Theory of Multiphoton Magnetoabsorption in Semiconductors

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We calculate direct interband transitions induced by a strong oscillating electric field, in the presence of a longitudinal or transverse magnetic field, including the largest, intraband effects of the electric field in the original wave functions. This is an extension of the Keldysh treatment to include the magnetic field, and also to include absorption of a second optical electric field, where we work in the effective-mass approximation, which is more suitable for the magnetic field case. The intraband effects, combined with an interband electric perturbation obtained from the effective-mass theory, induce Zener or photon-assisted (Franz-Keldysh) tunneling in parallel fields as the frequency of the strong electric field becomes small, and multiphoton transitions for relatively high frequencies and moderate electric fields. When a one-photon $\Delta n=0$ transition is allowed, the intraband effects induce multiphoton transitions with the same selection rule, except that in a transverse magnetic field $\mathbf{H}$, $\Delta n=\pm m$ transitions are allowed, but reduced by a factor proportional to H^m. The transition energies are those predicted earlier by Lax, where for a longitudinal magnetic field the electric-field energy shift is that obtained by Keldysh for zero magnetic field, and is modified for a transverse field by the cyclotron resonance frequencies.

I. INTRODUCTION

THE effect of a strong oscillating electric field on an intrinsic semiconductor depends on the frequency of the oscillation. For relatively high frequencies and moderate electric fields one observes interband transitions when the frequency is a submultiple of the interband transition frequency (multiphoton resonance). For low frequencies and strong electric fields Zener tunneling is observed, or photon-assisted tunneling when an additional optical field is absorbed. The tunneling case has been treated by Weiler *et al.*[1] including the effect of longitudinal and transverse magnetic fields, using the two-band model. In this paper we concentrate on the multiphoton limit, especially for crossed fields, since we work in the parabolic-band approximation and one of the principal results of the two-band calculation is that the parabolic approximation holds, for crossed fields, only for relatively low electric fields $[E/H<(\mathcal{E}_g/2m^*c^2)^{1/2}]$.

There have been two types of approach made to the calculation of multiphoton resonances. The first, worked out in detail by Braunstein *et al.*,[2] is to regard the oscillating electric field as a purely interband perturbation, with the transition rate and selection rules governed by products of interband matrix elements $\mathbf{p}_{ij}$ between the initial, final, and virtual intermediate states. In semiconductors with inversion symmetry, and in which a one-photon interband transition is allowed, this means that a two-photon interband transition is forbidden by parity. Then one of the interband matrix elements is proportional to $\mathbf{k}$, giving a transition $\sim(2\hbar\omega-\mathcal{E}_g)^{3/2}$ rather than $(2\hbar\omega-\mathcal{E}_g)^{1/2}$ as for an allowed transition. Zawadzki *et al.*[3] (ZHL) have shown that in this case a transverse magnetic field $\mathbf{H}$ induces resonant two-photon transitions between the Landau levels of the valence and conduction bands when the intermediate virtual state is another Landau level in either the valence or conduction band. The transition rate is proportional to H^2 and has the selection rule $\Delta n=\pm 1$. A tentative identification of such transitions, induced in PbTe by a CO_2 laser, was made by Button *et al.*[4] using a relatively crude photoconductivity technique. For a longitudinal magnetic field, ZHL obtain only nonresonant transitions.

The other approach to the oscillating electric field problem was made by Keldysh,[5] in the absence of a magnetic field. The electric field is regarded as a partly interband and partly intraband effect, with the largest, intraband effect taken into account by an exact solution of the time-dependent wave equation for the separate bands. Interband transitions are calculated to first order in the interband perturbation. Pantell *et al.*[6] have given an exact derivation of this formulation. This is a technique often used to calculate Zener tunneling in a dc electric field. The result depends on a parameter $\gamma=\omega\tau$ which relates the frequency ω of the electric oscillation to a semiclassical tunneling time $\tau=(2\mu\mathcal{E}_g)^{1/2}/eE$. Franz[7] shows how to calculate this tunneling time for a square potential barrier. For absorption of a weak optical beam of frequency ω', $\mathcal{E}_g$ in the parameter τ is replaced by $\mathcal{E}_g-\hbar\omega'$. When $\gamma\ll 1$, or the oscillation period is large compared to the tunneling time τ, the electrons have time during an oscillation to tunnel

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across the band gap. When $\gamma \gg 1$, transitions can take place only by direct absorption of one or more photons. In this multiphoton limit, Keldysh obtains multiphoton transitions with one-photon selection rules, where the higher-order transitions are caused by the time dependence of the states in the separate bands, introduced by the oscillating electric field. For the case where the one-photon transition is allowed, this gives a mechanism for two-photon transitions where the ordinary interband perturbation theory does not, although the transition rate is small compared to an allowed two-photon transition. This rate $\sim(2\hbar\omega - \mathcal{E}_g)^{1/2}$ near the edge, as for an "allowed" transition.

We have extended the Keldysh technique to the case of longitudinal and transverse magnetic fields, and also to include absorption of a second optical beam since most two-photon experiments have been done in this configuration. Although Keldysh worked with time-dependent Houston functions,[8] we have found it convenient to work in the Kohn-Luttinger effective mass representation[9] in order to include the magnetic field. In order to include both a magnetic field and an oscillating electric field we have found it necessary to work in the parabolic approximation since the diagonalization of the two-band Hamiltonian[1] does not commute with $i\hbar\partial/\partial t$. We work out this approximation in the Appendix, obtaining a time-dependent effective mass equation and an expression for the interband perturbation. We outline the Keldysh-type calculation in Sec. II, where we show that the effective-mass approximation does not seriously alter the results. For a longitudinal field the solution to the intraband problem (the effective-mass equation) is very similar to the Houston function used by Keldysh. In Sec. III we calculate both the multiphoton and tunneling limits along lines similar to his. For crossed fields when the electric and magnetic effects are not independent, the solution has been obtained in Sec. IV by the driven-harmonic-oscillator technique used by several authors,[10–13] where we have modified the results to neglect transient effects far from cyclotron resonance ($\omega \ll \omega_c$ or $\omega \gg \omega_c$), and to obtain solutions which reduce to those obtained by Aronov[14] in static crossed fields. In this case we have calculated only the multiphoton limit because of the limitation of the parabolic approximation in crossed fields. The multiphoton transition energies obtained in these calculations are those suggested by Lax.[15] In Sec. V we extend these calculations to include absorption of a second optical beam.

The results of this calculation give, for both orientations of magnetic field, resonant multiphoton transition rates proportional to H with the one-photon selection rules $\Delta n = 0$, and for a transverse field additional transitions $\Delta n = \pm m$ proportional to H^{m+1}. The anticipation of this result was the prime motivation for this work because Bierig and Weiler,[16] in a continuation of the work of Button *et al.*,[4] have observed multiphoton photoconductivity peaks in both PbTe and InSb for both orientations of magnetic field, with little dependence on the magnetic orientation. Of course, photoconductivity is not a very accurate measure of transition rates, but the spectra in InSb correlate quite well with one-photon results. Order-of-magnitude calculations for InSb indicate that the transitions we calculate, which seem to account for the results, are as strong or slightly stronger than the transitions proposed by ZHL for a transverse field only. A study of the PbTe results is in progress, using the results of one-photon experiments in progress at our laboratory, to determine whether the identification of $\Delta n = \pm 1$ transitions in a transverse field,[4] made using a simple two-band model, was premature in the light of the results for a longitudinal field, which cannot be accounted for by the theory of ZHL.

Patel *et al.*[17] in observing recombination radiation from PbTe produced by a CO_2 laser with no magnetic field, deduced multiphoton transitions and found that both the interband Braunstein mechanism and the intraband Keldysh mechanism could account for their results. Thus the selection rules observed in a magnetic field are the best means of distinguishing these two types of transitions.

