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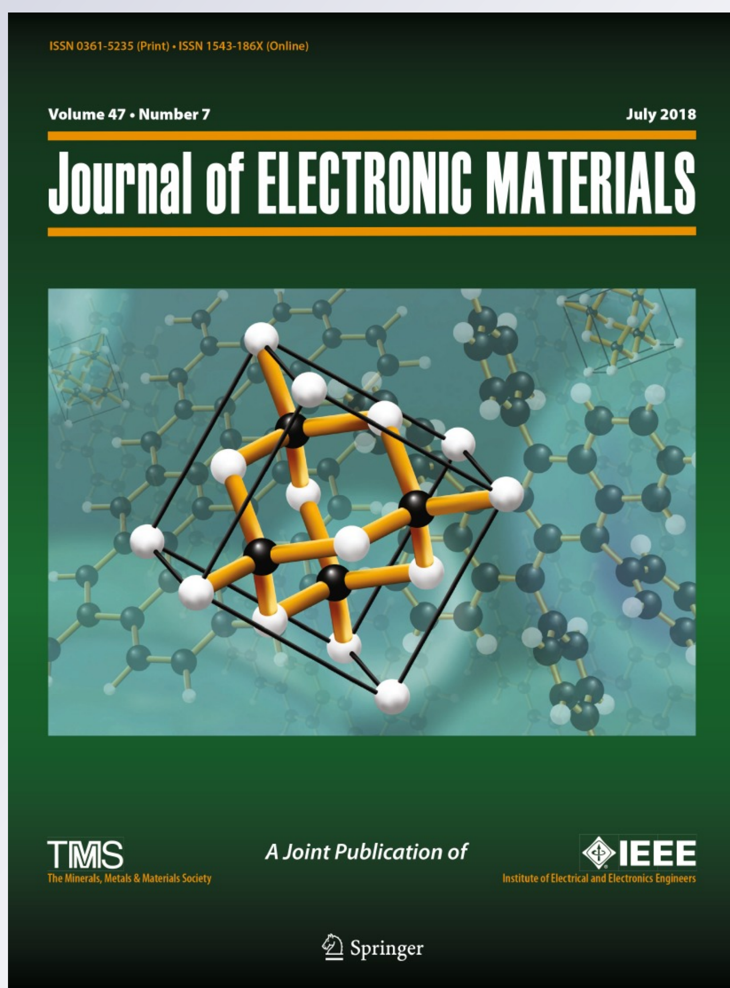

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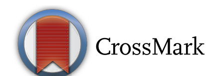

The Use of Different Pulsed Electron Irradiation for the Formation of Radiation Defects in Silicon Crystals

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This paper reports the formation of structural defects in the lattice of silicon (n-Si) single crystals, as a result of irradiation by different intensities and pulses of electrons. The samples were studied by means of Hall effect measurements of electro-physical parameters (specifically the concentration of the main charge carriers) as a function of temperature and radiation dose. The role of the radiation current density (pulse height) is discussed, which gives rise to a peculiar behavior in the electrical-physical properties of n-Si. In particular, thermal processes are found not to develop, due to the ultrafast (pulse duration in the range 10^{-12}–10^{-13}s) nature of the incident radiation, which causes an almost "pure" energy interaction to occur between the radiation and the atoms within the crystal, and the formation of cluster defects. A scheme for the time-scale of the formation of these radiation defects is presented. From the dose and temperature dependences of the concentration of main charge carriers, the radiation defects introduction rates were determined.

Key words: Silicon crystal, irradiation, electro-physical properties, radiation defects, pulse irradiation

INTRODUCTION

Studying the kinetics of formation and annealing of defects induced by different types of irradiation, particularly high density pulse irradiation, in semiconductors and semiconductor devices, is of special interest for solid state radiation physics, because it furnishes significant information about the temperature stability and time stability of radiation defects in materials and devices. The effect of high density pulse irradiation—which involves the intensive ionization of semiconductors, with the formation of additional non-equilibrium charge carriers—is important, since it may give rise to large non-stationary currents and electrostatic forces in actual electronic devices.

