Investigation of spin-dependent recombination in semiconductors

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The paper reports an experimental and theoretical investigation of spin-dependent recombination in plastically deformed Si. It is shown that the recombination occurs via the localized bound state of the electron-hole pair.

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The effect of a magnetic field on the recombination of non-equilibrium current carriers was discovered in the pioneer work of Lepine and Prejean.[1] In that work, the photoconductivity of n-Si located in a constant magnetic field $H_0 \approx 3$ kOe and a transverse—to it—microwave field h was investigated. The decrease in the photoconductivity due to the increase in the rate of recombination via surface impurity centers was recorded at the moment of resonance ($\gamma H_0 = \omega$). A new resonance method of investigating impurity centers was thus discovered which differs from the EPR method in that only the electrically active centers participating in the recombination process are detected in it.

In the initial model[1] it was assumed that the recombination proceeds via the paramagnetic centers, so that the constant magnetic field, which is responsible for the polarization of the free carriers ($p_n \approx \mu H_0/kT$) and the unpaired electrons at the centers ($p_N \approx \mu H_0/kT$), should decrease the rate R of recombination by an amount $\Delta R / R_0$ of the order of $p_n p_N \approx (\mu H_0/kT)^2$. The resonance microwave field destroys the polarization, and brings about at saturation the reestablishment of the initial rate R_0 of recombination.

Subsequently, spin-dependent recombination of the carriers was observed not only at the surface, but also at the volume, centers in plastically deformed[2–4] and amorphous[5] silicon. A number of experiments[6,7] have been performed in which the spin-dependent recombination of the nonequilibrium carriers in silicion p-n junctions was investigated, and this is especially interesting from the standpoint of the possible practical application of this effect. It was shown in these experiments that, as a rule, the relative change $\Delta R / R_0$ exceeds the simple estimate $(\mu H_0/kT)^2$, which is equal to 10^{-6} under the standard conditions of $H_0 \approx 3$ kOe and $T = 300$ K. In experiments on Si samples prepared in a special manner, the quantity $\Delta R / R_0$ attained a value of 5×10^{-4} under these conditions.[5,8]

To explain the large value of $\Delta R / R_0$, there was proposed in the model of recombination via paramagnetic centers a mechanism whereby the polarization p_N increases as a result of the combination of the centers into volume-ordered clusters.[9,10] Another mechanism, which is connected with the local warmup of the spin system, and can be responsible for the increase in the recombination rate R at paramagnetic resonance, is considered in Ref. 11. In this model we also have $\Delta R / R_0 \approx (\mu H_0/kT)^2$, but the coefficient of proportionality can be large. A fundamentally different model, which does not contain the small quantity $(\mu H_0/kT)^2$, was proposed for spin-dependent recombination in 1978 by Kaplan, Solomon, and Mott.[12] In their model, a recombination event precedes the formation of a bound state by an electron and a hole. Such a pair can either recombine or break up as a result of the electron's or hole's being thrown into the corresponding band. A fundamental assumption is that the recombination of the electron with a "foreign" hole does not occur in this case. The physical nature of these electron-hole pairs can vary. In a highly disordered semiconductor, their formation may be the result of the localization of the electron and hole at forbidden-band levels in close proximity to each other. If we neglect the weak spin-orbit interaction, then the recombination (in a pair) that gives rise to a state with zero total spin can proceed only from the singlet state. Therefore, the magnetic fields H_0 and h, which induced singlet-triplet transitions, can greatly increase the recombination rate. It is clear that in this model the polarization of the spin system as a whole is unimportant, only the spin correlation of the partners in a pair being important. Therefore, the parameter $\mu H_0/kT$ should not enter into the answer at all, and a fundamentally different dependence on the constant magnetic field H_0 should be expected.

In the present paper we carry out experimental and theoretical investigations of spin-dependent recombination in Si with a view to determining, and carrying, out as detailed a study as possible, of its mechanism. In §1 we develop a quantum theory of spin-dependent recombination with allowance for spin relaxation in the presence of a constant and a variable magnetic field. In §2 we describe the experimental data on plastically deformed silicon, obtained in the temperature range 100–300 K in different magnetic fields H_0 and a transverse resonance microwave field.

