Reflectivity of aluminum film upon exposure to current pulses and laser radiation

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The increase in aluminum reflectivity (λ = 280–317 nm) caused by the electric current (j ~ 100 MA/cm^2) and XeCl laser radiation ($I \approx 2$ MW/cm^2) is investigated. The effect can be both reversible and irreversible. We suppose that this effect can be explained by partial reflection of the radiation from free electrons falling into the conduction zone of the aluminum oxide film on the metal surface during electric current and laser radiation pulses.

Laser systems are now widely used in medicine, scientific investigations, and different industries mostly because of the feasibility of automatic control of laser radiation parameters due to the use of various types of high-speed modulators, such as Kerr and Pockels cells, as well as other optoacoustic transducers. Important elements of such devices are optically transparent solid or liquid media, which transmit radiation of different wavelengths. As is well-known, available optical materials are transparent only in some optical range. Therefore, every modulator can be applied in a rather narrow interval of electromagnetic waves. There are a sufficient number of materials for optoacoustic transducers in the middle and near IR, as well as in the visible region. The situation is somewhat worse in the UV spectral region and, especially, in the VUV range, for which it is difficult to find optically transparent materials having properties needed for modulators.

This problem can be solved by developing new electrooptical materials transparent for short-wave radiation. Another way to overcome the difficulties arising in an attempt to control the intensity of radiation in this spectral region is light modulation at its reflection from the surface, rather than transmission through a material. Theoretical investigations of Ivlev and Yakovlev into the effect of picosecond pulses on the optical parameters of conductors confirm, in principle, the possibility of such optical modulation.[1,2] Our experiments also demonstrate the possibility of changing the reflectivity of a conductor's surface during a current pulse. However, the investigations revealed that the current pulses with the density up to 100 MA/cm^2 and high-intensity radiation pulses lead also to irreversible changes in the optical parameters of films. The information about changes, in particular, improvement of the reflectivity of aluminum films can be found in Ref. 3. An object was exposed to UV radiation from a mercury quartz lamp for 5 h. In our experiments, such changes occurred much faster, for only several seconds.

The aim of this paper is to find regularities of irreversible and reversible changes in the optical parameters of aluminum film as a result of simultaneous exposure to nanosecond current pulses and radiation, as well as separate exposure to high-intensity radiation pulses.

1. Experimental setup

To determine the conditions for variation of the film reflectivity, experiments were conducted with a setup, whose schematic layout is shown in Fig. 1.

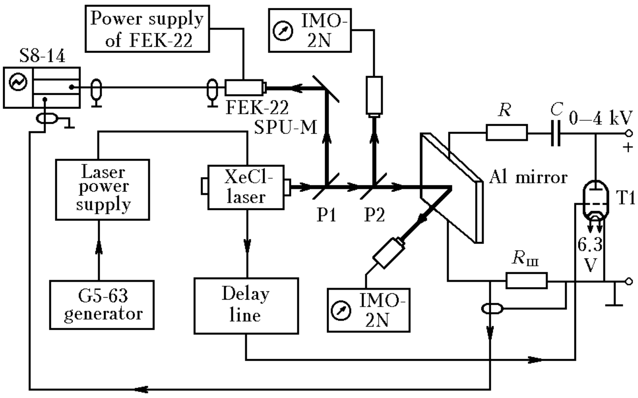

Fig. 1. Schematic layout of experimental setup.

The pulsed XeCl laser radiation (λ = 308 nm) passed through beam splitting plates P1 and P2, reflected from an 200–1000 Å aluminum film, and was then measured by an IMO-2N power and energy meter with and without high-power current pulses through the film. The pulse duration at half-maximum was equal, on the average, to 30 ns, and the pulse energy was 40 mJ. The more detailed description of laser characteristics can be found in Ref. 4. Since the energy varied within 10–15% from pulse to pulse, a part of the light pulse energy reflected from the beam-splitting plate P2 with the reflection coefficient r = 30% was used as a reference signal. This energy was measured by another IMO-2N power and energy meter. A 50–120 ns long current pulse up to 1000 A and higher was passed through the thin aluminum film as a capacitor C discharged through a current-limiting

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resistor R. The profiles of the current and radiation pulses were recorded by an S8-14 dual-beam oscilloscope. A padding resistor, which served for determination of the current, was assembled of low-inductance resistors of the TVO type.

