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Modeling of columnar recombination for high-energy photon generated electrons and holes in amorphous selenium

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Abstract An analytical model is developed to study the mechanisms of X-ray generated free Electron–hole pair (EHP) creation energy in amorphous selenium (a-Se) at high electric fields. The model is presented to show the electric field and temperature dependence of the charge extraction yield limited by the columnar recombination for the materials that have widely unequal drift mobility for electrons and holes, such as a-Se. The model is compared with Jaffe's columnar recombination model with widely varying field and temperature. In addition, the free EHP creation energy is calculated by incorporating the initial charge extraction yield and the charge collection efficacy of the free carriers. Also, the results of this model are compared with the recently published experimental results on EHP creation energy. The analysis of the results confirm that the proposed model gives the best possible explanation to the columnar recombination mechanisms in a-Se and the free EHP creation mechanisms at diagnostic X-ray exposures can be described by the columnar recombination.

1 Introduction

Amorphous selenium (a-Se) is widely used in direct conversion X-ray detectors and a highly promising photoconductor for optical light detection in very high-gain indirect conversion avalanche X-ray detectors [1, 2]. However, charge carrier transport and electron–hole pair creation mechanisms at high energy photons (e.g. X-ray energies) in a-Se are not yet clearly understood. The electron–hole pair (EHP) creation energy W_{ehp} in a-Se has a strong dependence on electric field F and a relatively weak dependence on the X-ray photon energy E and temperature T [3, 4]. The incident X-ray photon first creates an energetic primary electron and it generates many EHPs along its path but only a certain fraction of these are free to drift and the rest recombine. There are various possible explanations for the F dependence of the EHP creation energy. First, the simultaneously generated electron and its hole twin are attracted to each other by their mutual Coulombic force and may eventually recombine. This type of recombination is called *geminate recombination* (Gemini—the twins) [5, 6]. Another possible mechanism is *columnar recombination* that involves the recombination of nongeminate electrons and holes generated close to each other in the columnar track of the single high energy electron (*primary*) created by the absorption of an X-ray photon. This *columnar recombination* assumes that ionization along the path of the primary ionizing particle is dense enough that the geminate recombination is negligible and the mean separation between electron–hole pairs is much less than the column diameter.

In both geminate and columnar cases, the number of carriers escaping recombination should increase with increasing F that acts to separate the oppositely charged carriers. However, the X-ray photogeneration efficiency increases significantly with increasing the X-ray photon energy [7] and the *geminate recombination* model fails to describe this phenomenon [8]. This appears to be due to a reduction of recombination with increasing X-ray energy. The rate of deposition of energy per unit distance travelled by a primary electron decreases as a function of energy,

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decreasing the density of EHPs in the column around it [9]. This is expected to reduce columnar recombination—as is seen. Thus it appears that, at low energies, the contribution from columnar recombination is much higher than that from geminate and thus the *columnar recombination* theory is more appropriate for the diagnostic X-ray energies (12–120 keV).

The *columnar recombination* has been explained so far by the formulation of Jaffe's model [10]. He considered the carrier continuity equations of two charged species considering carrier drift, diffusion and bimolecular recombination between nongeminate electrons and holes. He solved the equations following an order; i.e., first neglected the drift and recombination terms, got the solution for the diffusion term only and then reintroduced the drift term and got the solution. He then reintroduced the recombination term. This procedure essentially emphasizes the diffusion term and underestimates the recombination. The diffusion term actually has less effect than the drift and recombination terms at moderate to high electric fields. For this reason, Kramers [11] reversed the Jaffe's procedure by neglecting the diffusion term and obtained an analytical solution for the remaining equations assuming the same mobility for both the carriers (equal mobility highly simplifies the formulation!). The effective mobility of electrons and holes are far different in a-Se. Therefore, the Kramers's formulation is not appropriate for a-Se.

In this paper, we have developed a model for the columnar recombination under an applied field, which are appropriate for the materials having widely unequal drift mobility for electrons and holes. We compare our model with Jaffe's model with widely varying field and temperature. The EHP creation energy is calculated by incorporating the initial charge extraction yield (Y) and the charge collection efficacy of the free carriers (see Sect. 2.3). We also compare our model calculations with the recently published experimental results [8] and present a critical discussion on the appropriateness of various models.

