Three-photon absorption and linear-circular dichroism of InAs

A. M. Danishevskiĭ, S. F. Kochegarov, and V. K. Subashiev

A. F. Ioffe Physico-technical Institute, USSR Academy of Sciences, Leningrad
(Submitted May 20, 1975)
Zh. Eksp. Teor. Fiz. 70, 158–162 (January 1976)

Three-photon absorption and photoconductivity associated with this absorption were observed in indium arsenide. A strong linear-circular dichroism was also observed near the three-photon absorption edge. This dichroism decreased rapidly away from the edge into the absorption band. The contributions of allowed-forbidden-forbidden and allowed-allowed-allowed transitions to the three-photon absorption were determined

PACS numbers: 71.30.Mw, 72.40.+w

Three-photon absorption has been observed in alkali halide crystals,[1] gases,[2] and semiconductors.[3] The polarization dependences of n-photon absorption in solids have been investigated experimentally only for the $n=2$ case (see, for example, [4,5]).

It is shown in theoretical papers[6,7] that in III-V crystals the three-photon transitions of light with circular polarization are forbidden for $k=0$ if we use the two-band approximation (valence band v and conduction band c). Allowance for intermediate states in a higher conduction band $\bar{c}$ lifts this forbiddeness. We can then have allowed–allowed–allowed (AAA) three-photon transitions of the $v-\bar{c}-v-c$ and $v-c-\bar{c}-c$ types. In the case of linear polarization, we can also have AAA transitions of the $v-c-v-c$ type. Their contribution to the three-photon absorption process is the strongest (for the linear polarization). The ratio of the probabilities of three-photon absorption of linearly and circularly polarized light $W_l^{(3)}/W_c^{(3)}$, considered in[7] at $k=0$ in the three-band model[1)] is of the order of $(P_{cv}{}^2E_0'/P_{\bar{c}v}{}^2E_g)^2$. We may assume that the matrix elements of the allowed transitions $P_{\bar{c}v}$ and P_{cv} do not differ greatly from one another and then the probability ratio is governed by $(E_0'/E_g)^2$. In the case of indium arsenide (InAs) at 293 °K the forbidden band width is $E_g=0.35$ eV and the energy of the upper conduction band $\bar{c}$, measured from the top of the valence band, is $E_0'=3.9$ eV.[8] Thus, the probability ratio $W_l^{(3)}/W_c^{(3)}\equiv\xi$ is $\sim 10^2$.

At $k\neq 0$ $(3\hbar\omega > E_g)$ in addition to the three transitions mentioned above, we can have six more transitions (in the linear and circular polarization) and three of them ($v-\bar{c}-\bar{c}-c$; $v-\bar{c}-c-c$; $v-v-\bar{c}-c$) are allowed–allowed–forbidden (AAF) in III-V compounds whereas the other three ($v-v-v-c$; $v-c-c-c$; $v-v-c-c$) are allowed–forbidden–forbidden (AFF) transitions. As the difference $(3\hbar\omega - E_g)$ increases and so does k and it then follows from the calculations of Arifzhanov and Ivchenko[7] that $W_c^{(3)}$ rises faster than does $W_l^{(3)}$ and, therefore, the linear-circular dichroism decreases.

The aim of the present study was to detect experimentally three-photon absorption in indium arsenide and to determine the value of linear-circular dichroism for various relationships between $3\hbar\omega$ and E_g. Three-photon absorption was investigated experimentally by measuring the transmission coefficient T and the photoconductivity $\Delta\sigma$ as a function of the intensity of incident light j_0. Our radiation source was a Q-switched CO_2 laser emitting at 10.6 or 9.5 μ. The transmission measurements were carried out under periodic conditions and also in the single-pulse regime. In the latter case there was no significant heating of the sample by the laser radiation. The photoconductivity measurements made it possible to observe signals at intensities several times lower than in the case of direct measurement of the nonlinearity of $T(j_0)$ and, therefore, they were carried out in the periodic regime. In all experiments careful measures were taken to avoid inhomogeneity of the distribution of the intensity of light in the investigated part of the sample.

The photoconductivity measurements were carried out under "static" conditions and the diameter of the illuminated spot exceeded the width of the illuminated face of the crystal. Thus, the ratio of the photoconductivity signals (for low values of $\Delta\sigma/\sigma$) obtained for linear and circular polarizations of the exciting radiation was equal to the ratio of the absorption probabilities[2)] $W_l^{(3)}/W_c^{(3)}=\xi$.

Figure 1 gives the dependences $j_h(j_0)$ obtained in our transmission measurements. Clearly, a deviation from linearity in the range $j_0>1.2\times10^{26}$ photons · cm^{-2} · sec^{-1}, was due to the three-photon absorption and the associated absorption by nonequilibrium holes. Figure 2 gives the dependences of the relative photoconductivity $\Delta\sigma/\sigma$, obtained for linear and circular polarizations of light at wavelengths 9.5 and 10.6 μ, on the intensity of the exciting light.

The cubic dependence of $\Delta\sigma/\sigma$ on j_0 for the linearly polarized radiation indicated that we were dealing with three-photon transitions from the valence to the conduction band.[3)] The use of oriented samples with the illuminated surface close to the (100) plane of the crystal indicated that two-photon transitions of the $v\to 2\hbar\omega+\hbar\omega\to c$ type made no significant contribution to the photoconductivity.

