Three- and four-photon excitation of nonequilibrium carriers in broad-band crystals

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Three- and four-photon interband excitation of nonequilibrium charge carriers in NaCl and KCl by picosecond laser pulses in the visible and UV ranges have been detected. The corresponding many-photon absorption coefficients and electron mobilities in the conduction band have been determined.

The many-photon ($k > 2$) excitation of nonequilibrium charge carriers in broad-band insulators (such as alkali-halide crystals) has essentially not been studied until now, despite a strong theoretical and practical interest stemming, in particular, from the problems involving the interaction of a strong laser light with transparent materials. There are only two studies, to our knowledge, in which experimental data on the three- and four-photon excitation of nonequilibrium carriers in alkali-halide crystals have been reported. Brost *et al.*[1] studied the excitonic luminescence in KI ($k = 3$) and Jones *et al.*[2] studied the absorption of light in NaCl by a photoacoustic method ($k = 4$).

In this letter we report the observation of three- and four-photon excitation of nonequilibrium carriers in NaCl and KCl by the picosecond laser photoconductivity method. We were able to detect and clearly identify these processes and to accurately measure the corresponding many-photon absorption coefficients.

The experimental arrangement for detecting the photoconductivity is shown in Fig. 1. The samples of the $3 \times 15 \times 15$-mm crystals to be tested were placed between the electrodes to which a 100-μs "bell-shaped" electric field with an amplitude $U_0 = 4$ kV was applied. This field was synchronized with the exciting light by single pulses of the fourth, third, and second harmonics of a picosecond Nd:Yag laser. The pulse length of the fundamental frequency ($\lambda = 1.064\,\mu$m) was 60 ps. The photoconductivity signal was removed by a cable line with a wave impedance $R = 50\ \Omega$. The signal under study was amplified by a broad-band samplifier and displayed on the screen of a high-speed oscilloscope. The time resolution of the detection channel was 1.5 ns. To eliminate the strong effect of photoionization of the forming F centers on the photoconductivity signal, we carried out all measurements at an intensity of the exciting light well below the threshold at which the laser destroys the test materials and each time in a different part of the crystal. The intensity of the exciting light was increased by contracting the beam by means of a long-focus lens, so that the diameter of the spot at the front side of the sample was 0.5–1 mm. Spatially uniform beams (which are basically necessary in experiments of this sort) with a nearly Gaussian intensity profile at all wavelengths were obtained by carefully filtering the laser light by spatial filters.

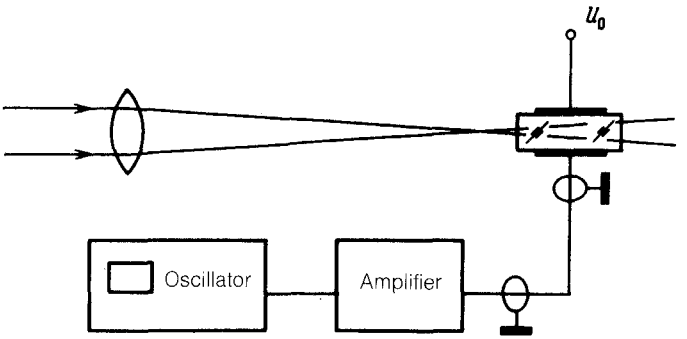

FIG. 1.

For a pulse with an intensity distribution

$$I(r, t) = I_0 \exp\left(-\frac{r^2}{r_0^2}\right) \exp\left(-\frac{t^2}{\tau^2}\right) \tag{1}$$

the total number of electrons, N_k, produced in the conduction band as a result of a k-photon excitation is, in the case of slight absorption,

$$N_k = \frac{\beta_k}{k\hbar\Omega} \frac{\pi r_0^2 l}{k} \frac{\sqrt{\pi}\tau}{\sqrt{k}} I_0^k . \tag{2}$$

Here β_k is the coefficient of the k-photon absorption, Ω is the light frequency, and l is the size of the region of interaction along the direction of the beam. On the other hand, the detected peak value of the voltage of the photoconductivity signal is given by

$$U_{pc} = \alpha N_k \frac{U_0 \mu e}{L^2} R \varphi\left(\frac{\tau_r}{\tau_x}\right), \tag{3}$$

where μ is the mobility, e is the electron charge, L is the electrode spacing, α is a coefficient of order unity in the parallel-plate-capacitor approximation which describes the deviation from this approximation, φ is the signal transmission function, τ_r is the electron recombination time, and τ_x is the detection-channel transient-characteristic time. Equation (3) was found by the nested-image method under the assumption that the external field is not screened by the free carriers. In the experiment we chose the excitation levels in such a way that the given condition would clearly be satisfied.

