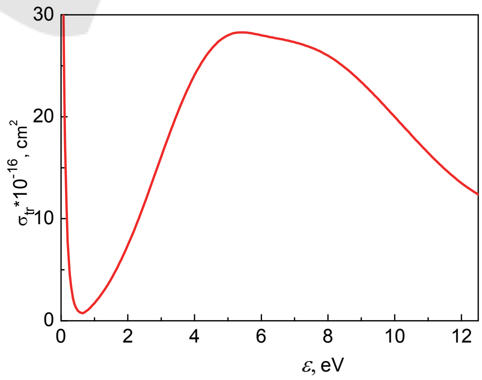

Figure 1: Transport cross section of xenon atoms.

Here σ^* is the cross section of excitation of a F_2 vibrational level with excitation energy $I^*\approx 0.1$ eV, $\langle\varepsilon\rangle$ is the averaged over EEDF electronic energy. If we assume that $\langle\varepsilon\rangle \approx 1$ eV and $\sigma_{tr}^{(1)}/\sigma^* \approx 10$ we obtain that (3) is fulfilled for the partial fraction of F_2 molecules in xenon being rather small, $\alpha << 10^{-3}$. Total transport cross section in such a mixture is very close to the partial transport cross section for Xe atoms (see fig. 1). This cross section was taken from (Hayashi, 1983). The most important thing for our consideration is the existence of energy interval with the positive derivative $d\sigma_{tr}/d\varepsilon > 0$ in the range of $0.6 \div 5.0$ eV.

The term Q_b is the source of slow electrons produced by fast electron beam in the ionization process. According to (Dyatko, 2007) it is chosen in a form

$$Q_b(\varepsilon) = q \times \frac{2}{I_{Xe}}\left(1 - \frac{\varepsilon}{I_{Xe}}\right) \qquad (4)$$

in the energy range of $0 < \varepsilon < I_{Xe}$ (I_{Xe} is the ionization potential of xenon atom) and $Q_b(\varepsilon) = 0$ outside this interval.

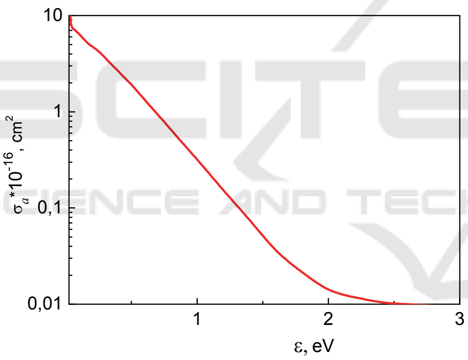

Figure 2: Cross section of electron attachment of F_2 molecule.

The energy spectrum of produced electrons is normalized according to

$$\int Q_b(\varepsilon)d\varepsilon = q \qquad (5)$$

So, q is the production rate, reflecting ionization of atoms by fast electrons. As the electron – electron collisions provide the tendency for maxwellization of the EEDF, the production rate q was chosen in a way to exclude contribution of ee-collisions to the formation of an electron energy spectrum. For Xe plasma it is possible only for ionization degree $\alpha = N_e/N \le 10^{-7}$ (Bogatskaya et al., 2013).

For low partial fraction of F_2 in a gas mixture only the following processes were taken into account: the elastic scattering of electrons on Xe atoms and dissociative attachment of electrons to F_2 molecules. Cross section for electron attachment to F_2 molecules was taken from (Morgan, 1992). This cross section is presented at fig. 2. Integration of the equation (2) over energy allows to obtain the equation for the electron density in the plasma:

$$\frac{dN_e}{dt} = q - \nu_a(t)N_e \qquad (6)$$

where

$$\nu_a(t) = \sqrt{2/m}\, N_{F_2}\int \sigma_a(\varepsilon) f(\varepsilon,t)\varepsilon d\varepsilon \qquad (7)$$

Here N_{F_2} is the concentration of F_2 molecules and $\sigma_a(\varepsilon)$ is the cross section for electron attachment.