In this paper, we discuss the formation of radiation defects by electron irradiation, giving special consideration to the influence of high intensity pulses of electrons, which are applied over a very short time interval (10^{-9} s or less), and how these affect the properties of a particular material—crystalline silicon. The high flux density of this radiation is able to stimulate novel effects which are practically absent when "long pulse duration" (about 10^{-6} s) irradiation is employed. Thus, when short pulses of radiation are applied, those processes which are initiated both in the bulk material and on its surface do not cease when the irradiation is terminated, but continue for a given time. These processes result in the formation of radiation defects, which are stable at a given temperature,

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but cause irreversible changes to occur in the characteristics of the irradiated materials and, hence, of devices made from them. The duration of these processes depends on both the sample temperature and the concentrations of impurity species in the material, which interact with the radiation defects.[1–4]

Conventional pulse sources employ pulses of electrons with duration in the region of 10^{-6}–10^{-7} s, which is within the timescale over which thermal processes occur.[5–9] Accordingly, the energy absorbed within the crystal is consumed by ionization processes, i.e., the formation of free electrons in the conduction band, and holes in the valence band, and by the creation of heat in the semiconductor material. If the radiation pulse is applied with a duration of the order of 10^{-12} s, from a very high density electron beam (about 100 Amp.), which is a unique feature of the present study, it is less probable that thermal processes will be developed than is the case when longer pulse durations are employed[10] and Fig. 1. However, it is important to be aware that such an increase in the radiation flux density may cause the following effects[11,12]:

1. A change in the charge carrier concentrations in the conduction and valence bands, which alters the charge state of any interacting defects and accordingly the secondary defect accumulation rate caused by the irradiation process.
2. An increase in the rate of production of Frenkel pairs (vacancies—V and interstitial atoms—I). The defect formation processes may also differ from those which arise from low density flux irradiation, since the individual primary defects begin to interact to form secondary complex defects, and partial annealing of the sample also occurs. While irradiation at low density fluxes results in a linear accumulation of Frenkel pairs with dose, the relationship becomes non-linear under conditions of high density flux radiation, but the form of the dependency can be fitted empirically.[4–6]
3. Saturation of the concentration of complex defects, which occurs as the irradiation dose is increased, occurs more rapidly at high current densities than at low current densities.

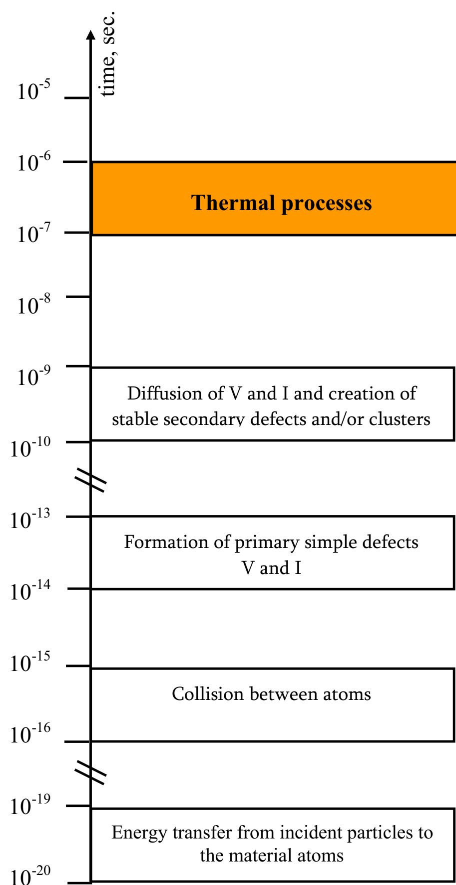

Fig. 1. Schematic diagram of the time—scales occupied by the different processes involved in the formation of radiation defects.

