Thermal expansion of the heavy-fermion superconductor $PuCoGa_5$

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We have performed high-resolution powder x-ray-diffraction measurements on a sample of $^{242}PuCoGa_5$, the heavy-fermion superconductor with the highest critical temperature of $T_c = 18.7$ K. The results show that the tetragonal symmetry of its crystallographic lattice is preserved down to 2 K. Marginal evidence is obtained for an anomalous behavior below T_c of the a and c lattice parameters. The observed thermal expansion is isotropic down to 150 K and becomes anisotropic for lower temperatures. This gives a c/a ratio that decreases with increasing temperature to become almost constant above ~ 150 K. The volume thermal expansion coefficient α_V has a jump at T_c, a factor ~ 20 larger than the change predicted by the Ehrenfest relation for a second-order phase transition. The volume expansion deviates from the curve expected for the conventional anharmonic behavior described by a simple Grüneisen-Einstein model. The observed differences are about ten times larger than the statistical error bars but are too small to be taken as an indication for the proximity of the system to a valence instability that is avoided by the superconducting state.

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I. INTRODUCTION

$PuCoGa_5$ has the highest T_c (18.7 K) of any heavy-fermion superconductor. Fifteen years after its discovery [1] our understanding of much of this material remains at best confused [2–8]. We do know from nuclear magnetic resonance (NMR) [2] and point-contact spectroscopy [6] measurements that the superconducting state has d-wave symmetry. Magnetic form-factor measurements with polarized neutron diffraction [5] have shown that the ground state is not the conventional $5f^5$ state found in many plutonium (Pu) intermetallics. Neutron inelastic scattering has failed to detect any sign of a resonance as found, for example, in the isostructural $CeCoIn_5$ compound [9], which also has d-wave symmetry, although the difficulty of performing these neutron experiments on Pu should not be overlooked. Recent theoretical efforts [10–12] have concluded that the driving mechanism for superconductivity is valence fluctuations. Electronic structure calculations combining the local-density approximation with an exact diagonalization of the Anderson impurity model [13] show an intermediate $5f^5$-$5f^6$-valence ground state and delocalization of the $5f^5$ multiplet of the Pu atom $5f$ shell. The $5f$ local magnetic moment is compensated by a moment formed in the surrounding cloud of conduction electrons, leading to a singlet Anderson impurity ground state.

The presence of valence fluctuations has recently been suggested by resonant ultrasound spectroscopy measurements, showing that the three compressional elastic moduli exhibit anomalous softening upon cooling, which is truncated at the superconducting transition [8]. These results have been interpreted as evidence for a valence transition at a $T_V < T_c$ that is avoided by the superconducting state, suggesting that $PuCoGa_5$ is near a critical end point involved in the unconventional superconductivity [8]. However, the identification of the fluctuating order parameter responsible for the observed anomalous softening requires information on the thermal expansion of the lattice, which is not available. Moreover, crystallographic studies at low temperatures have not been published, and it is important to verify that no lattice distortion occurs at or above T_c. These are the issues that we have addressed by performing high-resolution x-ray diffraction measurements in the T range between 2 and 300 K on a defect-free polycrystalline sample of $^{242}PuCoGa_5$. The results of our investigation show that no measurable structural distortion is associated with the stabilization of the superconducting phase. The thermal expansion is isotropic down to 150 K and anisotropic for lower temperatures, which is not surprising for a superconductor with an order parameter of d-wave symmetry. The T dependence of both a and c lattice parameters shows small anomalies at T_c and a behavior that deviates from the one expected by the simple quasiharmonic approximation commonly used to describe the thermal expansion in solids. However, no convincing evidence is found for an incipient valence transition of the Pu electronic configuration associated with the formation and condensation of Cooper pairs.

