ISSN 1063-7826, Semiconductors, 2016, Vol. 50, No. 5, pp. 694–698. © Pleiades Publishing, Ltd., 2016.
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FABRICATION, TREATMENT, AND TESTING OF MATERIALS AND STRUCTURES

Role of the Heat Accumulation Effect in the Multipulse Modes of the Femtosecond Laser Microstructuring of Silicon

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Submitted November 26, 2014; accepted for publication October 21, 2015

Abstract—The results of quantitative evaluation of the heat accumulation effect during the femtosecond laser microstructuring of the surface of silicon are presented for discussion. In the calculations, the numerical–analytical method is used, in which the dynamics of electronic processes and lattice heating are simulated by the numerical method, and the cooling stage is described on the basis of an analytical solution. The effect of multipulse irradiation on the surface temperature is studied: in the electronic subsystem, as the dependence of the absorbance on the excited carrier density and the dependence of the absorbance on the electron-gas temperature; in the lattice subsystem, as the variation in the absorbance from pulse to pulse. It was shown that, in the low-frequency pulse-repetition mode characteristic of the femtosecond microstructuring of silicon, the heat accumulation effect is controlled not by the residual surface temperature by the time of the next pulse arrival, which corresponds to conventional concepts, but by an increase in the maximum temperature from pulse to pulse, from which cooling begins. The accumulation of the residual temperature of the surface can affect the microstructuring process during irradiation near the evaporation threshold or with increasing pulse-repetition rate.

DOI: 10.1134/S1063782616050080

1. INTRODUCTION

The use of ultrashort pulses of laser radiation revealed new opportunities in a number of technological applications: in micro- and nanotechnologies, in micro- and nanomedicine, and in micro- and nanobiology [1]. Currently, the technology of the micro- and nanostructuring of the surfaces of metals and semiconductors upon exposure to femto- and picosecond pulses is topical [2, 3]. Depending on the laser-radiation parameters and material properties, various structures can be formed on the surface, i.e., a periodic ripple, ordered granular structure, microcolumns of various thickness and height, and oxide layers. In these modes, the technology for multipulse processing at various pulse-repetition rates is conventionally used [4–6]. A feature of the femtosecond multipulse structuring of semiconductors is a change in the optical properties of the surface layer during and after the pulse. Previously, the effect of heat accumulation in the modes of intense surface femtoablation [7, 8] or at high femtosecond pulse-repetition rates (>1 MHz) [9] was studied. Thus, the role of the heat accumulation effect for the case of low-frequency femtosecond microstructuring of a semiconductor surface requires detailed study.

In this paper, the results of numerical–analytical analysis, relating the effect of heat accumulation from pulse to pulse to a change in the silicon optical characteristics during a femtosecond pulse and between pulses are proposed for discussion.

2. NUMERICAL–ANALYTICAL METHOD FOR EVALUATING THE SURFACE TEMPERATURE

The numerical–analytical method is used in the calculations, within which the dynamics of electronic processes and lattice heating are simulated by a numerical method, and the cooling stage is described based on an analytical solution relating the thermophysical characteristics of a material and the pulse-repetition rate. This method makes it possible to significantly reduce the calculation time. The numerical model of single-pulse femtosecond excitation and heating of silicon [10–12] allows quantitative evaluation of the spatial-temporal change in the concentration of excited electrons, controlled by the two-photon absorption of light photons, carrier diffusion, and external emission, and the surface temperature based on the two-temperature model with varying specific heat of the electron gas. The calculations are performed using initial data corresponding to the experimental modes of femtosecond structuring of the silicon surface: the energy density Q = 0.4–0.8 J/cm^2, pulse duration τ = 80 fs, emission wavelength λ =

