Nonlinear optical properties of InSb: Hot-electron effects

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At high-power densities ($\gtrsim 2$ MW/cm^2) the temporal dependence of transmitted 10.6-μm pulses in InSb shows strong pump-depletion effects. The phenomena are on a 30-nsec time scale and are interpreted as due to hot-electronic kinetics and electron-hole excitation processes. A model calculation employing rate constants from independent measurements reported in the literature yields good agreement with the experimental time evolution of the transmitted signal.

I. INTRODUCTION

The output efficiency of InSb spin-flip lasers decreases rapidly with increasing pump power. Until recently this effect has been attributed to saturation of the InSb spins. However, recent studies of the time dependences of both the spin-flip output and the transmitted pump beam show unusual behavior, which is not compatible with a simple spin-saturation explanation.[1] In the present paper these effects are explained in terms of hot-electron kinetics. For simplicity, we address in this paper only the characteristics in zero magnetic field. Since the temporal dependence of the transmitted 10.6-μm CO_2 laser pump beam is essentially independent of magnetic field strength, we believe that this zero-field calculation explains the qualitative characteristics of hot-electron phenomena in InSb spin-flip lasers. The spin states and their H dependences play no essential role in the attenuation of the pump.

In a recent study of 12-μm spin-flip emission from InSb at ~4 °K, the time dependence of the 10.6-μm radiation transmitted through the sample was also measured.[1] At low incident laser power densities (~0.2 MW/cm^2), the transmitted signal is proportional to the incident laser pulse signal (with a peak at $t \sim 50$ nsec). However, if the incident laser power density is raised to ≥ 2 MW/cm^2 the transmitted radiation rises to a peak in 35–40 nsec and then falls essentially to zero in another 30–35 nsec. After remaining nearly at zero for 150 nsec, the transmission asymptotically returns to its original value. The time dependences of incident and transmitted pulses are shown in Fig. 1. Note that after 70 nsec the incident pump pulse intensity is still at 80% of its peak value, whereas the transmitted signal is nearly zero. This anomalous laser-induced absorption is due, we believe, to energetic conduction electrons produced by the strong laser field inside the sample.[2] In this paper we present a simple theory explaining the laser-induced absorption. A general formalism of laser absorption will be given in Sec. II. In Sec. III we describe a pair-excitation process ("Kane process") which contributes to the indirect laser generation of conduction electrons. The pertinent rate constant is calculated. In Sec. IV we use a simple two-band model with discrete sub-regions to calculate the conduction-electron population. The results of numerical calculations are discussed in Sec. V.

II. FORMALISM

Assume that $I_i(t)$ is the incident laser pulse intensity. Our pulse has a peak value I_0 at 50 nsec and a long tail (see Fig. 1). $RI_i(t)$ is the reflected laser pulse intensity. R is the reflection coefficient of the sample (includes net effect of en-

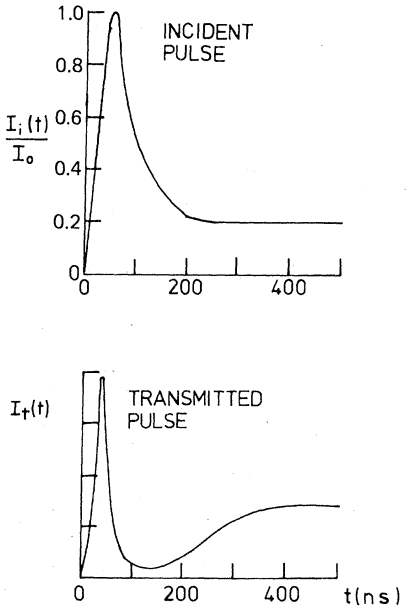

FIG. 1. Incident pulse and transmitted pulse profiles. Experimental result of 2.2-MW/cm^2 incident pulse.

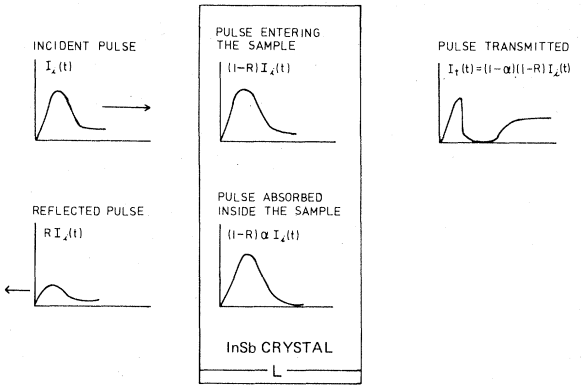

FIG. 2. Schematic representation of the transmission of CO_2 laser pulse through an n-InSb sample.

trance and exit faces and multiple reflections). $(1-R)I_i(t)$ is the laser pulse intensity inside the sample. $\alpha(1-R)I_i(t)$ is the laser energy loss per unit time inside the sample. α is the absorption coefficient of the sample. $I_t(t)=(1-\alpha)(1-R)I_i(t)$ is the transmitted pulse intensity. This is summarized in Fig. 2.

We will neglect all surface effects except reflection and assume that it is accounted for by an intensity-independent reflectivity. The optical properties can be described in terms of the complex dielectric constant ϵ and the complex index of refraction

$$N=\sqrt{\epsilon}=n+iK\,. \tag{1}$$

The coefficients R and α can be calculated by considering the sample as having two parallel plane surfaces of distance L (the thickness) and neglecting the coherence. The results are

$$R=R_\infty\{1+e^{-2\tilde{K}L}(1-R_\infty)^2/[1-e^{-4\tilde{K}L}R_\infty^2]\} \tag{2a}$$

and

$$\alpha=(1-e^{-2\tilde{K}L})(1+e^{-2\tilde{K}L}R_\infty)/(1-e^{-4\tilde{K}L}R_\infty)\,, \tag{2b}$$

