Quantum Paraelectric Glass State in SrCu$_3$Ti$_4$O$_{12}$

Jitender Kumar, Ram Janay Choudhary, and A.M. Awasthi*
UGC-DAE Consortium for Scientific Research, University Campus, Khandwa Road, Indore- 452 001, INDIA

Abstract
Magnetic and dielectric studies of SrCu$_3$Ti$_4$O$_{12}$ carried out over 5-300K confirm antiferromagnetic (AFM) ordering of Cu-spins at $T_N = 23$K. Dielectric constant $\varepsilon'$ measured across 1Hz-1MHz signifies quantum paraelectric character, Barrett-fittable almost down to $T_N$. Competition of athermal fluctuations and the literature-reported magneto-phonon-softening near $T_N$ manifests a quantum paraelectric glass (QPG) state. Emergent AFM-field tunes the otherwise quantum ordering (at absolute-zero) of the dipoles to finite-temperature kinetic glass transition; spectral dispersion of dielectric constant unambiguously manifested and characterized. Vogel-Fulcher glass-kinetics parameterization sets the almost relaxation-free QPG state in SrCu$_3$Ti$_4$O$_{12}$ apart from an emergent scaling-class, to which typical ferroelectric relaxors belong.

Keywords: Quantum Paraelectric, Magneto-Dielectricity, Glassy Kinetics.

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The physics of quantum fluctuations and its effects on the properties of the host-materials have recently gained prominence and attention of condensed matter researchers. Resultant emergent phenomena/states are fascinating and important from the viewpoint of basic and materials science; two of them being quantum paraelectrics\(^1\) (QP) and quantum spin-liquids.\(^2\) Quantum paraelectrics are important due to their huge piezoelectric effect at cryogenic temperatures,\(^3\) whereas quantum spin-liquids play dominant role in the low-temperature metal-insulator transitions, Mott-insulators, and superconductivity.\(^2\) Prime effect of quantum fluctuations is to prevent a macroscopic ordering down to 0K, by sustaining an athermal disordered state.\(^4\) In recent years, excellent articles have appeared on superconductivity of the hybrids of graphene and a magnetic material, wherein due to the proximity effects, a superconducting state is observed at the nano-scale.\(^5\) According to Loffe and Michael,\(^6\) dynamical inhomogeneity due to quantum fluctuations hinders the long range superconductivity in graphene. A general schematic of fig.1 represents the systems with quantum fluctuations having spin/dipolar degrees of freedom. The possible emergent matter-states include (i) magneto-electric multiferroics\(^7\) from the coupling of long-range-ordered magnetic and electrical degrees of freedom, (ii) quantum paraelectrics\(^1\) from athermal fluctuations of dipolar degrees of freedom, (iii) quantum spin liquids\(^2\) from athermal fluctuations of magnetic spins, and (iv) disordered states from the interplay of coupled spin and dipolar degrees of freedom and athermal fluctuations; quantum paraelectric glass (QPG), quantum spin glass (QSG), or a quantum multi-glass (QMG). Of these, QSG would be akin to a ‘multiferroic’ Griffith’s phase, driven by a long-range electrical order. For example, the emergent (magneto-electric) multiglass state\(^8\) in (Sr,Mn)TiO$_3$ is realized, though by static disorder (Mn-doping) in the parent SrTiO$_3$ quantum paraelectric.

* Corresponding Author e-mail: amawasthi@csr.res.in. Tel: +91 731 2463913.
In quantum paraelectrics (QP’s), the ferroelectric (FE) ground state is suppressed (no FE-$T_C$ down to 0K) by the quantum zero-point fluctuations;\(^1\) they are also called the *incipient ferroelectrics*.\(^{1,9}\) The prototype quantum paraelectrics\(^{1,10-11}\) are SrTiO$_3$, KTaO$_3$, and CaTiO$_3$. QP’s show huge and $T$-independent dielectric constant at cryogenic temperatures.\(^1\) This is realized due to the subtle balance of soft-phonon mode and quantum fluctuations; therefore, non-thermal external influences like electric field, pressure, or impurity-doping can create a ferroelectric or a relaxor ground state.\(^{10,12-13}\) In nearly all of the QP’s, a quantum critical phase transition\(^4\) (QPT) results from external-tuning of the (otherwise) 0K transition to finite temperatures; e.g., a ferroelectric or relaxor state is achieved in non-magnetic (Ba, Bi)-doped SrTiO$_3$,\(^{14-15}\) Pb-doped CaTiO$_3$,\(^{16}\) and in Li-doped KTaO$_3$.\(^{17}\) The disorder created by the doping of a magnetic atom is also quite able to suppress the quantum fluctuations of dipole-moments, resulting in the polar nano-regions (PNR’s) and/or spin-glass behavior.\(^9\) Another important quantum paraelectric EuTiO$_3$ shows AFM transition at the Nèel temperature $T_N$ =5.5K,\(^{18}\) below which the dielectric constant drops. EuTiO$_3$ shows magneto-dielectric effect near $T_N$, since Eu-spins are strongly coupled with the soft-phonon mode.