The question naturally arises, why the perturbation treatment of ZHL, which converges since the intraband electric perturbation acts between bound states in a transverse magnetic field, gives different results from the multiphoton limit of our Keldysh- or tunneling-type approach. At this point we can only make several remarks to indicate a possible resolution of this question. Our exact solution in a transverse field can be obtained to various powers of the electric field $\mathbf{E}$ by carrying out the intraband perturbation treatment of ZHL, giving results similar to those of Aronov.[14] We obtain couplings from a given Landau level n to other levels $n \pm m$, proportional to E^m, and the lowest-order correction to the nth level proportional to E^2. This latter correction is responsible for the $\Delta n = 0$ transitions we calculate. A straightforward interband-intraband perturbation treatment does not consider this transition because there is no intermediate virtual state involved. The transitions obtained by ZHL involve only the couplings to the levels $n \pm 1$, with the level n as intermediate state. These transitions are similar to the $\Delta n = \pm 1$ transitions we obtain, which are weaker than our $\Delta n = 0$ transition. Thus the ZHL perturbation treatment, which does not

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take into account the effect of the electric field on the wave functions themselves, seems to be a less rigorous approach.

II. OUTLINE OF THE CALCULATION

In order to include a magnetic field in the treatment of the intraband effects of an oscillating electric field, we find it necessary to work in the effective-mass representation.[9] The equation of motion for an electron in a periodic potential $V(\mathbf{r})$, an electric potential $-e\mathbf{E}\cdot\mathbf{r}\cos\omega t$ and a magnetic field $\mathbf{H}\|\mathbf{z}$ is

$$\left[\frac{\mathbf{P}^2}{2m_0}+V(\mathbf{r})-e\mathbf{E}\cdot\mathbf{r}\cos\omega t\right]\Psi(\mathbf{r},t)=i\hbar\frac{\partial\Psi}{\partial t}(\mathbf{r},t), \quad (1)$$

where $\mathbf{P}=i\hbar\partial/\partial\mathbf{r}+(e/c)\mathbf{A}(\mathbf{r})$ and $\mathbf{A}(\mathbf{r})=(0,Hx,0)$. We show in the Appendix that we can write $\Psi(\mathbf{r},t)$ approximately as

$$\Psi(\mathbf{r},t)\approx\sum_j\psi_j(\mathbf{r},t)u_j(\mathbf{r}), \quad (2)$$

where $u_j(\mathbf{r})$ is the band-edge Bloch function, with energy $\mathcal{E}_{j0}$, and the envelope function $\psi_j(\mathbf{r},t)$ obeys the approximate equation

$$\mathcal{H}_j\psi_j(\mathbf{r},t)=\left[\mathcal{E}_{j0}+\frac{\mathbf{P}^2}{2m_j}-e\mathbf{E}\cdot\mathbf{r}\cos\omega t\right]\psi_j(\mathbf{r},t)$$
$$=i\hbar\frac{\partial\psi_j}{\partial t}(\mathbf{r},t). \quad (3)$$

The above equation results from a diagonalization procedure which leaves an interband perturbation due to the electric field [see Appendix Eq. (A1)],

$$\mathcal{H}_{jk}'=V_{jk}\cos\omega t=\left[\frac{ihe\mathbf{E}\cdot\mathbf{p}_{jk}}{m_0(\mathcal{E}_{j0}-\mathcal{E}_{k0})}\right]\cos\omega t \quad (4)$$

which we use to calculate interband transitions between the approximate states j and k. The full equation is

$$\mathcal{H}_j\psi_j+\sum_{k\neq j}\mathcal{H}_{jk}'\psi_k=i\hbar\frac{\partial}{\partial t}\psi_j. \quad (4')$$

In Eq. (4), $\mathbf{p}_{jk}$ is the matrix element of $\mathbf{p}$ between the band-edge functions u_j, u_k. Note that $\mathcal{H}_{jk}'$ is the usual optical perturbation if we replace $(\mathcal{E}_{j0}-\mathcal{E}_{k0})$ by $\hbar\omega$.

The difference between our approximation and the method used by Keldysh[5] lies in this result for the interband perturbation. In the Bloch-function representation which Keldysh uses, the interband matrix elements are taken between the $\mathbf{k}$-dependent Bloch functions $u_{j,\mathbf{k}}$, $u_{k,\mathbf{k}}$. For zero magnetic field, the solutions ψ_j involve time-dependent $\mathbf{k}$'s, and in the saddlepoint calculation of $A_l(\omega)$ (see below), V_{cv} has a pole at each saddle point such that the value of $A_l(\omega)$ at each point is $\frac{1}{3}\hbar\omega$ times the exponential factors. In our case, the calculation for zero magnetic field is exactly the same except that we regard V_{cv} as constant (taken between the band-edge functions). This affects the resulting transition rate only by multiplying it by a slightly different numerical factor [$\approx(\hbar\omega)^2\pi/2l$ for an l-photon transition] which affects only estimations of transition strengths, not the selection rules. In fact, it can be argued (see Appendix) that the $\mathbf{k}$ values used in the solutions are large enough (far enough out in the Brillouin zone) so that the $\mathbf{k}$ dependence of V_{cv} is no longer accurate. The fact that both methods give essentially the same result indicates that the result is not very sensitive to the type of approximation used. In other words, the important properties of the wave functions for this calculation are included in the envelope functions, not the band functions.

Zeldovich[18] has shown that with equations like Eq. (1) or Eq. (3), where the Hamiltonian is periodic in time, we can always obtain quasistationary solutions in a form analogous to Bloch functions in a periodic lattice:

$$\psi_j(\mathbf{r},t)=\exp(-i\tilde{\epsilon}_jt/\hbar)\tilde{\psi}_j(\mathbf{r},t), \quad (5)$$

where $\tilde{\psi}_j(\mathbf{r},t)$ is periodic as $t\to t+T$, $T=2\pi/\omega$. Zeldovich calls $\tilde{\epsilon}_j$ a quasienergy analogous to the crystal momentum in a periodic lattice, because as $t\to t+T$, $\psi_j(\mathbf{r},t)\to\exp(-i\tilde{\epsilon}_jT/\hbar)\psi_j(\mathbf{r},t)$. Zeldovich shows that this quasienergy is conserved, modulo $[\hbar\omega]$, in transitions within the periodic system.

Zeldovich notes that the quasienergy is defined only up to an arbitrary constant $m\hbar\omega$ since as $\tilde{\epsilon}_j\to\tilde{\epsilon}_j+m\hbar\omega$, $\tilde{\psi}_j\to\tilde{\psi}_j\exp(im\omega t)$ which is still periodic in T. However, we can define $\tilde{\epsilon}_j$ unambiguously by requiring that $\tilde{\psi}_j$ contain no exponential terms which are linear in t. Then the quasienergy corresponds roughly to a time-averaged energy in the periodic field: If one can write the exponential part (the only nonperiodic part) of ψ_j as

$$\psi_j\sim\exp\left[-i\int_0^t dt\,\mathcal{E}_j(t)/\hbar\right],$$

where $\mathcal{E}_j(t)$ is periodic in T, then

$$\tilde{\epsilon}_j=\frac{\omega}{2\pi}\int_{-\pi/\omega}^{\pi/\omega}dt\,\mathcal{E}_j(t).$$

We shall obtain solutions of the form (5) for the conduction and valence bands in both longitudinal and transverse magnetic fields and calculate interband transitions induced by the perturbation (4), using first-order time-dependent perturbation theory. We write the perturbed wave function as $\Psi=\psi_vu_v+a_{cv}\psi_cu_c$ and use Eq. (4a) for $\psi_c\to(a_{cv}\psi_c)$. The result for the coupling

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from the valence to conduction band is

$$a_{cv}(t)=\frac{1}{i\hbar}V_{cv}\int_0^t d\tau\,\cos\omega\tau\int d\mathbf{r}\,\psi_c^*(\mathbf{r},\tau)\psi_v(\mathbf{r},\tau)$$

$$=\frac{1}{i\hbar}V_{cv}\int_0^t d\tau\,\cos\omega\tau M_{cv}(\tau)\times\exp[i(\tilde{\epsilon}_c-\tilde{\epsilon}_v)\tau/\hbar],\quad(6a)$$

where

$$M_{cv}(\tau)=\int d\mathbf{r}\tilde{\psi}_c^*(\mathbf{r},\tau)\tilde{\psi}_v(\mathbf{r},\tau).\quad(6b)$$

The coupling a_{cv} resembles the usual result for an interband optical perturbation, in that it contains the Bloch-function coupling V_{cv} multiplied by the overlap of the envelope functions ψ_c,ψ_v.

