Spin-dependent change in the rf photoconductivity of silicon crystals containing dislocations

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The radio-frequency photoconductivity of plastically deformed p-type silicon crystals is studied for $T = 1.4$–60 K. The results are attributed to conduction by carriers trapped in shallow dislocation states, which are probably produced by the deformation potential. The rf photoconductivity is observed to vary in a magnetic field in resonance with the dislocation dangling bonds. The results agree closely with the previously proposed model of spin-dependent recombination (SDR) via dislocation dangling bonds, which assumes that the carriers are first trapped in a shallow state.

INTRODUCTION

Much recent work has been done on spin-dependent recombination (SDR) of current carriers (cf. e.g. Refs. 1–4). This work is concerned with the change in the lifetime of the photoexcited electrons and holes which is observed when paramagnetic centers acting as recombination centers are in resonance with a magnetic field. Spin-dependent recombination was first observed in Ref. 1 for surface centers in silicon and was subsequently noted in plastically deformed silicon, where it was associated with carrier recombination via dislocation dangling bonds (DDB).[2] Spin-dependent recombination in plastically deformed silicon was also discovered independently in Ref. 3.

A specific model capable of describing the experimentally observed behavior was proposed in Ref. 4 on the basis of detailed studies of SDR at dislocations in silicon. This model generalizes the model in Ref. 5 to the case of DDB chains and utilizes the known energy spectrum of the dislocations. Essentially, the model in Ref. 4 is as follows. The recombination of the excess carriers via the DDB's involves preliminary trapping of the carriers in shallow intermediate states. The free carriers are first trapped in shallow states and then may either be trapped by a DDB chain or activated thermally back into the conduction band. According to Ref. 6, DDB's in silicon can trap holes and electrons into the bands $E_1 \approx E_v + 0.38$ eV and $E_2 \approx E_v + 0.65$ eV, respectively. The rf conductivity of crystals containing dislocations was measured in Ref. 7, where it was suggested that the electrons and holes trapped in the E_2 and E_1 levels, respectively, may move along the dislocations. This suggests that when an electron and ion are trapped by the same DDB chain, they can recombine quite rapidly. For definiteness, we will consider the spin-dependent recombination processes for electrons.

The probability for an electron to be trapped from a shallow state (which we call an E_{ed} state) into the E_2 state by a DDB chain depends on the projection of the polarization of the finite DDB chain near the electron on the electron spin vector. This is a consequence of the following two assumptions:

1. The $E_{ed} \to E_2$ transition is spin-preserving. Of course, the wave function of the electron in the E_{ed} state must be nonvanishing at the DDB in order for this to be true.

2. The electron in E_2 forms a single state with the DDB. This results in an accumulation of pairs (an electron in E_{ed} plus a DDB chain) which contain a large admixture of triplet configurations. These triplet and singlet states are mixed under conditions of magnetic resonance, with the result that there is an increase in the average probability W_c for electron capture $E_{ef} \to E_2$ followed by recombination. It is important to note that in this model, the SDR is independent of the static magnetic field to first order, in agreement with the experimental data.

Two requirements are imposed on the states E_{ed}. First, they must be localized in the immediate vicinity of the center of the dislocation. Second, they should be rather shallow and should have a large trapping cross section. According to the estimates in Ref. 4, ~ 10 meV is required to activate a carrier from E_{ef} into the conduction band, which is the same as the activation energy for trapping into the E_2 state. If we assume that the E_{ed} states are produced by the dislocation deformation potential, they should form a quasi-one-dimensional energy band. The carriers trapped in these bands can then move along the dislocations. If the model proposed in Ref. 4 is to be successful, clearly the DDB chains must be quite short and the electrons trapped in the E_{ed} states should not jump from one chain to another as they move along a chain. Such confinement might be a consequence of other types of defects which are known to be present at the dislocations, and/or to the existence of reconstructed regions which do not contain paramagnetic DDB's (Ref. 8). The average number of spins $\bar{N}$ in a chain was estimated to be 14–40 in Ref. 4; this corresponds to a chain length of 50–200 Å.

Thus if the shallow E_{ed} states in the model in Ref. 4 do in fact exist and are associated with the deformation potential, we anticipate that the carrier trapped in the E_{ed} states will dissipate large amounts of microwave power because of their rf conductivity; moreover, these losses will be spin-dependent. The purpose of our present work is to verify this experimentally.

