Physics of p-wave spin-triplet pairing with the experimental examples of Strontium Ruthenate

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Abstract

This paper deals with the unconventional superconducting properties of Sr_2RuO_4 , the best-known examples of p-wave superconductor with spin-triplet pairing. I shall first review the definition and rough classification of unconventional superconductivity. The formalism for p-wave spin-triplet pairing is examined. This will lead into the discussion of the most likely p-wave state for Sr_2RuO_4 , its additional symmetry-breaking quality – namely breaking of time-reversal symmetry, and the corresponding state in He³. The experimental evidences for both spin-triplet pairing and time-reversal symmetry breaking in Sr_2RuO_4 shall be presented next. Then I shall discuss two major controversies in Sr_2RuO_4 physics – the existence and location of gap nodes or deep minima and the possibilities of multiple superconducting phases.

I. INTRODUCTION

Since 1980's superconductor that cannot be explained by classical Bardeen-Cooper-Schrieffer (BCS) theory have attracted great amount of attention. The lion's share of this have gone to d-wave high cuprate superconductor, of course; however, other types of superconductor like strontium ruthenate (Sr_2RuO_4) and heavy-fermion have also proven to be fertile subject for both theoretical and experimental research.

In particular, Sr_2RuO_4 physics have attracted attention because of its widely believed similarity with that of He³, which means this may be the condensed matter system where some of the complex characteristic of He³ can be observed. Some of the recent theoretical researches on He³, such as half-quantum vortex physics with its potential application to quantum computing, are expected to be applicable to Sr_2RuO_4 as well. But this parallel with He³ is not the whole story; due at least partially to its crystal structure Sr_2RuO_4 also has interesting physics such as gap node (or deep minima) structure and multiple superconducting phase that are quite distinct from He³. This paper shall examine both aspects of Sr_2RuO_4 physics.

II. BASIC THEORETICAL IDEAS

A. Unconventional superconductivity

The onset of superconductivity occurs with the condensation of electron pairs. These electron pairs, called the Cooper pair, can be in a state of either total spin S=0 (spin singlet) or 1 (spin triplet). Being fermions, electrons anticommute. Therefore the antisymmetric spin-singlet state is accompanied by a symmetric orbital wave function (even parity) and vice versa, in order to preserve the antisymmetry of the total wave function. Therefore we get L=0 (s wave), 2 (d wave), etc for spin-singlet state and L=1 (p-wave), 3 (f-wave), etc for spin-triplet state.

Conventional superconductivity is characterized by s-wave Cooper pairs. The binding state of the Cooper pair tells us what are the symmetry-breaking properties of the condensate. The order parameter of superconductivity is in general represented by the gap function $\Delta(\mathbf{k})$. For an s-wave superconductor the phase of $\Delta(\mathbf{k})$ is constant irrespective of the direction of \mathbf{k} , although there may be some \mathbf{k} -dependent anisotropy in the magnitude of Δ . This reflects the fact that the condensate breaks only gauge symmetry at the transition into the superconducting state.

However the unconventional superconductivity is defined by the relation $\Sigma_{\mathbf{k}} \Delta(\mathbf{k}) = 0$ (where the summation is over the Fermi surface) – which is possible only with **k**-dependent change in the phase of the gap function, as in $\Delta(\mathbf{k}) = \Delta_d(k_x^2 - k_y^2)$ for the d-wave cuprate superconductor. This also requires additional symmetry breaking at the transition into the superconducting state. [1]

This definition of the unconventional superconductivity has a serious consequence. One of the most important features of an s-wave superconductor is its insensitivity to random scattering from disorder. However, in unconventional superconductor, this is no longer the case, as the order parameter can be averaged to zero by sufficiently strong scattering around the Fermi surface. Roughly speaking, the criterion for 'sufficiently strong' is that the elastic mean free path equals the superconducting coherence length. This implies that superconductivity can be observed only for materials with short coherence lengths. This is the main reason why this class of superconductor took such long time to observe; it is the pragmatic reason for the term 'unconventional'.

