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Electronic molecules in semiconductors and other novel materials

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Semiconductors are usually characterized by a concentration of impurities and defects. The electronic properties of these defects have a strong influence on the quality of devices made from these materials. Here it is demonstrated that under certain conditions there may arise electronic molecules, which behave similarly to quantum defects. The electronic molecules or e-molecules are formed through the formation of a confinement potential, which may trap one or more electrons. The electronic confinement arises either due to a lattice deformation or due to the formation of spontaneous magnetic topological defects or droplets. The shape and size of these e-molecules are arbitrary and depend on physical situations. In many cases they have linear form and one may therefore call them strings. The strings may also be closed into loops as happens with e-molecules in anti-ferromagnetic insulators. Some situations are reviewed in which e-molecules may arise and their properties are discussed. Relevant experiments are also discussed briefly.

1. Introduction

It was Lev Landau who, exactly 70 years ago, first suggested that electrons may be trapped by a lattice polarization [1]. The self-trapping of the electron by a polarization cloud created by this electron gave rise to a new quasi-particle named the polaron. During the 70 years after the first work by Landau [1] the polaron has been investigated thoroughly both theoretically and experimentally. The Landau idea has been generalized for all types of materials, liquids and solids, including insulators, semiconductors and metals. It was established that the polaron may have a small radius, of the order of the inter-atomic spacing of the lattice, or may have a very large size, much more than the single lattice spacing of a solid. The self-trapping confinement may be created not only by a lattice polarization but also by all types of lattice deformation. The first theory of large and small polarons was developed by a Russian school in the 1950s [2 – 4]. The large polarons introduced through Pekar's important contributions are often called Pekar polarons. Fröhlich developed Pekar's ideas to include weak and strong coupling with phonons. To unite the description of these two cases he derived a Hamiltonian now termed the Frölich Hamiltonian [5]. The proper theory of small polarons was developed by Holstein

and his collaborators in the 1960s [6]. Sir Nevill Mott proposed a bubble model of a polaron in a liquid metal-ammonia solution [7]. In this model an electron is self-trapped by a vacuum cavity, created by itself. Similar self-trapping of an electron in a vacuum cavity is observed also in liquid helium (see, more detail in review [8]) and is referred to as an electron bubble. Negative ions in helium also form similar bubbles [9]. They have been studied experimentally in detail [10,11]. In an electric field these (electron and ion) negatively charged bubbles are moving and their mobility can be measured. In particular it was found that the mobility of negative ions is substantially higher than normal electron bubbles [10,11].

In liquids of rare gases a free exciton is also self-trapped in a similar 'vacuum cavity' and it is referred to as an exciton bubble (see, more detail in the review [12]). The exciton is a neutral quasi-particle consisting of an electron and a hole in the bound state. In all such cases the electron or the exciton is self-trapped by the vacuum cavity due to the negative work function of the electron relative to the vacuum. Such self-trapped excitons may also arise in rare gas solids. If they arise in solids, then the local lattice deformation around the exciton will be so strong that there will be created lattice defects (interstitials or vacancies) and therefore, the creation of the self-trapped excitons may lead to the defect creation. Such an exciton mechanism for the defect creation in solids has been proposed in [13] (see, also,

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for details, [14]) and later observed in many experiments (see, for example, [15–23]). In these experiments the position of excitonic luminescence lines has been investigated. The creation of crystal defects is indicated by the red shift in the luminescence line, which is related to formation of the vacancy on which the exciton is localized. Such defects may be formed at the different stages of the exciton formation: either at the thermalization-relaxation stage or at the later stages of the self-trapping process.

It was De Gennes who first assumed that a single hole may change the orientation of magnetic moments, creating a canted state in an anti-ferromagnetic solid [24]. Nagaev then introduced the concept of a magnetic polaron, where an electron is self-trapped by a ferro-magnetic droplet [25]. This work was closely followed by similar papers from Kosuya [26] and Krivoglaz [27].

The single particle polaron states arise usually in ionic solids and they have been investigated in detail, mostly with the use of transport measurements [28,29]. The polarons having typically large effective mass are less effective in supporting electric current than free electrons. Therefore, the electric resistivity in materials with polarons is very large and the mobility of current carriers is very small, similar to the small measured mobility of electrons trapped in vacuum bubbles in liquid helium. Another important consequence of polarons is that the optical conductivity has no Drude peak [29].

The physical picture of the self-trapping with two particles or, in general, with many particles is far from complete. The idea of forming a two-particle polaron state, named the bi-polaron has attracted the attention of many people (see, [30] and references therein). This was especially stimulated by the possibility of the bipolarons, being bosons, to form a Bose–Einstein condensate with a consequence to form a superfluid or superconducting state. The formation of electronic clusters consisting of many electrons due to a deformation of the media has been known for a long time. For example, vacuum bubbles, like in the Mott polaron, but trapping of many electrons also arises in liquid helium and metal ammonia solutions [31,32]. In liquid helium these multi-electron bubbles have typically a radius of about $0.1–100$ μm and trap about $10^3–10^8$ electrons [31,32]. The bubble's diameter is determined by the balance between the surface tension of liquid helium and the Coulomb repulsion of electrons [33]. It is interesting that the electrons are not distributed throughout the volume of the bubble but form a nanometre thin and effectively 2D layer near the bubble surface [34].