We present SrCu$_3$Ti$_4$O$_{12}$ (SCTO) as the latest quantum paraelectric, which undergoes AFM-driven electrical vitrification, resulting from the competition of athermal quantum dipolar fluctuations and the relevant-phonon-softening accompanying the G-type antiferromagnetic (AFM) ordering\(^{19}\) at 23K. Here, the emergent magnetic field of the ordered spin-system tunes the otherwise 0K transition (as per indicated by the Barrett-fit permittivity) to the observed finite-temperature quantum critical kinetic phase transition, realizing the QPG state. Thus, SCTO is second to EuTiO$_3$ in deviating at finite-$T$ under internal field, from its parent QP character. SCTO belongs to the ACu$_3$Ti$_4$O$_{12}$ family of copper-titanates; better known as colossal dielectric constant (CDC) materials, viz., CaCu$_3$Ti$_4$O$_{12}$ (CCTO).\(^{20}\) Both SCTO and CCTO have cubic double-Perovskite structure with space group $Im3$,\(^{19,21}\) but have huge differences in their dielectric properties. Moreover, the low-temperature dielectric investigation of SCTO (down to liquid-Helium range) and the prospects of magneto-dielectricity have not been explored, which we present here.

The ceramic SCTO samples were prepared from high purity (99.99%) powders of SrCO$_3$, CuO, and TiO$_2$ by the conventional solid state route. For making of good quality samples we ground the mixed charge of precursors for more than 45 hrs. and calcined it at 1050°C for 24 hours. The pelletized samples (10 mm diameter and 1-3mm thick) were sintered at 1100°C for 24 hours and their flat faces were silver-coated to make good electrical contacts for the dielectric measurements. X-Ray diffraction of the samples has been done with Cu-$K_a$ radiation ($\lambda$ =1.54Å), using a Bruker D8 Advance X-ray Rotating-anode powder diffractometer. Dielectric measurements over 4.2K-room temperature spanning 1Hz to 1MHz with 1V ac-excitation were performed using (Alpha-A) High Performance Frequency Analyzer (NOVO-CONTROL). The magnetization data were collected from 2-300K using 7-Tesla SQUID-vibrating sample magnetometer (SVSM; Quantum Design Inc., USA).

Phase-purity and crystal structure of the samples were analyzed by the Rietveld analysis using the fullprof software. Rietveld refinement ($\chi^2 \sim 2.2$, goodness of fit 1.5) provided the lattice constant
7.4050(1)Å for the cubic space group Im3 without any secondary phase. The crystal structure (VESTA software) of SCTO is shown in fig.2, made using the fitting parameters obtained from the Rietveld refinement. Like other family members,20,22 SCTO too has tilted Ti-O₆ octahedra (direction $x=0→1$, $y=0.25→0.75$, $z=0.25→0.5$), as shown in fig.2 inserts. This octahedral-tilt makes the Ti-O-Ti bond-angle 141.83(14)° (as per determined from our results, resembling an earlier report19,21) instead of 180°, and forms square-planar arrangement of Cu-O₆, with Cu at the center and O’s at the corners.20 Quantum paraelectricity in SCTO is rooted in its crystal structure. For example, BaTiO₃ is ferroelectric while (Sr/Ca)TiO₃ are quantum paraelectrics. Ionic-size of Ba is larger than that of Sr/Ca, thus providing larger space for the Ti-O₆ octahedral cage to expand. Therefore, the Ti⁺⁺-ions rattle easily in the former and as a result BaTiO₃ undergoes series of phase transitions, which is not possible in (Sr/Ca)TiO₃.23 SrCu₃Ti₄O₁₂ (SCTO) also has the tilted Ti-O₆ octahedral structure and the Ti-O bond-length 1.958(4)Å is similar to that in SrTiO₃ (1.95Å); i.e., smaller compared to 2Å in BaTiO₃. Suppressed rattling/displacement of Ti⁺⁺ in the octahedral-cage kills bulk ferroelectricity in the centric SCTO, while fluctuating short-range dipolar correlations impart the system quantum paraelectricity.

