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Charge-Carrier Recombination in Silicon Irradiated with γ-Rays of Different Energies

By

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Comparative experiments are made to study the nonequilibrium charge-carrier recombination in silicon irradiated with γ-rays of ^{60}Co and of the bremsstrahlung spectrum of electrons with maximum energy 100 MeV. n- and p-Si crystals with $\varrho = 10$ to 1000 Ωcm grown by Czochralski and vacuum float-zone techniques are used. The temperature and injection dependences of nonequilibrium charge-carrier lifetimes measured by a phase method are analyzed. It is found that when irradiating with γ-rays of ^{60}Co a great number of recombination centers are produced which are mainly complexes of irradiation-generated vacancies and interstitials with dopants (phosphorus, boron) and technological (oxygen, carbon) impurities. As distinct from this, the dominant recombination centres in silicon irradiated with γ-rays of the bremsstrahlung spectrum are divacancies involved in defect clusters, surrounded by the potential barrier Ψ for the majority charge carriers. The parameters of recombination centres (energy level spectrum, annealing temperatures, charge-carrier capture coefficients) in crystals studied under γ-irradiation are determined. The peculiarities in the charge-carrier recombination processes due to the localization of recombination centres in defect clusters generated by photonuclear reaction products are discussed.

Выполнены сравнительные эксперименты по исследованию рекомбинации неравновесных носителей заряда в кремнии, облученном гамма-квантами Co^{60} и тормозного спектра электронов с максимальной энергией 100 MeV. Использовались кристаллы n- и p-кремния с $\varrho = 10$ до 1000 Ωcm, полученные по методу Чохральского и зонной плавкой в вакууме. Анализировались температурные и инжекционные зависимости времени жизни неравновесных носителей заряда, измеренные фазовым методом. Установлено, что при облучении гамма-квантами Co^{60} образуется большое количество рекомбинационных центров, представляющих собой, в основном, комплексы генерируемых облучением вакансий и междоузлий с атомами легирующих (фосфор, бор) и технологических (кислород, улгерод) примесей. В отличие от этого домирирующими рекомбинационными центрами в кремнии, облученном гамма-квантами тормозного спектра, являются дивакансии, входящие в состав кластеров дефектов, окруженных потенциальным баоьером Ψ для основных носителей заряда. Определены параметры рекомбинационных центров (спектр, энергетические уровни, температуры отжига, коэффициенты захвата носителей заряда) в исследуемых кристаллах при используемых энергиях гамма-квантов. Обсуждаются особенности в процессах рекомбинации носителей заряда из-за локализации рекомбинационных центров в кластерах дефектов, созданных продуктами фотоядерных реакций.

1. Introduction

Nature and properties of radiation defect in Si, and their spatial distribution are determined by inner (initial perfection of the crystal, impurity, and dislocation content) and outer factors (temperature, irradiation intensity, energy and type of bombarding particles) [1]. When irradiating by γ-rays of ^{60}Co ($E_\gamma \approx 1.3$ MeV), electrons with energy

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$E_e \lessapprox 10$ MeV, point radiation defects uniformly distributed in the bulk of the crystal are produced. As distinct from this, electrons with $E_e \gtrapprox 30$ MeV, high-energy protons ($E_p \gtrapprox 10$ MeV), and fast reactor neutrons produce radiation defects localized preferentially in radiation defect clusters [2]. Theoretical calculations show that radiation defect clusters can be formed when irradiating by γ-rays with energy in the giant dipole resonance region ($E_\gamma = 20$ to 40 MeV) [3]. The energy of photonuclear reaction products which appear in this case (photoprotons, photoneutrons, and recoil cores) has values close to the threshold energy of radiation defect clustering. In this work comparative experiments were carried out to elucidate the peculiarities of nonequilibrium charge-carrier recombination processes in silicon irradiated with γ-rays of different energies and to determine recombination centre parameters during their uniform distribution in the bulk of the crystal and localization in radiation defect clusters.