II. EXPERIMENTAL DETAILS AND RESULTS

The experiment was performed at the ID22 beamline of the European Synchrotron Radiation Facility (ESRF) in France. Data have been collected on a sample obtained by crushing a single crystal grown at the Karlsruhe establishment of the Joint Research Centre in a Ga flux using the ^{242}Pu isotope (99.932 wt% ^{242}Pu, 0.035 wt% ^{241}Pu, 0.022 wt% ^{240}Pu, 0.005 wt% ^{239}Pu, 0.004 wt% ^{238}Pu, and 0.002 wt% ^{244}Pu in December 2015) to avoid effects from radiation damage and self-heating. The total sample mass was 4.6 mg, corresponding to a plutonium mass of 1.7 mg and a total activity of ~ 760 kBq. Magnetic susceptibility and specific heat measurements show superconductivity below $T_c = 18.7$ K (see the

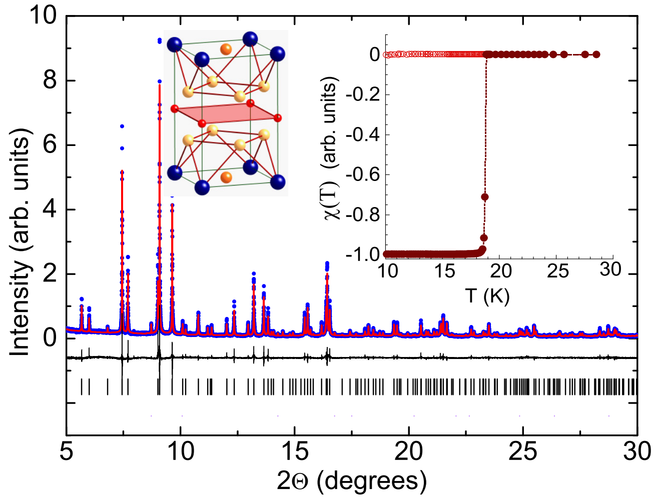

FIG. 1. X-ray-diffraction pattern recorded for $PuCoGa_5$ at 5 K; the blue points are the experimental data, the red line is the refined Rietveld profile ($R_{wp} = 8.77\%$, $R_{wp}/R_{exp} = 1.2$). The residual is given by the black line at the bottom; vertical ticks represent the angular position of Bragg peaks. No impurity phases were detected. The left inset: crystallographic unit cell (space-group $P4/mmm$, No. 123). Pu atoms (blue spheres) occupy the $1a$ Wyckoff position, Co atoms (red spheres) are located at the $1b$ position, halfway between the Pu atoms along the c direction, and the Ga atoms sit on two crystallographic positions, one (orange spheres) in the center of the basal planes ($1c$) and the other ($4i$) in the rectangular faces of the unit cell (yellow spheres) with the position (0, 1/2, z). The refined value of the z parameter at 5 K is $z = 0.3075(2)$. The right inset: temperature dependence of the magnetic susceptibility measured under zero-field cooling (closed brown circles) and field cooling conditions (open red circles, applied field of 10^{-3} T), providing a T_c of 18.7 K.

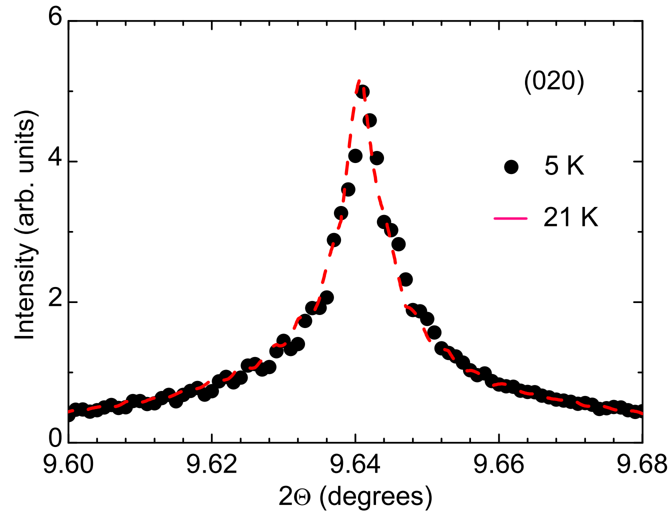

FIG. 2. The (020) Bragg peak measured at 5 K (black circles) and 21 K (red dashed line). Neither splitting nor broadening of the line shape is observable, indicating that the tetragonal symmetry is preserved in the superconducting phase.