Since $M_{cv}(\tau)$ is periodic in $2\pi/\omega$, we expand $M_{cv}(\tau)\cos\omega\tau$ in a Fourier series such that

$$A_l(\omega)=\frac{\omega}{2\pi}\int_{-\pi/\omega}^{\pi/\omega}dt\,\cos\omega t M_{cv}(t)e^{il\omega t}\quad(7)$$

or

$$\cos\omega\tau M_{cv}(\tau)=\sum_{l=-\infty}^{\infty}A_l(\omega)e^{-il\omega\tau}.$$

When this is substituted into (6a), the result for the total transition rate is

$$W_{cv}=\frac{2\pi}{\hbar}|V_{cv}|^2\sum_{\text{states }c,v}\sum_l|A_l(\omega)|^2\times\delta[l\hbar\omega-(\tilde{\epsilon}_c-\tilde{\epsilon}_v)].\quad(8)$$

Then using the δ function, we can rewrite the equation for $A_l(\omega)$ as

$$A_l(\omega)=\frac{\omega}{2\pi}\int_{-\pi/\omega}^{\pi/\omega}d\tau\,\cos\omega\tau M_{cv}(\tau)\exp[i(\tilde{\epsilon}_c-\tilde{\epsilon}_v)\tau/\hbar]$$

$$=\frac{\omega}{2\pi}\int_{-\pi/\omega}^{\pi/\omega}d\tau\,\cos\omega\tau\int d\mathbf{r}\psi_c^*(\mathbf{r},\tau)\psi_v(\mathbf{r},\tau).\quad(9)$$

The δ function in Eq. (8) gives the conservation of quasienergy mentioned above. If the one-photon matrix element V_{cv} is allowed, the result (8) gives a series of associated multiphoton transitions. It is the periodicity of the wave functions $\tilde{\psi}_c,\tilde{\psi}_v$ which introduces these higher-order transitions since if $M_{cv}(t)$ is constant, then A_l is zero unless $l=1$. In other words, the electronic motion, having the periodicity of the driving field, can absorb energy from that field.

The transition rate (8) represents a sum of rates for absorbing l photons $\hbar\omega$. If $\omega\to0$, we convert the sum over l to an integral, which yields an expression for interband tunneling.

In Secs. III and IV we solve Eq. (3) for the cases of a longitudinal and a transverse magnetic field, then compute the interband transition rates using Eqs. (8) and (9), converting the integral for $A_l(\omega)$ into a complex integral over $u=\sin\omega\tau$, and using the saddlepoint method. In Sec. V we make a simple extension of Eq. (8) to include the case of two-field absorption in both orientations of magnetic field.

III. LONGITUDINAL MAGNETIC FIELD

With the electric and magnetic fields along the z direction we write the solution to Eq. (3) as

$$\psi_j(\mathbf{r},t)=e^{ik_yy}\varphi_n(x+\lambda^2k_y)\psi_j(z,t),$$

where $\lambda=(\hbar c/eH)^{1/2}=(\hbar/m_j\omega_j)^{1/2}$ is the cyclotron orbit radius, $\omega_j=eH/m_jc$ is the cyclotron resonance frequency, and $\varphi_n(x)$ is a harmonic oscillator function. Then Eq. (3) becomes

$$\left[\mathcal{E}_{j0}+(n+\tfrac{1}{2})\hbar\omega_j+\frac{p_z^2}{2m_j}-eEz\cos\omega t\right]\psi_j(z,t)=i\hbar\frac{\partial\psi_j}{\partial t}(z,t).$$

The solutions to this equation are very similar to the Houston functions used by Keldysh,[5] except that, in Eq. (2), they multiply the band-edge Bloch function $u_j(\mathbf{r})$ rather than the k-dependent [and, through $k_z=k_z(t)$, time-dependent] periodic functions $u_{j\mathbf{k}}(\mathbf{r})$,

$$\psi_j(z,t)=\exp\left[i\left(k_z+\frac{eE}{\hbar\omega}\sin\omega t\right)z\right]\times\exp\left[-\frac{i}{\hbar}\int_0^t d\tau\mathcal{E}_j(\tau)\right],\quad(10a)$$

where

$$\mathcal{E}_j(t)=\mathcal{E}_{j0}+(n+\tfrac{1}{2})\hbar\omega_j+(\hbar^2/2m_j)[k_z+(eE/\hbar\omega)\sin\omega t]^2.\quad(10b)$$

We note that the solution (10) can be put in the form (5) with

$$\tilde{\epsilon}_{j,n}=\mathcal{E}_{j0}+(n+\tfrac{1}{2})\hbar\omega_j+(\hbar^2k_z^2/2m_j)+(e^2E^2/4m_j\omega^2).\quad(11)$$

We now proceed to calculate the interband transition rate using Eqs. (8) and (9). The space integral in Eq. (9) gives $\delta_{n,n'}$, $\delta_{k_y,k_y'}$, $\delta_{k_z,k_z'}$, and the energy δ function gives the transition energy, which was obtained by Lax[15] for $k_z\to0$,

$$l\hbar\omega=\tilde{\epsilon}_{c,n}-\tilde{\epsilon}_{v,n}\equiv\tilde{\epsilon}_n=\mathcal{E}_g+(n+\tfrac{1}{2})\hbar\omega_\mu+e^2E^2/4\mu\omega^2+\hbar^2k_z^2/2\mu,\quad(12)$$

where μ is the reduced mass, and $\omega_\mu\equiv\omega_c+\omega_v=eH/\mu c$. The Fourier component $A_l(\omega)$ becomes

$$A_l(\omega)=\frac{\omega}{2\pi}\int_{-\pi/\omega}^{\pi/\omega}d\tau\,\cos\omega\tau\times\exp\left\{\frac{i}{\hbar}\int_0^\tau d\tau'[\mathcal{E}_c(\tau')-\mathcal{E}_v(\tau')]\right\},\quad(13a)$$

where

$$\mathcal{E}_c(t)-\mathcal{E}_v(t)=\mathcal{E}_n[1+\zeta^2+(2\zeta/\gamma)\sin\omega t+(1/\gamma^2)\sin^2\omega t]$$

with

$$\mathcal{E}_n=\mathcal{E}_g+(n+\tfrac{1}{2})\hbar\omega_\mu,$$
$$\zeta^2=\hbar^2k_z^2/2\mu\mathcal{E}_n,\quad \gamma^2=2\mu\omega^2\mathcal{E}_n/e^2E^2. \quad (13b)$$

