

Ion displacement and electron-hole localization in perovskite-like layered cuprates

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The electron-hole localization can be realized on a boundary ab plane of layered crystals if the ion charge e_i and the c -direction force constant k_i satisfy the relation of $(1/S)\sum_{i \in V}(e_i^2/k_i) \geq 0.3 \text{ \AA}$, where S and V represent the boundary surface and the layer volume, respectively. In many perovskite-like layered cuprates, the relation is satisfied mainly due to the smallness of k_{Cu} compared to other k_i 's in typical perovskite crystals. This indicates the possibility of the electron-hole localization on the CuO_2 plane, where high- T_c superconductivity can be realized.

I. INTRODUCTION

The modulated structure of a displacive type has been observed in many perovskite-like layered cuprates, such as in BSCCO ,¹⁻³ TBCCO ,²⁻⁴ NCCO ,⁵⁻⁷ YSCBO ,⁸ and LSCO .⁹ In BSCCO , for example, the ion displacement in the c -axis direction is alternately modulated along the b -axis direction with the modulation period about 26 \AA . To explain the modulated structure, we previously proposed a two-dimensional electron-hole lattice model,¹⁰ where the electrons and holes are, to stabilize the local polarization caused by the ion displacement, pair created on the CuO_2 plane (and also the BiO plane in BSCCO) or transferred onto the CuO_2 (and the BiO) planes from their adjacent planes.

If the pair-creation energy and the charge-transfer energy are equal or less than about 1 eV , we found that the charge distribution of a stripe configuration can be realized;¹¹ the smallness of the pair-creation energy, which corresponds to the band gap on a semiconductive plane, in the formation of the electron stripe has been also emphasized by Kusmartsev,¹² where the electrons are trapped by an extended polarization well. From the stability condition of the total energy plus the chemical potential with respect to the electron lattice constants D_a and D_b , it has been found that the charge distribution of a checkerboard configuration, where the electrons and holes are aligned in both a - and b -axis directions, cannot be allowed,¹¹ resulting in the formation of the stripe configuration under certain region of the chemical potential. However, in calculating the electronic state of the localized charges, we assumed that the elastic energy due to the ion displacement is small enough compared with the Coulomb potential between the localized electrons (holes) and the polarization charges. If the elastic energy is comparable with the Coulomb potential, the assumption of the charge localization on the CuO_2 plane, in itself, may be violated, due to the overlap of the electron wave function between two adjacent CuO_2 planes. This is because as the force constant increases, the bound potential owing to the (background) polarization charges tends to weaken, so that the spatial spread of the wave function may elongate in the c -axis direction.

On the other hand, it has been pointed out that the localized charges are closely related to high- T_c superconductivity.¹³⁻¹⁵ Among them, Beschoten *et al.*¹⁵ have

suggested from the temperature dependence of the Hall mobility that the Cooper pairs can be formed by spatially localized states. Thus it is considered to be important to examine the possibility of the charge localization especially on the CuO_2 plane, where high- T_c superconductivity can be realized. Experimentally, the coexistence of the localized and itinerant charges has been indicated in normal and superconducting phases of YBCO by the femtosecond time-resolved spectroscopy and the transient photoconductivity measurements.¹⁶⁻¹⁸

Although the electronic state of the layered cuprates has been theoretically studied using *ab initio* calculations¹⁹ and a linear augmented-plane-wave method,^{20,21} the relationship between the ion displacement and the electron-hole localization is not manifestly clarified. In Sec. II we deal with the electronic state of the localized charge in the background of the local polarization charge caused by the ion displacement, where the ion elastic energy is taken into account. For the electron to be localized on the boundary surface, it will be found that the ion charge e_i and the c -direction force constant k_i should satisfy the relation of $(1/S)\sum_{i \in V}(e_i^2/k_i) \geq 0.3 \text{ \AA}$, where S and V represent the boundary surface and the layer volume, respectively. In Sec. III, we give summary and discussion.