2 Theoretical model

2.1 Jaffe's model

Jaffe assumed that the charge generated by high energy particles is initially (at time $t = 0$) in a dense column with a Gaussian distribution of initial charge carriers from the center of the cylindrical track as shown in Fig. 1 where,

$$n_0 \text{ or } p_0 = \left(\frac{N_0}{\pi b^2}\right) e^{-\left(x^2+y^2\right)/b^2}. \quad (1)$$

Here N_0 is the ionization line density (charges/cm), b is the radius of the Gaussian distribution, and n and p are the electron and hole concentrations, respectively. The subscript 0 on n or p denotes the initial carrier concentrations. The mechanism of recombination (or dissociation) of photogenerated charge carriers in amorphous selenium (a-Se) have been previously described by solving the carrier continuity equations of two charged species considering carrier drift, diffusion and biomolecular recombination between nongeminate electrons and holes defined by the following differential equations for the time variation of the carrier concentrations [10],

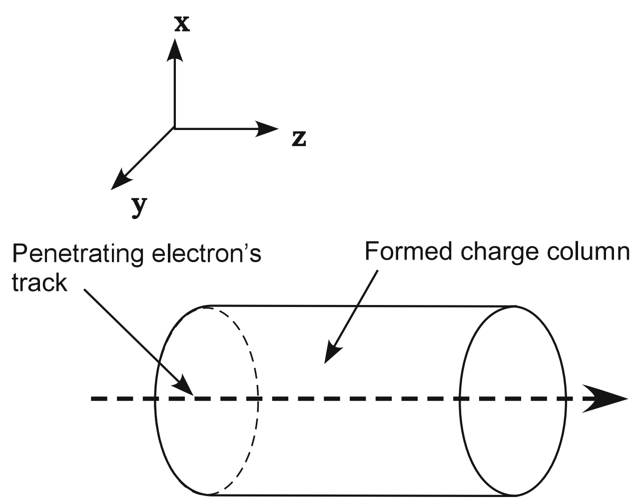

Fig. 1 The schematic illustrating the cylindrical column formation for the columnar recombination

$$\frac{\partial n}{\partial t} = \mu_e F \frac{\partial n}{\partial z} + D_e \frac{\partial^2 n}{\partial z^2} - C_r np, \quad (2)$$

$$\frac{\partial p}{\partial t} = -\mu_h F \frac{\partial n}{\partial z} + D_h \frac{\partial^2 p}{\partial z^2} - C_r np, \quad (3)$$

where C_r is the recombination coefficient, and D and μ are the diffusion coefficient and mobility of the charge carriers. The subscript e and h stand for electrons and holes, respectively. It is assumed in Eqs. (2) and (3) that the electric field is in the z-direction. The recombination rate in a-Se at low or nearby conventional operating field ($\sim$10 V/µm) is controlled by the diffusion of recombining carriers as described by the Langevin theory [12]. Thus, in a-Se, $C_r = C_L = e\,(\mu_h + \mu_e)/\varepsilon$, where C_L is the Langevin recombination coefficient, e is the elementary charge and ε (=$\varepsilon_o\varepsilon_r$, where ε_o is the absolute and ε_r is the relative permittivity) is the permittivity of the photoconductor. The hole mobility increases by almost an order of magnitude by increasing the electric field from 10 to 80 V/µm, and saturates at the level $\mu = 0.9$ cm^2/Vs [13]. One can expect the increase of mobility would lead to the gradual deviation of the Langevin recombination mechanism and the recombination coefficient C_r at high fields attains a constant value

C_0. Thus, Bubon et al. proposed an empirical expression for C_r, which is [8],

$$\frac{1}{C_r}=\frac{1}{C_L}+\frac{1}{C_0}. \tag{4}$$

With decreasing electric field, the carrier mobility decreases and the recombination coefficient gradually leads to the Langevin's value C_L.

