

Research Article

Regularities of Time-Fractional Derivatives of Semigroups Related to Schrodinger Operators with Application to Hardy-Sobolev Spaces on Heisenberg Groups

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In this paper, assume that $L = -\Delta_{\mathbb{H}^n} + V$ is a Schrödinger operator on the Heisenberg group \mathbb{H}^n , where the nonnegative potential V belongs to the reverse Hölder class $B_{Q/2}$. By the aid of the subordinate formula, we investigate the regularity properties of the time-fractional derivatives of semigroups $\{e^{-tL}\}_{t>0}$ and $\{e^{-t\sqrt{L}}\}_{t>0}$, respectively. As applications, using fractional square functions, we characterize the Hardy-Sobolev type space $H_L^{1,\alpha}(\mathbb{H}^n)$ associated with L . Moreover, the fractional square function characterizations indicate an equivalence relation of two classes of Hardy-Sobolev spaces related with L .

1. Introduction

It is well-known that the Hardy spaces H^p form a natural continuation of the Lebesgue spaces L^p to the range $0 < p \leq 1$. Correspondingly, let I_α and J_α denote the classical Riesz potentials and Bessel potentials, respectively. The Hardy-Sobolev spaces $I_\alpha(H^p)$ and $J_\alpha(H^p)$ can be seen as natural generalizations of homogeneous and inhomogeneous Sobolev spaces. Compared with Hardy spaces, the elements of Hardy-Sobolev spaces are of regularities and have been widely used in the research of partial differential equations, potential theories, complex analysis and harmonic analysis, etc. In the last decades, the theory of Hardy-Sobolev spaces was investigated by many researchers extensively. In [1], Strichartz proved that $I_{n/p}(H^p)$ was an algebra and found equivalent norms for the Hardy-Sobolev space or, more generally, for the corresponding space with fractional smoothness and Lebesgue exponents in the range $p > n/(n+1)$. The trace properties of the space $I_\alpha(H^p)$ were discussed by Torchinsky [2]. Miyachi [3] characterized the Hardy-Sobolev spaces in terms of maximal functions related to the mean oscillation of functions in cubes and obtained a counterpart of previous results of Calderón and of the general theory of De Vore and

Sharpley [4]. For further information on Hardy-Sobolev spaces and their variants on \mathbb{R}^d , or on subdomains, we refer the reader to [5–12].

The development of the theory of Hardy spaces with several real variables was initiated by Stein and Weiss. In [13], by use of square functions, Fefferman and Stein characterized the Hardy spaces $H^p(\mathbb{R}^n)$ for $0 < p \leq 1$. From then on, such characterizations were extended to other settings, see [14–16] and the references therein. Since the 1990s, the theory of Hardy spaces associated with second-order differential operators on \mathbb{R}^n attracts the attention of many researchers and has been investigated extensively, such as [15–22] and the references therein. In recent years, a lot of research has been done on the Hardy spaces associated with operators on the Heisenberg group and other settings, see [23–25].

Let $L = -\Delta_{\mathbb{H}^n} + V$ be a Schrödinger operator, where $\Delta_{\mathbb{H}^n}$ is the sub-Laplacian on \mathbb{H}^n and V belongs to the reverse Hölder class. Let $\{e^{-tL}\}_{t>0}$ be the heat semigroup generated by $-L$ and denote by $K_t^L(\cdot, \cdot)$ the integral kernels. Since V is nonnegative, the Feynman-Kac formula asserts that

$$0 < K_t^L(g, h) \leq \tilde{T}_t(g, h) := (4\pi t)^{-Q/2} e^{-|g^{-1}h|^2/4t}. \quad (1)$$

Lin-Liu-Liu [25] introduced the Hardy space associated with L , which is defined as follows. Let \mathcal{M}_L denote the semigroup maximal function: $\mathcal{M}_L(f)(g) := \sup_{t>0} |T_t^L f(g)|$, $g \in \mathbb{H}^n$. The Hardy space $H_L^1(\mathbb{H}^n)$ associated with L is defined to be

$$H_L^1(\mathbb{H}^n) = \{f \in L^1(\mathbb{H}^n): \mathcal{M}_L(f) \in L^1(\mathbb{H}^n)\}, \quad (2)$$

where $\|f\|_{H_L^1} = \|\mathcal{M}_L(f)\|_{L^1}$.

As an analogue of classical Hardy-Sobolev spaces, we introduce the following Hardy-Sobolev space associated with L on \mathbb{H}^n :

Definition 1. For $\alpha > 0$, the Hardy-Sobolev space $H_L^{1,\alpha}(\mathbb{H}^n)$ is defined as the set of all functions $f \in H_L^1(\mathbb{H}^n)$ such that $L^\alpha f \in H_L^1(\mathbb{H}^n)$ with the norm

$$\|f\|_{H_L^{1,\alpha}} := \|L^\alpha f\|_{H_L^1} + \|f\|_{H_L^1} < \infty. \quad (3)$$

Our motivation is inspired by the following square function characterization of $H_L^1(\mathbb{H}^n)$. For $k \in \mathbb{N}$, let

$$Q_t^k f(g) := t^{2k} \left(\partial_s^k T_s^L \Big|_{s=t^2} f \right)(g). \quad (4)$$

Define the square function associated with $\{Q_t^k\}$ as

$$S_k^L(f)(g) := \left(\int_0^\infty \int_{|g^{-1}h|<t} |Q_t^k(f)(h)|^2 \frac{dh dt}{t^{\mathcal{Q}+1}} \right)^{1/2}. \quad (5)$$

In [16], Hoffmann et al. obtained the following square function characterization of $H_L^1(\mathbb{H}^n)$:

Proposition 2. Let $k \in \mathbb{N}$. A function $f \in H_L^1(\mathbb{H}^n)$ if and only if $f \in L^1(\mathbb{H}^n)$ and the square function $S_k^L(f) \in L^1(\mathbb{H}^n)$. Moreover, $\|f\|_{H_L^1} \sim \|S_k^L(f)\|_{L^1} + \|f\|_{L^1}$.

The goal of this paper is to characterize $H_L^{1,\alpha}(\mathbb{H}^n)$ by the square functions generated by semigroups associated with L . It can be seen from Definition 1 that the elements of $H_L^{1,\alpha}(\mathbb{H}^n)$ have the regularities of order α . Based on this observation, we introduced the following fractional square functions associated with semigroup generated by L . For $\alpha > 0$, let $\partial_t^\alpha K_t^L$ and $\partial_t^\alpha P_t^L$ denote the time-fractional derivatives of the heat kernel and the Poisson kernel, respectively, (cf [26]), i.e.,

$$\begin{cases} \partial_t^\alpha K_t^L(g, h) := \frac{e^{in(m-\alpha)}}{\Gamma(m-\alpha)} \int_0^\infty \partial_t^m K_{t+s}^L(g, h) s^{m-\alpha} \frac{ds}{s}, & m = [\alpha] + 1; \\ \partial_t^\alpha P_t^L(g, h) := \frac{e^{in(m-\alpha)}}{\Gamma(m-\alpha)} \int_0^\infty \partial_t^m P_{t+s}^L(g, h) s^{m-\alpha} \frac{ds}{s}, & m = [\alpha] + 1, \end{cases} \quad (6)$$

For $\alpha > 0$, define the following family of operators:

$$\begin{cases} Q_{\alpha,t}^L(f) := t^{2\alpha} \partial_s^\alpha e^{-sL} \Big|_{s=t^2}(f), & t > 0; \\ D_{\alpha,t}^L(f) := t^\alpha \partial_t^\alpha e^{-t\sqrt{L}}(f), & t > 0. \end{cases} \quad (7)$$

Similar to ([27], Proposition 3.6), the regularities of the kernels of $\{Q_{\alpha,t}^L\}$ and $\{D_{\alpha,t}^L\}$ can be deduced from (6). In this paper, we apply a different method to derive the regularities. In Propositions 10 and 14, we estimate the regularities of $\{t^\alpha L^\alpha e^{-tL}\}$ and $\{t^\alpha L^{\alpha/2} e^{-t\sqrt{L}}\}$, respectively. Then, by the functional calculus, we deduce the following relations:

$$\begin{cases} t^\alpha L^\alpha e^{-tL}(\cdot, \cdot) = t^\alpha \partial_t^\alpha e^{-tL}(\cdot, \cdot); \\ t^\alpha L^{\alpha/2} e^{-t\sqrt{L}}(\cdot, \cdot) = t^\alpha \partial_t^\alpha e^{-t\sqrt{L}}(\cdot, \cdot), \end{cases} \quad (8)$$

see Lemmas 15 and 11. Hence, the desired regularities of $\{Q_{\alpha,t}^L\}$ and $\{D_{\alpha,t}^L\}$ are corollaries of Propositions 10 and 14.

Respect to $Q_{\alpha,t}^L$, we introduce the following fractional square functions:

$$\begin{cases} \mathfrak{g}_{H,\alpha}(f)(g) := \left(\int_0^\infty |Q_{\alpha,t}^L(f)(h)|^2 \frac{dt}{t} \right)^{1/2}; \\ \mathfrak{G}_{H,\alpha}(f)(g) := \left(\int_0^\infty \int_{B(g,t)} |Q_{\alpha,t}^L(f)(h)|^2 \frac{dh dt}{t^{\mathcal{Q}+1}} \right)^{1/2}; \\ \mathfrak{g}_{H,\alpha,\lambda}^*(f)(g) := \left(\int_0^\infty \int_{\mathbb{H}^n} \left(\frac{t}{t+|g^{-1}h|} \right)^{2\lambda} |Q_{\alpha,t}^L(f)(h)|^2 \frac{dh dt}{t^{\mathcal{Q}+1}} \right)^{1/2}. \end{cases} \quad (9)$$

In Section 3.1, we establish the characterizations of $H_L^1(\mathbb{H}^n)$ by the square function defined by (9), see Theorem 20. In Section 3.2, we introduce the fractional square functions as follows:

$$\begin{cases} g_{k,\alpha}^H(f) := \left(\int_0^\infty \left| t^{2k-2\alpha} \frac{\partial^k e^{-sL}}{\partial s^k} \Big|_{s=t^2} |f|^2 \frac{dt}{t} \right)^{1/2}, & k \geq \alpha > 0; \\ \mathfrak{G}_{k,\alpha}^H(f) := \left(\int_0^\infty \int_{B(g,t)} \left| t^{2k-2\alpha} \frac{\partial^k e^{-sL}}{\partial s^k} \Big|_{s=t^2} |f|^2 \frac{dh dt}{t^{\mathcal{Q}+1}} \right)^{1/2}, & k \geq \alpha > 0; \\ g_{k,\alpha,\lambda}^{H,*}(f) := \left(\int_0^\infty \int_{\mathbb{H}^n} \left(\frac{t}{t+|g^{-1}h|} \right)^{2\lambda} \left| t^{2k-2\alpha} \frac{\partial^k e^{-sL}}{\partial s^k} \Big|_{s=t^2} |f|^2 \frac{dh dt}{t^{\mathcal{Q}+1}} \right)^{1/2}, & k \geq \alpha > 0. \end{cases} \quad (10)$$

Let

$$D(M(L)) = \left\{ f \in L^2(\mathbb{H}^n): \int_0^\infty |M(\lambda)|^2 \langle dE_L(\lambda) f, f \rangle < \infty \right\}. \quad (11)$$

For every $f \in D(L^\alpha)$ and $L^\alpha f \in L^2(\mathbb{H}^n) \cap H_L^1(\mathbb{H}^n)$, we prove

$$\mathfrak{g}_{k-\alpha}^H(L^\alpha f) = g_{k,\alpha}^H(f), \quad \mathfrak{G}_{k-\alpha}^H(L^\alpha f) = \mathfrak{G}_{k,\alpha}^H(f), \quad \mathfrak{g}_{k-\alpha,\lambda}^{H,*}(L^\alpha f) = g_{k,\alpha,\lambda}^{H,*}(f), \quad (12)$$

The above relations, together with Theorem 20, indicate that

$$\|L^\alpha f\|_{H_L^\alpha} \sim \|g_{k,\alpha}^H(f)\|_{L^1} \sim \|S_{k,\alpha}^H(f)\|_{L^1} \sim \|g_{k,\alpha,\lambda}^{H,*}(f)\|_{L^1}, \quad (13)$$

see Proposition 23. Finally, in Theorem 24, we obtain the desired characterizations of $H_L^{1,\alpha}(\mathbb{H}^n)$ via the fractional square functions defined in (10): for every $f \in H_L^{1,\alpha}(\mathbb{H}^n)$,

$$\begin{aligned} \|f\|_{H_L^{1,\alpha}} &\sim \|f\|_{H_L^1} + \|g_{k,\alpha}^H(f)\|_{L^1} \sim \|f\|_{H_L^1} + \|S_{k,\alpha}^H(f)\|_{L^1} \\ &\sim \|f\|_{H_L^1} + \|g_{k,\alpha,\lambda}^{H,*}(f)\|_{L^1}. \end{aligned} \quad (14)$$

For the Poisson semigroup, via the operators $\{D_{\alpha,t}^L\}$, we can also obtain the corresponding square function characterizations of $H_L^1(\mathbb{H}^n)$ and $H_L^{1,\alpha}(\mathbb{H}^n)$, see Theorems 21 and 25 for the details.

Remark 3.

- (i) As far as the authors know, even on \mathbb{R}^n , the regularities of the time-fractional derivatives of the heat kernels obtained in Section 2.2 are new. The results obtained in Section 2.3 generalize those of [27] to the setting of Heisenberg groups. Moreover, all results in Sections 2.2 and 2.3 apply to some other operators, for example, the degenerate Schrödinger operators, the Schrödinger operators on stratified Lie groups, and so on
- (ii) Lemma 22 implies that the operators $Q_{\alpha,t}^L$ and $D_{\alpha,t}^L$ can be expressed by the spectrum integral of Schrödinger operator. In the sequel, sometime, we formally denote by $t^\alpha \partial_t^\alpha e^{-tL}$ and $t^\alpha \partial_t^\alpha e^{-t\sqrt{L}}$ by $Q_{\alpha,\sqrt{t}}^L$ and $D_{\alpha,t}^L$, respectively

The paper is organized as follows. In Section 2.1, we give some knowledge to be used throughout this paper. Sections 2.2 and 2.3 are devoted to the regularity estimates of $\{Q_{\alpha,t}^L\}$ and $\{D_{\alpha,t}^L\}$, respectively. In Sections 3.1 and 3.2, we establish the fractional square function characterizations of $H_L^1(\mathbb{H}^n)$ and $H_L^{1,\alpha}(\mathbb{H}^n)$. As an application, we deduce an equivalence of the norms of Hardy-Sobolev spaces associated with L .

1.1. *Notations.* Throughout this article, we will use c and C to denote the positive constants, which are independent of main parameters and may be different at each occurrence. By $B_1 \sim B_2$, we mean that there exists a constant $C > 1$ such that $1/C \leq B_1/B_2 \leq C$.

2. Preliminaries

2.1. *Heisenberg Groups and Hardy Spaces.* The $(2n+1)$ -dimensional Heisenberg group \mathbb{H}^n is the Lie group with

underlying manifold $\mathbb{R}^{2n} \times \mathbb{R}$ with the multiplication

$$(x, t)(y, s) = \left(x + y, t + s + 2 \sum_{j=1}^n (x_{n+j}y_j - x_jy_{n+j}) \right). \quad (15)$$

The Lie algebra of left-invariant vector fields on \mathbb{H}^n is given by

$$X_{2n+1} = \frac{\partial}{\partial t}, X_j = \frac{\partial}{\partial x_j} + 2x_{n+j} \frac{\partial}{\partial t}, X_{n+j} = \frac{\partial}{\partial x_{n+j}} + 2x_j \frac{\partial}{\partial t}, \quad j = 1, 2, \dots, n. \quad (16)$$

The sub-Laplacian $\Delta_{\mathbb{H}^n}$ is defined as $\Delta_{\mathbb{H}^n} = \sum_{j=1}^{2n} X_j^2$. The gradient $\nabla_{\mathbb{H}^n}$ is defined by $\nabla_{\mathbb{H}^n} = (X_1, \dots, X_{2n})$. The left-invariant distance is $d(h, g) = |h^{-1}g|$. The ball of radius r centered at g is denoted by $B(g, r) = \{h \in \mathbb{H}^n : |h^{-1}g| < r\}$ whose volume is given by $|B(g, r)| = c_n r^{2n+2}$, where c_n denotes the volume of the unit ball in \mathbb{H}^n and $2n+2$ is the homogenous dimension of \mathbb{H}^n . Let \mathbb{U}^n be the Siegel upper half-space in \mathbb{C}^{n+1} , i.e.,

$$\mathbb{U}^n = \left\{ z \in \mathbb{C}^{n+1} : \text{Im } z_{n+1} > \sum_{j=1}^n |z_j|^2 \right\}. \quad (17)$$

Then, \mathbb{U}^n is holomorphically equivalent to the unit ball in \mathbb{C}^{n+1} . It is well known that the Heisenberg group \mathbb{H}^n is a nilpotent subgroup of the automorphism group of \mathbb{U}^n , which consists of the translations of \mathbb{U}^n . The Heisenberg group \mathbb{H}^n can be also identified with the boundary $\partial\mathbb{U}^n$ via its action on the origin. We use the Heisenberg coordinates $(g, s) = (x, t, s)$ to denote the points in \mathbb{U}^n , where

$$\begin{cases} x_j + ix_{n+j} = z_j, & j = 1, \dots, n; \\ t = \text{Re } z_{n+1}; \\ s = \text{Im } z_{n+1} - \sum_{j=1}^n |z_j|^2. \end{cases} \quad (18)$$

A nonnegative locally L^q -integrable function V on \mathbb{H}^n is said to belong to the reverse Hölder class $B_q, 1 < q < \infty$, if there exists $C > 0$ such that the reverse Hölder inequality

$$\left(\frac{1}{|B|} \int_B V^q(h) dh \right)^{1/q} \leq \frac{C}{|B|} \int_B V(h) dh \quad (19)$$

holds for every ball $B \in \mathbb{H}^n$. In the sequel, we always assume that $0 \equiv V \in B_{2/2}$.

The following auxiliary function $\rho(g, V) = \rho(g)$ was first introduced by Shen [28] and widely used in the research of function spaces related to Schrödinger operators:

Definition 4. The auxiliary function $\rho(\cdot)$ is defined by

$$\rho(g) := \sup_{r>0} \left\{ r : \frac{1}{r^{Q-2}} \int_{B(g,r)} V(h)dh \leq 1 \right\}, g \in \mathbb{H}^n. \quad (20)$$

The following atomic characterization of $H_L^1(\mathbb{H}^n)$ was obtained by Lin-Liu-Liu [25].