The module for activation of the high-voltage pulse generator of the XeCl laser, depending on the experimental conditions, responded to a signal from either a pulse generator in the laser power supply in the single-pulse mode or from a G5/63 pulse generator.

A TGI1-500/16 gas thyratron T switched the charge accumulated by the capacitor C to the sample under study as a positive polarity pulse from the high-voltage pulse generator of the XeCl laser was applied to a grid through a delay line. The delay line (DL) served to control the delay of activation of the thyratron T with the step up to 10 ns. Owing to the use of DL, the radiation and current pulses could be shifted with respect to each other by a preset time interval from 0 to 500 ns.

A FEK-22 SPU-M coaxial photocell with the time resolution up to 100 ps served for conversion of the radiation pulse reflected from the quartz plate P1 into the electric signal. To decrease the power of a light pulse, we used an attenuator made of the properly arranged quartz plates of the KU1 type.

The reflection coefficients at different wavelengths were recorded by a BECMAN UV 5270 spectrophotometer. The resistance of the thin films was measured by an E6-181 milliohmmeter.

K 108 glass and KU-1 quartz plates were polished by the method of deep grinding-polishing. Before film evaporation, the plate surfaces were treated by glow discharge. Evaporation was conducted on a VU-1A vacuum system by the method of resistive evaporation.

2. Irreversible changes in aluminum film reflectivity as a result of exposure to current and radiation pulses

As was already noted, the reflectivity r of a metal film changes under the exposure to current and radiation pulses. Figure 2 depicts the dependence of r on the number of pulses. One can see that, first, r increases quite rapidly, but after 9–10 current and radiation pulses the reflection coefficient r saturates. The increase in the reflectivity of this sample at the wavelength of 308 nm was 13%. In this experiment the current and radiation pulses were generated synchronously.

When the technology of vacuum evaporation is used, the optical parameters of the films, all other conditions being the same, strongly depend on the degree of vacuum and the time of evaporation. For example, the reflection coefficient of the aluminum film at the wavelength $\lambda = 308$ nm can range from 22 to 92% [Ref. 5]. Since mirrors become dirty during storage and use, their reflectivity may be much worse.

The investigations showed that if films had the reflection coefficient close to 92%, then after the exposure their reflectivity almost did not change.

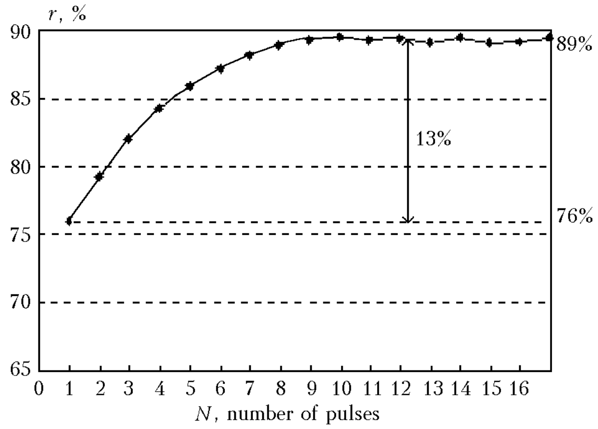

Fig. 2. Reflectivity of aluminum film as a function of the number of pulses N; mean power density $I = 2$ MW/cm^2.

At the same time, the reflectivity of samples with the low reflection coefficient became 20–25% higher after the exposure. This is clearly seen from Fig. 3, which depicts the wavelength dependence for the reflection coefficient r of three samples in the spectral range of 280–317 nm.