Temperature dependent magnetization $M(T)$ of SCTO at 100 Oe is shown in fig.3, resembling an earlier published report.19 The AFM order is observed at the Néel temperature $T_N=23±0.01$K close to that reported for CCTO,24 with little observable difference in ZFC and FC data. High magnetic field up to 7T does not affect the $T_N$, confirming rather robust exchange interaction responsible for the G-type AFM order. By Curie-Weiss linear-fit of the $1/\chi-T$ data, $\Theta_{CW}=-39.1±0.1$K and effective magnetic moment $\mu_{eff}/Cu$-ion = 2.09±0.0006μ₄B are evaluated (fig.3, right y-axis). The metric of magnetic frustration25 $f=|\Theta_{CW}|/T_N ≈ 1.73$ for SCTO is though larger than $f ≈ 1$ for CCTO; implying presence (absence) of AFM fluctuations above their respective $T_N$’s in SCTO (CCTO), also indicated by the deviation of the SCTO $1/\chi-T$ data from the perfect Curie-Weiss fit at $T \ge T_N$ (fig.3 inset). In SCTO, only Cu (Cu⁺⁺, d⁹) carrying $s =1/2$ spin in the 3d shell orders collinearly along the crystallographic [111] direction.19 The first- and third- nearest neighbors interact antiferromagnetically, whereas the interaction between the second-neighbor Cu-ions is ferromagnetic in nature. A direct interaction between Cu⁺⁺ ions is scarce, because the distance between these ions is quite large. Indirect super-exchange between Cu-ions is mediated through the Ti⁺⁺ ions (similar to CCTO),24 endowing SCTO more direct magneto-dielectricity; Ti⁺⁺ cations being also the constituent of electric-polarizability in the Ti-O₆ octahedra provide a platform for major influence of magnetic ordering on the electrical degrees of freedom.

Figure 4 shows the dielectric constant of SCTO from 4.5K to room temperature at 800 kHz, the high-frequency most clearly providing both the classical and quantum temperature-regimes as seen below. Sample quality plays an important role in the dielectric characterization of SCTO; e.g., the dielectric constant of impure samples is higher in comparison to the pure one.19 In the case of our specimen, the value of dielectric constant (~73) is close to the intrinsic value predicted by the first principles theory (for similarly-structured CCTO, which should be less than 100 at room temperature26), ensuring very good sample quality, free from any static/structural disorders.19 It is
therefore suggested that the extrinsic (Maxwell-Wagner) effects present in CCTO (responsible for its huge dielectric constant) are absent in SCTO over similar T-range. Almost down to $T_N$, the temperature dependence of real permittivity fits well the Barrett formula, the theoretical model available for the quantum paraelectrics, given as 

$$\varepsilon'(T) = A + C/[(T_i/2)\coth(T_i/2T) - T_c].$$

Here $T_c = -63.35 \pm 3.9$K is equivalent to $\Theta_{C,W} = -419 \pm 0.7$K, obtained from the classical (high-T) Curie-Weiss linear-fit. Below $\sim T_i$, the two (quantum and classical) behaviors are supposed to split; happening at $\approx 155$K, independent of the probed frequencies. Barrett-fit to the data at 800kHz provides $T_1 = 154.67 \pm 2.5$K, indicating that the high-temperature classical behavior is accurately reflected in the Barrett parameters derived from high frequency data. In the inset, we show the dramatic rise of the normalized deviance $[(\varepsilon'_b/\varepsilon'_c) - 1]$ below $T_1$, as a metric of the net quantum paraelectric (QP) character. Barrett’s turnover to plateau makes this QP-metric drop below $\sim 50$K. Barrett fit confers QP character to SCTO; high-T antiferroelectric correlations of dipoles ($\varepsilon'_b/\varepsilon'_c$ split below $T_1$ and -ve value of $T_c$) exclude their low-T organization into polar nano-regions (PNR’s of relaxors), or to a robust ferroelectric state.

