Many-photon absorption in p-Ge in the submillimeter range

S. D. Ganichev, S. A. Emel'yanov, E. L. Ivchenko, E. Yu. Perlin, and I. D. Yaroshetskiĭ
A. F. Ioffe Physicotechnical Institute, Academy of Sciences of the USSR, Leningrad

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The simultaneous participation of several n-photon absorption processes with $n \lesssim 10$ in nonlinear light absorption has been arranged in a crystal for the first time.

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In experiments on two-photon interband absorption in crystals the photon energy $\hbar\omega$ is usually choosen to satisfy $2\hbar\omega > \varepsilon_g$ but $\hbar\omega < \varepsilon_g$, where ε_g is the width of the energy gap. At $\hbar\omega > \varepsilon_g$ the single-photon absorption coefficient $K^{(1)}(\omega)$ for direct transitions is far larger than the two-photon absorption coefficient $K^{(2)}(\omega)$ (at light intensities up to the threshold for damage to the crystal). In this letter we report the first arrangement of conditions such that the coefficients for n-photon absorption with n less than or on the order of 10 are comparable in magnitude. To arrange these conditions we studied the absorption of light in p-type germanium crystals in the submillimeter range ($\hbar\omega = 13.5$ meV). In the lowest-order perturbation theory the ratio of the two-photon and linear absorption coefficients in the spherical approximation is given by the following expression, for the case of direct transitions between the subbands of heavy and light holes in the Γ_8^+ valence band:

$$\eta = K^{(2)}(\omega) / K^{(1)}(\omega)$$

$$= \frac{4\sqrt{2}\pi e^2}{\omega^2 \; n_\omega m_L} \frac{I}{\hbar\omega} e^{-\epsilon_0/k_BT} (1 + e^{-\hbar\omega/k_BT})(1 + \frac{P_{\rm circ}}{2}), \qquad (1)$$

where I is the light intensity, $\varepsilon_0 = \hbar\omega m_L/m_H - m_L$ is the energy of the heavy holes involved in the single-photon transition, $m_{L,H}$ are the effective masses of the heavy and light holes ($m_L \ll m_H$), n_ω is the refractive index for light at the frequency ω, and $P_{\rm circ}$ is the degree of circular polarization of the light. In calculating $K^{(2)}$, we took into account the intermediate states in other bands and the polarization state of the light, in contrast with Ref. 1. For germanium at a photon energy $\hbar\omega = 13.5$ meV, for a light intensity $I = 1$ MW/cm^2 in the medium, and for the temperature $T = 78$ K the many-photon parameter in $\eta \sim 1$. Perturbation theory cannot be used in this case. We note that for a CO_2 laser, i.e., in the IR range, the many-photon parameter is $\eta < 10^{-3}$ for the same photon flux density. By studying the absorption of light in crystals in the submillimeter range we thus find some new opportunities for studying the nonlinear interaction of electromagnetic radiation with matter.

The present experiments were carried out with an optically pumped, high-power, pulsed, submillimeter NH_3 laser.[2] The pulse length was 40 ns. Figure 1 shows the reciprocal transmission $\Lambda^{-1} = I_0/I(d)$ vs the incident light intensity I_0 from measurements in p-Ge at $T = 78$ K and $p = 5.8 \times 10^{16}$ cm^{-3} for three thicknesses d (the limit-

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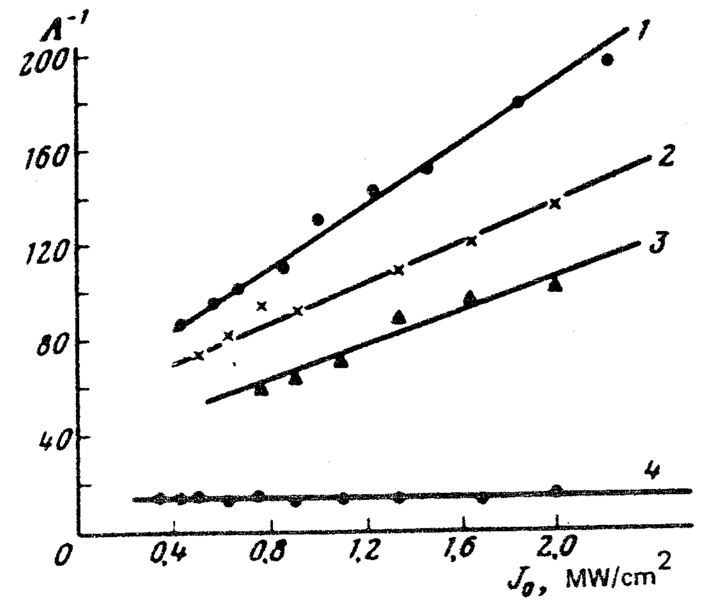