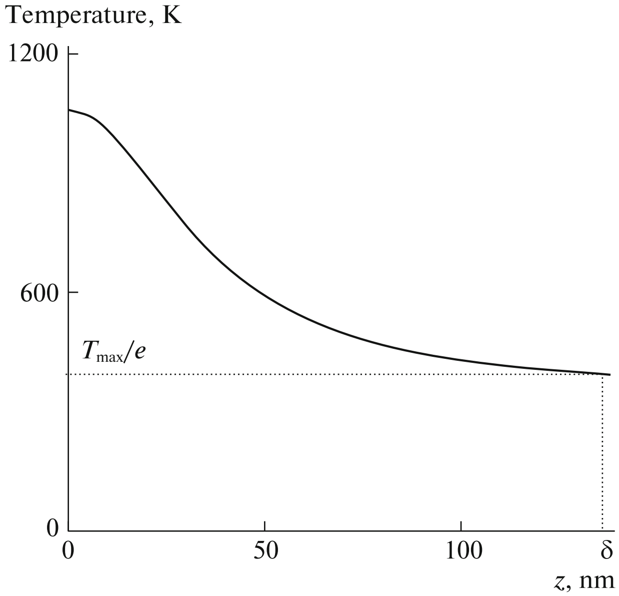

Fig. 1. Depth (z) distribution of the lattice temperature at the time point of the maximum lattice temperature.

1250 nm, pulse-repetition rate $f = 1$–1000 Hz, and the pulse numbers $N = 1$–1200 [13, 14].

The cooling stage is considered based on an analytical solution of the heat-conduction equation with a given initial temperature distribution. In this case, the change in the silicon surface temperature relative to the initial temperature by the arrival time of the

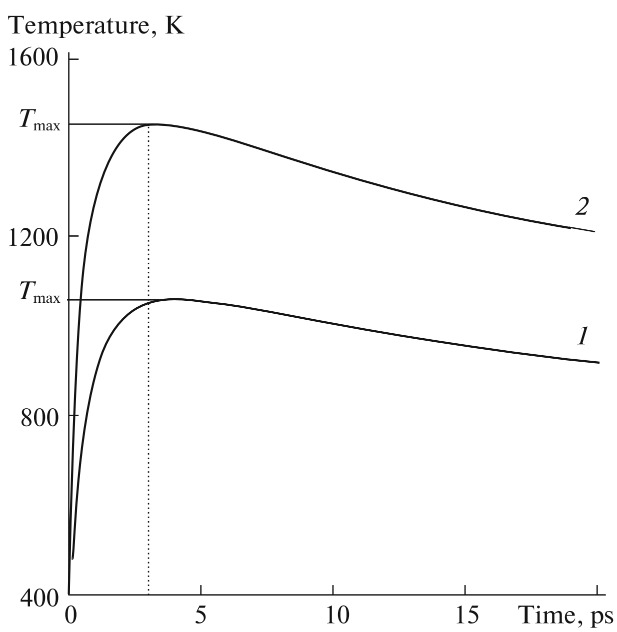

Fig. 2. Time dependence of the silicon-surface temperature after the end of the single pulse: (*1*) $A_0 = 0.2$ and (*2*) $A = A_0 + A(T_e)$.

$(N + 1)$th pulse (the residual surface temperature can be estimated according to [15] as

$$\Delta T(z = 0, t = N/f) = (T_{max} - T_{init})\frac{1}{\alpha\sqrt{\pi a/f}}\sum_{i=1}^{N}\frac{1}{\sqrt{i}}, \quad (1)$$

where T_{max} is the temperature to which the silicon surface is heated by the time of cooling onset, α is the silicon absorbance, a is the silicon thermal diffusivity, and T_{init} is the initial surface temperature.

Formula (1) shows that heat accumulation depends on the repetition rate and pulse numbers: the higher the rate and number of pulses, the greater the heat accumulation effect. At comparatively low frequencies (<1 kHz), the material absorbance and maximum heating temperature begin to play an important role.

3. ABSORBANCE AND MAXIMUM SILICON HEATING TEMPERATURE UPON EXPOSURE TO A SINGLE FEMTOSECOND PULSE

During the laser pulse, the semiconductor surface layer is metallized. The dynamics of the absorbance which is the sum of absorbances of the internal photoelectric effect, external photoelectric effect (emission phenomena), and free-electron absorbance, traces the change in the excess carrier density. As numerical calculations show, its value during the pulse exceeds 10^5 cm^{-1}. The absorbance after the pulse end at the instant the lattice-heating temperature reaches a maximum can be estimated by numerical calculation of the depth δ at which the maximum lattice temperature decreases by a factor of e. In Fig. 1, we can see that $\delta = 1.33 \times 10^{-5}$ cm, hence, $\alpha = 1/\delta = 7.5 \times 10^4$ cm^{-1}.