2. Experimental Procedure

n- and p-type silicon crystals with initial resistivity $\varrho = 10$ to 1000 Ωcm grown by the Czochralski (pulled) and vacuum floating-zone techniques were used. The samples $13 \times 3 \times 2$ mm³ in size were irradiated ($T_{irr} \lessapprox 30$ °C) with γ-rays of ^{60}Co (γ_c) or γ-rays of the bremsstrahlung spectrum of electrons with energy $E_e = 100$ MeV (γ_B). The dependences of nonequilibrium charge-carrier lifetime (τ) on temperature ($T = 80$ to 400 K) and the nonequilibrium charge-carrier excitation level ($\Delta n/n_0 = 10^{-5}$ to 10^0) were analyzed. The concentration of irradiation-induced radiation defects was determined from the temperature dependences of the Hall coefficient. τ was measured by a phase method [4] with exciting light of wave length $\lambda = 1.15$ μm and modulation frequency $f = 1$ to 100 kHz at different stages of irradiation or 15 min isochronous annealing of the samples studied.

Using the results obtained in [5, 6] one can show that in the general case τ measured in experiment is connected with lifetimes of nonequilibrium electrons τ_n and holes τ_p by the following relation:

$$\tau = \left(\frac{\tau_p \tau_{ps}}{1 + \omega^2 \tau_p^2} + \frac{\tau_n \tau_{ns}}{1 + \omega^2 \tau_n^2}\right)\left(\frac{\tau_{ps}}{1 + \omega^2 \tau_p^2} + \frac{\tau_{ns}}{1 + \omega^2 \tau_n^2}\right)^{-1},$$

where $\omega = 2\pi f$, and τ_{ns}, τ_{ps} are the stationary electron and hole lifetimes, respectively [4]. When trapping of charge carriers is absent, τ coincides with the nonequilibrium minority carrier lifetime and can be determined from the expression

$$\tau = \frac{\operatorname{tg} \Delta\varphi}{\omega}.$$

Here $\Delta\varphi$ is the phase difference between photoconductivity signal and exciting light.

The experimental results were treated on the basis of the Hall-Shockley-Read statistics [7, 8] with uniform distribution of recombination centres or using the model of nonequilibrium charge-carrier recombination at the centres localized in radiation defect clusters [9].

One of the important characteristics of recombination centres is the nonequilibrium charge-carrier capture coefficient. They were determined using the data on the energy spectrum of recombination centre levels and their concentration (capture of nonequilibrium minority charge carrier) and in the case of nonequilibrium majority charge carriers. The results of measurements of injection dependences of τ were used as well.

3. Experimental Results

3.1 Production efficiency and energy spectrum of recombination centre levels

The recombination centre production efficiency at different types of irradiation is determined by the coefficient K_τ of radiation variation in τ, which is proportional to

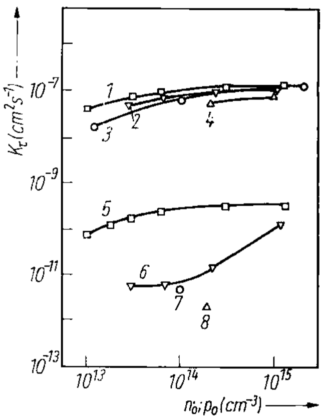

Fig. 1

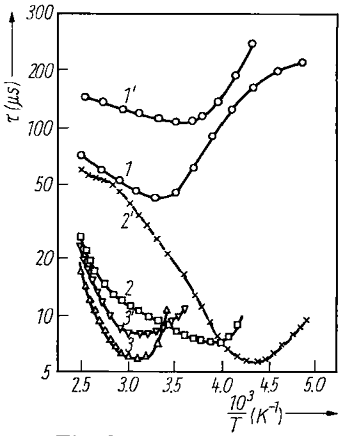

Fig. 2

Fig. 1. K_τ as a function of the majority charge carrier concentration n_0 or p_0. $T_{meas} = 330$ K. Irradiation: (1) to (4) γ_B-rays, (5) to (8) γ_c-rays. (1), (3), (5), (7) float-zone, (2), (4), (6), (8) pulled crystals. (1), (2), (5), (6) n-Si, (3), (4), (7), (8) p-Si

