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Parametric distortion of the optical-absorption coefficient of semiconductors by an additional infrared laser

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The distortion of the optical-absorption coefficient of a direct-gap semiconductor is investigated when the semiconductor is subjected to an additional ir laser field. It is shown that, when the ir laser intensity increases, the absorption coefficient develops a tail below the zero-field gap. An application is made for the case of the InSb irradiated by an intense CO_2 laser.

The development of high-power infrared (ir) lasers has stimulated the study of the interaction of intense radiation with atoms,[1-4] plasmas,[5,6] and solids.[7-12] In particular, in the case of solids, considerable effort has been devoted to the theoretical and experimental aspects of the multiphonon spectroscopy,[8-10] treating the disturbing ir laser as a perturbation. Of the "nonperturbative" methods (i.e., treatments which do not consider the ir laser as a perturbation), the most well-known work is the one carried out by Keldysh[11] almost two decades ago. More recently, Jones and Reiss[12] have proposed another nonperturbative method based upon the description of the band states in the presence of the laser field by a Volkov-type wave function.[13]

In this paper, we address ourselves to the investigation of the changes induced in the absorption coefficient of light of a direct-gap semiconductor due to the simultaneous irradiation of the sample by an ir laser of arbitrary intensity. Specifically, in what follows, we consider the excitation of a valence electron into the conduction band of a direct-band semiconductor by a light beam of frequency Ω and polarization $\vec{\epsilon}$. This exciting light beam is described by the vector potential

$$\vec{A}_{\mathrm{exc}}(t)=A'\vec{\epsilon}e^{-i\Omega t} \ . \tag{1}$$

The entire system is simultaneously irradiated by a circularly polarized ir laser beam of arbitrary intensity and frequency ω, described by

$$\vec{A}_L(t)=A\,(\hat{x}\cos\omega t+\hat{y}\sin\omega t) \ . \tag{2}$$

The ir laser frequency satisfies the condition $\hbar\omega < \epsilon_g$, where ϵ_g is the gap energy, so that it penetrates well through the sample. The semiconductor is assumed to be described by a two-parabolic-band model within the effective-mass approximation. The influence of the intense ir laser on the band states is taken into account by using a Volkov-type wave function for the band states.[12] This is justified by the fact that an electron in a solid in the effective-mass representation has properties which resemble those of a free electron. Thus we may take, to a good approximation, the band states to be described by the wave function of an electron in the presence of a radiation field[5,13,14] (i.e., the Volkov wave function). Under these conditions, the wave function for the valence band is

$$\phi_{v,k}=u_{v,k}(r)e^{i\vec{k}\cdot[\vec{r}-\vec{\delta}_v(t)]}e^{-i\epsilon_v t/\hbar} \ , \tag{3}$$

where $u_{v,k}$ is the Bloch function, $\vec{\delta}_v(t)$ is the amplitude of the electron oscillation in the ir laser field, namely,

$$\begin{aligned}\vec{\delta}_v(t)&=\frac{e}{m_v c}\int^t dt'\vec{A}_L(t')\\&=\frac{eA}{m_v c\omega}(\hat{x}\sin\omega t-\hat{y}\cos\omega t) \ ,\end{aligned} \tag{4a}$$

and

$$\epsilon_v=-\frac{\hbar^2k^2}{2m_v}+\frac{e^2A^2}{2m_v c^2} \ . \tag{4b}$$

As for the conduction-electron wave function, one has

$$\phi_{c,k'}=u_{c,k'}(\vec{r})e^{i\vec{k}\cdot[\vec{r}-\vec{\delta}_c(t)]}e^{-i\epsilon_c t/\hbar} \ , \tag{5}$$

where

$$\begin{aligned}\vec{\delta}_t(t)&=-\frac{e}{m_c c}\int^t dt'\vec{A}_L(t')\\&=-\frac{eA}{m_c c\omega}(\hat{x}\sin\omega t-\hat{y}\cos\omega t)\end{aligned} \tag{6a}$$

and

$$\epsilon_c=\epsilon_g+\frac{\hbar^2k'^2}{2m_c}+\frac{e^2A^2}{2m_c c^2} \ . \tag{6b}$$

Here, m_c and m_v are the effective masses for the electron in the conduction and valence bands, respectively.

