Resonant recombination of photoexcited light holes in germanium in a magnetic field

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Resonant capture of light holes on the ground and excited acceptor levels, with emission of optical phonons, was observed in the study of the photoconductivity of p-Ge in a magnetic field. An analysis of the experimental spectra yielded additional data on the energy dependence of the effective masses and g-factors of the light holes. It is shown that recombination takes place not from the very bottom of the magnetic subband, but from a discrete level separated from this subbband by the Coulomb field of the ionized center. It is shown that the cross sections for capture onto the ground and excited levels of the acceptor are of the same order in this process.

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1. INTRODUCTION

The study of nonradiative carrier recombination in semiconductors is made difficult by the fact that it is usually impossible to observe the elementary recombination acts themselves, since the recombination must be assessed by analyzing such macroscopic characteristics as, e.g., the temperature dependence of the electric resistivity. One of the recombination processes, however, namely carrier capture by an ionized center with emission of on optical phonon $\hbar\omega_0$, can proceed in a magnetic field in resonant fashion.[11] The resonance sets in because of singularities that appear in the state density in the magnetic subband when the condition

$$E_n - E^{(i)} = \hbar\omega_0 \qquad (1)$$

is satisfied, where E_n is the energy of the bottom of the n-th magnetic subband for the free carrier and $E^{(i)}$ is the energy of one of the carrier bound states on the impurity center, reckoned from one level. The

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latter level is best assumed to be the extremum of the free-carrier band in the absence of a magnetic field. In this case $E^{(i)}$ is negative. (For convenience, we renumber the $E^{(i)}$ levels in order of increasing excitation energy, assigning the superscript 0 to the ground state. The correspondence with the standard notation is explained in Table I below.)

As for the energy E_n, we have, for example, in the quasiclassical approximation

$$E_n=\hbar\Omega(n+\gamma)\pm{}^1/_2 g\mu H, \tag{2}$$

where n is a sufficiently large interger, γ is a constant, $\Omega=eH/mc$ is the cyclotron frequency, $m=(\hbar^2/2\pi)\partial S/\partial E$ is the cyclotron mass, S is the area of the extremal section of the equal-energy surface in k-space, g is the effective g-factor, and $\mu=e\hbar/2m_0c$ is the Bohr magneton.

The E_n levels go in succession through the energy values

$$E=\hbar\omega_0-|E^{(i)}| \tag{3}$$

in the quasiclassical limit periodically, in the reciprocal magnetic field scale

$$H_n^{-1}=nP+P(\gamma\pm{}^1/_4 g^*), \tag{4}$$

where the period is

$$P=2\pi e/c\hbar S(E), \tag{5}$$

$g^*=g(m/m_0)$ is the reduced g-factor, and m_0 is the mass of the free electron.

In the experiments described below we have succeeded in observing resonant capture of light holes in p-Ge on the ground level as well as on several excited acceptor levels. The description of the experimental technique is contained in the second section of the paper.

In the third section we identify the obtained resonant spectra. The identification is made complicated by the fact that we do not know beforehand that the spectra are determined just by relation (1), and to determine the energy E from the experimentally measured period we must know the function $S(E)$ contained in (5). This function is linear only in a parabolic band: $S_p=(2\pi m/\hbar^2)/E$. The function $S(E)$, however, was determined earlier[3] in the energy interval 40–150 meV by measuring the oscillations of the photocurrent in a magnetic field under monochromatic impurity excitation. After identifying with the aid of this function the resonant series, we actually obtained new experimental points on the $S(E)$ plot in the heretofore unfilled energy interval 25–40 meV. Therefore the present study is closely related to our earlier one,[3] which it supplements.

TABLE I.

