## Raman Scattering and Anomalous Current Algebra in Mott Insulators

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We present a theory of high energy large shift Raman scattering in Mott insulators and show that it provides an instrument for direct measurements of local chirality and anomalous terms in the electronic current algebra. On this basis we argue that the electric-dipole-forbidden electronic transition at energy just below the charge-transfer gap recently observed in inelastic light scattering in insulating cuprates can be interpreted as a zero mode bound state in the chiral spin liquid.

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Recent Raman studies of a number of insulating cuprates revealed resonant features just below the optical absorption peak [1]. The most robust feature was observed in crossed polarizations, i.e., in the  $A_2$  pseudoscalar scattering geometry. It was conjectured in [1] that the  $A_2$  and other features correspond to electric-dipoleparity-forbidden transitions, the orbital moment of an intermediate state of the oxygen atom being different from orbital moments of the initial and final states of the copper atom.

In the present Letter we discuss a mechanism of inelastic light scattering due to anomalous orbital motion of charge carriers in the Mott insulator. If one ignores possibilities of intra-atomic (Cu-Cu) as well as interatomic (Cu-O) transitions discussed by the authors of Ref. [1], then we conclude that the observed features suggest the following: (i) Within the Mott-Hubbard gap there is an excitonlike bound state and (ii) this bound state has an odd symmetry under reflection of a two dimensional square lattice.

Raman measurements have already yielded important data about magnetic and phonon properties of insulating cuprates [2-5]. We show that the new *high energy, large shift* Raman techniques [1] give a direct tool to investigate electronic current algebra and chirality of charged excitations, which are important ingredients of various topological mechanisms of superconductivity [6,7].

A process of inelastic Raman scattering is an absorption of an incident photon with a frequency  $\omega_i$ , a wave vector  $\mathbf{k}_i$ , and a polarization  $\mathbf{e}_i$  and a simultaneous emission of a scattered photon  $(\omega_f, \mathbf{k}_f, \mathbf{e}_f)$ . As a function of transferred energy  $(\Omega = \omega_i - \omega_f)$  and momentum  $(\mathbf{q} = \mathbf{k}_f - \mathbf{k}_i)$  the Raman scattering cross section is given by the Kramers-Heisenberg formula [8] which can be written in the form of the ground state correlation function

$$R(\mathbf{q},\Omega) = \frac{\omega_f^3 \omega_i}{2\pi h^2 c^4} \operatorname{Im}\langle 0 | M^{\dagger}(\mathbf{q},\Omega) M(\mathbf{q},\Omega) | 0 \rangle \qquad (1)$$

of the time dependent scattering tensor  $M(\mathbf{r}, \tau) = e_i^{\mu} e_f^{\nu} \times M_{\mu\nu}(\mathbf{r}, \tau)$ .

In the case of insulator the latter is given by the formula

$$M_{\mu\nu}(\mathbf{r},\tau) = i \sum_{\mathbf{r}}' \int_0^\infty d\tau' e^{i\mathbf{k}_i \cdot \mathbf{r}' - i\omega_i \tau'} \times [j_\mu(\mathbf{r} + \mathbf{r}', \tau + \tau'), j_\nu(\mathbf{r}, \tau)], \quad (2)$$

where  $j_{i,f} = \mathbf{j}(\mathbf{k}_{i,f})\mathbf{e}_{i,f}$  is an electromagnetic current along polarization of an incident (scattered) photon.

Since photon wavelengths are always much larger than an interatomic separation, one may neglect a spatial dispersion of the scattering tensor, putting q = 0 in (1).

In general, scattering rates appear to be strongly dependent on photon polarizations. The scattering tensor (2) can be decomposed into four one dimensional irreducible representations of the square lattice point group  $D_{4h}$  (see, e.g., [4]). Below we shall draw our main attention to the  $A_2$  scattering amplitude  $M_{A_2} = M_{xy} - M_{yx}$ , which is odd under reflection on the square lattice and gives the strongest signal in the experiments [1].