inset of Fig. 1). Following a protocol developed for powder-diffraction measurements at synchrotron radiation sources on other transuranium isotopes [14], the sample was put inside a hermetic holder providing four levels of containment. For this, we used a kapton capillary (1-mm diameter, ∼25 mm in length) half filled with Stycast (2850FT). The resin was allowed to cure before a 5-mm mixture of a second resin (Epofix), and the sample was inserted with a pipette. The Epofix was used because of the lower viscosity, allowing easier mixing with the powder sample and insertion into the narrow kapton capillary. The remainder of the capillary then was filled with Stycast and, once fully cured, it was inserted into a drilled-out plexiglass rod, which was sealed with a plexiglass plug, glued with further Stycast, and finally enveloped within a 4-mm polyimide tube. Due to the contamination risk generated by the plutonium element, all operations of preparation and encapsulation have been carried out in shielded gloveboxes under an inert nitrogen atmosphere following well-established safety procedures.

The channel-cut Si-111 monochromator of ID22 provided an incident beam wavelength of 0.354 155 Å. The sample capillary was mounted on the axis of the diffractometer inside a liquid-helium-cooled cryostat allowing reaching a base temperature of 2 K. To avoid any risks of mechanical failure of the containment, the sample was not spun within the cryostat. This did not result in preferred sample orientation issues in the data as the setting process in the resin eliminates any preferred orientation and provides a good sample average. A NIST 640c Si standard was used to calibrate the Si-111 multianalyzer stage. In the first part of the experiment, the diffraction pattern was measured at several temperatures from 2 to 300 K with acquisition times up to 4 h. The T dependence of the lattice parameters was obtained from the Rietveld refinement of diffraction patterns collected on warming from 5 K up to 260 K with a counting time of 1 h at each temperature. The experimental resolution was on the order of $\Delta d/d = 10^{-6}$. The main results are summarized below.

The best fit of the diffraction profile is obtained within the tetragonal $P4/mmm$ space group in the whole temperature range explored in this experiment. Close examination of the shape and width of individual Bragg peaks shows no evidence for the occurrence of a lattice distortion across T_c as shown for the (020) Bragg peak in Fig. 2.

The temperature dependence of the lattice parameters a and c is shown in Fig. 3 together with the thermal expansion of the unit-cell volume $V(T)$. The error bar on the experimental data represent the error σ_R estimated from the Rietveld refinement multiplied by a factor of 5. The solid line is a fit to a simple one-phonon Grüneisen-Einstein model,

$$\ln\left(\frac{V(T)}{V_0}\right) = \frac{k_B n}{B V_m} \frac{\gamma T_E}{\exp(T_E/T) - 1}, \quad (1)$$

and equivalent expressions for the lattice parameters $a(T)$ and $c(T)$. In Eq. (1), V_0 is the unit-cell volume at $T = 0$, k_B is the Boltzmann constant, $n = 7$ is the number of atoms per unit cell, $B \simeq 89$–100 GPa is the bulk modulus [8,15,16], $V_m = 7.23\times10^{-5}\ \mathrm{m^3/mol}$ is the molar volume, γ is the Grüneisen parameter, and T_E is the Einstein temperature. The best fit is obtained for $V_0 = 120.090(3)$ Å^3 [$c_0 = 6.7607(4)$, $a_0 = 4.2146(3)$ Å], $\gamma = 5.2(4)$, and $T_E = 197(4)$ K. The values estimated by this simple model are in line with those obtained by self-consistent calculations reported in Ref. [17]. Moreover,