The dipolar and spin degrees of freedom in SCTO are directly coupled as follows. Individual oxygens of Ti-O$_6$ octahedron are each bonded to a different Cu-atom and each Cu-O$_4$ forms a square-planar arrangement; and this sharing tilts the Ti-O$_6$ octahedra, making the Ti-O-Ti bond-angle 141.83° instead of 180°. As these oxygens are associated with the Ti-O phonon, their sharing between Ti-O$_6$ octahedra and Cu-O$_4$ square-planes (reflected in the octahedral-tilt) means a change in Cu-spin arrangement affects the Ti-O phonon. This spin-phonon coupling determines the magneto-electricity in SCTO, as also reported e.g., in DyMn$_2$O$_5$. Raman signatures of spin-phonon coupling in SCTO have been recently reported below the Néel temperature $T_N$, Cu-spins arrange antiferromagnetically and the associated Ti-O phonon ($A_g$1 rotation-like mode at 442cm$^{-1}$) softens, registering the observed drop (fig.4) in the dielectric constant. When the system undergoes AFM transition, the strong internal magnetic field tends to induce a long-range electrical ordering due to the spin-phonon coupling. On the other hand, the athermal QP-fluctuations oppose this tendency; the compromise being the medium-range dipolar-organization into nano-scale clusters. In the related Na$_{4/3}$Bi$_{1/2}$Cu$_3$Ti$_4$O$_{12}$, recently Ferrarelli et al. using infrared/THz spectroscopy have reported incipient-ferroelectric/quantum-paraelectric character. The soft-mode frequency was fitted by a modified Barrett formula, to determine the relevant temperatures $T_1$ and $T_c$; however, no effect of the AFM-ordering on the dielectric behavior was reported. Their dielectric data taken at GHz range (for intrinsic part sans huge Maxwell-Wagner contributions) is similar to ours’ presented here on SCTO.

With reference to fig.5a, the magnetic correlations onset their kinetic (frequency-dependent) effect of decreasing the dielectric constant (otherwise undergoing eventual Barrett level-off) almost 10K above $T_N$. While this dynamic manifestation of the clusters is triggered by the fluctuating AFM-correlations existing above $T_N$ ($f = |\Theta_{C,W}|/T_N > 1$), their static manifestation is the continued ω-
dependence of $\varepsilon'$ below, due to the underlying quantum fluctuations (QF). Without these QF’s, the magnetic-frustration alone may cause a magneto-dielectric $\varepsilon'$-dispersion strictly above $T_N$; with its sub-$T_N$ demise (i.e., $\varepsilon'(\omega)$-merger sans QF) at all frequencies, contrary to our results as per obtained. Clear low-$T$ frequency-dispersion in the dielectric constant of a spatially-uniform (sans defects/doping) QP-parent is observed here, reflecting nano-scale electrical-segmentation dynamically (statically) above (below) $T_N$. We attribute it to the magneto-electric competition product of the internal magnetic field, coupled with the quantum-fluctuating dipole-moments, and thermal energy. It is important to note that while in the classical FE-relaxors, the static disorder inhibits the long-range electrical ordering, the dynamical disorder here is caused by the athermal quantum fluctuations. Pure SCTO QP-parent thus becomes the first to feature vitreous dispersive-response character of a kinetic phase transition; the observed state is qualified to be coined as Quantum Paraelectric Glass (QPG).