As mentioned previously, Jaffe could not solve Eqs. (2) and (3) simultaneously, so he first neglected the recombination and drift terms. He then reintroduced the drift and recombination terms. Assuming that the initially charges were created in column length d and the electric field is parallel to the column axes, Jaffe derived the following analytical expression for charge extraction yield (i.e., the fraction of charges that escape columnar recombination),

$$Y_{//}(F)=\frac{\mu Fb^2}{2Dd}c_1e^{-c_1}[l_i(e^{c_2})-l_i(e^{c_1})], \tag{5}$$

where

$$c_1=\frac{8\pi D}{C_rN_o} \quad \text{and} \quad c_2=c_1+\ln\left(1+\frac{2Dd}{\mu Fb^2}\right) \tag{6}$$

Here $l_i(x)$ is the logarithmic integral function, $D=D_e+D_h$ and $\mu=\mu_e+\mu_h$.

If the electric field is at an angle θ with the column axis, the fraction of electrons that escape recombination is

$$Y_\theta(F)=\frac{1}{1+\frac{C_rN_o}{8\pi D}\sqrt{\frac{\pi}{\zeta}}S(\zeta)}, \tag{7}$$

where

$$S(\zeta)=\frac{1}{\sqrt{\pi}}\int_0^\infty\frac{e^{-s}ds}{\sqrt{s\left(1+\frac{s}{\zeta}\right)}}\quad,\ \text{and}\quad \zeta=\frac{b^2\mu^2(F\cos\theta)^2}{2D^2} \tag{8}$$

2.2 Proposed model

Jaffe's full treatment of this problem is very elegant despite the approximations he made as mentioned in the introduction. Kramers showed that the diffusion term is negligible compared to the drift term in Eqs. (2) and (3) if $F \gg kT/eb$, where k is the Boltzmann constant. That means, taking the smallest possible value for b to be of the order of 10 nm, F has to be much greater than 2.5 V/µm for neglecting the diffusion term in Eqs. (2) and (3). In fact, the operating electric field in a-Se detectors is 10 V/µm or higher. Thus, Kramers reversed the Jaffe's procedure by neglecting the diffusion term for moderate to high electric fields, and obtained an analytical solution for the remaining equations. However, he assumed the same mobility for both the carriers (equal mobility highly simplifies the formulation!). The effective mobility of electrons and holes are far different in a-Se and thus the original Kramers's formulation is not appropriate for a-Se. In this section, we propose a columnar recombination model following Kramers assumptions, which are appropriate for the materials having widely unequal drift mobility for electrons and holes.

2.2.1 *Electric field parallel to the column axis*

Amorphous Selenium is one of the semiconductors that have been characterized by the very different values of the electron and hole drift mobilities (the hole mobility is almost 40 times higher than the electron mobility) [14]. Therefore, the transport of electrons could be neglected within the time domain of the hole transport [15] and the remaining electrons after the hole transport are the escaped electrons from the columnar recombination between nongeminate electrons and holes. Thus, the transport equations for the electric field parallel to the column axis can be simplified as

$$\frac{\partial n}{\partial t}=-C_rnp \tag{9}$$

$$\frac{\partial p}{\partial t}=-\mu_hF\frac{\partial n}{\partial z}-C_rnp \tag{10}$$

The expression of $p(z,t)$ from (9) can be written as [16]

$$p(z,t)=-\frac{1}{C_r}\frac{d}{dt}\left\{\ln\frac{n(z,t)}{n_0(z)}\right\} \tag{11}$$

Substituting $p(z,t)$ in Eq. (10) gives

$$\frac{du(z,t)}{dt}+\mu_hF\frac{du(z,t)}{dz}=-C_rn_0(z)u^2(z,t), \tag{12}$$

where $u(z,t)=\frac{n(z,t)}{n_0(z)}$.

The free electron concentration,

$$n_{st}(z)=n_0\left[1+\frac{C_r}{\mu_hF}\int_0^z n_0(z')dz'\right]^{-1}. \tag{13}$$

The total number of electrons that are escaped the columnar recombination can be expressed as

$$\begin{aligned}Q&=\int_0^\infty 2\pi r\int_0^d n_{st}(z)dzdr\\&=\frac{2\pi\mu_hF}{C_r}\int_0^\infty\ln\left(1+\frac{C_rdn_0}{\mu_hF}\right)rdr\end{aligned} \tag{14}$$