Notice that γ is the same parameter as that used by Keldysh, except that $\mathcal{E}_g$ is replaced by $\mathcal{E}_n$. Then

$$\tilde{\epsilon}_n=\mathcal{E}_n[1+\zeta^2+(1/2\gamma^2)].$$

We define $u=\sin\omega\tau$, $v=\sin\omega\tau'$, and find

$$A_l(\omega)=\frac{1}{2\pi}\oint du\exp\left\{\frac{i}{\hbar\omega}\int_0^u dv\frac{[\mathcal{E}_c(v)-\mathcal{E}_v(v)]}{(1-v^2)^{1/2}}\right\}$$
$$=\frac{1}{2\pi}\oint du\exp[E(u)]$$

which we evaluate at the saddlepoint of the exponent, where $E'(u)=0$ or $\mathcal{E}_c(u)-\mathcal{E}_v(u)=0$, which gives

$$u_\pm=\pm i\gamma-\gamma\zeta$$

and

$$E''(u_\pm)\approx\mp 2\mathcal{E}_n/\hbar\omega\gamma(1+\gamma^2)^{1/2}.$$

Then

$$A_l(\omega)\approx\frac{1}{2\pi}\left\{\left[\frac{2\pi}{|E''(u_+)|}\right]^{1/2}\times\exp[E(u_+)]+\left[\frac{2\pi}{|E''(u_-)|}\right]^{1/2}\exp[-E(u_-)]\right\}$$

or, to order ζ^2,

$$A_l(\omega)\simeq\left[\frac{\hbar\omega\gamma(1+\gamma^2)^{1/2}}{\pi\mathcal{E}_n}\right]^{1/2}\exp\left[-l\sinh^{-1}\gamma+\frac{\mathcal{E}_n}{2\hbar\omega}\frac{(1+\gamma^2)^{1/2}}{\gamma}+\frac{\mathcal{E}_n\gamma\zeta^2}{\hbar\omega(1+\gamma^2)^{1/2}}\right]\cos f(\gamma)\zeta. \quad (14)$$

This is substituted into Eq. (8) for the transition rate. The sum over states becomes a sum over n and an integral over k_y, k_z or k_y, ζ. The rapidly oscillating factor $\cos^2 f(\gamma)\zeta$ contributes a factor $\frac{1}{2}$ to the ζ integral. We assume, for $|V_{cv}|^2$, that the one-photon matrix element $\mathbf{p}_{cv}$ is allowed, and, from the two-band model, $|\mathbf{p}_{cv}/m_0|^2\approx\mathcal{E}_g/2m^*\approx\mathcal{E}_g/4\mu$. The result for the transition rate, using the δ function for ζ^2, is

$$W=\frac{\hbar\omega^3}{8\pi^2\mathcal{E}_g}\left(\frac{eH}{\hbar c}\right)(2\mu)^{1/2}\frac{(1+\gamma^2)^{1/2}}{\gamma}\times\sum_n\sum_l\left[l\hbar\omega-\mathcal{E}_n\left(1+\frac{1}{2\gamma^2}\right)\right]^{-1/2}\times\exp\left\{-2l\left[\sinh^{-1}\gamma-\frac{\gamma}{(1+\gamma^2)^{1/2}}\right]-\frac{\mathcal{E}_n\gamma}{\hbar\omega(1+\gamma^2)^{1/2}}\right\}.$$

In the tunneling limit as $\omega\to 0$ or $\gamma^2\ll 1$, the sum over l is converted to an integral, which results in a tunneling current

$$j\approx\exp\left[-\frac{4}{3}\frac{(2\mu)^{1/2}\mathcal{E}_n^{3/2}}{\hbar eE}\right] \quad (15)$$

which is the same as the result of standard calculations using parabolic bands.[19] The exponential is multiplied by a prefactor which is not correct, as explained by Keldysh,[5] because our result is the limit as $\omega\to 0$ of an average tunneling current over a large time compared to a period $2\pi/\omega$.

The multiphoton limit, $\gamma^2\gg 1$, gives a sum of multiphoton resonances given by

$$W_n^{(l)}\approx\frac{\omega^2}{8l\pi^2}\left(\frac{eH}{\hbar c}\right)(2\mu)^{1/2}\left(\frac{e^2E^2}{8\mu\omega^2\mathcal{E}_n}\right)^l\times\left[l\hbar\omega-\left(\mathcal{E}_n+\frac{e^2E^2}{4\mu\omega^2}\right)\right]^{-1/2}\exp\left(2l-\frac{\mathcal{E}_n}{\hbar\omega}\right) \quad (16)$$

since $\sinh^{-1}\gamma\approx\ln(2\gamma)$, for $\gamma^2\gg 1$.

This result is similar to the standard effect of a magnetic field on a one-photon band edge: The edge absorption $\sim(l\hbar\omega-\mathcal{E}_g)^{1/2}$ for no magnetic field becomes a series of resonant absorptions $\sim(eH/\hbar c)(l\hbar\omega-\mathcal{E}_n)^{-1/2}$. Our expression differs from the perturbation theory result of ZHL for $\mathbf{E}\|\mathbf{H}$, which predicts a transition matrix element $\sim k_z$, hence a "forbidden" transition $\sim(l\hbar\omega-\mathcal{E}_n)^{1/2}$, which is nonresonant. The recent photoconductivity results referred to in the Introduction, which exhibit multiphoton peaks for $\mathbf{E}\|\mathbf{H}$, indicate that the Keldysh mechanism prevails.

IV. TRANSVERSE MAGNETIC FIELD

With the electric field along the x direction, the magnetic field along the z direction, we write

$$\psi_j(\mathbf{r},t)=e^{ik_yy}e^{ik_zz}\psi_j(x,t)$$

which gives, from Eq. (3),

$$\left[\mathcal{E}_{j0}+\frac{\hbar^2k_z^2}{2m_j}+\frac{p_x^2}{2m_j}+\tfrac{1}{2}m_j\omega_j^2(x+\lambda^2k_y)^2-eEx\cos\omega t\right]\psi_j(x,t)=i\hbar(\partial\psi_j/\partial t)(x,t).$$

We define raising and lowering operators

$$\eta=\frac{1}{\sqrt{2}}\left(\frac{x'}{\lambda}+i\lambda k_x\right),\quad \eta^\dagger=\frac{1}{\sqrt{2}}\left(\frac{x'}{\lambda}-i\lambda k_x\right),\quad [\eta,\eta^\dagger]=1,$$

where $x'\equiv x+\lambda^2k_y$ and $k_x\equiv i\partial/\partial x$. Defining $\theta=eE\lambda/\hbar$, where $\hbar\theta=eE\lambda$ is the electric energy an electron gains

[19] R. R. Haering and E. N. Adams, J. Phys. Chem. Solids 19, 8 (1961).

over one cyclotron orbit radius, we obtain

$$\left[\mathcal{E}_{j0}+\frac{\hbar^2k_z^2}{2m_j}+\hbar\omega_j(\eta^\dagger\eta+\tfrac{1}{2})\right.$$
$$\left.-\frac{\hbar\theta}{\sqrt{2}}\cos\omega t(\eta+\eta^\dagger)+eE\lambda^2k_y\cos\omega t\right]\psi_j(x,t)$$
$$=i\hbar(\partial\psi_j/\partial t). \quad (17)$$

We solve this equation using a trial solution similar to that of Budd[10]

$$\psi_j(x,t)=e^{\gamma_j(t)}e^{A_j(t)\eta^\dagger}e^{B_j(t)\eta}e^{-i\omega_jt(\eta^\dagger\eta+1/2)}\psi_{j0}(x). \quad (18)$$

Using the commutation relations for $\exp(A\eta^\dagger)$, $\exp(B\eta)$, and $\exp(C\eta^\dagger\eta)$,[20] we obtain the conditions

$$(\dot{A}_j+i\omega_jA_j)=(\dot{B}_j-i\omega_jB_j)=(i\theta/\sqrt{2})\cos\omega t. \quad (19)$$

We solve these equations for the particular solution only, $\sim\cos\omega t$, $\sin\omega t$, as we wish to express the wave function in the form (5), in terms periodic in ω only and with a single quasienergy $\tilde{\epsilon}_{j,n}$. This is the result of applying the electric field adiabatically,[10] rather than suddenly at $t=0$, and is equivalent to neglecting transient effects far from cyclotron resonance ($\omega\neq\omega_c$). We also assume $\psi_{j0}(x)=\varphi_n(x')$, a harmonic oscillator function. The result is a wave function which as $\omega\to 0$ reduces exactly to Aronov's solution[14] for static crossed fields.