II. MODEL HAMILTONIAN

If the energy of the two-dimensional (2D) electron-hole lattice system is composed of the ion elastic energy, the electron kinetic energy, and the Coulomb potential $U(\mathbf{x};\mathbf{x}')$ ($=1/|\mathbf{x}-\mathbf{x}'|$) between the electron and the (background) polarization charge caused by the ion shift, the model Hamiltonian H (in units of $\hbar=2m=e=1$, where \hbar , m , and e represent the Planck constant, the electron mass, and the elementary charge, respectively) can be written as

$$H = \frac{N}{2} \sum_{i \in V} k_i (\Delta z_i)^2 - \sum_{\alpha = \pm 1/2} \int \psi_{\alpha}^{\dagger}(\mathbf{x}) \nabla^2 \psi_{\alpha}(\mathbf{x}) d^3 \mathbf{x} + \frac{1}{2} \int : n^{(\text{tot})}(\mathbf{x}) U(\mathbf{x};\mathbf{x}') n^{(\text{tot})}(\mathbf{x}') : d^3 \mathbf{x} d^3 \mathbf{x}', \quad (1)$$

where N , V , k_i , and Δz_i represent the total electron lattice number, the volume occupied by one electron, the z -direction

force constant, and the z displacement of the i th ion, respectively. Furthermore, $n^{(\text{tot})}(\mathbf{x}) = n(\mathbf{x}) + \sum_{i \in V} \sigma_i(x, y) \delta(z)$ with $n(\mathbf{x}) = \sum_{\alpha = \pm 1/2} \psi_{\alpha}^{\dagger}(\mathbf{x}) \psi_{\alpha}(\mathbf{x})$ and $\sigma_i(x, y)$ representing the polarization charge density caused by the i th ion displacement. Noticing that σ_i and Δz_i are related as $\sigma_i = (1/V) e_i \Delta z_i$ (e_i is the effective charge of the i th ion) and that N can be written as $N = (1/S) \int dx dy$ (S is the surface area occupied by one electron), we can replace the first term in the right-hand side of Eq. (1) by

$$\frac{N}{2} \sum_{i \in V} k_i (\Delta z_i)^2 \rightarrow \frac{1}{2} \sum_{i \in V} \kappa_i \int \{\sigma_i(x, y)\}^2 dx dy, \quad (2)$$

where $\kappa_i = (k_i/e_i^2)(V^2/S)$. Substituting Eq. (2) into Eq. (1) and requiring the stability condition of $\delta\langle H \rangle / \delta\sigma_i(x, y) = 0$, we can rewrite H as (see Appendix A)

$$\begin{aligned} H \rightarrow & - \sum_{\alpha = \pm 1/2} \int \psi_{\alpha}^{\dagger}(\mathbf{x}) \nabla^2 \psi_{\alpha}(\mathbf{x}) d^3 \mathbf{x} \\ & + \frac{1}{2} \int : n(\mathbf{x}) [U(\mathbf{x}; \mathbf{x}') n(\mathbf{x}') \\ & - U'(\mathbf{x}; \mathbf{x}') \langle n(\mathbf{x}') \rangle] d^3 \mathbf{x} d^3 \mathbf{x}', \end{aligned} \quad (3)$$

where $U'(\mathbf{x}; \mathbf{x}') = 1/[1 - \lambda(\partial/\partial z)] U(\mathbf{x}_+; \mathbf{x}'_-)$ with $1/\lambda = 2\pi \sum_{i \in V} (1/\kappa_i)$ and $\mathbf{x}_{\pm} = (x, y, \pm|z|)$.