For numerical solution the EEDF was presented in a form $f(\varepsilon,t) = N_e(t)n(\varepsilon,t)$, where the function $n(\varepsilon,t)$ is normalized to unity: $\int n(\varepsilon,t)\sqrt{\varepsilon}d\varepsilon = 1$. The Boltzmann equation for $n(\varepsilon,t)$ and eq. (6) for electronic density were solved self-consistently using an explicit scheme in the energy range $\varepsilon = 0 - 12.5$ eV. Steps of integration in space and time domain were the same as in (Bogatskaya and Popov, 2013). Steady-state EEDF was obtained as a result of temporal evolution of the initial Maxwellian function with given temperature T_0 and electronic density $N_e(t=0)$. Calculations were performed for the gas mixture Xe:F_2 = $10^{-5} \div 10^{-4}$, gas pressure 4 atm ($N = 10^{20}$ cm^{-3}) and $q = 2 \div 8 \times 10^{17}$ cm^{-3} s^{-1}. Initial electron density was obtained from the stationary solution of eq. (6) with initial Maxwellian EEDF (with temperature T_0=1.0 eV) used to calculate the rate of attachment (6). Under considered conditions given q value is provided by the electron beam with the following characteristics (Cason et al., 1977): current density ~ 100 mA/cm^2 and energy of the beam ~ 300 keV.

Results of simulation of the EEDF in the Xe:F_2 mixture for the total concentration $N = 10^{20}$ cm^{-3} and the concentration of F_2 at a level of 4×10^{15} cm^{-3} and electron production rate $q = 8\times10^{17}$ cm^{-3} s^{-1} are presented at fig.3 for different time intervals from the initial instant of time corresponding to the Maxwellian distribution. First we note that for given conditions the relaxation time of the final steady-state distribution is about 200 ns. The obtained distribution has the energy range (~0.4 – 0.8 eV) with population inversion that appears as a result of slow electron losses in the attachment process, while the electron production mainly takes place in the energy range above 1 eV. The electron density corresponding to the given conditions is $N_e = 7.11\times10^{10}$ cm^{-3}.

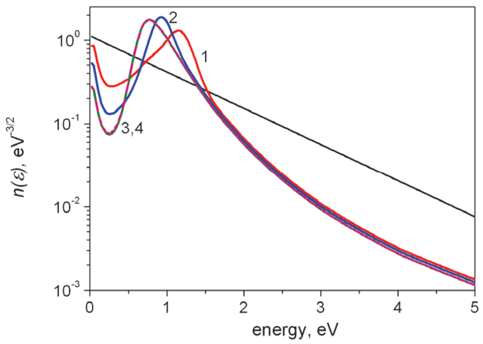

Figure 3: EEDF in Xe:F_2 plasma for different instants of time (ns): t=0 (1), 25 (2), 50 (3), 200 (4), 400 (5); Gas density is $N = 10^{20}$ cm^{-3}, fluorine concentration is N_{F2} = 4×10^{15} cm^{-3}, production rate is $q = 8\times10^{17}$ cm^{-3} s^{-1}.

The set of steady-state EEDFs and plasma parameters obtained for different F_2 molecule concentrations and different electron production rates q are presented at fig.4 and in the table 1. It can be seen that the increment of the F_2 concentration for a given q and vice versa the decrement of q for a given F_2 concentration results in electron density reduction. As about the EEDF it was found that it is the same one for any value of electron production rate q used for the modeling. Definitely, the reduction of the electron density will result in the decrement of a plasma gain factor. On the other hand, the less the plasma electron density, the higher is the position of the peak in energy spectrum. Later we will see that this fact allows to increase the frequency band of amplified RF radiation.

