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**19. Abstract**: Magneto-Optical studies of HgCdTe have provided a rich new group of spectroscopies for studying defects and impurities in narrow band semiconductors such as HgCdTe.
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"Two-Photon Absorption Characterization"

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According to the Time Phase Task Schedule for the third quarter on p. 26 of the proposed Technical Work, our research investigation and sample characterization has proceeded along four task categories (b), (c), (f), and (e):

(b) Investigate two-photon absorption...

(c) Identify impurity related energy levels and correlate...

(e) Investigate time resolved behavior in HgCdTe excited by two-photon absorption. Estimate the lifetimes of charge carriers.

(f) Analyze "lifetime" data to determine surface and bulk recombination probabilities.

We shall now address the progress in each of the tasks.

I. (b) Investigate two-photon absorption

As pointed out in the last quarter, a major breakthrough was achieved in observing two-photon magnetoabsorption (TPMA) in bulk samples of both n- and p-type Hg$_{1-x}$Cd$_x$Te. During this quarter, a major emphasis was placed on data interpretation. We now present TPMA results and analyses on four different samples of HgCdTe with different x-values (n-type samples of x = 0.238, 0.258, 0.276).

\[ n - \text{HgCdTe}(x = 0.236) \]

This sample was obtained from Honeywell and had the following properties:

\[ \mu(77K) = 1.6 \times 10^4 \text{cm}^2/\text{Vsec} \]

\[ n(77K) = 1.4 \times 10^{14} \text{cm}^{-3} \]

Infrared transmission \( x = 0.2406 \pm 0.001 \)

Figure 1 shows the photoconductive (PC) response of the sample versus magnetic field B for various CO$_2$ laser wavelengths. A variety of resonant structure is observed which we shall now describe in detail.
Table I. Two-Photon Magneto Absorption Transitions for Hg$_{1-x}$Cd$_x$Te Given From Low Energy to Higher Energies.

<table>
<thead>
<tr>
<th>Designation</th>
<th>Energy Level Transition</th>
<th>Polarization</th>
</tr>
</thead>
<tbody>
<tr>
<td>$R_1$</td>
<td>$a^- (2) \rightarrow a^c (0)$</td>
<td>$\sigma_R$</td>
</tr>
<tr>
<td>$R_2$</td>
<td>$b^- (2) \rightarrow b^c (0)$</td>
<td>$\sigma_R$</td>
</tr>
<tr>
<td>$\pi_1$</td>
<td>$a^+ (0) \rightarrow a^c (0)$</td>
<td>$\pi$ or $\sigma$</td>
</tr>
<tr>
<td>$L_1$</td>
<td>$a^+ (-1) \rightarrow a^c (1)$</td>
<td>$\sigma_L$</td>
</tr>
<tr>
<td>$\pi_2$</td>
<td>$a^- (1) \rightarrow a^c (1)$</td>
<td>$\pi$ or $\sigma$</td>
</tr>
<tr>
<td>$R_3$</td>
<td>$a^- (3) \rightarrow a^c (1)$</td>
<td>$\sigma_R$</td>
</tr>
<tr>
<td>$\pi_3$</td>
<td>$b^- (1) \rightarrow b^c (1)$</td>
<td>$\pi$ or $\sigma$</td>
</tr>
<tr>
<td>$L_2$</td>
<td>$b^+ (-1) \rightarrow b^c (1)$</td>
<td>$\sigma_L$</td>
</tr>
<tr>
<td>$\pi_4$</td>
<td>$b^+ (0) \rightarrow b^c (0)$</td>
<td>$\pi$ or $\sigma$</td>
</tr>
<tr>
<td>$\pi_5$</td>
<td>$a^- (2) \rightarrow a^c (2)$</td>
<td>$\pi$ or $\sigma$</td>
</tr>
<tr>
<td>$\pi_6$</td>
<td>$b^- (2) \rightarrow b^c (2)$</td>
<td>$\pi$ or $\sigma$</td>
</tr>
<tr>
<td>$R_4$</td>
<td>$a^+ (2) \rightarrow a^c (0)$</td>
<td>$\sigma_R$</td>
</tr>
<tr>
<td>$L_3$</td>
<td>$a^+ (0) \rightarrow a^c (2)$</td>
<td>$\sigma_L$</td>
</tr>
<tr>
<td>$\pi_7$</td>
<td>$a^+ (1) \rightarrow a^c (1)$</td>
<td>$\pi$ or $\sigma$</td>
</tr>
<tr>
<td>$\pi_8$</td>
<td>$a^- (3) \rightarrow a^c (3)$</td>
<td>$\pi$ or $\sigma$</td>
</tr>
<tr>
<td>$L_4$</td>
<td>$b^+ (0) \rightarrow b^c (2)$</td>
<td>$\sigma_L$</td>
</tr>
</tbody>
</table>

