MASTER**LASERS FOR ISOTOPE SEPARATION****Progress Report**

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ABSTRACT

The proposed research ~~research~~ associated with this contract is divided roughly into three parts: first, the study of Raman spin-flip devices and associated semiconductor physics; second, the free-electron laser; and finally, work related to specific isotope-separation processes.

The hot-electron properties of InSb irradiated by 10.6 μM CO_2 laser radiation and the anomalous transmission of the InSb device have been analyzed. In related work we have studied hot-electron effects both in Ge and in $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$ as probed by picosecond Nd:glass laser pulses. Extensive theoretical work has led to a satisfactory explanation of these effects, which are of crucial importance to the understanding of the behavior of semiconductor lasers, such as the InSb Raman spin-flip device. Our studies of Ge also currently include the design of an experiment which may produce direct-gap laser action in Ge.

The free-electron laser has been analyzed successfully using a purely classical approach, thus obviating the need for a much more difficult quantum-mechanical calculation. Substantial progress has been made in understanding both the linear and nonlinear aspects of the problem.

In theoretical studies of the atomic-beam laser-isotope-separation process, we showed that the gain had been substantially underestimated by previous researchers. Experimental work on SF_6 laser isotope separation did not proceed as planned because of equipment failure. Instead, we devoted our efforts to improving the CO_2 laser so as to produce clean single-mode short pulses for pulse-probe experiments on InSb.

Progress Report

The proposed research associated with this contract is divided roughly into three parts: first, the study of Raman spin-flip devices and associated semiconductor physics; second, the free-electron laser; and finally, work related to specific isotope-separation processes.

The first of these problems was investigated in collaboration with T. W. Nee, C. D. Cantrell, and J. F. Scott. In particular, they presented us with a much improved understanding of the problem of the hot-electron properties of InSb irradiated by 10.6 μM CO_2 laser radiation. The anomalous transmission of the InSb device has been studied in some detail and is the subject of the enclosed paper number 1. In closely related work along these lines which was partially supported by ERDA, and is reported in paper 2, we have studied hot-electron effects in germanium as probed by picosecond Nd:glass laser pulses. We have investigated the hot-electron effects not only in germanium but also in $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$, as reported in paper 3. Also, a detailed and extensive theoretical effort has explained satisfactorily the hot-electron processes associated with the passage of such intense laser pulses through matter. This work is reported in paper 4. All of these studies are directly relevant to, and, in fact, essential to proper understanding of semiconductor laser behavior when high fields are involved. The InSb Raman spin-flip device is one example of such a laser. Paper number 1 in this set of four is, of course, directly related to this device.

Experimentally, we are continuing with our work on transient effects in electron-hole plasmas in semiconductors using ultrashort pulses. Currently these experiments are aimed at two primary objectives: a) verification of the theory of Elci et al. (paper 4) through investigation of the predictions in the paper regarding transmission and other effects, and

b) continuation of our study of the feasibility of producing stimulated recombination of carriers and possibly direct-gap laser action in germanium. In the first area, in particular, we are seeing promising results, in that experimental observations of the dependence of transmission in germanium on the pulse width of the incident ($\lambda = 1.06 \mu\text{m}$) optical pulses are in agreement with predictions. In the second area, we are currently designing an experiment which is expected to produce stimulated emission of light of wavelength $\lambda \sim 1.5 \mu\text{m}$ (corresponding to the room temperature direct-band gap in Ge), using as a guide experimental and theoretical work recently completed here on spontaneous radiative recombination.

In regard to the problem of the free-electron laser, we were specifically requested by Keith Boyer to study this problem in some detail, so as to be able to understand the gain per centimeter to be expected as a function of beam current, magnetic field, wavelength, and so on. This we did in a purely classical fashion, demonstrating that the problem of the free-electron device can be treated satisfactorily without using esoteric quantum-mechanical machinery such as the Dirac equation and quantized radiation field. This is fortunate, for it is exceedingly difficult, if not impossible, to carry out a satisfactory calculation of the nonlinear saturation parameters associated with the free-electron device from a quantum mechanical point of view. Such a calculation has been attempted by Kapitza without success, and we are happy to report that we have made substantial progress towards understanding the problem in both its linear and nonlinear aspects. This work is outlined in papers 5 and 6.

