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Recombination of Electrons and Holes at Dislocations

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The phenomenon of hole-electron recombination at dislocations is examined, and it is demonstrated that the space charge barrier surrounding the dislocation may have a dominant effect in determining the characteristics of recombination. In particular, the inclusion of the space charge effect leads directly to the slow decay phenomena observed in silicon and n-type germanium. The characteristics of electrical fluctuations due to trapping at these levels are discussed on the basis of the model.

A. INTRODUCTION

Several recent experimental results suggest that electron-hole recombination in certain cases cannot be described wholly on the basis of the Shockley-Read theory.[1] It has been observed in many investigations[2] that under certain conditions instead of the simple exponential decay of injected carriers, a much slower decay is found. This deviation from the Shockley-Read theory arises in silicon at temperatures up to room temperature, and in germanium at temperatures the order of $-80°C$ and lower. In order to account for the slow, nonexponential behavior of the recombination of excess holes and electrons at low concentration, it has been suggested[3] that deep trapping centers are present. These traps communicate primarily with only one band, and act as a storehouse of injected carriers. When the cause of excess carriers is removed, there is immediate recombination of the added carriers present in the conduction and valence bands. However, excess holes or electrons remain in the deep center. These are ejected at a very low rate, and the corresponding photocurrent decays very slowly.

Another anomalous experimental result which we believe may be related is the recently observed $1/f$ noise, attributable to dislocations, which has been found under certain conditions in germanium at room temperature.[4] This latter result indicates that there is a slow decay process associated with dislocations, which, if expressed in terms of time constants, has time constants the order of 0.02 second (as the $1/f$ noise extends down to 10 cps at least). Thus dislocations have characteristic times at least up to the order of 0.02 second. In fact, from noise theory,[4] and the limits of present experiments, trapping at dislocations must be described by a spectrum of time constants τ, with spectral density $g(\tau)$, where $g(\tau)=1/\tau$ from $\tau=0.02$ second or higher to $\tau=10^{-4}$ second or lower.

It is apparent that time constants the order of 0.02 second and possibly higher, associated with dislocations, are the right order of magnitude to explain the slow decay processes observed in lifetime experiments. However, it cannot be assumed that levels at dislocations can only communicate with one band, as it is well known[5] that dislocation levels are probably the major contributing factor in bulk recombination of electrons and holes.

In this paper we will suggest a model to explain the various experimental results which does not depend on levels of unknown origin which can communicate with only one band. It will be shown that the barrier around the dislocation[6] or around an agglomerate of levels will cause such an effect. It will be found, using such a model, that although for large density of excess carriers the lifetime is low, at low density the apparent lifetime is sometimes high. In n-type germanium, at low injection density, excess holes may be "trapped" at the dislocation, and the recombination rate will be slow due to the fact that the rate of recombination is proportional to the density of holes left in the valence band, a much smaller density than the total injected carrier density. Thus the "trapping" of injected holes, which explains so many of the observed characteristics, is found in the present model also. A comparison of the theoretical predictions for germanium with the experimental observations will be shown to yield good agreement.

B. ANALYSIS OF RECOMBINATION AT DISLOCATIONS IN GERMANIUM

Consider the band structure about a dislocation or agglomerate of levels, as in Fig. 1. We can estimate the rate of capture of electrons and holes as a function of Z, the charge on the levels per unit length, or per agglomerate. The rate of capture of electrons and holes will be affected by the distorted band structure. We will analyze for the case of a dislocation in n-type germanium.