Table 1: Electron densities and EEDF peak position in the e-beam sustained discharge.

q , cm^{-3} c^{-1}	F_2 concentration, cm^{-3}	electron density, cm^{-3}	electron peak position, eV
8×10^{17}	2×10^{15}	1.10×10^{11}	0.68
8×10^{17}	4×10^{15}	7.11×10^{10}	0.78
8×10^{17}	8×10^{15}	4.89×10^{10}	0.88
8×10^{17}	1.6×10^{16}	3.44×10^{10}	1.00
4×10^{17}	8×10^{15}	2.45×10^{10}	0.88
2×10^{17}	8×10^{15}	1.22×10^{10}	0.88

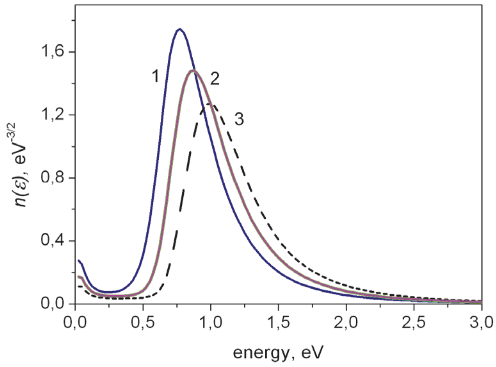

Figure 4: Steady-state EEDF in Xe:F_2 plasma for any value of the production rate and concentrations of F_2 molecules (cm^{-3}) 4×10^{15} (1), 8×10^{15} (2) and 1.6×10^{16} (3).

To provide more insight to the EEDF formation in Xe:F_2 plasma beam we will perform simple qualitative analysis of the EEDF in a beam-sustained plasma. Neglecting the contribution of transported RF field to eq. (2) as well as the term with non-zero value of the gas temperature, under above formulated assumptions the Boltzmann equation for the steady-state EEDF can be written in a form

$$\frac{2m}{M}\frac{d}{d\varepsilon}(\nu_{tr}(\varepsilon)\varepsilon^{3/2}n(\varepsilon)) - \nu_a(\varepsilon)n(\varepsilon)\sqrt{\varepsilon} + Q_b(\varepsilon)/N_e = 0. \tag{8}$$

Here M and ν_{tr} are the mass of Xe atom, and the transport frequency for electron scattering on Xe atoms, $\nu_a(\varepsilon) = N_{F_2}\sigma_a(\varepsilon)\sqrt{2\varepsilon/m}$ is the attachment frequency. Introducing new function

$$F(\varepsilon) = \frac{2m}{M}\nu_{tr}^{(Xe)}(\varepsilon)\varepsilon^{3/2}n(\varepsilon) \tag{9}$$

one derives

$$\frac{dF(\varepsilon)}{d\varepsilon} - A(\varepsilon)F(\varepsilon) = -Q_b(\varepsilon)/N_e \tag{10}$$

where $A(\varepsilon) = (M/2m)(\nu_a/\nu_{tr})/\varepsilon$. As the attachment process takes place only for slow electrons with energy $\varepsilon < \varepsilon^*$, we obtain for $\varepsilon > \varepsilon^*$

$$F(\varepsilon) = F^* - \frac{1}{N_e}\int_{\varepsilon^*}^{\varepsilon} Q_b(\varepsilon)d\varepsilon \tag{11}$$

(here $F^* = F(\varepsilon^*)$) which results in the parabolic dependence of F on energy. On the other hand, in low energy range $\varepsilon < \varepsilon^*$ the attachment process is of the most importance and solution of eq.(2) can be approximately expressed in the form

$$F(\varepsilon) = F^* \exp\left(-\int_{\varepsilon}^{\varepsilon^*} A(\varepsilon) d\varepsilon \right) \quad (12)$$

In the assumption that $A(\varepsilon) \approx A_0 = const$ in the low energy limit one obtains from (12)

$$F(\varepsilon) = F^* \exp(-A_0(\varepsilon^* - \varepsilon)) \, . \quad (13)$$

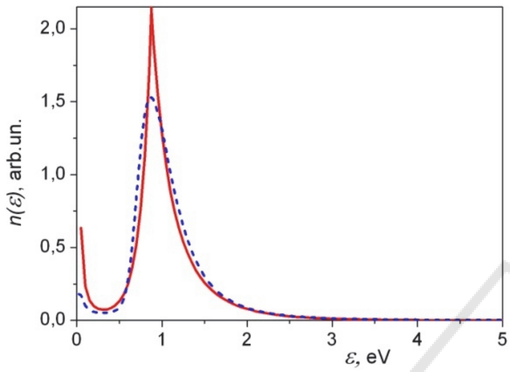

Figure 5: Analytical (solid curve) and numerical (dash curve) calculations of the EEDF in Xe:F_2 plasma sustained by an electron beam. See the text for details.