In experiments [1] performed on insulating cuprates  $Y(Pr)Ba_2Cu_3O_{6+x}$  and  $Gd(Nd)_2CuO_4$  at the incident light frequency  $\omega_i = 3.4 - 3.8$  eV the A<sub>2</sub> resonance was found at Raman shift  $\Omega \approx 1.5$  eV—which is about 0.15-0.20 eV below the optical absorption peak. Being of order of the magnetic exchange J this energy scale could be considered as an indication on a magnetic (manybody) origin of the observed features. To elucidate the mechanism of the  $A_2$  scattering we exaggerate the conditions of the experiment, assuming that energies of incident and scattered light are much larger than the widths of relevant electronic bands. In this limit the time separation between current operators in (2) is very small and in the case of the  $A_2$  geometry the scattering tensor is proportional to the equal-time current-current commutator,  $M_{A_2} = M_{xy} - M_{yx} \approx (1/\omega_i)[j_x(\tau), j_y(\tau)]$ , where  $\mathbf{j}(\tau) = \sum_{\mathbf{r}} \mathbf{j}(\mathbf{r}, \tau)$  is the spatial average of the current operator. Higher order corrections to the scattering tensor can be expressed in terms of time derivatives of the current operator  $(d^n/d\tau^n)\mathbf{j}(\tau)$ .

In what follows we choose the simplest Hubbard

Hamiltonian (with the on-site Coulomb repulsion U and the hopping amplitude t) as a description of the Mott insulator, thus ignoring the intra-atomic structure of CuO<sub>2</sub>. Introducing one particle translation operators  $T_{\mu}(\mathbf{r}) = \sum_{\sigma=1,1} c_{\sigma}^{\dagger}(\mathbf{r} + \hat{\mu})c_{\sigma}(\mathbf{r})$  such that  $j_{\mu}(\mathbf{r}) = (et/2ihc)[T_{\mu}(\mathbf{r}) - T_{-\mu}(\mathbf{r})]$  and keeping terms up to the second order in  $\omega_i^{-1}$  we obtain

$$M_{A_{2}} = \frac{1}{\omega_{i}} [j_{x}(\tau), j_{y}(\tau)] \left( 1 + \frac{U^{2}}{\omega_{i}^{2}} \right) + \frac{t^{2}}{\omega_{i}^{3}} \left[ j_{x}(\tau) \left( \sum_{\mathbf{r}, \mu} T_{\mu}(\mathbf{r}) \right)^{2} j_{y}(\tau) - j_{y}(\tau) \left( \sum_{\mathbf{r}, \mu} T_{\mu}(\mathbf{r}) \right)^{2} j_{x}(\tau) \right],$$

$$M_{A_{1}} = M_{xx} + M_{yy} = -\frac{2U}{\omega_{i}^{2}} [j_{x}^{2}(\tau) + j_{y}^{2}(\tau)], \quad M_{B_{2}} = M_{xy} + M_{yx} = -\frac{2U}{\omega_{i}^{2}} \{ j_{x}(\tau), j_{y}(\tau) \},$$
(3)

while the amplitude of the  $B_1$  scattering  $(M_{B_1} = M_{xx} \sqcap M_{yy})$  is even smaller.

Note that the last term in the formula for  $M_{A_2}$  contributes to the quasielastic  $A_2$  scattering  $(\Omega \rightarrow 0)$  which was considered by Shastry and Shraiman [4] under assumption  $|U - \omega_i| \ll \min(U, \omega_i)$ .

Charge excitations in the Mott insulator cause distortions of the magnetic state. As a result, a phase of the excitation wave function may depend not only on its location but also on a path, which the hole passed to arrive at the site. Therefore a hole inserted into the Mott insulator may acquire a phase while moving along the closed path. This property is the quantum holonomy which is determined by a spin chirality of the insulating ground state [9,10]. We define it as a measure of noncommutativity of one particle translation operators  $T_{\mu}$  when acting on the insulating ground state

$$T_x T_y |0\rangle = e^{i\hat{\Phi}} T_y T_x |0\rangle = e^{(i/2)\hat{\Phi}} T_{x+y} |0\rangle.$$
(4)

Using the relation  $\prod_C T_{\mu}(\mathbf{r}) = \mathbf{tr} \prod_C [1 + 2\sigma \mathbf{S}(\mathbf{r})]$ , where C is a lattice contour and  $\sigma$  are auxiliary Pauli matrices, one can express the chiral operator associated with an elementary plaquette,  $\langle 0|\exp(i\hat{\Phi})|0\rangle$  $\sim \langle 0|T_x T_y T_{-x} T_{-y}|0\rangle$ , as an expectation value of the local chirality operator. It turns out that high energy Raman scattering provides an instrument to measure matrix elements of the chirality operator.