Now we deal with a localized state. Even in the superconducting state, where the localized charges coexist with the itinerant charges,¹⁶⁻¹⁸ it will be found that the subsequent results may not be altered in a stripe phase, where the localized electrons and holes are equal in number. In the mixed phase of the localized state $|\psi^{(\text{loc})}\rangle$ and the itinerant state $|\psi^{(\text{itin})}\rangle$, the expectation value of H can be written as $\langle H \rangle = \sum_{s, s' \in \{\text{loc}, \text{itin}\}} [K^{(s)} + U^{(s, s')}]$, where $K^{(s)}$ represents the kinetic energy and $U^{(s, s')}$ corresponds to the Coulomb interaction between the states s and s' . In general, the Coulomb interaction of $U^{(\text{itin}, \text{loc})}$, which should be screened based on the Thomas-Fermi theory, cannot be neglected (in 2D electron gas, the screening radius, which, different from 3D gas, is independent of the carrier density, is on the order of 1 Å). However, if the localized electrons and holes are equal in number, as in the case of a stripe phase, $U^{(\text{itin}, \text{loc})}$ turns out to be vanishing from the stability condition of $\partial\langle H \rangle / \partial|\psi^{(s)}\rangle = 0$ (for more strict discussion, see Ref. 22). This can be understood by considering that the (screened) Coulomb interaction of the itinerant charge with the localized electron cancels that with the localized hole. Hence in the stripe phase, the electronic states of the localized and the itinerant phases can be treated independently, implying that the following results valid for the localized state can also hold for the mixed state.

In the localized state, $\langle \psi^{(\text{loc})} | H | \psi^{(\text{loc})} \rangle$ can be decomposed as $\langle \psi^{(\text{loc})} | H | \psi^{(\text{loc})} \rangle = \sum_i H_i + \sum_{i \neq j} H_{ij}$, where H_i and H_{ij} represent the intralattice energy and the interlattice energy, respectively. If the electron lattice constant D is much larger than the spatial spread Δ of the electron wave function, H_{ij} is negligible compared with H_i , that is, $H_{ij}/H_i = O(\Delta/D)$ or

$O[(\Delta/D)^3]$ for $\lambda \gg 1$ or $\lambda \ll 1$, respectively. This is because $H_i = O(1/\Delta)$, while $H_{ij} = O(1/D)$ or $H_{ij} = O(\Delta^2/D^3)$ for $\lambda \gg 1$ or $\lambda \ll 1$, respectively. In a practical case, where $D \sim 10$ Å, which corresponds to the modulation period, and $\Delta \sim 1$ Å, which is on the order of the Bohr radius, H_{ij} can be neglected.

In a singly or doubly occupied state, H_i can be written in the Hartree-Fock approximation as

$$\begin{aligned} H_i &= H_i^{(s)} = \langle -\nabla^2 \rangle - \frac{1}{2} \langle U' \rangle \quad (\text{for single occupation}), \\ &= H_i^{(d)} = 2H_i^{(s)} + \frac{1}{2} \langle U \rangle \quad (\text{for double occupation}). \end{aligned} \quad (4)$$

In estimating H_i , it is convenient to use a variational method, rather than to solve the original Hartree-Fock equation. Since we are concerned with the localized state, it seems natural to choose the trial function of the ground state $|\phi\rangle$ as a Gaussian form, that is, $\langle \mathbf{x} | \phi \rangle = \prod_{i=1}^3 [1/\sqrt{(2\pi)\Delta_i}] \exp(-x_i^2/4\Delta_i^2)$, where Δ_i 's (with $\Delta_1 = \Delta_2$) are variational parameters. Notice that Δ_3 is an increasing function of λ (this is because as λ increases, the polarization charge density decreases so that the bound potential in the z -axis direction can be reduced). Then it is sufficient to examine the limiting case such that $\lambda \gg \Delta_i$ (its validity will soon be verified). In this case, the doubly occupied state turns out to be unstable, because the potential energy $\langle -U' + \frac{1}{2}U \rangle$ tends to be positive due to the vanishing of $\langle U' \rangle$ in the limit of $\lambda \rightarrow \infty$.