We can easily estimate the rate of electron capture if we assume that the important variable is the barrier height. It will be the number of electrons with sufficient energy to cross the barrier multiplied by a proportionality factor $1/\tau_n$. The height of the barrier at some small distance λ will be, from the solution of Poisson's equation,

$$V=-(eZ/2\pi K)\ln(r_0/\lambda),$$

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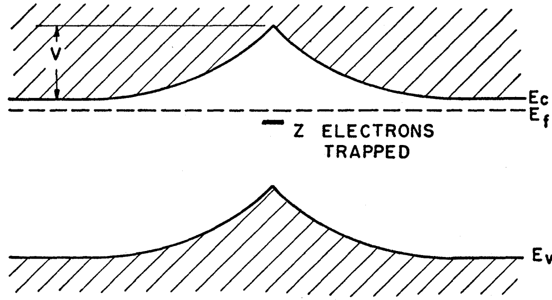

FIG. 1. Energy band diagram at a dislocation in germanium.

where $\pi N_D r_0^2 = eZ$, N_D the donor density. Thus the rate of electron capture can be approximately written

$$R_e = (1/\tau_n)[n \exp(-bZ) - n_0 \exp(-bZ_0)], \quad (1)$$

where $b = (e^2/2\pi KkT)\log(r_0/\lambda)$, λ being some small length related to the electronic wavelength. So b is the order of $10^{-7}/kT$ cm, where kT is in electron volts. The quantity Z should also appear in the coefficients of the exponentials. However, from Read's analysis[6] we find $bZ_0 > 1$ for T less than room temperature. We will therefore assume that the variation of the exponential factor with Z is much more rapid than the variation of the coefficients with Z.

The rate of electron capture can be alternatively calculated assuming a tunnelling process is the dominant mechanism. The expression obtained is similar, the constant being the same order, as it turns out that the energy of the incident electrons must be very close to the barrier height for tunnelling to occur. Another method which can be used to obtain the rate of electron capture is to use Read's[6] expression for the barrier height midway between two of the trapped electrons. By using the barrier height so obtained, a similar relation is again obtained, with again approximately the same value for b.

The rate of hole capture can be approximated by the expression

$$R_h = (1/\tau_p)\{p - p_0 \exp[b(Z_0 - Z)]\}, \quad (2)$$

with p_0 the equilibrium hole density, p the hole density, and where the last term, describing the rate of hole ejection from the levels, involves the barrier height bZ, the depth of the well in Fig. 1. Here again some simplifying assumptions have been used. The first is that there are essentially no holes trapped in the levels provided by the potential well shown, as in Fig. 1, to arise in the valence band. Thus the trapping of holes by the dislocation level arises chiefly by the direct transition of electrons in the dislocation to free holes in the valence band rather than the possible two-step process involving transitions first to a hole trapped in the potential well, and then to a free hole in the valence band. The validity of this latter assumption is questionable without a quantum-mechanical analysis of the number of levels in the potential well. It is apparent that if the well were truly three-dimensional, as arising from an agglomerate of impurities, the approximation would be excellent, as only one level of importance would exist for each trapped electron (using the hydrogen atom quantum-mechanical treatment), and the probability that this level is occupied by a hole is close to zero, the Fermi level being too high in the n-type material. Thus when the density of trapped electrons, Z, on the dislocation is small, so that approximately a Coulomb potential can be used for each, expression (2) is relatively accurate. With low temperature, hence high Z, and high injected hole density lowering the quasi-Fermi level, then the approximation may fail. A second approximation in (2) is that the exponential factor varies more rapidly with Z than the coefficients, so the latter are again assumed independent of Z.

A third approximation is that the capture cross section for holes is independent of the energy of the dislocation level, which varies with respect to the valence band as the barrier changes. An analogous approximation is inherent in the argument leading to Eq. (1). It is of interest to observe that with these approximations, the values of τ_n and τ_p can be related to the Shockley-Read[1] lifetimes τ_{n0} and τ_{p0}, obtaining for strongly n-type material $\tau_{n0} = \tau_n$, $\tau_{p0} = \tau_p \times Z/d$, where d is the number of states per unit length of dislocation. From experimental values[7] of τ_{n0} and τ_{p0} and estimated values[6] of Z/d, we can colclude that τ_p and τ_n are the same order of magnitude.