where

$$\tilde{K}=K\omega/c$$

and

$$R_\infty=\left|(1-N)/(1+N)\right|^2 \tag{3}$$

is the reflection coefficient for a sample of infinite thickness. To calculate ϵ, we assume that the absorption is due to free electrons in the conduction band; the electrical conductivity is

$$\sigma=\sigma_0/(1-i\omega\tau)\,, \tag{4}$$

where ω is the angular frequency of laser light; τ is the collision time of electrons in the conduction band; and the low-frequency electric conductivity is

$$\sigma_0=ne^2\tau/m^*\,. \tag{5}$$

(We assume that τ is independent of location in the conduction band.) In Eq. (5), n is the number of electrons in conduction band which contribute to absorption; m^* is the effective mass of the conduction electron (assumed constant over the populated region of the conduction band); and e is the electronic charge. Neglecting other polarization effects, we have the complex dielectric constant

$$\epsilon=\epsilon_\infty[1-(\omega_p^2/\omega^2)(1+i/\omega\tau)^{-1}]\,, \tag{6}$$

where

$$\omega_p=(4\pi ne^2/\epsilon_\infty m^*)^{1/2} \tag{7}$$

is the plasma frequency due to the free electrons in the conduction band, and ϵ_∞ is the optical dielectric constant (15.68 for InSb).

In this simple picture, the absorption coefficient α is an increasing function of the free-electron density n, and the enhanced absorption is due to the increased number of electrons in the conduction band. A fundamental problem is the explanation of the optical excitation of these extra electrons. These electrons must come from the valence band; however, at low temperatures, the band gap $E_g\sim 1900$ cm^{-1} is more than twice as large as the photon energy ($\lambda\simeq 10.6$ μm, $1/\lambda\sim 944$ cm^{-1}), so that direct particle-hole excitation due to single-photon or two-photon absorption is insignificant.[1] We will propose another possible particle-hole pair excitation process in Sec. III.

III. PAIR-EXCITATION PROCESS

In n-type InSb samples of Ref. 1 there exists an electron density of 10^{16} cm^{-3} at the bottom of the conduction band. These electrons will be excited to higher energy states in the conduction band due to absorption of photons from the incident laser beam. Those electrons excited to the states with energy $>2E_G$ can decay back to the bottom of the conduction band by exciting an additional electron from the valence band to the conduction band [due to the electron-electron Coulomb interactions (Fig. 8)]. This kind of pair-excitation process had been previously studied by Kane[2] for Si. In this section we estimate the pair-excitation rate for InSb. Because of the difference in density of states, the main contribution to this process is from the heavy-hole band rather than the light-hole band. Our model therefore neglects light holes.

Consider a two-band model. The Hamiltonian is

$$\hat{H}=\hat{H}_0+\hat{H}_1\,, \tag{8}$$

where

$$\hat{H}_0 = \sum_{\vec{k}\alpha} E_{k\alpha} \hat{C}^{\dagger}_{\vec{k}\alpha} \hat{C}^{\dagger}_{\vec{k}\alpha} \tag{9}$$

is the single-particle Hamiltonian and α is the band index (c for conduction band and v for the heavy-hole valence band). The electron-electron Coulomb interaction is

$$\hat{H}_1 = \sum_{\vec{k}\alpha} \sum_{\vec{k}'\alpha'} \sum_{\vec{p}\beta} \sum_{\vec{p}'\beta'} V_{k'\alpha' p'\beta', k\alpha p\beta} \times \hat{C}^{\dagger}_{\vec{k}'\alpha'} \hat{C}^{\dagger}_{\vec{p}'\beta'} \hat{C}_{\vec{p}\beta} \hat{C}_{\vec{k}\alpha} , \tag{10}$$

from which we obtain in Appendix A an expression for the lifetime τ_k of a highly energetic electron against decay by the Kane process.

Since we are interested in only the order of magnitude of the rate of pair excitation, we neglect the exchange term in (A15). We take the energy zero point at the top of valence band: then as an approximation, $E_{pv} \simeq 0$, because the effective mass of the heavy-hole valence band $m_v^* \simeq 0.5 m_e$ is much greater than the conduction electron mass $m_c^* \simeq 0.014 m_e$. Then we have

$$\frac{1}{\tau_k} \simeq \frac{\pi}{2\hbar} |I_{cv}(\vec{k})|^2 \sum_{\vec{k}'} |\tilde{V}(\vec{k} - \vec{k}')|^2 \times N_c(E_{kc} - E_{k'c}) , \tag{11}$$

where I_{cv}, the valence–conduction-band transition matrix element, is defined in Appendix A, Eq. (A16); and

$$N_c(E) = (1/2\pi^2)(2m_c^*/\hbar)^3 (E - E_G)^{1/2} \tag{12}$$

is the density of states of the conduction band. (We neglect the nonparabolic conduction band structure here.) The Fourier transform of the interaction potential is the screened Coulomb potential

$$\tilde{V}(\vec{q}) = (1/\epsilon_\infty) 4\pi e^2/(q^2 + k_s^2) , \tag{13}$$

where ϵ_∞ is the optical dielectric constant and the screening constant is

$$k_s = [4\pi n e^2/(E_F - E_G)]^{1/2} . \tag{14}$$

E_F is related to the conduction electron density n by

$$E_F = E_G + (\hbar^2/2m_c)(3\pi^2 n)^{2/3} . \tag{15}$$

Substituting Eqs. (12) and (13) into (11), we obtain

$$1/\tau_k = (2m_c^* e^4/3\pi\hbar^3 \epsilon_\infty^2) |I_{cv}(\vec{k})|^2 f(y, y_F) , \tag{16}$$

where

$$y = E_k/E_G \tag{17}$$

and

$$y_F = E_F/E_G . \tag{18}$$

From (15), we know that y_F is a number greater than 1. The function f is the integral

$$f(y, y_F) = \int_{x_-}^{x_+} dx \frac{x[(x_+ - x)(x - x_-)]^{3/2}}{(x^2 + x_s^2)^2} , \tag{19}$$

where

$$x_\pm = 1 \pm (1 - x_0^2)^{1/2} , \tag{20}$$

$$x_0 = 1/(y - 1)^{1/2} , \tag{21}$$

and

$$x_s = \frac{2e^2}{\pi\hbar} \left(\frac{2m_c^*(y_F - 1)}{E_G} \right)^{1/2} \frac{1}{y - 1} . \tag{22}$$