Hence we only deal with the singly occupied state, where $H_i^{(s)}$ can be written for $\lambda \gg \Delta_i$ as (see Appendix B)

$$H_i^{(s)} = \frac{1}{2} \left(\frac{1}{2} + \frac{1}{1+\beta} \right) \frac{1}{\Delta_3^2} - \frac{1}{2\lambda} \left[\ln \frac{\sqrt{2}\lambda}{\Delta_3} - g(\beta) \right]. \quad (5)$$

Here $\beta = (\Delta_1/\Delta_3)^2 - 1$ and $g(\beta) = \int_0^{\infty} \ln x \cdot (\partial/\partial x) f_{\beta}(x) dx$, where $f_{\beta}(x) = \text{erfc}^2(x) \exp(-2\beta x^2)$ with $\text{erfc}(x) = (2/\sqrt{\pi}) \int_x^{\infty} \exp(-t^2) dt$. From the stability condition of $\partial H_i^{(s)}/\partial \Delta_3 = \partial H_i^{(s)}/\partial \beta = 0$, we obtain

$$\Delta_3 = \sqrt{a_B \lambda \left(\frac{1}{2} + \frac{1}{1+\beta} \right)}, \quad \beta = 1.213, \quad (6)$$

where $a_B (= \hbar^2/m e^2)$, the Bohr radius, has been explicitly represented.

For the electrons and holes can be regarded as well localized on the xy plane, the z -direction spatial spread of the electron wave function, $2 \times \Delta_3$, should be smaller than the interplanar distance l , that is,

$$\Delta_3 \lesssim \frac{l}{2} \Leftrightarrow \frac{1}{S} \sum_{i \in V} \frac{e_i^2}{k_i} \gtrsim \tilde{a}_B, \quad (7)$$

where $\tilde{a}_B = (1/\pi)[1 + 2/(1+\beta)]a_B (= 0.321 \text{ Å})$, and we have used the relations of $l = V/S$ and Eq. (6). Originally, Eq. (7) is valid for $\lambda \gg \Delta_3$ or from Eq. (6) $\lambda \gg a_B$. In the case of $\lambda \ll a_B$, the value of Δ_3 should be smaller than that for $\lambda \gg a_B$ (because Δ_3 is an increasing function of λ , as men-

TABLE I. Values of S_0 , k_{Cu} , $(1/S_0)(e^2/k_{\text{Cu}})$, and r_{Cu} for various layered cuprates. The c -direction force constant k_i is evaluated using the interatomic force constant f_{ij} as $k_i = \sum_{j(\neq i)} f_{ij} \cos \theta_{ij}$, where θ_{ij} represents the angle between the c and the ij directions. The values of S_0 and k_{Cu} for $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$, $\text{YBa}_2\text{Cu}_3\text{O}_7$, $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$, and $\text{Tl}_2\text{Ba}_2\text{CaCu}_2\text{O}_2$ are taken from Refs. 23–26, respectively, while the values of θ_{ij} and $r_{\text{Cu},\text{O}}$ for these cuprates are taken from Refs. 27–29 and 26, respectively.

| Cuprate | S_0 (\AA^2) | k_{Cu} (kdyn/cm) | $\frac{1}{S_0} \frac{e^2}{k_{\text{Cu}}} (\text{\AA})$ | $r_{\text{Cu},\text{O}}$ (\AA) |
|---------|--------------------------|---------------------------|--|---|
| LSCO | 14.2 | 30 | 0.54 | 2.4 |
| YBCO | 14.8 | 40 ^a | 0.39 | 2.3 |
| BSCCO | 14.6 | 36 | 0.44 | 2.2 |
| TBCCO | 14.9 | 39 | 0.40 | 2.7 ^b |

^aDue to the lack of the force constant data for $f_{\text{Cu},\text{Ba}}$ and $f_{\text{Cu},\text{Y}}$, we approximate k_{Cu} as $f_{\text{Cu},\text{O}}$.

^bCalculated value by the same authors who derived the value of k_{Cu} .

tioned in the above paragraph). Hence the relation of Eq. (7) is automatically guaranteed in the case of $\lambda \ll a_{\text{B}}$, once the relation is satisfied for $\lambda \gg \Delta_3$.