Finally, the work on aspects of laser isotope separation was carried out by the Arizona group of Hopf and Scully, in collaboration with ERDA scientists Cantrell and Rhodes. In this regard, we showed (see paper 7) that

the atomic-beam laser-isotope-separation process led to a gain which had been underestimated by previous researchers by a substantial amount. We at the University of Arizona have continued to be involved with problems related to the SF_6 isotope-separation work and the general problem of driven nonlinear oscillators (of which the SF_6 molecule is an example).

The experimental side of this work (SF_6 laser isotope separation) is the one area that did not proceed exactly as planned. Shortly after we began to get our first results on SF_6 , the only mass spectrometer available to us broke down completely and had to be sent back to the factory. When it was returned four months later, it was evident to us that other groups had already substantially completed the studies that we had planned. Thus, we abandoned the SF_6 effort and devoted our efforts to making improvements on the CO_2 laser, so as to produce clean, single-mode short pulses for the InSb studies mentioned earlier. These modifications are now virtually complete, and we will soon be ready to do pulse-probe experiments on InSb. Another interesting application of the modified CO_2 laser is to explore the generation of ultra-short CO_2 pulses using the optical free-induction decay technique of Yablonovitch and Goldhar, Applied Physics Letters 25, [580 (1974)]. They used a hot 2-meter CO_2 cell to obtain 150 psec. CO_2 pulses. However, by using suitable molecular absorbers, it should be possible to obtain adjustable-length pulses using cells as short as 10 cm.

In conclusion, we should remark that the proposal we originally submitted contained more work than we could really do for the amount of monies involved. It was understood that we were to contribute to these efforts as we were called upon by our colleagues at Los Alamos and other branches of ERDA, in accord with the research activity of the group. It should be evident that we we have made substantial contributions to each of the three areas of

investigation discussed above, and we feel that we had the good fortune to live up to both the spirit and the letter of last year's proposal.

Nonlinear Hot Electron Effects on 10.6 μ Radiation in InSb[†]T. W. Nee, C. D. Cantrell[‡], J. F. Scott^{*}, and M. O. ScullyOptical Sciences Center
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ABSTRACT

At high power densities ($\geq 2 \text{ MW/cm}^2$) the temporal dependence of transmitted 10.6 μ pulses in InSb shows strong pump depletion effects. The phenomena are on a 50 nsec time scale and are interpreted as due to hot electron 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.

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ABSTRACT

At high power densities (≥ 2 MW/cm²) the temporal dependence of transmitted 10.6 μ pulses in InSb shows strong pump depletion effects. The phenomena are on a 30 nsec time scale and are interpreted as due to hot electron 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.

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I. Introduction

The output efficiency of InSb spin flip lasers decreases rapidly with increasing pump power. Until recently this effect has been attributed^{only} 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. 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 10.6μ CO_2 laser pump beam is essentially independent of magnetic field strength, we believe that this zero-field calculation ~~explains the qualitative char-~~ *O. K.* acteristics of hot electron phenomena in InSb spin-flip lasers. The spin states and their H-dependences play no important role in the attenuation of the pump.

In a recent study of 12μ spin-flip emission from InSb at $\sim 4^\circ\text{K}$, the time dependence of the 10.6μ radiation transmitted through the sample was also measured [1]. At low incident laser power densities ($\sim 0.2 \text{ MW/cm}^2$), the transmitted signal is proportional to the incident laser pulse signal (with a peak at $t \sim 50 \text{ ns}$). However, if the incident laser power density is raised to $\geq 2 \text{ MW/cm}^2$ the transmitted radiation rises to a peak in $35\text{-}40 \text{ ns}$ and then falls essentially to zero in another $30\text{-}35 \text{ ns}$. After remaining nearly at zero for 150 ns , 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 ns 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 the next section. In section III we describe a pair excitation process ("Kane process") which contributes to the indirect laser generation of conduction electrons. The pertinent rate constant is estimated. In section IV we use a simple two band model with discrete sub-regions to calculate the conduction electron population. The results of numerical calculations are shown in section V.