From a comparison of (2) and (3) we found the expressions for the absorption-coefficient ratios β_2/β_3 and β_2/β_4 (which depend exclusively on the space-time parameters and energy parameters of the exciting light) and we determined their values from the experimental functions $U_{ds} = f(I_0)$. Figure 2 shows the photoconductivity signal of KCl versus the intensity of light with a wavelength $\lambda = 0.266$ μm ($k = 2$), $\lambda = 0.355$ μm ($k = 3$), and $\lambda = 0.532$ μm ($k = 4$). We see that the measurement accuracy of the absorption coefficient ratios is actually determined by the measurement accuracy of the laser-pulse parameters. For the materials which were tested the values of the ratios β_2/β_3 and β_2/β_4 are respectively 4×10^{14} W/cm^2 and 9×10^{26}

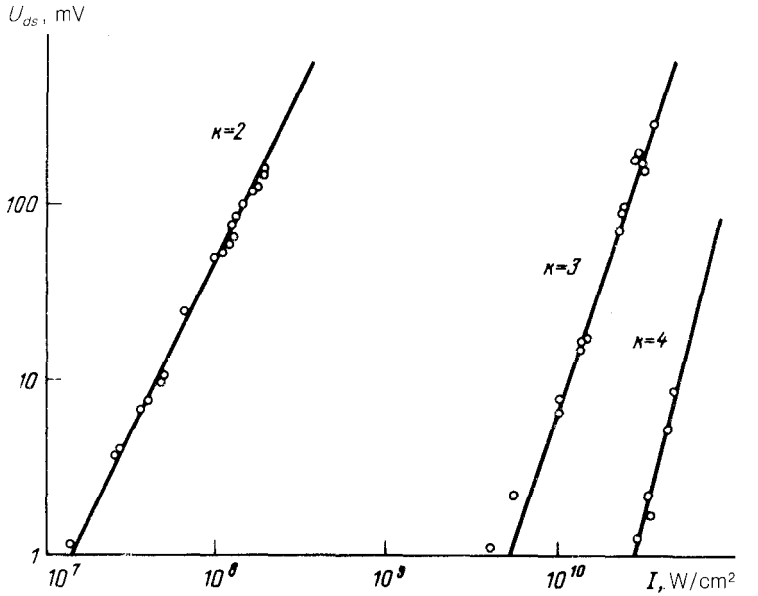

FIG. 2.

W^2/cm^4 for KCl and 5×10^{14} W/cm^2 and 4×10^{26} W^2/cm^4 for NaCl. To determine the absolute values of the coefficients β_3 and β_4, we used the values of β_2 found in Ref. 3 through direct measurement of the light absorption. We thus found $\beta_3 = 5\times10^{-24}$ cm^3/W^2 and $\beta_4 = 2\times10^{-36}$ cm^5/W^3 for KCl and 7×10^{-24} cm^3/W^2 and $\beta_4 = 8\times10^{-36}$ cm^5/W^3 for NaCl.

On the other hand, the drift mobilities of electrons in the conduction band can be determined from (2) and (3) on the basis of the measured values of U_{ds} if the values of β_2 are known. Analysis has shown that $\varphi(\tau_r/\tau_x) = \varphi(5/1.5) = 0.6$ for scale values of the time $\tau_r \simeq 5$ ns measured experimentally. The value of μ can then be determined unambiguously. We found $\mu = 1$ $cm^2/(V\cdot s)$ for KCl and $\mu = 0.9$ $cm^2/(V\cdot s)$ for NaCl. The measured values of the drift mobilities refer to thermalized electrons, since the photoconductivity signal measured under our experimental conditions forms almost entirely after the completion of the laser pulse. The data on the Hall mobility of electrons in alkali-halide crystals found in the literature[4] are $\mu_H = (1\text{–}2)\times10$ $cm^2/(V\cdot s)$. These values, as can be seen, differ substantially (by an order of magnitude) from those measured by us.

In conclusion we note that the picosecond laser photoconductivity method developed by us makes it possible to directly detect the nonequilibrium carriers with high sensitivity (the lowest measurable electron densities are $\sim10^{11}$–10^{12} cm^{-3}), to study their kinetics with a good time resolution (on the order of tens of picoseconds) and high relative accuracy, and to measure the many-photon absorption coefficients in various types of broad-band insulators. In order words, this method, in contrast with

the methods used by Brost *et al.*[1] and Jones *et al.*,[2] is a universal method which can be used to study many-photon excitation of nonequilibrium charge carriers.

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