

THEORY OF MULTIPHOTON ABSORPTION
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ABD-EL-RAHMAN EL-SAYED MOUSSA.M.Sc.

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S U M M A R Y

The purpose of the present thesis is to give a complete theoretical study on the effects of independent electric and magnetic fields as well as effects of both fields on the two-photon absorption coefficient in semiconductors. The process of the photon-assisted two-photon electronic transitions in semiconductors, is also considered. Numerical applications on some crystals are given.

Expressions for the two-photon absorption coefficient at the critical points M_0 and M_1 have been derived adopting a three-band model and the time-dependent perturbation theory in an electric field. At photon energies just below the threshold M_0 , the formula for the absorption coefficient tends to an exponential tail of Franz-Keldysh type. Above the threshold it gives an oscillatory structure around its zero-field value and reduces to the usual formula in the absence of the electric field. Using an Electronic Computer IBM 1130 at Ain Shams University, a numerical application has been made to the case of anisotropic ZnS crystals.

From the present theory of the two-photon absorption in the electric field, $|E|$, we get the following conclusions:

1) Deviations of at most 10 % from the zero-field two-photon absorption curves are observed in an electric field of 5×10^4 V/cm. 2) These deviations take the form of oscillations around the two-photon zero-field curve. The period of oscillations, τ , is approximately represented by $\tau \propto |E|^{2/3}$ at the edge point M_0 . 3) The theory reduces to known results of Franz-Keldysh effect before the absorption edge M_0 , and after the edge to the usual square-root energy dependence in zero electric field.

For the case of the saddle point M_1 , the two-photon absorption coefficient oscillates around its value at zero field only at energies after the gap, in agreement with the one-photon absorption process.

A theory is developed for indirect two-photon transitions in solids in a magnetic field. Three band models have been introduced, where the electronic transitions are defined by: a) interband-interband indirect transitions in the case of the four-band model; b) interband-intraband indirect transitions in the case of the three-band model, and; c) intraband-intraband indirect transitions in the case of the two-band model. The result shows

that the contributions from these band models agree with a recent experimental result in Insb. The four-band and two-band models give large contributions only at the limits of low and high magnetic fields, respectively.

The effect of temperature, T , on the process of the indirect two-photon absorption is to split up each step of the absorption into two different steps. The first at low temperatures, corresponding to phonon emission, while the second at high temperatures corresponding to phonon absorption.

The effect of crossed electric $|\underline{E}|$ and magnetic $|\underline{H}|$ fields on the two-photon electronic interband transitions in semiconductors is investigated. The approach we follow, is treat the electric field as a small perturbation in the presence of a magnetic field of different strengths. The general behaviour of the two-photon absorption coefficient depends mainly on the ratio: $|\underline{E}|/|\underline{H}|$. A numerical application to the case of Zns shows that: 1) for $|\underline{E}| \leq 10^3$ V/cm and $|\underline{H}| \geq 100$ KG, the magnetic field has the dominant effect. We obtain singularities corresponding to each pair of the Landau levels in the valence and conduction bands

with the corresponding Landau level in an intermediate band. The effect of the electric field is to reduce the value of the energy gap and to destroy the Landau selection rules. (2) As $|E| / |H|$ increases, the electric field becomes more effective. For example for $|H| = 75 \text{ KG}$ and 50 KG we get, first, the absorption starts at much lower energies because of raising up and lowering down the Landau levels in the valence and conduction bands respectively. Second, the separation between such levels decreases in such a way that we obtain the continuum for strong electric field. 3) For $|E| > 6 \times 10^3 \text{ V/cm}$, this procedure fails to give any results. The electronic transition becomes of an electric type and the present approach of the two-photon absorption is no longer valid. Instead we should use another treatment to obtain the Airy function behaviour of the electric field effect.

In parallel electric and magnetic fields, we obtained an electric type two-photon electronic interband transitions as for the case of one-photon absorption in parallel fields.

INTRODUCTION

Multiphoton absorption is that process in which at least two photons are simultaneously absorbed per elementary interaction of electromagnetic radiation with matter. The probability of such elementary interactions cannot formally be represented as a product of the probabilities of absorption of individual photons.

The theory of multiphoton absorption has been considered for the first time during the early years of quantum mechanics, but experimental techniques at that time did not make it possible to observe many of the predicted phenomena. The main difficulty in the study of multiphoton processes is their exceedingly low probability compared with ordinary single photon processes. In order to observe multiphoton processes it is necessary to have radiation densities that are many orders of magnitude larger than those used for the observation of single photon processes.