CONDITIONS AND CIRCUMSTANCES FOR THE INFLUENCE OF RADIATION PULSE DENSITY

It is possible to make a preliminary estimate of the effect of the radiation current density on the silicon crystal in terms of the abovementioned factors. For example, at an irradiation current density of $J \sim 3$ mA/cm^2 per s (pulse duration about one microsecond, with an energy in the range of 3–5 MeV), assuming an intrinsic concentration of silicon atoms (N_{Si}) = 5 × 10^{22} cm^{-3}, a cross-section for Frenkel pair production (Σ) = 5 × 10^{-24} cm^2, and the lifetime of vacancy capture by the impurity of τ = 3 × 10^{-6} s, we obtain for a stationary concentration of vacancies: $N_{\mathrm{v}} = N_{\mathrm{Si}} \times J \times \Sigma \times \tau = 1.6 \times 10^{10}$ cm^{-3}, which is significantly lower than the impurity concentration usually present in silicon crystals.[11,12] Hence, the average distance between vacancies is much larger than that between vacancy and impurity atoms, and the dominant process is the formation of vacancy—impurity atom complexes, such as A—center (oxygen concentration about 10^{18} cm^{-3}) or E—center (impurity concentration about 10^{17} cm^{-3}).[3,4,11–14] Clearly, this result is not compatible with the condition 2 above, and it is therefore necessary to apply a current density (J) of greater than 1 Ampere, which was achieved in the present study by means of pico-second pulse irradiation.

In the case of a very short irradiation pulse, the average time for the charge carriers to be collected from the bulk and from the peripheral areas of the surface is usually different, because the recombination rates of minority carriers in the bulk volume and on the surface are different. As a result of this

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phenomenon, radiation-generated electrons and holes move to the traps and provide a normal ionization current, which contains two components: immediate and delayed. Accordingly, it is necessary to consider such factors as the charge redistribution time, the rates of different carrier recombination processes (in the volume and on the surface), specific aspects of the sample geometry and other aspects, most of which cannot be anticipated in advance.

For those samples that are exposed to radiation at a high flux density—for example, γ-quanta with intensities of $\varphi_\gamma > 10^7$–10^8 Rad/s., or electrons with an energy of 3–5 MeV and an intensity of greater than 100 pico-Coulomb/pico-sec = 100 Ampere—it is possible that the large current that is generated, might create an accelerating electrical field that is able to influence the nature of the radiation—induced process.[12,15] The latter, in turn, will increase both the current and the electrical field tension. Such mechanism of current multiplication, rather like a positive feedback effect, can lead to instability and an exponential increase in the current over time. A stable condition is restored when, as a result of their recombination, the concentration of the non-equilibrium charge carriers decreases to a level at which the current from them is no longer supported by the electrical field.[11]

Interesting properties have been observed in silicon crystals following their irradiation by high energy particles, which create disordered regions in the crystal lattice.[16] In particular, there are high sensitivity to light and long relaxation time for the photocurrent observed which demonstrate a "memory effect", and potential technical applications.

We note that those recombination processes, which occur over longer timescales, are not related to low cross-section recombination at the trapping centers, but are determined by the maximum lifetime, as is expressed by[16]:

$$\tau_{\max} = (v \times S_{\min} \times \Delta n)^{-1}, \quad (1)$$

where v is the thermal velocity of charge carriers (at room temperature), Δn the concentration of non-equilibrium carriers, $S_{\min}$ the lowest value of cross-section of those carriers that are captured by the trapping centers (which is defined as 10^{-23} cm^2). Relaxation processes that occur over a timescale greater than $\tau_{\max}$, may be related to the longer timescale events, which are caused by the non-uniform distribution of carriers as a result of radiation defect formation.[12,17–21] In such cases, the following values apply: $v = 10^7$ cm/s, $\Delta n \sim 10^{13}$–10^{14} cm^{-3}, $S_{\min} \sim 10^{-23}$ cm^2, and, hence, from (1) we can obtain $\tau_{\max} \sim 10^2$—10^3 s., which is a timescale of sufficient length for non-equilibrium carriers to recombine.[9,16] In regard to immediate processes and respective carrier lifetimes, we can determine that the timescale for atoms to be moved to their appropriate inter-atomic distances, (see Fig. 1) is:

$$\tau_{\text{rem}} \sim 10^{-8}\text{cm}/10^6(\text{cm/s}) \sim 10^{-14}\ \text{s}. \quad (2)$$

Although this is difficult to set constant during the experiment, it is necessary to adopt this value in all cases in order to make the measurements reliable. The nature of long—term processes is characterized by an electrostatic potential barrier, U, which surrounds the radiation-induced defect regions and causes a spatial separation of minority and majority carriers. Minority carriers are attracted to the disordered regions where they recombine, while abundant majority carriers must penetrate to the interior of the disordered regions, surmounting the potential barrier, in order to return the system to a condition of equilibrium.[12]