Fig. 2. τ as a function of the temperature in n-Si ($n_0 = 7 \times 10^{13}$ cm^{-3}). (1), (1′) before irradiation, (2), (2′) γ_c-rays, (3), (3′) γ_B-rays. (1) to (3) float-zone crystals, (1′) to (3′) pulled. Φ = (2) 3×10^{14}, (2′) 1.5×10^{15}, (3) ,(3′) 9×10^{11} cm^{-2}

their concentration. As is known [10], $K_\tau = \Delta\tau^{-1}/\Phi$ (where Φ is the integral fluence of bombarding particles), therefore, the magnitude of K_τ can be determined from the analysis of the dose dependence of τ. Fig. 1 shows K_τ as a function of doping level (majority charge-carrier concentration n_0, p_0) of n- and p-type silicon. It is seen that when irradiating with γ-rays of ^{60}Co in float-zone n- and p-Si K_τ is higher than in pulled Si, although with increasing n_0 this difference is somewhat decreased. As a rule, K_τ has lower values in p-Si. With irradiation by high-energy γ-rays K_τ depends weakly on the content of residual (oxygen, carbon) and doping (phosphorus, boron) impurities. Note, however, that under this irradiation K_τ is much higher as compared to its values when irradiating with ^{60}Co γ-rays.

The energy level spectrum of γ-ray-induced recombination centres is determined from the analysis of the temperature dependence of τ. Some of them more typical,

Table 1

Recombination centre parameters in irradiated silicon ($T_{meas} = 330$ K)

γ-rays	silicon	level (eV)	T_{ann} (°C)	C_p (cm³ s⁻¹)	C_n (cm³ s⁻¹)
^{60}Co	n-type	$E_c - 0.43$	150	1.4×10^{-7}	1.6×10^{-8}
	float-zone	$E_c - 0.40$	300 to 320	1.8×10^{-7}	8.6×10^{-9}
	n-type	$E_c - 0.17$	350	—	—
	pulled	$E_c - 0.45$	350 to 400	2.0×10^{-7}	5.0×10^{-8}
	p-type	$E_v + 0.30$	330 to 360	2.6×10^{-10}	1.3×10^{-8}
	float-zone	$E_v + 0.30$	150	5.3×10^{-9}	2.5×10^{-7}
	p-type	$E_v + 0.35$	350 to 400	2.0×10^{-10}	2.0×10^{-8}
	pulled	$E_v + 0.18$	420	—	—
bremsstrahlung	n-type	$E_c - 0.40$	100 to 300	3.3×10^{-6}	1.7×10^{-9}
spectrum	p-type	$E_v + 0.30$	100 to 300	5.3×10^{-9}	2.5×10^{-6}

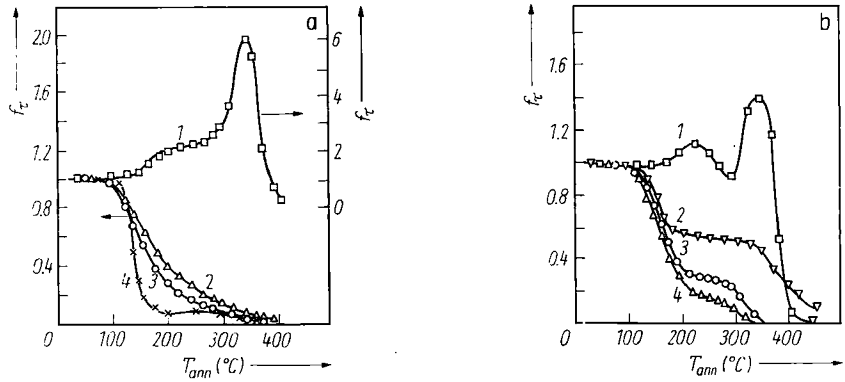

Fig. 3. f_τ as a function of the annealing temperature. a) n-Si, $n_0 = 7 \times 10^{13}\ \text{cm}^{-3}$. Irradiation: (1), (4) γ_C-rays, (2), (3) γ_B-rays. (1), (2) pulled, (3), (4) float-zone Si; b) p-Si, $p_0 = 1 \times 10^{14}\ \text{cm}^{-3}$. Irradiation: (1), (2) γ_C-rays, (3), (4) γ_B-rays. (1), (3) pulled, (2), (4) float-zone Si

are given in Fig. 2. They consist usually of two regions: high-temperature (recombination) and low-temperature which is due to the availability of trapping centres. Energy positions of all recombination centre levels obtained when treating the experimental results for n- and p-type silicon samples with different impurity contents after irradiation by γ-rays of ^{60}Co and the bremsstrahlung spectrum are given in Table 1. It should be underlined that in the case of irradiation by γ-rays of ^{60}Co the energy level spectrum of radiation centres is rich enough and depends on the impurity content of the crystals studied. At the same time under irradiation with high-energy γ-rays nonequilibrium charge carriers recombine in n-Si through level $E_c - 0.40$ eV and in p-Si through level $E_v + 0.30$ eV irrespective of the crystal growth method.

