Selection Rules for Cyclotron Resonance of Holes in Germanium and Silicon

HIROSHI KAMIMURA*
Bell Telephone Laboratories, Murray Hill, New Jersey
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The general selection rules for cyclotron resonance both in transverse and longitudinal geometries are derived for the magnetic field $\mathbf{H}_0$ along the [001], [111], and [110] directions in order to analyze experimental quantum effect spectra. The transverse transitions may occur if the final state differs from the initial one by $l\nu\pm1$ where $l=0, \pm1, \pm2, \ldots$ and ν is 4 for $\mathbf{H}_0\|[001]$, 3 for $\mathbf{H}_0\|[111]$, and 2 for $\mathbf{H}_0\|[110]$. On the other hand, the longitudinal transitions may occur if the final state differs from the initial one by $l\nu$. It is concluded that the longitudinal transitions are all harmonics resulting from the warped energy surface of the valence band, in agreement with the classical result. Further, it is shown that the k_H effect plays essential roles in causing transverse harmonics for $H_0\|[111]$ and longitudinal ones for all three directions of the magnetic field.

1. INTRODUCTION

SINCE quantum effects in the cyclotron resonance of holes were predicted by Luttinger and Kohn[1] and Luttinger[2] and then their prediction was experimentally substantiated by Fletcher, Yager, and Merritt,[3] the theory of quantum effects has been subsequently expanded.[4–7] Recently, detailed experiments of quantum effects in Ge have been performed by Hensel[8] at 1.3 and 4.2°K in the frequency range 50~60 kMc/sec.[9] Measurements have been made for the magnetic field $\mathbf{H}_0$ along the [001], [111], and [110] directions with linearly polarized microwave electric field $\mathbf{E}(t)$ in both the transverse $\mathbf{E}\perp\mathbf{H}_0$ and longitudinal $\mathbf{E}\|\mathbf{H}_0$ geometries. In his measurements, he has found out some new features of quantum lines. In particular, many absorption lines in the longitudinal mode have been observed, all of which correspond to harmonic transitions resulting from the warped energy surface of the valence band. The purpose of the present paper is to derive the general selection rules for these quantum transitions both in transverse and longitudinal geometries in order to analyze the experimental results.

2. THEORETICAL BACKGROUND

As was mentioned in Sec. 1, the quantum theory of cyclotron resonance in degenerate bands has been developed by Luttinger.[2] The valence band maxima in Ge and Si occur at the center of the Brillouin zone. The original sixfold degenerate band edge breaks up into a fourfold and a twofold degenerate one by the spin-orbit interaction. As the spin-orbit splitting in Ge between the higher fourfold and the lower twofold level is large enough, we shall deal with only the fourfold degenerate band. According to Luttinger, the effective mass Hamiltonian D for holes in the fourfold degenerate band in the presence of a constant external magnetic field $\mathbf{H}_0$ can be written as

$$\begin{aligned}D=&-(1/m)[(\gamma_1+\tfrac{5}{2}\gamma_2)(k^2/2)-\gamma_2(k_x^2J_x^2+k_y^2J_y^2+k_z^2J_z^2)\\&-2\gamma_3(\{k_xk_y\}\{J_xJ_y\}+\{k_yk_z\}\{J_yJ_z\}\\&+\{k_zk_x\}\{J_zJ_x\})+(e/c)\kappa\mathbf{J}\cdot\mathbf{H}_0\\&+(eq/c)(J_x^3H_{0x}+J_y^3H_{0y}+J_z^3H_{0z})],\end{aligned}\quad(1)$$

where γ_1, γ_2, and γ_3 are the effective mass parameters, κ is the g value of a hole, q is the parameter approaching zero as the spin-orbit coupling does. (The value of q has been estimated to be 0.01 by Kohn[10]; and, hence, the q term will be omitted in the following treatment.) And further $k_\alpha=p_\alpha+(e/c)A_\alpha$ $(\alpha=x, y, z)$, $\{k_\alpha k_\beta\}=\frac{1}{2}(k_\alpha k_\beta+k_\beta k_\alpha)$, and J_x, J_y, and J_z are 4×4 matrices for the case of $J=\frac{3}{2}$, satisfying commutation rules for angular momentum.

Solutions to the general effective mass equation

$$D\Phi=\epsilon\Phi\quad(2)$$

have the form

$$\Phi=\begin{Vmatrix}\sum_i a_iu_i\\\sum_j b_ju_j\\\sum_k c_ku_k\\\sum_l d_lu_l\end{Vmatrix},\quad(3)$$

where u_n is the nth harmonic oscillator wave function. Since the substitution of (3) into Eq. (2) yields an infinite number of coupled algebraic equations, Eq. (2) is not solvable, except for special cases given by Luttinger.