MEASUREMENT TECHNIQUE

We assume that at sufficiently low temperatures crystals with dislocations consist of a nonconducting matrix which contains fragments of dislocations. We assume that

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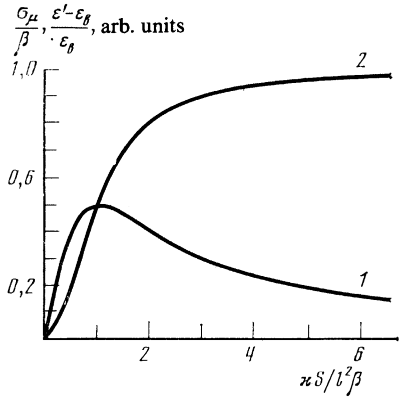

FIG. 1. Theoretical curves for the imaginary σ_μ/β (1) and real parts $(\varepsilon' - \varepsilon_b)/\varepsilon_b$ (2) of the dielectric permittivity of a sample calculated from (1), (2).

the matrix is an insulator with dielectric permittivity $\varepsilon = \varepsilon_b$ and contains randomly distributed, highly elongated conducting ellipsoids of length l, radius r_0, and specific conductivity σ. Using the formulas in Ref. 9, we have the expressions

$$\sigma_\mu \approx \nu l S \frac{\beta^2}{\beta^2 + (\varkappa S/l^2)^2}, \tag{1}$$

$$\varepsilon' \approx \varepsilon_b \left[1 + \frac{\nu l^3}{\varkappa} \frac{(\varkappa S/l^2)^2}{\beta^2 + (\varkappa S/l^2)^2} \right], \tag{2}$$

for the rf conductivity σ_μ and dielectric permittivity ε'; here $S = r_0^2\sigma$ is the "linear" conductivity of the ellipsoids, i.e., the conductivity per unit length of the ellipsoid; $\varkappa = 6[\ln(l/r_0) - 1]/\pi$; ν is the volume density of the "ellipsoids" in cm^{-3} (i.e., the density of the conducting dislocation fragments); $\beta = \varepsilon_0\varepsilon_b\omega$. We note that $\nu \bar{l}$ is $\sim N_D$, the density of the dislocations.

Figure 1 plots the ε' and σ_μ as functions of $\varkappa S/l^2\beta$ calculated from Eqs. (1) and (2). We see that $\sigma_\mu \sim \sigma_\omega = \nu l S$ only if $\beta \gg \varkappa S/l^2$. Therefore, both σ_μ and ε' must be measured to permit a more clear-cut interpretation of the experiments.

In our work we studied p-type silicon crystals doped with boron to $10^{13}/cm^3$; the crystals were plastically deformed by compressing them along the [110] axis at 680 °C. The samples were similar to the ones in Ref. 4 ($N_D \approx 10^{19}$ cm^{-3}), where spin-dependent recombination was studied. They were placed inside a cylindrical resonator (TM_{010} mode), which was connected to a superheterodyne EPR spectrometer operating in the 3-cm range ($f_0 \approx 9300$ MHz).

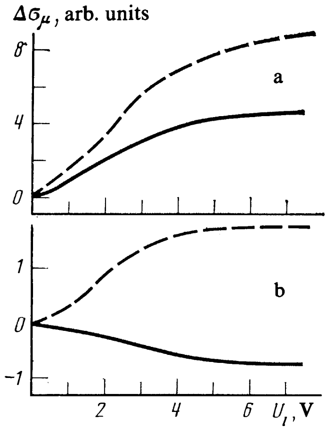

FIG. 3. Dependence of the rf losses $\Delta\sigma_\mu$ on the illumination (U_l is the incandescent lamp voltage) for several temperatures and rf electric fields: a) $T = 4.2$ K; b) $T = 1.4$ K; the solid and dashed curves are for $E_{mw} \approx 20$ V/cm and $E_{mw} \approx 500$ V/cm, respectively.