3.2 Annealing

Fig. 3 presents the dependences of the fraction of unannealed defects $f_\tau = (\tau_T^{-1} - \tau_0^{-1})/(\tau_\Phi^{-1} - \tau_0^{-1})$ (τ_0, τ_Φ, τ_T are the values of τ for nonequilibrium charge carriers before, after, and at various stages of isochronal annealing, respectively) on the annealing temperature in n- and p-type silicon. Annealing of recombination centres in ^{60}Co γ-ray irradiated crystals, especially Czochralski-grown, is seen to be rather complicated. Here it is characteristic that the type of the dependences and the temperature ranges of centre annealing are determined by the impurity content of the crystals studied. As distinct from this, recombination centres induced by high-energy γ-rays in n- and p-type Si anneal smoothly (without stages) in a sufficiently wide temperature

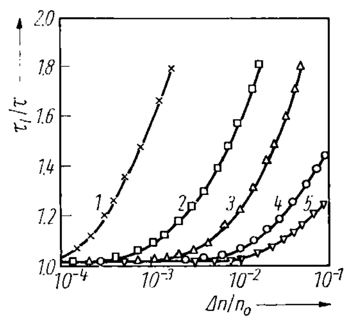

Fig. 4. τ_l/τ as a function of the non-equilibrium charge-carrier excitation level in float-zone n-Si. τ_l is τ under additional stationary illumination. Irradiation: (1) to (4) γ_B-rays, (5) γ_C-rays. $n_0 =$ (1) 3×10^{14}, (2), (5) 7×10^{13}, (3) 3×10^{13}, (4) $1 \times 10^{13}\ \text{cm}^{-3}$

range (100 to 300 °C). Annealing temperatures of all centres observed as well as the determined coefficients of nonequilibrium charge-carrier capture at them are presented in Table 1.

4. Discussion

From the results obtained it follows that in n- and p-type silicon irradiated with γ-rays of ^{60}Co some types of recombination centres with energy spectrum and annealing temperature different in float-zone and pulled material are produced. Taking into account the data available in literature [11 to 14] it can be concluded that in n-type float-zone Si crystals the main recombination centres are E-centres ($E_c - 0.43$ eV), $T_{ann} \approx$ ≈ 150 °C) and divacancies ($E_c - 0.40$ eV, $T_{ann} = 300$ to 320 °C) and in p-type Si these are interstitial carbon (C_I) and divacancies contributing close energy levels $\approx E_v +$ $+ 0.30$ eV to the forbidden gap, but being annealed in different temperature ranges ($T = 150$ °C and 330 to 360 °C, respectively). As follows from the estimates, in float-zone n-Si the contribution of divacancies to recombination processes at this irradiation is relatively small ($\approx 10\%$). In this case the E-centres dominate. In pulled n-Si the efficiency of E-centre and divacancy formation turns out to be lower as compared to A-centres and carbon-oxygen-divacancy complexes [C–O–W]. A-centres with level $E_c - 0.17$ eV are the main recombination centres in crystals with relatively low resistivity. Due to the decrease in electron occupancy of these centres, as the resistivity of crystal increases ($\varrho \gtrsim 1\ \Omega$ cm), the contribution to recombination of [C–O–W] complexes ($E_c - 0.45$ eV, $T_{ann} \approx 350$ to 400 °C) increases, which are sufficiently effective recombination centres in pulled p-Si as well (donor level $E_v + 0.35$ eV). The concentration of these complexes in n- and p-Si with increasing annealing temperature somewhat increases (Fig. 3) and this determines the sufficiently complicated character of the variation in τ when annealing in the temperature range 150 to 350 °C. An additional formation of [C–O–W] complexes at these temperatures takes place as a result of rearrangements of less stable radiation defects and interaction of free vacancies with complexes of the type CO electrically inactive under the experimental conditions, available in the initial crystals [15]. A complete annealing of irradiation- and heat-treatment-produced C–O–W complexes occurs at $T = 350$ to 400 °C in n- and p-Si. In the low temperature region and also in low resistivity pulled p-Si crystals ($\varrho \lesssim$ $\lesssim 1\ \Omega$ cm) nonequilibrium charge carriers recombine preferentially through the level $E_v + 0.18$ eV. The nature of the defect to which this level belongs is not yet established. It anneals at $T \gtrsim 420$ °C and constitutes oxygen atoms (possibly carbon atoms) and a vacancy. The latter is confirmed by the fact that the energy dependence of the production rate of these defects when irradiating by electrons with energy $E_e = 2.5$ to 100 MeV is much weaker than it is the case for the divacancy. Presumably, in pulled p-Si when annealed, a complex similar to A-centre, is formed.