For simplicity we will set $\tau_n = \tau_p = \tau$ and $\mu_n = \mu_p = \mu$ throughout most of the manuscript. Until other details of the model can be described more accurately, there seems no advantage in a distinction between these similar constants. The important formulas will be repeated at the end as they would appear without the above simplification.

The rate of hole capture in steady state must equal the rate of electron capture, whence, from Eqs. (1) and (2),

$$\exp(-bZ) = [p + n_0 \exp(-bZ_0)]/[n + p_0 \exp(bZ_0)]. \quad (3)$$

Substituting to obtain the rate of recombination, using $n_0 \gg p$, we find

$$R = (\Delta p/\tau)\{n_0/[n + p_0 \exp(bZ_0)]\}$$
$$= (n_0/\tau)[\exp(-bZ_0)][\exp(b\Delta Z) - 1]. \quad (4)$$

Here $\Delta p = p - p_0$, $\Delta Z \equiv Z_0 - Z$, and ΔZ is always positive in an injection process, corresponding to a net increase in hole density at the dislocations. In obtaining the last form in Eq. (4), n was set equal to n_0 in Eq. (1). For this approximation, it can be easily shown that $\tau_p \exp(-bZ_0)$ must be much less than τ_n. If τ_p is the same order as τ_n, as indicated above the approximation

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is obviously valid. If τ_p is much greater than τ_n, the approximation should be valid at some low temperature.

If holes are injected into n-type germanium, then, some of the injected holes will be in valence band (at a given instant), and some in the dislocation levels. If Δp_1 holes are injected per unit volume,

$$\Delta p_1 = \Delta p + N_t \Delta Z,$$

where N_t is the number of dislocations per unit area. The excess conductivity arising from the injection will arise from the Δp_1 electrons which are added to the conduction band (to neutralize the injected holes) plus the Δp holes added to the valence band. Thus

$$\Delta\sigma / e\mu = 2\Delta p + N_t \Delta Z. \quad (5)$$

If $N_t\Delta Z \gg \Delta p$, a simple exponential decay will not be observed. For, from (5) and (4),

$$\tau(e\mu)^{-1}(d\Delta\sigma/dt) = n_0 \exp(-bZ_0) \times[\exp(b\Delta\sigma/e\mu N_t)-1]. \quad (6)$$

If, on the other hand, $N_t\Delta Z \ll \Delta p$, a simple exponential decay will be observed and the Shockley-Read theory will be obeyed. From (5) and (4),

$$\tau(d\Delta\sigma/dt) = \Delta\sigma. \quad (7)$$

Here the expression for Z_0 developed by Read[3] has been used. With this expression, and the estimated value for b, it is found than $n_0 \exp(-bZ_0) \gg p_0$ for all temperatures in a 2 ohm cm n-type sample.

It remains to establish the value of $N_t\Delta Z/\Delta p$, to determine whether (6) or (7) holds under given conditions; that is, whether slow or fast decay will be observed. From Eq. (3), relating ΔZ to Δp,

$$N_t\Delta Z/\Delta p = (N_t/b\Delta p) \times \ln\{1+\Delta p/[n_0 \exp(-bZ_0)+p_0]\}, \quad (8)$$

whence with Δp low

$$N_t\Delta Z/\Delta p = N_t e^{bZ_0}/bn_0, \quad (9)$$

which is a function of temperature and the density of donors and dislocations. As the temperature increases, bZ_0 decreases, and the exponential term decreases rapidly. So at sufficiently high temperatures, the value of $N_t\Delta Z/\Delta p$ is always less than unity, and a simple exponential decay is obtained.