RESULTS AND DISCUSSION

Electron irradiation was performed at ambient temperatures with a linear accelerator which delivered a pico-second pulsed electron beam, with an energy of 3.5 MeV, at the « CANDLE » (Center for the Advancement of Natural Discoveries Using Light Emission) Synchrotron Research Institute (Armenia). The pulse frequency was 25 Hz, and the beam current was measured by charge accumulation in a Faraday cup; the beam intensity was 3×10^9 el/cm$^2 \times$ s. Other electron irradiations were performed using a pulse duration in the region of one microsecond from the linear accelerators housed in the Yerevan Physics Institute, with energies of 3.5 MeV and 50 MeV and a pulse frequency of 200 Hz.

Czochralski grown n-type silicon was chosen, with an oxygen concentration of 1.25×10^{18} cm^{-3} (defined by IR absorption) and a principal impurity (phosphorus) of less than 10^{16} cm^{-3}. Hall effect measurements were made on silicon samples with the following parameters: 0.5 mm thickness, 3 mm width, 10 mm length, 5 mm distance between conductivity contacts, which had a double-cross shape. The measurements were carried out, both before and after receiving defined radiation doses, over the temperature interval 80–300 K, with each temperature point being accurate to within 0.5 K. The electrical parameters of the samples were determined from Hall effect measurements with the following values: magnetic field induction 0.5 Tesla, direct current through sample from 100 μA to 2 mA, and an input resistance of the voltmeter of 10^{12} Ω. The determination of the irradiation doses and the calculation of the electro-physical parameters of the samples were carried out as we have described previously.[10]

From the known temperature dependence of the concentration of the charge carriers (n), the position of the radiation defect center energy level in the forbidden gap of the semiconductor zone structure was calculated, taking into account the location of the Fermi-level.

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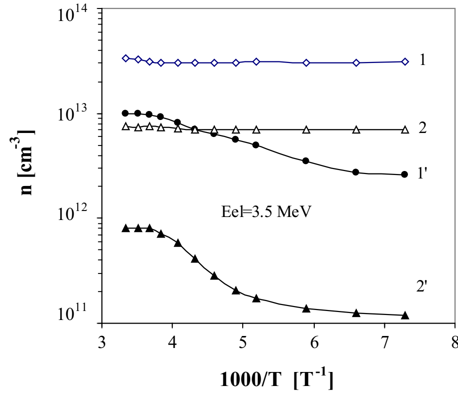

Fig. 2. Temperature dependence of *n*-type silicon main charge carrier concentration: before and after irradiation by pico-second pulse electron beam with energy of 3.5 MeV. The initial specific resistances of the samples were: $\rho = 124\ \Omega \times$ cm, 1—before irradiation, 1′—after irradiation with a dose of 6×10^{13} el/cm^2; $\rho = 710\ \Omega \times$ cm, 2—before irradiation, 2′—after irradiation with a dose of 1.2×10^{13} el/cm^2.

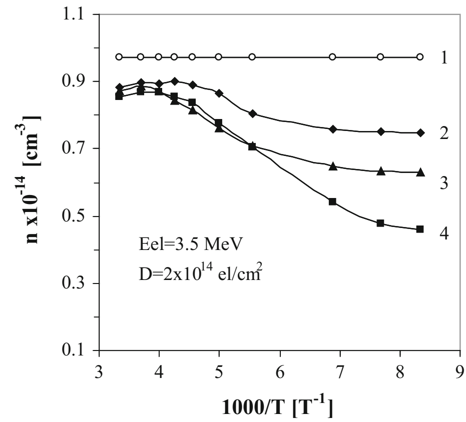

Fig. 3. Temperature dependence of *n*-type silicon main charge carrier concentration: before and after irradiation by microsecond pulse electrons with an energy of 3.5 MeV and dose 2×10^{14} el/cm^2, at different intensities: 1—before irradiation, 2—after irradiation with 2×10^{11} el/cm^2 $\times$ s; 3—after irradiation with 2×10^{12} el/cm^2 $\times$ s; 4—after irradiation with 2×10^{13} el/cm^2 $\times$ s; the initial specific resistance of the sample was: $\rho = 40\ \Omega \times$ cm.