Definition 5. A function a is called a $(1, q)$ -atom of the Hardy space $H_L^1(\mathbb{H}^n)$ related with a ball $B(g_0, r)$ if

- (i) $\text{supp } a \subset B(g_0, r)$;
- (ii) $\|a\|_{L^\infty} \leq |B(g_0, r)|^{1/q-1}$;
- (iii) if $r < \rho(g)$, then $\int_{B(g,r)} a(h)dh = 0$

The atomic norm of $H_L^1(\mathbb{H}^n)$ is defined by $\|f\|_{L\text{-atom},q} := \inf \{ \sum |c_j| \}$, where the infimum is taken over all decompositions $f = \sum c_j a_j$, and a_j are H_L^q -atoms.

Proposition 6. Let $1 \leq q \leq \infty$. The norms $\|f\|_{L\text{-atom},q}$ and $\|f\|_{H_L^1}$ are equivalent, that is, there exists a constant $C > 0$ such that $C^{-1}\|f\|_{H_L^1} \leq \|f\|_{L\text{-atom},q} \leq C\|f\|_{H_L^1}$.

Below, we give some results on the tent spaces introduced by Coifman-Meyer-Stein.

Definition 7. Assume that $0 < p, q < \infty$. The tent space $T_q^p(\mathbb{H}^n)$ is defined as the set of all functions $f(\cdot, \cdot)$ on \mathbb{H}^n satisfying $A_q(f)(\cdot) \in L^p(\mathbb{H}^n)$, where

$$A_q(f) := \left(\int \int_{\Gamma(g)} |f(h, t)|^q \frac{dhdt}{t^{Q+1}} \right)^{1/q}, \quad \Gamma(g) = \{(h, t) : |h^{-1}g| < t\}. \quad (21)$$

Coifman, Meyer, and Stein established the following atomic decomposition of $T_2^1(\mathbb{U}^n)$. A function $a(\cdot, \cdot)$ is called a T_2^1 -atom if (i) a is supported in \tilde{B} for some ball $B \subset \mathbb{H}^n$; (ii) $\int \int_{\tilde{B}} |a(g, t)|^2 (dgdt/t) \leq 1/|B|$.

The following proposition is one of the main results of tent spaces.

Proposition 8. Every element $f \in T_2^1(\mathbb{U}^n)$ can be written as $f = \sum_j \lambda_j a_j$, where a_j are T_2^1 -atoms, $\lambda_j \in \mathbb{C}$, and $\sum_j |\lambda_j| \leq C\|f\|_{T_2^1}$.

2.2. Time-Fractional Derivatives of the Heat Semigroup. In this part, we estimate the time-fractional derivatives of the heat kernel associated with L . For $k \in \mathbb{N}$, define

$$Q_{k,t}^L(g, h) := t^{2k} \partial_s^k K_s^L(g, h) \Big|_{s=t^2}. \quad (22)$$

In ([29], Proposition 2.9), the authors obtained the following estimates about the kernel $Q_{k,t}^L(\cdot, \cdot)$.

Proposition 9.

(i) For $M > 0$, there exists a constant $C_M > 0$ such that

$$|Q_{k,t}^L(g, h)| \leq C_M t^{-Q} e^{-c|g^{-1}h|^2/t^2} \left(1 + \frac{t}{\rho(g)} + \frac{t}{\rho(h)} \right)^{-M}. \quad (23)$$

(ii) Assume that $0 < \delta' \leq \min\{1, \delta\}$. For any $M > 0$, there exists a constant $C_M > 0$ such that, for all $|\omega| < \sqrt{t}$

$$|Q_{k,t}^L(g\omega, h) - Q_{k,t}^L(g, h)| \leq C_M \left(\frac{|\omega|}{t} \right)^{\delta'} t^{-Q} e^{-c|g^{-1}h|^2/t^2} \left(1 + \frac{t}{\rho(g)} + \frac{t}{\rho(h)} \right)^{-M}. \quad (24)$$

(iii) For any $M > 0$, there exists a constant $C_M > 0$ such that

$$\left| \int_{\mathbb{H}^n} Q_{k,t}^L(g, h)dh \right| \leq \frac{C_M (t/\rho(g))^{\delta'}}{(1+t/\rho(g))^M}. \quad (25)$$

Denote by $\tilde{Q}_{\alpha,t}^L(\cdot, \cdot)$ the kernel of $t^\alpha L^\alpha e^{-tL}$. In the following proposition, we investigate the regularities of $\tilde{Q}_{\alpha,t}^L(\cdot, \cdot)$.

Proposition 10. Let $\alpha > 0$.

(i) For $M > 0$, there exists a constant $C_M > 0$ such that

$$|\tilde{Q}_{\alpha,t}^L(g, h)| \leq C_M \min \left\{ \frac{1}{t^{Q/2}}, \frac{t^{\alpha/2}}{|g^{-1}h|^{Q+\alpha}} \right\} \left(1 + \frac{\sqrt{t}}{\rho(g)} + \frac{\sqrt{t}}{\rho(h)} \right)^{-M}. \quad (26)$$

(ii) Assume that $0 < \delta' \leq \delta$ with $0 < \delta' < \alpha$. For any $M > 0$, there exists a constant $C_M > 0$ such that for all $|\omega| \leq \sqrt{t}$

$$\begin{aligned} |\tilde{Q}_{\alpha,t}^L(g\omega, h) - \tilde{Q}_{\alpha,t}^L(g, h)| &\leq C_M \left(\frac{|\omega|}{\sqrt{t}} \right)^{\delta'} \min \\ &\cdot \left\{ \frac{1}{t^{Q/2}}, \frac{t^{\alpha/2}}{|g^{-1}h|^{Q+\alpha}} \right\} \left(1 + \frac{\sqrt{t}}{\rho(g)} + \frac{\sqrt{t}}{\rho(h)} \right)^{-M}. \end{aligned} \quad (27)$$

(iii) For any $M > 0$, there exists a constant $C_M > 0$ such that

$$\left| \int_{\mathbb{H}^n} \tilde{Q}_{\alpha,t}^L(g, h) dh \right| \leq \frac{C_M(\sqrt{t}\rho(g))^{\delta'}}{(1 + \sqrt{t}\rho(g))^M}. \quad (28)$$

Proof.

(i) The proof of (i) is divided into the following two cases.

Case 1. $\alpha \in (0, 1)$. For this case, it follows from functional calculus that

$$t^\alpha L^\alpha e^{-tL} = t^\alpha \int_0^\infty \int_0^s \partial_r e^{-(t+r)L} \frac{dr ds}{s^{1+\alpha}}. \quad (29)$$

By (i) of Proposition 9, we obtain

$$\begin{aligned} \left| \tilde{Q}_{\alpha,t}^L(g, h) \right| &\leq t^\alpha \int_0^\infty \int_0^s \left| Q_{\sqrt{t+r}}^L(g, h) \right| \frac{dr ds}{t+r s^{1+\alpha}} \\ &\leq t^\alpha \int_0^\infty \int_0^s \frac{e^{-|g^{-1}h|^2/(t+r)}}{(t+r)^{\mathcal{Q}/2}} \left(1 + \frac{\sqrt{t+r}}{\rho(g)} \right)^{-M} \\ &\quad \cdot \left(1 + \frac{\sqrt{t+r}}{\rho(h)} \right)^{-M} \frac{dr ds}{t+r s^{1+\alpha}}. \end{aligned} \quad (30)$$

On the one hand, a direct computation gives

$$\begin{aligned} \left| \tilde{Q}_{\alpha,t}^L(g, h) \right| &\leq t^\alpha \int_0^\infty \int_0^s \frac{1}{(t+r)^{\mathcal{Q}/2}} \left(\frac{\sqrt{t+r}}{\rho(g)} \right)^{-M} \left(\frac{\sqrt{t+r}}{\rho(h)} \right)^{-M} \frac{dr ds}{t+r s^{1+\alpha}} \\ &\leq t^\alpha \rho(g)^M \rho(h)^M \int_0^\infty \left(\int_r^\infty \frac{ds}{s^{1+\alpha}} \right) \frac{1}{(t+r)^{\mathcal{Q}/2+M+1}} dr \\ &\leq t^{-\mathcal{Q}/2} \left(\frac{\sqrt{t}}{\rho(g)} \right)^{-M} \left(\frac{\sqrt{t}}{\rho(h)} \right)^{-M}. \end{aligned} \quad (31)$$

On the other hand, because the heat kernel decays rapidly, we can get

$$\begin{aligned} \left| \tilde{Q}_{\alpha,t}^L(g, h) \right| &\leq t^\alpha \rho(g)^M \rho(h)^M \int_0^\infty \left(\int_0^s \frac{1}{(t+r)^{\mathcal{Q}/2+M+1}} \left(\frac{|g^{-1}h|^2}{t+s} \right)^{-(\mathcal{Q}+\alpha)/2} dr \right) \frac{ds}{s^{1+\alpha}} \\ &\leq \frac{t^\alpha \rho(g)^M \rho(h)^M}{|g^{-1}h|^{\mathcal{Q}+\alpha}} \int_0^\infty \left(\int_r^\infty \frac{ds}{s^{1+\alpha}} \right) (t+r)^{-M-1+\alpha/2} dr \\ &\leq \frac{t^\alpha \rho(g)^M \rho(h)^M}{|g^{-1}h|^{\mathcal{Q}+\alpha}} \int_0^\infty r^{-\alpha} (t+r)^{-M-1+\alpha/2} dr \\ &\leq \frac{t^{\alpha/2}}{|g^{-1}h|^{\mathcal{Q}+\alpha}} \left(1 + \frac{\sqrt{t}}{\rho(g)} \right)^{-M} \left(1 + \frac{\sqrt{t}}{\rho(h)} \right)^{-M}. \end{aligned} \quad (32)$$

Case 2. $\alpha > 1$. Let $\alpha - [\alpha] = \beta$. Write

$$t^\alpha L^\alpha e^{-tL} = t^\alpha L^{[\alpha]} L^\beta e^{-tL} = t^\alpha L^{[\alpha]} \int_0^\infty \int_0^s \partial_r e^{-(t+r)L} \frac{dr ds}{s^{1+\beta}}. \quad (33)$$

Since $m = [\alpha] + 1$, we can get

$$\begin{aligned} t^\alpha L^\alpha e^{-tL} &= t^\alpha L^{[\alpha]} \int_0^\infty \int_0^s (-L) e^{-(t+r)L} \frac{dr ds}{s^{1+\alpha-[\alpha]}} \\ &= t^\alpha \int_0^\infty \int_0^s (-L)^m e^{-(t+r)L} \frac{dr ds}{s^{2+\alpha-m}} \\ &= t^\alpha \int_0^\infty \int_0^s Q_{\sqrt{t+r},m}^L(g, h) \frac{dr ds}{(t+r)^m s^{2+\alpha-m}}. \end{aligned} \quad (34)$$

It can be deduced from (i) of Proposition 9 that

$$\begin{aligned} \left| \tilde{Q}_{\alpha,t}^L(g, h) \right| &\leq t^\alpha \int_0^\infty \int_0^s (t+r)^{-\mathcal{Q}/2} \left(\frac{\sqrt{t+r}}{\rho(g)} \right)^{-M} \\ &\quad \cdot \left(\frac{\sqrt{t+r}}{\rho(h)} \right)^{-M} \frac{dr ds}{(t+r)^m s^{2+\alpha-m}} \\ &\leq t^\alpha \rho(g)^M \rho(h)^M \int_0^\infty \int_0^s (t+r)^{-\mathcal{Q}/2-M-m} dr \frac{ds}{s^{2+\alpha-m}} \\ &\leq t^\alpha \rho(g)^M \rho(h)^M \int_0^\infty \left(\int_r^\infty \frac{ds}{s^{2+\alpha-m}} \right) (t+r)^{-\mathcal{Q}/2-M-m} dr \\ &\leq t^{-\mathcal{Q}/2} \left(1 + \frac{\sqrt{t}}{\rho(g)} \right)^{-M} \left(1 + \frac{\sqrt{t}}{\rho(h)} \right)^{-M}. \end{aligned} \quad (35)$$

Similarly, an application of (i) of Proposition 9 again yields

$$\begin{aligned} \left| \tilde{Q}_{\alpha,t}^L(g, h) \right| &\leq t^\alpha \rho(g)^M \rho(h)^M \int_0^\infty \left(\int_0^s (t+r)^{-\mathcal{Q}/2-M-m} \left(\frac{|g^{-1}h|^2}{t+s} \right)^{-(\mathcal{Q}+\alpha)/2} dr \right) \frac{ds}{s^{2+\alpha-m}} \\ &\leq \frac{t^\alpha \rho(g)^M \rho(h)^M}{|g^{-1}h|^{\mathcal{Q}+\alpha}} \int_0^\infty \left(\int_r^\infty \frac{ds}{s^{2+\alpha-m}} \right) (t+r)^{-M-m+\alpha/2} dr \\ &\leq \frac{t^\alpha \rho(g)^M \rho(h)^M}{|g^{-1}h|^{\mathcal{Q}+\alpha}} \int_0^\infty r^{m-\alpha-1} (t+r)^{-M-m+\alpha/2} dr \\ &\leq \frac{t^{\alpha/2}}{|g^{-1}h|^{\mathcal{Q}+\alpha}} \left(1 + \frac{\sqrt{t}}{\rho(g)} \right)^{-M} \left(1 + \frac{\sqrt{t}}{\rho(h)} \right)^{-M}. \end{aligned} \quad (36)$$

(ii) We first consider the case $\alpha \in (0, 1)$. By (ii) of Proposition 9, we obtain

$$\begin{aligned}
& \left| \tilde{Q}_{\alpha,t}^L(g\omega, h) - \tilde{Q}_{\alpha,t}^L(g, h) \right| \\
& \leq t^\alpha \int_0^\infty \int_0^s \left| Q_{\sqrt{t+r}}^L(g\omega, h) - Q_{\sqrt{t+r}}^L(g, h) \right| \frac{dr}{t+r} \frac{ds}{s^{1+\alpha}} \\
& \leq t^\alpha \int_0^\infty \int_0^s (t+r)^{-\mathcal{Q}/2} \left(\frac{|\omega|}{\sqrt{t+r}} \right)^{\delta'} e^{-|g^{-1}h|^2/t+r} \times \left(1 + \frac{\sqrt{t+r}}{\rho(g)} \right)^{-M} \\
& \quad \cdot \left(1 + \frac{\sqrt{t+r}}{\rho(h)} \right)^{-M} \frac{dr}{t+r} \frac{ds}{s^{1+\alpha}}.
\end{aligned} \tag{37}$$

Changing the order of integration, we obtain

$$\begin{aligned}
& \left| \tilde{Q}_{\alpha,t}^L(g\omega, h) - \tilde{Q}_{\alpha,t}^L(g, h) \right| \\
& \leq t^\alpha \int_0^\infty \int_0^s (t+r)^{-\mathcal{Q}/2} \left(\frac{|\omega|}{\sqrt{t+r}} \right)^{\delta'} \\
& \quad \cdot \left(\frac{\sqrt{t+r}}{\rho(g)} \right)^{-M} \left(\frac{\sqrt{t+r}}{\rho(h)} \right)^{-M} \frac{dr}{t+r} \frac{ds}{s^{1+\alpha}} \\
& \leq t^\alpha |\omega|^{\delta'} \rho(g)^M \rho(h)^M \int_0^\infty (t+r)^{-\mathcal{Q}/2-\delta'/2-M-1} \\
& \quad \cdot \left(\int_r^\infty \frac{ds}{s^{1+\alpha}} \right) dr \leq \left(\frac{|\omega|}{\sqrt{t}} \right)^{\delta'} \left(\frac{\sqrt{t}}{\rho(g)} \right)^{-M} \left(\frac{\sqrt{t}}{\rho(h)} \right)^{-M} t^{-\mathcal{Q}/2}.
\end{aligned} \tag{38}$$

Alternatively, we can also get

$$\begin{aligned}
& \left| \tilde{Q}_{\alpha,t}^L(g\omega, h) - \tilde{Q}_{\alpha,t}^L(g, h) \right| \\
& \leq t^\alpha |\omega|^{\delta'} \rho(g)^M \rho(h)^M \int_0^\infty \int_0^s (t+r)^{-\mathcal{Q}/2-\delta'/2-M-1} \\
& \quad \cdot \left(\frac{|g^{-1}h|^2}{t+r} \right)^{-(\mathcal{Q}+\alpha)/2} \frac{drds}{s^{1+\alpha}} \leq \frac{t^\alpha |\omega|^{\delta'} \rho(g)^M \rho(h)^M}{|g^{-1}h|^{\mathcal{Q}+\alpha}} \int_0^\infty \\
& \quad \cdot (t+r)^{-\delta'/2-M-1+\alpha/2} \left(\int_r^\infty \frac{ds}{s^{1+\alpha}} \right) dr \leq \left(\frac{|\omega|}{\sqrt{t}} \right)^{\delta'} \\
& \quad \cdot \left(\frac{\sqrt{t}}{\rho(g)} \right)^{-M} \left(\frac{\sqrt{t}}{\rho(h)} \right)^{-M} \frac{t^{\alpha/2}}{|g^{-1}h|^{\alpha+\mathcal{Q}}}.
\end{aligned} \tag{39}$$

For $\alpha \geq 1$, by (ii) of Proposition 9, we can get

$$\begin{aligned}
& \left| \tilde{Q}_{\alpha,t}^L(g\omega, h) - \tilde{Q}_{\alpha,t}^L(g, h) \right| \\
& \leq t^\alpha \int_0^\infty \int_0^s \left| Q_{\sqrt{t+r}}^L(g\omega, h) - Q_{\sqrt{t+r}}^L(g, h) \right| \frac{dr}{(t+r)^m} \frac{ds}{s^{2+\alpha-m}} \\
& \leq t^\alpha \int_0^\infty \int_0^s (t+r)^{-\mathcal{Q}/2} \left(\frac{|\omega|}{\sqrt{t+r}} \right)^{\delta'} e^{-|g^{-1}h|^2/t+r} \times \left(1 + \frac{\sqrt{t+r}}{\rho(g)} \right)^{-M} \\
& \quad \cdot \left(1 + \frac{\sqrt{t+r}}{\rho(h)} \right)^{-M} \frac{dr}{(t+r)^m} \frac{ds}{s^{2+\alpha-m}}.
\end{aligned} \tag{40}$$

Similar to the case $\alpha \in (0, 1)$, the rest of the proof can be finished by applying change of order of integration. We omit the details.