To obtain an order of magnitude for the critical temperature above which the slow decay is not observed (experimentally occurring at about 200°K for n-type germanium), we will take N_t the order of $10^6/\text{cm}^2$, n_0 the order of $10^{15}/\text{cm}^3$, b the order of 6×10^{-6}, whence at the critical temperature $\exp(bZ_0) = 6\times10^3$ or $bZ_0 = 8.7$. Using the values for Z_0 calculated by Read,[6] it turns out that the critical temperature is the order of 200°, which is fortuitous agreement with experiment, considering the approximations we have used. This excellent agreement tends to supply more confidence in the value of the sensitive parameter b, which was estimated from purely theoretical considerations, when used in the further comparisons with experiment below. Note that the parameter N_t occurs in the logarithmic form in this calculation, so the critical temperature is rather insensitive to N_t.

If the temperature is low enough so that slow decay is possible, it is apparent, from the inequality $(N_t/bn_0) \times \exp bZ_0 > 1$, and from Eq. (8), that for slow decay, the inequality $\Delta p < N_t/b$ must hold. Thus in order that a slow decay be observed, the injection must be small and the temperature low, in agreement with experiment. A more quantitative comparison with experiment of the predictions of the theory is presented in the next section.

C. VARIATION OF PHOTOCONDUCTANCE WITH INTENSITY OF ILLUMINATION

As the rate of hole capture in steady state must equal the rate of hole generation, we have, from Eq. (4),

$$\alpha I = \Delta p/\tau, \quad (10)$$

where $n_0 \gg p_0 \exp bZ_0$, following Read's analysis, α is a proportionality constant. From (4) and (5), we find the relation between $\Delta\sigma$ and Δp:

$$\Delta\sigma/e\mu = 2\Delta p + (N_t/b)\ln\{1+\Delta p/[n_0 \exp(-bZ_0)]\}. \quad (11)$$

Combining (10) and (11), we obtain

$$\Delta\sigma/e\mu = 2\tau(\alpha I) + (N_t/b) \times \ln\{1+(\tau\alpha I)/[n_0 \exp(-bZ_0)]\}. \quad (12)$$

It is seen there are three characteristic regions as I increases from zero. If the former term is dominant, as at high intensity of illumination (fast decay),

$$\alpha I = \Delta\sigma/2\tau e\mu, \quad (13)$$

whereas if the latter is dominant, in particular for very low intensity, Eq. (12) reduces to

$$\alpha I = [2bn_0/N_t \exp(bZ_0)]\Delta\sigma/2\tau e\mu. \quad (14)$$

It is seen that this is independent of the dislocation density, as $N_t\tau$ is approximately a constant. From Eq. (9) the bracketed factor must be less than unity for a slow decay, so the photoconductance is much higher for a given intensity of illumination. Comparing (14) and (13), the ratio of "lifetimes" at high *vs* low intensity is given approximately by the bracketed factor in (14).

The predictions of the model can now be compared with the results of Fan *et al.*[2] Consider first the temperature dependence of the slow lifetime [Eq. (14)] Fan and his co-workers found an activation energy of 0.29 ev controlling the increase in τ as the temperature was lowered, over a temperature range between 180° and 230°K. From Eq. (14), the factor in the expression for the apparent lifetime which is most sensitive to temperature is $\exp(bZ_0)$, where $b = 10^{-7}/kT$, Z_0 can be

approximated from Read's analysis as $Z_0=3\times10^6-0.8\times10^4T$. The apparent activation energy should thus be about 0.3 ev, in good agreement with experiment.

Below 180°K, Fan *et al.* found the activation energy begins to decrease with decreasing temperature. Our simple model does not predict such behavior at low temperatures, but indicates the activation energy should be roughly constant to temperatures below 40°K (applying Read's analysis of Z_0 *vs* T). The disagreement may arise due to the neglect, in the simple model presented, of the potential well in the valence band. At extremely low temperatures, where Z_0 is correspondingly high, an appreciable density of holes may be trapped in this well when holes are injected, thus increasing the rate of hole capture and lowering the apparent lifetime.