The electron spectrum $n(\varepsilon) \sim F(\varepsilon)/v_{tr}(\varepsilon)\varepsilon^{3/2}$ obtained from (11) and (13) for $A_0 = 10$ eV^{-1} and $\varepsilon^* = 0.85$ eV is presented in a liner scale at fig.5 and is found to be in a good agreement with the results of numerical simulations also displayed at the same figure.

3 AMPLIFICATION OF THE RF RADIATION IN A PLASMA

It is known that an absorption coefficient μ_ω (or gain factor $k_\omega = -\mu_\omega$) of electromagnetic radiation with frequency ω in a plasma with the EEDF $n(\varepsilon)$ is given by the expression (Bunkin et al., 1973; Ginzburg and Gurevich, 1960; Raizer, 1977):

$$\mu_\omega = \frac{2}{3}\frac{\omega_p^2}{c}\int_0^\infty \frac{\varepsilon^{3/2} v_{tr}}{\omega^2 + v_{tr}^2}\left(-\frac{dn}{d\varepsilon} \right) d\varepsilon. \quad (14)$$

Here $\omega_p^2 = 4\pi e^2 N_e / m$ is the plasma frequency squared. Typically the EEDF decreases with the increase of energy, i.e. $dn/d\varepsilon$ is negative and the integral in (14) is positive. Hence, the absorption coefficient in plasma is positive, $\mu_\omega > 0$. However, if energy intervals with positive derivative $dn/d\varepsilon$ dominate in the integral (14), it is possible to obtain negative absorption or amplification of electromagnetic radiation in a plasma. It was demonstrated in (Bekefi, et al, 1961; Bunkin, et al, 1973) that in order to obtain negative value of the integral (14) the condition

$$\frac{d}{d\varepsilon}\left(\frac{\varepsilon^{3/2} v_{tr}}{\omega^2 + v_{tr}^2} \right) < 0 \quad (15)$$

should be satisfied in the energy range with population inversion and also this energy range should mainly contribute to the integral (14). In the low frequency limit ($\omega << v_{tr}$) one derives from (15) $d(\varepsilon/\sigma_{tr})/d\varepsilon < 0$, i.e. transport cross section should grow up rapidly than linear dependence. Such a situation is realized for Xe atoms in the energy range $\varepsilon = 0.7 - 4$ eV.

Results of calculations of the gain factor in a Xe:F_2 plasma sustained by an electron beam are presented at fig.6. First we note that for xenon concentration of $N = 10^{20}$ cm^{-3} (≈ 4 atm) and concentration of F_2 molecules equal to 8×10^{15} cm^{-3} the positive value of gain factor is achieved for microwave radiation with frequencies $\omega \le 4 \times 10^{11}$ c^{-1} (see fig.6a). Electron density is found to be proportional to the total electron rate production q while the position of the peak in the electron energy spectrum do not depend on q. Hence the gain factor (absorption coefficient) is characterized by a linear dependence on q. The dependence of plasma amplification features on the F_2 concentration is more complicated (see fig.6b). First one notes that in order to achieve positive value of a gain factor the concentration of electronegative specie should be above some critical value $\approx 3 \times 10^{15}$ cm^{-3}. A gain factor reaches its maximum value ~0.0032 cm^{-1} for $N_{F2} \approx 6 \times 10^{15}$ cm^{-3} and then falls down with further increment of F_2 concentration due to the decrement of electron density. Such a nonmonotonous gain factor dependence on the F_2 concentration results from the sugnificant reconstruction of the EEDF in dependence on F_2 concentration. In particular, the position of the electron peak in the EEDF ε^* is shifted towards lower energies with the reduction of concentration of F_2 specie (see data in the Table 1). The possibility to obtain positive gain factor value disappears when ε^* is found to be out of the energy interval $\varepsilon = 0.7 - 4$ eV suitable for amplification.

