Two-photon interband absorption coefficients in tungstate and molybdate crystals

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***Abstract*—Two-photon absorption coefficients were measured in tungstate and molibdate crystals using 25ps laser pumping with 523.5 nm and 349 nm wavelengths.**

Keywords—two-photon absorption coefficients; stimulated Raman scattering; induced one-photon absorption

Two-photon absorption (TPA) coefficients were measured in tungstate and molybdate crystals – $BaWO_4$, KGW, $CaMoO_4$, $BaMoO_4$, $CaWO_4$, $PbWO_4$, and $ZnWO_4$ upon different orientations of excitation polarization with respect to the crystallographic axes (table 1). Trains of 25 ps pulses with variable radiation intensities of second (523.5 nm) and third (349 nm) harmonics of passively mode-locked 1047 nm Nd:YLF laser were used for interband two-photon excitation of the crystals.

TABLE I. TWO-PHOTON ABSORPTION COEFFICIENTS

Name of crystal	β, cm/GW at 349 nm	β, cm/GW at 523.5 nm	Excitation polarization
$PbWO_4$	1.6 1.5	1.2 0.6	$E\perp C_4$ $E\parallel C_4$
$CaWO_4$	0.4	–	$E\perp C_4$
$CaMoO_4$	0.6	0.1	$E\perp C_4$
$BaMoO_4$	1.0	–	unknown
KGW	1.6	–	$E\perp C$
$BaWO_4$	0.6 0.6	–	$E\perp C_4$ $E\parallel C_4$
$ZnWO_4$	2.6 2.4	1.0 0.6	E‖a $E\parallel b(C_2)$
$PbMoO_4$	–	4.0 1.6	$E\perp C_4$ $E\parallel C_4$

It was suggested that in the case, when 349 nm radiation pumping energy exceeds the bandgap width ($h\nu > E_g$), the nonlinear excitation process can be considered as two-step absorption. For both 523.5 and 349 nm wavelengths the initial reciprocal transmission for $PbWO_4$ and $ZnWO_4$ crystals increases from 1.5 to 60 depending on the pulse intensity and the azimuth of the linear excitation polarization with respect to the crystallographic axes (Fig. 1).

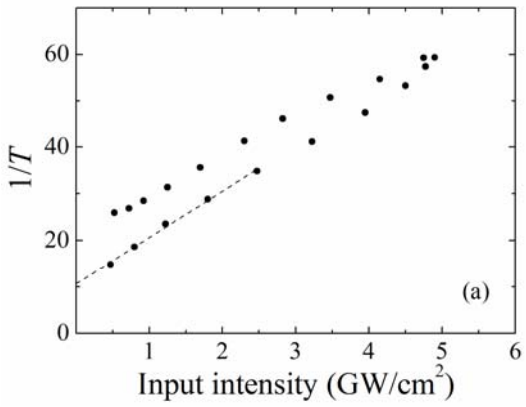

Fig. 1. Dependence of the reciprocal transmission of a $PbWO_4$ crystal 29 mm long on the effective input intensity I_0 at the 349 nm excitation wavelength

The interband two-photon absorption in all the studied crystals induces the following one-photon absorption from the exited states, which affects the nonlinear process dynamics and leads to a hysteresis in the dependence of the transmission on the excitation intensity (Fig. 1). This fact was taken into account when analyzing the experimental dependences of the reciprocal transmission on the excitation intensity, which allowed determining the TPA coefficients in the studied crystals. Laser excitation in the transparency region of the crystals caused stimulated Raman scattering (SRS) not for all the crystals studied. The measured nonlinear coefficients allowed explaining the suppression of SRS in crystals as a result of competition between the SRS and TPA.

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