B. Spin-triplet pairing

The complete pair wave function for S=1 spin can be written as:

$$\varphi = \varphi_{++}(\mathbf{r_1} - \mathbf{r_2}) \left| + + \right\rangle + \varphi_{--}(\mathbf{r_1} - \mathbf{r_2}) \left| - - \right\rangle + \varphi_0(\mathbf{r_1} - \mathbf{r_2}) \left(\left| + - \right\rangle + \left| - + \right\rangle \right)$$
(2.1)

Two cases can be treated almost as simply as the case of spin-singlet pairing. One is $\varphi_0=0$ case, commonly called "equal-spin-pairing" or ESP state. (It's possible to transform the pair wave function in a given region in space ESP. However, in general such transformation would not make the wave function ESP everywhere.) Another case, $\varphi_{++}=\varphi_{--}=0$ looks like the opposite of ESP but is actually belongs to an ESP state – by spin rotation we can obtain $\varphi_{++}=\varphi_{--}\exp(i\chi)$, where χ is a constant, and eliminate φ_0 . In the former case one can view it as two separate pairing – spin-up pairs and spin-down pairs – related, however, by the constant relative phase of two gap functions. In the latter case, however, one can merely view it as BCS with only spatial part antisymmetrized and odd part of the interaction potential contributing.

The above examination of ESP state suggests us a way to generalize the gap function. (Note that just as in BCS, the gap function is the pair wave function suitably normalized.) We may define the gap matrix:

$$\hat{\Delta} \equiv \begin{pmatrix} \Delta_{++} & \Delta_0 \\ \Delta_0 & \Delta_{--} \end{pmatrix}$$
(2.2)

We can use (3.2) to generalize the BCS excitation energy accordingly:

$$\hat{E}(\mathbf{k}) = (\varepsilon^{2}(\mathbf{k}) + \hat{\Delta}(\mathbf{k})\hat{\Delta}^{*T}(\mathbf{k}))^{1/2}$$
(2.3)

(T means transpose.) [2] The eigenvalues of (3.3) would give us the value we would expect from the BCS theory. One thing deserves mentioning – for ESP case, one can have unequal gaps for ++ and – pairing, which would lead to two values for excitation energy. Generalizing this to non-ESP case, the opposite of the ESP case just mentioned would be $\hat{\Delta}(\mathbf{k})\hat{\Delta}^{*T}(\mathbf{k}) = |\mathbf{c}(\mathbf{k})|^2\hat{1}$. In this case, one can show that if one sets $\hat{\Delta}(\mathbf{k})$ in a

spin basis where $\Delta_0(\mathbf{k})=0$, one would end up with $|\Delta_{++}(\mathbf{k})| = |\Delta_{--}(\mathbf{k})|$. Such states are called unitary states. Since this type of states in some sense keeps symmetry between ++ pairing and -- pairing, it would be energetically favored unless there exists some extra symmetry breaking mechanism.

As will be shown in the next section, transforming the spin basis is often helpful in gaining physical insight into a particular state. The gap matrix is inconvenient, however, when it comes to basis transformation. Things would be easier if we can express components of gap function in terms of vector components. This is done by d-vector formalism defined below:

$$\hat{\Delta} \equiv \begin{pmatrix} \Delta_{++} & \Delta_0 \\ \Delta_0 & \Delta_{--} \end{pmatrix} = \begin{pmatrix} -d_x + id_y & d_z \\ d_z & d_x + id_y \end{pmatrix}$$
(2.4)

In unitary states, we get $\hat{\Delta}(\mathbf{k})\hat{\Delta}^{*T}(\mathbf{k}) = \mathbf{d}(\mathbf{k})\cdot\mathbf{d}^{*}(\mathbf{k})\hat{1}$. Also $\mathbf{d}(\mathbf{k})$ has immediate physical meaning. Its direction defines the normal to the plane in which the electrons paired at $(\mathbf{k}, -\mathbf{k})$ are equal spin paired $(|++\rangle)$ and $|--\rangle$ relative to any quantization axis in that plane), and its magnitude is proportional to that of the energy gap at $(\mathbf{k}, -\mathbf{k})$. [1]

C. Sr₂RuO₄ and Time Reversal Symmetry Breaking



Fig. 1: Crystal structure of Sr₂RuO₄ [1]