The resonator was carefully shielded from external illumination (including radiation from the warm portions of the waveguide and cryostat). The sample was illuminated by a miniature incandescent lamp or GaAs light-emitting diode (LED). We measured $\Delta\varepsilon'$ and $\Delta\varepsilon''$ for the sample at the operating frequency f_0, where $\Delta\varepsilon'$ and $\Delta\varepsilon''$ are the changes in the real and imaginary parts of the dielectric permittivity, respectively. The familiar formula $\varepsilon'' = -\sigma_\mu/\omega\varepsilon_0$ holds, where $\omega = 2\pi f_0$ and σ_μ is the effective conductivity of the sample; $\Delta\varepsilon'$ was measured from the shift in the resonance frequency, and $\Delta\varepsilon''$ was found from the change in the Q-factor of the resonator. In the first case we stabilized the operating frequency relative to a reference resonator; in the second case, f_0 was stabilized relative to the resonator containing the sample. We measured the change in Q by recording the change in the microwave power reflected or transmitted by the resonator.

EXPERIMENTAL RESULTS

We first discuss how $\Delta\varepsilon'$ and $\Delta\sigma_\mu$ depend on the intensity of the light illuminating the sample (i.e., the current I through the LED). Figure 2a, b shows the data for one of the samples recorded at 1.4 K for microwave powers less than 0.1 mW ($E_{mW} < 20$ V/cm). Figure 2c plots the dependence of the dc conductivity σ_0 for the same sample; σ_0 is zero if the sample is unilluminated and is less than $1.5 \cdot 10^{-8}$ $(\Omega\cdot cm)^{-1}$ even when $I = 50$ mA (the microwave method was not sensitive enough to measure conductivities this small). Figure 3a,

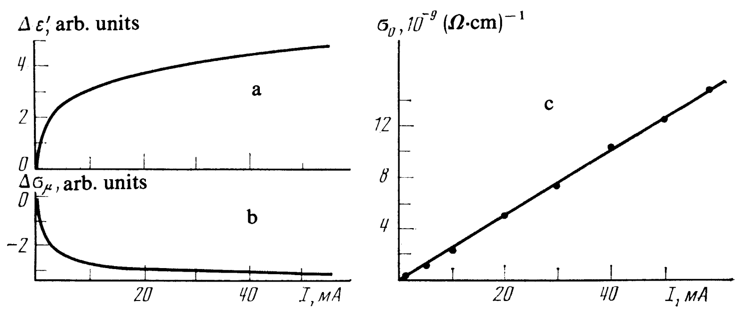

FIG. 2. Change ε' (a) and σ_μ (b) ($T = 1.4$ K, $E_{mw} \lesssim 20$ V/cm) and the dc conductivity σ_0 (c), as functions of the illumination intensity (I is the current through the GaAs LED).

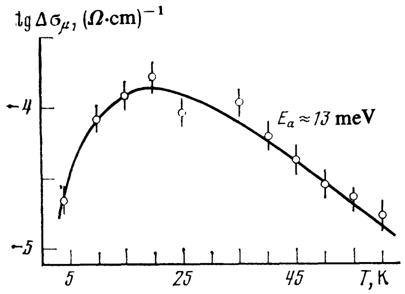

FIG. 4. Temperature dependence of $\Delta\sigma_\mu$ recorded for $E_{mw} \lesssim 20$ V/cm for an GaAs LED current $I = 50$ mA.

b shows the dependence $\sigma_0(I)$ for two microwave powers corresponding to $E_{mw} \approx 20$ and 500 V/cm for $T = 4.2$ and 1.4 K. Two facts are noteworthy: 1) the microwave power influences σ_μ appreciably; 2) the change $\Delta\sigma_\mu$ becomes positive when the temperature or the microwave power are increased.

Figure 4 shows the temperature dependence $\Delta\sigma_\mu(T)$; we see that σ_μ is almost independent of the microwave power for $T \gtrsim 10$ K. Unlike σ_μ, σ_0 is independent of temperature for $T = 1.4$–60 K.

We next discuss how $\Delta\varepsilon'$ and $\Delta\varepsilon''$ depend on the external magnetic field H_0. According to Fig. 5, $\Delta\varepsilon'$ decreases when H_0 passes through the resonant values corresponding to EPR for the dislocation dangling bonds. We note that following points.

1. The change $\delta\Delta\varepsilon$, at resonance was observed for microwave power much lower than the power for which the above-noted change in the losses $\Delta\sigma_\mu$ occurred in a vanishing magnetic field (cf. Fig. 3). The resonance change $\delta\Delta\varepsilon'$ increased with the microwave power and then saturated.