The validity of the conclusions made on the nature oi the main recombination centres introduced by γ-rays of ^{60}Co in n- and p-Si is also confirmed by the data on the magnitudes of the coefficients for majority and minority charge carrier capture at centre levels which in a number of cases are close to the known literature data. It is necessary to point out the fact that the majority of recombination centres produced under this annealing are complexes involving atoms of doping and technological (C, O) impurities. In this case the contribution of intrinsic radiation defects (divacancies) to nonequilibrium charge-carrier recombination appears to be rather small. For this reason in the experiment one observes the dependence of the coefficient K_τ, type of irradiation-produced recombination centres (spectrum of energy levels induced by them), and character of their annealing on the impurity content of the crystals studied (Fig. 1 to 3). The situation is somewhat different when irradiating the identical samples by γ-rays of the bremsstrahlung spectrum. Here doping and technological impurities exert negligible influence on the production processes of the main recombination centres contributing only two energy levels (E_c—0.40 eV and E_v+0.30 eV in n- and p-Si, respectively) in the forbidden gap. The dependence of K_τ on n_0 and p_0 within the framework of the Hall-Shockley-Read model is described qualitatively by the change in the occupancy function of recombination centre levels. In float-zone and pulled Si K_τ has approximately the same values and at annealing τ restores smoothly (without stages) over a wide temperature range irrespective of the crystal conductivity type and growth method. All this taken together suggests that at such irradiation nonequilibrium charge carriers recombine preferentially through the levels of intrinsic defects. The energy spectrum of levels (E_c — 0.40 and E_v + 0.30 eV) and the temperature of their complete annealing ($\lesssim$ 350 °C) show strong evidence for the fact that γ-rays of the bremsstrahlung spectrum produce effectively divacancies. However, during recombination of nonequilibrium charge carriers through their levels in this case one observes a number of peculiarities which were absent in silicon irradiated with ^{60}Co γ-ray fluences equivalent by the number of displaced atoms. Indeed, as is seen from Table 1, the coefficients for nonequilibrium minority charge carrier (electron or hole) capture at the level of divacancies [2]) introduced by γ-rays of the bremsstrahlung spectrum, have higher (by an order) values as compared to the ^{60}Co γ-ray -irradiation. Besides, in the experiment one observes (Fig. 4) an increased sensitivity of τ to the excitation level of nonequilibrium charge carriers, with this sensitivity being increased as the doping level of crystals increases. Taking into account these results and the smooth recombination centre annealing it can be concluded that divacancies generated by γ-rays of the bremsstrahlung spectrum are involved in radiation defect clusters surrounded by the potential barrier Ψ for majority charge carriers [16]. Bending of the potential relief around clusters creates favourable conditions for the capture of nonequilibrium minority charge carriers for which clusters are the potential wells (in the simplest case of rectangular form). The concentration of nonequilibrium minority charge carriers in the clusters appears to be exp (Ψ/kT) times higher than in the "undamaged" matrix. In a real crystal the cluster potential decays smoothly in the outer space charge region the extension of which (its volume V_{scr}) is determined by the Debye screening length in the crystal matrix and in some cases (high-resistivity materials) it can exceed the cluster volume (V_c). The minority carriers which appear inside V_{scr} will be attracted by the cluster. This will result in more effective capture of minority carriers at recombination centre levels of radiation defect clusters. One can assume that for this reason (nonrectangularity of the potential relief) the bulk recombination rate increases $K_s = V_{scr}/V_c$ times ($K_s = V_{scr}/V_c = \sum_k N_k f_{nk}/n_0$ where N_k is the concentration, f_{nk} the occupancy function of recombination centre levels in clusters [9]). Description of nonequilibrium charge carrier recombination processes at radiation defect clusters within the framework of the Hall-Shockley-Read model [7, 8], which does not take into account these factors, results in anomalously high values of coefficients for nonequilibrium charge carrier capture at recombination centres in clusters. It should be noted that irrespective of the spatial distribution of recombination centres their individual properties should be preserved. However, due to the spatial charge available around radiation defect clusters, conditions of nonequilibrium charge carrier recombination at centres localized within them are changed. Therefore, while treating the experimental results the so-called "effective" coefficients (C_p^*, C_n^*) of charge carrier capture by recombination centres are determined. The analysis shows [9] that the coefficients for nonequilibrium minority charge carrier capture at recombination

