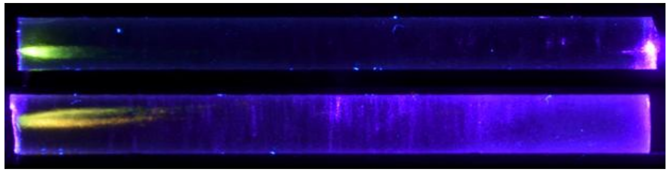
Aggregation and coagulation mechanisms of defects in the process of laser defect formation during filamentation in a crystalline medium

Luminescent defects are effectively created in lithium fluoride crystals under the action of femtosecond laser radiation in the near infrared region of the spectrum. These defects are aggregate color centers. The same defects are formed in lithium fluoride crystals and under the action of various types of ionizing radiation.

The internal photoelectric effect in the interaction of radiation from a femtosecond titanium-sapphire laser (photon energy $h\nu \approx 1{,}4$ эВ) with wide-gap lithium fluoride crystals ($\Delta E \approx 14$ эВ) is due to multiphoton-tunneling interband ionization. The degree of nonlinearity of the interaction of light with crystals at near-threshold intensities ($I \approx 10^{13}$ Вт/см^2) is in the range 8-10.

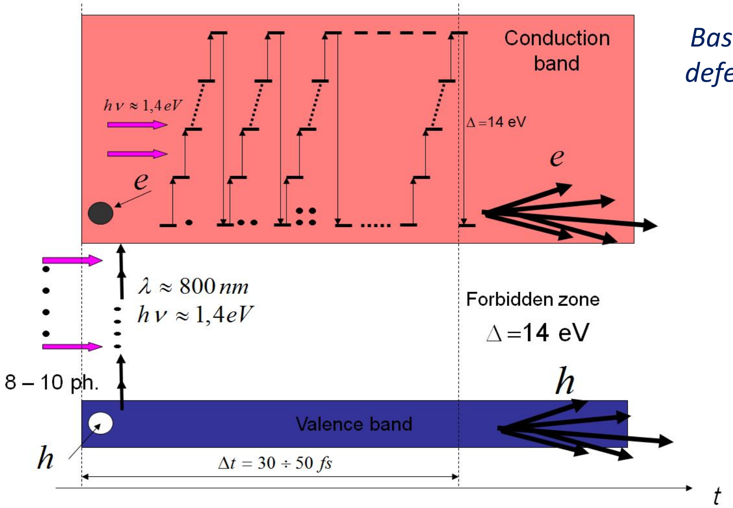
Color centers are distributed along the channels formed in the places of passage of light filaments (filaments) formed as a result of self-focusing of laser radiation.



Photographs of the spatial distribution of luminescence centers in LiF: Mg,Ti crystals irradiated by a series of 5 (top) and 125 (bottom) laser pulses with an energy of 5 mJ and a duration of 50 fs.

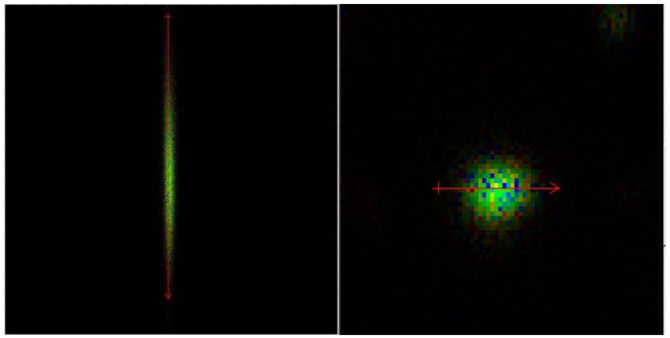
Pattern of the photoluminescence intensity distribution of defects in the head part of the channel induced by a single pulse with an energy of 5 mJ and a duration of 50 fs.
The large number of observed traces of filaments indicates that the peak power of the laser pulse at the entrance to the crystal is hundreds of times higher than the critical self-focusing power.

The mechanism for the creation of color centers under the action of femtosecond laser radiation includes highly nonlinear generation of electron-hole pairs in the region of passage of light filaments, their recombination with the formation of anionic excitons, the decay of excitons into Frenkel defects, their charge exchange, migration, and aggregation.



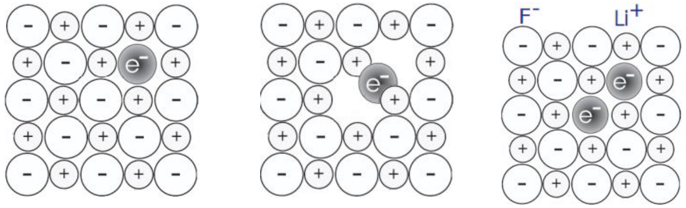
Basic photochemical reactions of defect formation. Arrows indicate defect mobility at room temperature.