The notation is as follows:

- $a, b$ denote different spin states; the superscript $-$ (+) denotes a heavy hole state (light hole state); the superscript $c$ denotes a conduction band state.

First of all, some of the structure in Fig. 1 have resonances whose magnetic field position depend upon the laser wavelength or photon energy. One example of this is shown by the arrow marked in the Figure. We identify this structure as arising from two-photon absorption transitions that take an electron from an initial hole Landau level ($E_H$) to a final conduction band Landau level ($E_C$). Possible transitions are given in Table I. Comparisons between the TPA and theoretical predictions are shown in Fig. 2 where $2 \hbar \omega$ is plotted versus $B$. The lines represent the theoretical variation calculated from $2 \hbar \omega = E_C - E_H$, where the Landau level energies are calculated with a modified Pidgeon-Brown energy band model using the following set of band parameters:

$$E_p = 19.0 \text{ eV}, \Delta = 1.0 \text{ eV}$$

$$\gamma_1 = 3.3, \gamma_2 = 0.1, \gamma_3 = 0.9$$
These are the set of parameters given by Weiler in her Semiconductors and Semimetal article. The energy gap was then adjusted to give the "best fit" to the observed transitions. Excellent agreement is attained with $E_g = 121.8 \pm 0.5$ meV which corresponds to an $x$-value of $0.2363 \pm 0.0003$ using the Hansen, Schmit, Casselman relationship to relate $x$ to $E_g$.

The excellent agreement between the data and the two-photon transition energy theoretical variations are one of the strongest evidences that these observed transitions are indeed two-photon related. Further proof as to this designation comes from polarization studies shown in Fig. 3 obtained using an infrared zero order wave plate that converts linear polarized light into either left ($\sigma_L$) or right ($\sigma_R$) circularly polarized light. These data were taken on a sample obtained from Texas Instruments (it had a similar $x$-value to the Honeywell sample). The strongest (TPA) transitions occur for $\sigma_L$ polarization as expected. These two $\sigma_L$ transitions seem to be the distinguishing feature of the two-photon spectra in Hg$_{1-x}$Cd$_x$Te. Two-photon spectra observed in InSb are quite different as seen by the data in Fig. 4. for $T = 50$K and $\lambda = 9.23$ $\mu$m. Note the much sharper linewidths and the wide variety of structure. The very broad linewidths of HgCdTe are due to inhomogeneities. This fact can be used to quantitatively estimate the amount of inhomogeneity broadening from the width of the resonance peaks.

Some of the structure (10.5 $\mu$m to 10.79 $\mu$m) present in Fig. 1 is independent of laser wavelength. We shall comment on this structure later in the report.