Some information about spin chiralities can be already obtained from the integrated Raman intensity  $R^{\text{int}} = \int_0^\infty R(\Omega) d\Omega$ . In a general case of noncollinear polarizations  $R^{\text{int}}$  is given in the leading order in  $\omega_i^{-1}$  by the static correlation function of equal-time current-current commutators:  $R^{\text{int}} \approx R_{A_2}^{\text{int}} \sim \langle 0||[j_x, j_y]|^2|0\rangle$ . Thus  $R^{\text{int}}$ is capable of measuring fluctuations of local chiralities:  $R_{A_2}^{\text{int}} \sim \sum_P \langle 0|(\sin \frac{1}{2} \hat{\Phi}_P)^2|0\rangle$ .

The amplitude of an elastic scattering  $R^{el} = R(\Omega \rightarrow 0)$ is a more informative object. In the leading order in  $\omega_i^{-1}$ it measures an averaged difference between holonomies associated with two oppositely oriented elementary closed contours which is equal to the average chirality of the ground state. Using (3) we obtain

$$R_{A_2}^{\text{el}}(\Omega) \sim \left(\frac{t}{\omega_i}\right)^4 \left|\sum_P \langle 0|\sin\hat{\Phi}_P|0\rangle\right|^2 \delta(\Omega)$$

The matrix element appearing in this formula yields the total solid angle formed by all spins in the ground state

$$R_{A_2}^{\rm el}(\Omega) \sim \left(\frac{t}{\omega_i}\right)^4 \left|\sum_{P} \langle 0 | (\mathbf{S}_1 \times \mathbf{S}_2) \cdot \mathbf{S}_3 | 0 \rangle \right|^2 \delta(\Omega) , \quad (5)$$

where 1, 2, 3 denote any three nearest neighboring sites on the plaquette P and the sum goes over all plaquettes.

Note that in other geometries elastic scattering occurs only in the next order in  $\omega_i^{-1}$  and involves only selfretracing paths (i.e., does not depend on chirality).

An observation of a separate peak in the  $A_2$  elastic scattering would mean a spontaneous parity breaking in the ground state [9,10]. Since elastic scattering measures a spatial average of chirality (if any) there are numerous obstacles for its observation [11]. Although an additional experimental study is desirable, it seems most likely that the ground state of *insulating* cuprates is parity even.

More interesting information can be obtained from inelastic scattering at energy shift close to the chargetransfer gap [1]. Let us suppose that inside the Hubbard band there exists a bound state  $|s\rangle$  of a hole and a doubly occupied site (doublon). Introducing notations for the hole-doublon bound state wave function  $\Psi_{\sigma,\sigma'}(\mathbf{r},\mathbf{r}')$  and a configuration of surrounding spins  $|\{\sigma\}\rangle$ , we can express our state as  $|s\rangle = \sum_{\mathbf{rr}'} \Psi_{\sigma,\sigma'}(\mathbf{r},\mathbf{r}')c^{\dagger}_{\sigma}(\mathbf{r})c'_{\sigma}(\mathbf{r}')|\{\sigma\}\rangle$ .

The energy of this state is by  $E_b \sim J$  lower than the optical absorption threshold  $\omega_T \approx U$  corresponding to the location of the upper Hubbard band. The leading contribution to the Raman intensity at large laser energy comes form the  $A_2$  amplitude which is proportional to the matrix element of the current-current commutator taken between the ground state and above bound state:

$$R_{A_2}(\Omega) \sim |\langle 0|[j_x, j_y]|s\rangle|^2 \delta(\Omega - \omega_T + E_b)$$