With the use of Eqs. (3) and (5) to describe the band states, the probability amplitude for the transition from a valence-band state $\phi_{v,k}$ to a conduction-band state $\phi_{c,w}$, owing to the interaction with the exciting light beam of frequency Ω, can be written as[5,6,12]

$$\begin{aligned}a_{v\to c}&=-\frac{i}{\hbar}\int_{-\infty}^{+\infty}dt\left\langle\Phi_{c,k'}\left|\frac{e}{mc}\vec{A}_{\mathrm{exc}}(t)\cdot\hat{p}\right|\Phi_{v,k}\right\rangle\\&=\sum_{\nu=-\infty}^{+\infty}a^{\nu}_{v\to c} \ ,\end{aligned} \tag{7}$$

where

$$a^{\nu}_{v\to c}=-2\pi i\frac{eA'}{mc}p_{cv}J_\nu(k_\perp a)\delta_{k',k}\delta\left(\frac{\hbar^2k^2}{2m}+\epsilon_g+\frac{e^2A^2}{2\mu c^2}-\hbar\Omega-\nu\hbar\omega\right) \ . \tag{8}$$

Here, $p_{cv}\langle u_c|\vec{\epsilon}\cdot\hat{p}|u_v\rangle$ is the momentum matrix element between the band states, $k_\perp=(k_x^2+k_y^2)^{1/2}$, $m^*=m_c m_v/(m_c+m_v)$ is the reduced mass, $\mu=m_c m_v/(m_v-m_c)$, and $a=eA/m^*c\omega$ is the amplitude of the electron oscillation in the ir laser field. Equation (8) is the probability amplitude for the excitation of an electron from the valence to the conduction band by radiation of frequency Ω with the simultaneous absorption ($\nu>0$) or emission ($\nu<0$) of $|\nu|$ photons of the ir laser. In other words, in the presence of an additional ir laser, the absorption of a light beam of frequency Ω now takes place with the simultaneous multiphoton absorption of emission of the ir laser field.

Knowing the probability amplitude and performing the standard calculations, the absorption coefficient α for the exciting light beam is readily obtained. One gets

$$\alpha=\frac{4e^2\epsilon_g}{n\hbar c\hbar\Omega}\sum_{\nu=-\infty}^{\infty}k(\nu)\int_0^{\pi/2}d\theta\sin\theta J_\nu^2[k(\nu)a\sin\theta] \ , \quad (9)$$

where n is the refractive index of the semiconductor, and

$$k(\nu)=\left[\frac{2m^*}{\hbar^2}\left(\hbar\Omega+\nu\hbar\omega-\epsilon_g-\frac{e^2A^2}{2\mu c^2}\right)\right]^{1/2} \ . \quad (10)$$

Here, $\bar{\nu}$ is the smallest value of ν for which energy conservation is satisfied, i.e., for which $k(\nu=-N)=0$. In arriving at Eq. (9), we have also used the effective-mass sum-rule[15] result for the momentum matrix element, namely, $p_{cv}^2/m=\epsilon_g/2m^*$. In the case of no ir laser field (i.e., $a=0$) and using the result $J_\nu^2(0)=\delta_{\nu,0}$, Eq. (9) reduces to the well-known expression

$$\alpha_0=\frac{4e^2}{n\hbar c}\left(\frac{2m^*}{\hbar}\right)^{1/2}\frac{\Omega_g}{\Omega}(\Omega-\Omega_g)^{1/2} \ , \quad (11)$$

where $\hbar\Omega_g=\epsilon_g$; i.e., in the absence of the ir laser field, there is absorption only for $\Omega>\Omega_g$, as expected. In contrast, the presence of the ir laser field introduces another channel for the electron to exchange energy, thereby making possible its excitation even for $\Omega<\Omega_g$. This is seen from Eq. (10) for which the condition $k(\nu)>0$ is satisfied provided

$$\nu\hbar\omega+\hbar\Omega-\hbar\Omega_g-e^2A^2/2\mu c^2>0 \ .$$

That is, for $\Omega<\Omega_g$ energy conservation can be satisfied provided the electron absorbs more than N photons from the ir laser, where

$$N\hbar\omega=\hbar\Omega_g-\hbar\Omega+e^2A^2/2\mu c^2 \ .$$

To better assess our results, we have calculated the absorption coefficient α for an InSb sample irradiated by a CO_2 laser with $\omega=1.77\times10^{14}\ s^{-1}$. The sum over ν in Eq. (9) has been carried out numerically using the following values for the physical parameters of InSb: $m_v=0.18m$, $m_c=0.013m$, $n=3.75$, $\epsilon_g=0.167$ eV. In our calculations we have introduced the parameter $\eta=\Omega/\omega$ as a measure of the exciting frequency in units of the ir laser frequency, and the parameter $\epsilon=e^2A^2/2m^*c^2\hbar\omega$ as a measure of the energy delivered by the ir laser to an electron of mass m^* in units of the ir quantum $\hbar\omega$. The parameter ϵ is related to the ir laser intensity by $I=ncm^*\hbar\omega^3\epsilon/2\pi e^2$, or, in W/cm^2,