Before discussing the p-wave state of the most famous example of Sr₂RuO₄, which is the most famous example of pwave superconductor, it would be useful to go over the essential characteristic of its normal state. The crystal structure of Sr₂RuO₄ is almost identical to the tetragonal crystal structure of the cuprate superconductor, and like the cuprate has two-dimensional strongly electronic structure. However electrons are not confined to planes - rather the Fermi surface consists of three weakly corrugated cylindrical sheets extending c-axis. This two-dimensional in characteristic of the electronic structure

fixes the possible direction of the orbital angular momentum, which actually makes Sr_2RuO_4 simpler than that most famous example of spin-triplet pairing – He³. (Actually it is possible for the crystal lattice to complicate things by lattice effects and spin-orbit coupling; however, it is widely believed that this is not the case with Sr_2RuO_4 . Also it is believed that unitary pairing states are still favored.) One would not have gap function without linear dependence on either k_x or k_y so we get $L_z = 1$ with the z-direction fixed to be parallel to the c-axis of the crystal.

The easiest guess on the pairing symmetry Sr₂RuO₄ of would be to guess that it would be one of the two possible states of He³, the one corresponding to B-phase (with $\mathbf{d}=\Delta(\hat{x} k_x + \hat{y} k_y)$) and the other to A-phase (with $\mathbf{d}=\Delta \hat{z} (k_x \pm ik_y)$). The explicit forms for the state vector of these two states are

$$\left|\psi\right\rangle_{A} = \frac{1}{\sqrt{2}}e^{i\phi}(\left|\hat{z};+-\right\rangle + \left|\hat{z};-+\right\rangle) = \frac{1}{\sqrt{2}}e^{i\phi}(\left|\hat{\alpha};++\right\rangle - \left|\hat{\alpha};--\right\rangle)$$
(2.5a)

$$\left|\psi\right\rangle_{B} = \frac{1}{\sqrt{2}} \left(e^{-i\phi} \left|\hat{z}; ++\right\rangle + e^{i\phi} \left|\hat{z}; --\right\rangle\right)$$
(2.5b)

where $\hat{\alpha} = (\cos \gamma, \sin \gamma, 0)$ is a unit vector in xy plane, and φ is the azimuthal angle in xy plane.

The equations (2.5a) and (2.5b) illustrate the very different characteristics of the two states. The B state has the spin angular momentum direction fixed in the opposite direction to the orbital angular momentum direction, so $J_z = 0$. In the A state, however, $J_z=1$ as $S_z = 0$. This non-zero total angular momentum implies the broken time-reversal symmetry. In fact it is the only allowed unitary p-wave states on a cylindrical Fermi surface on the tetragonal crystal to have broken time-reversal symmetry. [1] Also this pairing does not have any vertical line gap node.

Another difference between $\mathbf{d}=\Delta(\hat{\mathbf{x}} k_x + \hat{\mathbf{y}} k_y)$ and $\mathbf{d}=\Delta \hat{z} (k_x \pm ik_y)$ pairing deserves special mention. The former is a irreducible representation of the tetragonal symmetry group; however in case of the latter, in order to construct a state with time-reversal symmetry breaking, two different irreducible representations of the tetragonal symmetry, $\hat{z} k_x$ and $\hat{z} k_y$ has been combined. In general one requires one order parameter for one irreducible symmetry, [3] leading to the conclusion that two component order parameter is required to describe the A state while only one component is required for the B state.

In He³, except under pressure near the liquid-solid phase boundary the B state is energetically favored. This is partly because in the spherical Fermi surface of He³, a quick glance at the pairing $\mathbf{d}=\Delta \hat{z}$ ($k_x \pm ik_y$) shows that the A state requires two gap nodes at the north and south poles. However these nodes are not required in Sr₂RuO₄ due to its two-dimensional electronic structure. One cannot really make a prediction but to check experimentally whether the superconducting state has broken time-reversal symmetry. [4]

III. EXPERIMENTAL CONFIRMATION

So the theoretical consideration alone does not give conclusion on the pairing symmetry of Sr₂RuO₄. However, at present Sr₂RuO₄ is widely believed to have $\mathbf{d}=\Delta \hat{z} (k_x \pm ik_y)$ pairing. This is due to wide varieties of experimental result presented in this section.

A. Evidence for Triplet-pairing

A spin-singlet Cooper pair can be regarded as being spinless – it cannot have any spin susceptibility. Therefore an observation of temperature-independent spin susceptibility deep into the superconducting state can be consistent only with the existence of triplet pairing. The difficulty of measuring in superconductor the spin susceptibility χ_s (as opposed to neutral superfluids) is the Meissner effect. Even in the type-II superconductor for which strong field penetration can be achieved, the static susceptibility is dominated by the diamagnetism of the screening currents.