Thus, although the absorbance decreases at the cooling stage, it still remains sufficiently high by the time of reaching the maximum temperature of lattice heating, which, according to Eq. (1), decreases the residual surface temperature by the time of second-pulse arrival, hence, the role of the accumulation effect in the femtosecond structuring of silicon.

The residual surface temperature by the time of second-pulse arrival is also affected by the maximum temperature T_{max} to which the silicon surface is heated by the time of cooling onset. In turn, T_{max} is controlled by the incident-light flux density and the semiconductor absorbance. Using the value $Q = 0.4$ J/cm^2 in the calculations, let us evaluate the effect of the absorbance which varies during the pulse on T_{max}.

Indeed, the absorbance under ultrashort exposures depends on the frequency of electron–electron collisions and is a function of the electron-gas temperature (T_e). Taking into account the dependence of the absorbance on the electron gas temperature in the approximation of the weakly anomalous high-frequency skin effect [16], $A = A_0 + A(T_e)$, we can calculate the temporal temperature distribution of the silicon surface according to the two-temperature model

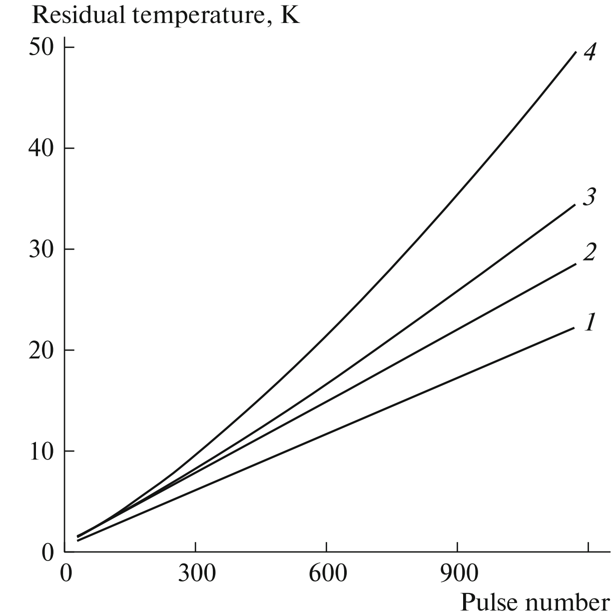

Fig. 3. Dependence of the residual temperature of the silicon surface on the pulse number: (*1*) $A = 0.2$, (*2*) $A = A_0 + A(T_e)$, (*3*, *4*) $A = A_0 + A(T_e) + N\rho$, $\rho =$ (*3*) 10^{-4} and (*4*) 5×10^{-4}.

[10–12]. In Fig. 2, we can see that the silicon surface temperature reaches a maximum a few picoseconds after the pulse end and increases by a factor of 1.3 if the temperature dependence of the absorbance is taken into account.

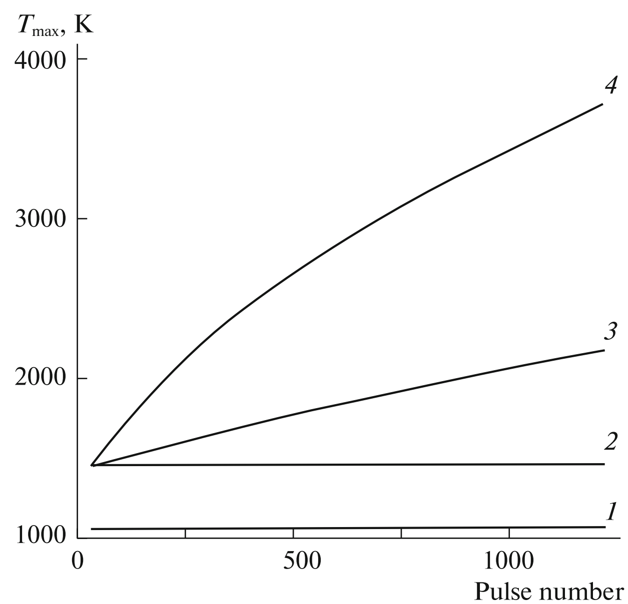

Fig. 4. Increase in the maximum heating temperature of the silicon surface with increasing pulse number: (*1*) $A = 0.2$, (*2*) $A = A_0 + A(T_e)$, (*3*, *4*) $A = A_0 + A(T_e) + N\rho$, $\rho =$ (*3*) 10^{-4} and (*4*) 5×10^{-4}.