Mathematically, for a real solution f, we require $x_\pm$ to be real and $x_0 < 1$. Then, from (21), we get $y > 2$. This means that the pair-excitation process cannot occur unless the electron is in a highly excited state with energy $E_k > 2E_G$. Physically, this is simply a result of energy conservation. The function $f(y, y_F)$ is an increasing function of $y = E_K/E_G$ and is of the order of magnitude of unity for $y \gtrsim 3$. $|I_{cv}(\vec{k})|^2$ is ≤ 1 for $\vec{K} \cdot \vec{r} \sim 1$. Then the pair-excitation rate parameter can be estimated from (16) to be

$$g_p \leq m_c^* e^4/\pi\hbar^3 \epsilon_\infty^2 \sim 5 \times 10^{11} \text{ sec}^{-1} \sim 5 \times 10^2 \text{ nsec}^{-1} . \tag{23}$$

This rate is faster than the ordinary conduction-electron relaxation rate[3] (10^{10} sec^{-1}) or the conduction–valence-band recombination rate (10^8 sec^{-1}) in this sample. (This will be justified and discussed in Sec. V.)

IV. SIMPLE RATE-EQUATION MODEL

It is convenient to separate the conduction band into three regions: a low-energy region c and an intermediate-energy region b, each with width $\sim\hbar\omega$; and a high-energy region a. Before the laser pulse ($t < 0$), all conduction electrons are in the bottom states of the conduction band, i.e., $n_a = n_b = 0$, $n_c = n_c^0 = n_0 AL$, where $n_0 = 10^{16}$ cm^{-3}, A is the cross-sectional area of the laser beam, and L is the sample thickness. After the laser pulse ($t \geq 0$) the conduction electrons in the lower-energy regions will be excited to the higher-energy regions, and there will be pair-excitation processes. There is a finite momentum change for a conduction electron decay from region a to c. This means the pair-excitation process will also involve a finite momentum change for an electron excited from the valence band to the conduction band (region c) because of momentum conservation. Furthermore, the recombination process of conduction electrons (in region c) to the valence band will involve negligible momentum change, because of the small momentum of the emit-

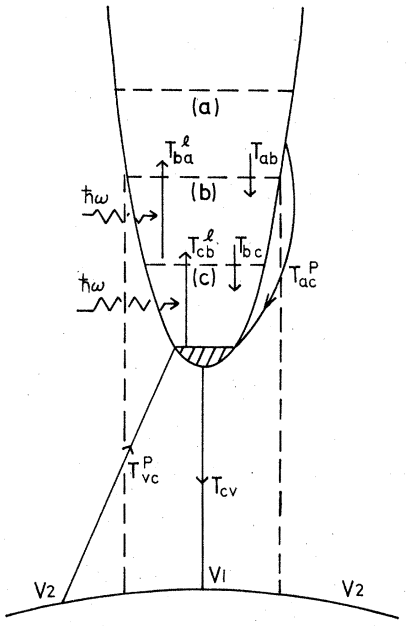

FIG. 3. Possible transitions in the two-band model of electronic states in InSb crystal in CO_2 laser field.

ted photon. Thus, it is a good approximation to separate the valence band into two regions: the region $v1$ extending to the wave vectors spanned by conduction band region c, and the region $v2$ outside the region $v1$. Then we may write the electron population rate equations as follows: For $t \geq 0$,

$$\dot{n}_a(t) = T^I_{ba} - T_{ab} - T^P_{ac}, \tag{24a}$$

$$\dot{n}_b(t) = -T^I_{ba} + T^I_{cb} + T_{ab} - T_{bc}, \tag{24b}$$

$$\dot{n}_c(t) = -T^I_{cb} + T_{bc} - T_{cv} + T^P_{ac} + T^P_{vc}, \tag{24c}$$

$$\dot{n}_{v1}(t) = T_{cv} - T_{12}, \tag{24d}$$

$$\dot{n}_{v2}(t) = -T^P_{vc} + T_{12}, \tag{24e}$$

where $n_a(t)$, $n_b(t)$, $n_c(t)$, $n_{v1}(t)$, and $n_{v2}(t)$ are the number of electrons in the three regions a, b, c of the conduction band and regions $v1$ and $v2$ of the valence band at given time t. T^I_{ba} (T^I_{cb}) is the rate of electron excitation from region b to region a (c to b) due to the single-photon absorption process. T_{ab}, T_{bc}, T_{12}, and T_{cv} are the relaxation rates for electron transitions from a to b, b to c, $v1$ to $v2$, and c to $v1$, respectively. T^P_{ac} and T^P_{vc} are the transition rates for pair excitation. The physical situations are shown in Fig. 3. We neglect here multiple-photon absorption processes (which are much slower than the single-photon absorption process). From the law of particle conservation, n_a, n_b, n_c, n_{v1}, and n_{v2} satisfy the following relation:

$$n_a(t) + n_b(t) + n_c(t) + n_{v1}(t) + n_{v2}(t) = 0 + 0 + n^0_c + N_{v1} + N_{v2}, \tag{25}$$

where at $t = 0$, $n_a = n_b = 0$, $n_c = n^0_c$, $n_{v1} = N_{v1}$, $n_{v2} = N_{v2}$, N_{v1} and N_{v2} denote the total number of electronic states in the two regions of the valence band. For the pair-excitation process,

