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Two-Photon Excitation Rate in Indium Antimonide*

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Kane's wave functions for InSb are used with second-order perturbation theory to obtain an accurate expression for the two-photon transition rate. The statistics of the crystal electrons are included in the general expression. A simplified expression is obtained under assumptions that fit our experimental conditions well, namely, the valence-band states and the conduction-band states involved in the transitions are completely filled and completely empty, respectively. The transition rate per unit volume at 77 °K is $0.36\vartheta^2$ and $0.064\vartheta^2$ $\sec^{-1}\,\mathrm{cm}^{-3}$ with irradiation wavelengths $\lambda = 9.6$ and 10.6 μm, respectively, and at 2 °K the rate is $0.25\vartheta^2$ $\sec^{-1}\,\mathrm{cm}^{-3}$ with $\lambda = 9.6$ μm, for irradiation intensity ϑ in erg $\sec^{-1}\,\mathrm{cm}^{-2}$. The rate for $T = 77$ °K and $\lambda = 10.6$ μm previously reported by Danishevskii is one order of magnitude larger than the present result. This discrepancy in the calculated values is rationalized. Measurements of the transition rates are described and the obtained values are given for the two temperatures and two wavelengths mentioned above. The agreement between our measured and theoretical values is within the experimental accuracy.

I. INTRODUCTION

Two-photon interband transitions in InSb have been observed recently in several laboratories.[1-4] Danishevskii and co-workers measured the transition rate in n-InSb at the 10.6-μm wavelength radiation from a Q-switched CO_2 laser.[2] Their result is in agreement with a theoretical estimate they obtained from an expression for the transition rate given by Basov and co-workers.[5] The other laboratories that have reported work on two-photon excitation in InSb did not give enough information to enable obtaining a value for the transition rate.[1,3,4] Danishesvkii's measurement and theoretical estimate do not agree with experimental values for the two-photon transition rate obtained in our laboratory. Furthermore, our calculations show that Danishevskii's theoretical estimate is one order of magnitude too large.

We have obtained a detailed expression for the two-photon transition rate in InSb by using the wave functions calculated by Kane.[6] The momentum matrix elements between the conduction and valence bands are written for any $\vec{k}$ vector close to the center of the Brillouin zone. The two-photon transition rate is calculated from these matrix elements with second-order time-dependent perturbation theory. Although this method is the simplest and most successful, as evidenced by the variety of materials to which it has been applied,[7] a detailed calculation for InSb, taking advantage of Kane's wave functions, does not appear to have been published previously. The rates obtained (Sec. II) at both the 10.6- and 9.6-μm excitation wavelengths are in agreement with our experimental results at these wavelengths (Sec. III).

The perturbation theory is subjected to the second-quantization formalism in this paper, since the statistics of the electrons are a natural part of this formalism. A direct result is that terms in the final expression for the transition rate that include matrix elements involving any intermediate states $|n\rangle$, different from either the initial or final states, will be multiplied by a factor $(1-f_n)$. The distribution function f_n gives the occupation probability for the state $|n\rangle$. Since f_n is very close to 1 for both the heavy- and light-hole bands in InSb for the excess carrier densities considered here, the terms that are multiplied by $(1-f_n)$ are negligible. By this procedure a simple expression for the two-photon transition rate in InSb is obtained in the last part of Sec. II. The final result has four adjustable parameters, namely, four material constants. These are the optical band-gap energy, the spin-orbit splitting of the valence band, and the effective masses of the heavy-hole band and the conduction band. The effective masses of the heavy-hole band and the conduction band are available from low-temperature Faraday-rotation measurements.[8] The optical band gap and the spin-orbit splitting are known from magnetoreflection, magnetoabsorption, and Faraday-rotation measurements.[8-10] The theoretical and the experimental results are discussed in Sec. IV.

Our calculations are intended for the temperature range 2–77 °K; however, they can easily be extended up to room temperature by using the technique outlined by Ehrenreich.[11] Since we use the parabolic-energy-band approximation, the upper limit of $\hbar\omega$ for which the nonparabolicity causes an error $\lesssim 10\%$ is $\hbar\omega = \frac{1}{2}E_g + 0.015$ eV, corresponding to an excitation wavelength down to approximately 9.6 μm at 77 °K and 9.3 μm at 2 °K. The band-gap energy E_g for InSb is 0.2357–

0.228 eV in the temperature range 4–77 °K.[9,10] The value $E_g = 0.2357$ eV is also used for 2 °K in the present work.

