Self-oscillations of particle yield from methane films irradiated with an electron beam: experiment and simulation

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Theoretical model

The system of equations for defect concentrations and temperature has the following form:

$$\frac{\partial n_1}{\partial t} = G_1 - K_{01}\exp(-E_{a1}/(\kappa T))n_1^2$$

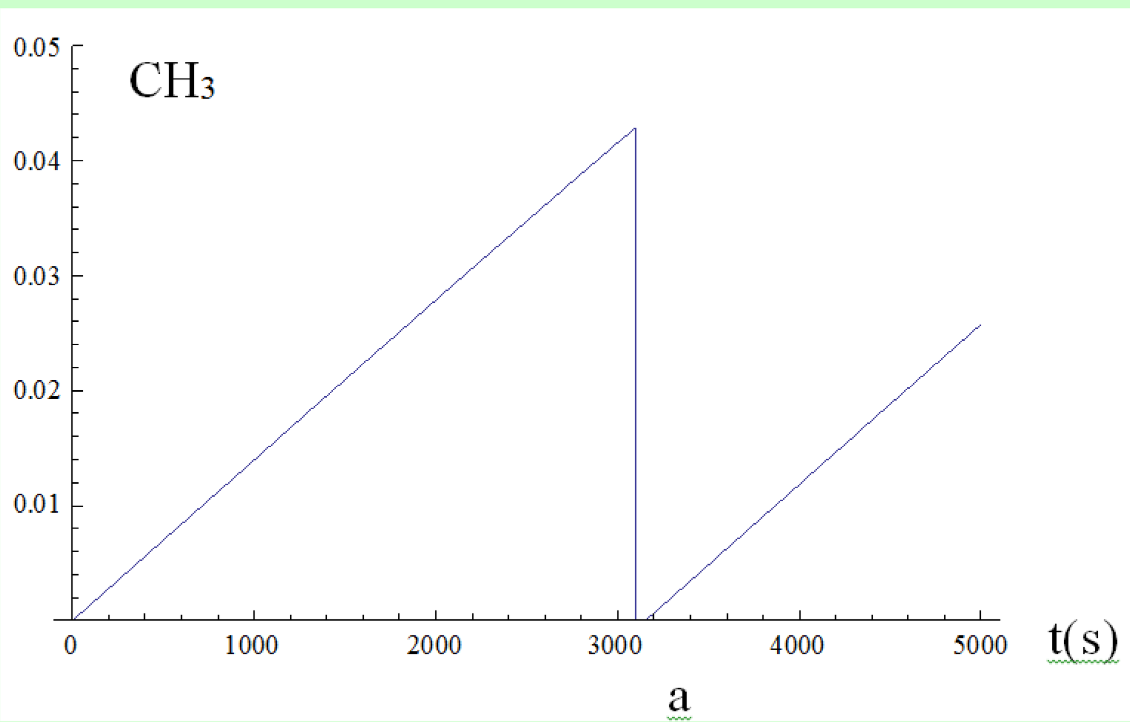
$$\frac{\partial n_2}{\partial t} = G_2 - K_{02}\exp(-E_{a2}/(\kappa T))n_2^2$$

$$lc(T)\frac{\partial T}{\partial t} = P + E_1K_{01}\exp(-E_{a1}/(\kappa T))n_1^2 l/v_0 + E_2K_{02}\exp(-E_{a2}/(\kappa T))n_2^2 l/v_0$$
$$-B\,(T - T_b),$$