The solutions to Eq. (19), then, are

$$A_j=(i\alpha_j-\beta_j)/\sqrt{2}, \quad B_j=(i\alpha_j+\beta_j)/\sqrt{2}, \quad (20a)$$

where

$$\alpha_j=\theta\omega\sin\omega t/(\omega^2-\omega_j^2), \quad \beta_j=\theta\omega_j\cos\omega t/(\omega^2-\omega_j^2). \quad (20b)$$

We note, along with Budd,[10] Hanamura *et al.*,[11] and Lax,[15] that α_j/λ and $\lambda\beta_j$ play the role of classical momentum and position, respectively, since the wave function can be rewritten

$$\psi_j(x,t)\sim e^{i\alpha_jx'/\lambda}e^{-i\lambda\beta_jk_x}\varphi_n(x')=e^{i\alpha_jx'/\lambda}\varphi_n(x'-\lambda\beta_j).$$

In fact, our solution is equivalent to Budd's, although we neglect his terms oscillating at ω_c. The expressions of Hanamura *et al.*[11] for α_j, β_j, contain terms oscillating at $(\omega\pm\omega_c)$; this is the result of writing the wave function in the form

$$\psi_j(x,t)\sim e^{-i\omega_jt(\eta^\dagger\eta+1/2)}e^{\gamma_j'(t)}e^{A_j'\eta^\dagger}e^{B_j'\eta}\psi_{j0}(x)$$

although Hanamura *et al.* incorrectly write the form (18). The result of commuting the $\exp(\eta^\dagger\eta)$ term to the left in (18) is only to introduce phase factors into the intraband matrix elements since they depend mainly on the quantity $\frac{1}{2}(\alpha_j^2+\beta_j^2)$ which is unaffected. However, Hanamura *et al.* make other errors so that their results are incorrect, as stated by Budd.

[20] W. Louisell, Ref. 12, pp. 98 ff.

We also obtain from Eq. (17) the condition for γ_j,

$$\dot{\gamma}_j=A_j(\dot{B}_j-i\omega_jB_j)-(ieE\lambda^2k_y/\hbar)\cos\omega t$$
$$-(i/\hbar)(\mathcal{E}_{j0}+\hbar^2k_z^2/2m_j).$$

Solving this using Eq. (20), the result for the normalized x part of the wave function is of the form (5), where

$$\tilde{\epsilon}_{j,n}=\mathcal{E}_{j0}+(n+\tfrac{1}{2})\hbar\omega_j+(\hbar^2k_z^2/2m_j)$$
$$+\hbar\theta^2\omega_j/4(\omega^2-\omega_j^2), \quad (21a)$$
$$\tilde{\psi}_{j,n}(x,t)=e^{\tilde{\gamma}_j(t)}e^{A_j(t)\eta^\dagger}e^{B_j(t)\eta}\varphi_n(x')$$

with

$$\tilde{\gamma}_j(t)=[-\theta^2/4(\omega^2-\omega_j^2)]$$
$$\times[\sin^2\omega t+i(\omega_j/\omega)\sin\omega t\cos\omega t+\omega_j^2/(\omega^2-\omega_j^2)]$$
$$-(ieE\lambda^2k_y/\hbar\omega)\sin\omega t. \quad (21b)$$

We use this result to calculate the interband transition rate using Eqs. (8) and (9). We note that the electric-field energy shift, which for a longitudinal magnetic field and also, from Keldysh, for no magnetic field, is $e^2E^2/4m_j\omega^2$, becomes for a transverse magnetic field from Eq. (21a), $e^2E^2/4m_j(\omega^2-\omega_j^2)$. The space integral in Eq. (9) gives $\delta_{k_yk_y'}\delta_{k_zk_z'}$ but not $\delta_{nn'}$. Then the energy δ function in Eq. (8) gives the transition energy obtained by Lax[15]:

$$l\hbar\omega=\tilde{\epsilon}_{c,n'}-\tilde{\epsilon}_{v,n}\equiv\tilde{\epsilon}_{n',n}$$
$$=\mathcal{E}_g+(n'+\tfrac{1}{2})\hbar\omega_c+(n+\tfrac{1}{2})\hbar\omega_v$$
$$+e^2E^2/4m_c(\omega^2-\omega_c^2)+e^2E^2/4m_v(\omega^2-\omega_v^2)$$
$$+\hbar^2k_z^2/2\mu, \quad (22)$$

where for $j=v$, we have used $m_j=-m_v$, $\omega_j=-\omega_v$.

To calculate $A_l(\omega)$ from Eq. (9) we need, from Eq. (7),

$$M_{cv}(t)=\int dx\tilde{\psi}_{c,n'}^*(x,t)\tilde{\psi}_{v,n}(x,t)$$
$$=\exp(\tilde{\gamma}_c^*+\tilde{\gamma}_v)\int dx\varphi_{n'}(x')$$
$$\times e^{B_c^*\eta^\dagger}e^{A_c^*\eta}e^{A_v\eta^\dagger}e^{B_v\eta}\varphi_n(x').$$

Since $A_c^*=-B_c$, we calculate

$$M_{cv}(t)=\exp[\tilde{\gamma}_c^*+\tilde{\gamma}_v-A_vB_c$$
$$+i(n-n')\tan^{-1}\alpha/\beta]I_{n'n}(t), \quad (23a)$$

where

$$I_{n'n}(t)=(-)^{n'-n}\left(\frac{n!}{n'!}\right)^{1/2}(z)^{(n'-n)/2}L_n^{n'-n}(z), \quad n'\geq n$$
$$=\left(\frac{n'!}{n!}\right)^{1/2}(z)^{(n-n')/2}L_{n'}^{n-n'}(z), \quad n'\leq n \quad (23b)$$

with $\alpha\equiv\alpha_v-\alpha_c$, $\beta\equiv\beta_v-\beta_c$, and $z=\frac{1}{2}(\alpha^2+\beta^2)$. The factor $L_n^{n'-n}$ is the associated Laguerre polynomial

$$L_n^{n'-n}(z)\equiv\sum_{q=0}^{n}\frac{(-1)^q}{q!}\binom{n'}{n-q}z^q=\frac{z^{n-n'}e^z}{n!}\frac{d^n}{dz^n}(z^{n'}e^{-z}).$$

We note that $I_{n'n}$ can also be expressed in terms of the summation coefficients $b_m(n',n)$ used by Aronov[14]; that is, his series can be rewritten into a standard series for the associated Laguerre polynomial. As $\omega\to 0$, the transition matrix element and energy shifts reduce exactly to those obtained by Aronov for stationary crossed fields.

Using this result for $I_{n'n}(t)$ and the result (23) for $M_{cv}(t)$, we proceed as for parallel fields to compute $A_l(\omega)$ [Eq. (9)] as a complex integral over $u=\sin\omega t$, and find the stationary point of the exponent which we express in terms of factors

$$\begin{aligned}&\epsilon=\hbar\theta^2\omega_\mu(\omega^2-\omega_c\omega_v)/4\mathcal{E}_{n'n}(\omega^2-\omega_c^2)(\omega^2-\omega_v^2),\\&\zeta^2=\hbar^2k_z^2/2\mu\mathcal{E}_{n'n},\\&\mathcal{E}_{n'n}=\mathcal{E}_g+(n'+\tfrac{1}{2})\hbar\omega_c+(n+\tfrac{1}{2})\hbar\omega_v,\end{aligned}\tag{24}$$

which gives

$$\tilde{\epsilon}_{n'n}=\mathcal{E}_{n'n}(1+\zeta^2+\epsilon).$$

In order to include a tunneling limit in this calculation we would require $\omega\ll\omega_c,\omega_v$ which gives

$$|\epsilon|\approx\frac{(m_c+m_v)E^2c^2}{4\mathcal{E}_gH^2}\ll\frac{(m_c+m_v)}{16\mu}\lesssim 1,$$

where the inequality comes from the parabolic approximation, $(E/H)^2\ll\mathcal{E}_g/2m^*c\approx\mathcal{E}_g/4\mu c^2$ (see Introduction). Thus tunneling ($|\epsilon|\gg 1$) does not occur in the parabolic approximation, but must be calculated using a two-band model.[1]

