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ИЗВЕСТИЯ

НАЦИОНАЛЬНОЙ АКАДЕМИИ НАУК
РЕСПУБЛИКИ КАЗАХСТАН

**СЕРИЯ
ФИЗИКО-МАТЕМАТИЧЕСКАЯ**

5 (255)

СЕНТЯБРЬ–ОКТЯБРЬ 2007 г.

ИЗДАЕТСЯ С ЯНВАРЯ 1963 ГОДА

ВЫХОДИТ 6 РАЗ В ГОД

АЛМАТЫ
НАН РК

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INFLUENCE OF PULSE ELECTRONIC IRRADIATION POWER ON PROCESSES OF F_2 CENTERS ACCUMULATION IN LiF CRYSTALS

The work presents the research results of F_2 - centers accumulation in LiF crystals under electron pulse irradiation and analytical description F_2 accumulation process in the range of integral doses not exceeding $2{\cdot}10^4$ Gy.

Introduction

Researches with the use of time-resolved pulse spectrometry methods carried out earlier determined that processes of F_2 centers formation and destruction under nanosecond electron pulses (EP) in LiF crystals at 300K have completely different kinetic parameters and they may be divided according to the period of their passing after the EP action is completed [1].

It has been found:

– at time interval $\Delta t \le 10^{-8}$s there take place the following reactions of F_2 centers transformation:

$$F_2(S_0)+H \Rightarrow F_1, \quad (1)$$

$$F_2(S_0)+ p \Rightarrow F_2^+ + e^o \Rightarrow (F_2^+)^* \Rightarrow F_2^+ + h\nu\,(F_2^+), \quad (2)$$

$$F_2^+ + e \Rightarrow F_2(S_j)...F_2(S_1) \Rightarrow F_2(S_0) + h\nu(F_2), \quad (3)$$
$$\searrow F_2(T_j)$$

$$F_2(T) + e \Rightarrow F_2^-; \quad (4)$$

– at the time interval $10^{-4} \ge \Delta t > 10^{-8}$ s after the EP action is completed:

$$F_2(S_0) + V_k \Rightarrow F_2^+ + e^o \Rightarrow (F_2^+)^* \Rightarrow F_2^+ + h\nu(F_2^+), \quad (5)$$

$$F_2^- + V_k \Rightarrow F_2 + e^o \Rightarrow (F_2)^* \Rightarrow F_2(S_0) + h\nu(F_2), \quad (6)$$

– at the time interval $10^{-1} \ge \Delta t > 10^{-4}$ s after the EP action is completed:

$$F_2(T_1) \Rightarrow F_2(S_0); \quad (7)$$

– at the time interval $10^2 \ge \Delta t > 10^{-1}$ s after the EP action is completed:

$$F_1 + \alpha \Rightarrow F_2^+ \quad (8)$$

$$F_1^- + \alpha \Rightarrow F_2(S_0), \quad (9)$$

$$F_2(S_0) + \alpha \Rightarrow F_3^+; \quad (10)$$

– at the time interval $10^4 \ge \Delta t > 10^2$ s after the EP action is completed:

$$F_2^+ + F_2^- \Rightarrow F_2(S_0), \quad (11)$$

where e^o – next to the center exciton; p and V_k – band hole and self-trapped hole respectively; S_j, S_1, S_0, T_i, T_j, T_1 –terms of singlet and triplet states of F_2 center respectively; H, α, F_1, F_2^+, F_2, F_2^-, F_3^+- notations of corresponding color centers.

It is obvious that efficiency of F_2 centers accumulation in the crystal under EP is determined by correlation between direct processes of creation and inverse processes of destruction of these centers. The existence of a great number of mechanisms of F_2 centers formation and destruction initiated by EP action causes considerable difficulties in trying to give analytical description of regularity of F_2 centers accumulation with the growth of integral absorbed dose. The solution of this task in corpora at present is impossible, as so-called feedback – the influence of different radiation defects, accumulated simultaneously with F_2 centers, both on the efficiency of already known creation and destruction processes, and on the efficiency of formation of primary defects – Frenkel pairs - hasn't been studied practically [2-7].