It is clear from Fig. 2 that in the case of the $\lambda=10.6$ μ radiation, corresponding to the three-photon absorption edge, there was a strong linear-circular dichroism reaching ~27 for minimum intensities used in these experiments. For $\lambda=9.5$ μ, this dichroism was much weaker (~1.4), which indicated a considerable contribution of AFF transitions to the probability of three-photon absorption away from the edge into the band.[4)]

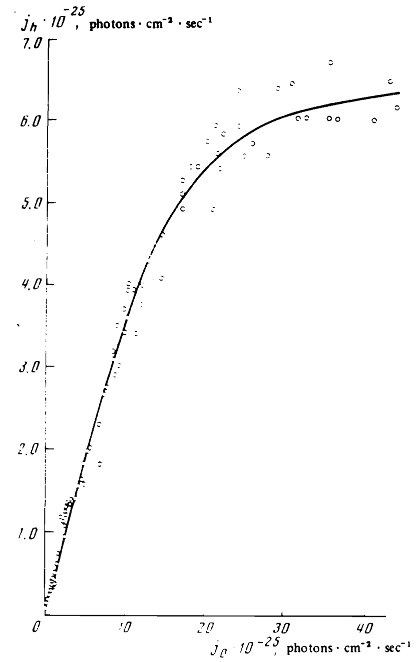

FIG. 1. Dependence of the intensity of the radiation transmitted by a sample j_h on the intensity of the radiation j_0 (linear polarization) incident on n-type InAs; $n = 8\times10^{16}$ cm^{-3}, $\lambda = 9.5\ \mu$, $h = 1$ mm, $T = 300$ °K.

This conclusion was also supported by the temperature dependence of $\Delta\sigma_l/\Delta\sigma_c$ plotted in Fig. 3. A relatively slight increase in temperature from 26 to 52 °C altered the ratio $\Delta\sigma_l/\Delta\sigma_c$ for $\lambda = 10.6\ \mu$ from 27 to 2, which was due to the temperature dependence of E_g and the strong dependence of $W_l^{(3)}/W_c^{(3)}$ on the value of $(3\hbar\omega - E_g)$ near the three-phonon absorption edge. The dichroism at $\lambda = 9.5\ \mu$ depended much less on temperature.

These results made it possible to explain the supercubic dependence $\Delta\sigma_c(j_0)$ for $\lambda = 10.6\ \mu$ (Fig. 2): in the case of radiation with circular polarization the main contribution to the three-photon absorption was due to AAA transitions, whose probability depended relatively on the value of $(3\hbar\omega - E_g)$, whereas in the case of circular polarization the contribution of AAA transitions was very small and AFF transitions played an important role and the probability of the latter depended strongly on the value of $(3\hbar\omega - E_g)$ and, consequently, on the heating of the crystal by laser radiation.

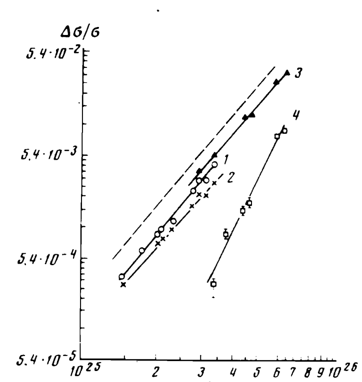

FIG. 2. Dependences of the relative photoconductivity $\Delta\sigma/\sigma$ on the intensity j_0 of light incident on n-type InAs ($n = 2.3\times10^{16}$ cm^{-3}): 1), 2) $\lambda = 9.5\ \mu$; 3), 4) $\lambda = 10.6\ \mu$; 1), 3) linear polarization; 2), 4) circular polarization; $T = 300$ °K. The dashed line represents the dependence $\Delta\sigma/\sigma \propto j_0^3$.

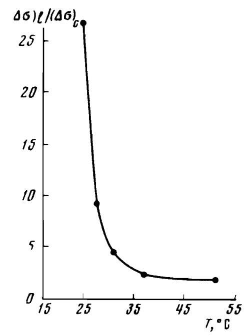

FIG. 3. Temperature dependence of the linear-circular dichroism deduced from the photoconductivity: $j_0 = 3.3\times10^{25}$ photons · cm^{-2} · sec^{-1}; n-type InAs ($n = 2.3\times10^{16}$ cm^{-3}); $\lambda = 10.6\ \mu$.

Thus, the uncontrolled rise of temperature of the sample with rising j_0 had little effect on the dependences of $\Delta\sigma_l$ and $\Delta\sigma_c$ on j_0 for $\lambda = 9.5\ \mu$ and on $\Delta\sigma_l(j_0)$ for $\lambda = 10.6\ \mu$ but it affected considerably the dependence $\Delta\sigma_c(j_0)$ for $\lambda = 10.6\ \mu$. Curves 3 and 4 in Fig. 2 were used to plot the dependence of $\Delta\sigma_l/\Delta c_c$ on j_0. This dependence was similar to that of $\Delta\sigma_l/\Delta\sigma_c$ on T (Fig. 3). A comparison of these curves made it possible to estimate the temperature of the sample in its illuminated part.

The authors are grateful to S. B. Arifzhanov and E. L. Ivchenko for numerous discussions, and to M. Bartenev for his help in the experiments.

[1)]In the two-band model, this ratio is $W_l^{(3)}/W_c^{(3)} = \infty$.

[2)]Since the rate of excitation in our experiments was relatively slow ($\Delta\sigma/\sigma < 10^{-2}$), the electron lifetime τ_{ph} making the main contribution to the photoconductivity was independent of the excess carrier density and was the same for linear and circular polarization of light.

[3)]The 0.9 confidence intervals in Fig. 2 were found allowing only for the random errors due to fluctuations of the observed signals. The points for which they are not given were characterized by very small fluctuations.

[4)]Estimates for this case indicated that the contribution due to AAF transitions was much smaller than due to AFF transitions.

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Translated by A. Tybulewicz