After the end of the first pulse, the residual surface temperature before second-pulse arrival is $\Delta T = 0.013$ K at $A = 0.2$ and $\Delta T = 0.015$ K at $A = A_0 + A(T_e)$.

4. EVALUATION OF SURFACE TEMPERATURE ACCUMULATION FROM PULSE TO PULSE

The initial data corresponding to the typical experimental modes of the femtosecond structuring of silicon were used in the calculation: $Q = 0.4$ J/cm^2, $f = 10$ Hz, and $N = 1200$.

Previously, we took into account the change in the absorbance during the pulse; however, upon irradiation in the surface microstructuring modes, the absorbance changes from pulse to pulse. This is caused by the appearance and development of a periodic relief on the semiconductor surface; from pulse to pulse, the relief is formed over the irradiated area [17], and the relief-height grows. Furthermore, the coefficient of incident radiation conversion to a surface electromagnetic wave increases, enhancing the polariton mechanism of relief formation. We propose complimenting the numerical–analytical model by taking into account the change in the surface relief and the possibility of accumulation of these changes from pulse to pulse as the appearance of the additional term A_s in the absorbance,

$$A = A_0 + A(T_e) + A_s, \quad A_s = N\rho, \tag{2}$$

where ρ is the phenomenological coefficient.

The proposed approach is consistent with the results of studying femtosecond-laser pulse-periodic surface structures [18], where the appearance of a significant antireflective effect of silicon irradiated in microstructuring modes was detected. The linear dependence of the additional absorbance component is consistent with the experimental results of [19], where a linear increase in the microrelief height on the pulse numbers was established. The phenomenological coefficient can be chosen from the experimental conditions of the silicon microstructuring mode, according to which surface structuring at pulse numbers of $N = 600$–1200 occurs under conditions of surface melting, but without intense evaporation and a change in the periodic relief type, and is $\rho = (1–5) \times 10^{-4}$.

Figure 3 shows the calculated dependence of the residual temperature of the silicon surface on the femtosecond-pulse numbers. We can see that consideration of the change in the absorbance from pulse to pulse due to surface modification leads to an increase in the relative heating per 1200 pulses by no more than 35–50 K, depending on the phenomenological coefficient ρ (Fig. 3, curves *3* and *4*). This is a rather weak accumulation effect on the residual surface temperature, which is not consistent with the experimentally

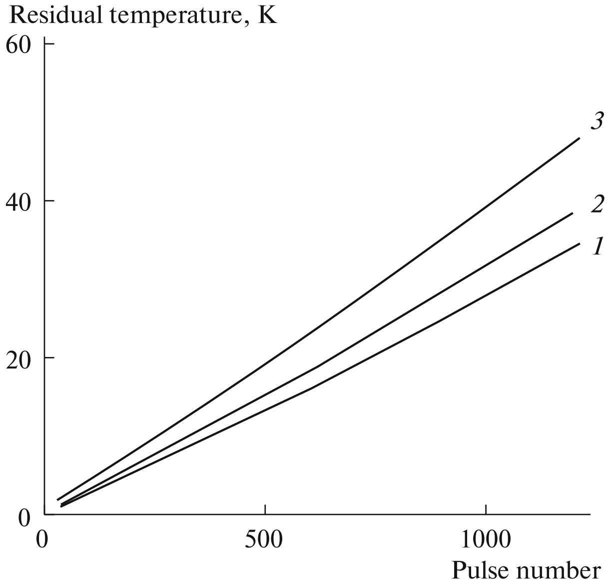

Fig. 5. Dependence of the residual temperature of the silicon surface at a frequency of $f = 10$ Hz on the pulse number at $A = A_0 + A(T_e) + N\rho$, $\rho = 10^{-4}$, $Q =$ (*1*) 0.4, (*2*) 0.5, and (*3*) 1 J/cm^2.

observed intensive relief development on the pulse number.

However, the accumulation effect on the maximum temperature manifests itself quite differently. Figure 4 shows the increase in the maximum temperature of the silicon surface with increasing pulse number for various functional dependences of the absorbance.