One way of overcoming this problem is by using the NMR Knight shift to measure the spin susceptibility. The Knight shift is the difference between the NMR frequency of a nucleus when it is in a metal or a superconductor rather than an insulator. It has an orbital part (K_{orb}) due to diamagnetism of bound and free electrons and a spin part (K_{spin}) due to the Pauli paramagnetism of the conduction electrons:

 $\omega = \gamma_{gyr}B_{int}(1 + K_{orb} + K_{spin})$ where ω is the NMR frequency, γ_{gyr} is the gyromagnetic ratio of the nucleus being studied and B_{int} is the average magnetic field in the sample. [1] What gets plotted as the result of the Knight shift experiment is of course K_{spin} , which would be non-zero only



Fig.2: Comparison of Knight shift [5]

for unpaired or spin-triplet paired electrons.

Fig.2 shows the first NMR test done by Ishida and his co-workers. [5] Unlike YBCO which has dwave (and therefore spin-singlet) pairing, shows finite spin susceptibility even at zero temperature where all electrons would be paired. That would be possible only for spin-triplet pairing.

Another way of measuring the spin susceptibility into the superconducting state is polarized neutron scattering. The idea is that in a magnetized material, neutron scattering occurs with Fourier components at reciprocal-lattice vectors because of both the periodicity of the nuclear position and the microscopic periodicity of the magnetization density. The two scattered waves interfere, and as described by Duffy *et al.*, the magnetic scattering can be isolated by measuring the flipping ratio R. This is defined as the ration of scattering cross sections for initial neutron states that are parallel or antiparallel to the applied magnetic field, and with an arbitrary final spin state. For a small induced moment

 $R = 1 + A[M(\kappa)/F(\kappa)]$

where κ is the scattering vector, $M(\kappa)$ is the component of magnetization parallel to the applied field, $F(\kappa)$ is the nuclear structure factor and $A = 1.16 \times 10^9$ J T m. [1]



Fig. 3: Polarized neutron scattering data for spin-singlet (a)V₃Si and (b) Sr₂RuO₄ [6]

The data of Duffy *et al.* [6] are summarized in Fig. 3. The control measurement on V_3Si shows the expected magnitude of change to the susceptibility due to singlet pairing. In contrast there is no resolvable change in the measured susceptibility in case of Sr_2RuO_4 at the transition to superconducting state, when the magnetic field is parallel to the ab-plane of the crystal.

B. Evidence for Time-Reversal Symmetry Breaking

When time-reversal symmetry breaks down electron pairs break down, electrons have definite angular momentum to a certain direction (in case of Sr_2RuO_4 , it can only be c-axis) and hence, a magnetic moment. However like in the case of spin susceptibility the Meissner effect screens this out on bulk level. It requires a sensitive local probe to observe this spontaneous magnetic field.



Fig.4: μ SR in different polarization direction (P_{μ}) [7]

One such probe is muon spin relaxation (μSR) . In μSR , fully spin-polarized positive muons are incedent on a specimen at a sufficiently low flux that they arrive individually on the scale of their decay time (~2.2 μ s). They come to rest very quickly, and their spins react to the local magnetic environment at the implantation site. When they decay, a positron is emitted in a direction that correlates with the spin direction of the muon at the time of decay. By studying many such positrons, one can deduce the muon polarization function as a function of time after implantation. The form of this function yields considerable information about the local magnetic-field distribution in the solid.

The first study of μ SR in Sr₂RuO₄ was reported by Luke and his co-workers. [7]

In addition to the standard relaxation caused by the dense array of randomly oriented nuclear dipole moments, they observed a spontaneous extra relaxation of the spinpolarization function at the superconducting transition temperature. This extra relaxation is suppressed by the application of a small longitudinal field, indicating that its cause is static on the μ s scale. Furthermore, the fact that it can be best modeled by an exponential rather than a Gaussian relaxation indicates that its source is a broad distribution of internal fields from dilute array of sources. The Fig.4 by this group shows the temperature dependence of the field. The fact that the spontaneous extra relaxation sets in at T_c shows that this is most probably an intrinsic feature of the superconductivity. The other signature of time-reversal symmetry breaking comes from the vortex lattice structure. In conventional SC, normally a triangular or hexagonal lattice is the most stable solution (the famous Abrisokov vortex lattice), but under special circumstance square lattices are possible. The situation is different for unconventional superconductor with broken time-reversal symmetry. Such superconductor requires the use of a two-component order parameter in Ginzburg-Landau treatments. It had been shown theoretically by Agteberg [8] that in this case square or rectangular lattices are

expected to be favored over the entire *H-T* plane in the presence of physically reasonable values of Fermi-surface