We work in the multiphonon limit $|\epsilon|\ll 1$, which gives the saddlepoints $u_\pm$ as

$$u_\pm\approx\mp i\gamma_\pm(1+\tfrac{1}{2}\zeta^2),$$

where

$$\gamma_\pm^2=\frac{(\omega^2-\omega_c\omega_v)}{2\epsilon(\omega\pm\omega_c)(\omega\pm\omega_v)}\approx\frac{1}{2\epsilon}\equiv\gamma^2,\quad\text{for}\quad\omega_c,\omega_v\ll\omega.\tag{25}$$

In this limit

$$\epsilon\approx\frac{e^2E^2}{4\mu\omega^2\mathcal{E}_{n'n}},\qquad\gamma^2\approx\frac{2\mu\omega^2\mathcal{E}_{n'n}}{e^2E^2},$$

so that γ^2 is approximately the parameter used by Keldysh,[5] with $\mathcal{E}_g$ now replaced by $\mathcal{E}_{n'n}$. At the saddlepoints the total exponent $E(u)$ in $A_l(\omega)$ has the second derivative

$$E''(u_\pm)\approx\pm 2\mathcal{E}_{n'n}/\hbar\omega\gamma^2$$

which gives

$$A_l(\omega)\approx\frac{1}{2\pi}\left\{\left(\frac{2\pi}{|E''(u_+)|}\right)^{1/2}I_{n'n}(u_+)\exp[-E(u_+)]+\left(\frac{2\pi}{|E''(u_-)|}\right)^{1/2}I_{n'n}(u_-)\exp[E(u_-)]\right\}.$$

Also

$$z(u_\pm)\approx-\frac{\mathcal{E}_{n'n}\omega_\mu}{\hbar(\omega\pm\omega_c)(\omega\pm\omega_v)}\approx-\frac{\omega_\mu}{l\omega},$$

for $\omega_c,\omega_v\ll\omega$ and $\mathcal{E}_{n'n}\approx l\hbar\omega$. Since $|z|\ll 1$, we take the lowest (constant) term in the Laguerre polynomial in $I_{n'n}$. The result for the total transition rate/volume is a sum of multiphoton transition rates

$$W_{n'n}^{(l)}\approx\frac{\omega^2}{4l\pi^2}\left(\frac{eH}{\hbar c}\right)\left(\frac{e^2E^2}{8\mu\omega^2\mathcal{E}_{n'n}}\right)^l\times\left[l\hbar\omega-\left(\mathcal{E}_{n'n}+\frac{e^2E^2}{4\mu\omega^2}\right)\right]^{-1/2}\exp\left(\frac{\mathcal{E}_{n'n}}{\hbar\omega}\right)\alpha_{n'n},\tag{26a}$$

where

$$\alpha_{n'n}=(|n'-n|\,!)^{-2}\left(\frac{n'!}{n!}\right)^{\pm 1}\left(\frac{\omega_\mu}{l\omega}\right)^{|n'-n|}\times\exp\left[\frac{2(n-n')(\omega_c-\omega_v)}{\omega}\right]\tag{26b}$$

with the upper sign for $n'>n$, the lower for $n'<n$.

This result is very similar to the parallel field result [Eq. (16)], especially since we have used the approximation $\omega_c,\omega_v\ll\omega$. It is larger by a factor of 2 because a $\cos f(\gamma)\zeta$ factor does not appear as in Eq. (14), and transitions $\Delta n=\pm m$ are allowed, but are reduced from the $\Delta n=0$ transition by a factor $(\omega_\mu/l\omega)^m\sim(H)^m$. The observation of $\Delta n=0$ transitions in InSb, discussed in the Introduction, can be accounted for by this result.[21]

V. TWO-FIELD ABSORPTION

When a weak optical electric field $\mathbf{E}'\cos\omega' t$ is applied in addition to the strong oscillating field $\mathbf{E}\cos\omega t$, the weak beam may be absorbed either through photon-assisted tunneling[22] or multiphoton absorption.[23] In the

[21] In a letter which came to our attention after the completion of this work, V. Shukovskii and A. Sokolov {Zh. Eksperim. i Teor. Fiz. Pis'ma v Redaktisyu **6**, 311 (1967) [English transl.: Soviet Phys.—JETP Letters **6**, 876 (1967)]} have made a calculation similar to ours for the two-field case (two optical beams) in a transverse magnetic field, but with the strong electric field circularly polarized. This gives their interband energy shift a different denominator: $(\omega+\omega_v)(\omega-\omega_c)$ rather than $(\omega^2-\omega_v^2)$, $(\omega^2-\omega_c^2)$. They also use the effective-mass approximation but do not consider its implied limitation on the strength of the electric field when discussing the enhancement they predict for the transition strength. In the case of circularly polarized light, the only oscillatory part of the wave function is in the coupling to other Landau levels $n'\neq n$ [$\gamma_j(t)$ is linear in t] such that a transition $\Delta n=m\neq 0$ corresponds to the absorption of m photons $\hbar\omega$ and one photon $\hbar\omega'$, similar to the results of ZHL when carried out to mth order in the intraband perturbation. Thus our $\Delta n=0$ transition occurs only for linearly polarized light.

[22] M. Reine, Q. H. F. Vrehen, and B. Lax, Phys. Rev. **163**, 726 (1967).

[23] J. J. Hopfield and J. M. Worlock, Phys. Rev. **137**, A1455 (1965); M. Matsuoka and T. Yajima, Phys. Letters **23**, 54 (1966); V. Koniukhov, L. Kulevskii, and A. Prokhorov, Phys. Status Solidi **21**, K107 (1967); Dokl. Akad. Nauk SSR **173**, 1048 (1967) [English transl.: Soviet Phys.—Doklady **12**, 354 (1967)].

usual experimental configuration, the absorption of the weak beam is observed as a function of ω', as the sample is subjected to a pulsed laser field or a dc electric field. The calculation of these effects is very similar to the calculations in Secs. III and IV for the strong electric field alone.

In this case the interband perturbation [Eq. (4)] is that due to the weak field

$$H_{cv}'=\left[\frac{ihe\mathbf{E}'\cdot\mathbf{p}_{cv}}{m_0\mathcal{E}_g}\right]\cos\omega' t=(E'/E)V_{cv}\cos\omega' t.$$

In the previous calculations we used, for both crossed and parallel fields, perturbation couplings of the form (6a). In the present case (6a) is multiplied by (E'/E), and $\cos\omega\tau$ is replaced by $\cos\omega'\tau$. Then we expand $M_{cv}(\tau)$ [not, as before, $\cos\omega\tau M_{cv}(\tau)$] in a Fourier series in ω. The result is, for absorption (rather than emission) of a photon $\hbar\omega'$,

$$W_{n'n}=\frac{2\pi}{\hbar}(E'/E)^2|V_{cv}|^2\sum_{\text{states }c,v}\sum_{l}|A_l'(\omega)|^2\times\delta(l\hbar\omega+\hbar\omega'-\tilde{\epsilon}_{n'n}),$$

where

$$A_l'(\omega)=\frac{\omega}{2\pi}\int_{-\pi/\omega}^{\pi/\omega}dt\tfrac{1}{2}M_{cv}(t)\exp[i(\tilde{\epsilon}_{n'n}-\hbar\omega')t/\hbar].$$

This result is exactly that used for both parallel and crossed fields, in Eqs. (8) and (9), except that we replace $\mathcal{E}_n$ or $\mathcal{E}_{n'n}$ by $\mathcal{E}_n-\hbar\omega'$ or $\mathcal{E}_{n'n}-\hbar\omega'$, and we divide the result of the saddlepoint calculation of $A_l(\omega)$ by $2\cos\omega t_\pm=2(1-u_\pm^2)^{1/2}\approx 2(1+\gamma^2)^{1/2}$, where γ^2 is defined either in Eq. (13b) or in Eq. (25), again with $\mathcal{E}_n$ or $\mathcal{E}_{n'n}$ reduced by $\hbar\omega'$. Thus we multiply the transition rate by $(E'/E)^2\times(4+4\gamma^2)^{-1}$. This result can also be applied to the zero-magnetic-field calculations of Keldysh.[5]

For photon-assisted tunneling in parallel fields, in the limit $\gamma^2\ll 1$, we replace $\mathcal{E}_n$ in Eq. (15) by $(\mathcal{E}_n-\hbar\omega')$ and obtain an absorption coefficient

$$\alpha(\omega')\sim\exp\left[-\frac{4}{3}\frac{(2\mu)^{1/2}}{\hbar eE}(\mathcal{E}_n-\hbar\omega')^{3/2}\right] \tag{27}$$

which is exactly the exponent obtained by Reine *et al.*[18] and by Weiler *et al.*[1] This result reduces to the Franz-Keldysh effect[24] as $H\to 0$.