§1. QUANTUM THEORY OF SPIN-DEPENDENT RECOMBINATION VIA BOUND STATES UNDER MICROWAVE-RESONANCE CONDITIONS

1. Introduction

As has already been noted, Kaplan, Solomon, and Mott[12] have proposed for spin-dependent recombination in semiconductors a new model based on the assumption that the recombination proceeds via the bound state of the electron-hole pair. In order to show that the microwave-field induced change in the recombination can be large, the auth-

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ors carrier out a simple $\Delta R/R$ calculation under microwave-resonance-saturation conditions, using the classical approach in which the pairs are described with the aid of the function $N(\theta)$ giving the distribution of the pairs over the angle between the spins. They considered the change that occurs in $N(\theta)$ as a result of the dissociation and recombination in the singlet state of the pairs. They found that

$$\Delta R/R=(R^{\text{sat}}-R)/R=\{(1+4\lambda)\,[1-2\lambda\ln(1+1/2\lambda)]^{-1}\}-1; \tag{1.1}$$

here $\lambda = W_D/W_s$, where W_D is the rate of dissociation of the pairs and W_s is the rate of their recombination in the singlet state.

Thus, the magnitude of the effect can indeed be large: it can attain a value of 0.1. The expression (1.1) does not exhibit the dependence of the effect on the constant magnetic field, a dependence which, as has already been noted, can serve as a criterion for the selection of a model for the recombination as realized in a particular situation. In order to describe this dependence and make a detailed comparison of the results of the theory with experiment possible, we developed in our previous paper[13] a classical theory that takes account of the change that occurs in the pair distribution function as a result of the motion of the spins in the constant and variable magnetic fields. The results of these calculations show, in particular, that the H_0 dependence of $\Delta R/R_0$ indeed saturates in sufficiently high magnetic fields H_0 if the difference between the Larmor-precession frequencies of the partners in a pair becomes greater than the reciprocal lifetime of the pair. In the opposite case $\Delta R/R \propto H_0^2$. This result of the theory is in qualitative agreement with the results obtained in our experiments.[14] From this viewpoint, the simple and graphic theory developed in Ref. 13 is satisfactory, but there are no guarantees that a classical (nonquantum) approach to the description of spin 1/2 systems will lead to qualitatively correct results.

2. Basic equations

In the quantum theory, instead of the pair distribution function, we introduce a density matrix in the singlet-triplet representation $\rho_{ik} = N\langle \alpha_k^* \alpha_i \rangle$, where N is the total number of pairs, the α are the coefficients of the expansion of the ψ function of one pair in terms of the singlet state ψ_s and the three triplet states: ψ_0, the state with zero spin component along the preferred axis, and $\psi_\pm$, the states with spin components equal to ± 1. Let us use the equation of motion for $\rho(t)$:

$$d\rho/dt=-i\hat{\mathscr{H}}\rho(t)+\hat{R}\rho(t), \tag{1.2}$$

in which $\mathscr{H}$ is the Hamiltonian for the interaction with the magnetic field $\mathbf{H}$:

$$\hat{\mathscr{H}}=-(\mu_1+\mu_2)\mathbf{H}={}^1/_2(\gamma_1\hat{\sigma}_1\mathbf{H}+\gamma_2\hat{\sigma}_2\mathbf{H}), \tag{1.3}$$

that takes account of the fact that the gyromagnetic ratios of the electron and hole in the pair are different ($\gamma_1 \neq \gamma_2$). The magnetic field $\mathbf{H}$ contains a constant component $\mathbf{H}_0$ and a circularly polarized microwave component perpendicular to the constant component:

$$H_x=h\cos\omega t, \quad H_y=-h\sin\omega t, \quad H_z=H_0. \tag{1.4}$$

In the basic equation (1.2) we phenomenologically take into account terms $\widetilde{R}\rho(T)$ describing the following stochastic processes:

(a) the thermal dissociation of the pairs, which does not depend on their spin state:

$$(\hat{R}_D\rho(t))_{ik}=-W_D\rho_{ik}; \tag{1.5}$$

b) the recombination of the pairs, which, by our assumption, occurs only in the singlet state:

$$(\hat{R}_R\rho(t))_{ik}=-W_{ik}\rho_{ik},$$

$$W_{ss}=W_s, \quad W_{si}=W_{is}={}^1/_2W_s \quad (i\neq s), \quad W_{ik}=0 \quad (i,\,k\neq s). \tag{1.6}$$

The thus obtained equation is solved in Ref. 15 with the initial condition $\rho_{ik} = \frac{1}{4}N\delta_{ik}$, which corresponds to an equiprobable distribution of the pairs over the spin states at the instant the magnetic fields are switched on.