Dispersion-kinetics of dielectric constant here confirms the Vogel-Fulcher-Tammann (VFT) glassy-slowdown\textsuperscript{31-32} (fig.5a, inset) of the characteristic frequency $\omega_p(T) = \omega_0 \exp[-E_d/(T-T_0)]$, which generally describes dispersion in the relaxation frequency of FE-relaxors. Note that the Arrhenius ($\omega_p \propto T^1$) plot here also serves the purpose of a thermo-spectral ‘dynamic phase-boundary’; separating the ‘liquidus & glassy’ regimes under & above the same, of the QP degrees of freedom. The curve marks upper (lower) cut-off frequency (temperature) at a particular temperature (frequency), for the dielectric and piezoelectric response of the liquidus (unarrested) phase to be manifested, probed, and manipulated. Moreover, the observed high-$T$ classical paraelectricity (sans dipolar-correlations) marks $T_1=155K$ as the upper cut-off temperature for this liquidus regime. Definitely, an electric-field ($E_{c,k}$) should nucleate & stabilize electrically-ordered regions in this liquidus regime (i.e., electrical ‘solidification’) besides altering the phase-boundary (i.e., narrowing the $\omega_p$-dispersion). Thus in SCTO too, electro-mechanical response\textsuperscript{33} ought to be observable under bias-field, across $T_N=23K$ to $T_1=155K$ and up to excitation frequency $\omega_p(T,E)$, in the piezoresponse force microscopy (PFM). For the SCTO-QPG, VFT temperature from the fit is $T_0=13K$ and the activation energy is $E_a=25meV$.

Over the range where the dielectric constant clearly displays measurable $\omega-T$ dispersion; fig.5b shows low-magnitude loss-background $\varepsilon''(T)$ without a peak-structure. This is understandable for two reasons. Firstly, for the antiparallelly-clustered dipoles, configurations directed along or opposite to an applied $E$-field are energetically equivalent. Therefore, the two local minima of their configurational potential-energy are degenerate; symmetric double-well rendering ‘relaxations’ mute for the non-polar nano-clusters, under the removal/flipping of the applied field. Secondly, at least above $T_N$, the AFM-correlations induce only dynamic/transient non-polar clusters, whose relaxation is meaningless. As such, $\omega_p(T)$ obtained from the dispersed $\varepsilon'(\omega,T)$-peaks here signifies characteristic response-frequency of these non-polar nano-clusters. In contrast, the same obtained for FE-relaxors (from either $\varepsilon'(\omega,T)$- or $\varepsilon''(\omega,T)$-peaks) refers to the relaxation-frequency of their polar nano-regions (PNR’s). Therefore, for applications, QPG’s seem more suitable than FE-relaxors, in that they provide a broadband (in both $\omega$ and $T$) high dielectric susceptibility, against the background of weakly-dispersive marginal-losses.
Dielectric losses ($\varepsilon''$) being negligent here (fig.5b), the QPG contrasts with the FE-relaxors, as revealed by the very distinct values for the two benchmarks used-in/describing their VFT behaviors. Generally, the ratio $T_0/T_s$ (limit 0 to 1) of ultimate $e_{ps}(T_0) \sim 0$ to ambient freezing temperatures ($T_p$ at 1kHz probing frequency, say) measures the non-Arrhenicity of dispersion-kinetics, whereas $E_d/k_B T_0$ (VFT-temperature-scaled barrier-activation energy) known as the glass-strength$^{34}$ is a metric of the resistance against devitrification of the glassy state by external means (c.f., pressure $P$ for the structural glasses and electric field $E$ for FE-relaxors). Table I compiles these metrics for a number of classical (statically-disordered) FE-relaxors, along with the same for the present (dynamically-disordered) SCTO-QPG, and those characterizing the glassy domain-wall freezing in KH$_2$PO$_4$ (KDP) crystal.$^{37}$ Apart from the qualitatively obvious$^{34}$ reverse-regression between these tabulated parameters across the types of relaxor/glassy specimens, we find that the family of FE-relaxors defines an exclusive scaling to which SCTO does not belong (fig.5b, inset). Therefore, clearly distinct anti-regressions between non-Arrhenicity and glass-strength delineate the categories of FE-relaxors and QPG. Moreover, the much-larger glass-strength for our SCTO-QPG translates into its feeble susceptibility to electrical-devitrification, which characterizes electrically-glassy FE-relaxors. Well-known electrical crystallization of the FE-relaxors under high $E_{dc}$-fields into robust ferroelectrics$^{36}$ is thus little expected for the pure SCTO, from its glassy-regime (i.e., below $\sim T_N$). In retrospect, this also explains why the internal magnetic field due to the long-range AFM-ordering too fails to induce a bulk electrical order in SCTO, expected of a non-local magneto-electric coupling, and rather settles for the nano-scale electrically-vitrified state.