Now we examine whether or not the relation of Eq. (7) is satisfied in perovskite-like layered cuprates. In general, the c -direction force constant of the Cu ion is small enough compared with that of other ions, so that it may be sufficient to show the following relation:

$$\frac{1}{S_0} \frac{e_{\text{Cu}}^2}{k_{\text{Cu}}} \gtrsim a_{\text{B}}, \quad (8)$$

where S_0 is the unit crystal lattice surface within which one Cu ion is occupied. Since $\sum_{i \in V} (e_i^2/k_i)$ is proportional to S , S can be chosen as unit crystal lattice surface S_0 . Using the experimental values of S_0 and k_{Cu} (see Table I), we find that the relation of Eq. (8) is satisfied for $e_{\text{Cu}} \gtrsim e$.

To be compared with a typical perovskite crystal such as BaTiO_3 , where two-dimensionally localized charges are not observed, we estimate the value of $(1/S_0)(e_i^2/k_i)$, in which i corresponds to the Ti ion in BaTiO_3 . Near the ferroelectric Curie temperature, when the soft-mode phonon of the Ti ion is observed, k_{Ti} can be written using the dipole interaction as³⁰ $k_{\text{Ti}} \approx -4(e_{\text{Ti}}e_{\text{O}}/r_{\text{Ti},\text{O}}^3)$, where $r_{\text{Ti},\text{O}}$ represents the distance between the Ti ion and the apical oxygen. Hence $(1/S_0)(e_{\text{Ti}}^2/k_{\text{Ti}})$ can be estimated as

$$\frac{1}{S_0} \frac{e_{\text{Ti}}^2}{k_{\text{Ti}}} \approx 0.78 \tilde{a}_{\text{B}} \quad (\text{near Curie temperature}), \quad (9)$$

where use has been made of $S_0 = a^2$, $r_{\text{Ti},\text{O}} = a/2$, and the relation of $e_{\text{Ti}}/|e_{\text{O}}| \approx 2$,³⁰ with a representing the a -lattice constant ($a \approx 4 \text{\AA}$). Considering the relation of $k_{\text{Ti}} \ll k_{\text{Ba}}, k_{\text{O}}$,³¹ we find that the charge localization is difficult to realize in BaTiO_3 .

III. SUMMARY AND DISCUSSION

We have found in layered cuprates, where the relation of Eq. (7) is satisfied, localized charges can be realized on the CuO_2 plane, mainly due to the smallness of the c -direction force constant of the Cu ion, compared with that of other ions in typical perovskite crystals. In layered cuprates $k_{\text{Cu}} \approx 40$ kdyn/cm, while in BaTiO_3 , for example, $k_{\text{Ti}} \approx 500$ kdyn/cm.³⁰ The condition of Eq. (7) is necessary for the charge localization, so that we should further require the stability condition of the total energy ($\langle H \rangle + \mu N$) with respect to the total lattice number N , where μ corresponds to the charge-transfer energy or the electron-hole pair creation energy. In the limit of vanishing k_i 's, it was found¹¹ for $\mu \lesssim 1$ eV that the localized charges whose charge distribution is of a stripe configuration can be realized. Even in the case of nonvanishing k_i 's, it is also expected that the localized charges of a stripe configuration be realized, although the allowed region of μ may be further restricted due to the nonvanishing k_i 's.