Experiment procedure

The given work was done in order to evaluate the possibility of using value of radiation average power as a parameter, unambiguously determining the result of radiation action, during analytical description of F_2 centers accumulation process.

Simulation of the radiation field of different average power was carried out in two ways: by changing of the frequency (f) of EP repetition, or by changing of the magnitude of electron fluence over pulse (Φ). The magnitude of electron energy (E) was invariable and equals to 200keV, duration of EP equels 20ns. EP repetition frequency varied over the range $1{\cdot}10^{-1}$–$1{\cdot}10^{-3}$Hz. The magnitude of electron fluence over pulse varied over the range 10^{10}–10^{13}см$^{-2}$ generating in crystal 10^{15}–10^{18}cm^{-3} of electron-hole pairs respectively. The research of F_2 centers accumulation as a function of integral absorbed dose was carried out in LiF crystals under irradiation by EP series.

Information about the processes, leading to F_2 centers accumulation in different parts of dose dependence, was taken during investigation of kinetic relaxation of absorption (KRA) in maximum of various bands in spectra of preliminary irradiated LiF crystals in wide time interval (10^{-8}–10^4 s) after the EP action is completed.

Experiment results

The action on crystal of EP series at 300K leads to accumulation of F_2 centers in the crystal. Dose dependence of a number of F_2 centers, accumulated

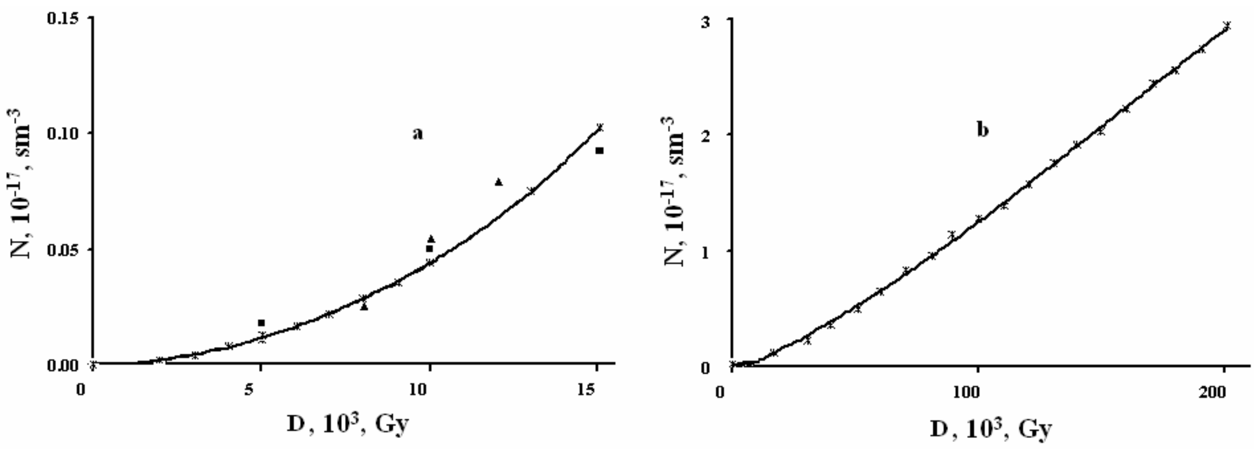

Fig. 1. Concentration of accumulated F_2 centers as a function of absorbed integral dose magnitude under irradiation of LiF crystal by EP series with repetition frequency $f=10^{-2}$ Hz in the area of doses $D\leq2\cdot10^4$ (a), $D\leq2\cdot10^5$ Gy (b)

in the crystal, has a complicated nature and may be presented in the form of superposition of two laws: parabolic – in the area of small values of integral absorbed dose ($D\leq2\cdot10^4$ Gy) and linear – in the area $10^5\geq D>2\cdot10^4$ Gy (Fig. 1).