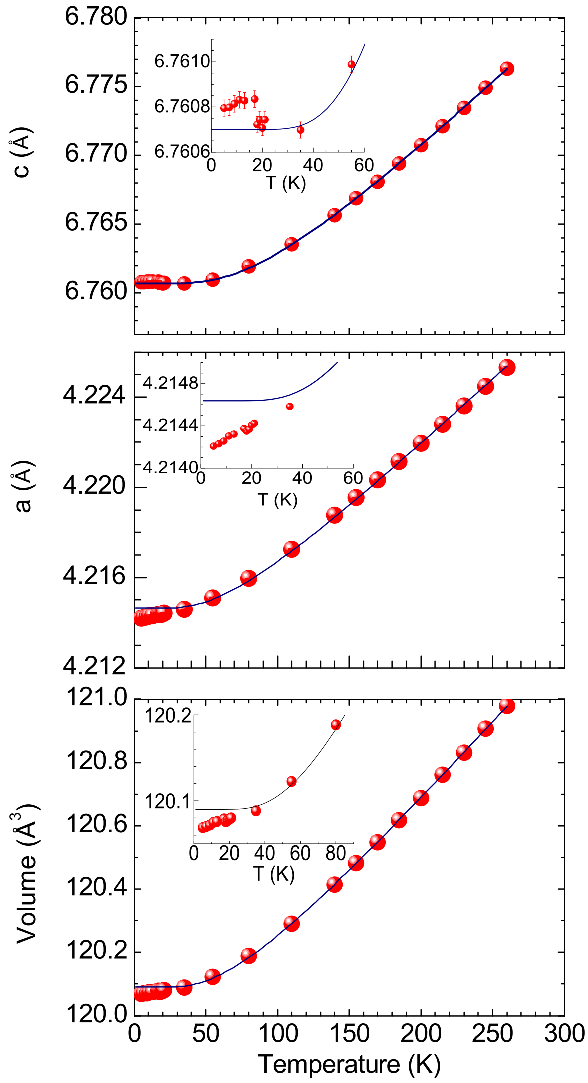

FIG. 3. Thermal expansion of the $PuCoGa_5$ lattice parameters and unit-cell volume. The solid lines represent a fit to a Grüneisen-Einstein model as explained in the text. The vertical bars give the statistical error multiplied by a factor of 5. If not visible, the error bars are smaller than the symbol sizes.

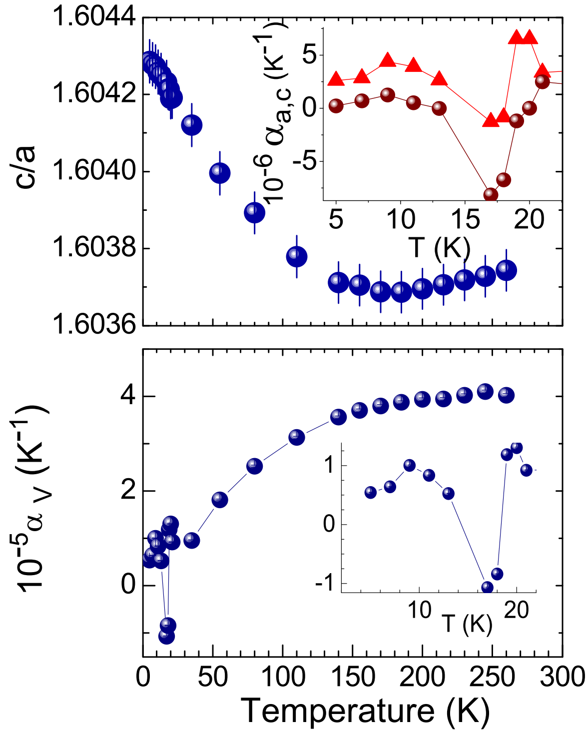

FIG. 4. Top panel: temperature dependence of the c/a ratio for $PuCoGa_5$. The inset shows the temperature dependence of the linear thermal expansion coefficients along the a (triangles) and c (circles) crystallographic directions. Bottom panel: Thermal expansion coefficient for the unit-cell volume (in an expanded scale around T_c in the inset). Error bars are estimated as five times the statistical error provided by the Rietveld refinement and are smaller than the symbol size if not shown. The solid lines are guides to the eye.

the Grüneisen parameter has the order of magnitude reported for other mixed-valent Ce and U compounds, for instance, $CePd_3$, $CeSn_3$, and UAl_2 [18].

Although the simple model above describes well the behavior at high temperatures, clear deviations from the predicted dependence are observed below T_c. Whereas a decreases linearly with decreasing T, c has a small expansion at T_c and becomes constant at lower temperatures, a behavior similar to the one calculated by Millis and Rabe for $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_7$ by taking into account Gaussian fluctuation corrections for the mean field superconducting free energy [19]. As a consequence, the volume expansion deviates from the curve expected for the conventional anharmonic behavior described by the Grüneisen-Einstein model with differences that are about two times larger than the error bars given by $5\sigma_R$. One must, of course, be aware that Eq. (1) has its roots in the Einstein approximation for the specific heat of a solid, which underestimates the contribution of long-wavelength vibrational modes at very low temperatures. However, as significant differences with the curve predicted by more sophisticated models are expected at temperatures much lower than the T_c in $PuCoGa_5$, the use of Eq. (1) to signal an anomaly in the observed experimental data at the onset of superconductivity is, in the present case, justified.