(iii) For $\alpha \in (0, 1)$, by (iii) of Proposition 9, we change the order of integration to obtain

$$\begin{aligned}
& \left| \int_{\mathbb{H}^n} \tilde{Q}_{\alpha,t}^L(g, h) dh \right| \leq t^\alpha \int_0^\infty \int_0^s \left| \int_{\mathbb{H}^n} Q_{\sqrt{t+r}}^L(g, h) dh \right| \frac{dr}{t+r} \frac{ds}{s^{1+\alpha}} \\
& \leq t^\alpha \int_0^\infty \int_0^s \left(\frac{\sqrt{t+r}}{\rho(g)} \right)^{\delta'} \frac{1}{(1 + \sqrt{t+r}/\rho(g))^M} \frac{dr}{t+r} \frac{ds}{s^{1+\alpha}}.
\end{aligned} \tag{41}$$

If $\sqrt{t} > \rho(g)$, then

$$\begin{aligned}
& \left| \int_{\mathbb{H}^n} \tilde{Q}_{\alpha,t}^L(g, h) dh \right| \leq t^\alpha \int_0^\infty \int_0^s \left(\frac{\sqrt{t+r}}{\rho(g)} \right)^{\delta'-M} \frac{dr}{t+r} \frac{ds}{s^{1+\alpha}} \\
& \leq t^\alpha \rho(g)^{M-\delta'} \int_0^\infty \left(\int_r^\infty \frac{ds}{s^{1+\alpha}} \right) \frac{dr}{(t+r)^{(M-\delta')/2+1}} \\
& \leq \frac{(\sqrt{t}\rho(g))^{\delta'}}{(1 + \sqrt{t}/\rho(g))^M}.
\end{aligned} \tag{42}$$

If $\sqrt{t} \leq \rho(g)$, then

$$\begin{aligned}
& \left| \int_{\mathbb{H}^n} \tilde{Q}_{\alpha,t}^L(g, h) dh \right| \leq t^\alpha \int_0^\infty \left(\frac{\sqrt{t+r}}{\rho(g)} \right)^{\delta'} \frac{1}{(1 + \sqrt{t+r}/\rho(g))^M} \frac{dr}{(t+r)r^\alpha} \\
& \leq t^\alpha \rho(g)^{-\delta'} \int_0^\infty (t+r)^{\delta'/2-1} \frac{dr}{r^\alpha} \\
& \leq \frac{(\sqrt{t}\rho(g))^{\delta'}}{(1 + \sqrt{t}/\rho(g))^M}.
\end{aligned} \tag{43}$$

For $\alpha \geq 1$, we have

$$\begin{aligned}
& \left| \int_{\mathbb{H}^n} \tilde{Q}_{\alpha,t}^L(g, h) dh \right| \\
& \leq t^\alpha \int_0^\infty \int_0^s \left| \int_{\mathbb{H}^n} D_{\sqrt{t+r}}^L(g, h) dh \right| \frac{dr}{(t+r)^m} \frac{ds}{s^{2+\alpha-m}} \\
& \leq t^\alpha \int_0^\infty \int_0^s \left(\frac{\sqrt{t+r}}{\rho(g)} \right)^{\delta'} \frac{1}{(1 + (\sqrt{t+r}/\rho(g)))^M} \frac{dr}{(t+r)^m} \frac{ds}{s^{2+\alpha-m}}.
\end{aligned} \tag{44}$$

If $\sqrt{t} > \rho(g)$, then

$$\begin{aligned} \left| \int_{\mathbb{H}^n} \tilde{Q}_{\alpha,t}^L(g,h) dh \right| &\leq t^\alpha \rho(g)^{M-\delta'} \int_0^\infty (t+r)^{(\delta'-M)/2-m} \left(\int_r^\infty \frac{ds}{s^{2+\alpha-m}} \right) dr \\ &\leq t^\alpha \rho(g)^{M-\delta'} \int_0^\infty (t+r)^{(\delta'-M)/2-m} r^{m-\alpha-1} dr \\ &\leq \frac{(\sqrt{t}\rho(g))^{\delta'}}{(1+\sqrt{t}\rho(g))^M}. \end{aligned} \tag{45}$$

If $\sqrt{t} \leq \rho(g)$, then

$$\begin{aligned} \left| \int_{\mathbb{H}^n} \tilde{Q}_{\alpha,t}^L(g,h) dh \right| &\leq t^\alpha \int_0^\infty \left(\frac{\sqrt{t+r}}{\rho(g)} \right)^{\delta'} \frac{1}{(1+\sqrt{t+r}\rho(g))^M} \frac{dr}{(t+r)^m r^{\alpha+1-m}} \\ &\leq t^\alpha \rho(g)^{-\delta'} \int_0^\infty (t+r)^{\delta'/2-1} \frac{dr}{r^\alpha} \leq \frac{(\sqrt{t}\rho(g))^{\delta'}}{(1+\sqrt{t}\rho(g))^M}. \end{aligned} \tag{46}$$

The following lemma can be deduced from the functional calculus immediately.

Lemma 11. *Let $\alpha > 0$. The operators $t^\alpha \partial_t^\alpha e^{-tL}$ and $t^\alpha L^\alpha e^{-tL}$ are equivalent.*

Proof. For $\alpha \in (0, 1)$, we have

$$\begin{aligned} t^\alpha L^\alpha e^{-tL} &= t^\alpha \int_0^\infty \int_0^s \partial_r e^{-(t+r)L} \frac{dr ds}{s^{1+\alpha}} = t^\alpha \int_0^\infty \left(\int_r^\infty \frac{ds}{s^{1+\alpha}} \right) \partial_r e^{-(t+r)L} dr \\ &= t^\alpha \int_0^\infty r^{-\alpha} \partial_r e^{-(t+r)L} dr = t^\alpha \partial_t^\alpha e^{-tL}. \end{aligned} \tag{47}$$

For $\alpha > 1$, let $\alpha - [\alpha] = \beta$. Since $m = [\alpha] + 1$, it holds

$$\begin{aligned} t^\alpha L^\alpha e^{-tL} &= t^\alpha L^{[\alpha]} L^\beta e^{-tL} = t^\alpha L^{[\alpha]} \int_0^\infty (-L) e^{-(s+t)L} s^{1-\beta} \frac{ds}{s} \\ &= t^\alpha \int_0^\infty (-L)^m e^{-(t+s)L} s^{m-\alpha} \frac{ds}{s} = t^\alpha \partial_t^\alpha e^{-tL}. \end{aligned} \tag{48}$$

Denote by $Q_{\alpha,t}^L(\cdot, \cdot)$ the integral kernel of $Q_{\alpha,t}^L$. By Proposition 10 and Lemma 11, we have the following result.

Corollary 12. *Let $\alpha > 0$.*

(i) *For $M > 0$, there exists a constant $C > 0$ such that*

$$\left| Q_{\alpha,t}^L(g,h) \right| \leq \frac{Ct^\alpha}{(|g^{-1}h| + t)^{\mathbb{Q}+\alpha}} \left(1 + \frac{t}{\rho(g)} + \frac{t}{\rho(h)} \right)^{-M}. \tag{49}$$

(ii) *Let $0 < \delta' \leq \delta$ with $0 < \delta' < \alpha$. For any $M > 0$ there exists a constant $C > 0$ such that, for all $|\omega| \leq \sqrt{t}$*

$$\begin{aligned} \left| Q_{\alpha,t}^L(g\omega, h) - Q_t^\alpha(g, h) \right| &\leq C \left(\frac{|\omega|}{t} \right)^{\delta'} \frac{t^\alpha}{(|g^{-1}h| + t)^{\mathbb{Q}+\alpha}} \\ &\cdot \left(1 + \frac{t}{\rho(g)} + \frac{t}{\rho(h)} \right)^{-M}. \end{aligned} \tag{50}$$

(iii) *For any $M > 0$, there exists a constant $C > 0$ such that*

$$\left| \int_{\mathbb{H}^n} Q_{\alpha,t}^L(g, h) dh \right| \leq \frac{C(t\rho(g))^{\delta'}}{(1+t/\rho(g))^M}. \tag{51}$$

2.3. Time-Fractional Derivatives of the Poisson Semigroup. In this part, our aim is to give some regularity estimates of the Poisson kernel associated with \sqrt{L} . For $k \in \mathbb{N}$, define $D_{k,t}^L(g, h) := t^k \partial_t^k P_t^L(g, h)$. In ([29], Proposition 2.12), the authors obtained the following estimates about the kernel $D_{k,t}^L(\cdot, \cdot)$.

Proposition 13 (see [29], Proposition 2.12).

(i) *For $M > 0$, there exists a constant $C_M > 0$ such that*

$$\left| D_{k,t}^L(g, h) \right| \leq \frac{C_M t^k}{(t^2 + |g^{-1}h|^2)^{(\mathbb{Q}+k)/2}} \left(1 + \frac{t}{\rho(g)} + \frac{t}{\rho(h)} \right)^{-M}. \tag{52}$$

(ii) *Assume that $0 < \delta' \leq \min \{1, \delta\}$. For any $M > 0$ there exists a constant $C_M > 0$ such that, for all $|\omega| < t$*

$$\begin{aligned} \left| D_{k,t}^L(g\omega, h) - D_{k,t}^L(g, h) \right| &\leq C_M \left(\frac{|\omega|}{t} \right)^{\delta'} \frac{t^k}{(t^2 + |g^{-1}h|^2)^{(\mathbb{Q}+k)/2}} \\ &\cdot \left(1 + \frac{t}{\rho(g)} + \frac{t}{\rho(h)} \right)^{-M}. \end{aligned} \tag{53}$$

(iii) *For any $M > 0$, there exists a constant $C_M > 0$ such that*

$$\left| \int_{\mathbb{H}^n} D_{k,t}^L(g, h) dh \right| \leq C_M \frac{(t\rho(g))^{\delta'}}{(1+t/\rho(g))^M}. \tag{54}$$

Denote by $\tilde{D}_{\alpha,t}^L(\cdot, \cdot)$ the kernel $t^\alpha L^{\alpha/2} P_t^L(\cdot, \cdot)$. Similar to Proposition 10, we have

Proposition 14. *Let $\alpha > 0$.*

(i) *For every M , there is a constant C_M such that*

$$\left| \tilde{D}_{\alpha,t}^L(g, h) \right| \leq C_M \min \left\{ \frac{1}{t^{\mathcal{Q}}}, \frac{t^\alpha}{|g^{-1}h|^{\mathcal{Q}+\alpha}} \right\} \left(1 + \frac{t}{\rho(g)} + \frac{t}{\rho(h)} \right)^{-M}. \quad (55)$$

(ii) Assume that $0 < \delta' \leq \delta$ with $0 < \delta' < \alpha$. For any $M > 0$ there exists a constant $C > 0$ such that for all $|\omega| \leq t$

$$\left| \tilde{D}_{\alpha,t}^L(g\omega, h) - \tilde{D}_{\alpha,t}^L(g, h) \right| \leq C_M \left(\frac{|\omega|}{t} \right)^{\delta'} \min \left\{ \frac{1}{t^{\mathcal{Q}}}, \frac{t^\alpha}{|g^{-1}h|^{\mathcal{Q}+\alpha}} \right\} \cdot \left(1 + \frac{t}{\rho(g)} + \frac{t}{\rho(h)} \right)^{-M}. \quad (56)$$

(iii) For any $M > 0$, there exists a constant $C_M > 0$ such that

$$\left| \int_0^\infty \tilde{D}_{\alpha,t}^L(g, h) dh \right| \leq \frac{C_M(t\rho(g))^{\delta'}}{(1+t\rho(g))^M}. \quad (57)$$

Proof. Let us prove (i) first. The following two cases are considered.

Case 1. $\alpha \in (0, 1)$. By the functional calculus, we obtain

$$t^\alpha L^{\alpha/2} e^{-t\sqrt{L}} = t^\alpha \int_0^\infty \int_0^s \partial_r e^{-(t+r)\sqrt{L}} \frac{dr ds}{s^{1+\alpha}}, \quad (58)$$

which, together with Proposition 13, implies that

$$\begin{aligned} \tilde{D}_{\alpha,t}^L(g, h) &= t^\alpha \int_0^\infty \int_0^s D_{t+r,1}^L(g, h) \frac{dr ds}{t+r s^{1+\alpha}} \\ &\leq t^\alpha \int_0^\infty \int_0^s \frac{t+r}{\left((t+r)^2 + |g^{-1}h|^2 \right)^{(\mathcal{Q}+1)/2}} \\ &\quad \cdot \left(1 + \frac{t+r}{\rho(g)} + \frac{t+r}{\rho(h)} \right)^{-M} \frac{dr ds}{t+r s^{1+\alpha}}. \end{aligned} \quad (59)$$

One the one hand, we use the change of order of integration to get

$$\begin{aligned} \left| \tilde{D}_{\alpha,t}^L(g, h) \right| &\leq t^\alpha \int_0^\infty \int_0^s (t+r)^{-\mathcal{Q}} \left(\frac{t+r}{\rho(g)} \right)^{-M} \left(\frac{t+r}{\rho(h)} \right)^{-M} \frac{dr ds}{t+r s^{1+\alpha}} \\ &\leq t^\alpha \rho(g)^M \rho(h)^M \int_0^\infty \left(\int_r^\infty \frac{ds}{s^{1+\alpha}} \right) (t+r)^{-\mathcal{Q}-2M-1} dr \\ &\leq t^\alpha \rho(g)^M \rho(h)^M \int_0^\infty r^{-\alpha} (t+r)^{-\mathcal{Q}-2M-1} dr \\ &\leq t^{-\mathcal{Q}} \left(\frac{t}{\rho(g)} \right)^{-M} \left(\frac{t}{\rho(h)} \right)^{-M}. \end{aligned} \quad (60)$$

One the other hand, for $\alpha \in (0, 1)$,

$$\begin{aligned} \left| \tilde{D}_{\alpha,t}^L(g, h) \right| &\leq t^\alpha \int_0^\infty \int_0^s \frac{(t+r)^{-\mathcal{Q}-1}}{\left(1 + \left(|g^{-1}h|^2 / (t+r)^2 \right)^{(\mathcal{Q}+\alpha)/2} \right)} \\ &\quad \cdot \left(\frac{t+r}{\rho(g)} \right)^{-M} \left(\frac{t+r}{\rho(h)} \right)^{-M} \frac{dr ds}{s^{1+\alpha}} \\ &\leq \frac{t^\alpha \rho(g)^M \rho(h)^M}{|g^{-1}h|^{\mathcal{Q}+\alpha}} \int_0^\infty \int_0^s (t+r)^{\alpha-2M-1} dr \frac{ds}{s^{1+\alpha}} \\ &\leq \frac{t^\alpha \rho(g)^M \rho(h)^M}{|g^{-1}h|^{\mathcal{Q}+\alpha}} \int_0^\infty r^{-\alpha} (t+r)^{\alpha-2M-1} dr \\ &\leq t^\alpha |g^{-1}h|^{-\mathcal{Q}-\alpha} \left(\frac{t}{\rho(g)} \right)^{-M} \left(\frac{t}{\rho(h)} \right)^{-M}. \end{aligned} \quad (61)$$

Case 2. $\alpha \geq 1$. Since, for $\alpha \in (0, 1)$, $t^\alpha L^{\alpha/2} e^{-t\sqrt{L}} = t^\alpha \int_0^\infty \int_0^s \partial_r e^{-(t+r)\sqrt{L}} (dr ds / s^{1+\alpha})$. We can get

$$t^\alpha L^{\alpha/2} e^{-t\sqrt{L}} = t^\alpha L^{[\alpha]/2} L^{(\alpha-[\alpha])/2} e^{-t\sqrt{L}}. \quad (62)$$

Setting $\beta = \alpha - [\alpha]$, we obtain

$$t^\alpha L^{\alpha/2} e^{-t\sqrt{L}} = t^\alpha L^{[\alpha]/2} L^{\beta/2} e^{-t\sqrt{L}} = t^\alpha L^{[\alpha]/2} \int_0^\infty \int_0^s \partial_r e^{-(t+r)\sqrt{L}} \frac{dr ds}{s^{1+\beta}}. \quad (63)$$

Since $m = [\alpha] + 1$,

$$\begin{aligned} t^\alpha L^{\alpha/2} e^{-t\sqrt{L}} &= t^\alpha L^{[\alpha]/2} \int_0^\infty \int_0^s (-L)^{1/2} e^{-(t+r)\sqrt{L}} \frac{dr ds}{s^{1+\alpha-[\alpha]}} \\ &= t^\alpha \int_0^\infty \int_0^s (-L)^{m/2} e^{-(t+r)\sqrt{L}} \frac{dr ds}{s^{2+\alpha-m}} \\ &= t^\alpha \int_0^\infty \int_0^s D_{t+r,m}^L(g, h) \frac{dr ds}{(t+r)^m s^{2+\alpha-m}}. \end{aligned} \quad (64)$$

It follows from Proposition 13 that

$$\begin{aligned} \left| \tilde{D}_{\alpha,t}^L(g, h) \right| &\leq t^\alpha \int_0^\infty \int_0^s (t+r)^{-\mathcal{Q}} \left(\frac{t+r}{\rho(g)} \right)^{-M} \left(\frac{t+r}{\rho(h)} \right)^{-M} \frac{dr ds}{(t+r)^m s^{2+\alpha-m}} \\ &\leq t^\alpha \rho(g)^M \rho(h)^M \int_0^\infty \int_0^s (t+r)^{-\mathcal{Q}-2M-m} dr \frac{ds}{s^{2+\alpha-m}} \\ &\leq t^\alpha \rho(g)^M \rho(h)^M \int_0^\infty \left(\int_r^\infty \frac{ds}{s^{2+\alpha-m}} \right) (t+r)^{-\mathcal{Q}-2M-m} dr \\ &\leq t^{-\mathcal{Q}} \left(1 + \frac{t}{\rho(g)} \right)^{-M} \left(1 + \frac{t}{\rho(h)} \right)^{-M}. \end{aligned} \quad (65)$$

Also, noticing that $\alpha < m$, we obtain

$$\begin{aligned}
 |\tilde{D}_{\alpha,t}^L(g, h)| &\leq t^\alpha \int_0^\infty \int_0^s \frac{(t+r)^{-\mathcal{Q}-m}}{\left(1 + (|g^{-1}h|^2/(t+r)^2)\right)^{(\mathcal{Q}+\alpha)/2}} \\
 &\quad \cdot \left(\frac{t+r}{\rho(g)}\right)^{-M} \left(\frac{t+r}{\rho(h)}\right)^{-M} dr \frac{ds}{s^{2+\alpha-m}} \\
 &\leq \frac{t^\alpha \rho(g)^M \rho(h)^M}{|g^{-1}h|^{\mathcal{Q}+\alpha}} \int_0^\infty \int_0^s (t+r)^{\alpha-2M-m} dr \frac{ds}{s^{2+\alpha-m}} \\
 &\leq \frac{t^\alpha \rho(g)^M \rho(h)^M}{|g^{-1}h|^{\mathcal{Q}+\alpha}} \int_0^\infty r^{m-\alpha-1} (t+r)^{\alpha-2M-m} dr \\
 &\leq \frac{t^\alpha}{|g^{-1}h|^{\mathcal{Q}+\alpha}} \left(1 + \frac{t}{\rho(g)}\right)^{-M} \left(1 + \frac{t}{\rho(h)}\right)^{-M}.
 \end{aligned} \tag{66}$$