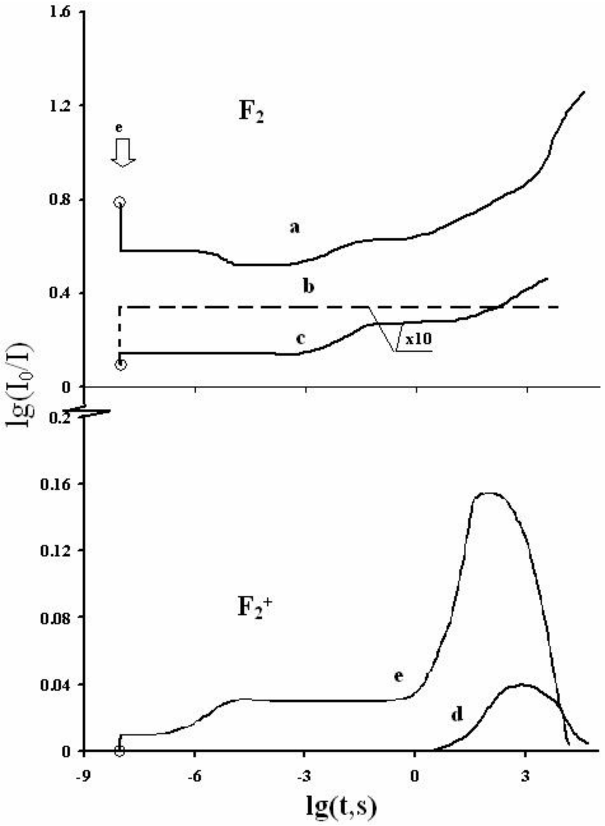

Fig. 2. Kinetics of relaxation of optical absorption in maximum F_2 (a-c) and F_2^+ (d, e) bands, initiated by the action of single EP at 300 (a,c-e) and 80K (b) on LiF crystals, integral doses of previous irradiation of which are $4\cdot10^3$ (b, c, d) and $2\cdot10^5$Gy (a, e)

During of the investigation of KRA in maximum of F_2 band, initiated by the action of single EP in different parts of dose dependence it has been found that in the area of small values of integral doses (parabolic part) in the crystal under EP mainly F_1 and F_2^+ centers are formed. KRA in maximum of F_2 band, initiated by the action of single EP on such a crystal, indicates that at 80K there exists an only noninertial (in relation to EP action) F_2 centers formation (Fig. 2, curve b) according to (3). At 300K F_2 centers formation takes place in nano-, milli- and second time intervals in relation to EP action (Fig. 2, curve c). Corresponding processes are given in (3), (7), (9).

The crystal, preirradiated in the area of doses>$2\cdot10^4$ Gy (the area of linear dependence of a number of accumulated F_2 centers in Fig. 1), contains the following set of accumulated centers: F_1, F_2^+, F_2, F_3^+, F_2^-. Action of single EP on the crystal initiates KRA in peak of F_2 band, which indicates about the dominance of the processes of destruction of accumulated F_2 centers at nano- and microsecond time intervals according to (1), (2), (5) and about the creation of F_2 centers at milli-, second and hour time intervals in relation to EP action (Fig. 2, curve a). Corresponding processes are described in (7), (9), (11).

From comparison of the results, presented in Fig. 2 (curves a, c), it becomes clear that at 300K in the area of small doses ($D\leq2\cdot10^4$ Gy) increase of a number of F_2 centers under single EP action takes place mainly while realizing millisecond mechanism, whereas in the area of large doses of preirradiation, millisecond component of F_2 centers creation initiated by EP does not provide an increase of number of

F_2 centers and even restoration of those F_2 centers destroyed by the same EP. Increase of a number of F_2 centers as a result of EP action in this case is carried out only as a result of inertial process at time $\leq 10^1$ s in relation to EP action at 300K.The discovered effect indicates that on parabolic and linear parts of dose dependence (Fig. 1) F_2 centers accumulation is determined by various set of mechanisms of their destruction and formation. This is one of the causes of the change of F_2 centers accumulation speed with the growth of integral absorbed dose.

