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The second hyperpolarizability of systems described by the space-fractional Schrödinger equation

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ABSTRACT

The static second hyperpolarizability is derived from the space-fractional Schrödinger equation in the particle-centric view. The Thomas–Reiche–Kuhn sum rule matrix elements and the three-level ansatz determines the maximum second hyperpolarizability for a space-fractional quantum system. The total oscillator strength is shown to decrease as the space-fractional parameter α decreases, which reduces the optical response of a quantum system in the presence of an external field. This damped response is caused by the wavefunction dependent position and momentum commutation relation. Although the maximum response is damped, we show that the one-dimensional quantum harmonic oscillator is no longer a linear system for $\alpha \neq 1$, where the second hyperpolarizability becomes negative before ultimately damping to zero at the lower fractional limit of $\alpha \to 1/2$.

1. Introduction

Kuzyk first discovered limits to the nonlinear optical responses of non-relativistic systems with position dependent potentials [1]. These limits are much greater than the largest responses obtained through experimentation [2]. Another gap has also been observed between the fundamental limits and the best optimized pseudo-potentials [3–5]. These reported gaps may be better understood by investigating more generalized quantum mechanical theories. The relativistically corrected Thomas–Reiche–Kuhn (TRK) sum rule [7,8] led to smaller intrinsic nonlinearities as compared to those calculated in the purely non-relativistic regime, where this decrease in the response is caused by higher-order momentum operators appearing from block diagonalization of the Dirac equation [9]. The first hyperpolarizability of systems described by the space-fractional Schrödinger equation has also been investigated [10].

Laskin discovered the space-fractional Schrödinger equation by generalizing the path integral formulation using a Lévy-type path [11]. Laskin further investigated the space-fractional Schrödinger equation, where he formulated a fractional generalization of the Heisenberg uncertainty principal, proved the Hermiticity of the fractional Hamiltonian operator, and determined the energy spectrum of space-fractional, hydrogen-like atoms [12,13]. The kinetic energy in the space-fractional Schrödinger equation depends on fractional momentum operators, which results in a fractional derivative. The Riesz fractional derivative [14] $\left(-\nabla^2\right)^\alpha$ appears in the space-fractional Schrödinger equation.

In this paper, we derive a sum-over-states expression for the second hyperpolarizability. The limit to the second hyperpolarizability from the space-fractional Schrödinger equation depends on the fractional parameter α, therefore we define an apparent intrinsic second hyperpolarizability to make comparisons between the space-fractional Schrödinger equation and the standard Schrödinger equation. Although the limit to the second hyperpolarizability decreases when α is reduced below unity, we show that some potentials with a small nonlinear optical response can gain a larger response magnitude. This is explicitly shown for the quantum harmonic oscillator, which has a non-zero second hyperpolarizability determined from the fractional Schrödinger equation within the Lévy index $1 < 2\alpha \leq 2$.

2. Theory

The time-independent space-fractional Schrödinger equation with a momentum operator given by the Riesz fractional derivative for a single particle system is given by

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$$\hat{H}_\alpha \psi = E\psi, \tag{1}$$

where $\hat{H}_\alpha$ is the space-fractional Hamiltonian with fractional parameter $1/2 < \alpha \leq 1$, E is the energy, and ψ is the wavefunction. The Hamiltonian considered in this paper has a kinetic energy described by the fractional momentum operator and a spatially dependent potential. The one-dimensional, space-fractional Hamiltonian may be written as

$$\hat{H}_\alpha = \frac{\hat{p}^2}{2m} + V\left(\hat{x}\right), \tag{2}$$

where m is the rest mass and $V(\hat{\boldsymbol{x}})$ is the potential energy.