Other absorption processes occur at lower magnetic fields and lower wavelengths as seen in Fig. 5. Two additional absorption mechanisms occur - one-photon interband absorption (OPA), and absorption due to electron transitions from a shallow acceptor to the lowest conduction band Landau level. These transitions occur at much lower intensities than the TPA transitions. The temperature dependence of the PC response at 9.69 $\mu$m is shown in Fig. 6. As can be seen, the impurity transition becomes much weaker at the higher temperatures. Verification of these identifications comes from the good agreement between the theoretically calculated one-photon transition energies and the experimental data as shown in Fig. 7. The one-photon transition energies are given by

$$\hbar \omega_{\text{OPA}} = E_C - E_V \text{ (interband, with no exciton corrections)}$$

and

$$\hbar \omega_{\text{IMP}} = E_C - E_a \text{ (impurity)}$$

With an energy zero at the top of the valence band, at $B = 0$, $E_V = 0$ and $\hbar \omega_{\text{OPA}} - \hbar \omega_{\text{IMP}} = E_a = \text{the acceptor impurity binding energy}$. The best fit to the data gives $E_a = 9.0$ meV.

Figure 8 shows the intensity dependence of the PC response at $T = 7$ K and $\lambda = 10.20$ $\mu$m, a wavelength where all three absorption mechanisms can be seen. First of all, at much lower intensities than used in Fig. 8 only OPA structure can be seen. As the intensity is increased to $I_0/4$, TPA structure can be seen at very high fields and the acceptor impurity structure becomes larger than the OPA structure which saturates at these high intensities. Finally, at the highest intensity, $I_0 = 1.8$ kW/cm$^2$, only the impurity and TPA structure can be observed.

The intensity dependence of the PC response at $T = 6$ K and $\lambda = 9.29$ $\mu$m, the region of the OPA and impurity structure is shown in Fig. 9. At low intensities only OPA structure dominates. At the highest intensity shown in Fig. 9, some unusual structure is observed at fields smaller than the highest field OPA structure. The photon energy dependence of these new transitions is shown in Fig. 10 on an expanded, more sensitive B-field scale. We note that the resonant positions of this structure do not depend greatly upon wavelength. During the next quarter, we shall work on interpreting this structure. For now we just note that it is there.
This bulk sample was obtained from Cominco and had the following properties:

300 K
IR-TCO

\[ \mu = 7.3 \times 10^4 \text{cm}^2/\text{V}.\text{sec} \]
\[ n = 7.2 \times 10^{15} \text{cm}^{-3} \]

77 K

\[ \mu = 7.6 \times 10^4 \text{cm}^2/\text{V}.\text{sec} \]
\[ n = 1.0 \times 10^{14} \text{cm}^{-3} \]

Crystal identification p4100.60

Figure 11 shows TPA spectra in the PC response for this sample at 7K. The dominant features are again two resonant absorption processes seen at high fields. Good agreement between the calculated two-photon transition energies vs. B field and the experimental data is obtained for \( E_g = 156.0 \pm 0.5 \text{ meV} \) or \( x = 0.2580 \pm 0.0003 \) as seen in Fig. 12.

The half-width of the resonant TPA structure can be used to determine the amount of inhomogeneity due to an \( x \)-value variation. As stated earlier, the TPA linewidths in n-InSb are much narrower (factor of 10) than in HgCdTe. Homogeneous samples of HgCdTe would have one \( x \)-value. Variations in \( x \)-value lead to variations in \( E_g \). Thus, if one determines the field positions of the full width at half-maximum of the resonances, these can be used to calculate values of \( E_g \) or hence \( x \). Figure 13 shows the experimental data for these half maximum positions for the two main transitions seen in Fig. 11. These in turn are fit to theoretical calculations with upper (160 meV) and lower (152 meV) values. This leads to an \( x \)-variation of \( \Delta X = 0.0025 \).

This bulk sample was obtained from Cominco and it had the following properties:

300 K
IR-TCO

\[ \mu = 5.8 \times 10^4 \text{cm}^2/\text{V}.\text{sec} \]
\[ n = 5.2 \times 10^{15} \text{cm}^{-3} \]

77 K

\[ \mu = 6.6 \times 10^4 \]
\[ n = 1.4 \times 10^{14} \]