II. CALCULATION OF TRANSITION RATE

A. General Description

For the calculations in this section it is assumed that the conduction band and the two valence bands in InSb have spherical energy surfaces with the extrema in the center of the Brillouin zone and that the band energies have a parabolic dependence on the wave vector $\vec{k}$. These assumptions are justified since the electrons involved in the transitions have a wave vector $|\vec{k}| \lesssim 10^6$ cm^{-1}. By comparison, the boundary of the first zone in the ⟨100⟩ directions is located approximately at $k = 10^8$ cm^{-1}. In our worst case, $k \approx 10^6$ cm^{-1}, the parabolic approximation gives a conduction-band energy referred to the bottom of the band that is approximately 10% higher than the nonparabolic value obtained by Kane.[6]

The Hamiltonian operator for the system including the conduction- and valence-band Bloch states, the photon modes, and the interaction between electrons and photons is

$$H = H_0 + H_1, \tag{1}$$

where

$$H_0 = \sum_{i,\vec{k}} b^\dagger_{i\vec{k}} b_{i\vec{k}} E_i(\vec{k}) + \sum_{\hat{\alpha},\omega} \hbar\omega (a^\dagger_{\hat{\alpha}\omega} a_{\hat{\alpha}\omega} + \tfrac{1}{2}) \tag{2}$$

and

$$H_1 = -\left(\frac{2\pi e^2\hbar}{L^3 m^2 \epsilon}\right)^{1/2} \sum_{\vec{k},\omega,\hat{\alpha}}^{i,j} \frac{(\vec{p}\circ\hat{\alpha})_{i,j}}{\omega^{1/2}} \times (a^\dagger_{\hat{\alpha}\omega} + a_{\hat{\alpha}\omega}) b^\dagger_{i\vec{k}} b_{j\vec{k}}. \tag{3}$$

Here $(\vec{p}\cdot\hat{\alpha})_{i,j}$ is the matrix element of the $\hat{\alpha}$ component of the momentum operator between the Bloch states $|i\rangle$ and $|j\rangle$, and $\hat{\alpha}$ is the polarization direction of the photon. The summation indices i and j are combined band and spin indices and are summed over the six electron states (each of the bands has twofold spin degeneracy) for each value of $\vec{k}$ in the first Brillouin zone. The energy of an electron in the state $|i\vec{k}\rangle$ is $E_i(\vec{k})$, $\hbar$ is Planck's constant divided by 2π, e is the charge of an electron, m is the free-electron mass, and ϵ is the dielectric constant. The operators $b^\dagger$, b, $a^\dagger$, and a are the creation and annihilation operators for the electrons and photons, respectively; for example, the term $a_{\hat{\alpha}\omega} b^\dagger_{i\vec{k}} b_{j\vec{k}}$ causes an electron transition from state j to i, with energy $\hbar\omega$ supplied by a photon.

The photon states of frequency ω are taken to be normal modes in a cavity of volume L^3. Their wave vector is neglected compared with the electron wave vector $\vec{k}$. Conservation of momentum therefore requires that the $\vec{k}$ vector be the same for both states i and j in Eq. (3).

The quantized interaction Hamiltonian H_1 is obtained from the interaction energy $(2m)^{-1} \times [-2(e/c)\vec{p}\cdot\vec{A} + (e/c)^2 |\vec{A}|^2]$, where c is the speed of light in a vacuum and $\vec{A}$ is the magnetic vector potential. The interaction term proportional to $|\vec{A}|^2$ vanishes when the photon momentum is neglected, because of the orthogonality of all the electron states.

B. Evaluation of Matrix Elements

To evaluate the momentum matrix elements in Eq. (3) we use the wave functions Kane obtained by diagonalizing the mutual interaction between the conduction and valence bands.[6] These wave functions and their corresponding eigenenergies describe the central part of the Brillouin zone quite accurately for all the relevant bands except the heavy-hole band. The accuracy of Kane's model is probably best illustrated by the good fit between absorption edge measurements and theoretical absorption calculations obtained by using Kane's wave functions.[6,12]