Because of the semiclassical character of light scattering at large  $\omega_i$  the hole **r** and the doublon **r'** must be located on the same diagonal of a plaquette, otherwise in the leading order in  $t/\omega_i$  the matrix element vanishes. Then the matrix element of the current commutator entering  $R_{A_i}(\Omega)$  acquires the form  $(\mathbf{r} \in P)$ :

$$\langle 0|[j_x, j_y]|_{\mathcal{S}} \rangle = 2 \sum_{\mathbf{r}, \sigma_1, \sigma_2} \langle 0_{\sigma_1, \sigma_2} |[T_{-x}, T_{-y}]T_{x+y}|_{\mathcal{S}} \rangle$$
$$\times \Psi_{\sigma_1, \sigma_2}(\mathbf{r}, \mathbf{r} + \mathbf{\hat{x}} + \mathbf{\hat{y}}) ,$$

where  $\hat{\mathbf{x}}$  and  $\hat{\mathbf{y}}$  are the lattice vectors and  $\langle \mathbf{0}_{\sigma_1,\sigma_2} |$  denotes the ground state with spins at sites  $\mathbf{r}$  and  $\mathbf{r}'$  being fixed.

The sum over  $\mathbf{r}$  is effectively restricted to sites close to the location of the center of mass of the hole-doublon pair.

Since a spin distortion caused by the localized bound state extends to a very few plaquettes, one may neglect a difference between the ground and the excited spin configurations when calculating an overlap factor  $\langle 0_{\sigma_1,\sigma_2} | \sigma \rangle$ . Then, we conclude that in the first approximation inelastic scattering gives a *local* value of the chirality in the ground state. Similar to the case of the elastic scattering (5) the expectation value of the holonomy operator  $\langle 0 | [T_{-x}, T_{-y}] T_{x+y} | 0 \rangle \sim \langle 0 | [\mathbf{S}(\mathbf{r}) \times \mathbf{S}(\mathbf{r} + \hat{\mathbf{x}})] \cdot \mathbf{S}(\mathbf{r} + \hat{\mathbf{x}} + \hat{\mathbf{y}}) | 0 \rangle$  is proportional to the solid angle subtended by three spins belonging to the plaquette which contains a charge excitation.

Other symmetries contribute in the next order in  $J/\omega_i$ and are independent of chirality.

Thus using the minimal one band Hubbard model we interpret the observation of the  $A_2$  peak in insulating cuprates as an evidence of long range correlations between triads of adjacent spins. It is important to notice that this kind of order does not necessarily mean parity breaking. The simplest possibility to get a nonzero holonomy, while saving the ground state invariance under the largest subgroup of the magnetic class including parity, is the so-called  $\pi$ -flux state. In this state  $\langle 0|\hat{\Phi}|0\rangle = \pi$  on every plaquette and translation operators anticommute,

$$\langle 0 | T_x T_y T_{-x-y} | 0 \rangle = - \langle 0 | T_{-y} T_x T_{-x+y} | 0 \rangle = i\Delta ,$$

$$(T_x T_y + T_y T_x) | 0 \rangle = 0 ,$$
(6)

In this state the solid angle of three adjacent spins alternates from one plaquette to another, so its spatial average vanishes:  $\langle 0 | (\mathbf{S}_1 \times \mathbf{S}_2) \cdot \mathbf{S}_3 | 0 \rangle = - \langle 0 | (\mathbf{S}_{1'} \times \mathbf{S}_{2'}) \cdot \mathbf{S}_{3'} | 0 \rangle = \Delta$ , where {1,2,3} and {1',2',3'} label any three nearest neighboring sites on adjacent plaquettes with the same orientation. Note that the staggered chiral order (6) is not necessarily inconsistent with the antiferromagentic ordering which is a property of two-spin correlations  $\langle \mathbf{S}_i \mathbf{S}_j \rangle \sim (-1)^{|i-j|}$  [12].

The observation of the  $A_2$  Raman resonance implies that the ground state and the excited state  $|s\rangle$  have different parities. At first glance it seems impossible in the framework of the one band Hubbard model on a square lattice. Indeed, in this case all eigenstates are doubly degenerate due to the reflection symmetry  $\hat{R}$ . Namely, if there exists an eigenstate  $|\Psi\rangle$  supporting currents  $j_x|\Psi\rangle, \langle\Psi|j_y$ , then there is also an eigenstate  $|\Psi^R\rangle = \hat{R}|\Psi\rangle$  with currents  $j_x|\Psi^R\rangle = j_y|\Psi\rangle, \quad \langle\Psi^R|j_y$  $= \langle\Psi|j_x$ . Then the current commutator vanishes:

$$[j_x, j_y] = \sum_{|\Psi\rangle} (j_x |\Psi\rangle \langle \Psi| j_y - j_y |\Psi\rangle \langle \Psi| j_x) = 0.$$

Apparently, the sum does not cancel out if one assumes a spontaneous parity breaking already in the ground state  $(\hat{R}|0\rangle \neq |0\rangle)$ . For example, this is the case of the uniform chiral spin liquid state characterized by a nonzero expectation value of the spatial average of chirality [9,10]. However, this state seems unlikely in insulating cuprates, particularly, because it is incompatible with the symmetry of the square lattice and also with an antiferromagnetic order.