Index	Levels of acceptor (Ga) Standard notation [6]	E, meV	$\hbar\omega_0-\|E^{(i)}\|$, meV [6,7]
0	8+0	26.9	26.7
1	8–01	33.5	33.45
2	8+1	34.6	34.75
3	8–02	35.5	35.2
4	8–11, 7–0	36.0	35.9

The fourth section of the article contains an analysis of the results. The topics discussed there can be divided into two groups. First, it is possible to obtain from the resonant spectra additional data on the spectrum of the light holes in the interior of the band, particularly the $g(E)$ dependence for magnetic field directions along [110] and [111]. Second, the narrowness of the resonance lines has made it possible to use in the analysis not only the oscillation period (5), but also directly the line positions on the magnetic-field scale. This has revealed a line shift due to the influence of the Coulomb field on the free-carrier spectrum in the immediate vicinity of this center. Since the magnetic field makes the motion of the free carriers finite in the plate perpendicular to **H**, the carriers at the bottom of the magnetic subband, whose velocity v_H along the field is low, turn out to be tied to the center, as it were. As a result, the degeneracy with respect to the orbit center is lifted for these carriers and discrete levels are separated from the bottom of the subband.[4] It is from these, properly speaking, that the capture by the center takes place. Third, an analysis of the spectrum leads to certain conclusions concerning the processes of recombination with participation of an optical phonon. Of greatest interest here is a comparison of the cross sections for the capture onto the ground and excited acceptor levels.

2. TECHNIQUE

We investigated in the experiments the dependence of the derivative $\partial J/\partial H$ of the transverse photocurrent on the magnetic field H. The measurements were made at 1.6 K. Nonequilibrium carriers were produced by infrared radiation incident on the sample from the upper warm parts of the cryostat. The samples of germanium doped with gallium [concentration $N_{Ga}=(1-7)\times10^{14}\,cm^{-3}$] [as well as for comparison with boron ($N_B=4\times10^{14}\,cm^{-3}$) and with indium $N_{In}=1.5\times10^{14}\,cm^{-3}$] were plates 0.5–1 mm thick. The contacts were produced by welding gold wire or by fusing-in and In-Ga alloy.[5] The distance between the contacts was of the order of 1 mm. Since the measurements were made in a **J**⊥**H** geometry, a voltage up to 1 V on the sample did not cause noticeable heating of the carriers. The sample was placed in superfluid helium at the center of a superconducting solenoid that produced a field up to 85 kOe. The light pipe was a stainless-steel tube connecting the sample holder with the upper flange of the cryostat. To make effective use of the light incident on the sample, the latter and its holder were placed in a spherical cavity with reflecting walls.

The magnetic field was modulated at 20 Hz at an approximate amplitude 10 Oe. The modulation was by an alternating component added to the superconducting-solenoid current. A standard amplification and synchronous detection circuit tuned to the modulation frequency registered the ac component of the photocurrent J, which was proportional to $\partial J/\partial H$. This component was plotted either against H or against H^{-1} with an $x-y$ recorder. To cancel the monotonic part of the H-dependence of the signal and to increase the amplification

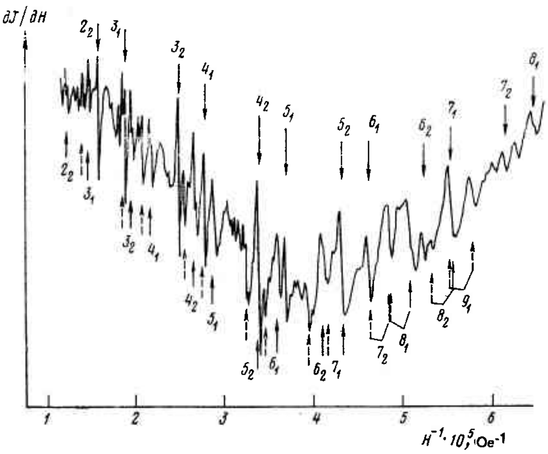

FIG. 1. Typical plot of the photocurrent derivative as a function of the reciprocal field. Here and in Figs. 4–6 the germanium is doped of gallium at a concentration 2×10^{14} cm^{-3}; the monotonic component, proportional to H^{-1}, was subtracted from the useful signal (the solid and dashed lower arrows correspond to $E^{(1)}$ and $E^{(2)}$, respectively).

thereby, a voltage proportional to H^{-1} was applied in to the Y coordinate in series with the measurement signal.