Kane improves the heavy-hole band description by including the perturbation caused by other bands. His second-order correction is not included in the present calculation since the description of the heavy-hole band does not affect our results significantly. Its description enters our result in two ways: (i) directly through the value of m_{v1} and (ii) indirectly through matrix elements involving heavy-hole band Bloch states. Here m_i is the effective mass for the band i with $i = c$, $v1$, and $v2$, the electron, heavy-hole, and light-hole bands, respectively. In connection with (i), our expression for the transition rate contains $(m_{cv1})^{1/2}$ as a factor, where m_{cv1} is the reduced electron-hole mass. Since $m_c \ll m_{v1}$, $m_{cv1} \approx m_c$ independent of the value of m_{v1}. The importance of (ii) is determined by making a very rough estimate of the admixture of higher band wave functions to the heavy-hole band wave functions. According to Kane's Eq. (12), the heavy-hole band has the same curvature as a free-electron band. In this estimate we require the second-order correction from the first higher band with Γ_{12} symmetry through the second-order $\vec{k}\cdot\vec{p}$ perturbation term to turn the curvature of the $v1$ band down until $m_{v1} = 0.18m$ in the ⟨110⟩ directions.[13] (Γ_{12} is referred to as Γ_3 in Kane's paper.) Then, assuming this Γ_{12} band to be located at least 3 eV above the valence band, only 5% or less of the Γ_{12} wave functions need to be mixed into the $v1$ wave functions to obtain $m_{v1} = 0.18m$. Thus the higher-order correction to the band structure is unimportant for the calculations in this paper.

Dresselhaus uses group theory to show that in the center of the Brillouin zone the cell-periodic part of the electron wave function corresponding to

the conduction band has the symmetry properties of the atomic s state under the operations of the tetrahedral group.[14] The two degenerate states are therefore designated by $S\uparrow$ (spin-up) and $S\downarrow$ (spin-down). Similarly, the valence-band wave functions at $\vec{k}=0$ have the symmetry properties of the atomic p state and are designated by $X\uparrow$, $Y\uparrow$, $Z\uparrow$, $X\downarrow$, $Y\downarrow$, and $Z\downarrow$, all independent of $\vec{k}$, to reflect the symmetry of the p state. For nonvanishing values of $\vec{k}$, the cell-periodic part of the electron wave functions are given by Kane as a linear combination of the eight $\vec{k}$-independent states just mentioned. The results are given by Eqs. (6)–(8), (14), and (15) in his paper.

The polarization direction of the laser beam is taken to be in the z direction, $\hat{\alpha}=\hat{z}$. Therefore we need only the matrix elements of the z component of the momentum operator in Eq. (3). Because of symmetry, $\langle S\uparrow | p_z | Z\uparrow\rangle$, $\langle S\downarrow | p_z | Z\downarrow\rangle$, and their complex conjugates are the only nonzero matrix elements of the z component of the electron momentum operator taken between these eight states. Using this selction rule together with Kane's wave functions, we have calculated the matrix elements of p_z. The results are given in Table I. In this table $\vec{k}$ is given by its spherical coordinates (k, θ, ϕ). The spin parts of the two degenerate wave functions in each band are now labeled separately by α and β. The constants A, B and Q are defined as follows:

$$A=(a_c c_{v2}+c_c a_{v2}),\quad B=(a_c b_{v2}-b_c a_{v2}), \tag{4}$$

and

$$Q=\langle iS \,|\, p_z \,|\, Z\rangle . \tag{5}$$

Here

$$\begin{aligned} a_i &= Q(\hbar k/m)(E_i'+\tfrac{2}{3}\Delta)/\Pi_i , \\ b_i &= (\sqrt{2}\,\Delta/3)(E_i'-E_g)/\Pi_i , \\ c_i &= (E_i'-E_g)(E_i'+\tfrac{2}{3}\Delta)/\Pi_i , \end{aligned} \tag{6}$$

where Π_i is a normalizing factor equal to the square root of the sum of the squares of the numerators. For the parabolic energy-band approximation the definition of E_i' is

$$\begin{aligned} E_c' &= E_c-\hbar^2k^2/2m = E_g+(\hbar^2k^2/2m)(m/m_c-1), \\ E_{vi}' &= E_{vi}-\hbar^2k^2/2m = -(\hbar^2k^2/2m)(m/m_{vi}+1), \end{aligned} \tag{7}$$

and Δ is the spin-orbit-splitting energy. Equation (6) is the same as Kane's Eq. (15), except that he uses the constant $P=Q\hbar/m$ instead of our Q.

