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Fractional approach, quantum statistics, and non-crystalline solids at very low temperatures

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Abstract. We consider nonlocal effects, obtained by incorporating fractional derivatives in the kinetic energy of a conventional Hamiltonian, to analyze physical properties of non-crystalline solids at very low temperature. By using thermal Green functions, we deduce some experimentally observable quantities such as the particle momentum distribution function, the particle energy distribution function, and the specific heat. The agreement between the results obtained for the specific heat and the experimental data suggests that the approach presented here may be useful as a phenomenological model to investigate thermal properties of non-crystalline solids at low temperature.

PACS. 05.30.-d Quantum statistical mechanics – 05.40.-a Fluctuation phenomena, random processes, noise, and Brownian motion – 05.90.+m Other topics in statistical physics, thermodynamics, and nonlinear dynamical systems

1 Introduction

The fractional approach has been used to face many physical situations characterized by a non-usual dynamical behavior. Some common examples are the anomalous transport in disordered systems [1], relaxation to equilibrium in systems (polymers chains and membranes) with long temporal memory [2–5], enhanced diffusion in active intracellular transport [6], random compressible flows [7], and tumor development [8], among many others. These situations have been faced by several approaches as, for instance, the fractional diffusion equations which emerge from the continuous time random walk formalism by considering a long-tailed waiting time distribution or jump length distribution with a diverging variance (Lévy flights). The variety of applications of these equations has also motivated the study of their solutions [9–14] by taking several scenarios into account, such as the analysis of the behavior at the origin [15], systems with trapping or recombination [16], the investigation of the changes produced by the presence of reaction terms [17,18] and the fractional Kramers equation [19] related to them. In this scenario, extensions of Schrödinger equation accomplishing the fractional approach [20–22] have been analyzed by considering the energy spectra of a hydrogenlike atom and of a fractional oscillator in the semiclassical approximation, the parity conservation law [20], quark−antiquark $q\overline{q}$ bound states treated within the non-relativistic potential picture [21] and the quantum scattering problem [22]. However, the quantum statistics (the many particle theory) was not properly considered, leading us to address this work to this issue, which may bring several applications to fractional approach. In this direction, we apply the formalism developed here to investigate the non-crystalline solids at very low temperature, where the thermal properties of these materials differ markedly from those of crystalline ones [23–25]. The differences are evident in the quasilinear temperature dependence of the specific heat and in the approximately quadratic temperature variation of the thermal conductivity. This important problem has been investigated during the last decades by several researchers which have proposed the tunnelling model of two-level systems [25–27], analogy with superfluid helium [28,29], roton-like excitations [30], and soft-potential model [31,32]. Others models such as the framework of a quasi-particle model [33,34] have been recently investigated. Although these models have been used quite successfully for decades to describe non-crystalline solids at very low temperature, the precise physical nature of these systems still deserves a careful investigation [34,35].

2 Fractional approach and quantum statistics

We shall consider the following effective Hamiltonian

$$\widehat{\mathcal{H}}_{Frac} = \frac{1}{2m_\alpha}\int d\mathbf{r}\psi^\dagger(\mathbf{r},t)\left(-\hbar^2\nabla^2\right)^{\alpha/2}\psi(\mathbf{r},t) + \int d\mathbf{r}\psi^\dagger(\mathbf{r},t)U(\mathbf{r},t)\psi(\mathbf{r},t) + \frac{1}{2}\int d\mathbf{r}\int d\mathbf{r}'\psi^\dagger(\mathbf{r},t)\psi^\dagger(\mathbf{r}',t)V(|\mathbf{r}-\mathbf{r}'|)\psi(\mathbf{r},t)\psi(\mathbf{r}',t), \quad (1)$$

where $\psi^\dagger(\mathbf{r},\mathbf{t})$ and $\psi(\mathbf{r},\mathbf{t})$ are second quantized operators,