Two-photon absorption is a nonlinear optical phenomenon in which two photons are simultaneously absorbed in an electronic transition. The two photons are chosen such as the electronic system could not absorb one photon alone to make a transition. The conservation of energy demands

that the sum of the energies of the two photons be equal to the energy of the electronic transition.

The matrix elements of a two-photon transition, unlike single photon absorption, is not zero only for initial and final states with the same wave-function symmetry. Accordingly, single photon transitions are forbidden in this case, and vice-versa. Two-photon spectroscopy, therefore, is capable for giving information about a solid not easily available from one-photon spectroscopy. For example, in electronic systems possessing inversion symmetry, direct examination of states having the same parity as the ground state is permitted.

By the recent developments of optical lasers with their out-put radiation, the properties of the two-photon absorption at band edges of solids have been extensively investigated.

The general theory of the two-photon electronic transition was early studied by Göppert-Mayer⁽¹⁾. Braunstein⁽²⁾ developed a theory to calculate the absorption coefficient near band edges in semiconductors. Braunstein's model

depends upon the excitation of an electron from an initial valence band, v , to a final conduction band, c , through an intermediate state, ℓ , by the simultaneous absorption of two photons $\hbar\omega_1$ and $\hbar\omega_2$. Basov and co-workers⁽³⁾ proposed another model for compounds in which the valence band, v , consists of three subbands denoted by v_1, v_2, v_3 . Electronic interband transitions from the v_i valence bands ($i = 1, 2, 3$) to the conduction band, c , are possible but with different probabilities depending on the wave-vectors ϵ_{-1} and ϵ_{-2} of the incident beams. The total probability of forming an electron-hole pair by the two-photon excitation consists of three terms each of which describes the probability of formation of an electron in the conduction band and a hole in one of the three valence bands. Gold et al.,⁽⁴⁾ developed a theory of two-photon transitions between a given pair of impurity levels of the same parity.

In a number of semiconducting crystals, the maximum of the valence band and the minimum of the conduction band are located in different points in the Brillouin zone (BZ). Phonon-assisted two-photon transitions may become important and directed towards lowest energy gaps

of the band structure. Kovarskii et al.,⁽⁵⁾ developed a theory of the phonon-assisted two-photon transitions in the case of ZnS crystals. The process has been described as a third-order process accompanied by the scattering of an electron to the conduction band and leaving a hole in the valence band. Their results were in good agreement with observations obtained by other authors⁽⁶⁾. Singolani et al.,⁽⁷⁾ observed the two-photon conductivity in ZnS crystals. Photocurrent characteristics have been explained by a two-stage absorption process in which the first photon reaches an excited state with a reasonably long lifetime and the second photon completes the transition. By the simultaneous absorption of two laser beams, they predicted the creation of an electron-hole pair as previously observed by Yee⁽⁸⁾ and Panizza⁽⁹⁾.

Bassani and Hassen⁽¹⁰⁾ developed a theory of the two-photon absorption near the critical points M_0 and M_1 . For the point M_1 , they noted a transition peak similar to that obtained in the case of one-photon transitions but it is sharper due to additional energy terms in the predicted formalism. Phonon-assisted two-photon transitions have been investigated in the cases of Ge and GaP

for the band-to-band (11) and exciton (12) transitions.

Polarization dependence of the process of the two-photon transitions have been investigated by many authors (13). Experimental observations have been obtained by Braunstein et al.,⁽¹⁴⁾ in the case of CdS crystals, Hopfield et al.,⁽¹⁵⁾ in the cases of KI and CsI, and Fröhlich et al.,⁽¹⁶⁾ in the case of Anthracene powder.

A shift of the absorption edge towards lower energies in the presence of an electric field $|E|$, was discovered theoretically by Franz⁽¹⁷⁾ and Keldysh⁽¹⁸⁾ and observed by many others⁽¹⁹⁾. Tharmalingam⁽²⁰⁾ used stationary wave-functions to compute interband matrix elements near the threshold M_0 in an electric field. The zero-field limiting case of the absorption coefficient has been examined and exponential tails due to Franz-Keldysh mechanism are deduced before the absorption edge. Callaway⁽²¹⁾ showed that above thresholds, optical functions undergo oscillations with a period π . Additional oscillations, orders of magnitude smaller, are predicted with a period γ due to Stark effect. Aspnes⁽²²⁾ investigated the Franz-Keldysh effect around all types of van