C. Transition Rate

We now use Eqs. (1)–(6) to calculate the transition rate for transitions between the two valence bands and the conduction band. The transition probability per unit time $w_{i,f}$ for the system described by Eqs. (1)–(3) to go from an initial state $|i\rangle$ of energy $\mathcal{E}_i$ to a final state $|f\rangle$ of energy $\mathcal{E}_f$ is given by Fermi's golden rule

$$w_{i,f}=(2\pi/\hbar)\,|M_{i,f}|^2\,\delta(\mathcal{E}_f-\mathcal{E}_i), \tag{8}$$

where

$$M_{i,f}=\sum_n \frac{\langle f | H_1 | n\rangle\langle n | H_1 | i\rangle}{\mathcal{E}_n-\mathcal{E}_i} . \tag{9}$$

Here $\mathcal{E}$ includes the energy of the photons. In Eq. (9) it is assumed that the photon energy $\hbar\omega$ is less than the band-gap energy E_g. The first-order term $\langle f | H_1 | i\rangle$ can therefore be neglected. Considering the transitions between the heavy-hole band $v1$ and the conduction band c for a particular value of $\vec{k}$, we see that the net transition probability per unit time from $v1$ to c consists of eight w terms:

$$\begin{aligned} w_{v1\rightleftarrows c} = {} & w_{v1\alpha,c\alpha}+w_{v1\beta,c\alpha}+w_{v1\alpha,c\beta}+w_{v1\beta,c\beta} \\ & -w_{c\alpha,v1\alpha}-w_{c\alpha,v1\beta}-w_{c\beta,v1\alpha}-w_{c\beta,v1\beta} . \end{aligned} \tag{10}$$

Each of these eight terms contains a summation over six intermediate states [see Eq. (9)]. These states are the four valence-band states and the two conduction-band states.

Higher and lower bands should also have been included among the intermediate states; however, with one exception, they are located so far away in energy space from the three considered bands that the denominator in Eq. (9) is sufficiently large to make their contribution negligible. The exception

TABLE I. Matrix elements of the z component of the momentum operator. The matrix is symmetric about the diagonal, so only one-half of the off-diagonal terms are given. For definition of A, B, and Q see Eqs. (4)–(7). The angle between the wave vector of the light and the z axis is denoted by θ.

$c\alpha$	$c\beta$	$v1\alpha$	$v1\beta$	$v2\alpha$	$v2\beta$	Bloch states
$(\hbar km/m_c)\cos\theta$	0	0	$-a_cQ\sin\theta/\sqrt{2}$	$AQ\cos\theta$	$BQ\sin\theta/\sqrt{2}$	$c\alpha$
	$(\hbar km/m_c)\cos\theta$	$-a_cQ\sin\theta/\sqrt{2}$	0	$-BQ\sin\theta/\sqrt{2}$	$AQ\cos\theta$	$c\beta$
		$(\hbar km/m_{v1})\cos\theta$	0	0	$-a_{v2}Q\sin\theta/\sqrt{2}$	$v1\alpha$
			$(\hbar km/m_{v1})\cos\theta$	$-a_{v2}Q\sin\theta/\sqrt{2}$	0	$v1\beta$
				$(\hbar km/m_{v2})\cos\theta$	0	$v2\alpha$
					$(\hbar km/m_{v2})\cos\theta$	$v2\beta$

is the third valence band, the "spin-orbit split-off" band; however, it cannot contribute as an intermediate state, since it is filled.

The evaluation of the terms in Eq. (10) is greatly simplified by grouping equal terms and using the selection rules contained in Table I. Also, since the number of photons in the incident laser beam N_ω is a large number, further simplification may be obtained by setting $N_\omega + 2 \approx N_\omega + 1 \approx N_\omega$. In the expression for the transition rate, the spontaneous emission term and a term that depends linearly on the radiation intensity thus vanish. These two terms are discussed by Chang and Ancker-Johnson for a two-band model.[15]

Once the vanishing matrix elements are eliminated from Eq. (10), the electron occupation numbers N_i occur only in combinations like $(N_{v1\alpha} - N_{c\alpha})$ and $(N_{v1\alpha} - N_{c\alpha})(1 - N_{v2\beta})$, where N_i can take only the value 1 or 0. Thus the N_i may be replaced by the distribution functions f_i to obtain average values. The distribution function does not contain the spin label, since the occupation probability is the same for two degenerate states.

The net two-photon transition rate from the heavy-hole to the conduction band $W_{v1 \rightarrow c} = \sum_{\vec{k}} w_{v1 \rightarrow c}$ is

$$W_{v1\rightarrow c} = \frac{16\pi^3 g^2}{\hbar m^4 \omega^4 \epsilon}\left(\frac{e^2}{\hbar c}\right)^2 \sum_{\vec{k}} (f_{v1} - f_c) \left\{ \frac{(P_{c\alpha,c\alpha} - P_{v1\beta,v1\beta})^2 (P_{c\alpha,v1\beta})^2}{\omega^2} + (1 - f_{v2}) \right.$$
$$\left. \times \left[\left(\frac{P_{c\alpha,v2\beta} P_{v2\beta,v1\alpha}}{\omega'} \right)^2 + \left(\frac{P_{c\alpha,v2\alpha} P_{v2\alpha,v1\beta}}{\omega'} \right)^2 - 2 \frac{(P_{c\alpha,c\alpha} - P_{v1\beta,v1\beta}) P_{c\alpha,v1\beta} P_{c\alpha,v2\alpha} P_{v2\alpha,v1\beta}}{\omega' \omega} \right] \right\} \delta(\mathcal{E}_c - \mathcal{E}_{v1}). \tag{11}$$