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m_α is an effective constant, $\left(-\hbar^2\nabla^2\right)^{\alpha/2}\psi(\mathbf{r},t) \equiv \int \frac{d\mathbf{p}}{(2\pi\hbar)^3} e^{i\mathbf{p}/\hbar\cdot\mathbf{r}}|\mathbf{p}|^\alpha\psi(\mathbf{p},t)$ is the quantum Reiz operator [22], $U(\mathbf{r},t)$ represents an external field present in the system and the last term is the interaction between the components of the system. Note that the first term of equation (1), which corresponds to the kinetic energy, has incorporated fractional derivatives. This extension of the kinetic energy to fractional operators incorporates a nonlocal behavior which is not present in the usual form. A direct consequence, via Heisenberg equation for $\psi(\mathbf{r},\mathbf{t})$, verified from equation (1) concerns the dynamical equation for $\psi(\mathbf{r},\mathbf{t})$, which is actually given by

$$i\hbar\frac{\partial}{\partial t}\psi(\mathbf{r},t) = \frac{1}{2m_\alpha}\left(-\hbar^2\nabla^2\right)^{\alpha/2}\psi(\mathbf{r},t) + U(\mathbf{r},t)\psi(\mathbf{r},\mathbf{t}) + \int d\mathbf{r}'V(|\mathbf{r}-\mathbf{r}'|)\psi^\dagger(\mathbf{r}',t)\psi(\mathbf{r}',t)\psi(\mathbf{r},t). \quad (2)$$

Equation (2) is a Schrödinger like equation with fractional derivatives applied on the spatial variable, instead of the usual ones. The fractional Schrödinger equation equation, without the interaction term, was analyzed in several scenarios, in a first quantized perspective, as previously mentioned. In particular, the solution for the free case (i.e., absence of external fields and without interaction) is the Lévy distribution, as we discuss below. By using the Fourier transform, we can simplify equation (2) to

$$i\hbar\frac{\partial}{\partial t}\psi(\mathbf{p},\mathbf{t}) = \frac{|\mathbf{p}|^\alpha}{2m_\alpha}\psi(\mathbf{p},t) + \int d\mathbf{p}U(\mathbf{p},t)\psi(\mathbf{p},t) + \frac{1}{2}V\int d\mathbf{p}'\psi^\dagger(\mathbf{p}',t)\psi(\mathbf{p}',t)\psi(\mathbf{p},t), \quad (3)$$

by considering, for simplicity, the interaction potential $V(|\mathbf{r}-\mathbf{r}'|) = V = \text{constant}$ (Hartree approximation). From this equation, we can observe that the presence of the fractional derivative changes the exponent of the momentum and the solution in the absence of external and interaction potentials is $\psi(\mathbf{p},t) = e^{-i|\mathbf{p}|^\alpha/(2m_\alpha\hbar)t}\psi(\mathbf{p},0)$. As mentioned above, this is the solution that may be related to the Lévy distribution, which is characterized by a power law asymptotic behavior in the $\mathbf{r}$ representation.

Now, let us analyze the quantum statistics which emerges from the above scenario. To perform this analysis, we use the thermal Green function approach, which may be directly related to the dynamical aspects of the $\psi(\mathbf{r},\mathbf{t})$ and may be used to obtain several physical quantities, such as density of particles, specific heat and other thermodynamical quantities. Following [36], we define the one-particle Green function as

$$\mathcal{G}(1,1') = \frac{1}{i}\langle\mathbf{T}(\psi(1)\psi^\dagger(1'))\rangle \quad (4)$$

where the thermodynamic averages, $\langle\cdots\rangle$, are evaluated by taking the grand canonical ensemble into account, $\mathbf{T}$ is the Dyson time-ordering operator and 1 and $1'$ correspond to the variables $\mathbf{r_1},\mathbf{t_1}$ and $\mathbf{r_{1'}},\mathbf{t_{1'}}$, respectively. From this equation, we can define the correlation functions

$$\mathcal{G}^>(1,1') = \frac{1}{i}\langle\psi(1)\psi^\dagger(1')\rangle,$$
$$\mathcal{G}^<(1,1') = \pm\frac{1}{i}\langle\psi^\dagger(1')\psi(1)\rangle, \quad (5)$$