Respectively, the position and momentum operators are given by

$$\hat{x} = \left(\frac{\hbar}{mc}\right)^{1-\alpha} |x|^\alpha \operatorname{sign}(x) \tag{3}$$

and

$$\hat{p} = -imc\left(\frac{\hbar}{mc}\right)^\alpha \frac{\partial^\alpha}{\partial x^\alpha}. \tag{4}$$

The operator $\partial^\alpha/\partial x^\alpha$ in Eq. (4) is a fractional derivative. There are many definitions of the fractional derivative; simulations performed in this paper are based on a numerical approximation to the Riesz fractional derivative. Note that the dimensions of linear space and momentum are preserved by the constants in Eqs. (3) and (4), where c is the speed of light in vacuum and $\hbar$ is the reduced Planck constant.

We use time-independent perturbation theory of the space-fractional Schrödinger equation in one dimension to determine the scalar, static, second hyperpolarizability in the zero frequency limit [15]. The perturbing term in the Hamiltonian caused by the constant electric field $\mathcal{E}$ is given by

$$\hat{H}_\alpha^{\text{pert}} = e\mathcal{E}\hat{x}, \tag{5}$$

where $\hat{H}_\alpha = \hat{H}_\alpha^{(0)} + \hat{H}_\alpha^{\text{pert}}$ with $\hat{H}_\alpha^{(0)}$ given by Eq. (2). We take a particle-centric approach, where the origin is placed at the expectation value of an electron in a potential well. Note that we may remove subscripts for single electron systems, where multi-electron systems will have different position operators based on the relative displacements between their origins at their respective expectation values. Only for $\alpha \to 1$ does the position operator and perturbation potential become linear.

The fourth-order correction to the energy from time-independent perturbation theory [16] is given by

$$E^{(4)} = \sum_{k,\ell,n}{}' \frac{\left(\hat{H}_\alpha^{\text{pert}}\right)_{0k}\left(\overline{H}_\alpha^{\text{pert}}\right)_{k\ell}\left(\overline{H}_\alpha^{\text{pert}}\right)_{\ell n}\left(\hat{H}_\alpha^{\text{pert}}\right)_{n0}}{E_{k0}E_{\ell 0}E_{n0}} - \sum_{k,\ell}{}' \frac{\left(\hat{H}_\alpha^{\text{pert}}\right)_{0k}\left(\hat{H}_\alpha^{\text{pert}}\right)_{k0}\left(\hat{H}_\alpha^{\text{pert}}\right)_{0\ell}\left(\hat{H}_\alpha^{\text{pert}}\right)_{\ell 0}}{E_{k0}^2 E_{\ell 0}}, \tag{6}$$

where the prime denotes the sum over all states *except* the ground state. Shorthand notation was introduced in Eq. (6), $E_{ij} = E_i^{(0)} - E_j^{(0)}$ and $\overline{\mathcal{O}}_{ij} = \hat{\mathcal{O}}_{ij} - \delta_{ij}\hat{\mathcal{O}}_{00}$ with δ representing the Kronecker delta function, where $\hat{\mathcal{O}}_{ij} = \langle i^{(0)} | \hat{\mathcal{O}} | j^{(0)} \rangle$ is the transition probability of the unperturbed system with $|i^{(0)}\rangle$ being the unperturbed state vector indexed from the ground state $i = 0$.

The static, third-order, scalar response is given by

$$\kappa^{(3)} = \frac{1}{(3)!}\frac{\partial^4}{\partial \mathcal{E}^4} E_0(\mathcal{E})\bigg|_{\mathcal{E}=0}, \tag{7}$$

where E_0 is the ground state energy. Thus, the sum-over-states expression for the static, scalar, second hyperpolarizability given in terms of the transition energies and fractional transition moments is

$$\kappa^{(3)} = 4e^4 \sum_{k,\ell,n}{}' \frac{\hat{x}_{0k}\overline{x}_{k\ell}\overline{x}_{\ell n}\hat{x}_{n0}}{E_{k0}E_{\ell 0}E_{n0}} - 4e^4 \sum_{k,\ell}{}' \frac{\hat{x}_{0k}\hat{x}_{k0}\hat{x}_{0\ell}\hat{x}_{\ell 0}}{E_{k0}^2 E_{\ell 0}}. \tag{8}$$

Because the theory is strictly particle-centric, the expectation value for an electron in its lowest energy state is always zero which allows us to neglect the bar operator in Eq. (8).