Similarly,

$$W_{v2\rightarrow c} = \frac{16\pi^3 g^2}{\hbar m^4 \omega^4 \epsilon}\left(\frac{e^2}{\hbar c}\right)^2 \sum_{\vec{k}} (f_{v2} - f_c) \left\{ \frac{(P_{c\alpha,c\alpha} - P_{v2\alpha,v2\alpha})^2 [(P_{c\alpha,v2\alpha})^2 + (P_{c\alpha,v2\beta})^2]}{\omega^2} \right.$$
$$\left. + (1 - f_{v1}) \left[\left(\frac{P_{c\alpha,v1\beta} P_{v1\beta,v2\alpha}}{\omega''} \right)^2 - 2 \frac{(P_{c\alpha,c\alpha} - P_{v2\alpha,v2\alpha}) P_{c\alpha,v2\alpha} P_{c\alpha,v1\beta} P_{v1\beta,v2\alpha}}{\omega'' \omega} \right] \right\} \delta(\mathcal{E}_c - \mathcal{E}_{v2}). \tag{12}$$

$P_{i,j}$ is the matrix element of the z component of the momentum operator given in Table I. The radiation intensity is given by $g = \hbar \omega N_\omega c/(L^3 \sqrt{\epsilon})$. In the denominators

$$\omega' \equiv \omega + [E_{v1}(\vec{k}) - E_{v2}(\vec{k})]/\hbar$$

and

$$\omega'' \equiv \omega - [E_{v1}(\vec{k}) - E_{v2}(\vec{k})]/\hbar .$$

The quantities $P_{i,j}$, ω', ω'', and f_i are all dependent on $\vec{k}$.

Equations (11) and (12) show that an intermediate band, either $v1$ or $v2$, plays an important part in the transition rate unless it is filled. However, for the excess carrier densities characteristic of our experiments it is a good approximation to set $f_{v1} = f_{v2} = 1$ and $f_c = 0$. Fermi-Dirac statistics then yield the following values at 77 °K for the k value corresponding to 10.6-μm irradiation: $f_{v1} = 0.9988$, $f_c \approx 0.1$, and $f_{v2} > f_{v1}$. At 2 °K with 9.6-μm irradiation, and for a heavy-hole band average effective mass of $0.39m$, $f_c = 0$ and $f_{v1} \approx 0.96$.

The final evaluation of Eqs. (11) and (12) needed to interpret our measurements ($f_{v1} = f_{v2} = 1$ and $f_c = 0$) require the following relations:

$$\sum \rightarrow \frac{V}{8\pi^3} \int_0^{2\pi} \int_0^{\pi} \int_0^{\infty} k^2 dk \sin\theta d\theta d\phi, \tag{13}$$

and an expression between the Dirac δ functions,

$$\delta(\mathcal{E}_c - \mathcal{E}_{vi}) = [m_{cvi}/(\hbar^2 k)]\, \delta(k - k_{cvi}), \quad i = 1, 2. \tag{14}$$

Here

$$k_{cvi} \equiv (1/\hbar)[2 m_{cvi}(2\hbar\omega - E_g)]^{1/2}. \tag{15}$$

The integration using Eqs. (11)–(15), Table I, $f_{v1} = f_{v2} = 1$, and $f_c = 0$ gives

$$\frac{1}{V} W_{vi\rightarrow c} = \frac{16\sqrt{2}}{15} \frac{\pi}{\epsilon}\left(\frac{e^2}{\hbar c}\right)^2 \frac{(C_i Q)^2}{\hbar^4 \omega^6 m^2} \times (m_{cvi})^{1/2} (2\hbar\omega - E_g)^{3/2} g^2, \tag{16}$$

where

$$Q = [\tfrac{3}{2} m E_g (m/m_c - 1)(E_g + \Delta)/(3E_g + 2\Delta)]^{1/2},$$
$$C_1^2 = a_c^2, \tag{17}$$

and

$$C_2^2 = 3A^2 + B^2 = 3(a_v c_{v2} + c_c a_{v2})^2 + (a_c b_{v2} - b_c a_{v2})^2. \tag{18}$$

The total transition rate per unit volume is $W/V = (W_{v1\rightarrow c} + W_{v2\rightarrow c})/V$. Equation (17) is equivalent to the first of Kane's Eqs. (12). Numerical values for W/V are listed in Table II for $T = 2$ and 77 °K, together with our measured values. The numerical values are obtained by taking $\Delta = 0.9$ eV, $m_c = 0.0145m$, $m_{v1} = 0.39m$, and $m_{v2} = 0.016m$.[8]

Discussion of these results is contained in Sec. IV. Next the measurements are described.