Recently, Evtuhov[6] has given the matrix form of D for the three directions of the magnetic field, namely, [001], [111], and [110] directions. From his matrices we can see that the Hamiltonian D is divided into two parts, $D=D_0+D_1$, where D_1 is the part containing only the terms of $\gamma_3-\gamma_2$ and D_0 is the remaining one, which

* On leave from Department of Physics, University of Tokyo, Tokyo, Japan.
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depends on k_H, the component of the momentum along the magnetic field. As was shown by Luttinger, D_0 can be exactly solved and its characteristic functions for a given n are given by

$$\Phi_n^{(0)}=\begin{Vmatrix} a(n,k_H)u_n \\ b(n,k_H)u_{n+2} \\ c(n,k_H)u_{n+1} \\ d(n,k_H)u_{n+3} \end{Vmatrix}, \tag{4}$$

where a, b, c, and d are numerical coefficients which depend parametrically on k_H. On the other hand, the off-diagonal matrix elements of D_1 which vanish by setting $\gamma_3=\gamma_2$ connect a state $\Phi_n^{(0)}$ with states $\Phi_{n\pm4}^{(0)}$ for $\mathbf{H}_0\|[001]$, with states $\Phi_{n\pm3}^{(0)}$ for $\mathbf{H}_0\|[111]$, and with states $\Phi_{n\pm2}^{(0)}$ and $\Phi_{n\pm4}^{(0)}$ for $\mathbf{H}_0\|[110]$. This corresponds to the fact that the energy surface of the valence band has fourfold symmetry about the [001] axis, threefold symmetry about the [111] axis, and twofold symmetry about the [110] axis in the Brillouin zone. Thus, in the specialized cases of the magnetic field along the [001], [111], and [110] directions the solution (3) has the following form:

$$\Phi_n=\begin{Vmatrix} \sum_i a_{i\nu}(n,k_H)u_{n+i\nu} \\ \sum_i b_{i\nu}(n,k_H)u_{n+2+i\nu} \\ \sum_i c_{i\nu}(n,k_H)u_{n+1+i\nu} \\ \sum_i d_{i\nu}(n,k_H)u_{n+3+i\nu} \end{Vmatrix}, \tag{5}$$

where ν is 4 for $\mathbf{H}_0\|[001]$, 3 for $\mathbf{H}_0\|[111]$, and 2 for $\mathbf{H}_0\|[110]$.

We should like to remark that for $\mathbf{H}_0\|[111]$ we have another type of solution given by the following form, instead of (5):

$$\Phi_n=\begin{Vmatrix} \sum_i a_i(n,k_H)u_{n+3i} \\ \sum_i b_i(n,k_H)u_{n+2+3i} \\ \sum_i c_i(n,k_H)u_{n-2+3i} \\ \sum_i d_i(n,k_H)u_{n+3i} \end{Vmatrix}. \tag{6}$$

As was shown by Luttinger, if we put $k_H=0$ in Eq. (2) with the magnetic field along the [111] direction, we can solve the effective mass equation exactly and its characteristic function has a form

$$\Phi_n^{(0)}=\begin{Vmatrix} a(n)u_n \\ b(n)u_{n+2} \\ c(n)u_{n-2} \\ d(n)u_n \end{Vmatrix}, \tag{7}$$

where numerical constants a, b, c, and d are all independent of k_H. And then if we take the part depending on k_H into account, we can obtain the solutions of the form (6). Therefore, for the case of small k_H and large $(\gamma_3-\gamma_2)$ we should use the functions (6), instead of (5). It should be also remarked that, in either case, ν is 3, because ν is determined from the symmetry of the energy surface.

3. THE DERIVATION OF THE SELECTION RULES

Now we shall choose a coordinate system x_1, x_2, x_3 such that the magnetic field lies along x_3 and we shall also select for the vector potential $\mathbf{A}$ the Landau gauge; $A_1=-H_0x_2$, $A_2=A_3=0$. It is well known that in this case the Hamiltonian for a single particle in the presence of the magnetic field is periodic in x_1 and x_3 but is not periodic in x_2. Thus, the wave function of the hole in the degenerate band under the external constant magnetic field can be written in the following form:

$$\psi^{(\rho)}(n,k_1,k_3)=e^{i(k_1x_1+k_3x_3)}f_{k_1k_3}{}^{(\rho)}(n), \tag{8}$$