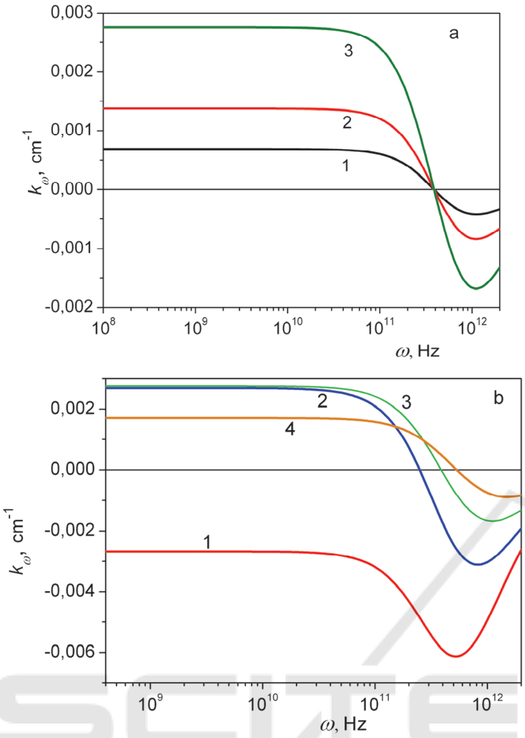

Figure 6: Gain factor (absorption coefficient) in Xe:F_2 plasma : (a) concentration of F_2 molecules is $8 \cdot 10^{15}$ cm^{-3}; production rates (cm^{-3}c^{-1}) are 2×10^{17} (1), 4×10^{17} (2) and 8×10^{17} (3) and (b) production rate is 8×10^{17} cm^{-3}c^{-1}; concentrations of F_2 molecules (cm^{-3}) are 2×10^{15} (1), 4×10^{15} (2), 8×10^{15} (3), 1.6×10^{16} (4).

In order to increase the frequency range of the amplification one also needs to shift the position of the maximum in the electron spectrum ε^* up to higher energies to satisfy the condition $\omega < \nu_{tr}(\varepsilon^*)$. This is possible (see data in the Table 1) if one increases concentration of F_2 molecules in the mixture, as the balance of electrons production and attachment will shift to higher energies. For example, for $q = 8\times10^{17}$ cm^{-3}c^{-1} the increment of F_2 concentration from 4×10^{15} cm^{-3} to 1.6×10^{16} cm^{-3} leads to the change in the energy peak position from 0.78 to 1.00 eV and subsequent expanding of the frequency band for amplification from 0.25 THz to 0.5 THz (see fig. 6b). On the other hand the increment of F_2 concentration for a given electron production rate results in reduction of the electron density and absolute value of the gain factor. For the above mentioned parameters the four-time-increment of the of F_2 concentration results in one and a half time reduction of the gain factor in low frequency $\omega << \nu_{tr}(\varepsilon^*)$ range.

All data above were obtained for rather low intensities of RF field that do not contribute to the EEDF evolution. In the two-term approximation used for analyzing of the EEDF temporal evolution external electromagnetic field also results in the diffusion spreading of initial photoelectron peak. This diffusion should be taken into account for low-frequency fields ($\omega << \nu_{tr}$) if the condition