But the steady-state solution of the equations for ρ_{ik} with a fixed rate of pair production corresponds better to the physical formulation of the spin-dependent-recombination problem. In the simplest case of isotropic pair production, the corresponding term has the form:

$$\text{(c)} \quad (\hat{R}_G\rho(t))_{ik}={}^1/_4G\delta_{ik}; \tag{1.7}$$

(d) it is also necessary to include the spin relaxation, neglected in Ref. 15, of the partners in Eq. (1.2):

$$(\hat{R}_{T_s}\rho(t))_{ik}=-T_s^{-1}(\rho_{ik}-{}^1/_4\delta_{ik}\,\mathrm{Sp}\,\rho). \tag{1.8}$$

The above-presented equations (1.2)–(1.8) allow us, in principle, to solve the problem, i.e., to find the density matrix of the pairs as a function of the amplitudes of the constant and microwave magnetic fields. We can then easily compute the rate R of recombination of the current carriers via the bound pairs:

$$R=W_s\rho_{ss}. \tag{1.9}$$

But the solution of this problem in the general form is somewhat difficult, for it requires the analysis of a linear system of sixteen complex equations for the elements of the density matrix. Let us show that the number of equations can be reduced to ten, if, instead of the elements ρ_{ik}, we use certain linear combinations of them, that have the classical meaning of the first coefficients of the expansion, in terms of the spherical harmonics, of a pair distribution function $N(\Omega_1,\Omega_2)$ that depends on two directions. Specifically, from the ρ_{ik} we go over to the combinations

$$N_{\alpha\beta}=\mathrm{Sp}(\hat{\rho}\,\hat{T}^{\alpha\beta}), \tag{1.10}$$

where the operators $\hat{T}^{\alpha\beta}$ are products of the Pauli matrixes σ_1^α and σ_2^β acting on the spin states of the electron and hole, respectively, in a pair. The superscripts α and β run through four values: 0 (in the case $\sigma^0 = 1$), z, and $\pm$ (in this case $\sigma^\pm = \sigma^x \pm i\sigma^y$). Then

$$R=W_s\rho_{ss}={}^1/_4W_s[N_{00}-N_{zz}-\mathrm{Re}\,N_{+-}].$$

In terms of the $N_{\alpha\beta}$, the equations for $N_{0\pm}$, $N_{\pm 0}$, N_{0z}, and N_{z0} split off, and it is sufficient to study the equations for the remaining ten $N_{\alpha\beta}$ components. The relations (1.10) allow us to relate them to the density matrix:

$$N_{00}=\mathrm{Sp}\,\rho=N,\quad N_{zz}=(\rho_{++}+\rho_{--})-(\rho_{ss}+\rho_{00}),$$
$$N_{++}=N_{--}^{*}=4\rho_{-+},\quad N_{+-}=N_{-+}^{*}=\sqrt{2}[\rho_{00}-\rho_{ss}+\rho_{0s}-\rho_{s0}],$$
$$N_{z+}=N_{z-}^{*}=\sqrt{2}[\rho_{s+}+\rho_{0+}+\rho_{-s}-\rho_{-0}],$$
$$N_{+z}=N_{-z}^{*}=\sqrt{2}[-\rho_{s+}+\rho_{0+}-\rho_{-s}-\rho_{-0}]. \qquad (1.11)$$

The sought equations following from (1.2)–(1.8) and (1.11) have the form

$$dN_{00}/dt=G-W_DN_{00}-R,$$
$$dN_{zz}/dt=\mathrm{Im}\{h[\gamma_1N_{-z}+\gamma_2N_{z-}]\}-N_{zz}/\tilde{T}+R,$$
$$dN_{z+}/dt=-i\gamma_2(H_0N_{z+}-hN_{zz})+{}^1/_2i\gamma_1(h^*N_{++}-hN_{-+})-N_{z+}/T^*+{}^1/_4W_sN_{+z},$$
$$dN_{+z}/dt=-i\gamma_1(H_0N_{+z}-hN_{zz})+{}^1/_2i\gamma_2(h^*N_{++}-hN_{+-})-N_{+z}/T^*+{}^1/_4W_sN_{z+}, \qquad (1.12)$$
$$dN_{+-}/dt=-i(\gamma_1-\gamma_2)H_0N_{+-}+i\gamma_1hN_{z-}-i\gamma_2h^*N_{+z}-N_{+-}/T^*+{}^1/_4W_sN_{-+}+2R,$$
$$dN_{++}/dt=i(\gamma_1+\gamma_2)H_0N_{++}+i\gamma_1hN_{z+}+i\gamma_2hH_{+z}-N_{++}/\tilde{T}.$$