Similar phenomena of multi-particle self-trapping may also arise in solids. However, their origination in solids is less transparent and the form of such clusters is very different. It is related to a new notion of electronic molecules (e-molecules) [35–40], which may arise in solids or fluids with strong or intermediate electron–phonon

interactions. The e-molecule consists of a few electrons bound by electron–phonon or by electron–electron interaction. The e-molecule is a generalization of the notion of a polaron or a self-trapping [1–14] to the many-body case, adapted originally to solids with narrow bands. It was shown that nearly all types of electron phonon interaction will lead to the creation of e-molecules, which may be created either in the ground state or in a metastable state and which have in many cases a linear, anisotropic shape. Due to their linear shape they have been named as strings [35–37]. The existence of the analogous multi-polaron instability and the consequential formation of electron strings in liquid polymers was originally discussed by Grigorov *et al.* [38–40] and named superpolarons. In this case the polymer molecules must have strong dipole moments. The superpolaron string is created due to a change of the orientation of the dipole-like polymer molecules around the electron string embedded in the polymer liquid. This reorientation of molecules creates a confinement potential in which many electrons are trapped.

The first formation of many-particle self-trapped states in magnetic semiconductors was also suggested by Nagaev and Krivoglaz [25,27]. They assumed that inside an anti-ferromagnetic solid a few electrons may polarize atomic magnetic moments and create a ferro-magnetic droplet, called a 'ferron' by Nagaev or a 'fluctuon' by Krivoglaz, which, in turn, will trap these electrons. These seminal works gave rise to the new fundamental concept of an electronic phase separation.

The studies of magnetic polarons, ferrons and fluctuons are related to a longstanding issue in the physics of doped antiferromagnetic solids, which still remains one of the most challenging issues of condensed matter physics. In some cases even a single hole added to the anti-ferromagnetic state may dramatically change the ground state from anti-ferromagnetic to ferromagnetic, creating a so-called Nagaoka state. More often, however, the hole changes the orientation of surrounding spins slightly, creating the canted state (DeGennes [24]). The hole may also completely polarize the surrounding spins only, creating a trapping potential in which the hole is self-trapped. Such a self-trapped hole is exactly the magnetic polaron or 'ferron', originally introduced by Nagaev *et al.* in magnetic semiconductors [25]. Condensation of many such magnetic polarons into droplets leads to the phenomenon electronic phase separation which, may arise in doped anti-ferromagnetic solids as well.

The discovery of novel oxide materials such as cuprates [41] and manganites (see [42–44] and references therein) has confirmed the existence of microscopic electronic phase separation in anti-ferromagnetic solids. Also these anti-ferromagnetic solids, well doped by holes, have displayed novel phenomena; high temperature superconductivity (in cuprates) [41,45] and colossal magneto-resistance (in

manganates) [42–44]. However, the origin of the super-conductivity and the colossal magneto-resistance arising in these materials remains unclear [44,45].

In the present paper we will show that in anti-ferromagnetic solids besides magnetic polarons and ferrons, there may arise a new topological quantum defect, which consists of a domain wall forming a loop with a few self-trapped holes. The domain loop creates effectively a linear potential well, which may trap many particles. Each hole trapped by the domain loop may perform a free motion along the loop. In a transverse direction this linear potential may be smeared by spin fluctuations, allowing a limited transverse hole motion, too.

At present there is a growing body of experimental evidence [46–49] indicating the existence of complex inhomogeneous micro- and mesoscopic structures created in oxide materials. Such structures are typically connected to static or dynamic stripe phases, which were independently studied by many researchers [46–54]. These structures have a natural explanation in the framework of the general notion of e-molecules considering the stripes made of the e-molecules. Recent experiments have also discovered the huge influence of isotope effect on the critical temperature of the stripe ordering [55] and strong lattice fluctuations in YBCO [56] which may be associated with the dynamics of the strings or e-molecules. With the isotope changes [55] the structure of individual strings is also changed (for example, the strings may become shorter due to a change in phonon frequency) and, therefore, the critical temperature of the stripe ordering must change. This indicates that the e-molecules and, in particular, strings, are a pertinent notion for oxides and, possibly, also for other solids. Here we estimate the size and the shape of the e-molecules for a few simple situations that may arise in semi-conducting and insulating materials. We show that under certain conditions ordinary electrons may form these special e-molecules, i.e. bound configurations within solids. These e-molecules arise due to a competition between an effective electron–electron attraction mediated by phonons and the usual Coulomb repulsion between electrons.