Hence, the charge extraction yield can be written as

$$Y_{//}=\frac{Q}{N_0d} \tag{15}$$

2.2.2 Electric field perpendicular to column axis

If the electric field is perpendicular to the Column axis (say along x-direction), the survived electron concentration,

$$n_{st}(x,y) = n_0\left[1 + \frac{C_r}{\mu_h F}\int_{-\infty}^{x} n_0(x',y)dx'\right]^{-1} \quad (16)$$

The total number of electrons per unit distance that are escaped the columnar recombination can be expressed as,

$$N = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} n_{st}(x,y)dxdy = \frac{2\mu_h F}{C_r}\int_0^{\infty} \ln\left(1 + \frac{C_r N_0}{\sqrt{\pi}\mu_h F b}e^{-y^2/b^2}\right)dy \quad (17)$$

Thus, the charge extraction yield can be written as

$$Y_{\perp} = \frac{2\mu_h F}{N_0 C_r}\int_0^{\infty} \ln\left(1 + \frac{C_r N_0}{\sqrt{\pi}\mu_h F b}e^{-y^2/b^2}\right)dy \quad (18)$$

If the electric field is at an angle θ with the column axis, the fraction of electrons that escape recombination can be written as,

$$Y_{\theta} = \frac{2\mu_h F}{N_0 C_r}\int_0^{\infty} \ln\left(1 + \frac{C_r N_0}{\sqrt{\pi} b \mu_h F \sin\theta}e^{-y^2/b^2}\right)dy \quad (19)$$

2.3 EHP creation energy

The average energy needed to create a single free EHP is called the EHP creation energy W_{ehp}. The W_{ehp} is usually calculated by W_0/Y, where W_0 is the average X-ray energy needed to create an EHP and Y is the charge extraction yield. It is assumed above that the free charge carriers are not lost during their transport across the photoconduction. This assumption is true at higher temperatures (e.g., at room temperature and above) and at higher fields (e.g., at above 10 V/µm), when the charge collection efficiency for the free carriers is close to unity. However, at low temperatures and/or at lower electric fields the electron mobility becomes very low and thus the charge collection efficiency even in a thin detector deviates considerably from unity. Therefore, in general, one can calculate W_{ehp} using the following expression,

$$W_{ehp} = \frac{W_0}{\eta_{cc} Y}, \quad (20)$$

where η_{cc} is the charge collection efficiency for the free carriers, which is given by [17],

$$\eta_{cc} = \frac{\mu_h F \tau_h}{L}\left[1 + \frac{1}{\eta(1/\alpha\mu_h\tau_h F - 1)}\left(e^{-L/\mu_h\tau_h F} - e^{-\alpha L}\right)\right] + \frac{\mu_e F \tau_e}{L}\left[1 - \frac{1}{\eta(1/\alpha\mu_e\tau_e F + 1)}\left(1 - e^{-\alpha L - L/\mu_e\tau_e F}\right)\right]. \quad (21)$$

Here $\eta = 1 - \exp(-\alpha L)$ is the quantum efficiency of the detector, τ is the lifetime of the free carriers, L is the a-Se layer thickness, and α is the linear attenuation coefficient of a-Se.

3 Results and discussion

In this section, we present the results of our analytical model to show the field and temperature dependence of the quantum yield limited by columnar recombination and compare them with the published models and the experimental data in order to validate the mechanisms of X-ray generated free EHP creation energy in a-Se at high electric field. Figure 2 shows the electron–hole pair creation energy (W_{ehp}) as a function of the electric field at room temperature ($T = 293$ K). The open circles represent the measured electron–hole pair creation energy (W_{ehp}) for 59.5 keV Am^{241} gamma rays, which were extracted from the recently published paper [8]. The dashed line represents the model calculation using Jaffe's model (Eq. (5)) with electric field parallel to the column axis for $N_0 = 5 \times 10^7$ cm^{-1}, $b = 2 \times 10^{-5}$ cm and $d = 15 \times 10^{-4}$ cm. The dash-dotted line represents the model calculation using Eq. (7) (i.e., Jaffe's model with electric field at an angle of 30° to the column axis) for $N_0 = 5 \times 10^7$ cm^{-1}, and $b = 5 \times 10^{-6}$ cm. The dotted and solid lines represent the proposed model calculations of Eqs. (15) and (19), respectively. The fitted parameters for both the dotted and