FIG. 1. Reciprocal transmission vs the incident light intensity I_0 at $T = 78$ K. 1, 2, 3—Measurements in p-Ge ($p = 5.8\times10^{16}$ cm^{-3}) for sample thicknesses 0.0695, 0.064, and 0.058 cm, respectively; 4—n-Ge ($n = 3.5\times10^{16}$ cm^{-3}, $d = 0.17$ cm).

ing energy flux density of the laser beam incident on the crystal was 2 MW/cm^2). With increasing I_0 there is a linear increase in the reciprocal transmission; the slope of the curves changes with the thickness d. The change in the reciprocal transmission in the intense radiation field is seen to be comparable to or even greater than Λ^{-1} at low values of I_0. To explain the effect we carried out some comparative experiments in n-type germanium with $n = 3.5\times10^{16}$ cm^{-3} at $T = 78$ and 300 K. We found that in the n-Ge the transmission Λ is independent of I_0 over the intensity range studied (Fig. 1). The Λ (I_0) dependence could thus be due only to a complex valence-band structure and possible direct optical transitions between the heavy- and light-hole subbands.

The basic features of this effect can be described by the following physical model. The absorption of the light actually consists of several simultaneous n-photon-absorption events:

$$K(\omega) = \sum_n K^{(n)}(\omega),$$

where $n = 1, \cdots, n_0$; and n_0 increases with increasing light intensity. Holes with a momentum $\hbar k_n = (2m_T\, n\varepsilon_0)^{1/2}$, determined from energy conservation, participate in the n-photon process. The coefficient for the n-photon absorption, $K^{(n)}$, is determined not only by n-photon processes but also by higher-order processes, in which $(n+m)$ photons are absorbed and m emitted in virtual transitions. These different pathways for n-photon absorption interfere and partially cancel each other out, with the result that the coefficient is no longer proportional to $I^{(n-1)}$, as it is in the lowest-order perturbation theory. It is instead a complex oscillatory function of the intensity I.

For quantitative estimates we can use the following expression for the n-photon absorption probability:

$$w_n(\omega) = nK^{(n)}I / \hbar\omega = \frac{m_L^{3/2}}{\pi \hbar^4} f\ (n\epsilon_0)\ (2n\hbar\omega)^{1/2}\ (M^{(n)})^2, \tag{2}$$

where the constituent matrix element for n-photon absorption is (higher-order processes are taken into account)

$$M^{(n)} \approx \frac{1}{4}\sqrt{\frac{2}{5}}\frac{1}{m_L}\left(\frac{eE}{\omega}\right)^2 \frac{n+1}{n-1} \sum_{m=-\infty}^{\infty} J_m(\rho_2^{(n)}) J_{n-2-2m}(\rho_1^{(n)}),$$

$$\rho_1^{(n)} = \left(\frac{8n}{3m_L \hbar\omega}\right)^{1/2} \frac{eE}{\omega}, \quad \rho_2^{(n)} = \left(\frac{eE}{\omega}\right)^2 \frac{1}{2m_L \hbar\omega} \frac{n^2}{n^2-1}, \tag{3}$$

$f(\epsilon)$ is the hole distribution function, $J_m(p)$ is the Bessel function, and E is the electric field of the light wave. In deriving (2) and (3) we used the method of Ref. 3 and the assumption $|M^{(n)}| \ll \hbar/\tau \ll \hbar\omega$, where τ is the hole lifetime in a state with a given wave vector $\mathbf{k}_n$. At $I = 2$ MW/cm^2, transitions with $n \lesssim 13$ are significant in the absorption. A numerical calculation shows that for an intense laser beam the theoretical intensity dependence of the nonlinear absorption coefficient can be approximated over the intensity range studied by $K(I) = \alpha + \beta I$, where α and β are no longer the single- and two-photon absorption coefficients. According to this $K(I)$ dependence, the reciprocal transmission A^{-1} is a linear function of I_0, in agreement with the experimental observations (Fig. 1). Incorporating a possible heating of free carriers in the field of the light wave does not substantially change α or β. The experimental value $\beta = 72$ cm^{-1} MW^{-1} agrees satisfactorily with the theoretical results.

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