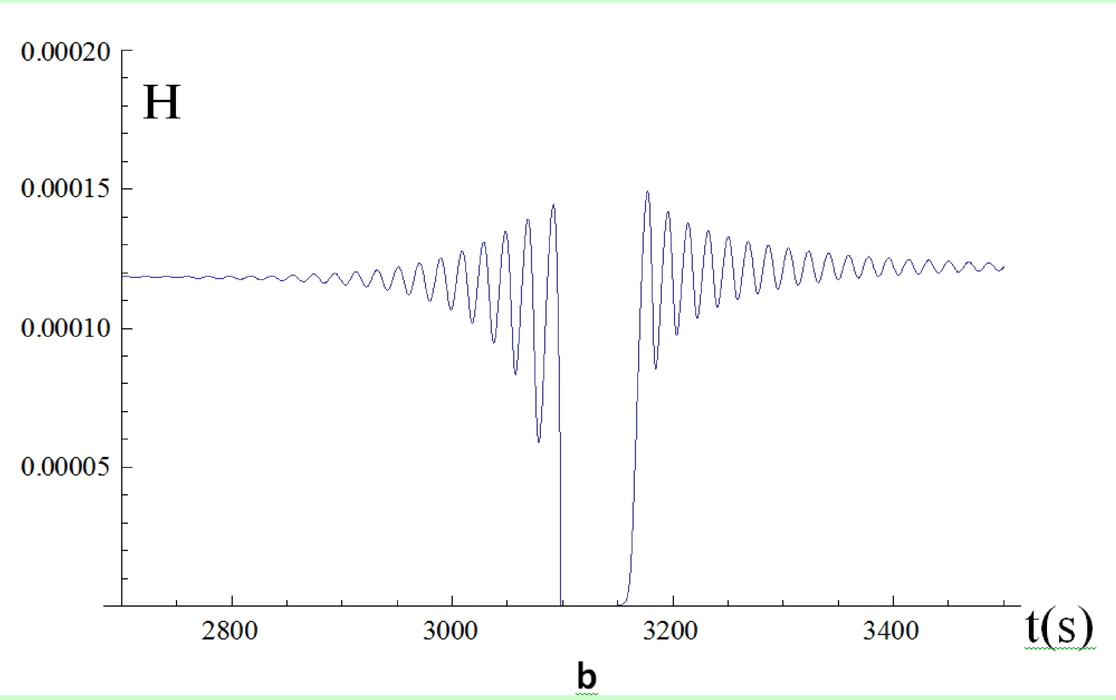
n_1 and n_2 – numbers of H and CH_3, relative to the number of CH_4, $G_1 = G_2$ - number of H and CH_3, which created by irradiation in unit time and in volume equal to CH_4, molecule volume, P – part of the electron energy spent to heating, E_{a1} and E_{a2} – migration activation energies of H and CH_3, B is the coefficient of energy transfer from the sample to the thermostat, $c(T)$ – the heat capacity of a CH_4 volume unit.

Results of the equations numerical solution

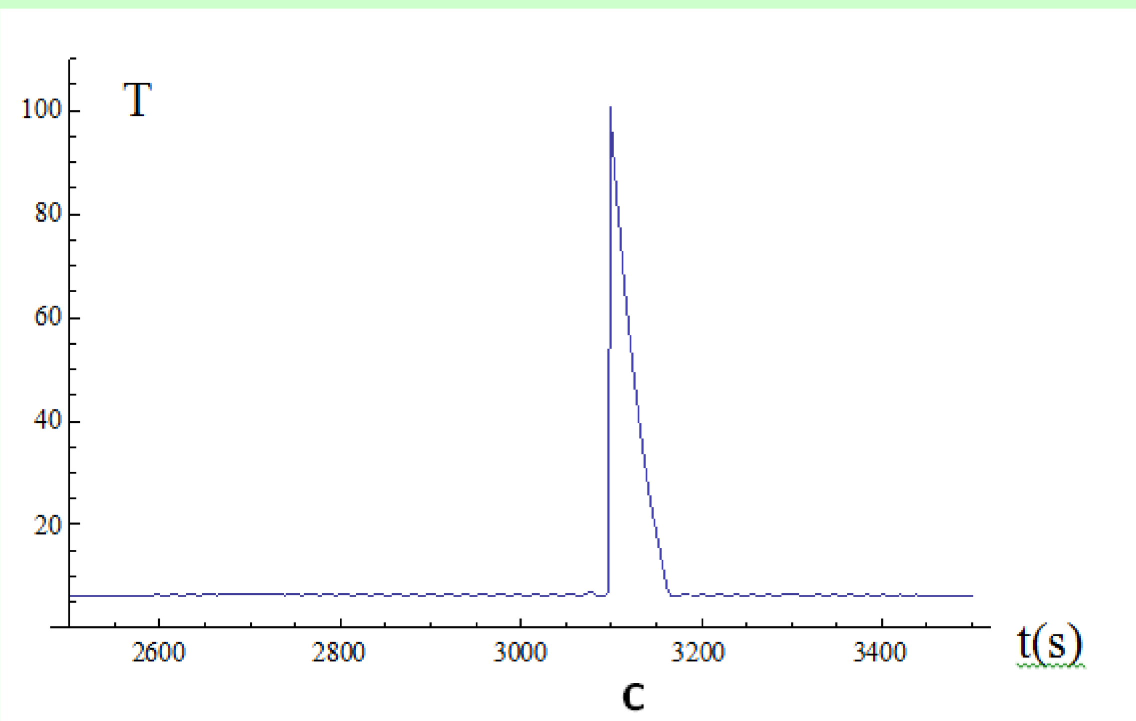
One example of the equations solution for a selected parameters set is shown below



a



b



c

The temporal dependences of densities of CH_3 radicals (a), H atoms (b) and T (c) at the following parameters: $G_1=G_2=1.4*10^{-5}$, E_{a2}=200K, E_{a1}=117.8K, T_b=4K.

The yield of H_2 from electron-irradiated CH_4 film arising from the binding of H atoms (reaction (5)), was calculated. It was shown that the temporal dynamics of desorption repeats the dynamics of self-oscillations.