$$T^P_{ac} = T^P_{vc} = T^P. \tag{26}$$

Because the pair-excitation process is very fast, it is expected that

$$n_a \ll n_b < n_c. \tag{27}$$

and $\dot{n}_a \ll \dot{n}_b, \dot{n}_c$. It is a good approximation to assume $\dot{n}_a \simeq 0$ for our calculation, i.e.,

$$T^P \approx T^I_{ba} - T_{ab}. \tag{28}$$

Then the rate equations [(24a)–(24e)] can be simplified as

$$\dot{n}_{v1} = T_{cv} - T_{12}, \tag{29a}$$

$$\dot{n}_{v2} = -T^I_{ba} + T_{ab} + T_{12}, \tag{29b}$$

$$\dot{n}_b = T^I_{cb} - T^I_{ba} + T_{ab} - T_{bc}, \tag{29c}$$

and

$$\dot{n}_c = -\dot{n}_{v1} - \dot{n}_{v2} - \dot{n}_b. \tag{29d}$$

Because of (27), the photon absorption due to electrons in region a is negligibly small in comparison with that due to electrons in regions b and c. Then we have

$$T^I_{ba} + T^I_{cb} \simeq \alpha(1-R)I_t(t)A, \tag{30}$$

where A is the cross-sectional area of the laser pulse beam. The absorptivity α is an increasing function of the density $n_{bc} = (n_b + n_c)/AL$ which contributes to the laser aborption. Hence, the effective static electrical conductivity in Eq. (5) can be approximated as

$$\sigma_0 \simeq (n_{bc}e^2/m^*)\tau. \tag{5'}$$

The effective electron density n_{bc} varies according to the rate equation

$$\dot{n}_{bc} = -\dot{n}_{v1} - \dot{n}_{v2} = T^P - T_{cv} \simeq T^I_{ba} - T_{ab} - T_{cv}. \tag{31}$$

This density is increased due to the generation of electrons from the valence band by the pair process T^P and decreased due to the recombination T_{cv}. The various rates are taken to be given by the following physically reasonable expressions:

$$T^I_{ba} = \gamma_I[n_b(N_a - n_a) - n_a(N_b - n_b)], \tag{32a}$$

$$T^I_{cb} = \gamma_I[n_c(N_b - n_b) - n_b(N_c - n_c)], \tag{32b}$$

$$T_{ab} = \gamma_c n_a(N_b - n_b), \tag{32c}$$

$$T_{bc} = \gamma_c n_b(N_c - n_c), \tag{32d}$$

$$T_{12} = \gamma_v n_{v1}(N_{v2} - n_{v2}), \tag{32e}$$

and

$$T^P = \gamma_p n_a (N_c - n_c)^2 n_v, \tag{32f}$$

where γ_I is the transition probability due to single-photon absorption, γ_c (γ_v) is the relaxation rate constant of electrons in the conduction (valence) band, and γ_p is the pair excitation rate constant. N_a, N_b, and N_c (N_{v1}, N_{v2}) are the total number of electronic states in the three (two) regions of the conduction (valence) band.

Because of momentum conservation, the recombination rate between the conduction band and valence band can be written

$$T_{cv} \lesssim \text{const} \times \text{smaller value of } \begin{cases} n_c, \\ N_{v1} - n_{v1} \end{cases}. \tag{33}$$

For the physical situation of the InSb experiment considered here, $n_c \gg N_{v1} - n_{v1}$; it is a good approximation to write

$$T_{cv} \approx \gamma_{cv}(N_{v1} - n_{v1}), \tag{34}$$

where γ_{cv} is a constant.

For numerical calculations we define the transition ratio

$$x = T^I_{ba}/T^I_{cb} = (N_a n_b - N_b n_a)/(N_b n_c - N_c n_b). \tag{35}$$

Then

$$T^I_{ba} = [x/(1+x)]\alpha(1-R)I_i(t)A \tag{36a}$$

and

$$T^I_{cb} = [1/(1+x)]\alpha(1-R)I_i(t)A, \tag{36b}$$

where A is the laser-beam cross-section area. From the relations (28), (32d), and (32f) the electron density $n_a (\ll n_b, n_c)$ can be calculated

$$n_a = T^I_{ba}/[\gamma_p(N_c - n_c)^2 n_v + \gamma_c(N_b - n_b)]. \tag{37}$$

V. NUMERICAL RESULTS

Using the preceding equations we have calculated the transmitted pulse intensity versus time. The results are shown in Fig. 4. The theoretical curve involves six parameters. However, these parameters may be calculated or at least estimated independently.

The first quantity g_I is given by

$$g_I = I_0 A/n_c^0, \tag{38}$$

where I_0 is the peak value of the incident pulse intensity $I_i(t)$; A is the cross-sectional area of the sample normal to the beam, and n_c^0 is the number of conduction electrons in the absence of the beam. In Fig. 4 the theory uses[1] $g_I = 24$ nsec^{-1} (i.e., 2.2 MW/cm^2).

g_I is a good characteristic parameter which can be adjusted to test the degree of nonlinearity. In our theory, the absorption or transmission coefficient is dependent on the electron population; the more conduction electrons, the more absorption and then the less transmission. This is shown in Fig. 5. For a comparison with experiment, we have calculated the curve for the low intensity case ($\langle I_i \rangle = 270$ kW/cm^2, i.e., $g_I = 3$), where very few electrons are generated by the pair-production processes, hence everything is linear and no nonlinear absorption effects can be observed. This is shown in Fig. 6.