Considerable differences of time parameters of F_2 centers transformation mechanisms allow, if to appropriately choose repetition frequency of EP, to exclude participation of long-time F_2 centers transformation mechanisms in accumulation. Then in the area of small absorbed doses under definite values of EP repetition frequency, it is possible to create conditions, under which

F_2 centers accumulation will be carried out upon realization of only short-time mechanisms of F_2 centers creation, appearing in the form of nano- and millisecond components of increase on KRA in peak of F_2 band. This, in its turn, makes possible analytical description of accumulation process.

It is obvious that maximal EP repetition frequency for realization of millisecond mechanism of F_2 centers formation in the main singlet state should not exceed $1/\tau$, where τ - characteristic time of the process.

F_2 centers formation at nano- and millisecond time intervals in relation to EP action by [8] takes place as a result of electron capture by F_2^+center. Whereas in the area 80K there are formed F_2 centers mainly in excited singlet state with the consequent relaxation by sublevels of the same multiplicity: S-S_o. In the area of high temperatures there occur predominant formations of F_2 centers with time delay with respect to EP because of relaxation of the formed center by sublevels of various multiplicity: S-T-S_o.

The corresponding processes are the following:

$$F_2^+ + e \Rightarrow F_2(S_j)...F_2(S_1) \overset{\tau(S)}{\Rightarrow} F_2(S_0) + h\nu(F_2) \quad (12)$$
$$\searrow \qquad \tau(T)$$
$$F_2(T_i)...F_2(T_1) \Rightarrow F_2(S_0),$$

where: $\tau(S)$, $\tau(T)$ - the life time of F_2 center in excited singlet and triplet states, which at 300K are equal to $17\cdot10^{-9}$ and $5\cdot10^{-3}$s respectively. Activation energes of intercombination S_j-T_i and T_1-S_o transitions were found to be equal to 0.06 and 0.04eV respectively.

As the life time of F_2 center in radiative singlet state is comparable to duration of EP, S_1-S_o transition, noninertial in relation to EP action, determines by itself nanosecond component of absorption growth on KRA in maximum of F_2 band (Fig. 2, curve b). T_1 -S_0 transition in F_2 center appears in the form of millisecond component of growth on KRA in peak of F_2 band (Fig. 2, curves a, b).

In [9] one can see that nanosecond stage of F_2 centers formation is not the result of spatial correlation, leading to the decay of electron excitation in the area of existing F_2 center according to the reaction:

$$F_1 + e^o \Rightarrow F_1 + F_1 + H \Rightarrow F_2(S_0) + H.$$

Analytical description of F_2 centers accumulation process

For quantitative description of the number of created by (12) F_2 centers in the ground singlet state as the result of electron capture on F_2^+ center, necessary information about the processes of creation in the crystal in the process of irradiation of F_2^+ centers has been received in [10] during studying KRA in maximum of F_2^+ absorption band initiated by EP action.

As appears from the presented in fig. 2 (curves d, e) results, in the general case one can observe three mechanisms of irradiated creation of F_2^+ centers: at nano-, micro- and second time intervals in relation to EP action at 300K. The first two processes lead to F_2^+ centers creation as a consequence of destruction of a part of F_2 centers as a result of band or self-trapped hole capture by (2) and (5). At the third time interval F_2^+ centers formation is described by the reaction (8), where α centers by [10] are secondary in LiF crystals, i.e. created as the result of holes capture by F_1 centers).