As shown in Fig. 4 (top panel), upon cooling, the expansion is isotropic down to 150 K and anisotropic for lower temperatures. This results in a c/a ratio that decreases with increasing T to become almost constant above $\sim$150 K. It is interesting to note that the marked increase in the c/a ratio below 150 K occurs in the temperature range where Ramshaw *et al.* [8] observe an anomalous softening of the bulk modulus and a significant temperature dependence of the in-plane Poisson ratio. Such a behavior was attributed in Ref. [8] to the development of in-plane hybridization between Pu $5f$ moments and conduction electrons.

The inset (top panel) of Fig. 4 shows the linear thermal expansion coefficients along the a and c directions around T_c. The temperature dependence of the volume thermal expansion coefficient α_V is shown in the bottom panel of Fig. 4. The presence of an anomaly with a minimum at T_c has been confirmed by repeating the sequence of measurements both on warming and on cooling cycles.

III. DISCUSSION

The anisotropic change in thermal expansion at T_c is not unexpected for a d-wave superconductor adjusting its crystal structure in order to minimize the lattice free energy. On the other hand, the observed deviation of the unit-cell volume with respect to the Grüneisen-Einstein prediction is much larger than the one obtained from the first Ehrenfest equation for second-order phase changes. Such an equation relates the difference between the temperature derivative of the volume at constant pressure calculated above and below the phase transition with the jump of the specific heat at T_c and the initial slope of the hydrostatic pressure dependence of T_c (which measures the average of the stress derivatives),

$$\left(\frac{\partial V_s}{\partial T}\right)_p - \left(\frac{\partial V_n}{\partial T}\right)_p = \Delta\alpha_V V_m = \frac{\partial T_c}{\partial p}\frac{C_s - C_n}{T_c}, \tag{2}$$

where $\Delta\alpha_V = \alpha_{Vs} - \alpha_{Vn}$ is the difference between the thermal expansion coefficients in the superconducting and normal phases. Previous studies have reported $\partial T_c/\partial p = 0.4(2)\times 10^{-9}$ K/Pa [20] and $(C_s - C_n)/T_c = 0.110(4)\,\mathrm{J\,mol^{-1}\,K^{-2}}$ [1], leading to $\Delta\alpha_V = 0.6\times 10^{-6}\,\mathrm{K^{-1}}$. This value for the thermal expansion discontinuity is comparable with those calculated for $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_7$ in Ref. [19], but it is smaller by one order of magnitude than the anomaly observed in the experimental curve shown in Fig. 4. Moreover, the positive jump at T_c of α_V (with increasing T), in conjunction with the negative jump of the specific heat, would indicate an initial negative value for $\partial T_c/\partial p$, in contrast to the direct measurement.

Such discrepancies in sign or magnitude have been observed in other superconductors, such as, for example, the A15 material V_3Si [21], the Chevrel phase $PbMo_6S_8$ [21], or the iron-based layered superconductor $Ba(Fe_{1-x}Co_x)_2As_2$ [22], where the thermal expansion is also highly anisotropic and the derivative $\partial T_c/\partial p$ deduced from the Ehrenfest relation is negative, whereas the pressure diagram is clearly displaying an increase in T_c with increasing pressure around $p \sim 0$ GPa. Several hypotheses have been invoked to account for these deviations, but no clear explanation has emerged yet, so we refer interested readers to references therein. One should also notice that $PuCoGa_5$ is a plutonium-based material and the Pu element is already at the origin of numerous exotic phenomena, such as a negative thermal dilatation in the δ phase [23] due to its specific electronic structure that is far from being fully understood.

Although we do not have any straightforward explanation for the departure from the predictions of the Ehrenfest relation, we think that this is an interesting finding that calls for further studies. Definitely, determining the thermal expansion of $PuCoGa_5$ single crystals along the main directions with a high-sensitivity technique, such as dilatometry, would be valuable to yield more details on this anomaly at T_c and stimulate theoretical work.