The remaining sections of the paper are organized as follows. In the section 2 we discuss the simplest model of the polaron, which is the electron or the exciton bubble. In section 3 we introduce magnetic polarons. In section 4 we introduce a simple example of many particle self-trapping, a string or an e-molecule, which arises due to local lattice distortions associated with the Jahn–Teller effect. Formation of the string due to other types of lattice distortion is discussed in section 5. Formation of e-molecules in doped antiferromagnetic solids is discussed in section 6. Finally in section 7 we discuss the possible implication of the novel ideas of e-molecules described in previous sections to the

physics different materials such as cuprates, manganites and others oxides.

2. Electron or exciton bubbles: e-molecules in metal–ammonia solutions, helium and in rare gas liquids and solids

An electron ejected into the liquid helium creates a special state, so-called an electron bubble. A similar electron bubble arises in metal-ammonia solutions, where it is called a Mott polaron.

The point is that the work function of this electron from bulk helium into the vacuum is negative, and approximately equal to 1 eV. To gain this energy the electron, therefore, expels the helium atoms around and creates a vacuum bubble of radius R in which this electron is self-trapped.

As a first approximation the energy of this self-trapped electron may be estimated as the following sum:

$$E = E_{e1} + E_{\text{surface}} + W \qquad (1)$$

where E_{el} is the work function of the electron into the vacuum plus the kinetic energy of the trapped electron, E_{surface} is the surface energy of the bubble, and W is the work performed to create a bubble against the external pressure $dW = p\,dV$.

If we measure the electron energy compared to the vacuum and assume that the bubble is inside an infinitely high potential well then the value E_{el} is equal to

$$E_{\text{el}} = \frac{h^2}{8\,m\,R^2} \qquad (2)$$

where m is the electron mass and h is Planck's constant. For a spherical bubble the surface energy has the standard form as $E_{\text{surface}} = \sigma\,S$ where S is the surface area of the bubble and σ is its surface tension, i.e. $E_{\text{surface}} = \sigma\,4\,\pi\,R^2$.

Finally the work against the external pressure p needed to create the bubble is $W = p\,V$, i.e. $W = p\,4\,\pi\,R^3/3$. If we neglect the external pressure and put $p = 0$, then the bubble radius can be found by the minimization of total energy, and is determined from the equation

$$\frac{\partial E_{\text{total}}}{\partial R} = 0 = \frac{-h^2}{4\,m\,R^3} + \sigma 8\pi R \qquad (3)$$

The solution of this equation gives the radius R_0 of the spherical bubble containing an electron in the ground state as

$$R_0 = \sqrt[4]{\frac{h^2}{32\pi\sigma m}} \qquad (4)$$

At this time there is an issue which is intensively discussed in the literature, namely, what is the structure of the excited state within such an electron bubble? The issue is related to the possible change of symmetry or to the question of the

existence and stability of a possibly dumbbell shaped electron bubble [57–59].

The considered model is equally applicable to a description of exciton self-trapping, i.e. the exciton bubble that is usually formed in helium and in rare gas liquids and solids irradiated by synchrotron radiation or by fast electrons. Of course for the estimate of the radius of the exciton bubble we have to substitute into equation (4) not the electron mass, but the mass of the exciton, equal to the sum of the electron and hole masses. It is obvious that due to large exciton mass (here the hole mass is very heavy) the radius of the exciton bubble will be much smaller than the radius of the electron bubble. It is typically of the order of the interatomic distance (see, for detail, [13,14]). In solids the formation of such exciton bubble will lead to defect creation, or, in general, to the aging of materials.

3. Magnetic polarons

A free electron on an anti-ferromagnetic lattice may create a special state, called a magnetic polaron [25]. Similar to the electron bubble discussed in the previous section, the magnetic polaron may be viewed as a magnetic bubble. The point is that a free electron on an anti-ferromagnetic lattice with a large exchange interaction J between magnetic moments has a very low mobility or, practically, is localized. However, when the electron polarization spins around on the magnetic lattice, it creates a ferro-magnetic region or a ferron bubble of radius R (figure 1). This electron is very mobile inside the bubble but is prevented from going outside and is therefore self-trapped. The energy of the magnetic polaron consists of the kinetic electron energy E_{el} (see, equation (2)), and the loss of exchange energy due to the created ferromagnetic bubble $E_{ex} = J 4 \pi R^3/3$. The total energy is then $E_m = E_{el} + E_{ex}$. The polaron radius is found from the equation obtained by minimization of the total energy E_m using
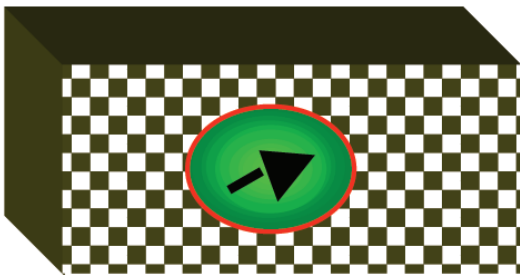