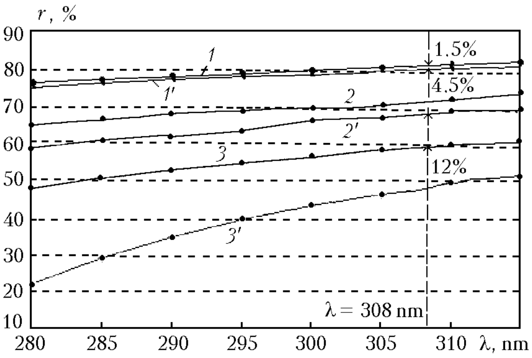

Fig. 3. Wavelength dependence of the reflectivity of aluminum film before (curves *1'*, *2'*, *3'*) and after (*1*, *2*, *3*) the exposure to radiation; radiation power density $I = 2$ MW/cm^2.

As the mirror was exposed to only current pulses, the reflectivity increased insignificantly (~ 2%), which suggests that the improvement of the reflectivity depends mostly on the radiation pulses or the joint effect of the both current and radiation pulses.

It should be noted that as the current density and the radiation power density exceeded certain values (> 100 MA/cm^2 and > 10 MW/cm^2, respectively), the samples were partly damaged and their reflectivity thus decreased.

As to the causes for changes in the reflection coefficients r, they depend on the methods of action on materials. The intense radiation cleans the surface from an oxide layer[6,7] and dirt, which is observed even visually. Our measurements showed that a high-power nanosecond current pulse led to the 10–15% decrease in the ohmic resistance of the films. As the current with the density $j \sim 100$ MA/cm^2 passes through the aluminum films, the changes improving the conductance likely occur in them, and electric properties of metals are known to be closely related to the optical properties.[8]

These results were put in the basis of the method for cleaning of the surface of optical elements.[9]

3. Reversible changes in aluminum film reflectivity as a result of exposure to current and radiation pulses

The experiments revealed that reversible increase in the aluminum film reflectivity occurs under the effect of current pulses up to 100 MA/cm^2.

The results of the experiments are tabulated below.

Laser pulse energy, mJ	28	29	31.4	33.8
Reflection coefficient, %				
with electrical current	89	87.6	85	83
without electrical current	88	86	82	79

We assume that this effect is caused by reflection of the laser radiation from the oxide film, which is always present on the aluminum surface in the atmosphere. The mechanism of reflection from the film is described in Ref. 10 with the V_2O_5 oxide film taken as an example. It consists in the following: laser radiation can reflect from free electrons falling, in some or other way, in the conduction zone of the oxide film. Under the condition $\omega_{\rm las} \sim \omega_{\rm p}$ ($\omega_{\rm las}$ is the laser radiation frequency, $\omega_{\rm p}$ is the frequency of plasma oscillations of free electrons) the reflection from the film becomes prevalent.

We have calculated the total reflection coefficient of the metal–film system (MFS). In the calculations, the reflection coefficient of the metal $r_{\rm m}$ was assumed constant. The reflection coefficient of the oxide film was calculated by the well-known equation

$$r_{\rm f} = \frac{(n-1)^2 + k^2}{(n+1)^2 + k^2}, \qquad (1)$$

where n and k are the refractive index and the absorption coefficient of the oxide film. They are calculated through numerical solution of the standard system of equations:

$$n^2 - k^2 = n_0 - \frac{\omega_{\rm p}^2 \tau^2}{1 + \omega_{\rm las}^2 \tau^2}, \qquad (2)$$

$$nk = \frac{\omega_{\rm p}^2 \tau}{2\omega_{\rm las}\left(1 + \omega_{\rm p}^2 \tau^2\right)}, \qquad (3)$$

where $n_0 = 1.57$ is the refractive index of Al_2O_3 under normal conditions, that is, at the virtually complete absence of free electrons in the conduction zone; τ is the mean relaxation time of electrons, which was taken equal to that of pure Al and calculated based on the data of Ref. 10.

The results shown in Fig. 4 demonstrate the increase of the MFS reflection coefficient with the increasing concentration of free electrons, that is, the ratio $\omega_{\rm p}/\omega_{\rm las}$. This increase becomes more significant (curves *2*, *3*), as the reflection coefficient of the metal decreases. This decrease can be caused by metal heating due to the laser irradiation, that is, as the energy of the laser pulse increases, the effect must be more pronounced, just which is observed experimentally.