3. SPECTRA AND THEIR INTERPRETATION

Figure 2 shows a plot of $\partial J/\partial H(H^{-1})$ obtained for a germanium sample doped with gallium ($N_{Ga}=2\times10^{14}$ cm^{-3}) at a field direction $\mathbf{H}\parallel[100]$. It shows a large number of resonance lines. The relative line width determined by the distance ΔH between the maximum and minimum, divided by H_{res}, was ~1%. This value was approximately the same for all the observed resonances and was practically independent of the acceptor density in the range covered by the employed sample. The resonant value H_{res} was taken to be the average of the fields corresponding to the maximum and the minimum. These resonant values did not vary from sample from sample within 0.1–0.2%.

The spectrum shown in Fig. 1 was interpreted in the following manner:

a) It is relatively easy to separate from the entire spectrum a series of lines with somewhat larger amplitudes, which constitutes a set of pairs of extrema periodic in H^{-1}. This series is marked in Fig. 1 by arrows under the curve. From the period (5) we obtained by formula (5) the value S of the extremal intersection of the equal energy surface of the valence band with the (100) plane. The dependence on the magnetic-field direction corresponded to the anisotropy of the light-hole spectrum. A preliminary estimate based on the formula of the parabolic model, using the mass of the light holes on the top of the band, shows in this case that E is of the order of 30 meV. At this energy it is already necessary to take into account the deviation from parabolicity.

Therefore the entire analysis that follows takes into account the results of Ref. 3, where the deviations from parabolicity in the light-hole band were measured.

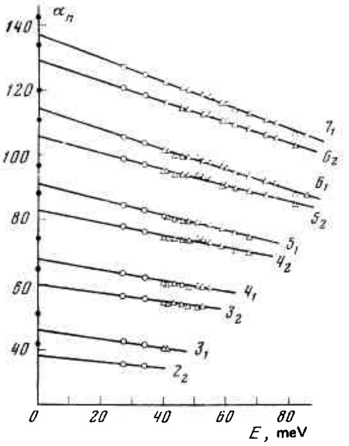

FIG. 2. Passage of the levels, from which the recombination proceeds, through fixed energy levels as the magnetic field is varied. Black circles on the ordinate axis–from Refs. 8 and 9, light circles–present study, triangles–from Ref. 3. The numbers on the right label the magnetic subbands in the notation of Ref. 8.

b) Using the $S(E)$ dependence known from Ref. 3, the measured value of S was used to determine E, which turned out to depend on the doping impurity (In, Ga, and B, respectively) in the following manner (we show in comparison the known[6,7] values of $\hbar\omega_0-|E^{(i)}|$):

E, meV	26.4	26.9	27.3
$\hbar\omega_0-\lvert E^{(0)}\rvert$, meV:	26.1	26.7	27.2 .

We estimate the accuracy of E at 1%. The differences between the values of E for the different impurities is approximately equal to the difference of their ionization energies $E^{(0)}$,[6] while the absolute values of E agree, with the same accuracy 1%, with relation (3), where $E^{(i)}$ is taken to be the ground-state energy $E^{(0)}$. For the energy of the optical phonon we use the value $\hbar\omega_0=37.8$ meV from Ref. 7. For gallium-doped samples this procedure was employed for three orientations of $\mathbf{H}$, [100], [110], and [111], and E is the average of the three obtained values. For boron- and indium-doped samples, the measurements were made only at $\mathbf{H}$ [100].