II. Formalism

$I_i(t)$ = Incident laser pulse intensity. Our pulse has a peak value I_0 at 50 ns and a long tail. See Fig. 1.

$RI_i(t)$ = Reflected laser pulse intensity.

R = Reflection coefficient of the sample (includes net effect of entrance and exit faces and multiple reflections).

$(1-R)I_i(t)$ = Laser pulse intensity inside the sample.

$\alpha(1-R)I_i(t)$ = Laser energy loss per unit time inside the sample.

α = absorption coefficient of the sample.

$I_t(t) = (1-\alpha)(1-R)I_i(t)$ = Transmitted pulse intensity.

~~This is summarized~~
~~The sample geometry is shown~~ in Figure 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,

$$N = \sqrt{\epsilon} = n + iK \quad (1)$$

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

$$R = R_{\infty} \left[1 + \frac{e^{-4KL} (1 - R_{\infty})^2}{1 - e^{-4KL} R_{\infty}^2} \right] \quad (2a)$$

and

$$\alpha = (1 - e^{-2KL}) \frac{1 + e^{-2KL} R_{\infty}}{1 - e^{-4KL} R_{\infty}^2} \quad (2b)$$

where

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

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 electric conductivity is

$$\sigma = \frac{\sigma_0}{1 - i\omega\tau} \quad (4)$$

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

$$\sigma_0 = \frac{ne^2\tau}{m} \quad (5)$$

[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.

$$\epsilon = \epsilon_{\infty} \left(1 - \frac{\omega_p^2}{\omega^2} [1 + 1/\omega\tau]^{-1} \right) \quad (6)$$

where

$$\omega_p = \sqrt{\frac{4\pi n e^2}{\epsilon_{\infty} m}} \quad (7)$$

is the plasma frequency due to the free electrons in the conduction band.

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 to explain the optical excitation of these extra electrons. These electrons must come from the valence band; however, at low temperatures, the band gap $E_g \sim 1900 \text{ cm}^{-1}$ is more than twice as large as the photon energy ($\lambda = 10.6 \mu$, $1/\lambda \sim 944 \text{ cm}^{-1}$), so that direct particle-hole excitation due to single-photon or two-photon absorption is insignificant. We will propose another possible particle-hole pair excitation process in the following section.

III. The Pair Excitation Process²

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 $> 2 E_g$ 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 (Appendix Fig. 1)]. 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

$$\hat{H} = \hat{H}_0 + \hat{H}_1 \quad (8)$$

where

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

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

$$\hat{H}_1 = \sum_{\vec{k}\alpha} \sum_{\vec{k}'\alpha'} \sum_{\vec{p}\beta} \sum_{\vec{p}'\beta'} v_{\vec{k}'\vec{k}\vec{p}'\vec{p}} \hat{C}_{\vec{k}'\alpha'}^{\dagger} \hat{C}_{\vec{p}'\beta'}^{\dagger} \hat{C}_{\vec{p}\beta} \hat{C}_{\vec{k}\alpha} \quad (10)$$

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,

$$E_{pv} = 0$$

because the effective mass of heavy hole valency band $m_v^* = 0.5 m_e$ is much greater than the conduction electron mass $m_c^* = 0.014 m_e$. Then we have

$$\frac{1}{\tau_k} = \frac{\pi}{2\hbar} |I_{cv}(\vec{k})|^2 \sum_{\vec{k}'} |\tilde{v}(\vec{k} - \vec{k}')|^2 N_c(E_{kc} - E_{k'c}) \quad (11)$$

where I_{cv} , the valence-conduction band transition matrix element, is defined

in the Appendix, Eq. (A16); and

$$N_c(E) = \frac{1}{2\pi^2} \left(\frac{2m^*}{\hbar} \right)^3 \sqrt{E - E_G} \quad (12)$$

is the density of state of the conduction band. [We neglect the non-parabolic conduction band structure here.] The Fourier transform of the interaction potential is the screened Coulomb potential