[2]) C_p^* and C_n^* coefficients are given for Si with $\varrho \approx 10\ \Omega$cm.

centre levels uniformly distributed in the bulk of the crystal and localized in radiation defect clusters can increase $K_s \exp(\Psi/kT)$ times and those for majority ones $K_s \exp(-\Psi/kT)$ times. Therefore, the relation of effective coefficients (cross-sections) for minority and majority charge carrier capture at the levels of divacancies localized in the radiation defect clusters is $\exp(2\Psi/kT)$ times higher as compared to their uniform distribution in the crystal. This explains the experimentally observed increased sensitivity of τ to the nonequilibrium charge-carrier injection level, as this sensitivity is proportional to the ratio C_p^*/C_n^* or C_n^*/C_p^* for n- and p-type silicon, respectively. With the increase in the doping level of a crystal the Fermi level (E_F) in the matrix is shifted to the boundaries of allowed bands and in the radiation defect clusters it retains a position near the divacancy level. As a result, the magnitude of the potential barrier of the radiation defect cluster (the difference between the position of E_F in the matrix of the crystal and in the radiation defect cluster) increases and, thereby, the sensitivity of τ to the nonequilibrium charge-carrier excitation level with increasing $n_0(p_0)$.

5. Conclusions

The results obtained show that photonuclear reaction products (photoprotons, photoneutrons, and recoil cores) generated by irradiation of Si with γ-rays of the bremsstrahlung spectrum are capable of creating radiation defect clusters. Indeed, with γ-ray energy $E_\gamma = 22$ to 26 MeV reaction cross-sections of Si(γ, p) and Si(γ, n) have maximum values 6×10^{-26} and 1×10^{-26} cm^2 [17]. The photoproton energy reaches thereby values $E_p \gtrsim 9$ MeV and the spectral distribution of photoneutrons has a maximum $E_n \approx 2$ MeV [18, 19]. The estimates show that the recoil cores in virtue of the pulse conservation law acquire energies 0.1 to 0.4 MeV high enough for radiation defect clustering in silicon.

Radiation defect clusters produced under this irradiation involve recombination centres determining the nonequilibrium charge carrier lifetime in silicon. Most of them are divacancies forming the radiation defect cluster core [20]. Due to the localization of divacancies in radiation defect clusters surrounded by the potential barrier Ψ for majority charge carriers, a number of peculiarities in nonequilibrium charge carrier recombination processes is observed in the experiment: anomalously high values of "effective" coefficients of charge carrier capture, increased sensitivity of τ to their excitation level, specific character of recombination centre annealing. The contribution of defects available in the peripheral region of radiation defect clusters to nonequilibrium charge carrier recombination process proves to be negligible. This is due to the fact that under the experimental conditions (Fermi level position in the initial material, measurement temperature, charge carrier injection level) defects forming the peripheral cluster region (A-, E-centres, [C–O–W] complexes, and others) are outside the bending of the potential relief of the cluster, although their production rate is comparable and the production rate of some defects is even higher than that of divacancies. When changing the experimental conditions the contributions of these defects as well as of isolated radiation defects in the "undamaged" crystal matrix can considerably increase.

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