TABLE II. Theoretical and experimental values for the two-photon excitation rate in InSb. $K_2 \equiv W/(V\mathcal{G}^2) = (W_{v1\rightarrow c} + W_{v2\rightarrow c})/(V\mathcal{G}^2)$.

T	(°K)	2	2	77	77
λ	(μm)	9.6	10.6	9.6	10.6
$W_{v1\rightarrow c}/(V\mathcal{G}^2)$	(cm sec/erg^2)	0.10	0	0.14	0.024
$W_{v2\rightarrow c}/(V\mathcal{G}^2)$	(cm sec/erg^2)	0.15	0	0.22	0.040
K_2 Theor.	(cm sec/erg^2)	0.25	0	0.36	0.064
K_2 Expt.	(cm sec/erg^2)	0.46–0.9	0	0.56–0.69	0.032–0.064

III. MEASUREMENTS OF TRANSITION RATE

A. Experimental Method

A Q-switched CO_2 laser is used to excite n-type InSb single-crystalline samples. The laser is tunable to either 9.6 or 10.6 μm radiation wavelength. To further ensure that only one of these wavelengths is present, dielectric bandpass filters are used.

For the transition rate experiments the laser beam is unfocused. The peak power density in the center of the beam, $\mathcal{G}_{max}$, is determined from measurements of the average total power, the time dependence of the laser, and the intensity distribution over the cross section of the beam. The laser is monitored by a high-speed gold-doped germanium detector. The time dependence of the laser pulse is shown in Fig. 1. The intensity distribution across the beam is measured by scanning with the same detector. The half-power diameter is 2.2 mm and its radial distribution is close to Gaussian.

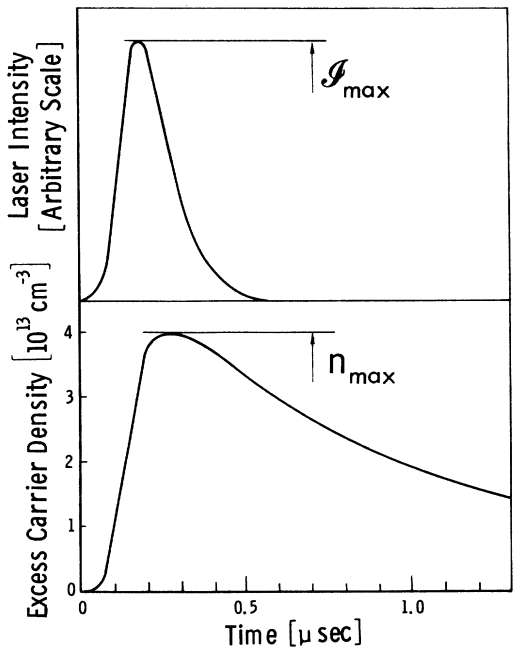

FIG. 1. Irradiation intensity and excess carrier density as a function of time.

The size of the sample surface facing the laser radiation is usually 1×1 mm. This dimension, compared with the half-power diameter of the laser, shows that the peak radiation intensity close to one of the edges is 15% lower than in the center of the sample, assuming perfect alignment. This deviation from uniformity means the measured transition rate is smaller than the actual rate by an estimated 10–20%. We obtain this estimate by an approximate numerical integration of $\mathcal{G}^2$ over the 1-mm^2 surface and comparison with $(\mathcal{G}_{max})^2 \times 1$ mm^2.

The InSb sample is immersed in either liquid N_2 or He. In the latter case, to eliminate He boiling and the consequent laser-beam scattering, the temperature is reduced below the λ point.