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

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

$$k_s = \sqrt{\frac{4\pi n e^2}{E_F - E_G}} \quad (14)$$

E_F is related to the conduction electron density n by

$$E_F = E_G + \frac{\hbar^2}{2m^*} (3\pi^2 n)^{2/3} \quad (15)$$

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

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

where $y = E_k/E_G$ (17)

and $y_F = E_F/E_G$ (18)

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

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

where

$$x_{\pm} = 1 \pm \sqrt{1 - x_0^2} \quad (20)$$

$$x_0 = 1/\sqrt{y-1} \quad (21)$$

and

$$x_s = \frac{2e^2}{\pi\hbar} \sqrt{\frac{2m_c^*(y_F - 1)}{E_G}} \frac{1}{y-1} \quad (22)$$

Mathematically, for a real solution f , we require $x_{\pm}$ to be real and $x_0 < 1$. Then, from (21), $y > 2$. 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 $\lesssim 1$ for $\vec{k} \cdot \vec{r} \sim 1$. Then the pair excitation rate parameter can be estimated from (16) to be

$$g_p \leq \frac{m_c^{*4} e^4}{\pi^3 \hbar^3 e_m^2} \sim 5 \times 10^{11} \text{ sec}^{-1} \sim 5 \times 10^2 \text{ (ns)}^{-1} \quad (23)$$

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 Section V.]

IV. A 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 ($t < 0$), we have $n_a = n_b = 0$, $n_c = n_0 = 10^{16} \text{ cm}^{-3}$; i.e., all conduction electrons are in the bottom states of the conduction band. 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 the momentum conservation law. 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 emitted 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 $t \geq 0$

$$\dot{n}_a(t) = T_{ba}^L - T_{ab} - T_{ac}^P \quad (24a)$$

$$\dot{n}_b(t) = -T_{ba}^L + T_{cb}^L + T_{ab} - T_{bc} \quad (24b)$$

$$\dot{n}_c(t) = -T_{cb}^L + T_{bc} - T_{cv} + T_{ac}^P + T_{vc}^P \quad (24c)$$

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

$$\dot{n}_{v2}(t) = -T_{vc}^P + T_{12} \quad (24e)$$

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. $T_{ba}^l(T_{cb}^l)$ 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_{ac}^P and T_{vc}^P 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 smaller than that of the single-photon absorption process). Then n_a , n_b , n_c , n_{v1} , and n_{v2} satisfy the particle conservation relation

$$n_a(t) + n_b(t) + n_c(t) + n_{v1}(t) + n_{v2}(t) = 0 + 0 + n_0 + N_{v1} + N_{v2} \quad (25)$$

where, at $t = 0$, $n_a = n_b = 0$, $n_c = n_0$, $n_{v1} = N_{v1}$, $n_{v2} = N_{v2}$; N_{v1} and N_{v2} denote the total number of electrons in the two filled regions of the valence band. According to the assumption of a fast pair excitation process,

$$T_{ac}^P = T_{vc}^P = T^P \quad (26)$$

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

$$n_a \ll n_b < n_c \quad (27)$$

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

$$T^P \approx T_{ba}^l - T_{ab} \quad (28)$$

Then the rate equations (24a-c) can be simplified as

$$\dot{n}_{v1} = T_{cv} - T_{12} \quad (29a)$$

$$\dot{n}_{v2} = -T_{ba}^l + T_{ab} + T_{12} \quad (29b)$$

$$\dot{n}_b = T_{cb}^l - T_{ba}^l + T_{ab} - T_{bc} \quad (29c)$$

and

$$\dot{n}_c = -\dot{n}_{v1} - \dot{n}_{v2} - \dot{n}_b \quad (29d)$$

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

$$T_{ba}^l + T_{cb}^l = \alpha(1 - R)I_l(t)A \quad (30)$$

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

$$\sigma_0 = \frac{n_{bc} e^2}{m} \tau \quad (5')$$

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

$$\dot{n}_{bc} = -\dot{n}_{v1} - \dot{n}_{v2} = T^P - T_{cv} = T_{ba}^l - T_{ab} - T_{cv} \quad (31)$$