where the extra index ρ specifies one of four ladders of Landau energy levels. By using (5), $f_{k_1k_3}{}^{(\rho)}(n)$ is expressed by

$$\begin{aligned} f_{k_1k_3}{}^{(\rho)}(n)=&(\textstyle\sum_i a_{i\nu}(n,k_1,k_3)u_{n+i\nu})\varphi_{3/2} \\ &+(\textstyle\sum_i b_{i\nu}(n,k_1,k_3)u_{n+2+i\nu})\varphi_{-1/2} \\ &+(\textstyle\sum_i c_{i\nu}(n,k_1,k_3)u_{n+1+i\nu})\varphi_{1/2} \\ &+(\textstyle\sum_i d_{i\nu}(n,k_1,k_3)u_{n+3+i\nu})\varphi_{-3/2}. \end{aligned} \tag{9}$$

Here the set of the wave functions $(\varphi_{3/2},\varphi_{-1/2},\varphi_{1/2},\varphi_{-3/2})$ corresponds to the four independent Bloch wave functions of the $J=\frac{3}{2}$ multiplet at the top of the valence band in the absence of the magnetic field.

The selection rules for cyclotron resonance are essentially determined by the matrix element of the electric dipole moment, $\langle\psi^{(\sigma)}(m,k_1',k_3')|x_\alpha|\psi^{(\rho)}(n,k_1,k_3)\rangle$, which appears in the expression of the absorption coefficient.[4] Here α indicates the direction of the applied microwave electric field.

A. Selection Rules for Transverse Cyclotron Resonance

Suppose the electric field is taken along the x_2 axis. Then the matrix element $\langle\psi^{(\rho)}(m,k_1',k_3')|x_2|\psi^{(\sigma)}(n,k_1,k_3)\rangle$ is given as follows, by using (8) and (9),

$$\begin{aligned} &\langle\psi^{(\rho)}(m,k_1',k_3')|x_2|\psi^{(\sigma)}(n,k_1,k_3)\rangle=\delta(k_1-k_1')\delta(k_3-k_3')(\hbar/2s)^{1/2} \\ &\times[\textstyle\sum_{ij}\{[a_{j\nu}{}^{*(\rho)}(m,k_1',k_3')a_{i\nu}{}^{(\sigma)}(n,k_1,k_3)(n+i\nu)^{1/2}+b_{j\nu}{}^{*(\rho)}(m,k_1',k_3')b_{i\nu}{}^{(\sigma)}(n,k_1,k_3)(n+2+i\nu)^{1/2} \\ &+c_{j\nu}{}^{*(\rho)}(m,k_1',k_3')c_{i\nu}{}^{(\sigma)}(n,k_1,k_3)(n+1+i\nu)^{1/2}+d_{j\nu}{}^{*(\rho)}(m,k_1',k_3')d_{i\nu}{}^{(\sigma)}(n,k_1,k_3)(n+3+i\nu)^{1/2}]\delta_{m+j\nu,n-1+i\nu} \\ &+[a_{j\nu}{}^{*(\rho)}(m,k_1',k_3')a_{i\nu}{}^{(\sigma)}(n,k_1,k_3)(n+1+i\nu)^{1/2}+b_{j\nu}{}^{*(\rho)}(m,k_1',k_3')b_{i\nu}{}^{(\sigma)}(n,k_1,k_3)(n+3+i\nu)^{1/2} \\ &+c_{j\nu}{}^{*(\rho)}(m,k_1',k_3')c_{i\nu}{}^{(\sigma)}(n,k_1,k_3)(n+2+i\nu)^{1/2}+d_{j\nu}{}^{*(\rho)}(m,k_1',k_3')d_{i\nu}{}^{(\sigma)}(n,k_1,k_3)(n+4+i\nu)^{1/2}]\delta_{m+j\nu,n+1+i\nu}\}], \end{aligned} \tag{10}$$

where $s=eH_0/c$. In deriving (10), we have made use of the well-known properties,

$$\int u_n^* x_2 u_{n+1} dx_2 = [(\hbar/2s)(n+1)]^{1/2}, \tag{11a}$$

$$\int u_n^* x_2 u_{n-1} dx_2 = [(\hbar/2s)n]^{1/2}, \tag{11b}$$

for the harmonic oscillator wave functions.