The excess density is determined by measuring the conductivity of the samples. The choice of n-type InSb is dictated by the fact that more nearly Ohmic contacts can be made on n-type material than on p type for a large range of excess carrier densities.[16] The contact material used is indium containing 1-wt.% tellurium which is alloyed onto the samples using Divco No. 300 rosen flux. The current-voltage relations of the samples at 77 °K show Ohmic behavior. At liquid-He temperatures, the relation becomes Ohmic when the applied electric field exceeds 0.1–0.2 V/cm. This current-voltage relation agrees qualitatively with earlier measurements and is ascribed to hot electron effects.[17] We are not able to correct for this deviation from Ohmic behavior; the resulting error in the excess conductivity may be as high as 50%.[16]

At 77 °K the average mobility for a boule is used to calculate the excess carrier density in a particular sample from the boule. The average value is taken since the size of the samples is too small to apply extra contacts for Hall measurements. At liquid-He temperatures the estimated value of the mobility is 1×10^5 cm^2/V sec.[18] The contribution to the conductivity from the excess holes can be neglected because of the large electron-to-hole mobility ratio in InSb.

B. Results

The excess carrier density in the sample can be described by the following rate equation[16]:

$$\frac{dn}{dt} = K_1 g + K_2 g^2 - n/\tau - \gamma n(n+n_0). \quad (19)$$

Here n is the excess carrier density in the sample; n_0 is the equilibrium carrier density in the conduction band; K_1 is the coefficient of linear generation of excess carriers, a quantity caused by single-photon excitation due to defect or impurity levels; K_2 is the proportionality factor between total transition rate per unit volume and the square of the laser intensity; $K_2 g^2 = W/V$; τ is the monomolecular recombination time constant, and γ is the bimolecular recombination coefficient. Equation (19) shows that for low laser intensities the excess carrier density is linearly proportional to the laser radiation intensity. As the intensity is increased, the term $K_2 g^2$ exceeds the term $K_1 g$ and a quadratic dependence between n and g results until γn^2 becomes appreciable compared with $n(1/\tau + \gamma n_0)$. This general behavior of n as a function of g is what we measure.

In this work we are concerned primarily with the value of K_2, therefore only the quadratic region is of interest. In this region we may neglect the first and the last term on the right-hand side of Eq. (19). The term n/τ may be neglected at liquid-N_2 temperatures because the measured time constant τ for the sample used is 730 nsec,[19] and the half-power full width of the laser pulse Δt is much shorter, 75 to 150 nsec. At liquid-He temperatures, neglecting n/τ is even more justifiable, since the excess carriers have a longer lifetime at these temperatures than at 77 °K. Figure 1 shows that neglecting the recombination terms is a good approximation, since the excess carrier density does not start to decrease until almost the entire laser pulse has passed. Thus Eq. (19) becomes simply

$$\frac{dn}{dt} = K_2 g^2. \quad (20)$$

Under these circumstances it is trivial to integrate Eq. (20) over the laser pulse. To get an analytical expression we approximate the time dependence of the laser intensity with a Gaussian curve, so

$$K_2 = (8/\pi)^{1/2}(\ln 2)^{1/2} n_{\max}/[\Delta t (g_{\max})^2 (1-R)^2]. \quad (21)$$

Here $n_{\max}$ is the maximum value of the excess carrier density and R is the reflectivity of InSb.

The experimental accuracy is primarily limited by the determination of the peak laser intensity, which is mainly determined by the accuracy of the average power measurement. The accuracy is estimated to be no better than within a factor 1.5 (− 33%, + 50%). Since the experimental determination of K_2 depends quadratically upon the measured value of $g_{\max}$, the experimental value of K_2 has an accuracy of approximately − 50%, + 100%. At 77 °K this large inaccuracy in the determination of $g_{\max}$ is the dominant source of error. Two other important error sources stem from the slightly inhomogeneous illumination of the sample and the neglect of the multiple reflections within the sample. Neither of these will change the − 50%, + 100% error significantly. At 2 °K the hot electron effects and the uncertainty in the value of the electron mobility may increase the error significantly.[16]

Since the data reduction is rather time consuming, only ten sets of data have been analyzed to yield a value for K_2. The analyzed data are for n-type boules at two temperatures and for two laser wavelengths. The lowest and highest values obtained from the analyzed data for each experimental condition are listed in Table II.

IV. DISCUSSION

A. Comparison between Calculated Transition Rates

Basov's expression $W_{i,\text{Basov}}$ given in his Eq. (5) for the two-photon transition rate between the ith valence band and the conduction band is used by Danishevskii[2] to obtain a theoretical estimate of the rate in InSb. We therefore compare $W_{i,\text{Basov}}$ with Eq. (16) in the present paper. To make this comparison, $W_{i,\text{Basov}}$ first has to be reduced by a factor of 3×16. The factor 16 arises because Basov apparently employs the peak value of the vector potential, instead of half of this value, and the factor 3 results from averaging the vector product $(\vec{k} \cdot \hat{\alpha})^2$ over all directions in k space.[20]