A second parameter is τ, the collision time in Eq. (5). We assume that τ is given by a mean free path divided by the electron velocity

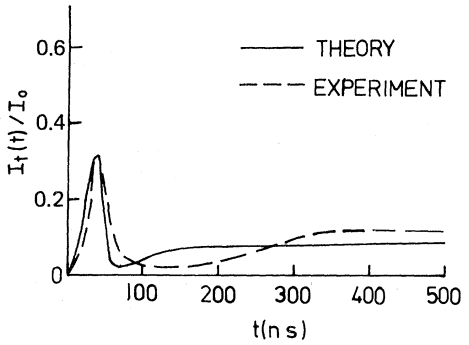

FIG. 4. "Best fit" to the experimental result ($I_i = 2.2$ MW/cm^2). The parameters in the theoretical curve are $g_I = 24$, $g_p = 500$, and $g_c = 0.22$, $g_r = 0.3$, $g_v = 8 \times 10^{-4}$ (units of nsec^{-1}) and $\omega\tau = 2000/(n/n_0)^{1/3}$. The experimental curve is scaled the same as the theoretical curve at the peak position.

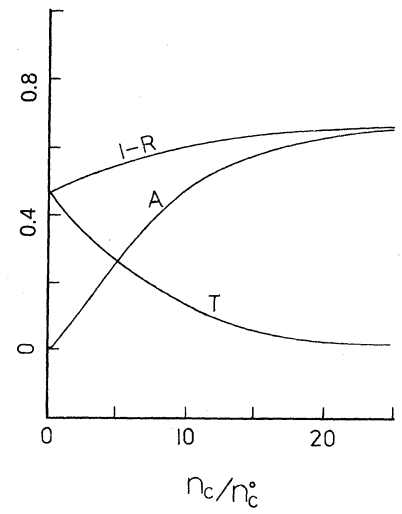

FIG. 5. Conduction-electron population dependence of the coefficients of reflection R, absorption A, and transmission T.

$$v=[2(E-E_G)/m^*]^{1/2}\,. \tag{39}$$

Since $(E-E_G)$ varies as $n^{2/3}$, we have

$$\tau \propto n^{-1/3}\,. \tag{40}$$

Of course n changes as a function of time during the pulse, and so does τ. Numerical computations which ignored this implicit time dependence of τ and instead treated τ as an adjustable but time-independent constant gave poorer agreement with experiment. Figure 4 uses a value of

$$\omega\tau = 2000(n/n_0)^{-1/3}\,. \tag{41}$$

This numerical value is taken from Kahlert and Bauer[4]; the functional form agrees with Eq. (40).

The four remaining parameters are as follows: g_r, which is the same as γ_{cv} in Eq. (34), is a radiative recombination-rate constant known to be $\sim 3\times 10^8$ sec^{-1} at 4 °K from the work of Fossum and Ancker-Johnson.[3] (Figure 4 uses this value of 0.3 nsec^{-1}.) g_c is the conduction-electron relaxation parameter equal to $\gamma_c n_c^0$. The fit in Fig. 4 with $g_c = 0.22$ nsec^{-1} corresponds to a conduction-electron relaxation time of 10^{-10} sec. This time τ_e for conduction-electron energy relaxation must be *long* compared to Kahlert and Bauer's[3] 10^{-12} sec, since that is a momentum-changing time, and most momentum-changing collisions do not change the energy. We further note that the time for an electron to leave region b is characterized by multiple- (not single-) phonon emission. Each phonon emission time is of the order of 10^{-12} sec, and each emission lowers the electron energy by roughly 0.01 eV. Thus approximately 30–50 phonon emissions are required to remove an electron from region b. The time τ_e must also be *short* compared with the value of 10^{-7} sec from Whalen and Westgate,[5] since their value corresponds to a physical situation in which no electron–optical-phonon relaxation is possible (only acoustical). Under the nonthermal conditions of hot electrons and hot phonons present in the experiments analyzed here, an estimate of the number of distribution of optical phonons is beyond the scope of the present paper, but τ_e must be such that $10^{-7} \gg \tau_e \gg 10^{-12}$ sec. Our value 10^{-10} sec is thus consistent with the physical requirements of the problem, within the limitations of a model employing a small number of discrete region to represent a continuum of electron states. For a further theoretical justification of this value, we make a microscopic numerical calculation of the optical-phonon-induced relaxation time, by assuming a time-dependent Fermi-Dirac distribution of conduction electron with temperature $T(t)$ and Fermi energy $\epsilon_F(t)$ (see Appendix B). In this calculation we have taken into account the time-dependent static screen effect. We find that the conduction electron relaxation time τ_e (defined as the average time that every conduction electron will lose energy $\hbar\omega$, i.e., an electron will relax from region a to b or b to c by emitting optical phonons to the crystal at low temperature) is exactly of the order of 10^{-10} sec, in agreement with our emperical constant g_c here. g_v is the valence-band hole replacement rate given by $\gamma_v n_c^0$. Our value $g_v = 8\times 10^{-4}$ nsec^{-1} used in Fig. 4 corresponds to a hole replacement time of 100 nsec. We know of no independent measurement of this parameter, and indeed its numerical value may be an artifact of our two-band model, which ignores the light-hole band. However, the physical meaning of g_v is a replacement rate for electron near the top of the valence band; thus, we expect the ratio g_c/g_v to scale not as the electron-to-hole lifetime ratio $(\tau_e/\tau_h \approx 1)$ but as the conduction-electron–to–heavy-hole-mobility ratio, which is $\sim 10^2$ due to the masses involved. Thus, the ratio of $g_c/g_v \approx 10^2$ used in Fig. 4 is in accord with the best estimate based on independent data. Figure 7 shows the effect of increasing or decreasing g_v by 10. Since the physi-