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:

$$T_{ba}^l = \gamma_l \{ n_b [N_a - n_a] - n_a [N_b - n_b] \} \quad (32a)$$

$$T_{cb}^{\pm} = \gamma_2 (n_c [N_b - n_b] - n_b [N_c - n_c]) \quad (32b)$$

$$T_{ab} = \gamma_c n_a [N_b - n_b] \quad (32c)$$

$$T_{bc} = \gamma_c n_b [N_c - n_c] \quad (32d)$$

$$T_{12} = \gamma_v n_{v1} [N_{v2} - n_{v2}] \quad (32e)$$

$$T^P = \gamma_p n_a [N_c - n_c]^2 n_v \quad (32f)$$

where γ_2 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, 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 the momentum conservation, the recombination rate between the conduction band and valence band can be written as

$$T_{cv} \approx \text{const.} \times \text{smaller value of } \left\{ \begin{matrix} n_c \\ N_{v1} - n_{v1} \end{matrix} \right\} \quad (33)$$

For the physical situation of the InSb experiment considered here,

$n_c \gg N_{v1} - n_{v1}$; it is a good approximation to write

$$T_{cv} \approx \gamma_{cv} [N_{v1} - n_{v1}] \quad (34)$$

where γ_{cv} is a constant.

For numerical calculations we define the transition ratio

$$x = \frac{T_{ba}^I}{T_{cb}^I} = \frac{N_a n_b - N_b n_a}{N_b n_c - N_c n_b} \quad (35)$$

Then

$$T_{ba}^I = \frac{x}{1+x} \alpha(1-R) I_i(t) A \quad (36a)$$

and

$$T_{cb}^I = \frac{1}{1+x} \alpha(1-R) I_i(t) A \quad (36b)$$

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

$$n_a = \frac{T_{ba}^I}{\gamma_p (N_c - n_c)^2 n_v + \gamma_c (N_b - n_b)} \quad (37)$$

V. Numerical Results

Using the preceding equations we have calculated a theoretical curve for 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_L is given by

$$g_L = I_0 A / n_0 \quad (38)$$

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_0 is the number of conduction electrons in the absence of the beam. In Fig. 4 the theory uses [1] $g_L = 24$ (i.e., 2.2 MW/cm^2)

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

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

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

$$\tau \propto n^{-1/3} \quad (40)$$

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

$$\omega \tau = 2000 \cdot \left(\frac{n}{n_0} \right)^{-1/3} \quad (41)$$

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 $\sim 3 \times 10^8 \text{ s}^{-1}$ at 4°K from the work of Fossum and Ancker-Johnson [3]. [Fig. 4 uses this value of 0.3 (ns)^{-1} .] g_c is the conduction-electron relaxation parameter equal to $\gamma_c n_o$. The fit in Fig. 4 with $g_c = 0.22 \text{ (ns)}^{-1}$ corresponds to a conduction-electron relaxation time of 10^{-10} sec. This is exactly the same value determined by Brueck [5]. g_v is the valence-band hole replacement time, given by $\gamma_v n_o$. Our value $g_v = 8 \times 10^{-4} \text{ (ns)}^{-1}$ used in Fig. 4 corresponds to a hole replacement time of 100 ns. 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 electrons near the top of the valence band; thus, we expect the ratio $\frac{g_c}{g_v}$ to scale not as the electron-to-hole lifetime ratio ($\frac{\tau_e}{\tau_h} \approx 1$) 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. Fig. 5 shows the effect of increasing or decreasing g_v by ten. Since the physical effect of small g_v is to prevent hole mobility and reduce the recombination process by shutting off the supply of electrons near the top of the valence band, values of $g_v \gg 10^{-2}$ 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 $\sim 5 \times 10^2 \text{ (ns)}^{-1}$ 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} s) 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 μ 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.

The limitations of the present work include the following:

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 $m \approx 0.014 m_e$) for the electric conductivity in the calculation. We have neglected the non-parabolicity of the conduction band in InSb.

