TWO-PHOTON EXCITATION OF LUMINESCENCE IN RUBY CRYSTALS

G. M. ZVEREV, T. N. MIKHAĬLOVA, and V. A. PASHKOV

Submitted February 21, 1968

Zh. Eksp. Teor. Fiz. 55, 141–143 (July, 1968)

Luminescence of the chromium R lines in ruby resulting from two-photon absorption of the radiation from a neodymium laser was observed ($^4A_2 - {}^4F_2$ transition). The dependence of the luminescence intensity on the intensity of the exciting radiation from the neodymium laser was measured, as well as the two-photon absorption coefficient for two polarizations of the exciting radiation.

IN[1,2] the pre-destruction luminescence and nonlinear absorption of radiation of a ruby laser in ruby crystals was associated with multi-photon transitions of the chromium ion. Such transitions can occur both from the ground level $^4A_2$ and from the metastable level ^2E, which complicates the interpretation and measurement of the transition probabilities.

We have observed two-photon excitation of the luminescence of the R lines of chromium in ruby by the radiation of a neodymium laser and measured the effective cross section of the double-quantum transition $^4A_2 \rightarrow {}^4F_2$ (Fig. 1). In the experiments we used a Nd-glass laser with an energy of about 1 J and pulse length 25 ns. It was Q-switched with saturable filters. In order to isolate a definite plane of polarization of the laser radiation, a stack of glass plates at the Brewster angle was placed in the resonator. The laser radiation was focused by a lens with focal length 25 mm in the volume of rectangular samples of ruby of dimensions $10 \times 10 \times 26$ mm. The optical axis of the crystals was perpendicular to their long axis, the content of chromium was 0.05%, and crystals without color centers, with a high threshold of destruction (2×10^{10} W/cm^2),[3] were chosen.

Luminescence was observed in the focal region of the lens with a minimum flux of exciting radiation of 4×10^9 W/cm^2. The spectrum and duration of the luminescence under excitation by the Nd laser did not differ from the spectrum and duration of the light measured under excitation in the green band of the ruby absorption.

We measured the dependence of the intensity of luminescence I_l on the intensity of the exciting radiation of the Nd laser I_e for two polarizations of the exciting radiation. Luminescence in the direction perpendicular to the incident radiation was isolated by interference filters and registered by an FÉU-28 photomultiplier. The signal from the photomultiplier was fed to an S-1-18 oscillograph and photographed. The intensity of the incident radiation was measured with a coaxial element on an S-1-11 oscillograph. The dependence of $\log I_l$ on $\log I_e$ for excitation by the ordinary ray is shown in Fig. 2. The straight line in the figure was obtained by the method of least squares assuming $I_l \sim I_e^n$. The value of $n = 2.0 \pm 0.12$ corresponding to this line is evidence of two-quantum excitation of the luminescence.

From the energy level diagram for Cr in ruby (Fig. 1) it follows that the excitation of luminescence occurs as a two-photon transition from level $^4A_2$ to

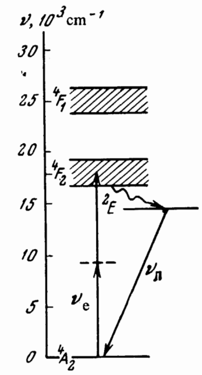

FIG. 1.

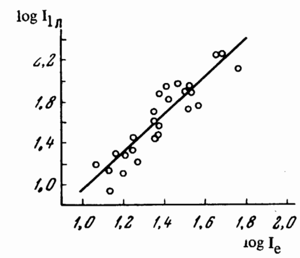

FIG. 2.

level $^4F_2$. Single-quantum absorption of radiation with frequency 9400 cm^{-1} by Cr ions in ruby is not observed; thus, a stepwise mechanism of exchange is excluded. With the powers that we used we observed no generation of the second harmonic of the exciting radiation in crystals of ruby and leucosapphire, so that the process of excitation by the harmonic is likewise excluded.

Measurement of the dependences of I_l on I_e for two directions of polarization of the exciting radiation with respect to the optic axis of the crystal allowed determination of the ratio of the cross sections of double-photon absorption for the ordinary ($\sigma_0^{(2)}$) and extraordinary ($\sigma_e^{(2)}$) rays: $\sigma_0^{(2)}/\sigma_e^{(2)} = 2 \pm 0.3$. A similar dichroism is also observed for the single-photon transition in the green band of ruby.

To determine the absolute magnitudes of the cross sections of two-photon absorption we measured the intensity of the red luminescence at a power of the exciting radiation close to the threshold of destruction of the ruby. The recording circuit in this case was calibrated using the luminescence excited by single-photon absorption of the second harmonic of the Nd laser (obtained by transformation in a KDP crystal). The intensity of the luminescence excited by the second harmonic can be determined by measuring the energy and absorption coefficient of the harmonic in ruby with the quantum yield of the luminescence taken into account, and in this manner the intensity of luminescence under excitation by the Nd laser can be measured.

During each flash of the Nd laser, 10^{12} Cr ions are excited in the ruby volume $V \approx 1.5 \times 10^{-5}$ cm^3 (length of luminescing region 2×10^{-1} cm and diameter 1.0×10^{-2} cm). From this we get the coefficient of two-photon absorption: $k^{(2)} = 5 \times 10^{-5 \pm 0.3}$ cm^{-1} and cross section for two-photon absorption $\sigma^{(2)}$ (cm^2) $= 2 \times 10^{-34 \pm 0.7} \times I_e$ (W/cm^2). The accuracy in the determination of $k^{(2)}$ and $\sigma^{(2)}$ is principally determined by the accuracy of measurement of the power flux I_e and its distribution in the focal region of the lens.

The measured values of $k^{(2)}$ and $\sigma^{(2)}$ are about five orders less than the corresponding values for single-photon absorption in the green band of ruby.

In conclusion, the authors express thanks to N. A. Ékonomov, who participated in some of the experiments.

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Translated by L. M. Matarrese
17