Crystal identification 15(603) - 51

Two-photon structure is again observed in the PC response of this sample as seen in Fig. 14. Two main features dominate as before, with two additional much broader resonances occurring at higher fields. The fan chart of \( 2 \hbar \omega \) vs. B in Fig. 15 shows a comparison between the data and the calculated transitions. The two main transitions are clearly in excellent agreement with the calculations demonstrating that they are related to TPA structure. However, the high field structure can not be explained by any known two-photon transitions. The three that are possible, \( R_1 \), \( R_2 \), and \( \pi_1 \).
show rather strong disagreement with the data. Consequently, we believe that the most plausible inter-
pretation of this structure is that they arise from electron transitions from two separate midgap levels to the lowest conduction band Landau level \( s_c(0) \). The deep level energies would not depend on magnetic field and so the transition energy would be

\[
\hbar \omega = E(s_c(0)) - E_d
\]

which would be governed by the variation of \( E(s_c(0)) \). In Fig. 16, \( \hbar \omega \) is plotted versus B showing the excellent agreement between the data and the theoretically calculated variation using \( E_d \) values of \( E_{d_1} = 100 \) meV and \( E_{d_2} = 94 \) meV. We note that midgap levels in HgCdTe are expected and these measurements are an important indication of their presence and provide an accurate determination of their location.
Figure 1. Wavelength dependence of photoconductive response showing high intensity structure.
Figure 2. Fan chart of transition energy vs. magnetic field. The lines are theoretical energies calculated from a modified Pidgeon-Brown model. They extrapolate at zero field to the band gap (121.8 meV). The labels for each transition represent the polarization of the incident light, and the energetic order.

\[ X = 0.2363 \pm 0.0003 \]

\[ E_g = 121.8 \pm 0.5 \text{ meV} \]

\[ T = 7.0 \text{K} \]
Figure 3. Polarization dependence of the photoconductive response showing clearly that the two dominant transitions arise from left circular polarization.
Figure 4. Two photon absorption spectra in n-InSb. The upper trace was analyzed with a boxcar averager, the lower with lock-in amplifier techniques which synthesize the second derivative.
Figure 5. Wavelength dependence of the photoconductive response in HgCdTe. One photon magneto-absorption (OPMA) and acceptor to conduction band (ACB) transitions are shown.
Figure 6. Temperature dependence of the photoconductive response of HgCdTe. Note that the ACB structure is more strongly temperature dependent than is the OPMA structure.
Figure 7. Fan chart of transition energies vs. magnetic field for HgCdTe. One-photon absorption (OPA), two-photon absorption (TPA), and acceptor to conduction band (Accepter Impurity) transitions are shown.
Figure 8. Photoconductive response of HgCdTe for various incident photon intensities. Note the damping out of the two-photon absorption (TPA) structure with decreasing intensity, and the separation of the impurity and one-photon absorption structures.
Figure 9. Incident photon intensity dependence of the photoconductive response of HgCdTe showing high intensity structure, acceptor to conduction band (ACB) structure, and one-photon magneto absorption (OPMA) structure.
Figure 10. Wavelength dependence of the high intensity structure showing the movement of the one-photon magneto absorption (OPMA) structure against the high intensity structure.
Figure 11. Wavelength dependence of the dominant two-photon magneto absorption (TPMA) structure in HgCdTe with $x = 0.26$. 
Figure 12. Fan chart of transition energies vs. magnetic field. The extrapolated band gap is 155.0 meV.
Figure 13. Fan chart of transition energy vs. magnetic field for the $x = 0.258$ sample. The typical full width at half max of the TPMA peaks was used as an estimate of the error. This analysis yields two band gap values which can be used to judge the relative accuracy of the $x$ value throughout the sample.
Figure 14. Photoconductive response vs. magnetic field for various wavelengths at 7 K. Note the two strong TPMA transitions.
Figure 15. Fan chart of transition energies vs. magnetic field. The extrapolated band gap is 195.0 meV which yields an x value of 0.2811. The two transitions which do not lie along any theoretical line are believed to arise from mid-gap to conduction band transitions.
Figure 16. The two mid-gap to conduction band transitions fit with theoretical Pidgeon-Brown calculations. The two levels lie at 100 and 94 meV above the valence band.