(ii) We first consider the case $\alpha \in (0, 1)$. Since

$$\tilde{D}_{\alpha,t}^L(g, h) = t^\alpha \int_0^\infty \int_0^s D_{t+r,1}^L(g, h) \frac{dr}{t+r} \frac{ds}{s^{1+\alpha}}, \tag{67}$$

we apply (ii) of Proposition 13 to obtain

$$\begin{aligned}
 &|\tilde{D}_{\alpha,t}^L(g\omega, h) - \tilde{D}_{\alpha,t}^L(g, h)| \\
 &\leq t^\alpha \int_0^\infty \int_0^s \left(\frac{|\omega|}{t+r}\right)^{\delta'} \frac{t+r}{\left((t+r)^2 + |g^{-1}h|^2\right)^{(\mathcal{Q}+1)/2}} \\
 &\quad \times \left(1 + \frac{t+r}{\rho(g)}\right)^{-M} \left(1 + \frac{t+r}{\rho(h)}\right)^{-M} \frac{dr}{t+r} \frac{ds}{s^{1+\alpha}}.
 \end{aligned} \tag{68}$$

One the one hand, we have

$$\begin{aligned}
 &|\tilde{D}_{\alpha,t}^L(g\omega, h) - \tilde{D}_{\alpha,t}^L(g, h)| \\
 &\leq t^\alpha \int_0^\infty \int_0^s \left(\frac{|\omega|}{t+r}\right)^{\delta'} \left(\frac{t+r}{\rho(g)}\right)^{-M} \left(\frac{t+r}{\rho(h)}\right)^{-M} \frac{dr}{(t+r)^{\mathcal{Q}+1}} \frac{ds}{s^{1+\alpha}} \\
 &\leq t^\alpha |\omega|^{\delta'} \rho(g)^M \rho(h)^M \int_0^\infty \left(\int_r^\infty \frac{ds}{s^{1+\alpha}}\right) (t+r)^{-\delta'-\mathcal{Q}-2M-1} dr \\
 &\leq t^{-\mathcal{Q}} \left(\frac{|\omega|}{t}\right)^{\delta'} \left(1 + \frac{t}{\rho(g)}\right)^{-M} \left(1 + \frac{t}{\rho(h)}\right)^{-M}.
 \end{aligned} \tag{69}$$

On the other hand, since $\alpha < 1$, it holds

$$\begin{aligned}
 &|\tilde{D}_{\alpha,t}^L(g\omega, h) - \tilde{D}_{\alpha,t}^L(g, h)| \\
 &\leq t^\alpha \int_0^\infty \int_0^s \left(\frac{|\omega|}{t+r}\right)^{\delta'} \frac{(t+r)^{-\mathcal{Q}}}{\left(1 + (|g^{-1}h|^2/(t+r)^2)\right)^{(\mathcal{Q}+\alpha)/2}} \\
 &\quad \cdot \left(\frac{t+r}{\rho(g)}\right)^{-M} \left(\frac{t+r}{\rho(h)}\right)^{-M} \frac{dr}{t+r} \frac{ds}{s^{1+\alpha}} \\
 &\leq \frac{t^\alpha |\omega|^{\delta'} \rho(g)^M \rho(h)^M}{|g^{-1}h|^{\mathcal{Q}+\alpha}} \int_0^\infty \int_0^s (t+r)^{-\delta'+\alpha-2M-1} dr \frac{ds}{s^{1+\alpha}} \\
 &\leq \frac{t^\alpha |\omega|^{\delta'} \rho(g)^M \rho(h)^M}{|g^{-1}h|^{\mathcal{Q}+\alpha}} \int_0^\infty r^{-\alpha} (t+r)^{-\delta'+\alpha-2M-1} dr \\
 &\leq \frac{t^\alpha}{|g^{-1}h|^{\mathcal{Q}+\alpha}} \left(\frac{|\omega|}{t}\right)^{\delta'} \left(1 + \frac{t}{\rho(g)}\right)^{-M} \left(1 + \frac{t}{\rho(h)}\right)^{-M}.
 \end{aligned} \tag{70}$$

For $\alpha \geq 1$, noticing

$$\begin{aligned}
 \tilde{D}_{\alpha,t}^L(g\omega, h) - \tilde{D}_{\alpha,t}^L(g, h) &= t^\alpha \int_0^\infty \int_0^s \\
 &\quad \cdot [D_{t+r,m}^L(g\omega, h) - D_{t+r,m}^L(g, h)] \frac{dr}{(t+r)^m} \frac{ds}{s^{2+\alpha-m}},
 \end{aligned} \tag{71}$$

we can use (ii) of Proposition 13 to get

$$\begin{aligned}
 &|\tilde{D}_{\alpha,t}^L(g\omega, h) - \tilde{D}_{\alpha,t}^L(g, h)| \\
 &\leq t^\alpha \int_0^\infty \int_0^s \left(\frac{|\omega|}{t+r}\right)^{\delta'} \frac{(t+r)^m}{\left((t+r)^2 + |g^{-1}h|^2\right)^{(\mathcal{Q}+m)/2}} \\
 &\quad \times \left(1 + \frac{t+r}{\rho(g)}\right)^{-M} \left(1 + \frac{t+r}{\rho(h)}\right)^{-M} \frac{dr}{(t+r)^m} \frac{ds}{s^{2+\alpha-m}}.
 \end{aligned} \tag{72}$$

The rest of the proof can be completed by the procedure of the case $\alpha > 1$ in (i), so we omit the details.

(iii) For $\alpha \in (0, 1)$, it follows from (iii) of Proposition 13 that

$$\begin{aligned}
 \left| \int_{\mathbb{H}^n} \tilde{D}_{\alpha,t}^L(g, h) dh \right| &\leq t^\alpha \int_0^\infty \int_0^s \left| \int_{\mathbb{H}^n} D_{t+r,1}^L(g, h) dh \right| \frac{dr}{t+r} \frac{ds}{s^{1+\alpha}} \\
 &\leq t^\alpha \int_0^\infty \int_0^s \left(\frac{t+r}{\rho(g)}\right)^{\delta'} \frac{1}{\left(1 + (t+r/\rho(g))\right)^M} \frac{dr}{t+r} \frac{ds}{s^{1+\alpha}}.
 \end{aligned} \tag{73}$$

If $t/\rho(g) \geq 1$, then

$$\begin{aligned}
\int_{\mathbb{H}^n} \tilde{D}_{\alpha,t}^L(g,h)dh &\leq t^\alpha \int_0^\infty \int_0^s \left(\frac{t+r}{\rho(g)}\right)^{\delta'-M} \frac{dr}{t+r} \frac{ds}{s^{1+\alpha}} \\
&\leq t^\alpha \rho(g)^{M-\delta'} \int_0^\infty \left(\int_r^\infty \frac{ds}{s^{1+\alpha}}\right) (t+r)^{\delta'-M-1} dr \\
&\leq \frac{(t/\rho(g))^{\delta'}}{(t/\rho(g))^M} \leq C \frac{(t/\rho(g))^{\delta'}}{(1+t/\rho(g))^M}.
\end{aligned} \tag{74}$$

If $t/\rho(g) < 1$, then

$$\begin{aligned}
\int_{\mathbb{H}^n} \tilde{D}_{\alpha,t}^L(g,h)dh &\leq t^\alpha \int_0^\infty \left(\frac{t+r}{\rho(g)}\right)^{\delta'} \frac{1}{(1+(t+r/\rho(g)))^M} \frac{dr}{r^\alpha(t+r)} \\
&\leq t^\alpha \rho(g)^{-\delta'} \int_0^\infty (t+r)^{\delta'-1} r^{-\alpha} dr \leq \left(\frac{t}{\rho(g)}\right)^{\delta'} \\
&\leq C \frac{(t/\rho(g))^{\delta'}}{(1+t/\rho(g))^M}.
\end{aligned} \tag{75}$$

For $\alpha \geq 1$, using (iii) of Proposition 13 again, we have

$$\begin{aligned}
\left| \int_{\mathbb{H}^n} \tilde{D}_{\alpha,t}^L(g,h)dh \right| &\leq t^\alpha \int_0^\infty \int_0^s \left| \int_{\mathbb{H}^n} D_{t+r,m}^L(g,h)dh \right| \frac{dr}{(t+r)^m} \frac{ds}{s^{2+\alpha-m}} \\
&\leq t^\alpha \int_0^\infty \int_0^s \left(\frac{t+r}{\rho(g)}\right)^{\delta'} \\
&\quad \cdot \frac{1}{(1+(t+r/\rho(g)))^M} \frac{dr}{(t+r)^m} \frac{ds}{s^{2+\alpha-m}}.
\end{aligned} \tag{76}$$

If $t/\rho(g) \geq 1$, we obtain

$$\begin{aligned}
\int_{\mathbb{H}^n} \tilde{D}_{\alpha,t}^L(g,h)dh &\leq t^\alpha \int_0^\infty \int_0^s \left(\frac{t+r}{\rho(g)}\right)^{\delta'-M} \frac{dr}{(t+r)^m} \frac{ds}{s^{2+\alpha-m}} \\
&\leq t^\alpha \rho(g)^{M-\delta'} \int_0^\infty \left(\int_r^\infty \frac{ds}{s^{2+\alpha-m}}\right) (t+r)^{\delta'-M-m} dr \\
&\leq \frac{(t/\rho(g))^{\delta'}}{(t/\rho(g))^M} \leq C \frac{(t/\rho(g))^{\delta'}}{(1+t/\rho(g))^M}.
\end{aligned} \tag{77}$$

If $t/\rho(g) < 1$, similarly, we can get

$$\begin{aligned}
\int_{\mathbb{H}^n} \tilde{D}_{\alpha,t}^L(g,h)dh &\leq t^\alpha \int_0^\infty \left(\frac{t+r}{\rho(g)}\right)^{\delta'} \frac{1}{(1+(t+r/\rho(g)))^M} \frac{dr}{r^{\alpha+1-m}(t+r)^m} \\
&\leq t^\alpha \rho(g)^{-\delta'} \int_0^\infty (t+r)^{\delta'-m} r^{m-\alpha-1} dr \leq \left(\frac{t}{\rho(g)}\right)^{\delta'} \\
&\leq C \frac{(t/\rho(g))^{\delta'}}{(1+t/\rho(g))^M}.
\end{aligned} \tag{78}$$

The following result can be obtained similar to Lemma 11.

Lemma 15. Let $\alpha > 0$. The operators $t^\alpha \partial_t^\alpha e^{-t\sqrt{L}}$ and $t^\alpha L^{\alpha/2} e^{-t\sqrt{L}}$ are equivalent.

Proof. For $\alpha \in (0, 1)$, we have

$$\begin{aligned}
t^\alpha L^{\alpha/2} e^{-t\sqrt{L}} &= t^\alpha \int_0^\infty \int_0^s \partial_r e^{-(t+r)\sqrt{L}} \frac{dr ds}{s^{1+\alpha}} \\
&= t^\alpha \int_0^\infty \left(\int_r^\infty \frac{ds}{s^{1+\alpha}}\right) \partial_r e^{-(t+r)\sqrt{L}} dr \\
&= t^\alpha \int_0^\infty r^{-\alpha} \partial_r e^{-(t+r)\sqrt{L}} dr \\
&= t^\alpha \partial_t^\alpha e^{-t\sqrt{L}}.
\end{aligned} \tag{79}$$

For $\alpha > 1$, let $\alpha - [\alpha] = \beta$. Noticing $m = [\alpha] + 1$, we obtain

$$\begin{aligned}
t^\alpha L^{\alpha/2} e^{-t\sqrt{L}} &= t^\alpha L^{[\alpha]/2} L^{\beta/2} e^{-t\sqrt{L}} \\
&= t^\alpha L^{[\alpha]/2} \int_0^\infty (-\sqrt{L}) e^{-(s+t)\sqrt{L}} s^{1-\beta} \frac{ds}{s} \\
&= t^\alpha \int_0^\infty (-\sqrt{L})^{[\alpha]+1} e^{-(t+s)\sqrt{L}} s^{1-\alpha+[\alpha]} \frac{ds}{s} \\
&= t^\alpha \int_0^\infty (-\sqrt{L})^m e^{-(t+s)\sqrt{L}} s^{m-\alpha} \frac{ds}{s} \\
&= t^\alpha \partial_t^\alpha e^{-t\sqrt{L}}.
\end{aligned} \tag{80}$$

Define an operator $D_{\alpha,t}^L(f) = t^\alpha \partial_t^\alpha P_t^L$. Denote by $D_{\alpha,t}^L(\cdot, \cdot)$ the integral kernel of $D_{\alpha,t}^L$. The following estimates are immediate corollaries of Proposition 14 and Lemma 15.

Corollary 16. Let $\alpha > 0$.

(i) For every M , there is a constant C_M such that

$$|D_{\alpha,t}^L(g,h)| \leq \frac{C_M t^\alpha}{(t^2 + |g^{-1}h|^2)^{(\alpha+\alpha)/2}} \left(1 + \frac{t}{\rho(g)} + \frac{t}{\rho(h)}\right)^{-M}. \tag{81}$$

(ii) Assume that $0 < \delta' \leq \delta$ with $0 < \delta' < \alpha$. For any $M > 0$ there exists a constant $C_M > 0$ such that for all $|\omega| \leq t$

$$\begin{aligned}
|D_{\alpha,t}^L(g\omega,h) - D_{\alpha,t}^L P_t^L(g,h)| &\leq \frac{C_M t^\alpha}{(t^2 + |g^{-1}h|^2)^{(\alpha+\alpha)/2}} \left(\frac{|\omega|}{t}\right)^{\delta'} \\
&\quad \cdot \left(1 + \frac{t}{\rho(g)} + \frac{t}{\rho(h)}\right)^{-M}.
\end{aligned} \tag{82}$$

(iii) For any $M > 0$, there exists a constant $C_M > 0$ such that

$$\left| \int_0^\infty D_{\alpha,t}^L(g, h) dh \right| \leq \frac{C_M(t/\rho(g))^{\delta'}}{(1+t/\rho(g))^M}. \tag{83}$$

3. Square Function Characterizations of Hardy-Sobolev Type Spaces

3.1. Fractional Square Functions Characterizations of $H_L^1(\mathbb{H}^n)$. Define

$$\begin{cases} \mathfrak{g}_{P,\alpha}(f)(g) := \left(\int_0^\infty |D_{\alpha,t}^L f(g)|^2 \frac{dt}{t} \right)^{1/2}; \\ \mathfrak{S}_{P,\alpha}(f)(g) := \left(\int_0^\infty \int_{B(g,t)} |D_{\alpha,t}^L f(g)|^2 \frac{dh dt}{t^{\mathcal{Q}+1}} \right)^{1/2}; \\ \mathfrak{g}_{P,\alpha,\lambda}^*(f)(g) := \left(\int_0^\infty \int_{\mathbb{H}^n} \left(\frac{t}{t+|g^{-1}h|} \right)^{2\lambda} |D_{\alpha,t}^L f(g)|^2 \frac{dh dt}{t^{\mathcal{Q}+1}} \right)^{1/2}. \end{cases} \tag{84}$$

In this section, we will characterize the Hardy space $H_L^1(\mathbb{H}^n)$ by the fractional square functions defined by (9) and (84). Now, we first prove the following reproducing formulas.

Lemma 17. *Let $\alpha > 0$.*

(i) *The operator $Q_{\alpha,t}^L$ defines an isometry from $L^2(\mathbb{H}^n)$ into $L^2(\mathbb{U}^n, dg dt/t)$. Moreover, in the sense of $L^2(\mathbb{H}^n)$,*

$$f = C_\alpha \lim_{\varepsilon \rightarrow 0} \lim_{N \rightarrow \infty} \int_\varepsilon^N (Q_{\alpha,t}^L)^2 f \frac{dt}{t}. \tag{85}$$

(ii) *The operator $D_{\alpha,t}^L$ defines an isometry from $L^2(\mathbb{H}^n)$ into $L^2(\mathbb{U}^n, dg dt/t)$. Moreover, in the sense of $L^2(\mathbb{H}^n)$,*

$$f = C_\alpha \lim_{\varepsilon \rightarrow 0} \lim_{N \rightarrow \infty} \int_\varepsilon^N (D_{\alpha,t}^L)^2 f \frac{dt}{t}. \tag{86}$$

Proof. The proofs of (i) and (ii) are standard and can be deduced from the spectral techniques. For completeness, we give the proof of (i) and omit the details of the proof of (ii). Since $e^{-t^2 L} = \int_0^\infty e^{-t^2 \lambda} dE(\lambda)$, we have

$$t^2 \frac{d}{ds} e^{-sL} \Big|_{s=t^2} = -t^2 L e^{-t^2 L} = - \int_0^\infty t^2 \lambda e^{-t^2 \lambda} dE(\lambda). \tag{87}$$

Thus, for all $f \in L^2(\mathbb{H}^n)$, we get

$$\begin{aligned} \|\mathfrak{g}_{H,\alpha} f\|_2^2 &= \int_{\mathbb{H}^n} \int_0^\infty |Q_{\alpha,t}^L(f)(g)|^2 \frac{dt dg}{t} \\ &= \int_0^\infty \left\langle (Q_{\alpha,t}^L)^2 f, f \right\rangle \frac{dt}{t} \\ &= \int_0^\infty \left[\int_0^\infty t^{4\alpha} \lambda^{2\alpha} e^{-2t^2 \lambda} \frac{dt}{t} \right] dE_{f,f}(\lambda) \\ &= C_\alpha \|f\|_2^2. \end{aligned} \tag{88}$$

For the second part, it suffices to show that, for every pair of sequences $n_k \rightarrow \infty$ & $\varepsilon_k \rightarrow 0$,

$$\lim_{k \rightarrow \infty} \int_{n_k}^{n_k+m} (Q_{\alpha,t}^L)^2 \frac{dt}{t} = \lim_{k \rightarrow \infty} \int_{\varepsilon_k}^{\varepsilon_k} (Q_{\alpha,t}^L)^2 \frac{dt}{t} = 0 \forall m \geq 1. \tag{89}$$

Indeed, if (89) holds, we can find $h \in L^2(\mathbb{H}^n)$ such that $\lim_{k \rightarrow \infty} \int_{\varepsilon_k}^{n_k} (Q_{\alpha,t}^L)^2 (dt/t) = h$. Therefore, it follows from a polarized version of the first part that for $g \in L^2(\mathbb{H}^n)$,

$$\begin{aligned} \langle h, g \rangle &= \lim_{k \rightarrow \infty} \int_{\varepsilon_k}^{n_k} \left\langle Q_{\alpha,t}^L f, Q_{\alpha,t}^L g \right\rangle \frac{dt}{t} \\ &= \int_0^\infty \left\langle Q_{\alpha,t}^L f, Q_{\alpha,t}^L g \right\rangle \frac{dt}{t} \\ &= C_\alpha \langle f, g \rangle, \end{aligned} \tag{90}$$

which implies $h = C_\alpha f$. To prove (89), we use again the functional calculus to deduce that

$$\left\| \int_{n_k}^{n_k+m} (Q_{\alpha,t}^L)^2 \frac{dt}{t} \right\|_2^2 \leq \int_0^\infty \left| \int_{n_k}^{n_k+m} t^{4\alpha} \lambda^{2\alpha} e^{-2t^2 \lambda} \frac{dt}{t} \right|^2 dE_{f,f}(\lambda). \tag{91}$$

Computing the integral inside one yields $\int_0^\infty (1 + 2\lambda n_k^2) e^{-2\lambda n_k^2} dE_{f,f}(\lambda)$ as $n_k \rightarrow \infty$, which by dominated convergence tends to 0. Observe that the last step makes use of the fact that 0 is not an eigenvalue of L because $V(g) > 0$ for almost every g , and $\langle Lf, f \rangle \geq \langle Vf, f \rangle > 0$ unless $f \equiv 0$. One proceeds similarly when $\varepsilon_k \rightarrow 0$.