As it appears from the presented in Fig. 2 (curves d, e) results, contribution of various reactions into general number of F_2^+ centers, accumulated in the crystal, depends on crystal preliminary irradiation dose. In the area of small values of absorbed doses ($D<10^4$ Gy) there prevails second component of creation and hour component of post irradiated destruction of centers (curve d). In the area of high doses of preliminary irradiation, the KRA in maximum of F_2 band indicates F_2^+ centers creation at nano- by (2) and in microsecond by (5) time intervals during simultaneous destruction of F_2 centers in the same time intervals (compare Fig. 2, curves a, e).

The expression, describing kinetics of F_2^+ centers number change in the area of small absorbed doses, initiated by the action of single EP number i presented in fig. 2 (curve d), is the following:

$$\Delta F_2^+(i,t)$$

$$\gamma\Phi E\left[F_1\right]_{i-1}(1-\exp(-t/\tau_1))\exp(-t/\tau_2), \quad (13)$$

where t – time from the moment of the end of i -EP action; τ_1 ,τ_2 – characteristic time of F_2^+ centers of formation and destruction processes respectively; γ- constant coefficient, independent of temperature [10]; E- electrons energy, Φ- fluence of electrons on pulse; $[F_1]_{i-1}$- number of F_1 centers, accumulated in the crystal as a consequence of action of the series of (i-1) pulses.

Values of characteristic time of F_2^+ centers creation and destruction process in (13) depend on crystal temperature during irradiation:

$$\tau_n^{-1} = \nu(n)\exp(-E(n)/kT),$$

where for n=1 (creation process) E(1)= 0.66eV, τ_1 =20s at 300K. For n=2 (destruction process) – E(2)=1eV , $\tau_2 = 2\cdot10^4$s at 300K [10].

Maximum of value $\Delta F_2^+(i,t)$, described in (13), appears when

$$t = t_0 = \tau_1 \ln(\tau_2/\tau_1). \quad (14)$$

During irradiation of crystal by EP series, as a result of each following EP action, all the existing in crystal F_2^+ centers, created in time interval between successive EP, capture electron and are transformed into F_2 centers. Characteristic time of F_2 centers formation process in ground singlet state by (12) is determined by their life time in excited singlet and triplet states: τ(S) and τ(T).

In the area of frequencies 1/f>>τ(T) F_2 centers creation process efficiency will be determined only by relation between 1/f and values of characteristic time of processes, determining F_2^+ centers creation and destruction: τ_1 and τ_2 in (13).

Taking into account (13), the number of accumulated in the crystal F_2 centers under the action of series of k electron pulses can be described by the expression, when t′=1/f ′ (f ′- EP repetition frequency):

$$\left[F_2\right]_k = \sum_{i=2}^{k} \Delta F_2^+(k-1,t') \quad (15)$$

$$\sum_{i=2}^{k} \gamma\Phi E\left[F_1\right]_{i-1}(1-\exp(-t'/\tau_1))\exp(-t'/\tau_2).$$

The number of F_2 centers destroyed under EP is by an order of magnitude smaller than the number of newly created F_2 centers by the same EP that is why in (15) we does not take into account F_2 destroy process.

In the range of small integral doses in (15) $[F_1]_{i-1} \approx iF_1$, where F_1 - the number of F_1 centers formed by single EP. Then:

$$\sum^{k}\left[F_1\right] \approx k^2\mu \; E\Phi, \quad (16)$$

where μ - coefficient, dependent on temperature.

When τ_2 >>1/f ′≈τ_1, limiting by the first two expansion terms of exponent in (15), and taking into account (16) we get:

$$\left[F_2\right]_k \approx \gamma \;\; \mu \;\; k^2\Phi^2E^2/f'\tau_1 \;\;\; \gamma \;\; \mu \;\; \Delta \; tD^2P/P'\tau_1, \quad (17)$$

where D=kEΦ – integral absorbed dose with crystal thickness more than electron penetration depth; W′=EΦf ′ – average power of irradiation; W=EΦ/δt – irradiation peak power; δt – electron pulse duration.