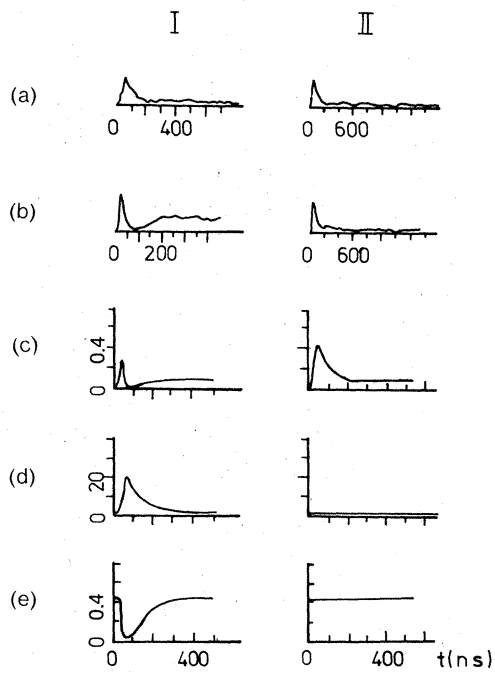

FIG. 6. Degree of nonlinearity and comparison between experiment and theory for the two cases: (I) $\langle I_i\rangle = 2.2$ MW/cm^2 and (II) $\langle I_i\rangle = 270$ kW/cm^2. (a) Experimental incident pulse profiles (Ref. 1). (b) Experimental transmitted pulse profiles (Ref. 1). (c) Theoretical transmitted pulse profiles $[I_t(t)/I_0]$. (d) Theoretical conduction-electron population profiles $[n_c(t)/n_c^0]$. (e) Theoretical transmission coefficient (T).

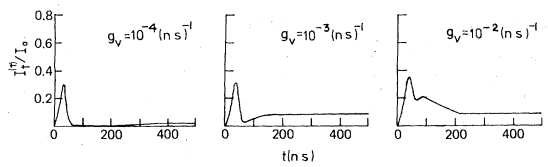

FIG. 7. Intra-band electron relaxation effect of heavy-hole band. The common parameters are $g_l=24$, $g_p=10^2$, $g_c=22$, $g_r=0.3$ (units of nsec^{-1}) and $\omega\tau=2000/(n_{bc}/n_0)^{1/3}$.

cal effect of small g_v is to prevent hole mobility and reduce the recombination processes by shutting off the supply of electrons near the top of the valence band, values of $g_v \gg 10^{-2}$ nsec^{-1} give only a small pump-depletion effect. For large values of g_v, more electrons can relax from region $v1$, causing electrons to decay radiatively from conduction-band region c and reducing the anomalous absorption of conduction electrons. The final parameter g_p, the Kane pair generation rate parameter, was calculated as $\sim 5\times 10^2$ nsec^{-1} in Eq. (23). This is the number used in Fig. 4. The calculation is not particularly sensitive to this number, as long as it corresponds to a characteristic time (in this case 10^{-11} sec) which is fast compared to all other relaxation times.

Thus, in summary, the six parameters used to calculate Fig. 4 are in no sense freely adjustable, and in all cases appear in agreement with either a calculation (for g_p) or independent values taken from the literature.

VI. DISCUSSION AND CONCLUSIONS

In this paper we have presented a very simple phenomenological theory to explain the anomalous absorption of high-power 10.6-μm laser radiation in InSb at low temperature. We consider the contribution due to an enhanced electron concentration in the conduction band which is generated indirectly by excitation from the valence band by means of the electronic Coulomb interaction.[2] Qualitatively, the results of simple model numerical calculations agree with the experimental results quite well. Quantitatively, we do not have an "exact" fit with the experimental curve because of our simple models and choice of parameters. However, our collision time agrees quantitatively with Kahlert and Bauer's calculation,[4] and other parameters seem equally plausible.

Some limitations of the present work are listed below:

First, the conductivity of holes in valence bands is neglected in comparison with that of electrons in the conduction band. This approximation is good only when the number of holes is much smaller than that of conduction electrons or the hole mobility is much smaller than the electronic mobility—this is good for heavy holes but not for light holes.

Second, this is only a phenomenological theory which separates the conduction band into three regions. A microscopic calculation is needed for an exact fit with the experimental result.

Third, we use the free-electron model (with effective mass $m\simeq 0.014m_e$) for the electric conductivity in the calculation. We have neglected the nonparabolicity of the conduction band in InSb.

Fourth, the T's, the transition rate parameters in the rate equations, are all phenomenological; there is no microscopic derivation here.

We plan to return to these and other points in later publications.

ACKNOWLEDGMENT

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APPENDIX A: PAIR-EXCITATION RATE CALCULATION

Assume

$$V_{\vec{k}'\vec{k}\vec{p}'\vec{p},\alpha'\alpha\beta'\beta}=\frac{1}{2}\int d^3r\,\phi^*_{\vec{k}'\alpha'}(\vec{r})\phi_{\vec{k}\alpha}(\vec{r}) \times\int d^3r'\phi^*_{\vec{p}'\beta'}(\vec{r}')\phi_{\vec{p}\beta}(\vec{r}) \times V(|\vec{r}-\vec{r}'|), \tag{A1}$$

where $\phi_{\vec{k}\alpha}$ is the single-particle wave function of state $\vec{k}$ in band α; the two-particle interaction can be written in the Fourier form

$$V(|\vec{r}-\vec{r}'|)=\sum_{\vec{q}}\tilde{V}(\vec{q})e^{i\vec{q}\cdot(\vec{r}-\vec{r}')}. \tag{A2}$$

Substituting (A2) into (A1) we have

$$V_{\vec{k}'\vec{k}\vec{p}'\vec{p},\alpha'\alpha\beta'\beta}=\frac{1}{2}\sum_{\vec{q}}\tilde{V}(\vec{q})I_{\vec{k}'\alpha',\vec{k}\alpha}(\vec{k}-\vec{k}'+\vec{q}) \times I_{\vec{p}'\beta',\vec{p}\beta}(\vec{p}-\vec{p}'-\vec{q}), \tag{A3}$$

where

$$I_{\vec{k}'\alpha',\vec{k}\alpha}(\vec{Q})=\int d^3r\,e^{i\vec{Q}\cdot\vec{r}}\phi^*_{\vec{k}'\alpha'}(\vec{r})\phi_{\vec{k}\alpha}(\vec{r}). \tag{A4}$$