where $>$ and $<$ represent the Green function to $t_1 > t_{1'}$, $\mathcal{G} = \mathcal{G}^>$ and $t_1 < t_{1'}$, $\mathcal{G} = \mathcal{G}^<$. The upper (lower) sign correspond to the bosonic (fermionic) case. From equations (1, 5) it is possible to show that $\mathcal{G}^<(1,1')|_{t_1=0} = \pm e^{\beta\mu}\mathcal{G}^>(1,1')|_{t_1=-i\beta}$ by using the cyclic invariance of the trace ($\text{Tr}(\hat{A}\hat{B}) = \text{Tr}(\hat{B}\hat{A})$). This result shows that the above Green function satisfies the same periodic boundary condition of the usual one [36], in contrast to the one [37] formulated within the Tsallis formalism [38]. Similarly to what is done in the usual case, we may introduce the spectral function, $\mathcal{A}(\mathbf{p},\omega)$, defined as

$$\mathcal{A}(\mathbf{p},\omega) = \mathcal{G}^>(\mathbf{p},\omega) \mp \mathcal{G}^<(\mathbf{p},\omega). \quad (6)$$

By using equation (6) and the boundary condition, we can express $\mathcal{G}^<$ and $\mathcal{G}^>$ as follows:

$$\mathcal{G}^>(\mathbf{p},\omega) = (1 \pm f(\omega))\,\mathcal{A}(\mathbf{p},\omega),$$
$$\mathcal{G}^<(\mathbf{p},\omega) = f(\omega)\mathcal{A}(\mathbf{p},\omega), \quad (7)$$

i.e., in terms of the spectral function, with $f(\omega) = 1/(e^{(\omega-\mu)} \pm 1)$. The equation of motion satisfied by equation (4) is

$$i\hbar\frac{\partial}{\partial t}\mathcal{G}(1,1') = \delta(1-1') + \frac{1}{2m_\alpha}\left(-\hbar^2\nabla^2\right)^{\alpha/2}\mathcal{G}(1,1') + U(\mathbf{r},t)\mathcal{G}(1,1') + \int d\mathbf{r}'V(|\mathbf{r}-\mathbf{r}'|) \times \psi^\dagger(\mathbf{r}',t)\mathcal{G}(1,2;1',2')|_{t_2=t_1}, \quad (8)$$

which may be obtained by employing the Heisenberg equation, where $\mathcal{G}(1,2;1',2')$ is given by

$$\mathcal{G}(12,1'2') = \frac{1}{i^2}\langle\mathbf{T}(\psi(1)\psi(2)\psi^\dagger(2')\psi^\dagger(1'))\rangle\,. \quad (9)$$

Note that to obtain a solution for equation (8) we may consistently approximate equation (9) in accordance with the propagator interpretation of $\mathcal{G}(1,1')$. In this direction, the first approximation to be considered to deal with equation (9) is the Hartree-Fock approximation, given by $\mathcal{G}(12,1'2') = \mathcal{G}(1,1')\;\mathcal{G}(2,2') \pm \mathcal{G}(1,2')\mathcal{G}(2,1')$. In addition, equation (8) recovers the usual form [36] for $\alpha = 2$. From the above results, we may identify $\mathcal{G}^<(\mathbf{p},\omega)$ with the average of particle density with momentum $\mathbf{p}$ and energy ω, i.e.,

$$\mathcal{G}^<(\mathbf{p},\omega) = \langle n(\mathbf{p},\omega)\rangle = \frac{\mathcal{A}(\mathbf{p},\omega)}{e^{\beta(\omega-\mu)} \mp 1} \quad (10)$$

and, as in the usual case, by means of equation (10) we may obtain the pressure for this system by using the equation $P(\beta,\mu) = \int_{-\infty}^{\mu} d\mu'\langle n(\mathbf{p},\omega)\rangle$. Other useful result which may be obtained in this framework concerns the grand canonical potential, $\varXi$. By writing a coupling constant, λ, in front of the interaction energy and, for simplicity, considering the absence of external potential, i.e., $U(\mathbf{r},t)=0$, $\widehat{\mathcal{H}}_{Frac}=\widehat{\mathcal{H}}_{Frac,\,o}+\lambda\widehat{V}$, where