From (17) it follows:

1) The number of accumulated F_2 centers is proportionate to the square of integral absorbed dose, that has experimental verification in the area of small doses (Fig. 1, curve a).

In particular case, when f=const and D=const the number of accumulated F_2 centers will be the same under EP with no equal values of peak power. This conclusion is verified by experiment results, presented in Fig. 3 (curves a, c)

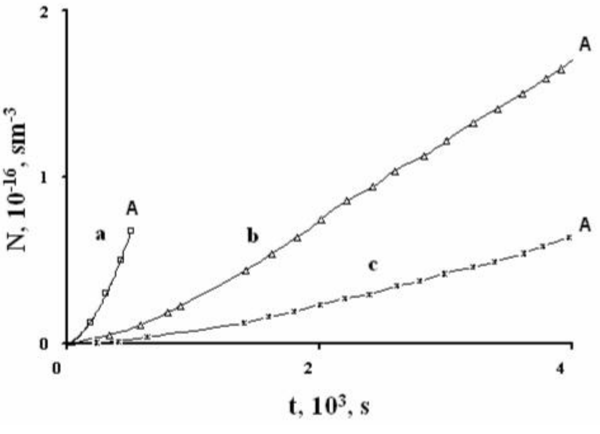

Fig. 3. Concentration of accumulated F_2 centers in LiF crystal as a function of radiation time at 300K. Values of average power of radiation: $8\cdot10^{-1}$ (a), $9\cdot10^{-2}$ W см$^{-3}$ (b, c). EP repetition frequency - $5\cdot10^{-2}$ (a, c), $6\cdot10^{-3}$Hz (b). Electron fluence over pulse - $1\cdot10^{13}$ (a, b), $1.2\cdot10^{12}$ см$^{-2}$ (c). (At points A on different curves D=const)

2) When W′=const and D=const the number of accumulated F_2 centers is increasing with the growth of peak power if there is equality of values of average radiation power. Conclusion is verified experimentally (Fig. 3, curves b,c)

3) When D=const and W=const the number of accumulated F_2 centers is decreasing with the growth of average radiation power. It means that F_2 centers accumulation efficiency increases with the decrease of EP repetition frequency under equality of peak power values. This conclusion also has experimental verification (Fig. 3, curves a, b). It should be noted that frequency $f_o=1/t_o$, where t_o is determined by (14), is cutoff frequency, below which the increase of the number of accumulated F_2 centers, created by (12), will not be observed.

So, the number of accumulated F_2 centers in crystal is the function of at least three parameters of radiation field: of integral absorbed dose of radiation (D), of peak power (W) and pulse repetition frequency (f). The result of F_2 centers accumulation under irradiation with equal values of average power (W') and radiation dose (D) differs greatly for radiation regimes with various peak values of power (W).

In conclusion, it should be noted that the change of type of particle without the change of their energy is equivalent to the change of radiation peak power. Consequently, the result of F_2 centers accumulation under action on material of fluxes of different types of particles on condition that two other parameters of irradiation - magnitudes of absorbed integral dose and average radiation power are equal, will be different. This effect can be used for analysis of type of radiation fields.

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Резюме

Импульстік электронды сәулелендірудің әртүрлі параметрлерінде LiF кристалындағы F_2-центрлердің жинақталу процестерін зерттеу нәтижелері көрсетілген. Интегралдық өлшемі 2Ч10^4 Гр-дан аспайтын аумағындағы центрлердің жинақталу процестерінің аналитикалық сипаттамасы берілген.

Резюме

Представлены результаты исследования процессов накопления F_2-центров в кристалле LiF при различных параметрах импульсного облучения. Дано аналитическое описание процесса накопления центров в области интегральных доз не превышающих 2·10^4 Гр.

L. N. Gumilyov Eurasian National university

Поступила 4.06.07г.

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