2. Significant magnetoresistance, i.e., a drop in the photoconductivity as H_0 increased from 0 to 12 kOe, was observed in the dc measurements of the spin-dependent recombination. We had to employ a special method[4] to identify the SDR-associated change in σ_0 against the background magnetoresistance. In contrast to the case for σ_0, we were unable to observe an appreciable difference in the values of $\Delta\varepsilon'$ or $\Delta\sigma_\mu$ for the fields $H_0 = 0$ and 12 kOe; in other words, the magnetoresistance for the rf conductivity was almost zero.

3. The was no resonance change in ε' when the sample was not illuminated. As the illumination (i.e., the LED current I) increased, $\delta\Delta\varepsilon'$ increased abruptly and then saturated for $I > 1$ mA.

For high microwave powers we were also able to observe a resonance change in $\Delta\sigma_\mu$. In both cases, the sign of $\delta\Delta\varepsilon'$ and $\delta\Delta\sigma_\mu$ was opposite to the change in ε' and σ_μ during illumination.

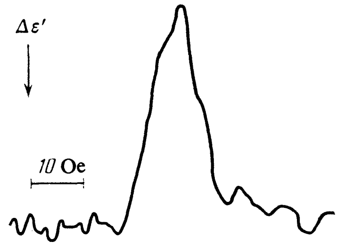

FIG. 5. Dependence of $\Delta\varepsilon'$ on the magnetic field H_0 near resonance with the DDB chains ($T = 1.4$ K).

DISCUSSION

The primary conclusion to be drawn from the experimental results is that the pronounced difference in the magnitudes of σ_0 and σ_μ and in their dependence on T and I clearly indicates that the conductivity of the sample is highly nonuniform. Because σ_0 is small compared to σ_μ, the sample may indeed be regarded as an insulating matrix with conducting inclusions. In this case we can define the average conductivity by

$$\langle\sigma_\omega\rangle = \frac{1}{V_s}\sum_i^M V_i\sigma_i,$$

where M is the number of conducting regions, σ_i and V_i are their specific conductivity and volume, and V_s is the volume of the sample. In terms of the quantities in Eqs. (1), (2), we can write $\langle\sigma_\omega\rangle = \nu\bar{S}\bar{l}$. The finding that ε' increases with the illumination I implies that $\langle\sigma_\omega\rangle$ must also increase, i.e., there is an increase in the number and/or conductivity S of the conducting regions.

As to the nature of these conducting regions, we believe that two descriptions are possible:

1. The carriers trapped in the dislocation states are responsible for the conductivity σ_ω. The conducting regions should then be segments of dislocations which are bounded by dislocation defects.

2. The conductivity σ_ω is due to free carriers is local regions of the crystal ("lakes") where the dislocation density is lower. Wide variations in the distribution of the dislocations could cause such lakes to be present.

Unlike the first case, the second explanation predicts that ε' and σ_μ should be influenced appreciably by the magnetic field. Since we did not detect any magnetoresistance for ε' and σ_μ in our work (whether the samples were illuminated or not), we conclude that the contribution of the lakes to the measured ε' and σ_μ is negligible. We note that we did observe a lake-associated rf conductivity for $T > 30$ K in samples doped to 10^{14} cm^{-3} or higher, as well as in samples doped to 10^{13} cm^{-3} but having a lower dislocation density. In this case the magnetoresistance was very large, and the difference $\sigma_\mu(0) - \sigma_\mu(H)$ was positive for $\sigma_\mu \ll \beta$ and negative for $\sigma_\mu > \beta$, where σ_μ is the conductivity of the undeformed sample [given by Eq. (10)]. These results are discussed in detail in Ref. 10.

We thus have reason to believe that the growth in $\langle\sigma_\omega\rangle$ during illumination is due to the increased conductivity associated with the dislocation states.