Here we have adopted the following notation for the characteristic times:

$$1/T^*=1/\tilde{T}+{}^1/_4W_s,\quad 1/\tilde{T}=1/T_s+W_D. \qquad (1.13)$$

Notice that the field terms (i.e., the terms proportional to H_0 and h) in the quantum-mechanical equations (1.12) coincide completely with the field terms in the classical equations for the moments $N_{\alpha\beta}$ of the angle distribution function $N(\Omega_1,\Omega_2)$ for the pairs. The terms describing the production (1.7), dissociation (1.5), and spin relaxation (1.8) processes also coincide. But the pair-recombination processes (1.6) occurring in the singlet state make different contributions in the classical and quantum-mechanical approaches. This difference will not change the qualitative aspect of the results obtained in Ref. 13, but, as we shall show below, the quantitative difference between the results obtained in these approaches is quite significant. Naturally, this, in the final analysis, is connected with the inapplicability of the classical description to spin 1/2 systems.

3. Dependence of the recombination rate on the constant magnetic field

If there is no microwave field, then $N_{z+}=N_{+z}=N_{++}=0$, and, to determine the recombination rate $R(H_0,0)$, we need only investigate the closed system of equations for the quantities N_{00}, N_{zz}, and N_{+-}. We find as a result that

$$R(H_0,0)=G\left[\frac{4(W_s+W_D)}{W_s}-\frac{3}{1+W_DT_s}-2W_D\tilde{T}f(H_0^2)\right]^{-1}, \qquad (1.14)$$

where

$$f(H_0^2)=\frac{(\omega_{12}\tilde{T})^2}{(\omega_{12}\tilde{T})^2+1+W_s\tilde{T}/2},\quad 0\leqslant f\leqslant 1, \qquad (1.15)$$
$$\omega_{12}=\omega_1-\omega_2,\quad \omega_i=\gamma_iH_0,\quad i=1,2.$$

It can be seen that the effect of the spin relaxation on the zero-field recombination rate $R(,0)$ is determined by the parameter W_DT_s. In the absence of spin relaxation (i.e., for $1\ll W_DT$)

$$R(0,0)={}^1/_4GW_s/(W_s+W_D) \qquad (1.16)$$

and attains its maximum value of $\frac{1}{4}G$ when the pair-dissociation rate is low (i.e., when $W_D\ll W_s$). The discriminating factor $\frac{1}{4}$ is connected with the fact that the recombination proceeds only via the singlet state of the pairs and the fraction of pairs in this state is equal to $\frac{1}{4}$. But if the spin relaxation proceeds faster than the dissociation (i.e., if $W_DT_s\ll 1$), then the recombination rate has a higher value:

$$R(0,0)=GW_s/(W_s+4W_D), \qquad (1.17)$$

attaining a maximum value of G when $W_D\ll W_s$. This is due to the fact that each pair has enough time to go over into the singlet state and recombine before it dissociates.

The magnetic field brings about additional mixing of the singlet and triplet states, and increases the recombination rate in accordance with (1.14). If this intermixing occurs at a rate (determined by the frequency difference ω_{12}) higher than all the remaining rates (i.e., if $1\ll\omega_{12}\tilde{T}$), then the dependence $R(H_0)$ saturates:

$$R(\infty,0)=G\left[\frac{2W_s+4W_D}{W_s}-\frac{1}{1+W_DT_s}\right]^{-1}. \qquad (1.18)$$

Now, in the absencef relaxation ($1\ll W_DT_s$), the recombination rate is equal to

$$R(\infty,0)={}^1/_2GW_s/(W_s+2W_D) \qquad (1.19)$$

and can attain a value of $\frac{1}{2}G$, i.e., a value twice as large as the maximum value in the case when $H_0=0$. This is due to the fact that a magnetic field that saturates the S-T_0 transitions opens a recombination channel for the pairs in the T_0 state. In the case of fast spin relaxation (i.e., for $W_DT_s\to 0$) the S–T_0 transitions become saturated without the participation of the magnetic field, and the effect of the field therefore disappears:

$$R(\infty,0)=R(0,0)\left[1+2W_DT_s\frac{W_s}{W_s+4W_D}\right]. \qquad (1.20)$$

Let us, in conclusion of this section, note that in the low-H_0 region, where $\omega_{12}\tilde{T}<1$, the increase $\Delta R(H_0,0)\equiv R(H_0,0)-R(0,0)$ in the recombination rate is, as can be seen from (1.14), proportional to the square of the field intensity.