In the latter half of the section, we discuss that the smallness of k_{Cu} is characteristic to the layered cuprates. If the c -direction Cu phonon is (nearly) a soft mode [the soft-mode phonon, in itself, is frequently observed in LSCO,^{32–35} TBCCO,^{36,37} and BSCCO (Ref. 37)], the c -direction force constant k_{Cu} can be roughly estimated, as in the case of BaTiO_3 , using the dipole interaction as $k_{\text{Cu}} \sim -2n_{\text{O}}(e_{\text{Cu}}e_{\text{O}}/r_{\text{Cu},\text{O}}^3)$, where n_{O} represents the number of the apical oxygen (in Tl-, Bi-, and Y-based cuprates, $n_{\text{O}} = 1$, while in La-based one $n_{\text{O}} = 2$). Actually, this relation is reasonably in agreement with the experimental value of k_{Cu} for $e_{\text{Cu}}e_{\text{O}} \sim e^2$. Hence it is found that $k_{\text{Cu}}/k_{\text{Ti}} \sim (n_{\text{O}}/2)(e_{\text{Cu}}/e_{\text{Ti}})(r_{\text{Ti},\text{O}}/r_{\text{Cu},\text{O}})^3$, where the two values of k_{O} are assumed to be almost equal to each other, for simplicity. In Y-, Bi-, and Tl-based cuprates ($n_{\text{O}} = 1$), the smallness of k_{Cu} originates from the small number of the apical oxygen. This at the same time is expected to induce the z displacement of the Cu ion, with the result of the elongation of $r_{\text{Cu},\text{O}}$, as is practically shown in Table I. Even in La-based cuprate ($n_{\text{O}} = 2$), $r_{\text{Cu},\text{O}}$ is still longer than $r_{\text{Ti},\text{O}}$ ($\approx 2 \text{\AA}$). This is because the La ion, located just above or below the apical oxygen of the CuO_6 octahedron, is surrounded by the pyramidal oxygen, not the octahedral oxygen, so that the La ion is expected to shift easily in the c -axis direction, which may further induce the elongation of $r_{\text{Cu},\text{O}}$. Thus the crystal structure that the Cu ions on the CuO_2 plane (or other cations lying in the same c -axis direction as the Cu ions, such as La ions in LSCO) are surrounded by the pyramidal oxygens may be the cause of longer $r_{\text{Cu},\text{O}}$ than the cation-oxygen distance $r_{\text{B},\text{O}}$ in a typical perovskite crystal ABO_3 , where the B ion is surrounded by octahedral oxygens.

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APPENDIX A: DERIVATION OF EQ. (3)

From the stability condition of $\delta\langle H\rangle/\delta\sigma_i(x,y)=0$, H can be written as

$$H=H(\sigma_i=0)+\frac{1}{2}\sum_i\int\frac{\sigma_i(x,y)\delta(z)n(\mathbf{x}')}{|\mathbf{x}-\mathbf{x}'|}d^3\mathbf{x}d^3\mathbf{x}', \quad (\text{A1})$$

where

$$H(\sigma_i=0)=-\sum_{\alpha=\pm 1/2}\int\psi_{\alpha}^{\dagger}(\mathbf{x})\nabla^2\psi_{\alpha}(\mathbf{x})d^3\mathbf{x}+\frac{1}{2}\int:n(\mathbf{x})U(\mathbf{x};\mathbf{x}')n(\mathbf{x}')d^3\mathbf{x}d^3\mathbf{x}'$$

and $\sum_i\sigma_i(x,y)$ is given by

$$(2\pi)^2\sum_i\sigma_i(x,y)=-\int\langle n(\mathbf{x}')\rangle\frac{e^{i[k(x'-x)+l(y'-y)]-|z'|\sqrt{k^2+l^2}}}{1+\lambda\sqrt{k^2+l^2}}dkdl d^3\mathbf{x}', \quad (\text{A2})$$

with $1/\lambda=2\pi\sum_j(1/\kappa_j)$. Substituting Eq. (A2) into the second term in the right-hand side of Eq. (A1) and integrating over \mathbf{x} , we obtain

$$\sum_i\int\frac{\sigma_i(x,y)\delta(z)}{|\mathbf{x}-\mathbf{x}'|}d^3\mathbf{x}=-\int\langle n(\mathbf{x})\rangle G_{\lambda}(\mathbf{x}_{\pm}-\mathbf{x}')d^3\mathbf{x}, \quad (\text{A3})$$

where $\mathbf{x}_{\pm}=(x,y,\pm|z|)$ and $G_{\lambda}(x,y,z)$ is given by

$$G_{\lambda}(x,y,z)=\frac{1}{2\pi}\int\frac{\exp(ikx+ily-z\sqrt{k^2+l^2})}{\sqrt{k^2+l^2}(1+\lambda\sqrt{k^2+l^2})}dkdl$$

$$=\frac{1}{1-\lambda\frac{\partial}{\partial z}}G_0(x,y,z)=\frac{1}{1-\lambda\frac{\partial}{\partial z}}\frac{1}{\sqrt{x^2+y^2+z^2}}. \quad (\text{A4})$$