The separated series of resonance lines is thus due to resonant capture of nonequilibrium light holes in the ground state of the acceptor, with emission of an optical phonon. The numbers above the arrows in Fig. 1 are the indices of the light-hole Landau levels in the Luttinger classification,[8] the resonant capture from which the corresponding lines are attributed.

c) Our next task is to show that the remaining lines in the experimentally observed spectrum are due to transitions to excited acceptor levels. We gathered for this purpose on a single plot, in addition to the data on the passage of the Landau levels through the energy value $E\approx27$ meV, also the data of Ref. 3 on their passage through fixed energy values in the 40–140 meV interval (see Fig. 2). For each value of the energy E on the abscissa axis, the ordinate is the value of E_n in the dimensionless units $\alpha_n=E_n/\hbar\omega_0$, where $\Omega_0=eH/m_0c$. The dark circles on the ordinate exits itself at the Luttinger values of α_n calculated without allowance for nonparabolicity on the basis of the parameters known from experiments on cyclotron resonance.[9]

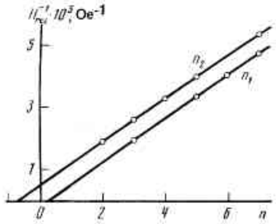

FIG. 3. Plot of H_{res}^{-1} vs. the quasiclassical number n for transitions to the third excited acceptor level from two light-hole ladders; **H**∥[111].

We shall return below to a discussion of the physical meaning of this plot and of its physical consequences. In the present section we use this plot only as an aid in the interpretation of the spectrum. Taking the energies of the two lowest excited acceptor levels $E^{(1)}$ and $E^{(3)}$, and drawing vertical lines through the points $\hbar\omega_0 = |E^{(1)}| = 33.45$ meV and $\hbar\omega_0 - |E^{(2)}| = 34.75$ meV on the abscissa axis, we have determined the sets of the resonant values of the magnetic field. They are marked by the arrows under the experimental curve in Fig. 1. Practically all the arrows corresponded to lines present in the spectrum, and the discrepancies the experimental and calculated resonance positions nowhere exceeded the line widths, i.e., 1–2% of a period.

d) It is easy to verify that all the remaining lines belong to a series of resonances corresponding to transitions to the third excited level. There are also traces of transitions to the forth level.

At field directions **H**∥[110] and **H**∥[111] the splitting due to the presence of two "ladders" of light holes is much smaller. This has simplified the spectrum and made it possible to interpret it directly, by successively separating from it line-pair series with periods P_i corresponding to transitions to different acceptor levels. Figure 3 shows by way of example a plot of H_{res}^{-1} against the Luttinger number for one of such series—for transitions to the third excited level, $n_{1,2} \to 3$. The plot shows only those lines that are not superimposed on lines of other series. The value of En determined from the slope of the lines is 35.5 meV as against[6,7] $\hbar\omega_0 - |E^{(3)}|$ = 35.2 meV (see Table I, where the values of E obtained from different resonant series for the gallium impurity from the plots with **H**∥[111] are compared with the known values of $\hbar\omega_0 - E^{(i)}$).

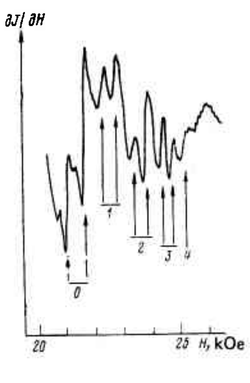

FIG. 4. Section of 2J/2H (H) spectrum at H∥[111]. The lower arrows mark the acceptor levels to which the transitions take place (to the ground state - from the magnetic subbands 5_2 and 6_1, to the excited ones from the subbands 6_2 and 7_1).

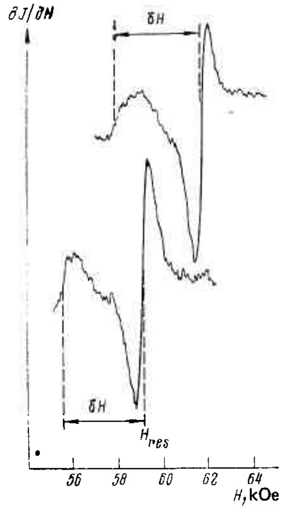

FIG. 5. Lines of $2_1 \to 0$ resonant transition at field directions H∥[100] and H∥[110] for the upper and lower curves, respectively.

Figure 4 shows a section of the spectrum where lines from different series do not overlap. Transitions from the ground level to four excited levels are clearly seen (the transition to the highest level is shown in the form of a weak unresolved line).