4. Dependence of the recombination rate on the variable magnetic field in the region of saturating constant magnetic fields

In a saturating constant magnetic field, when $1\ll\omega_{12}T$, the components N_{++} and N_{+-} are small in comparison with N_{zz}, and they can be neglected. The condition $1\ll\omega_{12}\tilde{T}$ also implies that the separation of the microwave resonances at the frequencies ω_1 and ω_2 exceeds their widths. Therefore, the only important quantity will, depending on the field frequency ω, be either N_{+z}, or N_{z+}. All this substantially simplifies the analysis of the system of equations (1.12). Assuming, for definiteness, that ω is close to ω_1, so that $1 \ll (\omega_2 - \omega)\tilde{T}$, we set $n_{++} = N_{+-} = N_{z+} = 0$ in (1.12), and obtain

$$R(\infty, h) = G\left[2 + \frac{4W_D}{W_s} - \frac{1}{1+W_D T_s} - W_D \tilde{T} f_1(h^2)\right]^{-1}, \quad (1.21)$$

where

$$f_1(h^2) = \frac{\gamma_1^2 h^2 T^* \tilde{T}}{1 + [(\omega_1 - \omega) T^*]^2 + \gamma_1^2 h^2 T^* \tilde{T}}; \quad 0 \leqslant h_1 \leqslant 1. \quad (1.22)$$

In the vicinity of the other resonance, where ω is close to ω_2, the expression for $R(\infty, h)$ has a form similar to (1.21). It is only necessary to make the substitutions $\omega_1 \to \omega_2$ and $\gamma_1 \to \gamma_2$. It can be seen from (1.21) that the microwave field gives rise to an additional increase in the recombination rate. This is due to the mixing of the states S and $T_\pm$ by the microwave field. At saturation of the microwave resonance, when $1 \ll \gamma_1^2 h^2 T^* \tilde{T}$ and $f_1(h^2) = 1$, we have

$$R(\infty, \infty) = G W_s / (W_s + 4W_D). \quad (1.23)$$

It can be seen that this expression does not depend on T, and coincides with the formula (1.17) for $R(0,0)$ in the fast-relaxation case, when $T_s \to 0$. This coincidence is explained by the fact that, in strong microwave and constant fields, both the S-T_0 and the S-$T_\pm$ transitions are saturated, which is entirely equivalent to the effect of the spin relaxation. It follows from (1.23) and (1.18) that

$$\frac{\Delta R(\infty)}{R(\infty, 0)} = \frac{R(\infty, \infty) - R(\infty, 0)}{R(\infty, 0)} = \frac{W_s}{W_s + 4W_D} \frac{W_D T_s}{1 + W_D T_s}. \quad (1.24)$$

The second cofactor in (1.24) describes the decrease of the effect as a result of the spin relaxation.

It is useful to have a formula for $\Delta R(h)/R(\infty, 0)$ in the region of weak microwave fields in case we have to analyze experiments in which microwave saturation cannot be achieved:

$$\frac{\Delta R(h)}{R(\infty, 0)} = \frac{4W_D W_s \tilde{T}^3 \gamma_1^2 h^2}{[1 + (\Delta\omega T^*)^2][4 + W_s \tilde{T}][W_D W_s \tilde{T} + W_s + 4W_D]}. \quad (1.25)$$

The resonance width is determined by the total spin-relaxation time T^*, contributions to which are, according to (1.13), made not only by the natural relaxation of the spins in a pair, but also by the processes of recombination and dissociation into unpolarized bands.

5. Spin-dependent recombination in intermediate magnetic fields

It is possible to consider another physically interesting case, in which the microwave resonances are not yet separated ($\omega_{12}\tilde{T} < 1$), but the constant field H_0 is high enough so that the resonance curve has already been formed ($\Delta\omega\tilde{T} > 1$). In this case all the $N_{\alpha\beta}$ components entering into Eq. (1.12) are important, and the solution of the problem becomes substantially complicated. But in the present case the saturation of the microwave resonance occurs at higher microwave-power levels, which are difficult to attain in experiment: $\gamma_{12}^2 h^2 \tilde{T}^2 > 1$. Therefore, we shall limit ourselves to the case of microwave fields that are weak in this sense. This allows us to construct a perturbation theory in terms of not only the amplitude H_0, but also the amplitude h. After tedious calculations we obtain

$$\frac{\Delta R(H_0, h)}{R(H_0, 0)} = \frac{4W_D W_s \gamma_{12}^2 h^2 \tilde{T}^3}{(2 + W_3 \tilde{T})(3W_D W_s \tilde{T} + W_s + 4W_D)}\left[1 - \frac{4\bar{\gamma}^2 H^2 \tilde{T}^2}{(2 + W_s \tilde{T})}\right]. \quad (1.26)$$