Fan and his co-workers have obtained a plot of the photoconductance against intensity of illumination which can be compared directly with Eq. (12). With the exception of the quantity $(\alpha\tau)$ in Eq. (12), all the quantities can be estimated. The quantity $(\alpha\tau)$ can be obtained for Fan's sample by considering the photoconductance at high intensity (no slow-decay effects). The two linear regions of the curve, represented in the theory by Eqs. (13) and (14) are apparent in the experimental results. Quantitatively, the predictions of the theory are in good agreement. The ratio of the slow to fast lifetimes at 210°K, given by the bracketed factor in Eq. (14), can be calculated as 3, if one uses $N_t=10^6/\text{cm}^2$, and Read's value of Z_0, together with $b=10^{-7}kT$ cm. Experimentally the ratio turns out to be about 9. The range of photoconductance where the decay cannot be considered a simple exponential function, can be compared with experiment as follows. The value of the photoconductance where the contribution of the two terms in (12) is equal can be calculated. This corresponds experimentally to the case where the photoconductance is double that to be expected on the basis of the fast lifetime. If one uses our estimated values for the parameters in (12), the two terms are equal when $\Delta p=10^{11}$, or $\Delta\sigma/\sigma=2.8\times10^{-3}$. The corresponding experimental value is about 2×10^{-3}, in close agreement, considering the approximations which have been made.

For completeness, the formulas obtained without the simplifications $\tau_n=\tau_p$ and $\mu_n=\mu_p$ are listed below. Equation (6), describing the rate of the "slow" decay, becomes

$$\tau_n(e\mu_n)^{-1}d\Delta\sigma/dt = n_0\exp(-bZ_0)[\exp(b\Delta\sigma/N_te\mu_n)-1], \quad (15)$$

while Eq. (12) describing the photoconductance becomes

$$(e\mu_n)^{-1}\Delta\sigma=(1+\mu_p/\mu_n)\tau_p(\alpha I)+(N_t/b)\times\log\{1+[\tau_n(\alpha I)/n]\exp(-bZ_0)\}. \quad (16)$$

D. NOISE DUE TO DISLOCATIONS

Small fluctuations of the value of Z from the equilibrium value Z_0 will occur at a dislocation. When such a fluctuation occurs, the rates of hole and electron capture, as given by Eqs. (1) and (2), will become different from zero. Thus the concentrations of electrons and holes will vary due to the fluctuations in the value of Z. The decay of a fluctuation in Z will have the form, from Eqs. (1) and (2),

$$d\Delta Z/dt=B(e^{b\Delta Z}-1), \quad (17)$$

where B and b depend on whether equilibrium is being restored by hole or electron capture processes. The two processes can be considered independently in connection with noise, the resulting concentration fluctuations each giving rise to its own characteristic noise spectrum.

It has been shown by Morrison[8] that a decay law of the form of (17) may give rise to $1/f$ noise. The inverse frequency spectrum will be observed to an angular frequency $\omega>bB$. For electron capture this corresponds to $\omega>(bn_0/N_t\tau)\exp(-bZ_0)$ and for hole capture $\omega>bp_0/N_t\tau$.

From Read, who calculated the equilibrium value of Z_0, it is apparent that hole capture is the more promising mechanism to explain low frequency $1/f$ noise, as $p_0<n_0\exp(-bZ_0)$. However, even in this case, the lower limit to the $1/f$ spectrum at room temperature appears too high, for using Kulin *et al.*'s estimate[5] of $N_t\tau=300$ sec/cm, together with $b=10^{-7}/kT$ cm, it turns out that at room temperature $\omega>10^{-8}p$ sec^{-1}, where p_0 is the order of $6\times10^{11}/\text{cm}^3$. Experimentally, at room temperature $1/f$ noise has been measured[4] down to $\omega=60$ sec^{-1}, two orders of magnitude lower, on a bent germanium sample. A possible reason for the discrepancy is more complicated behavior of the dislocations if the space charge cylinders overlap, or if a large density of impurities is trapped at the dislocations. At somewhat lower temperatures, of course, $1/f$ noise over a wide frequency range is predicted, as p_0 decreases rapidly with temperature.