The entire observed resonant structure is due thus to transitions from the light-hole levels. The same can be stated even in the strong-field region: wherever there are no transitions from the light-hole levels there are

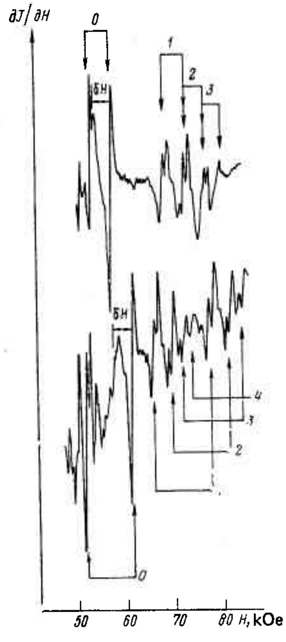

FIG. 6. Resonance spectra in strong fields at field directions H∥[111] and H∥[100] for the upper and lower curves, respectively. The acceptor levels to which the transitions from the magnetic subbands 2_2 and 3_1 take place are indicated.

no resonant signals whatever (for examples, the field intervals 60–66 kOe and 80–85 kOe at **H** ‖[100] (see Fig. 1).

To conclude this section, we consider the shapes of the resonance lines. Figure 5 show in enlarged scale, plotted in the coordinates $\partial J/\partial H$ and H, two relatively isolated lines corresponding to $2_2 \to 0$ transitions. Their characteristic shape (a minimum on the side of the weak fields and a maximum on the strong side) means that the photocurrent has a minimum at resonance. It appears that the additional slowly varying maximum on the weak fields in both plots is not accidental. A similar maximum exists on the left of the $2_2 \to 0$ line also at **H**‖[111] (see Fig. 6); traces of such a maximum can be discerned also near other lines. We shall return to the possible nature of this maximum in the next section, in the discussion of the influence of the Coulomb field.

In strong fields, the spectrum becomes more complicated and the lines acquire a fine structure (Fig. 6). It is due to Zeeman splitting of the acceptor levels. If the lines from the different series do not overlap, then this splitting can be measured. For example, from the splitting of the $3_1 \to 1$ line at H ‖[111] (Fig. 6) it follows that the splitting of the level 1 is of the order of 0.5 meV, in sufficiently good agreement with the data of Ref. 10. It should be noted in this connection that at all three field directions the $2_2 \to 0$ line has no fine structure (Figs. 5 and 6). This confirms the previously made[11] statement that the ground level of a shallow acceptor in germanium has an unusually small Zeeman splitting.

4. DISCUSSION

1. The foregoing identification of the resonant structure does not make it possible to obtain directly additional information on the nonparabolicity of the spectrum of the light holes. As seen from Figs. 1 and 3, the arrangement of the resonant lines in the reciprocal-field scale is equidistant with high accuracy, up to the transitions to the levels 2_2 and 3_1. This means that the obtained spectra can be used to determine the quasiclassical characteristics of the spectrum $S(E)$ and $g(E)$. The measured $S(E)$ agrees within 1% with the calculation by the previously obtained[3] interpolation formulas, and hence also with the $m(E)$ dependences obtained by us. The latter are needed, in particular, for the calculation of the functions $g(E)$ obtained from the directly measured $g^*(E)$ on the basis of formula (4) in terms of the ratio of the splitting in the H^{-1} scale to the period P.

The measured $g(E)$ are shown in Fig. 7 together with the known data pertaining to the top of the valence band (dark circles),[9] and with part of the data from Ref. 3 (triangles). The values from Ref. 3 in the region at large E at **H**‖[100] are not shown on the plots. The entire aggregate of the experimental data for this field direction can be described by the interpolation formula

$$g(E)=27.9-7.2\cdot 10^{-2}E+3.5\cdot 10^{-4}E^2 \quad (\mathbf{H}\|[100],\ 0<E<150\,\mathrm{meV}).$$

Figure 7 shows the E-dependences not only of the effective g, but also of the reduced g^*. This is done not only because it is g^* which is measured in the experiment.