Here

$$\gamma_{12} = \gamma_1 - \gamma_2, \quad \bar{\gamma} = (\gamma_1 + \gamma_2)/2. \quad (1.27)$$

It can be seen that the formula (1.26) for the magnitude of the effect, in contrast to the formula (1.25) for the case of separated resonances, contains the small factor $(\gamma_{12} h \tilde{T})^2$. In the absence of spin relaxation (i.e., for $1 \ll W_D T_s$), the expression (1.26) assumes the simpler form

$$\frac{\Delta R(H_0, h)}{R(H_0, 0)} = \frac{\gamma_{12}^2 h^2 [1 - \bar{\omega}^2/(W_D + {}^1/_2 W_s)^2]}{2W_D (W_D + {}^1/_2 W_s)(1 + W_D/W_s)}. \quad (1.28)$$

If, on the other hand, the spin relaxation is strong (i.e., if $W_D T_s \ll 1$), then the magnitude of the effect should be significantly smaller:

$$\frac{\Delta R(H_0, h)}{R(H_0, 0)} = \frac{2\gamma_{12}^2 h^2 (1 - \bar{\omega}^2 T_s^2) W_D T_s}{1 + 4W_D/W_s + (4W_D/W_s) W_D T_s}. \quad (1.29)$$

6. Spin-dependent recombination in the regions of weak magnetic fields

The effect of a weak field in the absence of a microwave field has already been considered in Subsec. 3. From (1.14) we easily obtain

$$\frac{R(H_0, 0) - R(0, 0)}{R(0, 0)} = \frac{2\gamma_{12}^2 H_0^2 \tilde{T}^2}{(1 + W_s \tilde{T}/2)(3 + 1/W_D \tilde{T} + 4/W_s \tilde{T})}. \quad (1.30)$$

We can, by analyzing the basic equations (1.11) for $H = 0$ and small h, obtain a formula for $[R(0, h) - R(0, 0)]/R,(0, 0)$. It can be obtained from (1.30) by making the substitution $H_0^2 \to h^2/(1 + (\omega\tilde{T})^2)$. At low frequencies, i.e., when $\omega\tilde{T} \ll 1$, there is no difference between H_0 and h, and the expression for $\Delta R(H_0, h)/R(0, 0)$ then contains the sum of the squares of the intensities of the mutually perpendicular fields, i.e., the square of the intensity of the resultant field. In other words,

$$\frac{\Delta R}{R} = \frac{R(H_0, h) - R(0, 0)}{R(0, 0)} = \frac{2\gamma_{12}^2 (H_0^2 + h^2) \tilde{T}^2}{(1 + W_s \tilde{T}/2)(3 + 1/W_D \tilde{T} + 4/W_s \tilde{T})}. \quad (1.31)$$

A comparison of the results obtained with the results of the semiclassical calculation[13] shows that the magnitude of the effect in the quantum model is almost an order of magnitude greater.

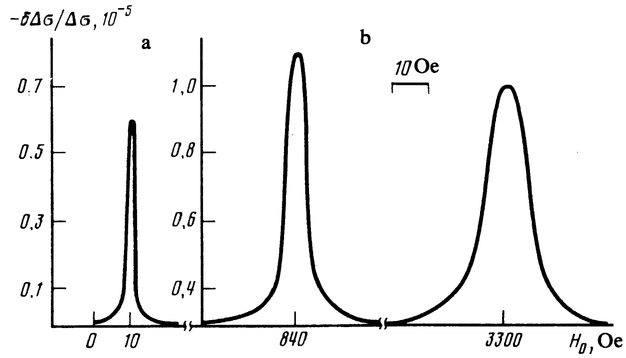

FIG. 1. Resonance photoconductivity curves at frequencies of: a) 30 MHz; b) 2.4 and 9.4 GHz.

