

Nonlinear optical susceptibility of EIT systems with a degenerate Rydberg level

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It has been demonstrated that very large optical nonlinearities can arise in cold Rydberg gases from strong Rydberg–Rydberg interactions. The interactions between atoms excited to a degenerate Rydberg level are described by a large number of molecular potentials which greatly complicates the theoretical treatment of these systems. We here present a method for very accurate calculation of the third order interaction-induced optical nonlinearities that fully includes the angle-dependent mixing of molecular states by the control optical field. In addition, we investigate how an effective potential can be introduced to describe the third-order optical susceptibility arising from the underlying multi-potential Rydberg–Rydberg interactions. We show that a single effective potential can replace a manifold of asymptotically degenerate potentials of the same sign. Therefore, one effective potential has to be defined for attractive interactions and another for repulsive ones. As an example, we have calculated effective C 6 coefficients of $nd + nd$ asymptotes of rubidium and cesium. We compare accurately calculated collisional integrals with those obtained using effective potentials.

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