The single-particle wave function can be written in the Bloch form

$$\phi_{\vec{k}\alpha}(\vec{r})=u_{\vec{k}\alpha}(\vec{r})e^{i\vec{k}\cdot\vec{r}}, \tag{A5}$$

with the atomic-wave approximation

$$u_{\vec{k}\alpha}(\vec{r})\approx\frac{1}{\sqrt{N}}\sum_{j=1}^{N}\phi_\alpha(\vec{r}-\vec{R}_j), \tag{A6}$$

where N is the total number of atoms, $\vec{R}_j$ is the lattice point of the jth atom, and $\phi_\alpha(\vec{r}-\vec{R}_j)$ is the

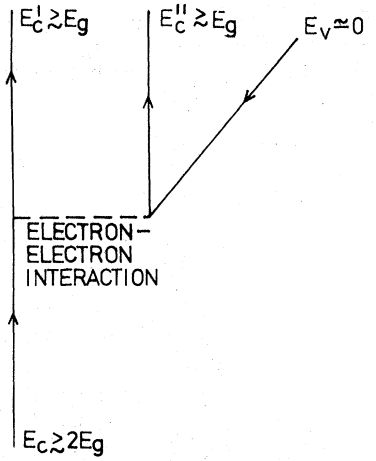

FIG. 8. Pair-excitation process.

atomic wave function about the lattice R_j. In our simple mode, $\alpha = 5s$ for the conduction band and $\alpha = 5p$ for the valence band.

Substituting Eqs. (A5) and (A6) into (A4), we have

$$I_{k'\alpha',k\alpha}(\vec{Q}) = g_{\alpha'\alpha}(\vec{Q})\,\frac{1}{N}\sum_{j=1}^{N} e^{i\vec{Q}\cdot\vec{R}_j}\ , \tag{A7}$$

where

$$\frac{1}{N}\sum_{j=1}^{N} e^{i\vec{Q}\cdot\vec{R}_j} = \delta_{\vec{Q},\vec{K}}\ . \tag{A8}$$

K is a reciprocal-lattice vector, and the transition-matrix element

$$g_{\alpha',\alpha}(\vec{Q}) = \int_{\Omega_a} d^3r\, \phi^*_{\alpha'}(\vec{r}) e^{i\vec{Q}\cdot\vec{r}} \phi_\alpha(\vec{r}) \tag{A9}$$

Ω_a is the atomic volume. We have two cases: (i) intraband transition, $\alpha = \alpha'$, the dominant contribution is at $K = 0$; then we have

$$I_{k'\alpha,k\alpha}(\vec{Q}) = \delta_{\vec{Q}0}\,; \tag{A10}$$

(ii) interband transition, $\alpha \neq \alpha'$, the term $\vec{K} = 0$ will vanish, we have

$$I_{\alpha'\alpha}(\vec{Q}) = g_{\alpha'\alpha}(\vec{Q})\delta_{\vec{Q},\vec{K}}\ . \tag{A11}$$

In this paper, we are interested in the pair-production process where $\alpha = \alpha' = \beta' = c = 5s$ and $\beta = v = 5p$; see Fig. 8. For the model interaction Hamiltonian

$$\hat{H}_1 = \sum_{kk'}\sum_{pp'} V_{k'kp'p}\hat{C}^\dagger_{k'c}\hat{C}^\dagger_{p'c}\hat{C}_{pv}\hat{C}_{kc}\ , \tag{A12}$$

the interaction matrix element is

$$V_{k'kp'p} = \frac{1}{2}\sum_{\vec{q}} \tilde{V}(\vec{q}) I_{k'c,kc}(\vec{k} - \vec{k}' + \vec{q}) \times I_{p'cpv}(\vec{p} - \vec{p}' - \vec{q})\,. \tag{A13}$$

Next, we consider the lifetime τ_k of a high-energy electron due to this pair excitation process. From the Fermi Golden Rule

$$\frac{1}{\tau_k} = \frac{2\pi}{\hbar}\sum_f |\langle f|\hat{H}_1|i\rangle|^2 \delta(E_i - E_f)\,, \tag{A14}$$

and using the results of Eqs. (A10)–(A13), we obtain

$$\frac{1}{\tau_k} = \frac{\pi}{2\hbar}|I_{cv}(\vec{K})|^2 \sum_{k'pp'} \delta(E_{kc} + E_{pv} - E_{k'c} - E_{p'c})\delta_{\vec{p}-\vec{p}'-\vec{k}'+\vec{k},\vec{K}} \times \delta_{\vec{p}-\vec{p}'-\vec{k}'+\vec{k},\vec{K}} n_{\vec{k}c} n_{\vec{p}v}(1 - n_{\vec{k}'c})(1 - n_{\vec{p}'c}) \times [|\tilde{V}(\vec{k}' - \vec{k})|^2 - \tilde{V}^*(\vec{p}' - \vec{k})V(\vec{k}' - \vec{k})]\,, \tag{A15}$$

where

$$I_{cv}(\vec{K}) = \langle c|e^{i\vec{K}\cdot\vec{r}}|v\rangle \tag{A16}$$

is the valence–conduction-band transition matrix element. There are two terms: the first term $\propto|\tilde{V}(\vec{k}' - \vec{k})|^2$ is the direct, and the second term, proportional to $\tilde{V}^*(\vec{p}' - \vec{k})V(\vec{k}' - \vec{k})$, is the exchange term.