For the multiphoton limit in both longitudinal and transverse magnetic fields, where $\gamma^2\gg 1$, we multiply the transition rate by a factor

$$(E'/E)^2\frac{1}{4\gamma^2}=\frac{e^2E'^2}{8\mu\omega^2(\mathcal{E}_{n'n}-\hbar\omega')}.$$

For parallel fields, from Eq. (16) we obtain

$$W_n^{(l+1)}\approx\frac{\omega^2}{8l\pi^2}\left(\frac{eH}{\hbar c}\right)(2\mu)^{1/2}\frac{(e^2E'^2)(e^2E^2)^l}{[8\mu\omega^2(\mathcal{E}_n-\hbar\omega')]^{l+1}}\times\exp\left[\frac{(2l\hbar\omega+\hbar\omega'-\mathcal{E}_n)}{\hbar\omega}\right]\times\left[l\hbar\omega+\hbar\omega'-\left(\mathcal{E}_n+\frac{e^2E^2}{4\mu\omega^2}\right)\right]^{-1/2} \tag{28}$$

and for crossed fields, from Eq. (26),

$$W_{n'n}^{(l+1)}\approx\frac{\omega^2}{4l\pi^2}\left(\frac{eH}{\hbar c}\right)(2\mu)^{1/2}\frac{(e^2E'^2)(e^2E^2)^l}{[8\mu\omega^2(\mathcal{E}_{n'n}-\hbar\omega')]^{l+1}}\times\exp\left(\frac{\mathcal{E}_{n'n}-\hbar\omega'}{\hbar\omega}\right)\alpha_{n'n}\times\left[l\hbar\omega+\hbar\omega'-\left(\mathcal{E}_{n'n}+\frac{e^2E^2}{4\mu\omega^2}\right)\right]^{-1/2}. \tag{29}$$

From these equations we obtain the transition rate and selection rules for the absorption of $(l+1)$ photons, with a total energy change of $(l\hbar\omega+\hbar\omega')$, with the absorption coefficient at ω' proportional to the lth power of the intensity at ω.

It has been pointed out[25] that the polarization dependence of "allowed" two-photon absorption, using two beams, gives information about the symmetry properties of the band-edge states (the tensor properties of $\mathbf{E}'\cdot\mathbf{p}_{cj}\mathbf{p}_{jv}\cdot\mathbf{E}$) which cannot be obtained from single-photon measurements. For the case we treat, where the two-photon transition is of the "forbidden" type, the dependence of the polarization of the weak beam $\mathbf{E}'$ is just that of the one-photon matrix element $\mathbf{E}'\cdot\mathbf{p}_{cv}$. In a magnetic field, the $n'-n$ selection rules depend on the polarization of the strong field $\mathbf{E}$ with respect to the magnetic field. Also, for nonspherical bands, the transition rate will depend on the orientation of $\mathbf{E}$ and $\mathbf{H}$ with respect to the effective-mass tensor.

VI. CONCLUSION

Using the effective-mass approximation, we have obtained solutions to the intraband effects of an oscillating electric field in the presence of longitudinal and transverse magnetic fields, and used these to calculate interband transitions by means of first-order time-dependent perturbation theory. We have shown that an allowed one-photon interband transition has associated with it a series of l-photon transitions with the same selection rules, caused by the intraband effects of the electric field, proportional to $(\mathcal{E}_E/2\mathcal{E}_{cv})^l$, where $\mathcal{E}_E$ is the electric-field shift of the transition energy $\mathcal{E}_{cv}$.

[24] W. Franz, Z. Naturforsch. **13a**, 484 (1958); L. V. Keldysh, Zh. Eksperim. i Teor. Fiz. **34**, 1138 (1958) [English transl.: Soviet Phys.—JETP **7**, 788 (1958)].

[25] M. Inoue and Y. Toyozawa, J. Phys. Soc. Japan **20**, 363 (1965).

Our calculation can be carried out to higher order in the interband perturbation, following Braunstein's method,[2] but using the time-dependent wave functions. This will result in higher-order multiphoton transitions associated with any allowed interband-type multiphoton transition.

Two-photon experiments in a magnetic field, where the selection rules for n could be obeyed, would provide a test of the above results. The photoconductivity experiments of Button *et al.*[4] are not accurate enough to provide firm evidence, although they do tend to support our theory as opposed to the perturbation theory of ZHL. In any case our theory should provide a method for analyzing multiphoton transitions whenever the interband-type transition is forbidden.

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APPENDIX

The Luttinger-Kohn effective-mass approximation[9] has been carried out by Zak and Zawadzki[26] to include both electric and magnetic fields. The diagonalization procedure they use leaves certain interband perturbing terms, including one due to the electric field, which they do not write out explicitly. In this Appendix we give an outline of their procedure, obtain an expression for the interband electric-field perturbation, and discuss the approximations made in relation to our solutions in Secs. III and IV.

The equation of motion we treat is Eq. (1), which we write as

$$\mathcal{H}\psi(\mathbf{r},t)=\left[\frac{\mathbf{P}^2}{2m_0}+V(\mathbf{r})+U(\mathbf{r},t)\right]\psi(\mathbf{r},t)=i\hbar\frac{\partial\psi}{\partial t}(\mathbf{r},t),$$

where $U(\mathbf{r},t)=-e\mathbf{E}\cdot\mathbf{r}\cos\omega t$. In the Kohn-Luttinger representation this becomes, in the notation of Callaway,[27]

$$\begin{aligned}(n\mathbf{k}|\mathcal{H}|n'\mathbf{k}')&=[\mathcal{E}_n\delta(\mathbf{k}-\mathbf{k}')+(1/2m_0)(\mathbf{k}|\mathbf{P}^2|\mathbf{k}')\\&\quad+(\mathbf{k}|U(\mathbf{r},t)|\mathbf{k}')]\delta_{nn'}\\&\quad+(1/m_0)\mathbf{p}_{nn'}\cdot(\mathbf{k}|\mathbf{P}|\mathbf{k}')\\&=\mathcal{H}_0^{(\mathrm{D})}+\mathcal{H}_1^{(\mathrm{D})}+\mathcal{H}_2^{(\mathrm{D})}+\mathcal{H}_3^{(\mathrm{ND})}.\end{aligned}$$

The potential u, being linear in $\mathbf{r}$, has only diagonal matrix elements. We remove the nondiagonal term by a canonical transformation

$$\tilde{\mathcal{H}}=e^{-S}\mathcal{H}e^{S}=\mathcal{H}+[\mathcal{H},S]+\tfrac{1}{2}([\mathcal{H},S],S)+\cdots,$$

requiring

$$[\mathcal{H}_0^{(\mathrm{D})},S]=-\mathcal{H}_3^{(\mathrm{ND})}$$

or

$$\begin{aligned}(n\mathbf{k}|S|n'\mathbf{k}')&=(\mathbf{p}_{nn'}/m\omega_{nn'})\cdot(\mathbf{k}|\mathbf{P}|\mathbf{k}'),&n'\neq n\\&=0,&n'=n.\end{aligned}$$

