PHOTOCONDUCTIVITY OF DIELECTRICS UNDER THE INFLUENCE OF LASER RADIATION

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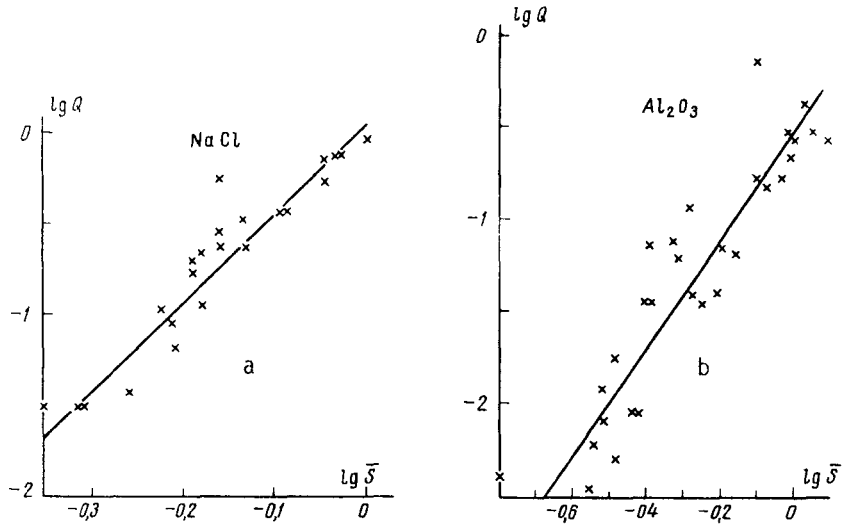
One of the possible methods of recording many-photon absorption in condensed media is to observe the conductivity due to excited electrons. This method was successfully employed to observe two-photon absorption in anthracene [1]. Its main advantage is sensitivity, which should be the decisive factor in the observation of transitions in which more than two or three photons participate. On the other hand, the use of pulsed lasers as excitation sources for the study of the properties of semiconductors and dielectrics has several advantages; for example, in the investigation of transparent dielectrics it becomes unnecessary to use the complicated vacuum-ultraviolet experimental technique.

We present in this communication preliminary results of experiments aimed at observing the photoconductivity induced in uncolored NaCl and Al_2O_3 single crystals by radiation from a ruby laser. The investigated sample was placed in a parallel-plate capacitor charged to a voltage $E_0 \sim 1$ kV. The laser flash induced in the capacitor a charge which was observed on an oscilloscope. To increase the radiation intensity (by a factor of ~5) and to reduce the beam dimensions, a cylindrical telescopic system was used. To avoid effects connected with the space charges, the voltage was applied to the capacitor only just before the flash; the capacitor was short-circuited during the intervals between the flashes.

The figure shows the maximum charge Q against the radiation density S. The straight lines were obtained by statistically reducing the experimental data by the method of least squares, assuming a relation $Q \sim S^n$. The slopes of the lines correspond to $n = 4.9 \pm 0.4$ for NaCl and $n = 3 \pm 0.3$ for Al_2O_3 (the errors indicated are due only to the scatter of the experimental points). The charge growth time was ~0.2 msec in both cases, this being apparently due to the presence of shallow traps.

A possible explanation of the observed effect is many-photon excitation of the electrons in the conduction band [1)]. The first maximum of single-photon absorption in NaCl is located at $\Delta_1 = 7.9$ eV, so that $\langle \Delta_1/\hbar\omega + 1 \rangle = \langle 4.3 + 1 \rangle = 5$ ($\langle x \rangle$ denotes the integer part of x), and the electron excitation probability should be proportional to S^5, which agrees with the figure. There exists an opinion, however, that the absorption at 7.9 eV is connected with exciton production. Thus, Ferguson's experiments [2] have shown that the limit of single-photon conductivity of NaCl is located in the region of the second absorption maximum at $\Delta_2 = 9.4$ eV, which corresponds to $\langle \Delta_2/\hbar\omega + 1 \rangle = \langle 5.3 + 1 \rangle = 6$. This discrepancy can be attributed to the fact that, at the radiation intensity employed, the excitons produced by the absorption of 5 photons



Total charge Q on the capacitor with the sample vs. the average laser radiation density $\bar{S}$ in the case of NaCl (a) and Al_2O_3 (b).

have become ionized almost instantaneously. The absorption band in Al_2O_3 begins at ~6 eV, so that $\langle \Delta/\hbar\omega + 1 \rangle = \langle 3.4 + 1 \rangle = 4$. Here, too, the observed relation $Q \sim S^3$ can be ascribed to stepwise excitation via exciton or impurity levels whose ionization proceeds at a rapid rate.