Figure 1. Single electron bubble (green) in liquid helium and also a ferron (green) arising in antiferromagnetic (chessboard) background of magnetic semiconductors.

$$\frac{dE_m}{dR} = 0 = \frac{-h^2}{4\,m\,R^3} + J4\pi R^2 \qquad (5)$$

The solution of this equation gives the radius of the magnetic polaron R_0:

$$R_0 = \sqrt[5]{\frac{h^2}{16\pi Jm}} \qquad (6)$$

One sees a striking similarity between magnetic polarons and electron bubbles in helium or in metal–ammonia solutions. The exchange constant in magnetic semiconductors plays the role of an external pressure in liquid helium. The bubbles discussed here (sections 2 and 3) are spherically symmetric because we have neglected the discrete details of the lattice and band structures of the materials. But most semiconductors have degenerate valence bands. For the self-trapping of holes, this band degeneracy leads to the phenomenon of spontaneous symmetry breaking. For example, if we take into account the effect of this band degeneracy on the formation of magnetic or other polarons, we will find that the shapes of these polarons will be dramatically changed. The polarons will then have cigar or disc shapes (see, for example, [60,61]). From this consideration, we also see that when the values of m or J increase, the polaron radius decreases. But the polaron size cannot be smaller than atomic size. This is exactly when the polaron of small radius arises.

We continue to discuss polarons having small radius.

4. Strings arising due to Jahn–Teller deformations

There are some crystal lattices, which consist of Jahn–Teller ions. Jahn–Teller ions are usually ions originally associated with degenerated electronic spectra. However, this degeneracy is broken due to the Jahn–Teller effect [62]. The essence of the Jahn–Teller effect is to remove degeneracy in the electronic spectrum as a result of a decrease in local symmetry arising due to distortions of the crystal lattice. Before symmetry breaking the Jahn–Teller ions are typically in the centre of some high symmetry object, as in cuprates where each copper atom is the Jahn–Teller ion located at the centre of a oxygen octahedron configuration CuO_6. For the Jahn–Teller ions the electron energy levels that lead to the formation of the band in the crystal are degenerate. Therefore, due to the Jahn–Teller effect the symmetry must decrease, i.e. associated Jahn–Teller lattice distortions arise which decrease the symmetry of the oxygen octahedron. The two types of lattice distortion may cause these octahedra to become prolate (elongated) or oblate leading to a chessboard lattice configuration [63,64]. These distortions also lead to the formation of Jahn–Teller polarons and to an effective polaron–polaron attraction. In such Jahn–Teller solids a

large variety of different types of electronic molecules may arise. Here, we consider a simple model of Jahn–Teller distortions, which allows us to perform exact analytic estimations of the binding energy of different electronic molecules. The realistic Hamiltonian consists of

$$H_{el} = -t \sum_{<ij>} a_i^+ a_j + \sum_{<ij>} \omega(q) b_q^+ b_q + hc + H_{el-ph} + H_{el-el}$$

$$(7)$$

where the electron–phonon Hamiltonian is defined as:

$$H_{el-ph} = \sum_{iq} n_i g_q [\exp(iqR_i) b_q + hc], \qquad (8)$$

where a_i and b_q are fermionic and bosonic annihilation operators associated with electrons and phonons, with the following commutation relations, $a_i^+ a_j + a_j a_i^+ = \delta_{ij}$ and $b_q^+ b_p - b_p b_q^+ = \delta_{pq}$ for electrons and phonons, respectively. The electron–electron interaction is defined as

$$H_{el-el} = \sum_{i<j} \frac{n_i n_j}{\varepsilon_\infty \mid R_i - R_j \mid} \qquad (9)$$

with $n_i = a_i^+ a_i$ a fermionic occupation number operator. Using a standard procedure to shift the phonon variables as

$$b_q \Rightarrow b_q - \sum_{iq} n_i g_q \exp(iqR_i)/\omega(q) \qquad (10)$$

we obtain, instead of a plain Coulomb potential ($\sim 1/(\varepsilon R)$), an effective electron–electron interaction in the form:

$$V(R_i - R_j) = \frac{1}{\varepsilon_0 \mid R_i - R_j \mid} - \frac{1}{N} \sum_q g_q^2 \cos(q \mid R_i - R_j \mid)/\omega(q)$$

$$(11)$$

The same expression may be obtained using the Lang–Firsov transformation [65] or by use of a coherent states method [66]. The essence of Lang–Firsov transformation is the change in the vacuum or a canonical shift of the qth harmonic of the field made with the use of the unitary transformation:

$$U(p_k, q_k) = \exp(b_k^+ d_k - b_k d_k^*)$$

where the value d_k characterizes the shift in the vacuum or change in the average value of lattice distortions as well as in the amplitude of lattice vibrations. Depending on the type of electron–phonon interaction the form of this effective potential $V(R_i - R_j)$ may be different. For an electron interacting with Jahn–Teller phonons, or with quantized Jahn–Teller lattice distortions [63,64], using the formula for the effective potential $V(R_i - R_j)$, (equation (11)) we obtain the potential for the inter-polaron interac-

tion $U(R) = V(R)$, presented in figure 2. It consists of a short-range (inter-site) effective attraction (producing the deep minimum in figure 2) and a long-range electron–electron Coulomb repulsion:

$$U(R \geq \sqrt{2}) = \frac{h\sqrt{2}}{R}$$

where the distance R is measured in units of the lattice spacing a. The strength of the Coulomb repulsion depends on the screening due to lattice polarization, and is characterized by the height of the Coulomb peak,

$$h = \frac{e^2}{\varepsilon \cdot a\sqrt{2}}$$

(figure 2), where ε is a static dielectric constant of the solid. The depth of the minimum, d, depends on the strength of the Jahn–Teller lattice distortions or on the value of vacuum shift in Lang–Firsov transformations [65]. The stronger the distortion, and the larger the CuO_6 octahedral elongation, the deeper the minimum. For strong coupling of electrons with Jahn–Teller lattice distortions, the polarons are small (the size of a single atom) but have very large masses. In the limit of very narrow electron bandwidth $t \Rightarrow 0$, the polaron band collapses [12,65] and, therefore, the kinetic energy of polarons may be neglected. Electronic molecules are then formed due to the effect of the inter-polaron interaction. The heavy polarons then fall into the deep minimum of the potential $U(R)$ (figure 2) forming an electronic molecule or e-molecule.
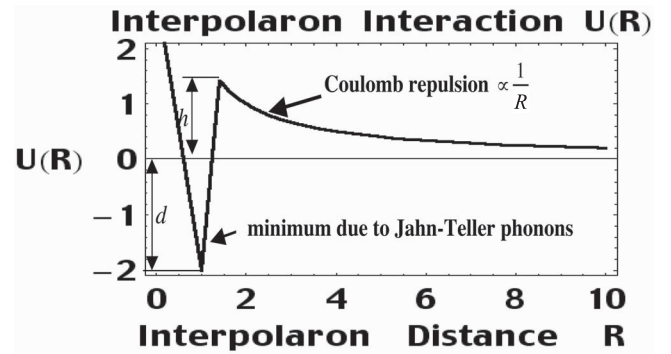

Figure 2. Potential of the interpolaron interaction in oxide materials, which consists of a short-range attraction (producing the sharp deep minimum) associated with the Jahn–Teller lattice distortions, and a long-range Coulomb repulsion (producing the peak and the shallow slope from the right side of the peak). The depth of the minimum is given by $d \sim K Q^2$, where Q is a local strain. The minimum is located at $R = 1$. The height of the Coulomb peak is given by $h = \frac{e^2}{\varepsilon \cdot a\sqrt{2}}$. The peak is located at $R = \sqrt{2}$. All distances are measured in units of one lattice spacing a.

To illustrate the main principals involved in the formation of e-molecules, let us consider for simplicity a square crystal lattice with polarons located on these lattice sites. We neglect the change of lattice symmetry and a formation of the checkerboard pattern due to two types of Jahn – Teller lattice distortions. This will not change the main principles of e-molecules formation. Conditions for the formation of these molecules are completely defined by the parameters of the potential of the inter-polaron interaction $U(R)$: d and h. Any multi-electron molecule can be considered in terms of the interaction of all possible pairs of electrons, which make up the molecule. Each pair interacts via a potential $U(R)$. Due to the very non-monotonic behaviour of the function $U(R)$ (compare with interatomic potentials, such as one formed due to the Van der Waals interaction) the energy contribution to the binding energy from each pair may be positive or negative. This contribution depends on the distance between the particles in a given pair. If the pair of particles is localized on neighbouring sites, i.e. located in the minimum of $U(R)$, they gain energy equal to the value d. If the distance between two given particles in the electronic complex is larger than the lattice spacing a, then these two particles repel each other, and the contribution from this pair of the e-molecule to the binding energy is negative. That is, due to such repulsion the binding energy of the chosen e-molecule decreases by an amount $\sim c\,h$, where the constant c is associated with the sum of inverse distances of all pairs in the molecule where the electrons are not located on neighbouring sites. In this simplest model there is a very general expression for the binding energy per particle of any multi-electron molecule. For an e-molecule consisting of n particles this expression has the following form:

$$E_b = (kd - c\,h\sqrt{2})/n \qquad (12)$$

Here k is the number of pairs of adjacent electrons, where the two electrons in the pair are located on neighbouring sites in this n-electron molecule, and $c = 1/x_1 + 1/x_2 + \ldots$ is the sum of inverse distances $x_i > 1$ (measured in units of a) for all pairs for which the electrons in each pair are farther away from each other than a single lattice spacing a.