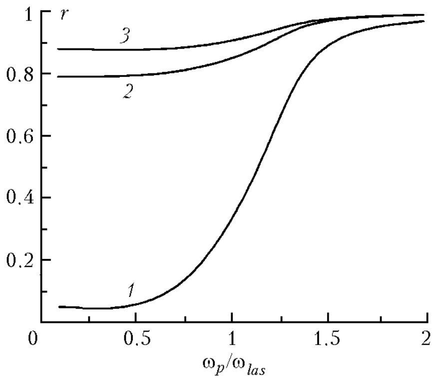

Fig. 4. Dependence of the reflection coefficients of the oxide film (*1*) and the metal–film system (*2*, *3*) on the parameter $\omega_{\rm p}/\omega_{\rm las}$ at different reflection coefficients of the metal: 79% (*2*) and 88% (*3*).

We cannot indicate with certainty the cause for appearance of free electrons in the conduction zone of the oxide film. In our opinion, this can be explained by the following reasons. Heating of aluminum by the current pulse can cause changes in the stoichiometric and phase composition of the oxide film, that is, formation of phases with a narrow forbidden zone. The reduction of the forbidden zone favors the penetration of electrons into the conduction zone directly from the metal, since, as known, the Fermi level of the metal lies at the center of the forbidden zone of the dielectric film covering the metal surface. Besides, sharp expansion of the metal heated by the short current pulse can cause deformation of the oxide film. In the intensity, this deformation is similar to the effect of a shock wave, which, as known (see Ref. 11 and references therein) causes the shift of energy zones in a dielectric. The decrease of the width of the forbidden zone significantly increases the probability of short-time overlap of the valence band with the conduction zone and transition of electrons into the latter.

Conclusion

The results presented in this paper demonstrate the ways to improve the reflectivity r of the aluminum film both by the simultaneous exposure to laser radiation and current pulses and by the separate exposure to only laser radiation. The degree of interaction was not fully optimized, and it can be supposed that, with the proper choice of the current and radiation pulse parameters, the increase in r will be more significant.

The experiments have revealed the effect of reversible increase of the aluminum reflection coefficient up to 4% during the high-density current pulse, and the qualitative explanation to this phenomenon has been proposed.

References

1. I.V. Ivlev and M.A. Yakovlev, Zh. Tekh. Fiz. **65**, No. 4, 142–149 (1995).
2. I.V. Ivlev, K.B. Pavlov, and M.A. Yakovlev, Zh. Tekh. Fiz. **64**, No. 9, 50–59 (1994).
3. G. Haas and R.E. Thun, eds., *Physics of Thin Films* (Academic Press, New York, 1966).
4. A.Yu. Kropanev, A.N. Orlov, and V.V. Osipov, Quant. Electron. **26**, No. 3, 218–221 (1996).
5. G. Haas and J.E. Waylons, J. Opt. Soc. Am. **51**, No. 7, 719–722 (1961).
6. A.M. Bonch-Bruevich, Ya.A. Imas, G.S. Romanov, M.N. Libenson, and L.N. Mal'tsev, Zh. Tekh. Fiz. **38**, No. 5, 851–855 (1968).
7. G.V. Rozenberg, *Optics of Thin-Coat Finishes* (Fizmatgiz, Moscow, 1958), 570 pp.
8. Sokolov, *Optical Properties of Metals* (Fizmatgiz, Moscow, 1961), 464 pp.
9. V.V. Osipov, A.N. Orlov, and V.V. Tsarapkin, "*Method for Cleaning of Optical Surfaces*," Patent of the RF No. 2119217 dated to 10.16.98.
10. A.M. Prokhorov, V.I. Konov, I. Ursu, and I.N. Mikheilesku, *Interaction of Laser Radiation with Metals* (Nauka, Moscow, 1988), 537 pp.
11. Yu.N. Vershinin, *Electron-Thermal and Detonation Processes at Electric Breakdown of Solid Dielectrics* (UrB RAS, Ekaterinburg, 2000), 258 pp.