Table I. A Compilation of glass-kinetics parameters in arrested electrical degrees of freedom.

| Materials$^{[Ref]}$ | Non-Arrhenicity $(T_0/T_s)$ | Glass-Strength $(E_d/k_B T_0)$ |
|----------------------|-----------------------------|--------------------------------|
| SrCu$_3$Ti$_4$O$_{12}$ (QPG)$^{[\text{present}]}$ | 0.482 | 22.1 |
| 0.22BS-0.25PMN-0.53PT$^{[38]}$ | 0.916 | 1.91 |
| 0.9PMN-0.1PT$^{[39]}$ | 0.922 | 1.63 |
| PZN$^{[40-41]}$ | 0.942 | 1.37 |
| KH$_2$PO$_4$ (Domain-Wall Freezing)$^{[37]}$ | 0.957 | 0.20 |
| 0.75PMN-0.25PT$^{[42]}$ | 0.969 | 0.51 |

A functional-interest of the QPG state is the character of its electrical quality factor, defined as the inverse loss-tangent, $Q = \cot \delta = \varepsilon' / \varepsilon''$. Large value and spectrally/thermally benevolent behavior (enabling calibrations) of this $Q$-metric is practically important and desirable for the use of a dielectric in microwave/high-frequency device components such as resonators, oscillators, phase-shifters, and mixers for narrow-band applications. A major benefit of replacing the air-filled metallic-voids/cavities etc. by a dielectric is the size-downscaling of particulate structures by the refractive index $n = \sqrt{\varepsilon}$,
crucial for miniaturization and large-scale integration. Moreover, due to the lower thermal expansion vs. metals, size-specific precision spectral parameters (e.g., operational frequency) of the device-structures incorporating the dielectrics remain sturdier against thermal variations. To this end, fig.6 shows this quality-factor spectrum for the SCTO-QPG, at key temperatures across the observed $\varepsilon'$-dispersion range. Note the rather high $\sim O(10^3)$ magnitude ($\pm 4\%$) and regular $(\omega, T)$-variation (-10dB drop over 10Hz-1MHz and +7dB increase across 20-35K) functional-features of this $Q$-factor for SCTO-QPG. Remarkable too is the positive temperature-coefficient $(dQ/dT > 0)$ of the quality factor.

To conclude, we have observed a quantum paraelectric glass (QPG) state in pure SrCu$_2$Ti$_2$O$_7$. The high-temperature QP-liquid state in competition with the AFM-order-driven phonon-softening $(A_y(1) \text{ rotation-like mode})$ is witnessed to undergo kinetic glass-phase transition near $T_N=23K$. Strong spin-phonon coupling due to the Ti$^{3+}$ cations, common to both the indirect Cu-Ti-Cu exchange and the Ti-O bond-polarizability, together with the frustrated magnetic correlations above $T_N$ results in peculiar magneto-dielectricity of this material. High-temperature antiferroelectric-like correlations intrinsic to the QP-parent exclude any polar-organization of the dipoles at low-temperatures. Essentially capacitive, low-loss magneto-dielectric response around $T_N$ features glassy Vogel-Fulcher frequency-dispersion (electrical-vitreousity), traced to non-polar nano-clusters. An interesting classification scheme sets the QPG state in SCTO distinctly apart as a strong electrical-glass-former, less susceptible to long-range electrical ordering under an $E$-field, versus the FE-relaxors defining a family of fragile glass-formers. Rather small ($\leq 5\%$) peak-anomaly in the dielectric constant near $T_N$ despite a direct (Ti-mediated) magneto-dielectric coupling is attributable to this “ideal-glass-like” and “non-polar” characters of the QPG state here; reflecting only the polarizability-change due to the electrical nano-clustering, without their polar-alignment/conglomeration (typical of the FE-relaxors). However, the nearly negligible-losses ($\tan\delta \sim 10^{-4}$) and the temperate $(\omega, T)$-dependent high-$Q$ $(=\varepsilon'/\varepsilon'')$ characteristics of the QPG state ensure even this small (magneto-dielectric) $\Delta\varepsilon'$ to be robust/integral against time and other (electrical, magnetic, mechanical etc.) disturbances, meriting QPG preferable to the “polar-base” multiferroics, for prospective applications.