One the other hand, the current-current commutator may not vanish if there is an excited state  $|\Psi_0\rangle$  which is annihilated by the reflection operator  $\hat{R}|\Psi_0\rangle = 0$ , while the ground state respects parity. Then a nonzero contribution to the current commutator  $[j_x, j_y] = j_x |\Psi_0\rangle \langle \Psi_0| j_y$  $-j_y |\Psi_0\rangle \langle \Psi_0| j_x$  comes from the excited state having no partner under reflection, which is known as a zero mode [13].

Thus, the zero mode contribution is the only one present in the case of the resonant  $A_2$  scattering:

$$R_{\mathcal{A}_2}(\Omega) \sim \sum_{\mathbf{r}} \Delta^2 |\Psi_0(\mathbf{r}, \mathbf{r})|^2 \delta(\Omega - U) .$$
<sup>(7)</sup>

A zero mode (if it exists) is always a bound state (in fact the lowest one). In our case it is a bound state of a hole and doublon with energy by order of J lower than the charge transfer gap. Moreover, it exists if there is a local chiral order.

In order to see that zero mode is always a bound state we consider the case of a charge excitation moving in a spin background which evolves adiabatically.

Following Refs. [14,15] we describe a charged excitation in the Mott insulator by a coherent state of a hole  $\psi(\mathbf{r})$  and a hard core boson  $z_{\sigma}(\mathbf{r})$  subjected to the local constraint:  $|z_1|^2 + |z_2|^2 = 1 - \psi^{\dagger}\psi$ . Then  $c_{\sigma}(\mathbf{r}) = \psi(\mathbf{r})$  $\times z_{\sigma}^{\dagger}(\mathbf{r})$  represents an electron operator and  $\mathbf{S}(\mathbf{r}) = \bar{z}_{\sigma}(\mathbf{r})$  $\times \sigma_{\sigma\sigma'}z_{\sigma'}(\mathbf{r})$  serves for a local spin. The hopping Hamiltonian is  $H = t \sum_{(\mathbf{a},\mathbf{b})} \psi_{\sigma}^{\dagger}(\mathbf{a}) \Delta_{\mathbf{a},\mathbf{b}} \psi_{\sigma}(\mathbf{b}) + c.c.$ , where **a** and **b** are sites of sublattices A and B and  $\Delta_{\mathbf{a},\mathbf{b}} = \sum_{\sigma} \bar{z}_{\sigma}(\mathbf{a}) z_{\sigma}(\mathbf{b})$ .

If the hopping amplitude t is bigger than the exchange constant J, then antiferromagnetic spin dynamics cannot be treated adiabatically, because every hole jump changes a spin configuration abruptly by flipping sublattice spins. However, after two consecutive jumps, a hole appears on the same sublattice, so the spin configuration remains approximately unchanged. To implement the adiabatic approach we introduce a staggered chemical potential for holes on different sublattices  $\mu(\sum_a c_a^{\dagger} c_a - \sum_b c_b^{\dagger} c_b)$ . If the hopping energy t is less than  $\mu$ , then a hole and a doublon appear on different sublattices only virtually. Therefore in the leading order in the adiabatic parameter  $t/\mu$  one can consider only processes of two consequent hoppings described by the effective Schrödinger equation for the hole-doublon wave function:

$$t' \sum_{\mathbf{b}', i=1,2} \Delta_{\mathbf{a}_i, \mathbf{b}'} \Delta_{\mathbf{b}', \mathbf{a}'_i}^{\dagger} \Psi_0(\mathbf{a}_1', \mathbf{a}_2') = (E - \mu^2) \Psi_0(\mathbf{a}_1, \mathbf{a}_2) \, .$$

As long as  $t' \sim t^2/\mu > J$  one can treat magnetic fluctuations adiabatically. After all, we suggest to consider this Hamiltonian as phenomenological.

