observing Bennett holes, say in spontaneous emission, can be successfully used to determine the radiative and collision widths, to investigate the performance of a laser in a magnetic field, and also to stabilize the frequency of a powerful gas laser. The use of molecular systems pumped by chromatic radiation in accordance with a three-level scheme will apparently permit amplification at new wavelengths with supernarrow lines.

The authors are deeply grateful to S. G. Rautian and A. K. Popov for useful advice during the course of the discussion of interference effects.

[1] W. R. Bennett, Jr., Phys. Rev. 126, 580 (1962).
[2] V. M. Kontorovich and A. M. Prokhorov, Zh. Eksp. Teor. Fiz. 33, 1428 (1957) [Sov. Phys.-JETP 6, 1100 (1958)].
[3] A. Javan, Phys. Rev. 107, 1579 (1957).
[4] S. G. Rautian and I. I. Sobel'man, Zh. Eksp. Teor. Fiz. 41, 456 (1961) [Sov. Phys.-JETP 14, 328 (1962)].
[5] T. Ya. Popova and A. K. Popov, ibid. 52, 1517 (1967) [25, 1007 (1967)].
[6] G. E. Notkin, S. G. Rautian, and A. A. Feoktistov, ibid. 52, 1673 (1967) [25, 1112 (1967)].
[7] V. P. Chebotaev, I. M. Beterov, and V. N. Lisitzyn, Paper at 5th International Conference on Quantum Electronics, Miami, May 1968.

TWO-PHOTON ABSORPTION IN GERMANIUM

B. V. Zubov, L. A. Kulevskii, V. P. Makarov, T. M. Murina, and A. M. Prokhorov
P. N. Lebedev Physics Institute, USSR Academy of Sciences
Submitted 28 November 1968
ZhETF Pis. Red. 9, No. 4, 221-224 (20 February 1969)

This paper is devoted to an investigation of two-photon absorption in germanium single crystals exposed to light from a Q-switched $CaF_2:Dy^{2+}$ laser (λ = 2.36 μ) [1]. We observed for the first time recombination radiation of Ge under two-photon excitation, and measured as well as estimated theoretically the two-photon absorption coefficient.

Figure 1a shows a spectrogram of recombination radiation (indirect transitions) in Ge at T = 77°K under two-photon excitation. Figure 1b shows for comparison the recombination radiation spectrogram obtained under single-photon excitation of Ge by light from a DKSSh-1000 xenon lamp.

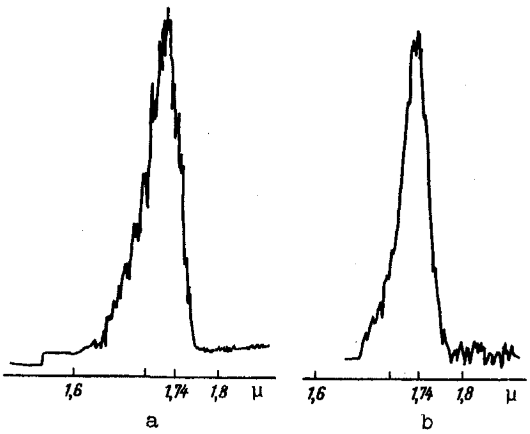

Fig. 1. Spectrum of recombination radiation of Ge (indirect transition): a - two-photon excitation, b - one-photon excitation.

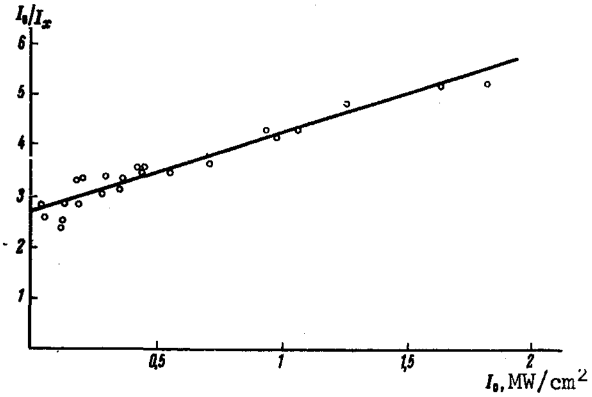

Fig. 2. Dependence of I_0/I_x on I_0.

For a direct determination of the characteristics of two-photon absorption in Ge, we investigated the dependence of the intensity I_x of the light passing through the crystal on the intensity I_0 of the laser emission incident on the crystal. For a crystal of length x, at normal incidence of the light on the crystal, this dependence is given by

$$\frac{I_0}{I_x} = \frac{e^{\alpha x}}{(1-R)^2} + \frac{e^{\alpha x}-1}{\alpha(1-R)}\beta I_0, \tag{1}$$

where α and β are respectively the single-photon and two-photon absorption coefficients, and R is the reflection coefficient.

Figure 2 shows a plot of I_0/I_x against I_0. From the slope of the straight line we obtained the two-photon absorption coefficient $\beta_{exp} = 1$ cm/MW (β was measured at room temperature).