The following boundedness of square functions can be deduced from the spectral theorem immediately.

Lemma 18. *Let $\alpha > 0$ and $\lambda > \mathcal{Q}/2$.*

- (i) *The operators $\mathfrak{g}_{H,\alpha}$, $\mathfrak{G}_{H,\alpha}$ and $\mathfrak{g}_{H,\alpha,\lambda}^*$ are bounded on $L^2(\mathbb{H}^n)$. Moreover, there exist constants C, C_1 and C_2 such that $\|\mathfrak{g}_{H,\alpha} f\|_{L^2} = C \|f\|_{L^2}$, $\|\mathfrak{G}_{H,\alpha} f\|_{L^2} \leq C_1 \|f\|_{L^2}$, $\|\mathfrak{g}_{H,\alpha,\lambda}^* f\|_{L^2} \leq C_2 \|f\|_{L^2}$*
- (ii) *The operators $\mathfrak{g}_{P,\alpha}$, $\mathfrak{G}_{P,\alpha}$ and $\mathfrak{g}_{P,\alpha,\lambda}^*$ are bounded on $L^2(\mathbb{H}^n)$. Moreover, there exist constants C, C_1 and*

C_2 such that $\|\mathfrak{g}_{P,\alpha}f\|_{L^2} = C\|f\|_{L^2}$, $\|\mathfrak{G}_{P,\alpha}f\|_{L^2} \leq C_1\|f\|_{L^2}$, $\|\mathfrak{g}_{P,\alpha,\lambda}^*f\|_{L^2} \leq C_2\|f\|_{L^2}$

Proof. We only prove (i), and (ii) can be done similarly. For $\mathfrak{G}_{H,\alpha}$, using the reproducing formula on $L^2(\mathbb{H}^n)$, we can get

$$\begin{aligned} \|\mathfrak{G}_{H,\alpha}f\|_{L^2}^2 &= \int_0^\infty \langle Q_{\alpha,t}^L f, Q_{\alpha,t}^L f \rangle \frac{dt}{t} \\ &= \int_0^\infty \langle (Q_{\alpha,t}^L)^2 f, f \rangle \frac{dt}{t} \\ &= \int_0^\infty \left[e^{-i\pi\alpha} \int_0^\infty t^{4\alpha} \lambda^{2\alpha} e^{-2t^2\lambda} \frac{dt}{t} \right] dE_{f,f}(\lambda) \\ &= C\|f\|_{L^2}^2. \end{aligned} \quad (92)$$

For $\mathfrak{G}_{H,\alpha}$, we have

$$\begin{aligned} \|\mathfrak{G}_{H,\alpha}(f)\|_{L^2}^2 &\leq \int_0^\infty \int_{\mathbb{H}^n} \left[\frac{1}{t^{\mathcal{Q}}} \int_{\mathbb{H}^n} \chi_{\Gamma(g)}(h,t) dg \right] |Q_{\alpha,t}^L f(g)|^2 \frac{dhdt}{t} \\ &\leq \int_0^\infty \int_{\mathbb{H}^n} |Q_{\alpha,t}^L f(g)|^2 \frac{dhdt}{t} = \|\mathfrak{G}_{H,\alpha}f\|_{L^2}^2 \\ &\leq C_1\|f\|_{L^2}^2. \end{aligned} \quad (93)$$

For $\mathfrak{g}_{H,\alpha,\lambda}^*$, the relation: $\mathfrak{g}_{H,\alpha,\lambda}^*f(g) \leq C\mathfrak{G}_{H,\alpha}(f)(g)$ indicates that $\|\mathfrak{g}_{H,\alpha,\lambda}^*f\|_{L^2} \leq C_2\|f\|_{L^2}$.

Proposition 19. Let $\alpha > 0$ and $\lambda > \mathcal{Q}/2$.

(i) There exists a constant C such that for any function f which is a linear combination of H_L^1 -atoms

$$\|\mathfrak{G}_{H,\alpha}f\|_{L^1} \leq C\|f\|_{H_L^1}, \|\mathfrak{g}_{H,\alpha}f\|_{L^1} \leq C\|f\|_{H_L^1}, \|\mathfrak{g}_{H,\alpha,\lambda}^*\|_{L^1} \leq C\|f\|_{H_L^1}. \quad (94)$$

(ii) There exists a constant C such that for any function f which is a linear combination of H_L^1 -atoms

$$\|\mathfrak{G}_{P,\alpha}f\|_{L^1} \leq C\|f\|_{H_L^1}, \|\mathfrak{g}_{P,\alpha}f\|_{L^1} \leq C\|f\|_{H_L^1}, \|\mathfrak{g}_{P,\alpha,\lambda}^*\|_{L^1} \leq C\|f\|_{H_L^1}. \quad (95)$$

Proof. We only prove (i), and (ii) can be dealt with similarly. Firstly, by Lemma 18, we can get $\|\mathfrak{G}_{H,\alpha}(f)\|_{L^2} = C\|f\|_{L^2}$. For $f \in H_L^1(\mathbb{H}^n)$, it holds an atomic decomposition: $f = \sum_j c_j a_j$. Then,

$$\begin{aligned} \mathfrak{G}_{H,\alpha}(f)(g) &= \left(\int_0^\infty \int_{B(g,t)} \left| \sum_j c_j (Q_{\alpha,t}^L a_j)(h) \right|^2 \frac{dhdt}{t^{\mathcal{Q}+1}} \right)^{1/2} \\ &\leq \sum_j |c_j| \mathfrak{G}_{H,\alpha}(a_j)(g). \end{aligned} \quad (96)$$

So we only need to verify that $\mathfrak{G}_{H,\alpha}(a)$ is in $L^1(\mathbb{H}^n)$ for any H_L^1 -atom a uniformly. By Lemma 18,

$$\begin{aligned} \|\mathfrak{G}_{H,\alpha}(a)\|_{L^2}^2 &\leq \int_0^\infty \int_{\mathbb{H}^n} |Q_{\alpha,t}^L a(h)|^2 \frac{dhdt}{t} \\ &= \|\mathfrak{G}_{H,\alpha}(f)\|_{L^2}^2 \leq C\|a\|_{L^2}^2 \\ &\leq C|B(g_0, r)|^{-1}. \end{aligned} \quad (97)$$

Write $\|\mathfrak{G}_{H,\alpha}(a)\|_{L^1} = A + B$, where $A = \int_{B(g_0, 4r)} |\mathfrak{G}_{H,\alpha}a(g)| dg$ and $B = \int_{B^c(g_0, 4r)} |\mathfrak{G}_{H,\alpha}a(g)| dg$. For A , it is clear that

$$\begin{aligned} A &\leq |B(g_0, 4r)|^{1/2} \left(\int_{B(g_0, 4r)} |\mathfrak{G}_{H,\alpha}a(g)|^2 dg \right)^{1/2} \\ &\leq |B(g_0, 4r)|^{1/2} C|B(g_0, r)|^{-1} \leq C. \end{aligned} \quad (98)$$

For the estimate of B , the following two cases are considered.

Case 1. $r < \rho(g_0)/4$. By the cancelation property of the atom a , we have $\mathfrak{G}_{H,\alpha}a(g) \leq B_1 + B_2$, where

$$\begin{cases} B_1 := \left(\int_0^{|\mathcal{g}^{-1}g_0|/2} \int_{|\mathcal{g}^{-1}h|<t} \left(\int_{B(g_0,r)} |Q_{\alpha,t}^L(h,z) - Q_{\alpha,t}^L(h,g_0)||a(z)||dz \right)^2 \frac{dhdt}{t^{\mathcal{Q}+1}} \right)^{1/2}; \\ B_2 := \left(\int_{|\mathcal{g}^{-1}g_0|/2}^\infty \int_{|\mathcal{g}^{-1}h|<t} \left(\int_{B(g_0,r)} |Q_{\alpha,t}^L(h,z) - Q_{\alpha,t}^L(h,g_0)||a(z)||dz \right)^2 \frac{dhdt}{t^{\mathcal{Q}+1}} \right)^{1/2}. \end{cases} \quad (99)$$

For B_1 , since $0 < t < |\mathcal{g}^{-1}g_0|/2$ and $|\mathcal{g}^{-1}h| < t$, we can get $|h^{-1}g_0| \sim |\mathcal{g}^{-1}g_0|$. For $z \in B(g_0, r)$ and $g \in B^c(g_0, 4r)$, we have $|\mathcal{g}_0^{-1}z| < r \leq C|\mathcal{g}_0^{-1}h|/4$. Using (ii) of Corollary 12 and the symmetry, we can get

$$\begin{aligned} B_1 &\leq \left(\int_0^{|\mathcal{g}^{-1}g_0|/2} \int_{|\mathcal{g}^{-1}h|<t} \left(\int_{B(g_0,r)} C_M \frac{t^\alpha}{(t+|\mathcal{g}_0^{-1}h|)^{\mathcal{Q}+\alpha}} \right. \right. \\ &\quad \left. \left. \cdot \left(\frac{|z^{-1}g_0|}{t} \right)^{\delta'} |a(z)| dz \right)^2 \frac{dhdt}{t^{\mathcal{Q}+1}} \right)^{1/2} \\ &\leq C_M \left(\int_0^{|\mathcal{g}^{-1}g_0|/2} \int_{|\mathcal{g}^{-1}h|<t} \frac{t^{2\alpha}}{(t+|\mathcal{g}_0^{-1}g|)^{2(\mathcal{Q}+\alpha)}} \left(\frac{r}{t} \right)^{2\delta'} \frac{dhdt}{t^{\mathcal{Q}+1}} \right)^{1/2} \\ &\leq C_M \frac{r^{\delta'}}{|\mathcal{g}_0^{-1}g|^{\mathcal{Q}+\alpha}} \left(\int_0^{|\mathcal{g}^{-1}g_0|/2} \frac{1}{t^{2\delta'-2\alpha+1}} dt \right)^{1/2} \leq \frac{C_M r^{\delta'}}{|\mathcal{g}_0^{-1}g|^{\mathcal{Q}+\delta'}}. \end{aligned} \quad (100)$$

The above estimate for B_1 implies that

$$\begin{aligned} \int_{B^c(g_0, 4r)} B_1 dg &\leq C_M \sum_{k=2}^{\infty} \int_{2^k r \leq |g^{-1}g_0| < 2^{k+1}r} \frac{r^{\delta'} dg}{|g^{-1}g_0|^{\mathcal{Q}+\delta'}} \\ &\leq C_M \sum_{k=2}^{\infty} \frac{r^{\delta'} (2^{k+1}r)^{\mathcal{Q}}}{|2^k r|^{\mathcal{Q}+\delta'}} \leq C. \end{aligned} \quad (101)$$

Next, we estimate B_2 . Since $|z^{-1}g_0| \leq r < |g^{-1}g_0|/2 \leq t$, the estimate

$$\begin{aligned} B_2 &\leq C_M \left(\int_{|g^{-1}g_0|/2}^{\infty} \int_{|g^{-1}h| < t} \left(\int_{B(g_0, r)} \frac{t^\alpha}{(t+|g_0^{-1}h|)^{\mathcal{Q}+\alpha}} \right. \right. \\ &\quad \cdot \left. \left. \left(\frac{|z^{-1}g_0|}{t} \right)^{\delta'} |a(z)| dz \right)^2 \frac{dhdt}{t^{\mathcal{Q}+1}} \right)^{1/2} \\ &\leq C_M \left(\int_{|g^{-1}g_0|/2}^{\infty} \int_{|g^{-1}h| < t} \left(\frac{r}{t} \right)^{2\delta'} \frac{t^{2\alpha}}{(t+|g_0^{-1}h|)^{2(\mathcal{Q}+\alpha)}} \frac{dhdt}{t^{\mathcal{Q}+1}} \right)^{1/2} \\ &\leq \frac{C_M}{|g^{-1}g_0|^{\mathcal{Q}}} \frac{r^{\delta'}}{|g^{-1}g_0|^{\delta'}} \end{aligned} \quad (102)$$

implies that

$$\int_{B^c(g_0, 4r)} B_2 dg \leq C_M \int_{|g^{-1}g_0| \geq 4r} \frac{r^{\delta'} dg}{|g^{-1}g_0|^{\mathcal{Q}+\delta'}} \leq C. \quad (103)$$

Case 2. $\rho(g_0)/4 \leq r < \rho(g_0)$. In this case, we write $(\mathfrak{G}_{H, \alpha} a(g))^2 = D_1 + D_2 + D_3$, where

$$\begin{cases} D_1 := \int_0^{r/2} \int_{|g^{-1}h| < t} |Q_{\alpha, t}^L a(h)|^2 \frac{dhdt}{t^{\mathcal{Q}+1}}; \\ D_2 := \int_{r/2}^{|g^{-1}g_0|/4} \int_{|g^{-1}h| < t} |Q_{\alpha, t}^L a(h)|^2 \frac{dhdt}{t^{\mathcal{Q}+1}}; \\ D_3 := \int_{|g^{-1}g_0|/4}^{\infty} \int_{|g^{-1}h| < t} |Q_{\alpha, t}^L a(h)|^2 \frac{dhdt}{t^{\mathcal{Q}+1}}. \end{cases} \quad (104)$$

We first estimate the term D_1 . Since $|g^{-1}g_0| > 4r$, $|g_0^{-1}z| < r$ and $|g^{-1}h| < t < r/2$, $|h^{-1}g_0| > 7r/2$. For $z \in B(g_0, r)$, $|z^{-1}g_0| < r < |g^{-1}g_0|/4$. Using the triangle inequality, we apply (i) of Corollary 12 to estimate D_1 as follows.

$$\begin{aligned} D_1 &\leq C_M \int_0^{r/2} \int_{|g^{-1}h| < t} \left(\int_{B(g_0, r)} \frac{t^\alpha}{(t+|g^{-1}h|)^{\mathcal{Q}+\alpha}} |a(z)| dz \right)^2 \frac{dhdt}{t^{\mathcal{Q}+1}} \\ &\leq C_M \int_0^{r/2} \int_{|g^{-1}h| < t} \frac{t^{2\alpha}}{(t+|g^{-1}g_0|)^{2(\mathcal{Q}+\alpha)}} \frac{dhdt}{t^{\mathcal{Q}+1}} \\ &\leq \frac{C_M}{|g^{-1}g_0|^{2\mathcal{Q}+2\alpha}} \int_0^{r/2} t^{2\alpha-1} dt \leq \frac{C_M r^{2\alpha}}{|g^{-1}g_0|^{2\mathcal{Q}+2\alpha}}. \end{aligned} \quad (105)$$

For D_2 , since $z \in B(g_0, r)$, $|z^{-1}g_0| < r < \rho(g_0)$, then $\rho(z) \sim \rho(g_0) \sim r$. We have

$$\begin{aligned} D_2 &\leq C_M \int_{r/2}^{|g^{-1}g_0|/4} \int_{|g^{-1}h| < t} \left(\int_{B(g_0, r)} \frac{t^\alpha}{(t+|g^{-1}h|)^{\mathcal{Q}+\alpha}} \frac{|a(z)| dz}{(1+(t/\rho(h))+(t/\rho(z)))^M} \right)^2 \frac{dhdt}{t^{\mathcal{Q}+1}}, \\ &\leq C_M \int_{r/2}^{|g^{-1}g_0|/4} \int_{|g^{-1}h| < t} \frac{t^{2\alpha}}{(t+|g^{-1}h|)^{2(\mathcal{Q}+\alpha)}} \left(1 + \frac{t}{\rho(g_0)} \right)^{-2M} \frac{dhdt}{t^{\mathcal{Q}+1}} \\ &\leq C_M \int_{r/2}^{|g^{-1}g_0|/4} \left(\frac{r}{t} \right)^{2M} \frac{t^{2\alpha-1}}{|g^{-1}g_0|^{2\mathcal{Q}+2\alpha}} dt \leq \frac{C_M r^{2M}}{|g^{-1}g_0|^{2\mathcal{Q}+2M}}. \end{aligned} \quad (106)$$

At last, we estimate D_3 . For $|z^{-1}g_0| < r < \rho(g_0)$, we have $\rho(g_0) \sim \rho(z)$. Then, we can get

$$\begin{aligned} D_3 &\leq C_M \int_{|g^{-1}g_0|/4}^{\infty} \int_{|g^{-1}h| < t} \left(\int_{B(g_0, r)} \frac{t^\alpha}{(t+|h^{-1}z|)^{\mathcal{Q}+\alpha}} \frac{|a(z)| dz}{(1+(t/\rho(h))+(t/\rho(z)))^M} \right)^2 \frac{dhdt}{t^{\mathcal{Q}+1}} \\ &\leq C_M \int_{|g^{-1}g_0|/4}^{\infty} \int_{|g^{-1}h| < t} \left(\int_{B(g_0, r)} \left(1 + \frac{t}{\rho(g_0)} \right)^{-M} \frac{1}{(t+|h^{-1}z|)^{\mathcal{Q}}} |a(z)| dz \right)^2 \frac{dhdt}{t^{\mathcal{Q}+1}} \\ &\leq C_M \frac{r^{2M}}{|g^{-1}g_0|^{2\mathcal{Q}}} \int_{|g^{-1}g_0|/4}^{\infty} \frac{1}{t^{2M+1}} dt \leq \frac{C_M r^{2M}}{|g^{-1}g_0|^{2\mathcal{Q}+2M}}. \end{aligned} \quad (107)$$

The above estimates for D_i , $i = 1, 2, 3$, indicate that

$$\begin{aligned} \int_{B^c} \mathfrak{G}_{H, \alpha} a(g) dg &\leq \sum_{k=2}^{\infty} \int_{2^k r \leq |g^{-1}g_0| < 2^{k+1}r} \\ &\quad \cdot [D_1^{1/2}(g) + D_2^{1/2}(g) + D_3^{1/2}(g)] dg \leq C. \end{aligned} \quad (108)$$

Now, we give the following characterizations of $H_L^1(\mathbb{H}^n)$.