Then

$$\tilde{\mathcal{H}}=\mathcal{H}_0^{(\mathrm{D})}+\mathcal{H}_1^{(\mathrm{D})}+\mathcal{H}_2^{(\mathrm{D})}+[\mathcal{H}_1^{(\mathrm{D})},S]+[\mathcal{H}_2^{(\mathrm{D})},S]+[\mathcal{H}_3^{(\mathrm{ND})},S]+\tfrac{1}{2}[[\mathcal{H}_0^{(\mathrm{D})},S],S]+\cdots.$$

We neglect the term $[\mathcal{H}_1^{(\mathrm{D})},S]$ and remove the nondiagonal parts of

$$[\mathcal{H}_3^{(\mathrm{ND})},S]=-([\mathcal{H}_1^{(\mathrm{D})},S],S)$$

by a higher-order transformation leaving a diagonal part which contributes to the effective mass. The term $[\mathcal{H}_2^{(\mathrm{D})},S]$ is the interband perturbation due to the electric field:

$$\begin{aligned}&(n\mathbf{k}|[\mathcal{H}_2^{(\mathrm{D})},S]|n'\mathbf{k}')\\&\quad=-i(\mathbf{p}_{nn'}/m\omega_{nn'})\cdot(\mathbf{k}|\nabla U(\mathbf{r},t)|\mathbf{k}')\\&\quad=i(\mathbf{p}_{nn'}/m\omega_{nn'})\cdot e\mathbf{E}\cos\omega t\delta(\mathbf{k}-\mathbf{k}')\\&\quad\equiv iV_{nn'}\cos\omega t\delta(\mathbf{k}-\mathbf{k}').\end{aligned}\tag{A1}$$

The result for $\tilde{\mathcal{H}}$ is

$$(n\mathbf{k}|\tilde{\mathcal{H}}|n'\mathbf{k}')=\left[\mathcal{E}_n\delta(\mathbf{k}-\mathbf{k}')+\frac{1}{2m_{\alpha\beta}}(\mathbf{k}|\mathbf{P}^\alpha\mathbf{P}^\beta|\mathbf{k}')+(\mathbf{k}|U(\mathbf{r},t)|\mathbf{k}')\right]\delta_{nn'}+iV_{nn'}\cos\omega t\delta(\mathbf{k}-\mathbf{k}'),$$

where the effective-mass tensor is defined by

$$1/m_{\alpha\beta}=1/m_0+(2/m_0^2)\sum_{n''}p_{nn''}{}^{\alpha}p_{n''n}{}^{\beta}/\omega_{nn''}.$$

We transform from k space to r space, assuming spherical bands, and obtain Eq. (3) for the envelope function, with the interband term $[\mathcal{H}_2^{(\mathrm{D})},S]$ giving the perturbation in Eq. (4).

This effective-mass approximation is valid only under certain conditions, which have been enumerated by Zak and Zawadzki[26]: (a) The interband perturbation $[\mathcal{H}_2,S]$ and also the term $[\mathcal{H}_1,S]$ must be small with respect to the zero-order terms. (b) The envelope functions $\psi_j(\mathbf{r},t)$ must have Fourier components $\mathbf{k}$ such that S is small, i.e., $\mathbf{k}\cdot\mathbf{p}_{nn'}/m\omega_{nn'}\ll1$, where $\mathbf{k}$ represents $(\mathbf{k}|\mathbf{P}|\mathbf{k}')$ which includes the magnetic potential. If we define $a\equiv p_{nn'}{}^{\alpha}/m\omega_{nn'}$, this means $ka\ll1$.

Zak and Zawadzki show that in an external magnetic field, $[\mathcal{H}_1^{(\mathrm{D})},S]$ is small if

$$(2n+1)^{1/2}(a/\lambda)\ll1$$

which restricts the results to the first few Landau levels in large magnetic fields (50–100 kOe). They also obtain for crossed electric and magnetic fields the condition

$$eEa/\hbar\omega_c\ll1$$

[26] J. Zak and W. Zawadzki, Phys. Rev. **145**, 536 (1966).
[27] J. Callaway, *Energy Band Theory* (Academic Press Inc., New York, 1964), p. 246.

which restricts our results to the multiphoton limit for the transverse case. We know that the interband electric term $[\mathcal{H}_2^{(D)},S]$ is small since it leads either to tunneling or multiphoton absorption, both of which are relatively weak effects.

We estimate the Fourier components $\mathbf{k}$ of our solutions in Secs. II and III at the saddlepoints $u_\pm=\sin\omega t_\pm \approx i\gamma$. For a longitudinal magnetic field, k_z is increased by $(eE/\hbar\omega)\sin\omega t_\pm\approx i(2\mu\mathcal{E}_n)^{1/2}/\hbar$, so the restriction $|k_z|a\ll 1$ gives the condition

$$(p_{cv}/m_0)^2\ll\mathcal{E}_g^2/2\mu\mathcal{E}_n. \tag{A2}$$

We obtain essentially the same condition in a transverse magnetic field, where we use $\Delta k_x\approx\alpha_j/\lambda\approx i(2\mu\mathcal{E}_{n'n})^{1/2}/\hbar$ at the saddlepoints $u_\pm$. But, from the two-band model $(p_{cv}/m_0)^2\approx\mathcal{E}_g/4\mu$, which gives the condition

$$\mathcal{E}_{n'n},\mathcal{E}_n\ll 2\mathcal{E}_g \tag{A3}$$

which again limits our results to the first few Landau levels in high magnetic fields. The accuracy of our approximation is not very good: At $H\to 0$, $\mathcal{E}_{n'n}$, $\mathcal{E}_n\to\mathcal{E}_g$ which gives $|k|a\approx\frac{1}{2}$ which is not very much smaller than 1. Thus our results must be used carefully, in that the conditions (A2) and (A3) must be checked for a given material, and estimates of transition strengths can be expected to be only approximate.

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Calculation of the Formation Energy of a Schottky Defect in Germanium*

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The formation energy of a Schottky defect in germanium has been calculated from first principles by using the valence-bond method of Heitler and London. The formation energy of a vacancy is given by the difference between the ground-state energy of the crystal containing the defect and that of the perfect crystal. The latter is derived by using a method based on the general directional theory of valence. The energy of the defect crystal is obtained by using the same general method but taking into account the possible pairing schemes for the vacancy electrons and applying the method of resonance. The tetrahedral valence state of germanium is used as the reference level for the various energies in the calculation. The numerical result obtained depends on the value chosen for the cohesive energy of germanium. Using an average of the three reported values for this quantity, we find for the formation energy at a Schottky defect $E_v=2.21\pm0.18$ eV. This is in good agreement with the experimental values.

1. INTRODUCTION

A KNOWLEDGE of the energy required to form a Schottky defect in a semiconductor is necessary for the analysis of experiments on diffusion, quenching, and radiation damage. Calculations of this energy for germanium have been reported by Swalin,[1] Scholz,[2] Scholz and Seeger,[3] and Bennemann.[4] Swalin assumed a Morse potential to describe the covalent bond in the crystal while Scholz, and Scholz and Seeger used a combination of the harmonic approximation to Born's lattice potential with a Morse potential. Bennemann[4] developed a method using first principles. The purpose of this paper is to present a calculation, also from first principles, based on the method of atomic functions developed initially by Heitler and London. Experimental values for the energy of formation of a singly charged negative vacancy in germanium have been reported by a number of workers.[5–11] The values range from 1.7 to about 2 eV. The corresponding values for the neutral vacancy can be calculated by the method used by Kröger.[11]

The calculations by Swalin, by Scholz, and by Scholz and Seeger are open to criticism on a number of grounds. The use of a Morse function to represent the potential is questionable on theoretical grounds. Even assuming that the Morse function could be used as a reasonable approximation, the reference level for the dissociation energy of a covalent bond should be the sp^3 valence state of the constituent atom of the solid, not the free ^3P state used by these authors. This can be seen most

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