$$I=n\epsilon\frac{m^*}{m}\omega^3\times1.98\times10^{-33} \ , \quad (12)$$

which for a CO_2 laser reduces to

$$I=10.98n\epsilon(m^*/m)\ \mathrm{GW/cm^2} \ .$$

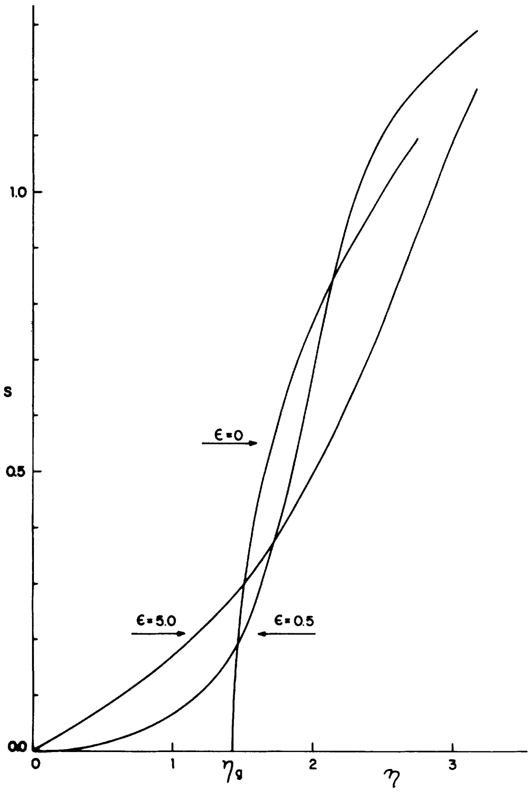

FIG. 1. Absorption coefficient (S), of an InSb sample as a function of the exciting frequency (η), for several values of the dressing CO_2-laser intensity (ϵ).

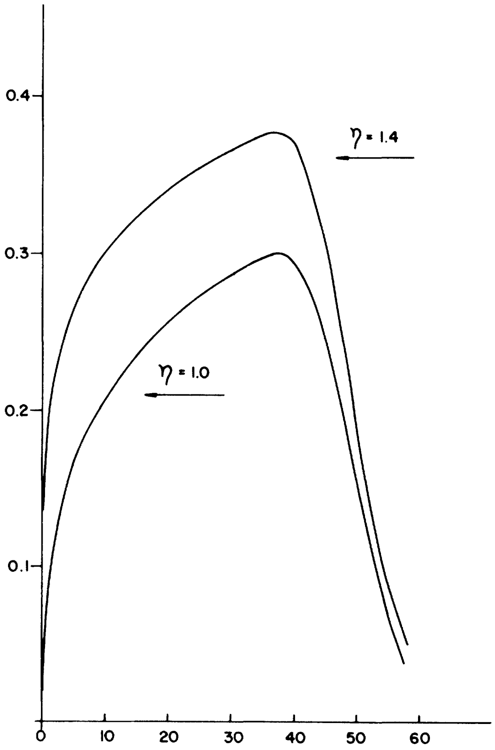

FIG. 2. Absorption coefficient (S) of InSb, as a function of the dressing CO_2-laser intensity (ϵ), for the exciting frequencies $\eta=1.0$ and 1.4.

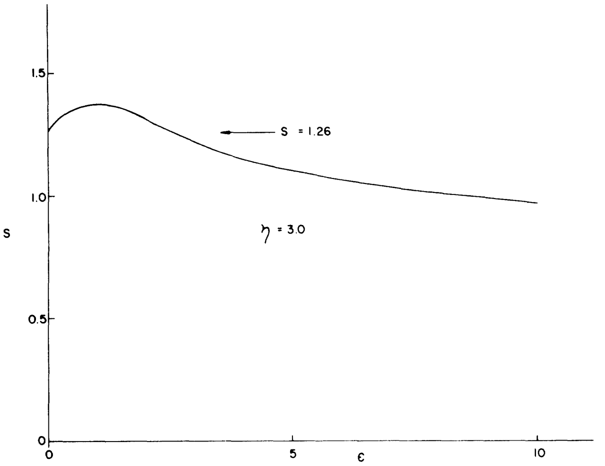

FIG. 3. Absorption coefficient (S) of InSb, as a function of the dressing CO_2-laser intensity (ϵ), for the exciting frequency $\eta = 3$. $S_0 = (\eta - \eta g)^{1/2}$ is the result without the ir laser.