Theorem 20. Let $\alpha \geq 1/2$ and $\lambda > \mathcal{Q}/2$. The following assertions are equivalent:

- (i) $f \in H_L^1(\mathbb{H}^n)$;
- (ii) $f \in L^1(\mathbb{H}^n)$ and $\mathfrak{g}_{H, \alpha}(f) \in L^1(\mathbb{H}^n)$;
- (iii) $f \in L^1(\mathbb{H}^n)$ and $\mathfrak{G}_{H, \alpha}(f) \in L^1(\mathbb{H}^n)$;
- (iv) $f \in L^1(\mathbb{H}^n)$ and $\mathfrak{g}_{H, \alpha, \lambda}^*(f) \in L^1(\mathbb{H}^n)$

Moreover, for every $f \in H_L^1(\mathbb{H}^n)$,

$$\begin{aligned} \|f\|_{H_L^1} &\sim \|f\|_1 + \|\mathfrak{g}_{H, \alpha}(f)\|_1 \sim \|f\|_1 + \|\mathfrak{G}_{H, \alpha}(f)\|_1 \\ &\sim \|f\|_1 + \|\mathfrak{g}_{H, \alpha, \lambda}^*(f)\|_1. \end{aligned} \quad (109)$$

Proof. By Proposition 19, for $f \in H_L^1(\mathbb{H}^n)$, we know that $\mathfrak{g}_{H,\alpha}(f) \in L^1(\mathbb{H}^n)$, $\mathfrak{G}_{H,\alpha}(f) \in L^1(\mathbb{H}^n)$, and $\mathfrak{g}_{H,\alpha,\lambda}^*(f) \in L^1(\mathbb{H}^n)$, respectively.

For the reverse, we first show that for $\mathfrak{G}_{H,\alpha}(f) \in L^1(\mathbb{H}^n)$, $f \in H_L^1(\mathbb{H}^n)$. Assume that $f \in L^1(\mathbb{H}^n) \cap L^2(\mathbb{H}^n)$. When $\mathfrak{G}_{H,\alpha}(f) \in L^1(\mathbb{H}^n)$, we can see that

$$\int_{\mathbb{H}^n} |\mathfrak{G}_{H,\alpha} f(g)| dg = \int_{\mathbb{H}^n} \left(\int_0^\infty \int_{B(g,t)} |Q_{\alpha,t}^L f(h)|^2 \frac{dh dt}{t^{Q+1}} \right)^{1/2} dg, \quad (110)$$

which implies that $Q_{\alpha,t}^L f(g) \in T_2^1$, where $Q_{\alpha,t}^L f(g) := \int_{\mathbb{H}^n} Q_{\alpha,t}^L(g,h) f(h) dh$. By Proposition 8, $Q_{\alpha,t}^L f(g) = \sum_k \lambda_k a_k(g,t)$, where $a_k(\cdot, \cdot)$ are T_2^1 -atoms and $\sum_k |\lambda_k| < \infty$. Assume that the atom $a(\cdot, \cdot)$ is supported on $\tilde{B}(g_0, r)$. By Lemma 17,

$$f(g) = C \int_0^\infty Q_t^\alpha \left(\sum_{k=1}^\infty \lambda_k a_k(g,t) \right) \frac{dt}{t} := \sum_{k=1}^\infty \lambda_k T_k(g), \quad (111)$$

where $T_k(g) = \int_0^\infty Q_{\alpha,t}^L a_k(g,t) (dt/t)$. For simplicity, we denote $T_k(g)$ by $T(g)$ for $k=1, 2, \dots$. Write

$$\begin{aligned} \left\| \sup_{t>0} |e^{-tL} T(g)| \right\|_{L^1} &\leq \left\| \left(\sup_{t>0} |e^{-tL} T(g)| \right) \chi_{B^*} \right\|_{L^1} \\ &\quad + \left\| \left(\sup_{t>0} |e^{-tL} T(g)| \right) \chi_{(B^*)^c} \right\|_{L^1} \\ &= I_1 + I_2, \end{aligned} \quad (112)$$

where $B^* = B(g_0, 2r)$. For I_1 , we use Hölder's inequality to deduce that

$$\begin{aligned} \|T\|_{L^2} &= \sup_{\|\phi\|_2 \leq 1} \int_{\mathbb{H}^n} \left(\int_0^\infty Q_{\alpha,t}^L a(g,t) \frac{dt}{t} \right) \bar{\phi}(g) dg \\ &\leq \sup_{\|\phi\|_2 \leq 1} \left(\int_0^\infty \int_{\mathbb{H}^n} |a(g,t)|^2 \frac{dg dt}{t} \right)^{1/2} \\ &\quad \cdot \left(\int_0^\infty \int_{\mathbb{H}^n} |Q_{\alpha,t}^L \bar{\phi}(g)|^2 \frac{dg dt}{t} \right)^{1/2} \\ &\leq \sup_{\|\phi\|_2 \leq 1} |B|^{-1/2} \|\phi\|_2 \leq |B|^{-1/2}, \end{aligned} \quad (113)$$

which gives $I_1 \leq |B^*|^{1/2} |B|^{-1/2} \leq C$.

Now, we deal with I_2 . For $s > 0$, by functional calculus and Proposition 2.9, we have

$$\begin{aligned} \left| e^{-sL} \left(\int_0^\infty Q_{\alpha,t}^L a(g,t) \frac{dt}{t} \right) \right| &= \left| \int_0^\infty \int_0^\infty t^{2\alpha} \partial_s K_{s+t}^L |_{s=t^2} a(g,t) \lambda^{1-\alpha} \frac{d\lambda}{\lambda} \frac{dt}{t} \right| \\ &= \left| \int_0^\infty t^{2\alpha} |\partial_s^\alpha K_{s+t}^L |_{s=t^2} a(g,t) \frac{dt}{t} \right| \\ &\leq C \int_0^\infty \frac{t^\alpha |a(h,t)|}{((s+t^2)+|g^{-1}h|)^{Q+\alpha}} \frac{dh dt}{t}. \end{aligned} \quad (114)$$

When $h \in B(g_0, r)$ and $g \in (B^*)^c$, we have $|g^{-1}h| \sim |g^{-1}g_0|$, and

$$\begin{aligned} \left| e^{-sL} \left(\int_0^\infty Q_{\alpha,t}^L a(g,t) \frac{dt}{t} \right) \right| &\leq C |g^{-1}g_0|^{-(Q+\alpha/2)} \\ &\quad \cdot \left(\int_0^r \int_B t^{2\alpha-1} dh dt \right)^{1/2} \\ &\quad \cdot \left(\int_0^r \int_B |a(h,t)|^2 \frac{dh dt}{t} \right)^{1/2} \\ &\leq C |B|^{-1/2} |g^{-1}g_0|^{-(Q+\alpha)} \\ &\quad \cdot \left(\int_0^r \int_B t^{2\alpha-1} dh dt \right)^{1/2} \\ &\leq Cr^\alpha |g^{-1}g_0|^{-(Q+\alpha)}. \end{aligned} \quad (115)$$

Finally, we get

$$I_2 \leq \int_{B^*(g_0,r)} \frac{r^\alpha}{|g^{-1}g_0|^{Q+\alpha}} dg \leq C. \quad (116)$$

When $f \in H_L^1(\mathbb{H}^n)$, let $\tilde{\mathfrak{G}}_{H,\alpha}$ be the bounded extension of $\mathfrak{G}_{H,\alpha}(f)$ from $L^2 \cap H_L^1(\mathbb{H}^n)$ to $H_L^1(\mathbb{H}^n)$. Since $L^2 \cap H_L^1(\mathbb{H}^n)$ is dense in $H_L^1(\mathbb{H}^n)$, there exists a sequence $\{f_n\} \subset L^2 \cap H_L^1(\mathbb{H}^n)$ such that $f_n \rightarrow f$ as $n \rightarrow \infty$ in $H_L^1(\mathbb{H}^n)$. By Corollary 12, we conclude that $\mathfrak{G}_{H,\alpha}(f_n) \rightarrow \mathfrak{G}_{H,\alpha}(f)$ as $n \rightarrow \infty$. By the definition of $\tilde{\mathfrak{G}}_{H,\alpha}$, we know that $\mathfrak{G}_{H,\alpha}(f_n) \rightarrow \tilde{\mathfrak{G}}_{H,\alpha}(f)$ as $n \rightarrow \infty$. Therefore, $\mathfrak{G}_{H,\alpha}(f) = \tilde{\mathfrak{G}}_{H,\alpha}(f)$ for $f \in H_L^1(\mathbb{H}^n)$, which gives

$$\begin{aligned} \|f\|_{H_L^1} &= \left\| \lim_{n \rightarrow \infty} f_n \right\|_{H_L^1} \leq \lim_{n \rightarrow \infty} \|\mathfrak{G}_{H,\alpha}(f_n)\|_{L^1} \\ &= \left\| \tilde{\mathfrak{G}}_{H,\alpha}(f) \right\|_{L^1} = \|\mathfrak{G}_{H,\alpha}(f)\|_{L^1}. \end{aligned} \quad (117)$$

For the Littlewood-Paley \mathfrak{g} -function, it is sufficient to prove $\|\mathfrak{G}_{H,\alpha}(f)\|_{L^1} \leq C \|\mathfrak{g}_{H,\alpha}(f)\|_{L^1}$. For $\beta > 0$, we define $\tilde{\mathfrak{G}}_{H,\beta}(f)$ by

$$\tilde{\mathfrak{G}}_{H,\beta}(f)(g) = \left(\int_0^\infty \int_{|g^{-1}h| < \beta t} Q_{\alpha,t}^L f(h) \frac{dh dt}{t^{Q+1}} \right)^{1/2}. \quad (118)$$

Similarly, we can prove that $f \in H_L^1(\mathbb{H}^n)$ if and only if $\tilde{\mathfrak{G}}_{H,\beta}(f) \in L^1(\mathbb{H}^n)$ and $f \in L^1(\mathbb{H}^n)$. Moreover, $\|f\|_{H_L^1} \sim \|\tilde{\mathfrak{G}}_{H,\beta}(f)\|_{L^1}$.

Let $F(g)(t) := (\partial_s^\alpha e^{-sL}|_{s=t^2} f)(g)$ and $V(g, s) := e^{-sL} F(g)$. Then $V(g, s)(t) = (\partial_r^\alpha e^{-(s+r)L}|_{r=t^2} f)(g)$. Therefore,

$$\begin{aligned} \int_0^\infty |V(g, s)(t)|^2 \frac{dt}{t^{1-4\alpha}} &= \int_0^\infty \left| (\partial_r^\alpha e^{-(s+r)L}|_{r=t^2} f)(g) \right|^2 \frac{dt}{t^{1-4\alpha}} \\ &= \int_{\sqrt{s}}^\infty \left| (\partial_r^\alpha e^{-rL}|_{r=t^2} f)(g) \right|^2 \frac{tdt}{(t^2 - s)^{1-2\alpha}}. \end{aligned} \tag{119}$$

When $\alpha \geq 1/2$, we have $(t^2 - s)^{2\alpha-1} \leq t^{4\alpha-1}$. Hence,

$$\begin{aligned} \sup_{s>0} \int_0^\infty |V(g, s)(t)|^2 t^{4\alpha-1} dt &\leq \int_0^\infty \left| (\partial_r^\alpha e^{-rL}|_{r=t^2} f)(g) \right|^2 t^{4\alpha-1} dt \\ &= (\mathfrak{G}_{H,\alpha} f(g))^2. \end{aligned} \tag{120}$$

Let $\mathbf{X} = L^2((0, \infty), t^{4\alpha-1} dt)$. Then, $\sup_{s>0} \|e^{-sL} F(g)\|_{\mathbf{X}} \leq \mathfrak{G}_{H,\alpha} f(g) \in L^1(\mathbb{H}^n)$. Therefore, $F \in H_{\mathbf{X}}^1(\mathbb{H}^n)$, where $H_{\mathbf{X}}^1(\mathbb{H}^n)$ can be seen as a vector-valued Hardy space (cf. [30]). This shows that $\tilde{\mathfrak{G}}_2^{\mathbf{X}} F(g) \in L^1(\mathbb{H}^n)$, where

$$\tilde{\mathfrak{G}}_2^{\mathbf{X}} F(g) = \left(\int_0^\infty \int_{|g^{-1}h|<2t} \|Q_{\alpha,t}^L F(h)\|_{\mathbf{X}}^2 \frac{dhdt}{t^{\mathcal{Q}+1}} \right)^{1/2}. \tag{121}$$

We can assume that $1/2 \leq \alpha < 1$. Then, the identity (6) gives

$$\begin{aligned} (\partial_t^\alpha K_t^L|_{t=s^2}) (\partial_s^\alpha K_s^L|_{s=t^2}) &= C \int_0^\infty \int_0^\infty (\partial_a K_{a+t}^L|_{t=s^2}) \\ &\quad \cdot (\partial_b K_{s+b}^L|_{s=t^2}) a^{-\alpha} b^{-\alpha} da db \\ &= C \int_0^\infty \int_0^\infty (\partial_a^2 K_{a+b+s+t}^L|_{s=t^2, t=s^2}) a^{-\alpha} b^{-\alpha} da db \\ &= C \int_0^\infty (\partial_\lambda^2 K_{\lambda+s+t}^L|_{s=t^2, t=s^2}) \lambda^{1-2\alpha} d\lambda. \end{aligned} \tag{122}$$

When $\alpha \geq 1/2$, we get $\partial_t^\alpha K_t^L|_{t=s^2} \partial_s^\alpha K_s^L|_{s=t^2} = \partial_t^{2\alpha} K_{s+t}^L|_{s=t^2, t=s^2}$. Via integration by substitution, we can change the orders of integration to obtain

$$\begin{aligned} [\tilde{\mathfrak{G}}_2^{\mathbf{X}} F(g)]^2 &= \int_0^\infty \int_{|g^{-1}h|<2t} \int_0^\infty |t^{2\alpha} \partial_s^{2\alpha} e^{-sL}|_{s=t^2} F(h)(s)|^2 \frac{s^{4\alpha-1} ds dh dt}{t^{\mathcal{Q}+1}} \\ &\geq \int_0^\infty \int_0^{\sqrt{3}t/2} \int_{|g^{-1}h|<2\sqrt{t^2-s^2}} |\partial_s^{2\alpha} e^{-sL}|_{s=t^2} f(h)|^2 \frac{ts^{4\alpha-1} dh dt ds}{(t^2 - s^2)^{1+\mathcal{Q}/2-2\alpha}} \\ &\geq \int_0^\infty \int_0^{\sqrt{3}t/2} \int_{|g^{-1}h|<t} |\partial_s^{2\alpha} e^{-sL}|_{s=t^2} f(h)|^2 t^{4\alpha-1} s^{4\alpha-1} dh ds dt \\ &= C \int_0^\infty \int_{|g^{-1}h|<t} |t^{4\alpha} \partial_s^{2\alpha} e^{-sL}|_{s=t^2} f(h)|^2 \frac{dh dt}{t^{\mathcal{Q}+1}} \\ &= C (\tilde{\mathfrak{G}}_L^1 f(g))^2, \end{aligned} \tag{123}$$

which implies $\mathfrak{G}_{H,\alpha}(f) \in L^1(\mathbb{H}^n)$, and therefore, $f \in H_L^1(\mathbb{H}^n)$. Since $(t/(|g^{-1}h|+t))^{2\lambda} > 2^{-2\lambda}$ in the cone $\Gamma(g) = \{(h, t): |g^{-1}h|<t\}$, we have

$$\begin{aligned} \mathfrak{G}_{H,\alpha}(f)(g) &\leq \left[\int_{\Gamma(g)} 2^{2\lambda} \left(\frac{t}{|g^{-1}h|+t} \right)^{2\lambda} |Q_t^\alpha f(h)|^2 \frac{dh dt}{t^{\mathcal{Q}+1}} \right]^{1/2} \\ &\leq 2^\lambda \mathfrak{G}_{H,\alpha,\lambda}^*(f)(g). \end{aligned} \tag{124}$$

This completes the proof of Theorem 20.

Theorem 21. Let $\alpha \geq 1/2$ and $\lambda > \mathcal{Q}/2$. The following assertions are equivalent:

- (i) $f \in H_L^1(\mathbb{H}^n)$
- (ii) $f \in L^1(\mathbb{H}^n)$ and $\mathfrak{g}_{P,\alpha}(f) \in L^1(\mathbb{H}^n)$
- (iii) $f \in L^1(\mathbb{H}^n)$ and $\mathfrak{G}_{P,\alpha}(f) \in L^1(\mathbb{H}^n)$
- (iv) $f \in L^1(\mathbb{H}^n)$ and $\mathfrak{g}_{P,\alpha,\lambda}^*(f) \in L^1(\mathbb{H}^n)$

Moreover, for every $f \in H_L^1(\mathbb{H}^n)$,

$$\begin{aligned} \|f\|_{H_L^1} &\sim \|f\|_1 + \|\mathfrak{g}_{P,\alpha}(f)\|_1 \sim \|f\|_1 + \|\mathfrak{G}_{P,\alpha}(f)\|_1 \\ &\sim \|f\|_1 + \|\mathfrak{g}_{P,\alpha,\lambda}^*(f)\|_1. \end{aligned} \tag{125}$$

Proof. This theorem can be proved similarly as the proof of Theorem 20, so we omit it.

3.2. Fractional Square Functions Characterizations of $H_L^1(\mathbb{H}^n)$. In this part, we will give the characterizations of Hardy-Sobolev space $H_L^{1,\alpha}(\mathbb{H}^n)$ by fractional square functions. Firstly, we give the following Lemma, which will be used in the sequel. Similar to ([31], Proposition 2.4), we can express the operators $\partial_t^\alpha e^{-tL}$ and $\partial_t^\alpha e^{-t\sqrt{L}}$ as follows.

Lemma 22. Let $\alpha > 0$.