The Leibniz rule and chain rule known from integer calculus do not take the same form in fractional calculus, and therefore $[\hat{x}, \hat{p}]$ will not, in general, be equal to the constant $i\hbar$ when $\alpha \neq 1$. The TRK sum rule [17–19] for the mechanical Hamiltonian found in the fractional Schrödinger equation results in a wavefunction-dependent form. For a single electron, the fractional TRK sum rule, calculated from the transition probability of the second commutation relation of the Hamiltonian with the position operator $\langle k^{(0)} | [\hat{x}, [\hat{H}_\alpha^{(0)}, x]] | \ell^{(0)} \rangle$, follows as

$$\sum_{q=0}^{\infty} \hat{x}_{kq}\hat{x}_{q\ell}\left[E_q^{(0)} - \frac{1}{2}\left(E_k^{(0)} + E_\ell^{(0)}\right)\right] = \frac{\hbar^2}{2m}\lambda_\alpha(k,\ell), \tag{9}$$

where

$$\lambda_\alpha(k,\ell) = \int \psi_k^{(0)\dagger}(x)\left[\frac{1}{2}\hat{\xi}^2(x)\frac{\partial^{2\alpha}}{\partial x^{2\alpha}} + \frac{1}{2}\frac{\partial^{2\alpha}}{\partial x^{2\alpha}}\hat{\xi}^2(x) - \hat{\xi}(x)\frac{\partial^{2\alpha}}{\partial x^{2\alpha}}\hat{\xi}(x)\right]\psi_\ell^{(0)}(x)\,dx \tag{10}$$

with $\psi_i^{(0)}(x) = \langle x | i^{(0)} \rangle$ and $\hat{\xi}(x) = |x|^\alpha \operatorname{sign}(x)$. The normalized wavefunction of the unperturbed system has the usual property,

$$\delta_{k\ell} = \int_{-\infty}^{\infty} \psi_k^{(0)\dagger}(x)\,\psi_\ell^{(0)}(x)\,dx. \tag{11}$$

Note that the summation over the state q is introduced into Eq. (10) through the use of closure.

The $(k = 0, \ell = 0)$ TRK sum rule element gives,

$$E_{10}\left|(\hat{x})_{10}\right|^2 = \frac{\hbar^2}{2m}\lambda_\alpha(0,0) - \sum_{q=2}^{\infty} E_{q0}\left|(\hat{x})_{q0}\right|^2. \tag{12}$$

It is clear from Eq. (12) that the largest possible ground state transition moment allowed by the TRK sum rule happens when all of the oscillator strength is in the transition to the first excited state. Setting all terms in the sum for $q \geq 2$ equal to zero gives the maximum value of the ground state to first excited state transition moment,

$$\hat{x}_{10}^{\max} = \frac{\hbar}{\sqrt{2mE_{10}}}\sqrt{\lambda_\alpha(0,0)}, \tag{13}$$

where transition moments of a bound electron described by the space-fractional Schrödinger equation with the Riesz fractional derivative and mechanical Hamiltonian are real, and therefore, $\hat{x}_{ij} = \hat{x}_{ji}$.

The maximum hyperpolarizability derived from the TRK sum rule with only three levels has traditionally been regarded as the fundamental limit. The three-level ansatz appears to hold when the response is near the fundamental limit for a mechanical Hamiltonian in the standard Schrödinger equation. For the case of the fractional Schrödinger equation, the fractional TRK sum rule gives a reduced value which lowers the limit while the transition dipole moment and energy eigenvalue dependencies are of the same form

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