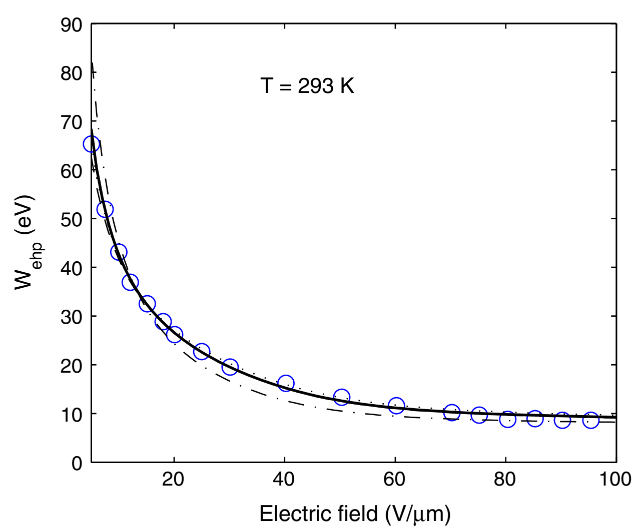

Fig. 2 The free EHP creation energy (W_{ehp}) as a function of electric field. *Symbols*: experimental results [8], *dashed line*: Jaffe's model with electric field parallel to the column axis, *dash-dotted line*: Jaffe's model with electric field at an angle of 30° to the column axis, *dotted line*: proposed model with electric field parallel to the column axis and *solid line*: proposed model with electric field at an angle of 30° to the column axis

solid lines are $N_0 = 3 \times 10^7$ cm^{-1}, $b = 1.55 \times 10^{-6}$ cm and $d = 10^{-5}$ cm. The parameter W_0 is taken as 7 eV in all calculations.

The incident X-ray photons create energetic electrons and these electrons interact in discrete collisions resulting in distinct energy deposition events known as *spurs*. In the simplest version of the model, spurs are considered to be uniformly spaced and spherically shaped regions that contains few EHPs. At diagnostic X-ray energies, the linear energy transfer of the primary electrons is high enough that the spurs are overlapped and create a column along the track of the primary electron. Mah et al. [7] estimated the average spur radius to be ~10 nm and thus the value of b should be in the same order of the spur radius and the value of d should be several times higher than b.

As shown in Fig. 2, the solid line (i.e., the proposed model with electric field at an angle of 30° to the column axis) gives the best fit to the experimental data. For $N_0 = 3 \times 10^7$ cm^{-1}, the mean separation of EHPs is ~0.3 nm (the interatomic separation in a-Se is 0.23 nm), which is reasonable. Again the fitted value of b is 15.5 nm, which is close to the average spur radius (10 nm) as mentioned by Mah et al. [7]. Note that Jaffe's model with electric field parallel to the column axis (dashed line in Fig. 2) gives the next closer fit to the experimental results. However, the fitted values of b (200 nm) and d (15 μm) are too high and thus far from the reality because of unrealistic assumptions in Jaffe's formulation. Again the proposed model with electric field parallel to the column axis (dotted line in Fig. 2) also gives a reasonable fit with the experimental results. The fitted value of d is 0.1 μm (this value is ~6 times of b, which is quite reasonable) and other fitted parameters are the same as in solid line curve in Fig. 2. Therefore, the proposed model (Eqs. 14, 15, 18, 19) gives the best possible explanation to the columnar recombination mechanisms in a-Se.

The temperature dependencies of W_{ehp} at various applied fields are shown in Fig. 3. The symbols (open circles), dashed lines and solid lines represent the experimental data, the model calculation without η_{cc} and the model fit including a correction for η_{cc}, respectively. The experimental data are extracted from Ref. [8]. The model calculation considering the charge collection efficiency (solid lines in Fig. 3) shows a very good fit to the experimental results. The fitted values of carrier lifetimes in calculating the charge collection efficiency (Eq. 21) are $\tau_e = 52$ μs and $\tau_h = 10$ μs, which are very reasonable for a-Se [14]. The temperature dependencies of drift mobility at various applied fields are adapted from Ref. [18] and [19]. All other fitting parameters in Fig. 3 are the same as in Fig. 2. The corresponding charge collection efficiency versus temperature at various applied fields is shown in Fig. 4. As evident from Fig. 4, the η_{cc} at 10 V/μm field