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

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Appendix A Pair Excitation Rate Calculation

Assume

$$V_{\mathbf{k}'\mathbf{k}\mathbf{p}'\mathbf{p}}^{\alpha'\alpha\beta'\beta} = \frac{1}{2} \int d^3r \phi_{\mathbf{k}'\alpha'}^*(\vec{r}) \phi_{\mathbf{k}\alpha}(\vec{r}) \int d^3r' \phi_{\mathbf{p}'\beta'}^*(\vec{r}') \phi_{\mathbf{p}\beta}(\vec{r}') V(|\vec{r} - \vec{r}'|) \quad (\text{A1})$$

where $\phi_{\mathbf{k}\alpha}$ is the single particle wave function of state $\mathbf{k}$ in band α ;

the two particle interaction can be written in the Fourier form

$$V(|\vec{r} - \vec{r}'|) = \sum_{\vec{q}} \hat{V}(\vec{q}) e^{i\vec{q} \cdot (\vec{r} - \vec{r}')} \quad (\text{A2})$$

Substituting (A2) into (A1) we have

$$V_{\mathbf{k}'\mathbf{k}\mathbf{p}'\mathbf{p}}^{\alpha'\alpha\beta'\beta} = \frac{1}{2} \sum_{\vec{q}} \hat{V}(\vec{q}) I_{\mathbf{k}'\alpha', \mathbf{k}\alpha}(\vec{k} - \mathbf{k}' + \vec{q}) I_{\mathbf{p}'\beta', \mathbf{p}\beta}(\vec{p} - \mathbf{p}' - \vec{q}) \quad (\text{A3})$$

where

$$I_{\mathbf{k}'\alpha', \mathbf{k}\alpha}(\vec{Q}) = \int d^3r e^{i\vec{Q} \cdot \vec{r}} \phi_{\mathbf{k}'\alpha'}^*(\vec{r}) \phi_{\mathbf{k}\alpha}(\vec{r}) \quad (\text{A4})$$

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

$$\phi_{\mathbf{k}\alpha}(\vec{r}) = u_{\mathbf{k}\alpha}(\vec{r}) e^{i\vec{k} \cdot \vec{r}} \quad (\text{A5})$$

with the atomic wave approximation

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

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

$$I_{\vec{k}'\alpha',\vec{k}\alpha}(\vec{Q}) = I_{\alpha',\alpha}(\vec{Q}) \left[\frac{1}{N} \sum_{j=1}^N e^{i\vec{Q} \cdot \vec{R}_j} \right] \quad (\text{A7})$$

where

$$\frac{1}{N} \sum_{j=1}^N e^{i\vec{Q} \cdot \vec{R}_j} = \delta_{\vec{Q},\vec{K}} \quad (\text{A8})$$

$\vec{K}$ is a reciprocal lattice vector, and the transition matrix element

$$I_{\alpha',\alpha}(\vec{Q}) = \int_{\Omega_a} d^3r \phi_{\alpha'}^*(\vec{r}) e^{i\vec{Q} \cdot \vec{r}} \phi_\alpha(\vec{r}) \quad (\text{A9})$$

Ω_a is the atomic volume. We have two cases,

(i) Intraband transition, $\alpha = \alpha'$, the dominant contribution is at

$\vec{K} = 0$. Then we have

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

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

$$I_{\alpha',\alpha}(\vec{Q}) = I_{\alpha',\alpha}(\vec{Q}) \delta_{\vec{Q},\vec{K}} \quad (\text{A11})$$

In this paper, we are interested in the pair production process where $\alpha = \alpha' = 8s = c = 5s$ and $\beta = v = 5p$; see Appendix Fig. 1. For the model interaction Hamiltonian,

$$\hat{H}_1 = \sum_{\mathbf{k}\mathbf{k}'} \sum_{\mathbf{p}\mathbf{p}'} v_{\mathbf{k}'\mathbf{k}\mathbf{p}'\mathbf{p}} \hat{c}_{\mathbf{k}'\mathbf{c}}^\dagger \hat{c}_{\mathbf{p}'\mathbf{c}}^\dagger \hat{c}_{\mathbf{p}\mathbf{v}} \hat{c}_{\mathbf{k}\mathbf{v}} \quad (\text{A12})$$