The results of the measurements are presented in Figs. 2, 3, 4, and 5, which show the variation of the major electrical parameters for silicon, each determined as a function of temperature and irradiation dose. Strikingly, the values and trends in the parameters are the same as those previously measured for radiation defect clusters, i.e., a cluster of oxygen containing centers, when pico-second pulse irradiation was used (Fig. 2).[10] In Fig. 3 are presented the temperature dependences of the concentrations of the main charge carriers in *n*-Si irradiated with 3.5 MeV electrons (pulse duration about 5 microseconds) which show the formation of different radiation defects at different intensities even for the same dose. In addition, comparison of Figs. 2 and 3 shows that a large difference between the saturation levels of carrier concentrations before and after irradiation exists. In the case of pico-second pulse irradiation, this difference is almost an order of magnitude, while in the case of microsecond pulse irradiation this difference is much smaller. Note, that the slopes of the curves indicate the energy levels.

The radiation dose dependences of charge carrier concentrations for samples irradiated by picosecond pulses of energy 3.5 MeV and microsecond pulses of energy 50 MeV, presented in Fig. 4a, b are shown on a large scale because the behavior of the concentration of charge carriers, and; hence, the radiation defects introduced in samples with a lower specific resistance, is more precisely expressed at large radiation doses. The behavior of both temperature and dose dependences show a cluster character of radiation defects at a picosecond pulse of energy 3.5 MeV and a microsecond pulse of an energy and 50 MeV irradiation, despite the irradiation energy being much lower in the first case.

In Fig. 5 is presented the concentration of radiation defects produced by $\Delta N_{\mathrm{def}}\ (D) = n_0 - n(D)$, where n_0 is the concentration of main charge carriers before irradiation, n(D) is the concentration of main charge carriers after irradiation to a given dose, which shows the "sensitivity to defect formation" of the irradiated material according to the irradiation dose and the specific resistance of the sample.

To interpret the results in more detail, we may consider the influence of the charge state of the reacting defects in regard to the barrier (U) for the formation of defect complexes (for example A- or E-centers) as a function of irradiation intensity (J). When two defects, with charges Z_1e and Z_2e, interact, the barrier U abruptly changes to $Z_1Z_2e^2/r$ (Coulomb energy, r—distance between defects, e—electron charge), but the charge on the defect is defined by the difference between the energy level on the defect and the Fermi level (quasi-level) in the forbidden gap.[17] Due to the ionization changes in the crystal, the proportion of defects for a given charge state changes quasi-continuously, so altering the accumulation rate of complexes, similarly to showing how the interaction of two individual defects is altered when their charge is changed. Hence, in the dependency of defect formation on irradiation intensity (J), it may be assumed that U is a linear function of J,[12,18] i.e.:

$$U = U_0 + kxJ; \tag{3}$$

where U_0 is the potential barrier at $J = 0$, and k is the proportionality coefficient (number of electron–hole pairs produced by one irradiating particle). In this treatment, temperature dependencies are not considered because the relevant radiation processes have a low probability on the picosecond timescale. Thus, (given that the accumulation rate of A- or E-centers depends linearly on radiation intensity) the

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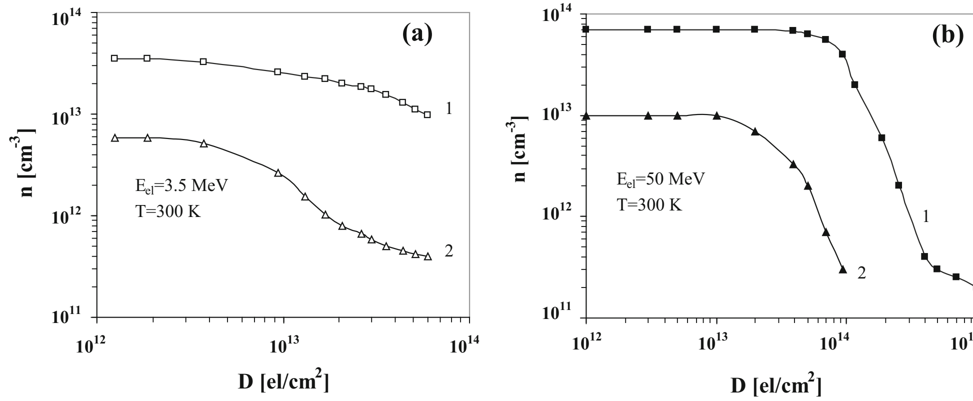