Introduction

The ongoing interest in research of radiation effects in solid CH_4 is driven by its significant role in astrophysics, technology of cryogenic moderators, radiation physics and chemistry. The phenomenon of delayed desorption was observed during irradiation with neutron flux [1, 2], ions [3] and electrons [4]. In our experiments [4], unlike the previous ones, two types of self-oscillations in the yield of particles from solid CH_4 were detected. The first theoretical model of the self-oscillations with short and long periods was suggested [5]. Here we present an outgrowth of this research.

Excited electronic states of CH_4 – dissociative

$$CH_4 + \Delta E \rightarrow CH_3 + H \quad (1)$$
$$CH_4 + \Delta E \rightarrow CH_2 + H_2 \quad (2)$$
$$CH_4 + \Delta E \rightarrow CH_2 + 2H \quad (3)$$
$$CH_4 + \Delta E \rightarrow CH + H + H_2 \quad (4)$$

We studied the system already considered in [1,2] and described by reactions (5), (6) and found two types of self-oscillations under e-beam.

For self-oscillations to arise, the movement and recombination of radicals are necessary. The H atoms and CH_3 radicals have an activation character of diffusion and the largest diffusion coefficients. When they recombine, energy is released.

$$H + H \rightarrow H_2 + \Delta E_1 \quad (5)$$
$$CH_3 + CH_3 \rightarrow C_2H_6 + \Delta E_2 \quad (6)$$

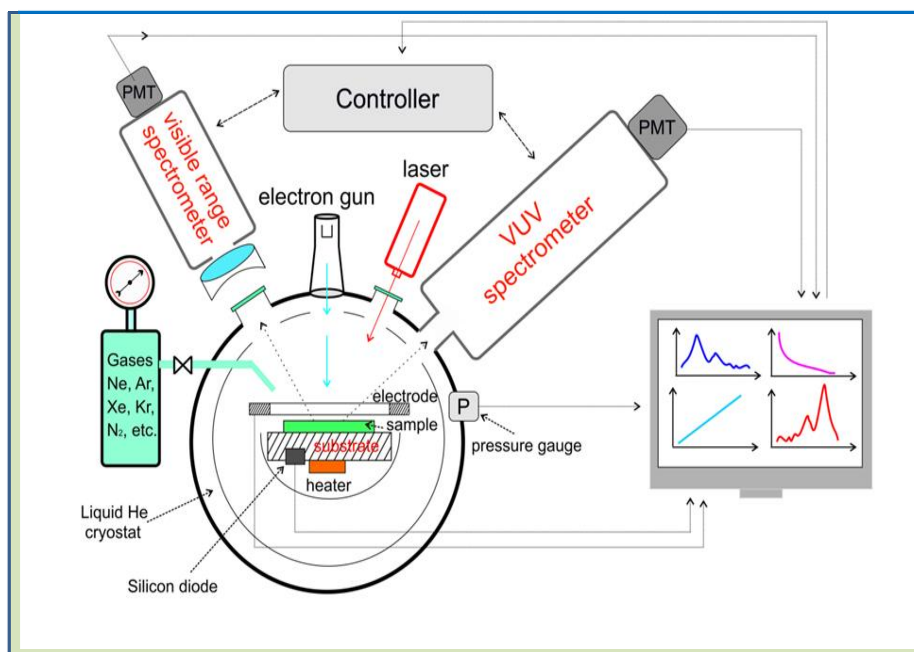
With increasing T, processes of diffusion and recombination are accelerated. Subsequent energy release leads to a further increase in T. The stationary state of the system becomes unstable and self-oscillations occur. A simplified task – self-oscillations in a thin CH_4 layer with a thickness of the order of the electron range, was considered.

Findings

Self-oscillations and, in general, explosive instability are realized in a limited region on the temperature-pump plane. The main findings for the studied case are listed below:

- ✓ A model is proposed that qualitatively describes the appearance of two types of self-oscillations upon electronic excitation of solid CH_4, their periods and delay time of burst.
- ✓ Two types of self-oscillations found are a periodic change of temperature and concentration with time of CH_4 decay products – H atoms and CH_3 radicals upon irradiation.
- ✓ The found patterns of self-oscillations was shown to determine the temporal dynamics of the delayed explosive desorption.
- ✓ The effect of decreasing the concentration of CH_4 over time was found to influence the temporal dynamics. Due to the different reactions (1)-(4) stimulated by electron irradiation, the number of molecules CH_4 should drop. The calculations show that the such decrease of the methane density causes the reduction of number of the self-oscillation cycles.