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Figure Captions:

Fig.1. Schematic of the possible emergent matter-states from the interaction of spin and dipolar degrees of freedom, in the presence of their quantum fluctuations. QPG (quantum paraelectric glass), QSG (quantum spin glass), and QMG (quantum multiglass).

Fig.2. Rietveld-refined X-ray diffraction pattern of SrCu$_3$Ti$_4$O$_{12}$, along with its crystal structure in the direction (x=0→1, y=0.25→0.75, z=0.25→0.5), with tilted Ti-O$_6$ octahedra. Right sketch shows the coplanar arrangements of oxygens around the Cu, responsible for the octahedral-tilting.

Fig.3. Magnetization $M(T)$ of SrCu$_3$Ti$_4$O$_{12}$ with antiferromagnetic phase transition at $T_N$ =23K in little-different FC and ZFC runs (left y-axis). Inverse susceptibility $1/\chi$ vs. temperature (right y-axis) and the Curie-Weiss fit ($\Theta_{CW}$ = -39.1K, $\mu_{eff}$/Cu-ion =2.09$\mu_B$, also enlarged in the right-inset). Top inset compares zoomed-in $M/H$ at 100 Oe and 7T, confirming no change in $T_N$ at high-fields, indicative of rather robust exchange interaction. Symbol-sizes represent the raw-data uncertainty.

Fig.4. Left y-axis: dielectric constant of SrCu$_3$Ti$_4$O$_{12}$ vs. temperature at 800 kHz, with Barrett fit (quantum paraelectric behavior) and showing the drop near $T_N$. Right y-axis: inverse electrical-susceptibility $(\varepsilon'-1)^{-1}$ vs. temperature; Curie-Weiss straight line and the Barrett fits split below $T \approx 155$K (the quantum regime) and merge within uncertainty at higher temperatures (classical regime). Symbol-size represents the raw-data uncertainty. Inset: a normalized metric $[(\varepsilon'_B/\varepsilon'_{CW})-1]$ of the net quantum paraelectric (QP) character, rising sharply at ~155K, maximizing at ~50K, and dipping at lower-$T$'s (reflecting Barrett’s turnover to plateau-behavior).

Fig.5. (a) Glassy dispersion of the dielectric constant over a wide range (~ 6 decades) of frequency. Inset: Arrhenius-plot of probing frequency vs. inverse of $\varepsilon'$-peak temperature ($1/T_p$) fits the Vogel-Fulcher glassy slowdown with cooling. Regions under (above) the curve represent liquidus-QPL (glassy-QPG) dynamical regimes of the quantum paraelectric degrees of freedom. (b) Low-valued/featureless losses ($\varepsilon'' \sim 10^{-1}$ at $T_p$’s) signify no “polar-like” relaxations. Symbol-sizes represent the raw-data uncertainty. Inset: a ‘phase’ diagram of correlated glass-metrics (strength-against-devitrification vs. non-Arrhenicity of glassy kinetics) brings out the apartheid of QPG-SCTO presented here (open star) and the classical FE-relaxors (open dots, defining a clear family-locus), along with the data representing the domain-wall freezing (solid dot).

Fig.6. Spectral character at key temperatures of the quality factor $Q = \varepsilon'/\varepsilon''$ illustrates its regular -10dB drop over five decades in frequency and +7dB increase across 20-35K, for the SCTO-QPG. Highly-desirable positive temperature-coefficient of $Q$ is in contrast to its negative values for the metal-based structures, used in the high-frequency applications.
The graph shows the permittivity (\(\varepsilon'/800\text{kHz}\)) of a material as a function of temperature (K). The permittivity is plotted on the y-axis, with values ranging from 80 to 120. The temperature is plotted on the x-axis, ranging from 0 to 300 K.

There are two distinct regions indicated on the graph:

1. A region near \(T_1 \approx 155\text{K}\) where the permittivity decreases sharply as the temperature increases.
2. A region where the permittivity exhibits a Curie-Weiss behavior, indicated by the linear curve with the formula \(\theta_{\text{CW}} = -419\text{K}\). The Curie-Weiss law is represented by the blue dotted line.

Additionally, there is an inset graph in the top right corner showing the ratio of the permittivity at a particular frequency to the permittivity at a reference frequency, plotted against temperature. The inset graph uses a logarithmic scale for the y-axis, ranging from \(10^0\) to \(10^{-2}\).