APPENDIX B

The hot-electron energy relaxation processes, the relaxations from region a to b and b to c in our model, for example, are principally due to the electron–optical-phonon interaction. To calculate this relaxation time, we start from the definition of total conduction electronic energy

$$U(t) = \sum_k \epsilon_k n_k(t)\,, \tag{B1}$$

where the summation is over the states of conduction band. Then the rate of energy change is

$$\dot{U}(t) = \sum_k \epsilon_k \dot{n}_k(t)\,. \tag{B2}$$

The energy relaxation time τ_e is defined as the average time that each conduction electron will lose energy $\hbar\omega$ (i.e., an electron will relax from region a to b or b to c) by emitting optical phonons to the crystal, i.e.,

$$1/\tau_e = [1/Vn(t)\hbar\omega]\dot{U}(t)\,, \tag{B3}$$

where $n(t)$ is the density of conduction electrons. $\hbar\omega$ is the photon energy of CO_2 laser pulse. At low lattice temperature (4 °K), by considering only the spontaneous phonon emission process, $\dot{n}_k(t)$ can be calculated from the first-order time-dependent theory

$$\dot{n}_k = (1/\tau_k^{\rm in})(1 - n_k) - (1/\tau_k^{\rm out})n_k\,, \tag{B4}$$

where

$$\frac{1}{\tau_k^{\text{in}}}=\frac{2\pi}{\hbar}\sum_q |\alpha_q|^2 n_{k+q}\delta(\epsilon_k-\epsilon_{k+q}+\hbar\Omega_0), \quad \text{(B5a)}$$

$$\frac{1}{\tau_k^{\text{out}}}=\frac{2\pi}{\hbar}\sum_q |\alpha_q|^2(1-n_{k+q}) \times\delta(\epsilon_k-\epsilon_{k+q}-\hbar\Omega_0). \quad \text{(B5b)}$$

α_q is the matrix element of the electron–optical-phonon interaction (it can be calculated that the electron–acoustical-phonon interaction can be neglected in the high-electron-temperature region[4] which is practically the interesting region in this experiment), in the screening form, we have

$$|\alpha_q|^2=(\hbar/Vq^2)(4\pi Fe)^2q^4/(q^2+k_s^2)^2 \quad \text{(B6)}$$

and

$$F^2=(\Omega_0/8\pi)(1/\epsilon_\infty-1/\epsilon_0). \quad \text{(B7)}$$

k_s is the screening constant, which is a fractional of electron density n. Ω_0 is the average optical phonon frequency. V is the volume of the sample. ϵ_∞ and ϵ_0 are the optical and static dielectric constants.

Substituting (B4), (B5a), and (B5b) into (B2), we have

$$\dot{U}(t)=-\frac{2\pi}{\hbar}\sum_k\sum_q |\alpha_q|^2(\epsilon_k-\epsilon_{k+q})n_k(1-n_{k+q}) \times\delta(\epsilon_k-\epsilon_{k+q}-\hbar\Omega_0). \quad \text{(B8)}$$

If we assume the nonequilibrium electronic population to be the time-dependent Fermi distribution,[6] i.e.,

$$n_k(t)=f(\epsilon_k,t)=(e^{[\epsilon_k-\epsilon_F(t)]/k_BT(t)}+1)^{-1}, \quad \text{(B9)}$$

with time-dependent electronic temperature $T(t)$ and Fermi energy $\epsilon_F(t)$. $T(t)$ will be much higher than the lattice temperature (4 °K) during the laser-pulse irradiation.

We assume further that the conduction electrons have the energy dispersion

$$\epsilon_k=\hbar^2k^2/2m_c=\epsilon, \quad \text{(B10)}$$

with the origin at the bottom of conduction band. Substituting (B6)–(B10) into (B3), after angular integration and some algebra, we obtain

$$\frac{1}{\tau_e}=-\frac{2\Omega_0^2m_c^2e^2}{n\pi^2\hbar^4\omega(e^{\hbar\Omega_0/k_BT}-1)}\left(\frac{1}{\epsilon_\infty}-\frac{1}{\epsilon_0}\right) \times\int_0^\infty d\epsilon[f(\epsilon,t)-f(\epsilon+\hbar\Omega_0,t)] \times\int_{(k^2+2m_c\Omega_0/\hbar)^{1/2}-k}^{(k^2+2m_c\Omega_0/\hbar)^{1/2}+k}\frac{q^3}{(q^2+k_s^2)^2}dq. \quad \text{(B11)}$$

The total conduction electron density n and the screening constant k_s are given by

$$n(t)=\int_0^\infty N(\epsilon)\,d\epsilon\,f(\epsilon,t) \quad \text{(B12)}$$

and

$$k_s^2=4\pi e^2\int_0^\infty N(\epsilon)\,d\epsilon\left(-\frac{\partial}{\partial\epsilon}f(\epsilon,t)\right), \quad \text{(B13)}$$

where

$$N(\epsilon)=(m_c^{3/2}/\pi^2\hbar^3)(2\epsilon)^{1/2} \quad \text{(B14)}$$

is the density of states.

From (B9) and (B11)–(B13) we know that $\tau_e(t)$ and $n(t)$ are function of $T(t)$ and $\epsilon_F(t)$. In our model, the electrons are heated and generated by laser pulse. For the case of 2.2-MW/cm^2 pulse, the electron density $n(t)$ is in the range $n_0 \sim 20n_0$ ($n_0=10^{16}$ cm^{-3}) during the pulse period, i.e., temperature $T(t)$ and Fermi energy $\epsilon_F(t)$ will vary accordingly. For a quantitative estimate of τ_e, we work the numerical calculation with the following characteristic constants[4,5,7]:

$$\omega=1.78\times10^{14}\ \text{sec}^{-1},$$

$$\Omega_0=3.03\times10^{13}\ \text{sec}^{-1},$$

$$\epsilon_g=0.235\ \text{eV},\quad \epsilon_0=17.88,$$

$$\epsilon_\infty=15.68,\quad m_c=0.0139m_e.$$

For the case $\epsilon_F(t)=1.3\times10^{-4}$ eV and $T=800$ °K, from (B11) and (B12), we get

$$n(t)\simeq 20n_0$$

and

$$1/\tau_e\simeq 8\times10^9\ \text{sec}^{-1}$$

or

$$\tau_e\simeq 10^{-10}\ \text{sec}.$$

This is just the value calculated from $g_c=0.22$ of our previous phenomenological model kinetic calculation.

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