From (10) we can see that transverse transitions may occur only if the final state, m, differs from the initial state, n, by $l\nu\pm1$, where l is positive and negative integers and zero. Transitions are possible between all four ladders for $k_3\neq0$. As is well known, the two δ functions in the coefficient mean that k_1 and k_3 are conserved during transitions. From (4) and (5) it is clear that if the energy surface is taken to be spherical, that is, if γ_2 and γ_3 are taken to be equal in Eqs. (1) and (2), $\nu=0$. In this case we can observe only the fundamental transitions resulting from the usual selection rule $\Delta n=\pm1$ in the transverse cyclotron resonance. The mixing of the harmonic oscillator wave functions in (5) is caused from the off-diagonal terms containing the factor $(\gamma_3-\gamma_2)$ for $\mathbf{H}_0\|[001]$ and $\mathbf{H}_0\|[110]$, while for $\mathbf{H}_0\|[111]$ the mixing arises from the interplay of the $k_H(=k_3)$ effect and the $\gamma_3-\gamma_2$ effect, as was shown in the previous section. Thus we may conclude that, though all harmonics in transverse cyclotron resonance arise from the warped energy shape of the valence band, besides this fact those for $H_0\|[111]$ will vanish unless $k_H\neq0$. This conclusion coincides with that given by Okazaki.[7]

B. Selection Rules for Longitudinal Cyclotron Resonance

In the case of the electric field parallel to the magnetic field, the matrix element of the electric dipole moment along the x_3 axis, is calculated in the following:

$$\begin{aligned}
&\langle\psi^{(\sigma)}(m,k_1',k_3')|x_3|\psi^{(\rho)}(n,k_1,k_3)\rangle \\
&=\int d\tau\, e^{i(k_1-k_1')x_1}e^{i(k_3-k_3')x_3}f_{k_1'k_3'}{}^{*(\sigma)}(m)f_{k_1k_3}{}^{(\rho)}(n)x_3 \\
&=\frac{\partial}{\partial k_3}\left[\int d\tau\, e^{i(k_1-k_1')x_1}e^{i(k_3-k_3')x_3}f_{k_1'k_3'}{}^{*(\sigma)}(m)f_{k_1k_3}{}^{(\rho)}(n)\right]-\int d\tau\, e^{i(k_1-k_1')x_1}e^{i(k_3-k_3')x_3}\left[f_{k'_1k_3'}{}^{*(\sigma)}(m)\frac{\partial}{\partial k_3}f_{k_1k_3}{}^{(\rho)}(n)\right] \\
&=\frac{\partial}{\partial k_3}\delta(k_1-k_1')\delta(k_3-k_3')\delta_{mn}-\delta(k_1-k_1')\delta(k_3-k_3')\Big[\sum_{ij}\Big\{a_{j\nu}{}^{*(\sigma)}(m,k_3')\frac{\partial}{\partial k_3}a_{i\nu}{}^{(\rho)}(n,k_3)+b_{j\nu}{}^{*(\sigma)}(m,k_3')\frac{\partial}{\partial k_3}b_{i\nu}{}^{(\rho)}(n,k_3) \\
&\qquad+c_{j\nu}{}^{*(\sigma)}(m,k_3')\frac{\partial}{\partial k_3}c_{i\nu}{}^{(\rho)}(n,k_3)+d_{j\nu}{}^{*(\sigma)}(m,k_3')\frac{\partial}{\partial k_3}d_{i\nu}{}^{(\rho)}(n,k_3)\Big\}\delta_{m+j\nu,n+i\nu}\Big].
\end{aligned} \tag{12}$$

In deriving (12), we have used the orthogonality relation

$$\langle f_{k_1k_3}{}^{(\sigma)}(m)|f_{k_1k_3}{}^{(\rho)}(n)\rangle=\delta_{mn}\delta_{\sigma\rho} \tag{13}$$

of the function $f_{k_1k_3}{}^{(\sigma)}(n)$ for the same $\mathbf{k}$. The first and the second terms in (12) represent the nonperiodic and the periodic parts of the electric dipole moment, respectively. The first term leads to transitions between states of the same energy and thus does not contribute to the absorption. Therefore, the selection rule for longitudinal cyclotron resonance can be derived from the second term in (12) and thus is given by $\Delta n=m-n=l\nu$, where $l=0,\ \pm1,\ \pm2,\ \cdots$. In this case transitions are also possible between all four ladders, and k_1 and k_3 are also conserved during transitions. We can see that, if we put $\nu=0$ in (12), the longitudinal transition does not occur. Thus, we may conclude that cyclotron resonance lines observed in the longitudinal geometry are all harmonics caused from the warped energy surface which depends explicitly on $\gamma_3-\gamma_2$. This coincides with the classical results given by Zeiger, Lax and Dexter.[11] Further, it should be mentioned that the $k_H(=k_3)$ effect is essentially important in causing longitudinal transitions on account of the factor $\partial f_{k_1k_3}(n)/\partial k_3$ in (12).