Fig.5: Diffraction pattern of flux-line lattice in Sr₂RuO₄ measured by smallangle neutron scattering [9]

anisotropy. Using small-angle neutron scattering, Kealey and Forgan [9] obtained the diffraction pattern shown in the Fig. One can see clearly that vortices form square lattices. And it had been observed that this is so for all fields and temperature. Furthermore the field distribution observed is different from the possible square lattice emerging from a single component Ginzburg-Landau treatment.

The important feature to be emphasized here is that in both cases, the time-reversal symmetry broke down with the onset of superconductivity. It is possible to construct a two-component state with time-reversal symmetry breaking with d state such as $d_x^2 \cdot y^2 + id_{xy}$. However, in this case these two components have different symmetry-breaking properties, and are not degenerate in a tetragonal crystal field. Consequently the time-reversal symmetry-broken state would be entered as a second transition well below T_c, a clear contradiction with the experiment. [1] Also among the unitary p-wave states only has time-reversal symmetry breaking. All this indicates that pairing of should be.

IV. CONTROVERSIES

So far, Sr_2RuO_4 looks just like He³ with two-dimensional electronic structure and nothing more. If one considers the fact that the crystal structure can cause lots of different symmetry breakings, this actually looks a little amazing. And sure enough, the recent experiments are showing that this is not the whole story.

A. Gap structure: Existence and position of gap nodes (or deep minima)

In its simplest form, the conventional superconductor has isotropic gap. There is absolutely no available state with the energy less than the absolute value of the energy gap. That means at T<<T_c quasi-particles only exist by if thermal energy overcomes the gap energy Δ_0 , so the quasi-particle density would vary as exp(- Δ_0/k_BT). This exponential suppression at low temperature shows up in all experiment sensitive to either the number density of thermally excited quasi-particles or the quasi-particle density.

On the other hand, if there exist nodes in the gap, the thermodynamics would be altered substantially. Around the nodes there are now allowed states for all energy value, and the quasiparticle density now varies as the power laws rather than exponentially suppressed in T<<T_c regime.



Fig.6: Comparison of Sr₂RuO₄ electronic heat capacity with prediction for isotropic gap and line node [10]

To be able to say something about not only the existence of nodes/deep minima but also its position, we would need a direction sensitive probe. One of such probes is ultrasound attenuation. In superconductor, this phenomenon occurs because phononquasiparticle interaction dissipates phonon propagation. Now in very low temperature, for superconductor with gap nodes, the quasiparticles would exist only near the nodes. In this situation, the attenuation would be due mostly to the coupling between phonon and the nodal quasiparticle, hence the attenuation would have power-law dependence on temperature. The exponent of this power-law however is direction dependent, for the coupling would be direction dependent. For instance, if neither the phonon wave vector nor its polarization is perpendicular to nodal direction the coupling is maximal, and the node is 'activated'. Otherwise, the node is 'inactivated'. Obviously, the 'activated' node would have stronger attenuation, which means that the exponent on temperature

and The direct obvious most experimental probe of the thermodynamic state is the electronic heat capacity. The experiment by Nishizaki et al. [10] shows no exponential suppression up to very low temperature. Although it is guite similar to the prediction for the case with line node, with the power-law dependence at the low temperature down to 0.1K (T_c =1.48K), there is a significant difference in the normalized jump at T_c. Further more the heat capacity follows the power-law over too large T range to fit with the line node prediction.

Similar results have been also observed for NMR relaxation rate experiment.



Fig.7: Ultrasonic attenuation measured in terms of viscosity

would be smaller. J. Moreno and P. Coleman had found out that the difference of exponent between the attenuation by the 'active' and 'inactive' node should be equal to 2. [12] If there is a vertical node, it would mean that the 'active' and 'inactive' nodes should be detected among phonons with wave vector and polarization in xy plane.