- (i) For every $f \in L^2(\mathbb{H}^n)$,

$$\partial_t^\alpha e^{-tL} f = e^{i\pi\alpha} \int_0^\infty \lambda^\alpha e^{-t\lambda} dE_L(\lambda) f, \quad t > 0. \tag{126}$$

- (ii) For every $f \in L^2(\mathbb{H}^n)$,

$$\partial_t^\alpha e^{-t\sqrt{L}} f = e^{i\pi\alpha} \int_0^\infty \lambda^{\alpha/2} e^{-t\sqrt{\lambda}} dE_L(\lambda) f, \quad t > 0. \tag{127}$$

Proof. Let $E(\lambda)$ denote a resolution of the identity. It follows from the spectral decomposition:

$$e^{-tL} f = \int_0^\infty e^{-\lambda t} dE_f(\lambda) \quad \forall f \in L^2(\mathbb{H}^n) \tag{128}$$

that

$$\partial_t^k e^{-tL} f = e^{-in\pi k} \int_0^\infty \lambda^k e^{-\lambda t} dE_f(\lambda), \quad k = 1, 2, \dots \quad (129)$$

By (6) and (129), we have

$$\partial_t^\alpha e^{-tL} f = \frac{e^{-in\alpha}}{\Gamma(k-\alpha)} \int_0^\infty \int_0^\infty \lambda^k e^{-(t+s)\lambda} dE_f(\lambda) s^{k-\alpha-1} ds, \quad (130)$$

where k is the smallest integer satisfying $k > \alpha$. Then, the integral

$$\int_0^\infty \int_0^\infty \lambda^k e^{-(t+s)\lambda} |dE_f(\lambda)| s^{k-\alpha-1} ds \quad (131)$$

is absolutely convergent. By the fact that $\|\partial_t^\alpha e^{-tL} f\|_{L^p} \leq C_\alpha \|f\|_{L^p/t^\alpha}$, the integral in (6) is absolutely convergent in $L^2(\mathbb{H}^n)$. Hence, by (130), we can get for $g \in L^2(\mathbb{H}^n)$,

$$\begin{aligned} \langle \partial_t^\alpha e^{-tL} f, g \rangle &= \left\langle \frac{e^{-in\alpha}}{\Gamma(k-\alpha)} \int_0^\infty \int_0^\infty \lambda^k e^{-(t+s)\lambda} dE_f(\lambda) s^{k-\alpha-1} ds, g \right\rangle \\ &= \frac{e^{-in\alpha}}{\Gamma(k-\alpha)} \int_0^\infty \int_0^\infty \lambda^k e^{-(t+s)\lambda} dE_{(f,g)}(\lambda) s^{k-\alpha-1} ds \\ &= \frac{e^{-in\alpha}}{\Gamma(k-\alpha)} \int_0^\infty \int_0^\infty \lambda^k e^{-(t+s)\lambda} s^{k-\alpha-1} ds dE_{(f,g)} \\ &= \left\langle e^{-in\alpha} \int_0^\infty \lambda^\alpha e^{-t\lambda} dE_f(\lambda), g \right\rangle, \end{aligned} \quad (132)$$

which implies (i). The assertion (ii) can be obtained by the aid of functional calculus similarly.

The following result can be deduced from Lemma 22 immediately.

Proposition 23.

(i) Let $0 < \alpha < k$, $k \in \mathbb{N}$ and $\lambda > \mathcal{Q}$. If $f \in D(L^\alpha) \cap H_L^1(\mathbb{H}^n)$ and $L^\alpha f \in L^2(\mathbb{H}^n) \cap H_L^1(\mathbb{H}^n)$. Then,

$$\|L^\alpha f\|_{H_L^1} \sim \|g_{k,\alpha}^H(f)\|_{L^1} \sim \|S_{k,\alpha}^H(f)\|_{L^1} \sim \|g_{k,\alpha,\lambda}^{H,*}(f)\|_{L^1}. \quad (133)$$

(ii) Let $0 < \alpha < k$, $k \in \mathbb{N}$ and $\lambda > \mathcal{Q}$. If $f \in D(L^\alpha) \cap H_L^1(\mathbb{H}^n)$ and $L^\alpha f \in L^2(\mathbb{H}^n) \cap H_L^1(\mathbb{H}^n)$. Then,

$$\|L^{\alpha/2} f\|_{H_L^1} \sim \|g_{k,\alpha}^P(f)\|_{L^1} \sim \|S_{k,\alpha}^P(f)\|_{L^1} \sim \|g_{k,\alpha,\lambda}^{P,*}(f)\|_{L^1}. \quad (134)$$

Proof. We only prove (i), and (ii) can be dealt with similarly.

Using Lemma 22, we can get

$$\partial_s^{k-\alpha} e^{-sL} \Big|_{s=t^2} (L^\alpha f) = L^{(k-\alpha)} e^{-t^2 L} (L^\alpha f) = L^k e^{-t^2 L} (f) = \partial_s^k K_s^L \Big|_{s=t^2} (f), \quad (135)$$

therefore,

$$g_{k-\alpha}^H(L^\alpha f) = g_{k,\alpha}^H(f), S_{k-\alpha}^H(L^\alpha f) = S_{k,\alpha}^H(f), g_{k-\alpha,\lambda}^{H,*}(L^\alpha f) = g_{k,\alpha,\lambda}^{H,*}(f). \quad (136)$$

Using Theorem 20, we can get

$$\|L^\alpha f\|_{H_L^1} \sim \|g_{k,\alpha}^H(f)\|_{L^1} \sim \|S_{k,\alpha}^H(f)\|_{L^1} \sim \|g_{k,\alpha,\lambda}^{H,*}(f)\|_{L^1}. \quad (137)$$

Let $G_{\alpha,L} = \{f \in H_L^1(\mathbb{H}^n): L^\alpha f \in C_c^\infty(\mathbb{H}^n)\}$. Since $C_c^\infty(\mathbb{H}^n)$ is dense in $H_L^1(\mathbb{H}^n)$, $G_{\alpha,L}$ is dense in $H_L^{1,\alpha}(\mathbb{H}^n)$. Note that $G_{\alpha,L} \subset D(L^\alpha) \cap H_L^1(\mathbb{H}^n)$, and

$$L^\alpha G_{\alpha,L} = C_c^\infty(\mathbb{H}^n) \subset L^2(\mathbb{H}^n) \cap H_L^1(\mathbb{H}^n). \quad (138)$$

Using Proposition 23, $g_{k,\alpha}^H$, $S_{k,\alpha}^H$, and $g_{k,\alpha,\lambda}^{H,*}$ can be extended to $H_L^{1,\alpha}(\mathbb{H}^n)$ as bounded operators from $H_L^{1,\alpha}(\mathbb{H}^n)$ to $L^1(\mathbb{H}^n)$. Let $\widetilde{g}_{k,\alpha}^H$ be the extension of $g_{k,\alpha}^H$ to $H_L^{1,\alpha}(\mathbb{H}^n)$ as a bounded operator from $H_L^{1,\alpha}(\mathbb{H}^n)$ to $L^1(\mathbb{H}^n)$. Then, there exists $C > 0$ such that for $f \in H_L^{1,\alpha}(\mathbb{H}^n)$,

$$\|f\|_{H_L^1} + \|\widetilde{g}_{k,\alpha}^H(f)\|_{L^1} \leq C \|f\|_{H_L^{1,\alpha}}. \quad (139)$$

Below, we give the square function characterizations of the Hardy-Sobolev space $H_L^{1,\alpha}(\mathbb{H}^n)$ as follows.

Theorem 24. Let $\alpha \geq 1/2$, $k \in \mathbb{N} \setminus \{0\}$, and $\lambda > \mathcal{Q}$. Then, the following assertions are equivalent:

- (i) $f \in H_L^{1,\alpha}(\mathbb{H}^n)$
- (ii) $f \in H_L^1(\mathbb{H}^n)$ and $g_{k,\alpha}^H(f) \in L^1(\mathbb{H}^n)$ for $k > \alpha$
- (iii) $f \in H_L^1(\mathbb{H}^n)$ and $S_{k,\alpha}^H(f) \in L^1(\mathbb{H}^n)$ for $\alpha < k - (\mathcal{Q} + 1)/2$
- (iv) $f \in H_L^1(\mathbb{H}^n)$ and $g_{k,\alpha,\lambda}^{H,*}(f) \in L^1(\mathbb{H}^n)$ for $\alpha < k - (\mathcal{Q} + 1)/2$

Moreover, for every $f \in H_L^{1,\alpha}(\mathbb{H}^n)$,

$$\begin{aligned} \|f\|_{H_L^{1,\alpha}} &\sim \|f\|_{H_L^1} + \|g_{k,\alpha}^H(f)\|_{L^1} \sim \|f\|_{H_L^1} + \|S_{k,\alpha}^H(f)\|_{L^1} \\ &\sim \|f\|_{H_L^1} + \|g_{k,\alpha,\lambda}^{H,*}(f)\|_{L^1}. \end{aligned} \quad (140)$$

Proof. We first prove $\|f\|_{H_L^1} + \|g_{k,\alpha}^H(f)\|_{L^1} \leq C \|f\|_{H_L^{1,\alpha}}$. By (139), it is sufficient to prove $\widetilde{g}_{k,\alpha}^H(f) = g_{k,\alpha}^H(f)$. For $N \in \mathbb{N}$ and $h \in H_L^1(\mathbb{H}^n)$, by the subordination formula, we obtain

$$\left| \frac{\partial^k}{\partial t^k} K_t^L(\mathbf{h})(g) \right| \leq C t^{-2k} \sup_{t>0} |T_{t^2/2}^L(\mathbf{h})(g)|. \quad (141)$$

Then,

$$\begin{aligned} & \left(\int_{1/N}^{\infty} \left| t^{2k-2\alpha} \frac{\partial^k}{\partial s^k} K_s^L \right|_{s=t^2} (\mathbf{h})(g) \right)^2 \frac{dt}{t} \Bigg|^{1/2} \\ & \leq C \left(\int_{1/N}^{\infty} t^{-1-4\alpha} dt \right)^{1/2} \sup_{t>0} |T_{t^2/2}^L(\mathbf{h})(g)| \\ & \leq C N^{2\alpha} \sup_{t>0} |T_{t^2/2}^L(\mathbf{h})(g)|. \end{aligned} \quad (142)$$

By the definition of $H_L^1(\mathbb{H}^n)$, we conclude that the operator

$$\mathbf{h} \rightarrow \left(\int_{1/N}^{\infty} \left| t^{2k-2\alpha} \frac{\partial^k}{\partial s^k} K_s^L \right|_{s=t^2} (\mathbf{h})(g) \right)^2 \frac{dt}{t} \Bigg|^{1/2} \quad (143)$$

is bounded from $H_L^1(\mathbb{H}^n)$ to $L^1(\mathbb{H}^n)$. Therefore, if $f = L^{-\alpha}\mathbf{h}$, where $\mathbf{h} \in H_L^1(\mathbb{H}^n) \cap L^2(\mathbb{H}^n)$, we have

$$\begin{aligned} & \left\| \left(\int_{1/N}^{\infty} \left| t^{2k-2\alpha} \frac{\partial^k}{\partial s^k} K_s^L \right|_{s=t^2} (L^{-\alpha}\mathbf{h})(g) \right)^2 \frac{dt}{t} \right\|_{L^1}^{1/2} \\ & = \left\| \left(\int_{1/N}^{\infty} \left| t^{2k-2\alpha} \frac{\partial^{k-\alpha}}{\partial s^{k-\alpha}} K_s^L \right|_{s=t^2} (\mathbf{h})(g) \right)^2 \frac{dt}{t} \right\|_{L^1}^{1/2} \\ & \leq C \|\mathbf{h}\|_{H_L^1}, \end{aligned} \quad (144)$$

where the positive constant C is independent of $N \in \mathbb{N}$. Letting $N \rightarrow \infty$ yields

$$\begin{aligned} \left\| \widetilde{\mathfrak{g}}_{k,\alpha}^H(f) \right\|_{L^1} & = \left\| \left(\int_0^{\infty} \left| t^{2k-2\alpha} \frac{\partial^k}{\partial s^k} K_s^L \right|_{s=t^2} (f)(g) \right)^2 \frac{dt}{t} \right\|_{L^1}^{1/2} \\ & \leq C \|f\|_{H_L^{1,\alpha}}. \end{aligned} \quad (145)$$

Since $G_{\alpha,L}$ is dense in $H_L^{1,\alpha}(\mathbb{H}^n)$, for $f \in H_L^{1,\alpha}(\mathbb{H}^n)$, we obtain

$$\begin{aligned} \left\| \mathfrak{g}_{k,\alpha}^H(f) \right\|_{L^1} & = \left\| \left(\int_0^{\infty} \left| t^{2k-2\alpha} \frac{\partial^k}{\partial s^k} K_s^L \right|_{s=t^2} (f)(g) \right)^2 \frac{dt}{t} \right\|_{L^1}^{1/2} \\ & = \left\| \widetilde{\mathfrak{g}}_{k,\alpha}^H(f) \right\|_{L^1} \leq C \|f\|_{H_L^{1,\alpha}}. \end{aligned} \quad (146)$$

The proofs for $S_{k,\alpha}^H$ and $\mathfrak{g}_{k,\alpha,\lambda}^{H,*}$ are similar, and so is omitted.

For the reverse, we only deal with the case of $\mathfrak{g}_{k,\alpha}^H$ for simplicity.

Step I. We prove

$$\left\| \partial_s^\alpha K_s^L|_{s=t^2}(f) \right\|_{H_L^1} \leq C t^{-2\alpha} \|f\|_{H_L^1}. \quad (147)$$

For $m \in \mathbb{N}$ and $m > \alpha$, by (141), we obtain

$$\begin{aligned} & \left| \sup_{\beta>0} T_\beta^L \left(\partial_s^\alpha K_s^L|_{s=t^2}(f)(g) \right) \right| \\ & \leq \left| \sup_{\beta>0} T_\beta^L \left(\int_0^{\infty} u^{m-\alpha-1} \frac{\partial^m}{\partial u^m} K_{u+s}^L|_{s=t^2}(f)(g) ds \right) \right| \\ & \leq C \left| \sup_{\beta>0} \int_0^{\infty} s^{m-\alpha-1} T_{(t^2+u)/2+\beta}^L(f)(g) (t^2+s)^{-m} ds \right| \\ & \leq C \sup_{t>0} |T_{t^2/2}^L(f)(g)| \int_0^{\infty} \frac{s^{m-\alpha-1}}{(t^2+s)^m} ds \\ & \leq C t^{-2\alpha} \sup_{t>0} |T_{t^2/2}^L(f)(g)|. \end{aligned} \quad (148)$$

Therefore, (147) follows from the definition of $H_L^1(\mathbb{H}^n)$.

Step II. Assume that $f \in H_L^1(\mathbb{H}^n)$ and $\mathfrak{g}_{k,\alpha}^H(f) \in L^1(\mathbb{H}^n)$. Let $\{f_n\}$ be a sequence in $C_c^\infty(\mathbb{H}^n)$ such that $\lim_{n \rightarrow \infty} f_n = f$ in $H_L^1(\mathbb{H}^n)$. For fixed $t > 0$, set $u(t, \cdot) := e^{-tL}(f)(\cdot)$ and $u_n(t, \cdot) := e^{-tL}(f_n)(\cdot)$, $n \in \mathbb{N}$. Then, $u(t, \cdot)$ and $u_n(t, \cdot)$ belong to $H_L^1(\mathbb{H}^n)$. By Lemma 22 and (147), we have

$$\partial_t^\alpha u_n(s, \cdot)|_{s=t^2} = L^\alpha u_n(t^2, \cdot) \in H_L^1(\mathbb{H}^n), \quad (149)$$

which implies that $u_n(t, \cdot) \in H_L^{1,\alpha}(\mathbb{H}^n)$ with $\|u_n(t^2, \cdot)\|_{H_L^{1,\alpha}} = \|u_n(t^2, \cdot)\|_{H_L^1} + \|\partial_t^\alpha u_n(s, \cdot)|_{s=t^2}\|_{H_L^1}$. By (147) again,

$$\lim_{n \rightarrow \infty} \|\partial_t^\alpha u_n(s, \cdot)|_{s=t^2} - \partial_t^\alpha u(s, \cdot)|_{s=t^2}\|_{H_L^1} = 0. \quad (150)$$

This indicates that $\{u_n(t^2, \cdot)\}$ is a Cauchy sequence in $H_L^{1,\alpha}(\mathbb{H}^n)$. Therefore, there exists $v(t, \cdot) \in H_L^{1,\alpha}(\mathbb{H}^n)$ such that $\|u_n(t^2, \cdot) - v(t, \cdot)\|_{H_L^{1,\alpha}} \rightarrow 0$ as $n \rightarrow \infty$. Hence, $\|u_n(t^2, \cdot) - v(t, \cdot)\|_{H_L^1} \rightarrow 0$ as $n \rightarrow \infty$, which yields $u(t^2, \cdot) = v(t, \cdot) \in H_L^{1,\alpha}(\mathbb{H}^n)$ and $\|u_n(t^2, \cdot) \rightarrow u(t^2, \cdot)\|_{H_L^{1,\alpha}} \rightarrow 0$ as $n \rightarrow \infty$.