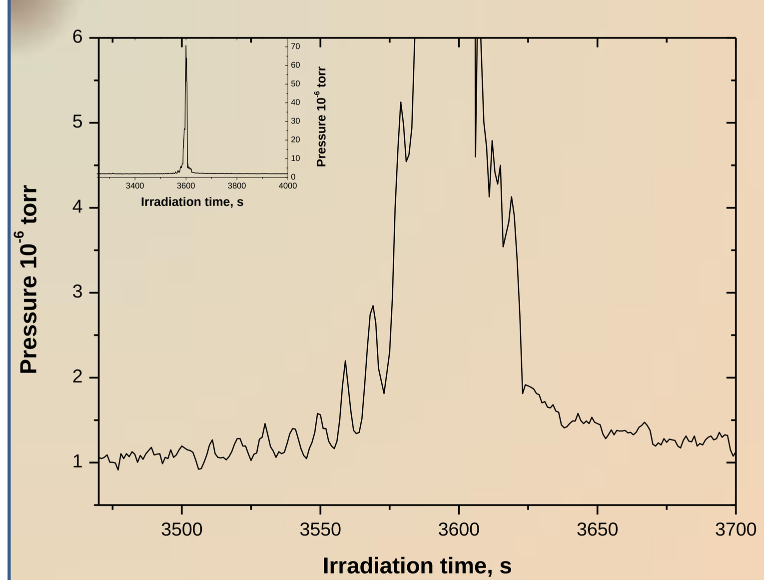
Experimental setup



Sample deposition from the gas phase

Sample thickness 100 μ

The irradiation was performed in dc regime with electrons of subthreshold energy (1 keV) to avoid the knock-on defect formation and sputtering of samples. The current density j was set to 3 mAcm^{-2}.

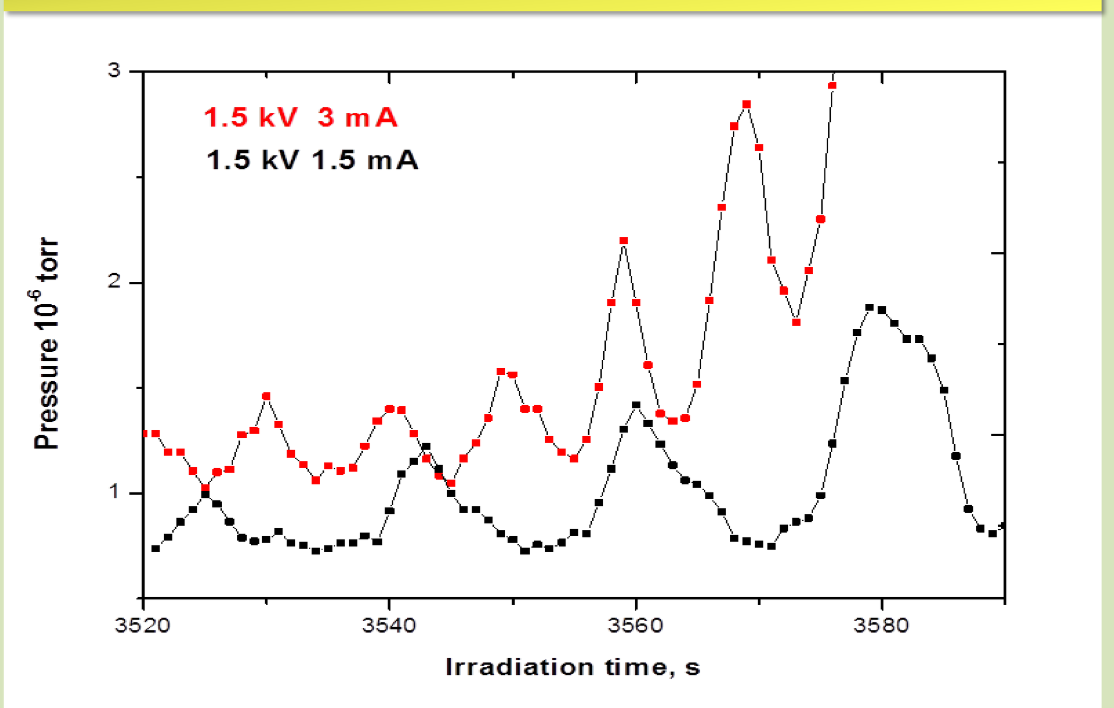


Delay in the giant burst of particles with respect to the start of irradiation indicates the need to accumulate the radiolysis products for this effect.

Two types of oscillations of the particle yield with long and short periods. One burst is shown in the Figure

The total yield of particles from the surface of solid CH_4 was characterized by an increase in P in the vacuum chamber. After a long exposure to an electron beam at LHe T, we observed an explosive emission of particles from an irradiated CH_4 film. This “explosion” resulted in a drop in the chamber vacuum by two orders of magnitude. The central peak of explosive particle emission was preceded by oscillations in the yield of particles with increasing amplitude. The oscillation period τ depended on the current density j and decreased with increasing j.

Self-oscillation period



Change in the self-oscillation period with j.

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