The effective Schrödinger operator is a square of the elementary translation operator so one can apply stan-

dard arguments about its zero modes (see, e.g., [16]). The only nondegenerate state with the energy  $E = \mu^2$  is the one annihilated by either  $\Delta_{\mathbf{a},\mathbf{b}}$  or  $\Delta_{\mathbf{b},\mathbf{a}}^{\dagger}$ . If the ground state possesses a *local* chirality then  $\langle 0|(\mathbf{S}_1 \times \mathbf{S}_2) \cdot \mathbf{S}_3|0\rangle \neq 0$  and operators  $\Delta_{\mathbf{a},\mathbf{b}}$  and  $\Delta_{\mathbf{b},\mathbf{a}}^{\dagger}$  do not commute. Therefore only one of them can have a zero mode for a given spin configuration. Thus we obtain two candidates for a zero mode which satisfy equations (i)  $\Delta_{\mathbf{a},\mathbf{b}}\Phi_0(\mathbf{b})=0$  $[\Phi_0(\mathbf{a})=0]$  and (ii)  $\Delta_{\mathbf{b},\mathbf{a}}^{\dagger}\Phi_0(\mathbf{a})=0$   $[\Phi_0(\mathbf{b})=0]$ . Charge excitations occupying the zero mode stay on only one of the two sublattices.

Many properties of zero modes are universal and depend only on the crystal symmetry of the lattice. They constitute the subject of the "index" theorem [16]. In particular, the index theorem states that a spin configuration which supports a zero mode (i) must have a non-trivial topology (i.e., chirality of the excited state differs from that of the ground state by one flux quantum) and (ii) the local fermion density of the occupied zero mode  $\rho(\mathbf{r}) = \Psi_0(\mathbf{r}, \mathbf{r}) = 2|\Phi_0(\mathbf{r})|^2$  equals topological charge density  $q(\mathbf{r}) = (1/2\pi)[(d/dx)\mathbf{n}(\mathbf{r}) \times (d/dy)\mathbf{n}(\mathbf{r})] \cdot \mathbf{n}(\mathbf{r})$ , written in terms of the antiferromagnetic vector  $\mathbf{n}(\mathbf{r})$ . Thus the spin configuration accompanying the hole-doublon bound state can be viewed as a magnetic hedgehog which carries one flux quantum on the top of the flux distribution in the ground state.

Combining previous results, we obtain another formulation of the index theorem: The equal time currentcurrent commutator acquires the anomalous term given by the topological density of the spin excitation,

$$[j_x(\mathbf{r}), j_y(\mathbf{r}')] \sim i\Delta q(\mathbf{r})\delta(\mathbf{r} - \mathbf{r}').$$
(8)

As a result, a local density of topological charge explicitly appears in the scattering rate (7).

Going beyond the adiabatic approximation we estimate the width of the  $A_2$  Raman peak as the inverse spin relaxation time which remains of the order of J even at low temperatures. This does not contradict the experiments which show that the width of the peak only slightly narrows as temperature decreases.

Note that according to (3), there should be no scattering in the  $B_2$  geometry if translations anticommute [see (6)]. Indeed no  $B_2$  contribution to the resonant scattering at  $\Omega \approx U$  has been detected in Ref. [1].

In contrast to the  $A_1$  feature the anomalous  $A_2$  peak depends on the symmetry of the magnetic structure and not on details of the Hamiltonian. It was also observed and stressed in Ref. [1].

However, an additional experimental study is necessary to conclude whether these features are due to local interatomic transitions or anomalous orbital motion of charge excitations in the Mott insulator.

The presence of zero modes in the insulating state together with anomalous current algebra imply important properties of the doped state. Because of the particlehole symmetry of the half-filled band the spin configuration  $|s\rangle$  in the presence of two holes contains a spin soliton in the same way as in the case of a hole-doublon pair. Therefore at small doping, which does not destroy the magnetic ground state, two holes couple with each other inside the topological spin bag. A residual interaction between zero modes lifts their degeneracy and opens a narrow midgap band. Although this band is always filled, it remains compressible [6]. This phenomenon may provide a basis for a topological mechanism of superconductivity.

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