Let us consider examples of simplest e-molecules on the square lattice. If there are only two particles located on neighbouring sites, then they are localized in the deep minimum of $U(R)$ and, therefore, are bound into a two-electron molecule (a bi-polaron) with binding energy per particle $E_b = d/2 \equiv -U(1)/2$ (figure 3). Here the parameter values are $m = 1$, $c = 0$ and $n = 2$. A three-electron molecule may also exist. The three-particle molecule may be considered as three pairs. Two of these pairs can be located on neighbouring sites. The size of the third pair is always larger than the lattice spacing a. Therefore, due to Coulomb repulsion $U(R > \sqrt{2})$, the contribution to the

binding energy of this three-electron molecule arising from this third pair is always negative. The binding energy per particle is maximal if the distance between these two particles is the largest value possible (provided that the inter-particle distance in other pairs of the three electron molecule is equal to the lattice spacing a). There is only one such configuration and this is the linear molecule (the string configuration [29 – 31], see figure 3). The binding energy per particle of such a string is equal to $E_b = (2d - h/\sqrt{2})/3$. This is consistent with equation (12) when the parameter values are $k = 2$, $c = 1/2$ and $n = 3$. From a comparison of this binding energy with the binding energy of the bi-polaron one may find that the bi-polaron has a lower binding energy per particle than the three-electron string if $d < h\sqrt{2}$.

Four polarons may create a four-electron molecule whose optimal shape depends on the relation between the height of the Coulomb peak h and the depth of the phonon minimum d. Using equation (12) one may readily estimate the binding energy for the square electronic molecule (figure 3) as $E_{\text{square}} = (4d - 2h)/4$; for the 'zig-zag' molecules as $E_{\text{zig-zag}} = (3d - (2 + \sqrt{2}/\sqrt{5})h)/4$; and finally for the string molecule as $E_{\text{string}} = (3d - 4\sqrt{2}\,h/3)/4$. From these estimates it is clear that the 'zig-zag' molecule always has a binding energy smaller than either the square molecule or the string molecule. The string and the square molecules are associated with local minima, which are separated by a barrier associated with other shapes of the molecule, which is the 'zig-zag' and 'corner' shapes. Therefore these electronic molecules, the string and the square, may coexist with each other. In a ground state the molecule has a square shape if $d > (2 - 4\sqrt{2/3})h$. A comparison of the binding energy of the square electronic molecule with the binding energy of a bi-polaron shows that the square molecule has a lower binding energy than a bi-polaron if $d > h$.

When the screening is strong, the Coulomb repulsion is weak, $d > > h$, and the optimal shape is a square. With increase of the Coulomb repulsion or a decrease in the dielectric constant, the electronic molecule may form a string or it may decay directly into bi-polarons. In the framework of the simple model chosen we may estimate the optimal shape of e-molecules on a square lattice. Depending on the strength of the electron – phonon interaction and the Coulomb screening (or the ratio d/h), the optimal molecules (i.e. having the lowest energy) are the two-particle ones, or dumbell bi-polarons (see the green dumbbell in figure 3) and four-particle molecules having the shape of the square lattice plaquette (see the green square in figure 3). The optimal shape for a many electron molecule is a droplet having the form a dense lattice of squares (see, for example, figure 4).

In general, the form and the number of electrons in electronic molecules may be arbitrary and is largely determined by the overall interaction consisting of the
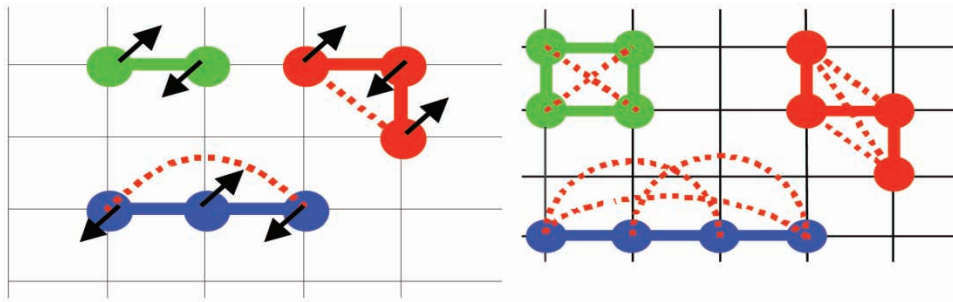
Figure 3. Possible shapes of e-molecules on a square lattice. The bi-polaron has the dumbbell shape. The lowest energy state of the three electron molecule is the string. The bold solid lines indicate interpolaron attraction. The dashed lines indicate Coulomb repulsion, which gives a negative contribution to the binding energy of electronic molecules.
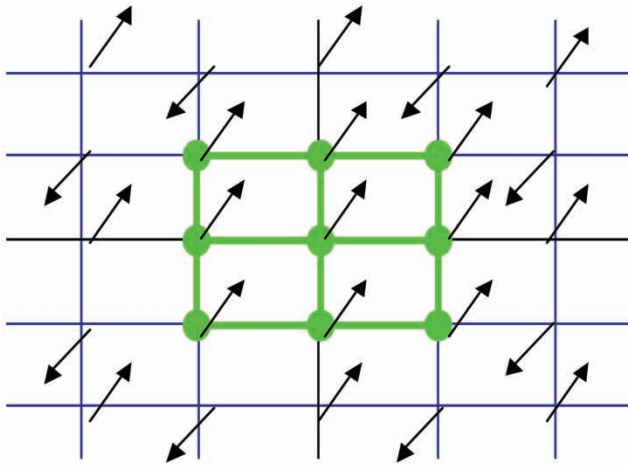