$$\widehat{\mathcal{H}}_{Frac,\,o}=\frac{1}{2m_\alpha}\int d\mathbf{r}\psi^\dagger(\mathbf{r},t)\left(-\hbar^2\nabla^2\right)^{\alpha/2}\psi(\mathbf{r},t) \quad (11)$$

and

$$\widehat{V}=\frac{1}{2}\int d\mathbf{r} \\ \times\int d\mathbf{r}'\psi^\dagger(\mathbf{r},t)\psi^\dagger(\mathbf{r}',t)V(|\mathbf{r}-\mathbf{r}'|)\psi(\mathbf{r},\mathbf{t})\psi(\mathbf{r}',\mathbf{t}), \quad (12)$$

it is possible to write the grand canonical potential as follows:

$$\varXi(\beta,V,\mu;\lambda)=\varXi(\beta,V,\mu;0) \\ -\beta V\int_0^1\frac{d\lambda}{\lambda}\int\frac{d\mathbf{p}}{(2\pi\hbar)^3} \\ \times\int\frac{d\omega}{2\pi}\frac{\omega-|\mathbf{p}|^\alpha/(2m_\alpha)}{2}\mathcal{A}_\lambda(\mathbf{p},\omega)f(\omega). \quad (13)$$

The first term corresponds to the grand canonical potential in the absence of potential energy and the second term is due to the interaction among the particles.

3 Application

As an application, we use the above formalism to explore the temperature dependence of the specific heat of non-crystalline systems (glasses). In addition to the phonons present in these systems, at very low temperature their specific heat shows a non-usual behavior connected with the noncrystalline structure of the material [34,35]. This non-usual temperature behavior of the specific heat, which is usually fitted at low temperature by the expression $\mathcal{C}=\widetilde{\mathcal{A}}T^n+\widetilde{\mathcal{B}}T^3$, has been investigated by several models as mentioned at the introduction. In this direction, we expect that this expression for the specific heat naturally arise from the phenomenological model investigated here, in contrast, for example, to the model of tunnelling states [40].

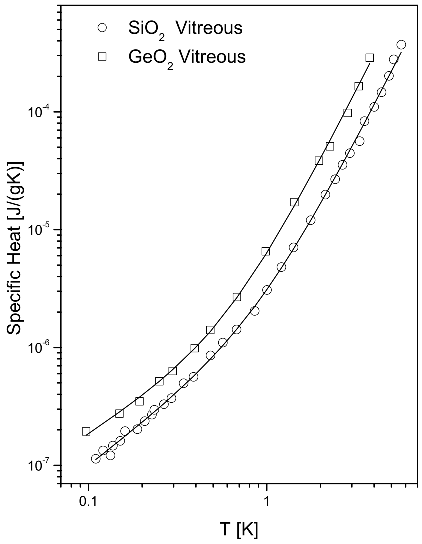

Fig. 1. Behavior of the specific heat for two non-crystalline systems versus temperature. These experimental data (circles and squares) are fitted by the expression $\mathcal{C}=a\,T^{3/\alpha}+b\,T^3$ (α, a and b are parameters) obtained from the formalism developed here based on the fractional approach. The values of parameters a, b and α for SiO$_2$ are 1.40×10^{-6} [J/(g K$^{1+3/\alpha}$)], 1.70×10^{-6}[J/(g K^4)] and 2.60, respectively. For GeO$_2$, we have that $a=2.05\times10^{-6}$[J/(g K$^{1+3/\alpha}$)], $b=4.48\times10^{-6}$[J/(g K^4)] and $\alpha=2.80$.