TABLE I. The selection rules for transverse and longitudinal cyclotron resonance of holes for the magnetic field along the [001], [111], and [110] directions.[a]

General rules	ν	Transverse ($\mathbf{E}\perp\mathbf{H}_0$) $\Delta n=\nu l^b\pm1$	Longitudinal ($\mathbf{E}\|\mathbf{H}_0$) $\Delta n=\nu l^b$
$\mathbf{H}_0\|[001]$	4	$4l^b\pm1$	$4l^b$
$\mathbf{H}_0\|[111]$	3	$3l^b\pm1$	$3l^b$
$\mathbf{H}_0\|[110]$	2	$2l^b\pm1$	$2l^b$

[a] Recently, Okazaki (reference 7) and Stickler, Zeiger, and Heller (reference 9) have given the selection rules for transverse and both for transverse and longitudinal resonance for $\mathbf{H}_0\|[111]$, respectively.
[b] $l=0,\ \pm1,\ \pm2,\ \cdots$.

4. CONCLUSION

We have derived the selection rules for the transverse and the longitudinal cyclotron resonance of holes in the

[11] H. J. Zeiger, B. Lax, and R. N. Dexter, Phys. Rev. **105**, 495 (1957).

degenerate band. The obtained results for the selection rules governing all transitions are summarized in Table I. All transitions will be possible between all four ladders of Landau energy levels for $k_H \neq 0$. These are the quantum-mechanical analogs of the classical result derived by Zeiger, Lax, and Dexter.[11]

We have seen that if the energy surface is taken to be spherical, that is, if γ_2 and γ_3 are taken to be equal, $\nu=0$. Thus we have concluded that cyclotron resonance lines observed in the longitudinal geometry are all harmonics caused from the warped energy surface which depend explicitly on $\gamma_3-\gamma_2$. Further, it has been remarked that the k_H effect plays an essential role in causing transverse harmonics for $\mathbf{H}_0\|[111]$ and longitudinal transitions for three directions of the magnetic field, namely, [001], [111], and [110] directions.

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Self-Diffusion in Silver-Gold Solid Solutions*

W. C. MALLARD,† A. B. GARDNER, RALPH F. BASS,‡, AND L. M. SLIFKIN
University of North Carolina, Chapel Hill, North Carolina
(Received 23 August 1962)

The diffusion of silver and gold tracers in silver-gold in crystals of 0, 8, 17, 35, 50, 66, 83, 94 and 100 at. % gold has been measured. It is shown that the limiting error in such measurements is due to temperature uncertainty rather than to the sectioning process. The activation energies obtained do not vary in proportion to the melting point or heat of fusion, and the deviations cannot be rectified in terms of lattice parameter arguments. The activation energies in the pure metals are better accounted for by the theory of Turnbull and Hoffman than by that of Swalin. The suggestion that the vacancy migration energy should vary as $(c_{11}-c_{12})$ is not confirmed. From the change in frequency factor with composition it is deduced that the activation entropy of migration of a vacancy decreases linearly with composition by $1.5R$ from pure silver to pure gold. The dependence of diffusion coefficient on gold content is compared with the theories of Hoffman, Turnbull, and Hart, of Reiss, of Manning, and of Lidiard, and impurity correlation factors of reasonable magnitudes are obtained. Particularly difficult to treat, however, is the decrease in diffusion coefficients resulting from additions of the rapid diffuser silver to the slow diffuser gold. An experiment to measure the effects of vacancy flux directly is proposed.

A LARGE part of present-day theory of atomic diffusion in crystals[1] is still sufficiently approximate and uncertain that the guidance and validation provided by simple and thorough experiments are welcome. In particular, much can be learned about interactions between vacancies and impurity atoms by the study of self-diffusion in binary alloys as a function of composition. There have been several such investigations in recent years, dealing with various alpha phase solutions in silver,[2] the gold-nickel system,[3] transition metal solutions,[4] and the lead-thallium system.[5] All of these alloys present various complexities—limited single-phase miscibility, thermodynamic or electronic complications, or difficulties in experimental procedure. The silver-gold system, on the other hand, appears much more amenable to theoretical interpretation. Both ions are monovalent, of essentially equal radii, and form a single face-centered cubic solid solution over the entire range of composition, with a phase diagram showing no major thermodynamic complexities.[6] From an experimental point of view, the near coincidence of the solidus and liquidus curves assures the ready production of single crystals of rather uni-

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† Present address: Emory University, Atlanta, Georgia.
‡ Present address: Oak Ridge National Laboratory, Oak Ridge, Tennessee.

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