§2. THE EXPERIMENTAL RESULTS AND THEIR DISCUSSION

Spin-dependent recombination was experimentally studied by us in plastically deformed silicon. Samples of the parent n-type material with a resistance of 60 Ω /cm at room temperature were prepared in the form of bars with the long edges along the $\langle 110\rangle$ direction. Dislocations were introduced by plastic deformation during compression along the $\langle 110\rangle$ direction. The method of preparing the investigated samples is described in greater detail in Ref. 8. The dislocation density in the thus obtained samples was 10^7–10^9 cm^{-2}. The relative change $\Delta R/R$ in the spin-dependent-recombination rate was determined by measuring the relative change in the photoconductivity under spin-resonances conditions ($\Delta R/R = -\delta\Delta\sigma/\Delta\sigma$). The spin-dependent photoconductivity was measured, using the same procedure employed in the experiments reported in Refs. 8 and 10. The photoconduction was produced by an incandescent lamp with an optical filter having a radiation-transmission band extending to 1.3 μm. The change $-\delta\Delta\sigma/\Delta\sigma$ was studied in the broad range of variation of the magnetic-field intensity from 10 to 3300 Oe and at the frequencies corresponding to these fields (30 MHz–9.3 GHz). Figure 1 shows the resonance curves of the photoconductivity change for resonance frequencies of 30 MHz, 2.4 GHz and 9.3 GHz, and Fig. 2 shows the dependence of the change $-\delta\Delta\sigma/\Delta\sigma$ observed at resonance on the power of the resonance electromagnetic radiation. The halfwidth ($\gamma\Delta H = 1/T^*$) of the resonance line at half-maximum of its amplitude depends on the intensity of the constant magnetic field. Figure 3 shows the experimental $\Delta H(H_0)$ curves for the variable fields corresponding to the linear section of the $\delta(P)$ curve. The parameters of the resonance signal in the 3300-Oe field were studied in the temperature range 100–300 K. It turned out that H and the threshold microwave-field intensities, i.e., the intensities at which saturation of the signal begins, practically did not depend on temperature. The temperature dependence of $-\delta\Delta\sigma/\Delta\sigma$ is shown in Fig. 4. In the region of weak constant magnetic fields, the photoconductivity depends on H_0 and h in a complicated manner.

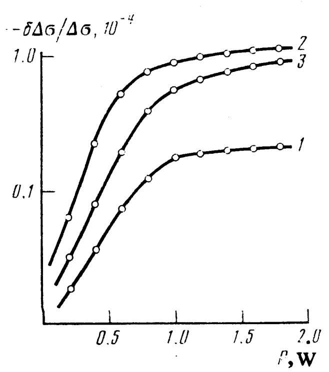

FIG. 2. Dependence of $-\delta\Delta\sigma/\Delta\sigma$ at resonance on the power of the electromagnetic radiation at frequencies of 30 MHz (curve 1), 2.4 GHz (curve 2), and 9.4 GHz (curve 3).

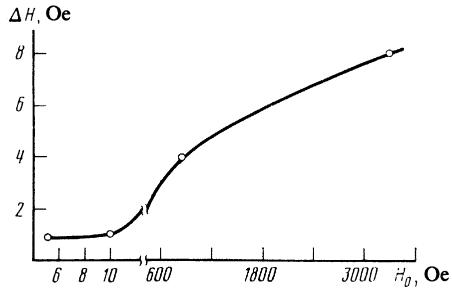

FIG. 3. Dependence of the halfwidth of the resonance line (in the linear region of the dependence in Fig. 2) on the resonance-field strength.

The following experimental fact is of fundamental importance for the determination of the mechanism underlying spin-dependent recombination. Under conditions close to saturation, the value of $-\delta\Delta\sigma/\Delta\sigma$ does not change by more than an order of magnitude in the constant-magnetic-field range 10-3300 Oe. The models proposed in Refs. 10 and 11 predict a five-to-six order-of-magnitude change in $-\delta\Delta\sigma/\Delta\sigma$ in the indicated field range.

The observed H_0 dependence of $-\delta\Delta\sigma/\Delta\sigma$ is consistently explained only by the model developed in §1 on the basis of the assumption that spin-dependent recombination proceeds via paired states. Indeed, as can be seen from the formulas (1.24), the effect should not depend on the magnetic-field intensity when $1 \ll \omega_{12}\widetilde{T}$, i.e., in the case of separated resonances. True, two separate resonance curves are not observed in our experimental data (see Fig. 1). In our opinion, this may be due to the inhomogeneous line broadening, which is neglected in our simple theory. Let us recall that we consider in the theory electron transitions between two levels fixed with respect to energy, g factor, and other parameters (W_s, W_D, T_s, etc.). In fact, for plastically deformed

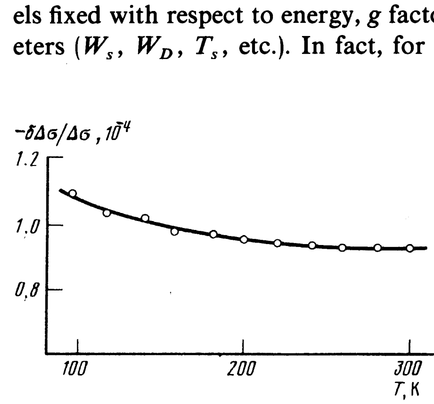

FIG. 4. Temperature dependence of $-\delta\Delta\sigma/\Delta\sigma$.