Step III. Noting that $u_n(t^2, \cdot) \in L^2(\mathbb{H}^n) \cap H_L^1(\mathbb{H}^n)$ and $L^\alpha u_n(t^2, \cdot) \in L^2(\mathbb{H}^n) \cap H_L^1(\mathbb{H}^n)$, by Proposition 23, we get

$$\|u_n(t^2, \cdot)\|_{H_L^1} + \|\mathfrak{g}_{k,\alpha}^H(u_n(t^2, \cdot))\|_1 \sim \|u_n(t^2, \cdot)\|_{H_L^{1,\alpha}}. \quad (151)$$

Letting $n \rightarrow \infty$, we have $\|u(t^2, \cdot)\|_{H_L^{1,\alpha}} \leq C(\|u(t^2, \cdot)\|_{H_L^1} + \|\mathfrak{g}_{k,\alpha}^H u(t^2, \cdot)\|_1)$. Since

$$\begin{aligned}
\mathfrak{g}_{k,\alpha}^H(f^t)(g) &= \left(\int_0^\infty \left| s^{2k-2\alpha} \frac{\partial^k}{\partial t^k} K_t^L \right|_{t=s^2} (u(t^2, \cdot))(g) \right)^2 \frac{ds}{s} \Big)^{1/2} \\
&= \left(\int_0^\infty \left| e^{-t^2 L} s^{2k-2\alpha} \frac{\partial^k}{\partial t^k} K_t^L \right|_{t=s^2} (f)(g) \right)^2 \frac{ds}{s} \Big)^{1/2} \\
&\leq e^{-t^2 L} \left[\left(\int_0^\infty \left| s^{2k-2\alpha} \frac{\partial^k}{\partial t^k} K_t^L \right|_{t=s^2} (f)(\cdot) \right)^2 \frac{ds}{s} \right]^{1/2} (g),
\end{aligned} \tag{152}$$

we get $\|\mathfrak{g}_{k,\alpha}^H(u(t, \cdot))\|_1 \leq \|\mathfrak{g}_{k,\alpha}^H(f)\|_1$. Furthermore, this gives

$$\|u(t^2, \cdot)\|_{H_L^{1,\alpha}} \leq C \left(\|u(t^2, \cdot)\|_{H_L^1} + \|\mathfrak{g}_{k,\alpha}^H(f)\|_1 \right), \tag{153}$$

where $C > 0$ is independent of t . By (153), we know $\{u(t^2, \cdot)\}$ are uniformly bounded in $H_L^{1,\alpha}(\mathbb{H}^n)$, i.e., $\{L^\alpha(u(t^2, \cdot))\}$ are uniformly bounded in $H_L^1(\mathbb{H}^n)$. Since $H_L^1(\mathbb{H}^n)$ is a Banach space, we can find $g \in H_L^1(\mathbb{H}^n)$ such that $L^\alpha(u_j(t^2, \cdot)) \rightarrow g$ as $j \rightarrow \infty$, where $\{u_j(t^2, \cdot)\}$ is a subsequence of $\{u(t^2, \cdot)\}$. Since $H_L^1(\mathbb{H}^n)$ is the dual space of $VMO_L(\mathbb{H}^n)$ and $C_c^\infty(\mathbb{H}^n)$ is dense in $VMO_L(\mathbb{H}^n)$ with norm of $VMO_L(\mathbb{H}^n)$ (cf. [32]), we get $\lim_{j \rightarrow \infty} \langle L^\alpha(u_j(t^2, \cdot)), \phi \rangle = \langle g, \phi \rangle, \phi \in C_c^\infty(\mathbb{H}^n)$. Let $h = L^{-\alpha}g$. Then, $h \in H_L^{1,\alpha}(\mathbb{H}^n)$ and $\lim_{j \rightarrow \infty} \langle (u_j(t^2, \cdot)), \phi \rangle = \langle h, \phi \rangle, \phi \in C_c^\infty(\mathbb{H}^n)$. By the arguments analogous to ([33] page 776), which rely on the decay of the kernel of e^{-tL} , we can get

$$\lim_{t \rightarrow 0} \langle u(t^2, \cdot), \phi \rangle = \langle f, \phi \rangle, \quad \phi \in C_c^\infty(\mathbb{H}^n). \tag{154}$$

It follows that $f = h$ and

$$\|f\|_{H_L^{1,\alpha}} \leq C \left(\|f\|_{H_L^1} + \|\mathfrak{g}_{k,\alpha}^H(f)\|_{L^1} \right). \tag{155}$$

This completes the proof of Theorem 24.

For the Poisson semigroup $\{P_t^L\}_{t>0}$, we define the fractional square functions as follows:

$$\begin{cases} \mathfrak{g}_{k,\alpha}^P(f) := \left(\int_0^\infty \left| t^{k-\alpha} \frac{\partial^k P_t^L}{\partial t^k} f \right|^2 \frac{dt}{t} \right)^{1/2}, & k \geq \alpha > 0; \\ \mathfrak{S}_{k,\alpha}^P(f) := \left(\int_0^\infty \int_{B(g,t)} \left| t^{k-\alpha} \frac{\partial^k P_t^L}{\partial t^k} f \right|^2 \frac{dhdt}{t^{2\alpha+1}} \right)^{1/2}, & k \geq \alpha > 0; \\ \mathfrak{g}_{k,\alpha,\lambda}^{P,*}(f) := \left(\int_0^\infty \int_{\mathbb{H}^n} \left(\frac{t}{t+|g^{-1}h|} \right)^{2\lambda} \left| t^{k-\alpha} \frac{\partial^k P_t^L}{\partial t^k} f \right|^2 \frac{dhdt}{t^{2\alpha+1}} \right)^{\frac{1}{2}}, & k \geq \alpha > 0. \end{cases} \tag{156}$$

Similar to the proof of Theorem 24, we can apply (ii) of Proposition 23 to establish the following characterization of $H_L^{1,\alpha}(\mathbb{H}^n)$ via the fractional square functions related to the Poisson semigroup. We omit the proof.

Theorem 25. Let $\alpha \geq 1/2$, $k \in \mathbb{N} \setminus \{0\}$ and $\lambda > \mathcal{Q}$. Then, the following assertions are equivalent:

- (i) $f \in H_L^{1,\alpha}(\mathbb{H}^n)$
- (ii) $f \in H_L^1(\mathbb{H}^n)$ and $\mathfrak{g}_{k,\alpha}^P(f) \in L^1(\mathbb{H}^n)$ for $k > \alpha$
- (iii) $f \in H_L^1(\mathbb{H}^n)$ and $\mathfrak{S}_{k,\alpha}^P(f) \in L^1(\mathbb{H}^n)$ for $\alpha < k - (\mathcal{Q} + 1)/2$
- (iv) $f \in H_L^1(\mathbb{H}^n)$ and $\mathfrak{g}_{k,\alpha,\lambda}^{P,*}(f) \in L^1(\mathbb{H}^n)$ for $\alpha < k - (\mathcal{Q} + 1)/2$

Moreover, for every $f \in H_L^{1,\alpha}(\mathbb{H}^n)$,

$$\begin{aligned}
\|f\|_{H_L^{1,\alpha}} &\sim \|f\|_{H_L^1} + \|\mathfrak{g}_{k,\alpha}^P(f)\|_{L^1} \sim \|f\|_{H_L^1} + \|\mathfrak{S}_{k,\alpha}^P(f)\|_{L^1} \\
&\sim \|f\|_{H_L^1} + \|\mathfrak{g}_{k,\alpha,\lambda}^{P,*}(f)\|_{L^1}.
\end{aligned} \tag{157}$$

3.3. Equivalent Norms of Hardy-Sobolev Spaces. We define the following Hardy-Sobolev space $\mathcal{H}_L^{1,\alpha}(\mathbb{H}^n)$ as the set of all functions $f \in H_L^1(\mathbb{H}^n)$ such that $(I + L)^\alpha f \in H_L^1(\mathbb{H}^n)$, with the norm

$$\|f\|_{\mathcal{H}_L^{1,\alpha}} = \|(I + L)^\alpha f\|_{H_L^1} + \|f\|_{H_L^1}. \tag{158}$$

The purpose of this section is to characterize $\mathcal{H}_L^{1,\alpha}(\mathbb{H}^n)$ by the fractional square functions defined by (10) and (156), respectively. As an application, it follows from the fractional square function characterizations of $\mathcal{H}_L^{1,\alpha}(\mathbb{H}^n)$ and $H_L^{1,\alpha}(\mathbb{H}^n)$ that the two Hardy-Sobolev spaces are equivalent.

Let E_L be the spectral decomposition of the operator L . For a bounded function M on $(0, \infty)$, the spectral multiplier $M(L)$ is defined by

$$M(L)f = \int_0^\infty M(\lambda) dE_L(\lambda)f, \quad f \in D(M(L)), \tag{159}$$

where $D(M(L))$ denotes the domain, i.e.,

$$D(M(L)) = \left\{ f \in L^2(\mathbb{H}^n) : \int_0^\infty |M(\lambda)|^2 \langle dE_L(\lambda)f, f \rangle < \infty \right\}. \tag{160}$$

We say that a function M on $(-\infty, +\infty)$ belongs to the space $C(s)$, $s > 0$, if

$$\|M\|_{C(s)} := \begin{cases} \sum_{k=0}^s \sup |M^{(k)}(\lambda)| < \infty, & s \in \mathbb{Z}; \\ \left\| M^{(s)} \right\|_{Lip(s-|s|)} + \sum_{k=0}^{|s|} \sup |M^{(k)}(\lambda)| < \infty, & s \notin \mathbb{Z}. \end{cases} \tag{161}$$

We have the following version of spectral multiplier theorems.

Proposition 26 (see [34], Theorem 1.11). *Let M be a bounded continuous function on $(0, \infty)$. If for some $\varepsilon > 0$ and a non-zero function $\phi \in C_c^\infty(0, \infty)$, there exists a constant $C > 0$ such that for every $t > 0$,*

$$\|\phi(\cdot)M(t \cdot)\|_{C(\mathbb{R}/2+\varepsilon)} \leq C, \quad (162)$$

then the operator $M(L)$ is bounded on $H_L^1(\mathbb{H}^n)$.

Let $\alpha, \beta > 0$. For $\lambda > 0$, define

$$M_1(\lambda) = \frac{\lambda^\alpha}{(1+\lambda)^\alpha}, \quad M_2(\lambda) = \frac{(1+\lambda)^\alpha}{1+\lambda^\alpha}, \quad M_3(\lambda) = (\beta + \lambda)^{-\alpha}. \quad (163)$$

Then, it is clear that $M_i, i = 1, 2, 3$, are smooth and bounded on $(0, \infty)$. It follows from Proposition 26 that

Proposition 27. *Let $\alpha, \beta > 0$. The operators $M_i(L), i = 1, 2, 3$, can be extended to bounded operators on $H_L^1(\mathbb{H}^n)$.*

Theorem 28. *Let $0 < \alpha < k, k \in \mathbb{N}$ and $\lambda > \mathcal{Q}$. If $f \in D((I+L)^\alpha) \cap H_L^1(\mathbb{H}^n)$ and $(I+L)^\alpha f \in L^2(\mathbb{H}^n) \cap H_L^1(\mathbb{H}^n)$,*

$$\begin{aligned} \|(I+L)^\alpha f\|_{H_L^1} &\sim \|f\|_{H_L^1} + \|g_{k,\alpha}^H(f)\|_{L^1} \\ &\sim \|f\|_{H_L^1} + \|S_{k,\alpha}^H(f)\|_{L^1} \sim \|f\|_{H_L^1} + \|g_{k,\alpha,\lambda}^{H,*}(f)\|_{L^1}. \end{aligned} \quad (164)$$

Proof. We give the proof of $\|(I+L)^\alpha f\|_{H_L^1} \sim \|f\|_{H_L^1} + \|g_{k,\alpha}^H(f)\|_{L^1}$. The proofs for the cases of $S_{k,\alpha}^H(f)$ and $g_{k,\alpha,\lambda}^{H,*}(f)$ are similar. By Proposition 27, we know that the operators $L^\alpha (I+L)^{-\alpha}$ and $(I+L)^\alpha (I+L^\alpha)^{-1}$ are bounded on $H_L^1(\mathbb{H}^n)$. Then, following from Proposition 23, we obtain

$$\begin{aligned} \|(I+L)^\alpha f\|_{H_L^1} &= \left\| (I+L)^\alpha (I+L^\alpha)^{-1} (I+L^\alpha) f \right\|_{H_L^1} \\ &\leq \|(I+L^\alpha) f\|_{H_L^1} \\ &\leq C \left(\|f\|_{H_L^1} + \|L^\alpha f\|_{H_L^1} \right) \\ &\leq C \left(\|f\|_{H_L^1} + \|g_{k,\alpha}^H(f)\|_{L^1} \right). \end{aligned} \quad (165)$$

For the reverse, we take the function $M_1(\lambda) = \lambda^\alpha (1+\lambda)^{-\alpha}$, $\lambda > 0$. For any $r \in (0, \infty)$,

$$\begin{aligned} \int_0^r \lambda^\alpha dE_L(\lambda) f &= \int_0^r \frac{\lambda^\alpha}{(1+\lambda)^\alpha} (1+\lambda)^\alpha dE_L(\lambda) f \\ &= M_1(L) \int_0^r (1+\lambda)^\alpha dE_L(\lambda) f. \end{aligned} \quad (166)$$

Letting $r \rightarrow \infty$, we get $L^\alpha(f) = M_1(\lambda)(I+L)^\alpha(f)$. By Proposition 27 again, we obtain $\|L^\alpha f\|_{H_L^1} \leq C\|(I+L)^\alpha f\|_{H_L^1}$, and

$$\|f\|_{H_L^1} = \|(I+L)^{-\alpha}(I+L)^\alpha f\|_{H_L^1} \leq C\|(I+L)^\alpha f\|_{H_L^1}. \quad (167)$$

Theorem 28 follows from Proposition 23.

Similar to Theorem 28, we also can obtain

Theorem 29. *Let $0 < \alpha < k, k \in \mathbb{N}$ and $\lambda > \mathcal{Q}$. If $f \in D((I+L)^{\alpha/2}) \cap H_L^1(\mathbb{H}^n)$ and $(I+L)^{\alpha/2} f \in L^2(\mathbb{H}^n) \cap H_L^1(\mathbb{H}^n)$,*

$$\begin{aligned} \|(I+L)^{\alpha/2} f\|_{H_L^1} &\sim \|f\|_{H_L^1} + \|g_{k,\alpha}^P(f)\|_{L^1} \\ &\sim \|f\|_{H_L^1} + \|S_{k,\alpha}^P(f)\|_{L^1} \sim \|f\|_{H_L^1} + \|g_{k,\alpha,\lambda}^{P,*}(f)\|_{L^1}. \end{aligned} \quad (168)$$

Let

$$G_{\alpha,L} = \{f \in H_L^1(\mathbb{H}^n): (I+L)^\alpha f \in C_c^\infty(\mathbb{H}^n)\}. \quad (169)$$

Since $C_c^\infty(\mathbb{H}^n)$ is dense in $\mathcal{H}_L^1(\mathbb{H}^n)$, $G_{\alpha,L}$ is dense in $\mathcal{H}_L^{1,\alpha}(\mathbb{H}^n)$. Note that $G_{\alpha,L} \subset D((I+L)^\alpha) \cap H_L^1(\mathbb{H}^n)$, and

$$(I+L)^\alpha G_{\alpha,L} = C_c^\infty(\mathbb{H}^n) \subset L^2(\mathbb{H}^n) \cap H_L^1(\mathbb{H}^n). \quad (170)$$

Using Theorem 28, $g_{k,\alpha}^H, S_{k,\alpha}^H$, and $g_{k,\alpha,\lambda}^{H,*}$ can be extended to $H_L^{1,\alpha}(\mathbb{H}^n)$ as bounded operators from $\mathcal{H}_L^{1,\alpha}(\mathbb{H}^n)$ to $L^1(\mathbb{H}^n)$. Let $\widetilde{g_{k,\alpha}^H}$ be the extension of $g_{k,\alpha}^H$ to $\mathcal{H}_L^{1,\alpha}(\mathbb{H}^n)$ as a bounded operator from $\mathcal{H}_L^{1,\alpha}(\mathbb{H}^n)$ to $L^1(\mathbb{H}^n)$. Then, there exists $C > 0$ such that for $f \in \mathcal{H}_L^{1,\alpha}(\mathbb{H}^n)$, $\|f\|_{H_L^1} + \|\widetilde{g_{k,\alpha}^H}(f)\|_{L^1} \leq C\|f\|_{\mathcal{H}_L^{1,\alpha}}$.

Similar to Theorems 24 and 25, we will give the following characterizations of the Hardy-Sobolev space $\mathcal{H}_L^{1,\alpha}(\mathbb{H}^n)$ as follows. We omit the proof.

Theorem 30. *Let $\alpha \geq 1/2, k \in \mathbb{N} \setminus \{0\}$ and $\lambda > \mathcal{Q}$. The following assertions are equivalent:*

- (i) $f \in \mathcal{H}_L^{1,\alpha}(\mathbb{H}^n)$
- (ii) $f \in H_L^1(\mathbb{H}^n)$ and $g_{k,\alpha}^H(f) \in L^1(\mathbb{H}^n)$ for $k > \alpha$
- (iii) $f \in H_L^1(\mathbb{H}^n)$ and $S_{k,\alpha}^H(f) \in L^1(\mathbb{H}^n)$ for $\alpha < k - (\mathcal{Q} + 1)/2$
- (iv) $f \in H_L^1(\mathbb{H}^n)$ and $g_{k,\alpha,\lambda}^{H,*}(f) \in L^1(\mathbb{H}^n)$ for $\alpha < k - (\mathcal{Q} + 1)/2$

Moreover, for every $f \in \mathcal{H}_L^{1,\alpha}(\mathbb{H}^n)$,

$$\begin{aligned} \|f\|_{\mathcal{H}_L^{1,\alpha}} &\sim \|f\|_{H_L^1} + \|g_{k,\alpha}^H(f)\|_{L^1} \sim \|f\|_{H_L^1} + \|S_{k,\alpha}^H(f)\|_{L^1} \\ &\sim \|f\|_{H_L^1} + \|g_{k,\alpha,\lambda}^{H,*}(f)\|_{L^1}. \end{aligned} \quad (171)$$

Theorem 31. *Let $\alpha \geq 1/2, k \in \mathbb{N} \setminus \{0\}$ and $\lambda > \mathcal{Q}$. The following assertions are equivalent:*

- (i) $f \in \mathcal{H}_L^{1,\alpha}(\mathbb{H}^n)$

- (ii) $f \in H_L^1(\mathbb{H}^n)$ and $g_{k,\alpha}^p(f) \in L^1(\mathbb{H}^n)$ for $k > \alpha$
- (iii) $f \in H_L^1(\mathbb{H}^n)$ and $S_{k,\alpha}^p(f) \in L^1(\mathbb{H}^n)$ for $\alpha < k - (\mathcal{Q} + 1)/2$
- (iv) $f \in H_L^1(\mathbb{H}^n)$ and $g_{k,\alpha,\lambda}^{p,*}(f) \in L^1(\mathbb{H}^n)$ for $\alpha < k - (\mathcal{Q} + 1)/2$

Moreover, for every $f \in \mathcal{H}_L^{1,\alpha}(\mathbb{H}^n)$,

$$\begin{aligned} \|f\|_{\mathcal{H}_L^{1,\alpha}} &\sim \|f\|_{H_L^1} + \|g_{k,\alpha}^p(f)\|_{L^1} \sim \|f\|_{H_L^1} + \|S_{k,\alpha}^p(f)\|_{L^1} \\ &\sim \|f\|_{H_L^1} + \|g_{k,\alpha,\lambda}^{p,*}(f)\|_{L^1}. \end{aligned} \quad (172)$$

Theorems 24, 25, 30, and 31 indicate the following equivalence relation:

Corollary 32. Let $\alpha \geq 1/2$. $\mathcal{H}_L^{1,\alpha}(\mathbb{H}^n) = H_L^{1,\alpha}(\mathbb{H}^n)$.

Data Availability

The data used to support the findings of this study have not been made available because this is a mathematical article, which is pure theoretical proof and derivation, no specific data information.

Conflicts of Interest

The authors declare that there is no conflict of interest regarding the publication of this paper.

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