Let us estimate the probability of n-photon absorption with the aid of the concept of effective energy of the virtual states (cf. e.g., [3]), which we shall assume equal to $\Delta \approx n\hbar\omega$. In this case the transition matrix element is equal to $(H_{sk}^n)/(n-1)!(\hbar\omega)^{n-1}$, where $H = e\vec{r}\cdot\vec{E}/2$. Let the wave function of the initial state be $(\pi a^3)^{-1/2}\exp(-r/a)$, and that of the final state $\exp(i\vec{k}\cdot\vec{r})$, and then if $a^2k^2 \ll 1$ and n is odd

$$|H_{sk}^n| = 2^3\pi^{1/2}(n+1)!\,\frac{n+3}{2}\,a^{n+\frac{5}{2}}\left(\cos\theta\,\frac{eE}{2}\right)^n k, \qquad (1)$$

where θ is the angle between $\vec{E}$ and $\vec{k}$. The transition probability per valence electron is then

$$W = \frac{2^4[n(n+1)(n+3)]^2}{2n+1}\,\omega\left(n - \frac{\Delta}{\hbar\omega}\right)^{3/2}\left(\frac{\hbar\omega}{I_H}\right)^{5/2}\left(\frac{a}{a_0}\right)^5\left(\frac{eaE}{2\hbar\omega}\right)^{2n}, \qquad (2)$$

where $I_H = 13.6$ eV and $a_0 = 0.53$ Å; it is assumed that the effective mass in the conduction band is equal to the mass of the free electron.

The experimental value of W can be estimated in the following manner. The time t_d needed for the electron to traverse the distance d from the illuminated layer to the positive electron is equal to $d/\mu E_0$, where the mobility μ is 0.05 cm^2/V-sec in the case of Al_2O_3 [4] and 12 cm^2/V-sec in the case of NaCl [5], so that t_d is equal to 7×10^{-7} and 2×10^{-4} sec at d = 1 mm and $E_0 = 1.2$ kV. The carrier lifetime τ in NaCl is of the order of 4×10^{-8} sec [5], and therefore $\tau \ll t_d$ (this is apparently valid also in the case of Al_2O_3. In the estimate below we assume $\tau(Al_2O_3) = 10^{-8}$ sec), and if we neglect the thermal ionization of the

traps, then the induced charge is t_d/τ times smaller than the initial charge $eN_0WV\Delta t$ (N_0 is the density of the excited electrons, $V \sim 0.05$ cm^3 the effective volume of the emitting layer of the sample, and $\Delta t \sim 10^{-8}$ sec the effective laser-pulse duration), so that $W_{exp} = Qt_d/eN_0V\Delta t\tau$ [2]. In the case of NaCl we have $Q = 5 \times 10^{-12}$ Coul (at a radiation energy, averaged over the beam cross section, $\bar{S} \approx 100$ MW/cm^2), and if it is assumed that N_0 is equal to the density of the valence electrons (1.3×10^{23} cm^{-3}), then $W_{exp} \sim 0.8 \times 10^{-4}$ sec^{-1}.

From (2) with $n = 5$, $n - \Delta/\hbar\omega = 0.5$, and $a = 2$ Å we get $W_{theor} = 2.5 \times 10^{-48} \cdot (S[W/cm^2])^5$ sec^{-1}, so that $W_{theor} = W_{exp}$ when $S = 500$ MW/cm^2, which is five times the experimental value of $\bar{S}$. This excess can be attributed to the spatial structure of the laser radiation field.

Analogous estimates for Al_2O_3 yield when $\bar{S} \sim 20$ MW/cm^2 a value $W_{exp} = 4.2 \times 10^{-3}$ sec^{-1} (if we assume that N_0 is the density of the valence electrons, $N_0 = 2.5 \times 10^{23}$ cm^{-3}), and $W_{theor} = 1.6 \times 10^{-23} \cdot (S[W/cm^2])^3$ sec^{-1} (for $n = 3$, $n - \Delta/\hbar\omega = 0.5$, $a = 1.4$ Å), so that $W_{theor} = W_{exp}$ when $S = 60$ MW/cm^2, which is three times the value of $\bar{S}$ (the spatial structure plays, naturally, a smaller role when $n = 3$ than when $n = 5$). We note that calculation by the Keldysh formula [6] yields 6 GW in the case of NaCl and 30 MW in the case of Al_2O_3.

The foregoing estimates do not contradict the "many-photon" interpretation of the observed effects. It should be noted that the observation of many-photon absorption in laser media is of interest for the study of the mechanism whereby they become damaged at large generation levels, and for the determination of the limiting laser power. Thus, the experiments allow us to estimate the limiting radiation density S_{max} at which the gain in ruby is offset by three-photon absorption. Let α be the gain, and then $\bar{S}_{max} = (\alpha/\beta)^{1/2}$, where in accord with the described experiments $\beta = 3\hbar\omega N_0 W_{exp}/S^3 \sim 10^{-19}$ W^{-2} cm^3 and for $\alpha = 1$ cm^{-1} we have $S_{max} = 3 \times 10^9$ W/cm^2, which is two orders of magnitude smaller than the value of S_{max} calculated by Bunkin and Prokhorov [7].

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[1] The observed amplitude and time characteristics of the signals cannot be readily related to other possible factors (heating of the sample, the photodielectric effect, nonlinear corrections to the dielectric constant). For example, according to a very simple estimate, the contribution of the thermal effects to the magnitude of the observed signal does not exceed 1% at the lowest radiation level employed.

[2] We disregard here the effect of space charge. A more rigorous calculation of the connection between Q and W calls for additional experiments.