APPENDIX B: DERIVATION OF EQ. (5)

Using the x representation of the state $|\phi\rangle$ as $\phi(\mathbf{x})=\prod_{i=1}^3[1/\sqrt{(2\pi)^{1/2}\Delta_i}]\exp(-x_i^2/4\Delta_i^2)$ with $\Delta_1=\Delta_2$, we find that $\langle-\nabla^2\rangle$ is calculated as $\langle-\nabla^2\rangle=-\int\phi^*(\mathbf{x})\nabla^2\phi(\mathbf{x})d^3\mathbf{x}=\frac{1}{4}\sum_{i=1}^3(1/\Delta_i^2)$. In a similar way, $\langle U'\rangle$ can be written as

$$\langle U'\rangle=\int|\phi(\mathbf{x})|^2U'(\mathbf{x};\mathbf{x}')|\phi(\mathbf{x}')|^2d^3\mathbf{x}d^3\mathbf{x}'$$

$$=\int_0^{\infty}\frac{1}{1+\lambda r}\exp(-\tilde{\Delta}^2r^2)\text{erfc}^2\left(\frac{\Delta_3}{\sqrt{2}}r\right)dr, \quad (\text{B1})$$

where $\tilde{\Delta}^2=\Delta_1^2-\Delta_3^2$, and we have used for $U'(\mathbf{x};\mathbf{x}')$ the integral representation as $U'(\mathbf{x};\mathbf{x}')=\int_0^{\infty}[1/(1$

$+\lambda r)]\exp(-\zeta r)J_0(r\sqrt{\xi^2+\eta^2})dr$, with $\xi=x-x'$, $\eta=y-y'$, $\zeta=|z|+|z'|$, and J_0 the Bessel function of the first kind. In the limiting case of $\lambda\gg\Delta_i$, $\langle U'\rangle$ can be decomposed into $\langle U'\rangle=U'_1+(1/\lambda)U'_2$, where

$$U'_1=\int_0^{r_0}\frac{1}{1+\lambda r}dr, \quad (\text{B2})$$

$$U'_2=\int_{r_0}^{\infty}\frac{1}{r}\exp(-\tilde{\Delta}^2r^2)\text{erfc}^2\left(\frac{\Delta_3}{\sqrt{2}}r\right)dr,$$

with r_0 satisfying the relation of $1/\lambda\ll r_0\ll 1/\Delta_3$. In calculating U'_2 , change the variable r as $(\Delta_3/\sqrt{2})r\rightarrow r$ and introduce the function $f(r)$ as $f(r)=\text{erfc}^2(r)\exp(-2\beta r^2)$ with $\beta=(\Delta_1/\Delta_3)^2-1$, then we obtain

$$U'_2=\int_{r_0\Delta_3/\sqrt{2}}^{\infty}(\ln r)'f(r)dr$$

$$=-\ln\frac{r_0\Delta_3}{\sqrt{2}}\cdot f\left(\frac{r_0\Delta_3}{\sqrt{2}}\right)-\int_{r_0\Delta_3/\sqrt{2}}^{\infty}\ln r\cdot f'(r)dr$$

$$\approx -\ln\frac{r_0\Delta_3}{\sqrt{2}}\cdot f(0)-\int_0^{\infty}\ln r\cdot f'(r)dr, \quad (\text{B3})$$

where we have assumed that $\beta\geq 1$. As a result, $\langle U'\rangle$ can be expressed as

$$\langle U'\rangle\approx\frac{1}{\lambda}\left[\ln\frac{\sqrt{2}\lambda}{\Delta_3}-\int_0^{\infty}\ln r\cdot f'(r)dr\right] \quad (\text{for } \lambda\gg\Delta_i), \quad (\text{B4})$$

which is indeed independent of the choice of r_0 such that $1/\lambda\ll r_0\ll 1/\Delta_3$.

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