silicon, such an approximation is justified only at the first stage of the development of the theory. If we take account of, for example, the statistical spread in the values of the g factors, then this will lead to the smearing of the observed $\sigma_1 = \gamma_1 H_0$ and $\omega_2 = \gamma_2 H_0$ resonance lines right up to the point of their coalescence. It is important to note that, when the condition $1 \ll \omega_{12}\tilde{T}$ is fulfilled, the recombination rates for each pair of centers do not depend on ω_{12}. Therefore, all the conclusions about the broadening will remain unchanged, except that the resonance curves can merge completely. The absolute magnitude of the effect then decreases by a factor of $\delta\omega_H T^*$, which takes account of the fraction of centers that are simultaneously in resonance. If the inhomogeneous line width $\delta\omega_H$ is due to the g-factor spread, then $\delta\omega_H \sim H_0$. Such a field dependence of the resonance-line width is indeed observed in Fig. 3. This allows us to estimate the magnitude of the inhomogeneous g-factor spread: $\delta g_H/g = 5\times10^{-3}$. The difference $g_{12} = g_1 - g_2$ between the g factors in each pair should not exceed this value. Then the expected inhomogeneous line width in a field of intensity $H_0 = 10$ Oe is of the order of 0.05 Oe, which is significantly smaller than the measured width ≈ 1 Oe. Taking account of the fact that the amplitude of the measured effect (i.e., $\delta\Delta\sigma/\Delta\sigma$) begins to decrease noticeably in this field region, we should assume within the framework of the above-developed theory that the individiual widths $1/T^*$ of the two resonance lines in a pair become close to the separation ω_{12} of the lines. Thus, we obtain the estimate $1/T \approx 1$ Oe at room temperature.

In fields of intensity lower than 5 Oe, the line width is comparable to the resonance-field intensity. This leads to the distortion of the shape of the resonance curve, and makes the interpretation of the experimental data significantly difficult and, at this stage, inexpedient. Let us only note that the tendency of the magnitude of the effect to decrease with decreasing H_0 is clearly observed in this field region. The obtained estimate $1/T^* \approx 1$ Oe is in good agreement with the resonance-saturation data for fields $h = h_{\text{sat}}$ of the order of 1–2 Oe in the microwave region. Taking account of the fact that $h^2 T^* \tilde{T} \approx 1$, we find in addition that

$$W_s < 1/\tilde{T} = 1/T_s + W_D.$$

Since the quantity h_{sat} practically does not depend on temperature, and W_D should depend on temperature exponentially, we can conclude that the dominant contribution to $\tilde{T}$ (and, consequently, to T^*) is made by the spin relaxation:

$$T^* \approx \tilde{T} \approx T_s, \quad W_D T_s < 1, \quad W_s T_s < 1.$$

When $W_D T_s < 1$, the formula (1.24) can be simplified to the form

$$\Delta R(\infty, \infty)/R(\infty, 0) = W_D T_s/(1 + 4W_D/W_s).$$

It is known from experiment (see Fig. 4) that this quantity depends weakly on temperature. This means that, in the experimentally realized situation in plastically deformed silicon, the pair states recombine at a rate W_s lower than the rate W_D of their dissociation, at least in the temperature range 200–300 K. Finally, assuming that the considered spin-dependent channel is the principal channel, we can estimate from the absolute value of the effect the important characteristic W_s of the pair recombination in the singlet state. Taking the inhomogeneous broadening in our region of values of the parameters into consideration, we obtain

$$\frac{\Delta R(\infty, \infty)}{R(\infty, 0)} = \frac{W_s T_s}{4} \frac{1}{\delta\omega_H T_s} \approx \frac{W_s}{4\delta\omega_H}.$$

Substituting into this expression the values $\Delta R/R = 10^{-4}$ and $\delta\omega_H = 10^8$ sec^{-1}, we obtain $W_s \approx 4\times10^4$ sec^{-1}. The value obtained for W_s clearly indicates that the paired centers are located fairly far away from each other (slight overlap of the wave functions). The quantity $1/W_s$ can be compared with the time constant τ of the spin-dependent recombination. The latter was determined by us from the variation of the photoconductivity under resonance conditions with the frequency of modulation of the microwave radiation. It turned out that τ and $1/W_s$ have the same order of magnitude. This serves as additional corroboration of the correctness of the above-developed ideas about the mechanism underlying, and the character of, spin-dependent recombination of carriers in plastically deformed silicon.

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