To test the potentiality of the formalism presented above, we assume that a system in this situation may be described by the following Hamiltonian $\widehat{\mathcal{H}}=\widehat{\mathcal{H}}_{Frac}\bigoplus\widehat{\mathcal{H}}_{Debye}$. The first Hamiltonian $\widehat{\mathcal{H}}_{Frac}$, given by equation (1), is expected to play an important hole at very low temperature, where the non-crystalline characteristics of the system manifest quantum effects, leading to a non-usual behavior for the specific heat. This temperature behavior is different from the one of a crystalline solid, which is well accounted for by the Debye model and may be related to the energy excitation intrinsic to the amorphous state. The second Hamiltonian, $\widehat{\mathcal{H}}_{Debye}$, is related to the presence of phonons in the system and, similarly to what occurs in the conventional crystalline case, gives the Debye contribution for the specific heat, but now including an excess term [39], typical of glasses. For the $\widehat{\mathcal{H}}_{Frac}$, we consider the spectral function $\mathcal{A}(\mathbf{p},\omega)=2\pi\delta(\omega-|\mathbf{p}|^\alpha/(2m_\alpha))$, which implies absence of interactions terms. Thus, the specific heat obtained from the Hamiltonian $\widehat{\mathcal{H}}=\widehat{\mathcal{H}}_{Frac}\bigoplus\widehat{\mathcal{H}}_{Debye}$ is given by $\mathcal{C}=\mathcal{C}_{Frac}+\mathcal{C}_{Debye}$, where $\mathcal{C}_{Frac}\propto T^{\frac{3}{\alpha}}$ and $\mathcal{C}_{Debye}\sim T^3$ for very low temperatures. The parameters α and m_α are obtained from the connection of the above expression for the specific heat (in a simplified form, $\mathcal{C}=a\,T^{3/\alpha}+b\,T^3$) and the experimental data, suggesting that the values of α and m_α are related to the characteristic of the system. In Figure 1, we show the experimental data of two non-crystalline systems (SiO$_2$ and GeO$_2$ obtained from [23]) and the specific heat $\mathcal{C}$ obtained within the previous formalism. The agreement between the specific heat and the experimental data suggests that the formalism based on the fractional derivatives may find applications, as a phenomenological model, in non-crystalline systems at very low temperature. Another important property whose temperature behavior is considered anomalous is the thermal conductivity κ. To obtain the temperature behavior for this quantity we follow the procedure employed in [34] which is based on the kinetic formula $\kappa=\int_0^\infty\mathcal{C}(\omega)cl(\omega)d\omega$ where c is the sound velocity, $l(\omega)$ is the mean free path, and $\mathcal{C}=\int_0^\infty\mathcal{C}(\omega)d\omega$ is the heat capacity. After some calculations by considering, for simplicity, $l(\omega)\propto\omega^\gamma$, it is possible to show that the thermal conductivity may be written as $\kappa=\mathcal{K}_1T^{\frac{3}{\alpha}+\gamma}+\mathcal{K}_2T^{3+\gamma}$ where $\mathcal{K}_1$ and $\mathcal{K}_2$ are constants.

4 Summary and conclusions

To sum up, the quantum statistics emerging from the Hamiltonian given by equation (1) was analyzed. This

Hamiltonian was formulated in order to incorporate the fractional derivative in the kinetic term, instead of the usual one, and to analyze its consequences. It also has as a particular case the fractional Schrödinger worked out in references [22]. The quantum statistics obtained from equation (1) preserves several properties present in the usual case, in particular, the boundary condition. This last feature is not verified to the quantum statistics which emerges from the nonextensive statistics [37]. In this direction, it is necessary to mention that a perturbation theory for the Green functions, obtained for the fractional case, can be formulated as the usual case just by using the boundary condition to obtain the collision effects on the thermal Green function. Other remarkable aspect about this formalism concerns its eventual application, as a phenomenological model, to physical systems such as non-crystalline solids at very low temperature. In this direction, we also expect that this phenomenological model may be used to discuss the energy excitation intrinsic to the amorphous state. The complete scenario could be then as follows. At very low temperature, the dynamics of the glassy system is governed by a Hamiltonian in the form of a sum of two terms: one of them represented by (1), and another one by a anharmonic term like the one discussed in [41]. At low temperature, i.e., near the temperature of the boson peak (between 5 and 50 K), the dynamics is still governed by the complete Hamiltonian (i.e. the sum), but the contribution coming from (1) being negligible, and the term represented by the anharmonic part playing the dominant role. In this framework, to cover a wide range of temperature, the specific heat of the system could be well described by a Hamiltonian written as the sum of two contributions, with each term contributing more significantly or not according to the ranges of temperatures considered. Further investigations should be performed along these lines. Anyway, we hope that the formalism presented above may be useful to investigate systems presenting non-usual behavior and requiring the analysis of a non-conventional Hamiltonian.

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