Figure 4. Typical optimal form of the multi-electron complex consisting of nine electrons on the square lattice. The green square may also represent a magnetic polaron or ferron, where the green atoms are polarized magnetic moments; only one or two electrons are trapped in the green magnetic droplet area.

effective (phonon mediated) electron–electron attraction at short distances, and the form of Coulomb repulsion at long distances. The electron screening effects may fundamentally change the shape and size of these molecules. If there is Debye screening, then the long-range Coulomb repulsion takes the form $U(R > \sqrt{2}) = \frac{h\sqrt{2}}{R} \exp(-R/R_D)$, where R_D is the Debye radius. In this case the optimal shape may have the form of a string. Moreover at any finite value of the Debye radius R_D there exists instability resulting in the formation of stripes. If the electron–phonon coupling (or the ratio d/h) increases to some critical value, there arise strings which are then ordered into stripes. For example, if the Debye radius $R_D = 2$, then this instability arises if $d > h\sqrt{2}$. In order for stripes to be formed, the electron–phonon interaction must be strong, the dielectric constant ε must be large, and the Debye radius must be small. The larger the Debye radius R_D, the stronger the electron–

phonon coupling has to be for the formation of electronic stripes.

5. Strings arising due to other lattice deformations

It has been shown above that, in the presence of a strong Jahn–Teller electron lattice interaction, the electrons have a tendency to create e-molecules and strings. Each string consists of M charged particles that are self-trapped by local lattice deformation and polarization in a linear array of N lattice sites. In the range of physical parameters relevant to the doped perovskites such as cuprates and maganites these strings may be formed due to other (non-Jahn–Teller) lattice distortions, for example, just like plain stress and strain and these strings may have lower energy than isolated polarons [4–31]. It was recently demonstrated that due to interaction induced by the lattice distortions (by a plain strain) arising around point-like impurities, they may be self-organized into a stripe-like pattern [67].

Now let us estimate the characteristic size of the strings which may be created by a local strain. In fact, a non-uniform nanoworld of sign-varying textures in strain and charge has been revealed in many complex electronic materials [68]. Charge doping acts as a local stress that deforms the lattice without generating defects and producing polaronic elasto-magnetic textures or nanoscale phase separation associated with the formation of strings.

In the very low density limit we can consider a system of non-interacting strings where in each string the elastic deformation Q is proportional to the number of trapped charges M. The elastic energy of the lattice is proportional to $KQ^2 \sim KM^2$; here K is an elastic modulus. The electron energy of the self-trapped particles, and therefore the lattice adiabatic potential of the string state, decreases as

$$E_e = -E_p M^2$$

where E_p is the energy for trapping a single charged particle. This localization energy is opposed by the

Coulomb repulsion between particles trapped by the string potential well for which the electrostatic energy has an additional factor log M (in a model of linear charged string). So the Coulomb energy is approximately equal to

$$E_C = VM^2 \log M$$

where V is a constant of the inter-site inter-electron Coulomb repulsion. A balance between these energies which is determined by a minimization of the total energy gives a stationary many particle self-trapped string state where the string length is equal to [35,53]:

$$N \sim M \sim \exp(E_p/V) \tag{13}$$

Let us estimate the length of these strings in the perovskite materials. If we take the elastic modules of the order of $c_{11} = 11 \times 10^{10}$ erg cm^{-3}, the inter-atomic distance is $a = 4$ Å, and we get $K \sim 4.1$ eV. If the deformation potential is approximated as $D \sim e^2/a = 3.4$ eV, then for the electron–phonon coupling we obtain $E_p = D^2/K = 2.5$eV. Taking a dielectric constant of $\varepsilon = 5$ for a doped system, we get $V = e^2/\varepsilon a = 0.68$ eV. This gives an estimate for the length of the string of the order of 10 inter-atomic distances.