Figure 2b shows that the rf losses σ_μ drop abruptly at low temperatures (the size of the drop is $3\cdot10^{-6}$ $(\Omega\cdot\text{cm})^{-1}$). This implies that the dark conductivity σ_μ^{dark} of the sample prior to illumination must already exceed $3\cdot10^{-6}$ $(\Omega\cdot\text{cm})^{-1}$. For lightly doped p-type samples with a high dislocation density, the position of the Fermi level almost coincides with the deep dislocation level E_1. According to Ref. 7, the dark rf conductivity observed in these samples at low temperatures may be attributed to movement of the additional holes trapped in the dislocation state E_1. The number of holes and electrons trapped in the E_1 and E_2 states should decrease during illumination, because carriers of opposite sign are more likely to be trapped and recombination of the electrons and holes trapped by a DDB chain is more probable. The drop in the number of trapped carriers should then be accompanied by a decrease in the trapped carrier contribution to the measured values ε' and σ_μ. However, ε' is found to increase during illumination, which indicates that $\langle\sigma_\omega\rangle$ increases.

These results suggest that there exist shallower bands E_{ed} and E_{hd} (for electrons and holes, respectively) which have large free-carrier trapping cross sections but low $E_{ed} - E_{hd}$ recombination probabilities, and that these carriers move in the bands and contribute to the conduction. We will assume below that these bands coincide with the shallow intermediate states postulated in Ref. 4, and we will attempt to explain the experimental findings in the framework of this model. These bands are most likely produced by the elastic stress fields near the dislocations.

The carrier mobility in the shallow state may be assumed to be much greater then in the deep bands E_1, E_2, which is ~ 10 cm^2/V·s). The linear conductivity S of a section of dislocation may thus increase during illumination due to trapping of carriers in the E_{ed} or E_{hd} states. We now estimate the average length of the conducting sections for the case when the dark conduction is associated with carriers in the E_1 state. As we noted above, σ_μ^{dark} is $\gtrsim 3\cdot 10^{-6}$ $(\Omega\cdot\text{cm})^{-1}$. If we assume that the density of the conducting holes trapped in the DDB chains is 10^{13} cm^{-3} and that their mobility is 10 cm^2/(V·s), we find from (1) that $l \approx 200$ Å. Moreover, since $\varkappa S/l^2 \approx \beta$, the increase in S during the illumination will increase ε' but cause σ_μ to drop in accordance with Eqs. (1), (2).

We next discuss how the microwave power and temperature affect $\Delta\sigma_\mu$. Figure 3 shows that $\Delta\sigma_\mu$ changes sign for $T = 1.4$ K as the microwave power is increased, but it increases for $T \gtrsim 4.2$ K. The effect of increasing the microwave power is thus similar to increasing T (cf. Fig. 4). This suggests that the carriers trapped in the one-dimensional bands E_{ed} (E_{hd}) are heated. We can understand these results qualitatively by invoking a model in which modulation of the deformation potential by various types of defects causes the one-dimensional dislocation bands to be nonideal. We consider two kinds of perturbations:

1. "Macro" defects in the dislocation chains (constrictions, reconstructed regions, etc.), which produce very high potential barriers that block the carriers moving along the deformation potential band.

2. Defects which modulate the band less strongly, e.g., impurity atoms and point defects located near a dislocation.

At low temperatures both types of defects tend to localize the carriers in the one-dimensional band and thereby alter the phenomenological parameters l and S in Eqs. (1), (2). The influence of defects of the second type was considered in detail in Refs. 11, 12. The localization length increases somewhat as the carriers heat up; l therefore increases until it becomes limited by defects of the first type. According to Eq. (1), the accompanying drop in $\varkappa S/l^2$ should cause $\Delta\sigma_\mu$ to increase, as is observed experimentally.

According to Fig. 4 $\Delta\sigma_\mu$ starts to decrease for $T > 10$ K, and the formal activation energy E_a is ≈ 13 meV. This can hardly be attributed to a drop in the conductivity; a more likely explanation is that fewer carriers are trapped in the shallow bands. We have the balance equations

$$dn_0/dt = G - n_0 W_t + n_d W_D, \quad dn_d/dt = n_0 W_t - n_d W_D - n_d W_c,$$

for n_0 and n_d, the number of free electrons and the number of electrons in E_{ed}, respectively. We thus find that

$$n_d = G/W_c, \tag{3}$$

$$n_0 = G(1 + W_D/W_c)/W_t, \tag{4}$$

where G is the rate of photogeneration of electrons; W_t is the trapping probability for free electrons by E_{ed}; W_D is the probability that electrons will be trapped from E_{ed} into the E_2 band (i.e., by a DDB chain) and then recombine. Thus, $W_c \sim \exp(-\Delta/kT)$ where $\Delta \approx 13$ meV, in good agreement with the data in Ref. 4.