the interaction matrix element is

$$v_{\mathbf{k}'\mathbf{k}\mathbf{p}'\mathbf{p}} = \frac{1}{2} \sum_{\vec{q}} \hat{V}(\vec{q}) I_{\mathbf{k}'\mathbf{c},\mathbf{k}\mathbf{c}}(\vec{k} - \vec{k}' + \vec{q}) I_{\mathbf{p}'\mathbf{c},\mathbf{p}\mathbf{v}}(\vec{p} - \vec{p}' - \vec{q}) \quad (\text{A13})$$

Next, we consider the lifetime $\tau_{\mathbf{k}}$ of a high energy electron due to this pair excitation process. From the Fermi's Golden Rule

$$\frac{1}{\tau_{\mathbf{k}}} = \frac{2\pi}{\hbar} \sum_{\mathbf{f}} |\langle \mathbf{f} | \hat{H}_1 | \mathbf{i} \rangle|^2 \delta[E_{\mathbf{i}} - E_{\mathbf{f}}] \quad (\text{A14})$$

and using the results of eqs. (A10), (A11), (A12), and (A13), we obtain

$$\begin{aligned} \frac{1}{\tau_{\mathbf{k}}} = & \frac{\pi}{2\hbar} |I_{\mathbf{cv}}(\vec{k})|^2 \sum_{\mathbf{k}'\mathbf{p}\mathbf{p}'} \delta(E_{\mathbf{k}\mathbf{c}} + E_{\mathbf{p}\mathbf{v}} - E_{\mathbf{k}'\mathbf{c}} - E_{\mathbf{p}'\mathbf{c}}) \delta_{\vec{p}-\vec{p}',-\vec{k}'+\vec{k},\vec{k}} \\ & \times \delta_{\vec{p}-\vec{p}',-\vec{k}'+\vec{k},\vec{k}} \frac{n_{\vec{k}\mathbf{c}} n_{\vec{p}\mathbf{v}} (1 - n_{\vec{k}'\mathbf{c}})(1 - n_{\vec{p}'\mathbf{c}})}{\vec{k}'\mathbf{c} \vec{p}'\mathbf{c}} \\ & \times (|\hat{V}(\vec{k}' - \vec{k})|^2 - \hat{V}^*(\vec{p}' - \vec{k})\hat{V}(\vec{k}' - \vec{k})) \end{aligned} \quad (\text{A15})$$

where

$$I_{\mathbf{cv}}(\vec{k}) = \langle \mathbf{c} | e^{i\vec{k}\cdot\vec{r}} | \mathbf{v} \rangle \quad (\text{A16})$$

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

Figure Captions

- Fig. 1. The incident pulse and transmitted pulse profiles. Experimental result of 2.2 MW/cm^2 incident pulse.)
- Fig. 2. A schematic representation of the transmission of CO_2 laser pulse through an n-InSb sample.
- Fig. 3. The possible transitions in the two band model of electronic states in InSb crystal in CO_2 laser field.
- Fig. 4. A "best fit" to the experimental result ($I_i = 2.2 \text{ MW/cm}^2$). The parameters in the theoretical curve are $g_i = 24$, $g_p = g_c = .22$, $g_r = .3$, $g_v = 8 \times 10^{-4}$ [in units of $(\text{ns})^{-1}$] 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. The intra-band electron relaxation effect of heavy hole band. the common parameters are $g_i = 24$, $g_p = 10^2$, $g_c = .22$, $g_r = .3$ [in units of $(\text{ns})^{-1}$] and $\omega\tau = 2000/[(n_{bc}/n_0)^{1/3}]$.

Appendix

- Fig. 1. The pair excitation process diagram.

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INCIDENT PULSE

REFLECTED PULSE

PULSE ENTERING
THE SAMPLE

PULSE ABSORBED
INSIDE THE SAMPLE

InSb CRYSTAL

$\longleftrightarrow L \longrightarrow$

PULSE TRANSMITTED