We now proceed to a theoretical estimate of the two-photon absorption coefficient β. The coefficient β is expressed in terms of the probability W/4 of two-photon absorption in the following manner:

$$\beta = \frac{\kappa W}{2c^2 N^2 \hbar\omega}, \tag{2}$$

where W is determined from formulas (2) and (3) of [2], in which it is necessary to put $N_2 = N_1 \equiv N$, $\omega_1 = \omega_2 \equiv \omega$, $\vec{e}_1 = \vec{e}_2 \equiv \vec{e}$, and $\kappa_1 = \kappa_2 \equiv \kappa$ 1). If ϵ_0 is the width of the forbidden band at the point $\vec{k} = 0$ and Δ is the spin-orbit splitting of the valence bands of Ge at $\vec{k} = 0$, then the inequalities $\hbar\omega < \epsilon_0 < 2\hbar\omega < \epsilon_0 + \Delta$ are satisfied for the quantum $\hbar\omega = 0.53$ eV of the $CaF_2:Dy^{2+}$ laser used by us. Therefore the index in (II, 2-3) runs through the values of ℓ and h, i.e., the two-photon absorption proceeds only from the bands of light holes ($\ell \to c$) and heavy holes ($h \to c$). Since germanium is a crystal having a symmetry center, and the single-photon $v \to c$ transition is allowed in it, the composite matrix element (II, 3) is proportional to k at small values of $\vec{k}$ ($kd \ll 1$), i.e., $M_{cv}(\vec{k}) \sim k$. As the intermediate bands j in (II, 3) we consider only the conduction band c, the bands of light and heavy holes ℓ and h, and the valence band s split by the spin-orbit interaction from the ℓ and h bands. The contribution made to β by other j bands is neglected, since they lie more than 10 eV higher and lower than the tops of the ℓ and h bands [3]. The matrix elements $\vec{p}_{jv}(\vec{k})$ contained in the composite matrix element $M_{cv}(\vec{k})$ (II, 3) were calculated by us in the approximation of spherical valence bands $\gamma_2 = \gamma_3 \equiv \bar{\gamma}$ [4].

In the indicated approximation, the coefficient β is expressed in terms of the known parameters of the band spectrum of germanium. (We note that the matrix element $\zeta \equiv \langle S|p_x|X\rangle$ of the interband transition in germanium can be estimated if we know ϵ_0, Δ, and the mass m_c of the electron in the conduction band [5].) Using the values of the parameters determining

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1) Reference [2] is cited henceforth as II. For example, the citation (II, 3) denotes formula (3) of [2]. We use throughout the notation introduced in [2].

β, namely $|\zeta^2| = 1.9 \times 10^{-38}$ g^2cm^2/sec^2, $\hbar\omega = 0.53$ eV, $\Delta = 0.29$ eV, $\kappa = 16$, $m_c = 0.037m$ [5], $\gamma_1 = 13.2\bar{\gamma} = 4.9$ [4], and $\epsilon_0 = 0.8$ eV (at room temperature), we obtained an estimate of the value of the coefficient of the two-photon absorption in germanium, namely $\beta_{theor} = 0.1$ cm/MW.

The discrepancy between the experimentally and theoretically obtained values of β can probably be attributed to two causes. First, in the experiment we were unable to determine the distribution of the intensity of light over the cross section of the beam. The nonequilibrium distribution of the light intensity over the cross section should lead only to an overestimate of β. Second, one cannot exclude the possibility of an appreciable contribution to β from indirect two-photon transitions. Both these factors call for further study.

In conclusion, we are grateful to N. A. Penin and T. I. Galkina for a discussion of the work and V. V. Kostin for help with the measurements.

[1] V. V. Kostin, L. A. Kulevskii, T. M. Murina, A. M. Prokhorov, and A. A. Tikhonov, ZhPS 6, 33 (1967).
[2] M. S. Bespalov, L. A. Kulevskii, V. P. Makarov, A. M. Prokhorov, and A. A. Tikhonov, Zh. Eksp. Teor. Fiz. 55, 144 (1968) [Sov. Phys.-JETP 28, 77 (1969)].
[3] G. Dresselhaus, A. F. Kip, and C. Kittel, Phys. Rev. 98, 368 (1955).
[4] J. M. Luttinger, ibid. 102, 1030 (1956).
[5] L. M. Roth, B. Lax, and S. Zwerdling, ibid. 114, 90 (1959).

SIMULTANEOUS GENERATION OF TWO WAVES OF DOUBLE FREQUENCY IN A NONLINEAR CRYSTAL

M. P. Golovei and G. I. Kosourov
Institute of Crystallography, USSR Academy of Sciences
Submitted 16 December 1968
ZhETF Pis. Red. 9, No. 4, 225-228 (20 February 1969)

The question of effective frequency conversion with satisfaction of the so-called condition of vector synchronism was considered in many papers, for example, as applied to the study of the generation of harmonics in focused beams [1-3] and to the generation of sum and difference frequencies [4,5]. In these studies, sight was lost of the fact that it is possible to satisfy the synchronism condition for several directions simultaneously. In the present paper we demonstrate such a possibility, using the KDP crystal as an example.

When two plane monochromatic electromagnetic waves with frequency ω and with wave vectors $\vec{k}_1$ and $\vec{k}_2$ making an angle Δ propagate in a nonlinear medium, a wave of quadratic polarization is produced and breaks up into three plane polarization waves of frequency 2ω and wave vectors $2\vec{k}_1$, $\vec{k}_1 + \vec{k}_2$, and $2\vec{k}_2$, propagating in the directions of the initial waves and in the direction of the bisector of the angle between their normals. For optically negative crystals such as KDP, the initial waves are ordinary waves and the moduli of the wave vectors of the polarization waves are respectively

$$2k = 2(\omega/c)n_1^o, \quad 2k\cos\frac{\Delta}{2} = 2(\omega/c)n_1^o\cos\frac{\Delta}{2}, \quad 2(\omega/c)n_1^o.$$

Each of these polarization waves can effectively radiate an electromagnetic harmonic wave of frequency 2ω, with the same normal, only if its propagation velocity is equal to the wave