The rf photoconductivity σ_ω is observed to vary when the external magnetic field H_0 is swept near resonance with the DDB chains. The maximum change $\delta\Delta\varepsilon'/\Delta\varepsilon' \approx 2\cdot 10^{-2}$ occurred for $T = 1.4$ K. The magnitude of the SDR at dc current mass $\Delta n_0/n_0 \approx 8\cdot 10^{-3}$ in Ref. 4. According to the model used there, spin-dependent recombination occurs because of the increase in W_c at magnetic resonance. Equations (3) and (4) then imply that

$$(\Delta n_d/n_d)\,(\Delta n_0/n_0) \approx (W_D + W_c)/W_D.$$

Substituting the values of W_c and W_D from Ref. 4, we get $(W_c + W_D)/W_D \approx 4$, which is in satisfactory agreement with the experimental data.

We now want to make two observations.

1. The phenomenological value of l in (1), (2) is evidently somewhat greater than the actual length of the conducting regions. Indeed, (1) and (2) assume that the conducting regions are randomly distributed and uncorrelated. Although the average volume density of the conducting regions in our samples was in fact low, they were concentrated near the dislocation lines, so that their interaction cannot be neglected. This should make the effective length $\bar{l}$ in Eqs. (1), (2) greater than the physical length.

The finding that the DDB chains are very short is consistent with the large deep level transient spectroscopy (DLTS) signal observed from the chains.[13] Indeed, if the conducting chains were longer than the width of the depleted layer ($\approx\mu$m in the DLTS experiments), no DLTS signal would be observed.

2. The Coulomb interaction among the carriers trapped at a dislocation must be included in calculations of W_c, W_D, and W_t. For instance, an increase in n_d should result in an extra Coulombic contribution to the deformation potential which tends to make the E_{ed} band shallower. Coulomb interactions should give rise to negative feedback in the system which tends to stabilize the density n_d. This would lead to an

increase in W_c (and hence in W_D) as the illumination I increases (i.e., the dependence $\Delta\varepsilon'(I)$ should become sublinear, cf. Fig. 2) and n_0 should depend slightly on T. Among other things, this implies that the activation energies corresponding to W_c and W_D which were found in our work and in Ref. 4 probably cannot be used to estimate the actual depth of the bands produced by the deformation potential for neutral dislocations.

Our data thus support the model proposed in Ref. 4 and suggest that carriers recombine along DDB chains in silicon crystals containing dislocations, where they are captured after preliminary trapping in shallow one-dimensional dislocation bands produced by the deformation potential. The observed change in the rf photoconductivity when the magnetic field is swept through resonance with the DDB chains is the consequence of the depopulation of carriers in the shallow one-dimensional bands.

The above SPR model readily predicts that the rate of thermal activation of carriers from the deep dislocation levels E_2 or E_1 (produced by the DDB chains) into the conduction (or valence) band will also be spin-dependent. Indeed, it is reasonable to assume that an electron will first be excited from the E_2 state into an E_{ed} state, from which it will either be activated into the conduction band or else will be trapped back into the DDB chain. Because the thermal activation gives rise to a singlet pair, the probability for back-trapping of the electron by the DDB chain can be decreased if a resonant magnetic field "mixes" the singlet and triplet configurations during the lifetime of the pair. The result will be an increase in the thermal generation rate at resonance, which can be detected experimentally as an increase in the dark conductivity of samples which contain depleted regions in which the current is determined by the rate of thermal activation. Such depleted regions may be present in n-type silicon with a highly irregular dislocation distribution, or in specially fabricated junctions and Schottky diodes.

Observations of this spin-dependent current transport were described in Ref. 14 for samples with irregularly distributed dislocations. Spin-dependent current transport in p-n junctions was noted in Ref. 15.

Finally, optical nuclear polarizations (ONP) was observed in Ref. 16 when plastically deformed silicon samples were irradiated by unpolarized light. Analysis revealed that the formation of centers with spin $s > 1/2$ in dislocation chains was responsible for the nuclear polarization. As we have noted above, illumination is accompanied by accumulation of DDB chain-E_{ed} electron pairs which process a strong triplet component. The results in Ref. 16 are in excellent agreement with the proposed model for spin-dependent recombination and can be explained in terms of the interaction of the nuclear system with these pairs.

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