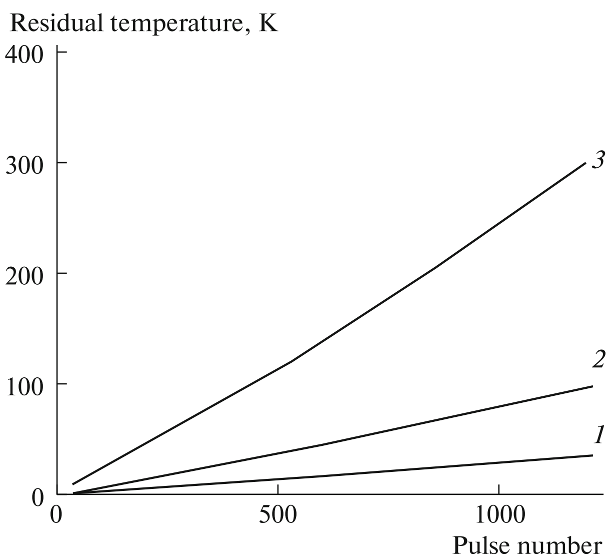

Fig. 6. Dependence of the residual temperature of the silicon surface on the pulse number at $A = A_0 + A(T_e) + N\rho$, $\rho = 10^{-4}$, $Q = 0.4$ J/cm^2, $f =$ (*1*) 10, (*2*) 100, and (*3*) 1000 Hz.

In the case of constant absorbance or absorbance varying due to processes in the electronic subsystem of the photoexcited semiconductor, T_{max} remains unchanged with increasing pulse number, and the heat accumulation effect is described, according to conventional concepts [20], by the residual temperature. In this case, the heat accumulation effect, as mentioned above, is smallest.

If the interpulse change in the absorbance is taken into account in the calculation, the maximum temperature increases in proportion to the pulse number. Comparing the results obtained with concepts on the "polariton" microrelief, we can say that the performed evaluations suggest the possibility of the development of hydrodynamic or evaporative mechanisms of relief formation with increasing pulse number, which is consistent with experimental data. Furthermore, the results obtained allow us to explain the experimentally determined fact of the extremely complex formation of microrelief seeds in the first 1–3 pulses.

5. DEPENDENCE ON THE LIGHT FLUX AND PULSE-REPETITION RATE

All previous calculations were performed for a low pulse-repetition rate, $f = 10$ Hz, and a light flux density below the silicon melting threshold, $Q = 0.4$ J/cm^2. Figures 5 and 6 show the dependences of the residual temperature ΔT on the pulse number for various energy densities of laser radiation and pulse-repetition rates. We can see that the effect of residual surface-temperature accumulation can have an effect on surface modification at frequencies of ~1000 Hz (see Fig. 6, curve *3*). This conclusion is indirectly confirmed by the known experimental data [21, 22]. Indeed, surface-relief formation at these frequencies occurs for a significantly smaller pulse number $N =$ 50–200.

6. CONCLUSIONS

The results of quantitative evaluation of the effect of heat accumulation on a silicon surface during multipulse laser irradiation in femtosecond microstructuring modes allowed us to determine the contribution of changes in the optical characteristics of a semiconductor to the final result.

An absorbance close to those of metals by the time of cooling onset, leads to a decrease in the residual surface temperature (a hundredth of a degree per pulse), hence, a weak accumulation effect. An increase in the absorbance during the pulse due to an increase in the electron-gas temperature causes an increase in the maximum temperature to which the lattice can be heated. However this value does not change with increasing pulse number; therefore, the accumulation effect is controlled by the residual surface temperature and appears weak, which is poorly compared with the experimentally observed microrelief development.

The change in the absorbance between pulses in microstructuring modes, first of all, due to relief growth, increases the residual surface temperature by a factor of 1.5–2, and can have an effect on microstructuring during irradiation near the evaporation threshold or with increasing pulse-repetition rate. A crucial role in the accumulation effect during the low-frequency femtosecond structuring of silicon is played by accumulation of the maximum surface temperature, caused by a linear change in the absorbance from pulse to pulse.

ACKNOWLEDGMENTS

This study was supported by the Russian Science Foundation, project no. 14-12-00351 and state support leading universities of the Russian Federation (grant no. 074-U01).

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Translated by A. Kazantsev