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***Validity of Two-Level Approximation for Two-Photon Absorption in the Lowest Electronic Transition of Dipolar Molecules***

In dipolar molecules two-photon absorption (2PA) tensor contains a term proportional to the change of permanent dipole moments between the final and ground states. We employ simple asymmetrical potential well models to investigate the contribution of this two-level term relative to other higher-level contributions. Our simple model of dipolar molecules consists in placing N=10 π-electrons in a one-dimensional infinite potential well of width A with asymmetric floor. We study different model potentials for which analytical expressions for the 2PA amplitudes can be obtained. These include linear slope, rectangular step, and antisymmetric sinusoidal potentials. We solve the corresponding Schrödinger equation both analytically and numerically, and show that for a reasonable values of the potential amplitudes (ΔV not larger than 5E1(0) = 5π2ïƒ‘2/ 2mA2), the “two-level” contribution to the 2PA cross section is at least an order of magnitude larger than the contribution from all other terms.