Feasibility of experimental identification of the thermophysical mechanism of formation of periodic surface structures

I. A. Avrutskii, P. V. Bazakutsa, V. L. Maslennikov, A. M. Prokhorov, and V. A. Sychugov

Institute of General Physics, Academy of Sciences of the USSR, Moscow

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An analysis is made of the results of observations which led to important conclusions on thermophysical mechanisms of formation of periodic surface structures which appeared on germanium under the action of high-power laser radiation.

In spite of the existence of electrodynamic models of formation of periodic structures on the surfaces of metals, semiconductors, and insulators,\textsuperscript{1,2} the problem of the specific thermal mechanism of the formation of these structures has not yet been solved. A theoretical solution of the problem is difficult because of the absence of reliable information on optical, mechanical, and other properties of materials, particularly semiconductors interacting with high-power laser radiation. It would therefore be of interest to report experiments and observations of characteristic features of changes in the state of the surface which occurred as a result of interaction with high-power laser radiation. We investigated samples of germanium which were subjected to radiation of wavelengths $\lambda = 9.6 \mu m$ and $1.06 \mu m$.

It is known that irradiation of germanium with light of the $\lambda \approx 10 \mu m$ wavelength creates structures with a period $\Lambda$ very different from the wavelength of light. The lines in the structures are oriented parallel to the vector $E$ of the incident radiation and the period varies from 0.8 to 3 $\mu m$. A diffraction mechanism\textsuperscript{3} of such "longitudinal" structures was used by us in calculations in which we estimated the difference between the evolution of the energy at the humps and troughs $\Delta Q$ as a function of $\lambda / \Lambda$. It was then found that the function $\Delta Q(\lambda / \Lambda)$ had a negative minimum of large amplitude at $\lambda / \Lambda \geq n$, where $n$ is the refractive index of the material (Fig. 1a). This extremum could give rise to an anomalously rapid rise of the corresponding spatial harmonics and the appearance of corrugations with such periods under the action of laser radiation. It should be stressed that a thermal mechanism when a relatively large amount of energy is dissipated at the tops of the corrugations rather than at the bottom should be responsible for the longitudinal corrugations.\textsuperscript{4}

We calculated the dependences $\Delta Q(\lambda / \Lambda)$ for substances with the optical constants $n$ and $\kappa$, and they varied from 0 to 4 (Fig. 1b). It was established that the above extremum of the function $\Delta Q(\lambda / \Lambda)$ appeared only for the samples for which $n$ and $\kappa$ in this diagram were above the curve corresponding approximately to $\text{Re}(\varepsilon) = 1$. The optical constants of solid germanium at $\lambda = 9.6 \mu m$ ($n = 4, \kappa \approx 10^{-2}$) were such that the formation of corrugations on the surface as a result of the laser interaction were quite feasible (according to our calculations), whereas molten germanium was known to go over to the metallic state, i.e., so that $\text{Re}(\varepsilon) < 1$, and, consequently, formation of periodic structures with $\Lambda_1 = \lambda / n$ was not possible in liquid germanium.

Figure 2a shows a photograph of the surface of germanium with a periodic structure. The structure had a bright region where the intensity of the interacting radiation was maximal and the germanium surface melted, as indicated by the absence in this region of scratches and projections expected after mechanical polishing and clearly visible in that part of the surface which was not subjected to radiation. Attention should be drawn to the fact that a periodic structure appeared along the perimeter of the molten region and in the intervals between such corrugation lines there were still scratches and irregularities, i.e., the material itself did not melt, because there would have been corrugations on the surface of a molten substance. This necessarily indicated that the structure in question formed in solid germanium.
and not in liquid, and this confirmed the results of our theoretical analysis.

We made similar observations on a structure with a period $A_z = \lambda / \cos \theta$, where the lines were oriented along the plane of incidence of the interacting light. A photograph of such a structure (Fig. 2b) shows clearly that the scratches and irregularities on the surface were retained in the intervals between the troughs of the corrugations. This confirmed the conclusion reached in Ref. 5 that the structure $A_z$ appeared because of the processes occurring on the surface of solid germanium.

Figure 2c shows a photograph of the relief obtained on the surface of germanium as a result of its irradiation with s-polarized light. A surface scratch, identified by an arrow, became smeared out in the corrugation troughs and was quite visible at its humps. In our opinion, such changes in the nature of a scratch were due to the fact that the depth of melting of germanium during formation of corrugations at their humps was less and in the troughs it was greater than the depth of the scratch. Hence, the mechanism of formation of structures of the type shown in Fig. 2c was inhomogeneous melting of the surface of germanium.

It was pointed out in Ref. 6 that the appearance of corrugations on the surface of germanium after the action of a laser pulse could be due to the loose structure of the surface layer of Ge. The formation of troughs required heating of Ge simply to the melting point. Evaporation of a material and thermocapillary effects clearly did not play an important role in the formation of structures shown in Fig. 2.

The photographs and their analysis presented above indicate that the influence of radiation on previously created surface defects could give major information on thermophysical mechanisms of formation of periodic surface structures, and it stimulated the search for the mechanisms of formation of periodic structures which could describe accurately the situation.

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Optothermal spectroscopy of ethylene using a tunable waveguide CO$_2$ laser

S. T. Kornilov, I. V. Ostreǐkovskii, and E. D. Prot senko

Engineering Physics Institute, Moscow

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The results are given of a study of optothermal linear and nonlinear absorption spectra of ethylene. An investigation was made of the dependence of the optothermal signal on the pressure of the absorbing gas and on the modulation frequency. Measurements were made of the collisional self-broadening coefficient of ethylene: 13.2 ± 0.7 MHz/Torr.

Optothermal and optoacoustic spectroscopic techniques can be used to record directly the energy absorbed by a gas. Whereas the optoacoustic method has been developed and is widely used, the optothermal method has been comparatively little studied. However, various publications indicate that it is potentially useful, since substantially lower pressures can be used compared with the optoacoustic method and the sensitivity is high. Optothermal detection was studied by us and its possible applications in laser spectroscopy were investigated using as an example the recording of the linear and nonlinear absorption spectra of ethylene.

The apparatus is shown schematically in Fig. 1. Radical...