$$\frac{e^2 E_0^2}{3m\nu_{tr}^2} \geq \frac{2m}{M} T_g \tag{16}$$

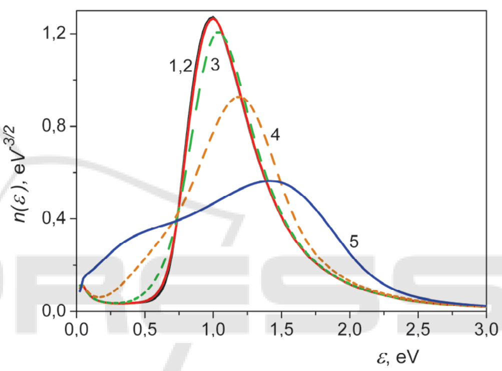

Figure 7: Steady-state EEDF in Xe:F_2 plasma for production rate 8×10^{17} cm^{-3}c^{-1} and F_2 concentration are 1.6×10^{16} cm^{-3}. RF field intensities (W/cm^2) are 0 (1), 0.1 (2), 1 (3), 10 (4) and 100 (5).

is fulfilled. For example, for xenon plasma with energy peak position $\varepsilon^* \approx 1$ eV, gas temperature $T_g = 0.03$ eV and xenon concentration $N = 10^{20}$ cm^{-3} one derives that the inequality (16) is valid for RF radiation with the intensity greater than $1 \div 3$ W/cm^2. The results obtained from the numerical integration of the Boltzmann equation (see fig. 7) are in agreement with the above estimations. Above the intensity of 1 W/cm^2 the reconstruction of the EEDF by RF field is of importance.

Results of calculations of a gain factor for different values of radiation intensity in dependence on RF frequency are presented at fig. 8 and demonstrate that the absolute value of gain factor value reduces significantly with the growth of amplified RF field. The intensity ~100 W/cm^2 destroys the amplification process for any value of frequency. Also the increment of RF field intensity leads to the shift of the upper boundary of the amplification band.

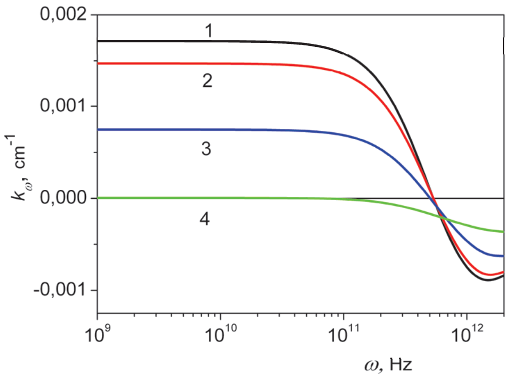

Figure 8: Gain factor (absorption coefficient) in Xe:F_2 plasma for: (a) concentration of F_2 molecules 1.6×10^{16} cm^{-3}, production rate 8×10^{17} cm^{-3}c^{-1}. RF field intensities (W/cm^2) are 0 (1), 1 (2), 10 (3) and 100 (4).

4 CONCLUSIONS

Thus, it was shown that a plasma sustained by an electron beam in a gas mixture with effective attachment process and the energy range with increasing transport cross section can be used as a media for amplification of RF radiation up to subterahertz frequency band. For the mixture of Xe:F_2 for the pressure ~4 atm it was found that the gain factor $\sim 1\div3\times10^{-3}$ cm^{-1} can be achieved in the stationary regime. The amplification up to intensity ~1-10 W/cm^2 is possible in the examined mixture. For the discharge chamber with transverse size of ~30 cm it provides the possibility to amplify subterahertz pulses up to 1-10 kW power. To achieve terahertz range one needs to increase electron collisions transport frequency in the energy range corresponding to the peak position in electron energy spectrum. The simplest way to do it is to increase the gas pressure. More complicated but probably more effective way is to choose optimal mixture components. For example, if xenon as a gas that characterized by the energy interval with increasing transport cross section $0.7-4$ eV is used, it is desirable to create the peak in the electron spectrum near the upper limit of this interval as the transport cross section will be of order of value larger than that used in our simulations. Hence, one needs the electronegative component in the mixture with effective attachment of electrons up to energies 2 - 3 eV.

The absence of spontaneous emission in terahertz and subterahertz frequency band does not allow to use an e-beam sustained plasma also as a generator of subterahertz radiation as it is typically possible for visible and IR radiation. One still needs the additional source of RF radiation to be used for amplification.

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