E. DISCUSSION

In the above analysis it has been shown that the unique characteristic of dislocations in n-type germanium can give rise to the observed slow-decay phenomena, both in lifetime and noise measurements. The slow-decay phenomena is not predicted for p-type germanium, in agreement with experiment. The required characteristic is the presence of a space charge region around the dislocation which does not occur in p-type germanium. It must be emphasized that an agglomerate of impurity levels will produce a completely analogous situation. This latter case cannot be analyzed completely at this time as there are too many unknown variables associated with the problem, such as the

[8] S. R. Morrison, Phys. Rev. 99, 1904 (1955).

number of impurities to be expected per agglomerate and the depth of the energy levels.

The analysis performed has been based on two important conditions. The first is that hole capture is the rate-limiting step in recombination, that is, $p \ll n \times \exp(-bZ_0)$. This was assumed following Read's analysis for dislocations in germanium. If this is not true, as may be the case for other materials, and in n-type germanium for high resistivity material, a somewhat different analysis must be performed. The second condition is that the potential well for minority carriers at the dislocation is essentially unoccupied by minority carriers. This condition may fail at extremely low temperatures and high injection concentrations. But at moderate temperature or low injection, it appears from a rough estimate of the density of levels in the well, that few holes will be trapped in the well.

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Antiferromagnetism and Antiferromagnetic Resonance in $CuBr_2 \cdot 2H_2O$ at 9800 Mc/sec

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Antiferromagnetic resonance was observed in a single crystal of $CuBr_2 \cdot 2H_2O$ at liquid helium temperature. This crystal shows an isomorphous structure with that of $CuCl_2 \cdot 2H_2O$ and the experimental results are analogous to those for the latter. However, the resonance field shifts to the higher field side and this is considered to be the result of a stronger molecular field than that of $CuCl_2 \cdot 2H_2O$. The Néel point of $CuBr_2 \cdot 2H_2O$ was inferred to lie between 5°K and 6.5°K.

We have recently observed antiferromagnetic resonance absorption in hydrated copper bromide $CuBr_2 \cdot 2H_2O$ at liquid helium temperature. As is well known, the antiferromagnetic substances whose resonance absorption can be observed under readily realizable experimental conditions are limited to a few appropriate antiferromagnetics. The difficulties usually encountered are that for most antiferromagnetic substances, one must use the microwaves in the submillimeter region or magnetic fields as high as 10^5–10^6 oersteds. Accordingly, in spite of the considerable success of the theory of antiferromagnetic resonance,[1–6] the experimental work has been performed only with $CuCl_2 \cdot 2H_2O$,[7–13] and more recently with $MnBr_2 \cdot 4H_2O$.[14] The former sample was thoroughly investigated by the Leiden group. Many other antiferromagnetic substances have been also investigated[15–17]; however, the results observed are not true antiferromagnetic resonance absorption but rather show the quenching effects of paramagnetic resonance just above the Néel temperature.

We prepared a single crystal of hydrated copper bromide $CuBr_2 \cdot 2H_2O$, which is isomorphous with $CuCl_2 \cdot 2H_2O$, by slowly growing it from the aqueous solution at room temperature. Considerable care must be devoted to prevent the precipitation of the anhydrous salt. The measuring microwave circuit is the usual one of the reflection type and can be employed for observations in the temperature range between 1.39°K and 300°K, and the frequency used is about 9800 Mc/sec.

The experimental results are summarized as follows:

1. $CuBr_2 \cdot 2H_2O$ shows antiferromagnetic resonance in the temperature range of liquid helium and shows paramagnetic resonance near 6.5°K. Accordingly, it may be inferred that the Néel point lies in the temperature range of about 5–6.5°K which is higher than that of $CuCl_2 \cdot 2H_2O$. It is noteworthy that the Néel temperatures of bromides are higher than those of chlorides both for the cupric and for the manganous

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