After this correction is made, we compare Basov's and our results for the transition rate by dividing the two expressions by each other:

$$W_{i,\text{Basov}}/W_{vi\rightarrow c} = 5[\,|\,(\vec{p}\cdot\hat{\alpha})_{c,vi}\,|^2/(C_i Q)^2].$$

The factor of 5 results from Basov's assumption that the matrix element $(\vec{p}\cdot\hat{\alpha})_{c,vi}$ is independent of the angle between $\vec{k}$ and $\hat{\alpha}$. Table I shows the angular dependence of the matrix elements $P_{i,n}$. The average of $(\vec{k}\cdot\hat{\alpha})^2$ over all directions in k space multiplied by the maximum value of the matrix element squared is five times greater than the average of the product $(\vec{k}\cdot\hat{\alpha})^2 \,|\, P_{i,n}\,|^2$. The value $|\,(\vec{p}\cdot\hat{\alpha})_{c,vi}\,|^2$ used by Basov is slightly more than two times the value of $(C_i Q)^2$. The corrected expression for $W_{i,\text{Basov}}$ is therefore one order of magnitude larger than $W_{vi\rightarrow c}$.

A factor 2 is apparently missing in front of the constant k_2 in Eq. (1) in Danishevskii's paper, and $I(x)$ in his Eq. (3) should read $I(x)^2$. The relation between Danishevskii's "two-photon cross-section" W, hereafter designated by W_D, and the two-photon creation rate coefficient K_2 defined by Eq. (19), is obtained from the continuity equation

$$W_D = (c/\sqrt{\epsilon})(\hbar\omega)^2 K_2, \tag{22}$$

or, since $K_2 g^2 = \sum_i W_{vi\to c}/V$,

$$W_D = (c/\sqrt{\epsilon})(\hbar\omega/g)^2 \sum_i W_{vi\to c}/V. \tag{23}$$

Danishevskii apparently used the following procedure to obtain his expression for W_D: Basov's equation for $W_{i,\text{Basov}}$ is divided by 16 to correct for Basov's use of the peak value of the vector potential. Then $|P_{c,vi}/m|^2 = E_g/(2m_c)$ is used instead of three times this value to correct for Basov's failure to average $(\vec{k}\cdot\hat{\alpha})^2$ over all directions in k space. Finally, Danishevskii apparently multiplied by 2 to account for the two valence bands and used the relation given by Eq. (23). His Eq. (13) then reads

$$W_D = \frac{2^{9/2}\pi e^4 m_{cv}^{1/2} E_g (2\hbar\omega - E_g)^{3/2}}{(\hbar\omega)^4 \epsilon^{3/2} c m_c}.$$

From this expression Danishevskii calculated $W_D = 1.5\times10^{-16}$ cm^2 for $T \approx 80$ °K and 10.6-μm laser irradiation. With $E_g = 0.228$ eV, $W_D = 1.5\times10^{-16}$ cm^2 corresponds to $K_2 = 0.57$ cm sec/erg^2. This K_2 is one order of magnitude larger than the value calculated in Sec. II for the same temperature and laser wavelength (see Table II).

B. Experimental Transition Rates

The calculated and measured values for the transition rate obtained in the present work are given in Table II for two different temperatures and two irradiation wavelengths. The agreement between our theoretical and experimental values is within the accuracy of the experiment at 77 °K. One of the measured values at 2 °K is larger than expected, however, as pointed out in Sec. III, at this temperature there are large possible sources of error in the measurements. Table II shows that the small change in photon energy caused by changing the laser wavelength from 9.6 to 10.6 μm has a large effect on the two-photon creation rate coefficient K_2. From the measured values of K_2 at these two wavelengths, we see that this effect is also clearly demonstrated experimentally.

At 2 °K a region with quadratic dependence between excess carrier density and incident 10.6-μm laser irradiation could not be obtained.[15] This indicates that the quadratic creation term in Eq. (19) is absent, i.e., $K_2 = 0$. The theoretical value is also zero, as shown in Table II, because the band-gap energy at 2 °K is sufficiently large so that the 10.6-μm laser irradiation is below cutoff for the two-photon transition.

Danishevskii reports a measured value for W_D at 80 °K with 10.6-μm irradiation wavelength, namely, $W_D = (8\pm2)\times10^{-17}$ cm^2. This corresponds to $K_2 = (0.3\pm0.08)$ cm sec/erg^2. Danishevskii's result does not agree with our measurements, since we obtain $K_2 \approx (0.032-0.064)$ cm sec/erg^2 at 77 °K and with 10.6-μm laser irradiation. We have no explanation for this discrepancy. At the present time these appear to be the only results available for the two-photon transition rate in InSb.

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