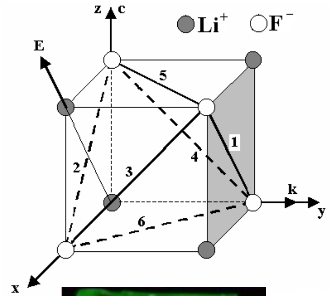
$$m\bullet h\nu \rightarrow e + p$$
$$e + p \rightarrow ex$$
$$ex \rightarrow F + H$$
$$F + p \rightarrow V_a^{+}\uparrow$$
$$V_a^{+}\uparrow + F \rightarrow F_2^{+}\uparrow$$
$$F_2^{+}\uparrow + F \rightarrow F_3^{+}$$
$$F_2^{+}\uparrow + e \rightarrow F_2$$



Longitudinal (left) and transverse (right) distribution of the luminescence intensity of color centers in the track (the place of passage of the light filament), induced by a single pulse (data from a MicroTime 200 microscope). The track length is 32 μm, the diameter is 1.4 μm.

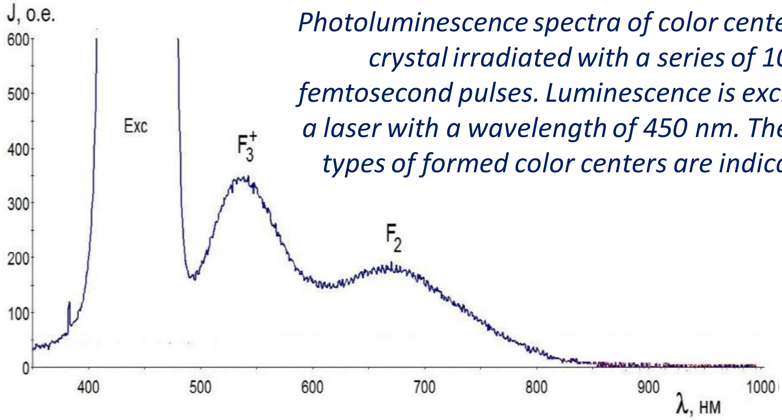
Structure of F (left), F_2^+ (center) and F_2 (right) color centers





Orientation of aggregate color centers in the crystal lattice:

- F_2, F_2^+ and F_2^- - centers, oriented along the diagonal of the cube face;
- F_3, F_3^+ and F_3^- - centers form a plane in the form of an equilateral triangle.



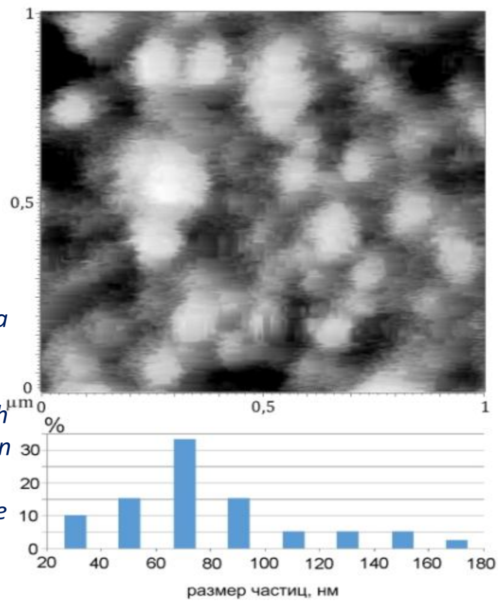
Photoluminescence spectra of color centers of a crystal irradiated with a series of 10 femtosecond pulses. Luminescence is excited by a laser with a wavelength of 450 nm. The main types of formed color centers are indicated.

A large aggregate, for example F_3, the color center, forms a cavity in the crystal lattice, inside which there are cations with electrons distributed on them. This state is unstable; a metal bond arises between the metal atoms with the formation of a colloid - lithium nanoparticles.

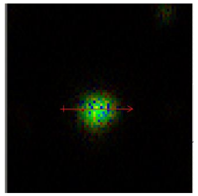
The investigated physical processes can be applied in the technology of recording optical images (information) in crystals.

As an example, the photograph shows an image of a "fairy dragon" recorded in a lithium fluoride crystal by a femtosecond laser in the high-aperture radiation focusing mode. Image size 5x5 mm. To visualize the luminescent image, a laser with a radiation wavelength of 405 nm is used, the shooting is made through a green light filter. In the absence of excitation, the crystal is transparent (photo on the right). In fact, a latent image is formed in the crystal.

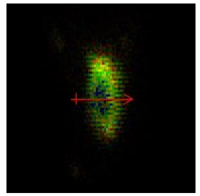
The picture of the spatial and statistical size distribution of colloidal lithium nanoparticles in the cross section of the channel induced by a series of 3000 femtosecond laser pulses, obtained with a Certus Light scanning probe microscope.



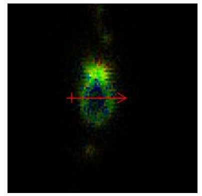
Dynamics of changes in the profile of the track cross section with an increase in the number of femtosecond laser pulses in a series.



1 pulse
diameter -1.4 µm



1000 impulses
diameter - 5.9 µm



3000 impulses
diameter - 8.3 µm

The concentration of defects increases with approaching the center of the channel formed by the light filament. At the periphery of the filament trace, there are predominantly stable F centers, then an area with F_2 color centers. As we approach the center of the filament trace, the place of localization during irradiation of the maximum temperature and concentration of primary defects, a region with a high concentration of complex F_2, F_3^-, F_3^+, F_3, F_4 aggregate centers is formed. In the center, there is a heavily damaged lattice region containing colloidal lithium nanoparticles.