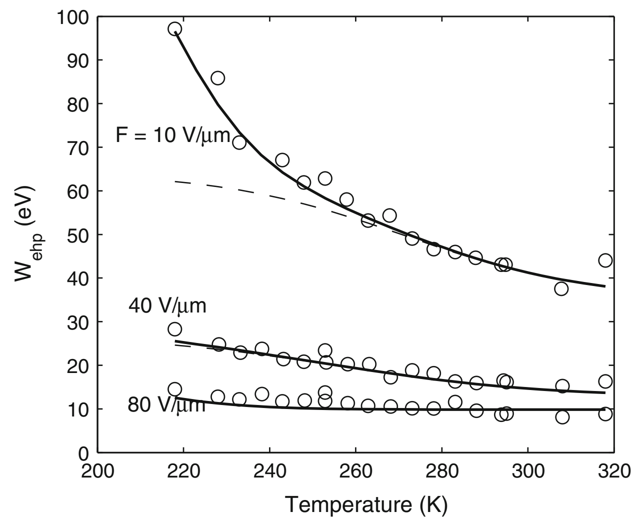

Fig. 3 W_{ehp} versus temperature at various electric fields. *Symbols*: experimental results [8], *dashed line*: W_{ehp} calculation without considering free charge collection efficacy and *solid line*: the model fit including a correction for free charge collection efficiency (η_{cc})

decreases abruptly by lowering the temperature below 260 K because of very low mobility of electrons at low temperatures [18]. At 59.5 keV γ-ray excitation on a 15 μm thin detector, the normalized absorption depth (the absorption depth/thickness) becomes 64 and thus the charge collection of holes and electrons are almost equally important [20]. The electron collection at low temperatures is severely affected by its low mobility, which reduces the overall charge collection efficiency. However, the electron mobility increases abruptly with increasing the applied field beyond 20 V/μm and the charge collection efficacy has a less significant effect on W_{ehp} at higher fields.

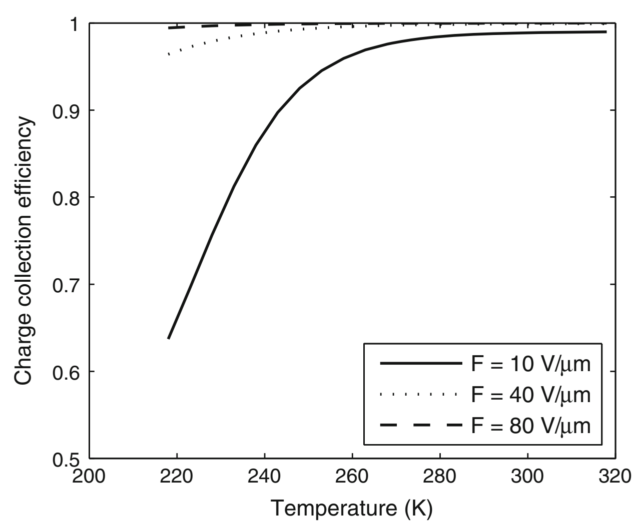

Fig. 4 Charge collection efficiency of free carriers versus temperature at various electric fields

4 Conclusions

The theoretical model for describing the columnar recombination at moderate to high electric fields in the materials that have widely unequal drift mobility for electrons and holes has been described. The EHP creation energy has been calculated by incorporating the initial charge extraction yield and the charge collection efficacy of the free carriers. The results of the model have been compared with the recently published experimental results on temperature and field dependent EHP creation energy. The proposed model with electric field at an angle of 30° to the column axis gives the best fit to the experimental data with reasonable fitting parameters. Although Jaffe's model with electric field parallel to the column axis gives the second best fit to the experimental results, the fitted values of b and d are too high and unreasonable because of unrealistic assumptions in Jaffe's formulation (i.e., emphasizes diffusion rather than drift even at high fields). The charge collection efficacy for free carriers has a significant effect on determining the EHP creation energy when the carrier mobility is too low (e.g. at low temperature and/or at low field in a-Se). The results of this paper, combined with data in [8], have shown that the proposed model gives a possible alternative explanation to the columnar recombination mechanisms in a-Se. The free EHP creation mechanisms at high-energy photon excitation in a-Se can be described by the columnar recombination.

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