Thermal expansion measurements have been reported for the isostructural heavy-fermion superconductor $CeCoIn_5$ ($T_c = 2.3$ K) [24]. Also in that case, the thermal expansion shrinks for the [100] direction in the superconducting state, whereas it expands for [001]. The volume thermal expansion decreases linearly down to T_c with decreasing temperature and more rapidly so in the temperature range from T_c down to 1.5 K [24]. This is similar to what we report for $PuCoGa_5$, but for $CeCoIn_5$ the coefficient of the volume thermal expansion shows an anomaly with a λ shape, which is not the case for $PuCoGa_5$. Moreover, applying the Ehrenfest relation to $CeCoIn_5$ leads to a correct estimate for $\partial T_c/\partial p$ (both in sign and in magnitude), again in contrast with what we report for the Pu analog.

The linear decrease in the unit-cell volume with decreasing temperature below T_c is qualitatively similar to the one observed in $CeRu_2Si_2$, a compound where the Kondo screening (changing the $4f^1$ localized state to a nonmagnetic $4f$-itinerant state) is accompanied by a volume contraction below the Kondo temperature of $T_K = 20$ K [25,26]. In that case, the phenomenon can be interpreted in the framework of a theory describing critical valence fluctuations involving $4f^1$ and $4f^0$ electronic configurations [27], although $CeRu_2Si_2$ is thought to be relatively far from criticality [28]. According to Ref. [27], the variation of the f-shell occupation number $\Delta n_f = n_f(T) - n_f(0)$ is proportional to the volume change $\Delta V(T)$, $\Delta n_f \propto \zeta\chi_0\Delta V/(\eta_0 + bT^{\xi})$, where ζ is a temperature-independent constant that relates the energy variation of the f-electron levels to the volume change, χ_0 is the noninteracting susceptibility of the order of the quasiparticle density of states, η_0 is a parameter characterizing the degree of departure from the critical point, and ξ is a critical exponent.

Although a theory for valence transitions between electronic configurations with occupation numbers higher than 1 is not yet available, the behavior should be qualitatively similar, and the observed volume shrinkage could be an indication that the valence of the Pu atom changes below T_c. However, it must be emphasized that the observed departure of the volume from the Grüneisen-Einstein model is on the order of 10^{-4} and, in the absence of a quantitative theory, we cannot claim that $PuCoGa_5$ is at the verge of a critical valence transition on the basis of our results.

Our attempts to separate electronic and vibrational contributions from the observed thermal expansion failed. In $PuCoGa_5$, T_c is relatively high, whereas its linear specific heat capacity is relatively small compared to other heavy-fermion materials. As a consequence, the phonon contribution to the thermal expansion cannot be considered as a small correction as in many heavy-fermion superconductors where the critical temperature is in the subkelvin range and the specific heat Sommerfeld coefficient γ is very high. Therefore, in the absence of a precise estimate of the vibrational term, a reliable separation of the different contributions to the thermal expansion was not feasible.

IV. CONCLUSIONS

X-ray-diffraction measurements with a resolution of $\Delta d/d \sim 10^{-6}$ in the lattice spacing show that the tetragonal symmetry exhibited by the $PuCoGa_5$ unconventional superconductor is preserved down to 2 K, well below the critical temperature of $T_c = 18.7$ K. The lattice thermal expansion is isotropic down to 150 K and anisotropic for lower temperatures. This gives a c/a ratio that decreases with increasing T to become almost constant above ~ 150 K. The volume thermal expansion coefficient α_V has a jump at

T_c, a factor of $\sim$20 larger than the change predicted by the Ehrenfest relation. At low temperatures, the expansion of the unit-cell volume deviates from the curve corresponding to a simple one-phonon Grüneisen-Einstein model and shows, below T_c, a continuous linear shrinking of the volume. In the case of the $CeRu_2Si_2$ Kondo system, a similar trend has been attributed to critical valence fluctuations. Although the deviations observed for $PuCoGa_5$ are about ten times larger than the statistical errors, in the absence of a quantitative theory, it is not possible to establish the occurrence of critical valence fluctuations near T_c. The determination of thermal expansion along the main directions in $PuCoGa_5$ single crystals with a technique affording higher sensitivity and a higher density of experimental points, such as dilatometry, would be welcome to study more precisely this anomaly at T_c, confirm and refine our observations, and stimulate theoretical works.

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