In terms of these dimensionless parameters, Eq. (7) can be rewritten as

$$\alpha = \frac{4e^2}{n\hbar c}\frac{\eta_g}{\eta}\left[\frac{2m^*\omega}{\hbar}\right]^{1/2} S \ , \tag{13}$$

where η_g is the value of η for $\Omega = \Omega_g$, and $S = S(\eta, \epsilon)$ is the dimensionless form of the summation over ν of Eq. (9), i.e., the dimensionless absorption coefficient.

In Fig. 1 we plot the dependence of S on η for several values of the laser intensity (ϵ). Also indicated in Fig. 1 is the value of η corresponding to the energy gap for the case of an InSb sample irradiated by a CO_2 laser, namely, $\eta_g = 1.4$. We note from Fig. 1 that on increasing the ir laser intensity, the absorption coefficient gets distorted so as to give rise to an absorption tail below the zero-field forbidden gap. This tail is such that the absorption coefficient increases and spreads more below the gap, on increasing the ir laser intensity. In Figs. 2 and 3 we plot the dependence of the absorption coefficient as a function of the ir laser intensity. In Fig. 2, we show this dependence for two cases which, in the absence of the ir laser field, namely, the cases where $\Omega < \Omega_g$ (or $\eta < \eta_g$), would give us no absorption of radiation. We note that in these cases the absorption coefficient starts from zero, as should be expected. In contrast, in Fig. 3 we plot the absorption coefficient as a function of ϵ for a case in which, in the absence of the ir laser field, we already have a finite absorption coefficient, namely, the case $\eta > 1.4$. In this case, the absorption starts from the ir laser-free value $S_0 = (\eta - \eta_g)^{1/2}$. Figures 2 and 3 show, however, the same general type of behavior with the laser intensity; namely, after starting from the corresponding ir field-free value, the absorption coefficient increases on increasing the ϵ, reaches a peak, and finally, for higher fields, decreases asymptotically as $\epsilon^{-1/2}$. This behavior is reminiscent of other phenomena involving multiphonon processes, such as multiphonon ionization in atomic systems[1-4] and high-field heating of electrons in plasmas,[5-7] where one also encounters a drop in the effectiveness of the laser at very large fields. To explain this result, one can argue that for small fields both absorption and emission of ir photons by the electrons, while absorbing the radiation of frequency Ω, are equally possible. This opening of extra "channels" for the electron to exchange energy makes possible the absorption below the gap and results in an initial increase of the absorption cross section. However, at intermediate fields, the emission of ir photons reaches a saturation since the energy conservation puts an upper limit equal to ν on the maximum of photons that can be emitted. At this point the absorption cross section tends to level off. For even higher fields, in addition to this limit, the energy gap is increasing linearly with the intensity, due to the last term on the right-hand side of Eq. (10) (namely, $\epsilon_g \rightarrow \tilde{\epsilon}_g = \epsilon_g + e^2A^2/2\mu c^2$). Furthermore, in the high-field limit, the electron is moving very fast so that its interaction with the environment tends to weaken. These two facts thus point in the direction of a reduction of the absorption cross section at very large ir laser intensity.

TABLE I. Peak values of the absorption coefficient of an InSb sample and the corresponding CO_2-laser intensity, at the frequencies $\eta = 1$, 1.4, and 3.

η	ϵ	S	α (cm^{-1})	I (GW/cm^2)
1.0	37	0.3	0.47×10^4	19.81
1.4	35	0.38	0.59×10^4	18.74
3.0	1	1.38	2.15×10^4	0.53

To estimate the size of the effects reported above, let us consider the absorption at the peaks of Figs. 2 and 3. For $\eta = 1.0$ and 1.4, the absorption peaks at $\epsilon = 37$ and 35, and the corresponding values of S are 0.30 and 0.38, respectively. For $\eta = 30$, the absorption peak occurs at $\epsilon = 1$ and S is equal to 1.38. Substituting these values of η, ϵ, and S in Eqs. (12) and (13), one finally gets both the absorption coefficient and the corresponding CO_2 laser intensity needed. The results from our calculations are summarized in Table I. It follows from this table that, even below the zero-field gap, the absorption coefficient is greatly enhanced as the dressing ir laser intensity increases. This result suggests that when using semiconductors such as InSb as an ir window or as a lens in experiments involving high-power ir lasers (e.g., laser fusion), one should be careful about the ir laser intensity crossing the window, otherwise severe damage or malfunction of the window may occur.

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