In their Sr₂RuO₄ ultrasound attenuation experiment, Lupien *et al.* [11] has tested all four modes, L100, L110, T100 and T110 (where L refers to longitudinal, T transverse), satisfying this condition (1996) as is shown in Fig. 7. They have found the power-law at the low temperature (down to $T_c/30$) to be $T^{1.8}$ for L100, T110 and L110, and $T^{1.4}$ for T100. They failed to found the crucial difference of 2 between temperature exponents, which amounts to serious argument against vertical line nodes.

Another direction dependent probe into quasiparticle density – thermal conductivity along different directions – has yielded similar conclusion.



Fig.8: Proposed Fermi surface with a horizontal line node [1]

From experiments performed so far it seems to be logical to conclude that gap nodes or deep minima (at least 1/30 of full value, according to ultrasound attenuation) exist as a horizontal line around the cylindrical Fermi surface as sketched in Fig. 8. [1]

One thing that should be mentioned is that while it may be difficult to distinguish nodes with deep minima, theoretically the difference would be quite telling; gap minima, however deep, would not really demand any theoretical correction while any gap nodes would demand non-trivial explanation.

It can also be argued that if there are indeed nodes rather than minima, they should be locations where gap phase should change by π . Ideally Josephson junction experiment should be able to detect such

phase change. However the crystal proved to be very difficult to cut cleanly, and defects thus created often nests magnetic fluxes which render the result suspect. Consequently the effort by van Harlingen group, among others, has not yielded very definite result so far. [13]

B. Multiple superconducting phases

In triplet pairing, it often happens possible states are either degenerate or separated only by small amount of energy. This means that by adjusting external parameters, one can observe many different phases.

In case of Sr₂RuO₄, $\mathbf{d}=\Delta \hat{z} (k_x \pm ik_y)$, which is believed to be the basic state, is itself twofold degenerate. Agterberg pointed out that this degeneracy would lift in magnetic fields near H_{c2} applied in the ab plane; [8] the applied field would stabilize a state with a vertical line node corresponding to $\Delta \hat{z} k_x$, where the x-direction is along the field. The nodes would thus rotate with an in-plane field, and one of the signals of this state was predicted to be in-plane anisotropy of the upper critical field. Mao *et al.* [14] did succeed in observing such anisotropy; however, its magnitude was not in accord with Agterberg's prediction, and was temperature dependent to the extent that it even changed sign near T_c.

The study by also uncovered preliminary evidence for multiple phase behavior not predicted by Agtergerg. It appeared that there may be a second phase at high fields and low temperatures. with а bicritical point somewhere near 0.8K. Since then, highspecific resolution heat and thermal conductivity measurements have uncovered considerable evidence that such phase does indeed exist. Among these two, the most direct thermodynamic evidence is from the specific heat. [15] If the field is aligned in the plane within 0.5 degree, the single peak seen at all





Fig.9: Phase diagram with second critical field deduced from specific heat measurement by Deguchi *et al.*[15]

V. CONCLUSION

The hypothesis of $\mathbf{d} = \Delta \hat{z} (k_x \pm i k_y)$ spin-triplet pairing in Sr₂RuO₄ has been successful in wide varieties of experiments. But recent controversies on gap structure and multiple phase indicates that there still remains quite a bit of missing pieces in our picture; one cannot rule out that we would need entirely different pairing hypothesis.

It should be pointed out that the phase-sensitive probe that was mentioned for detecting gap nodes are even more meaningful for a most basic purpose – checking that the orbital wave function is indeed antisymmetric. [1] Concerning the gap node position question, it should be pointed out that experiments done so far may be too indirect to draw a definite conclusion. Also to answer 'gap node or deep minima' question, experiments need to probe into lower temperature.

Finally, it should also be pointed out that on the theoretical side, the most basic question of superconductivity mechanism has not been addressed properly. What we need is to incorporate the spin-orbit coupling due to crystal into the spin-fluctuation-driven pairing of He³. Certainly overlooking such microscopic details in order to concentrating on the basic symmetry of pairing was a strategy that paid off handsomely up to certain point. But it may be possible that we have already reached a level at which at least certain amount of such details are essential for understanding certain details of macroscopic phenomena.

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