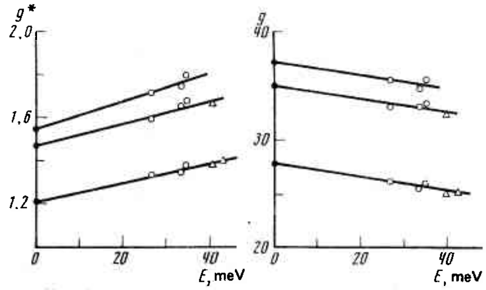

FIG. 7. Plots of $g(E)$ and $g^*(E)$ at magnetic field directions H‖[111], H‖[110], and H‖[100] for the upper, middle, and lower lines, respectively. Dark circles—from Refs. 8 and 9, white—present work, triangles—from Ref. 3.

The $g^*(E)$ plots demonstrate also the degree of proximity of g^* to two. This proximity is no accident[12]: in all the known cases, anomalously small cyclotron masses are accompanied by large g. In our experiments, the increase of E at all the directions of H brings g^* closer to two. It is not clear, however, whether this tendency is accidental or whether the system tends asymptotically with increasing E to the total compensation condition $gm=2m_0$.

2. We return now to the plots of Fig. 2. The straight lines passing through the experimental points show how the resonance lines from which the recombination proceeds move towards higher energies with increasing H. In the case of a parabolic spectrum this motion should be described by straight lines $\alpha_n=$const parallel to the abscissa axis. The appearance of an inclination $\alpha_n=\alpha_n^{(0)}-b_nE$ is the result of nonparabolicity of the spectrum. In the quasiclassical approach, for example,

$$\alpha_n=\left(n+\gamma\pm\frac{1}{4}g^*\right)\frac{m_0}{m},\quad b_n=\alpha_n\frac{m'}{m}. \tag{6}$$

The prime denotes here differentiation with respect to energy. It seems natural for extrapolation of the experimental plots to $E=0$ to lead to a system of Luttinger levels based on the data on cyclotron resonance in germanium.[9] In other words, the straight lines should seemingly pass through the black circles on the ordinate axis. Yet the difference $\delta\alpha_n$, which amounts to approximately one-third of a cyclotron period, undoubtedly greatly exceeds the experimental errors. The very large change in the hole-spectrum parameters in the energy region $E<25$ meV seems improbable. The only reasonable explanation is therefore that the experiment reveals the influence of the Coulomb field of an ionized center on the spectrum of the free carriers in its vicinity, and that the capture by the center is not from the bottom of the magnetic subband, but from a level split away from this bottom.

An analytic solution of the problem of the Coulomb spectrum in a magnetic field[4] exists only for a simple band within the strong-field limit

$$\hbar\Omega\gg|E^{(0)}|, \tag{7}$$

when the electric field can be regarded as a small correction compared with the magnetic one, and the ordinary system of excited states of the center no longer exists. It can be formally assumed that the condition

(7) is satisfied in our experiments, if Ω is taken to mean the cyclotron frequency of the light holes. The electronic structure of the acceptor, however, is formed with participation of heavy holes. Therefore, condition (7) notwithstanding, the field H for the acceptors is still weak in the sense that the Zeeman level splittings $\Delta E^{(i)} \ll |E^{(0)}|$. The problem was solved in this formulation only as applied to a diamagnetic exciton.[13,14]

Unfortunately, we have at present no reliable algorithm for the reduction of the experimental data, capable of separating in Fig. 1 the contribution of the nonparabolicity from the Coulomb correction, and of determining the distance δE_n from the split-off level to the true bottom of the magnetic subband. As a rough estimate we can draw through the dark circles of Fig. 2 straight lines parallel to the experimental lines, and obtain the value of δE_n as the difference between the abscissas of the intersections of the obtained pair of lines with the line $\alpha = E/\hbar\Omega_0$:

$$\delta E_n = \delta\alpha_n \hbar\Omega_0 (1 + b_n \hbar\Omega_0)^{-1}.$$

Such an estimate yields, for example for the level 2_2 in a field 60 kOe, a value of δE on the order of 2 meV. If now, starting from the field in which the resonance $2_2 \to 0$ is observed, we calculate the field in which the true bottom of the magnetic subband 2_2 will pass through the level $E = \hbar\omega_0 - |E^{(0)}|$, then we land precisely in the field region where the broad maximum is observed (see Figs 5 and 6). This gives grounds for assuming that this maximum is due to the passage of the bottom of the magnetic subband, or of higher discrete levels split-off from this subband, passes through this maximum.[4] From the value of δH on Fig. 5 or 6 we then obtain, at all three directions of $\mathbf{H}$, one and the same value $\delta E = 1.65 \pm 0.1$ meV at $H \approx 60$ kOe.

Thus, the observed resonant transitions start out from levels split-off from magnetic subbands. It follows from the relative width of the resonances that the width of these levels is $\Delta E \approx 0.2-0.3$ meV. This corresponds to a lifetime $\tau \approx 2 \times 10^{-12}$ sec. We cannot tell as yet whether this time is determined by the resonant capture or whether this is the discrete-level lifetime due to the transition of the carrier to a higher or lower magnetic subband. It is appropriate to point out here that the described experiments showed no resonant capture from the heavy-hold band. Of course, owing to the differences of the effective masses, the transitions from the heavy-hole band should have produced at the same absolute width a structure that is almost ten times more frequent. It must furthermore be borne in mind that although the average state densities in the bands differ by a factor $(m_h/m_e)^{3/2} \approx 23$, the difference between the heights of the maxima of the state densities at the bottom of the magnetic subbands is much less, since it is proportional to $(m_h/m_e)^{1/2}$. The small distance between the resonances is in no way compensated by the high intensity. Nevertheless, this does not explain completely the absence of resonant capture of heavy holes. It is probable that in the investigated field interval the recombination proceeds actually mainly via the light-hole band precisely because this band contains split-off levels, whereas in there are no such levels as yet in the heavy-hole band.

3. It follows thus from the experiments described above that recombination from the interior of the band, with participation of an optical phonon, proceeds differently than the Lax recombination from the bottom of the band.[15] Capture from the bottom of the band takes place mainly from highly excited levels of the center with a subsequent transition to deeper levels, with emission of long-wave phonons. On the contrary, capture from the interior of the band in a strong magnetic field proceeds in two stages: first the carrier lands on the split-off level of the magnetic subband, and then already on one of the acceptor levels proper. The probability of resonant transitions to the ground state is here of the same order as to a highly excited state. This follows from the comparison of the amplitudes of the resonance lines on Figs. 1, 4, and 6. To be sure, this comparison can so far be regarded only as qualitative, since capture onto different acceptor levels is from different energy levels E, and the carrier distribution function $f(E)$ in the band has not been investigated. Allowance for the function $f(E)$, however, can hardly change this qualitative conclusion.

The approximate equality of the probabilities of recombination via different levels can be explained by means of an estimate that does not take the magnetic field strength into account, but reveals nonetheless the gist of the matter.

An increased of the characteristic dimensions a_i of the excited states compared with the size of the ground state should lead to an increase of the matrix element of the transition, accompanied by emission of an optical phonon. However, the wave vector of such a phonon should satisfy the inequality $q \lesssim a_i^{-1}$. Therefore the increase of the matrix element is offset by a decrease of the phase volume of the phonons that can participate in the transition. If the wave function of the electron in the band is assumed to be a plane wave $\exp(ikr)$ and the potential U of the interaction of the hole with the optical phonon is assumed constant, then the transition probability is

$$W \sim \int d^3q \left| \int \psi^* U e^{iqr} e^{ikr} d^3r \right|^2 \sim U \int |\psi^*_{q+k}| d^3q = U.$$

Here ψ is the wave function of the bound state and ψ_{q+k} is its Fourier component; if the initial function ψ is normalized, then the last integral is equal to unity regardless of the concrete form of the function ψ and of its effective size a_i.

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