Fig. 4. Dependence of *n*-type silicon main charge carrier concentration on electron irradiation dose. (a) Irradiation by Picosecond pulse electrons beam with an energy of 3.5 MeV; The initial specific resistances of samples were: 1—ρ = 124 Ω × cm; 2—ρ = 710 Ω × cm. (b) Irradiation by microsecond pulse electrons beam with an energy of 50 MeV; The initial specific resistances of samples were: 1—ρ = 100 Ω × cm; 2—ρ = 450 Ω × cm.

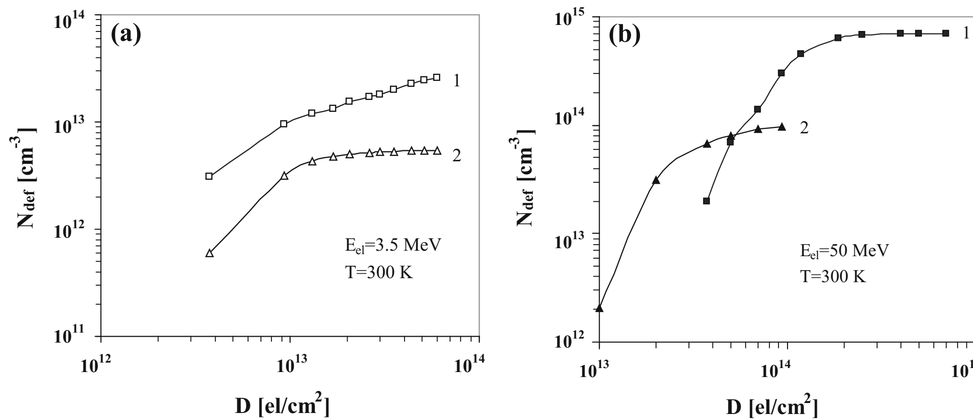

Fig. 5. Concentration of radiation defects N_{def} introduced in *n*-type silicon according to electron irradiation dose. (a) Irradiation by picosecond pulse electrons beam with an energy of 3.5 MeV; The initial specific resistances of samples were: 1—ρ = 124 Ω × cm; 2—ρ = 710 Ω × cm. (b) Irradiation by microsecond pulse electrons beam with an energy of 50 MeV; The initial specific resistances of samples were: 1—ρ = 100 Ω × cm; 2—ρ = 450 Ω × cm.

number of vacancies increases with the irradiation dose (Fermi quasi-level is located below the defect level). At a sufficiently high rate of generation of non-equilibrium carriers, which is easily attained by irradiation with high density pulses of electrons, the energy of the Fermi quasi- level will be located above the energy level of the defect.[9,17–19] Accordingly, the filling of a given level, which to a first approximation is determined by the electron–hole recombination rate, will not change as J increases. The latter explains the saturation behavior for the rate of production of complex radiation defects observed as J increases (Fig. 2).

CONCLUSION

In summary, the above results show that the application of high density pulse irradiation introduces structural defects in semiconductor crystals, which cause transition processes that are able to essentially change the electro-physical properties of materials, and result in serious damage to the efficiency of semiconductor devices fabricated from them. However, the relevant transition processes have a dual character: namely, there is a redistribution of radiation-induced structural defects within the material, but also the occurrence of relaxation of the excitation of electronic centers. These redistribution processes can influence the working power of devices over a timescale of perhaps hundreds of seconds. The rates for the introduction of radiation defects for different irradiation pulse durations were determined according to particular irradiation doses and specific resistances for the samples.

Since ultrafast irradiation is applied with a pulse duration of the order of picoseconds, while thermal effects occur on a micro-econd timescale, then the latter will not be observed in experiments that operate on the far shorter timescale. This is in sharp

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contrast to the effect of radiation as applied from conventional radiation sources (pulse duration 10^{-6}–10^{-7} s), where thermal effects occur significantly (Fig. 1).

We suggest that the uniquely high level of detail that has proved possible in determining the influence of irradiation on the crystal may be described as "fine structure of radiation defects creation in materials".

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CONFLICT OF INTEREST

The authors declare that they have no conflict of interest.

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