TWO-PHOTON DICHROISM IN NITROBENZENE

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A unique dichroism, whereby the absorption coefficient may depend on the mutual orientation of the polarization of the two photons, can arise in two-photon absorption even in an isotropic medium. Observation of such a dichroism affords an opportunity of revealing the anisotropy of optical transitions in molecules of an isotropic liquid. We have observed its existence for nitrobenzene.

The magnitude of this dichroism can be readily found in the case of limiting molecule anisotropy, when the dipole moments of all the intermediate transitions that determine the two-photon absorption have but a single direction that is rigidly connected to the molecule. Let $\vec{p}$ be a vector directed along the dipole moment of the transitions, and $\vec{E}_1$ and $\vec{E}_2$ the electric vectors of the two absorbed light beams. In this case the coefficient of two-photon absorption is proportional to the quantity $(\vec{p}\cdot\vec{E}_1)^2(p\cdot E_2)^2$. We denote by $\alpha_\parallel$ the absorption coefficient at $\vec{E}_1 \parallel \vec{E}_2$ and $\alpha_\perp$ at $\vec{E}_1 \perp \vec{E}_2$. Then, for our case, $\alpha_\parallel \sim \cos^2(\vec{p}\cdot\vec{E})$ and $\alpha_\perp \sim \cos^2(\vec{p}\cdot\vec{E}_1)\cos^2(\vec{p}\cdot\vec{E}_2)$. It is necessary here to average over all possible directions of $\vec{p}$, since the moduli are randomly oriented. With such an averaging, we get $\alpha_\parallel/\alpha_\perp = 3$.*

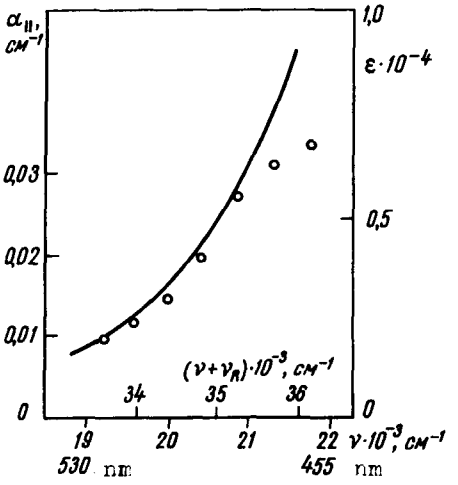
For other models, fore example when intermediate transitions with other momentum directions participate, $\alpha_\parallel/\alpha_\perp$ can assume other values. This quantity is therefore a certain characteristic of the anisotropy of the transitions that determine the two-photon absorption.

The experiment was carried out with a pulsed ruby laser with energy flux approximately 7×10^{25} photons/cm^{2}sec. Light absorption from another source (flash lamp synchronized with the laser pulse) was observed during the time that the giant laser pulse passed through the liquid, i.e., the experimental procedure was similar to that used in [1,2]. Both light beams passed through a cell with a liquid, 10 cm long, in opposite directions. The ruby laser produced linearly polarized light. The direction of polarization of the light from the flash lamp could be established either parallel or perpendicular to the polarization of the laser light by rotating the polarizer.

The light from the flash lamp passed through the cell and then was directed to a monochromator. A photomultiplier was mounted at the exist slit of the monochromator. The photomultiplier signal was fed in parallel to two oscilloscopes. An S1-16 scope registered the relatively slow signal from the flash lamp (the laser signal from another photomultiplier was fed to the other beam). An S1-11 oscilloscope (bandwidth 100 MHz) registered the fast signal, fed from a capacitor which blocked the slow signal almost completely. This fast signal, produced at the instant of the laser pulse, yielded the value ΔI of the decrease in the intensity of the transmitted light. By measuring with the first oscilloscope the intensity I transmitted at that instant, it is possible to determine $\Delta I/I$ and the corresponding absorption coefficient.

We measured the absorption in the spectral region 530 - 450 nm (19 000 - 22 000 cm^{-1}), i.e., in the region where the nitrobenzene is transparent in the conventional sense. With allowance for the ruby-laser frequency, this corresponds to a two-photon transition in the

spectral region near 33 000 - 36 000 cm^{-1}. It has turned out that the edge of the two-photon absorption spectrum of nitrobenzene coincides closely with the edge of its single-photon absorption, with a shift by the value of the ruby-laser frequency (see the figure). The slight deviations on the short-wave side pertain to the region where single-photon absorption already took place and the measurement accuracy was much lower.



Two-photon absorption of nitrobenzene (points, left ordinate scale, lower scale of frequencies ν); single-photon spectrum (continuous curve, right scale, upper scale of frequencies $\nu+\nu_R$).

The spectrum shown in the figure pertains to the case $\vec{E}_1 \parallel \vec{E}_2$. When $\vec{E}_1 \perp \vec{E}_2$ the absorption was much lower, with $\alpha_{\parallel}/\alpha_{\perp} = 1.8 \pm 0.2$.

Thus, two-photon dichroism does not reach in nitrobenzene a value corresponding to the limiting anisotropy. A contribution is apparently made here by transitions with other directions of the dipole moment.

It should be noted that the two-photon absorption in nitrobenzene is itself quite large, in agreement with its observed large nonlinear polarizability [3]. Measurement of two-photon absorption in a number of other liquids is made very difficult by the light scattering caused by variations of the refractive index, due to nonlinear effects and to heating by the laser pulse [3].

[1] J. J. Hopfield, J. M. Worlock, and K. Park, Phys. Rev. Lett. 11, 414 (1963).

[2] D. Frolich and H. Mahr, Phys. Rev. Lett. 16, 895 (1966).

[3] A. P. Veduta, JETP Letters 5, 154 (1967), transl. p. 124.

*It can be noted that this ratio of $\alpha_{\parallel}$ to $\alpha_{\perp}$ recalls the case of polarized luminescence of extremely anisotropic molecules in a liquid, when the oscillators responsible for the absorption and emission coincide in direction. In this case, the polarization ratio for the luminescence intensities is also equal to 3.

MEASUREMENT OF THE REACTION Nd(n, α) WITH RESONANT NEUTRONS

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Reactions of the (n, α) type with heavy nuclei and at low neutron energies have been the subject of relatively few investigations, namely work by Macfarlane [1], Cheifetz [2], and Andreev [3] at thermal neutron energy and the authors' work [4] on the Sm(n, α) reaction in the resonant region. This is caused by methodological difficulties brought about by the extremely small cross sections of the (n, α) reaction and the large gamma background.

We investigated the reaction (n, α) with a natural mixture of neodymium isotopes, and also with enriched Nd^{145} and Nd^{143}, using the IBR pulsed reactor of the Neutron Physics Laboratory of JINR and a multilayer xenon scintillation detector [5]. We measured simultaneously the (n, γ) reaction for each sample (for details see [6]).