6. Loops of e-molecules in anti-ferromagnetic insulators

Let us consider a 2D anti-ferromagnetic (AF) state on a square lattice. In the ground state, all spins are anti-ferromagnetically ordered creating a chessboard plane, as in figure 1. The low energy excitations of AF are spin waves. Besides spin waves there are topological excitations like spin-vortices and domain walls. In the 2D AF the domain walls are linear defects. They may originate from the boundary, for example, due to a twist in the boundary conditions. The microscopic domain forming a loop (figure 5) may be nucleated inside of the AF by thermal fluctuations. Such an event is very rare due to the small probability $\propto \exp\left(-\frac{JL}{kT}\right)$ associated with large loop energy.

The energy of this loop depends on its perimeter and it is equal to $E_{dl} = J \cdot L$, where L is the loop perimeter measured in units of the lattice spacing a and J is an exchange constant of the AF. The loop energy decreases when the loop size decreases, therefore such a loop is unstable and its size should decrease until the loop eventually vanishes. The situation may be drastically changed when we will dope the AF by holes. The motion of a single hole is usually limited and accompanied by the breaking of AF bonds. Therefore the hole is attracted to an initial position where there are no extra broken bonds except for the initial four bonds missing due to the presence of the hole (therefore, the energy lost due to a single hole is ~ 4 J). However, the hole located near the domain wall will have a lower energy (~ 2J) than a hole

inside the well-ordered AF state (~ 4 J). This hole is mobile along the domain wall. In the direction transverse to the domain wall line, the hole is localized (figure 6).

The transverse localization length is of the order of a few inter-atomic spacings. The hole performs a relatively free motion along the whole loop perimeter. With such motion at the start of each lap, one kink and one anti-kink must be created (see the schematic structure of the kink anti-kink pair on figure 6). These kinks and anti-kinks will disappear at the end of each lap, and then be created at the start of the
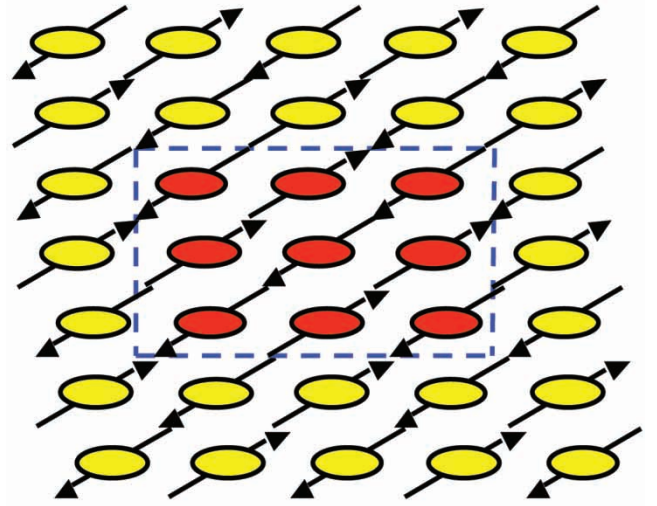

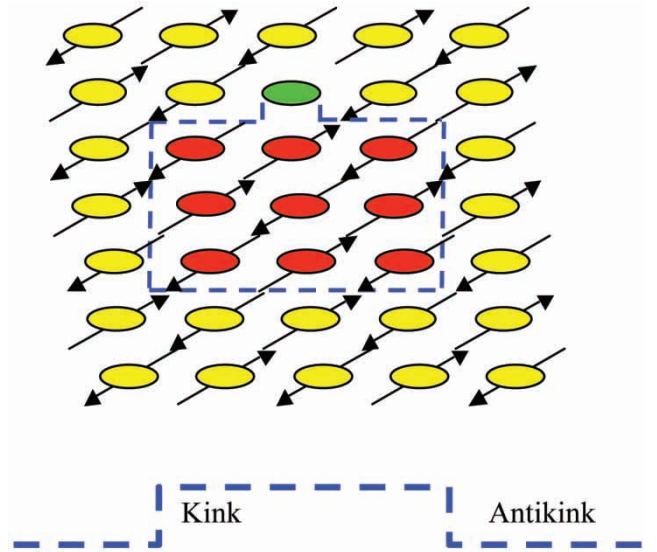**Figure 5. Domain wall forming a square loop sketched as the blue line.**

Figure 6. Domain wall loop with a single hole self-trapped by the loop.

next lap and, for the hole in motion, the process of kink–anti-kink creation and annihilation will be repeated.

The domain loop with the trapped hole located on the kink may correspond to the ground state energy of the doped AF. It may also exist in a metastable state if it is locally stable. Indeed, if the kinetic energy of the single hole with the kink is

$$E_k = \frac{h^2}{2mL^2a^2}$$

then the total energy of the loop with the single hole will be

$$E = \frac{h^2}{2mL^2a^2} + JL - J$$

This expression has a minimum when

$$L = \sqrt[3]{\frac{h^2}{ma^2J}}$$

The loop energy is then given by

$$E_{gr} = \frac{3}{2}\sqrt[3]{\frac{h^2J^2}{ma^2}} - J$$

Estimating on the basis of a model similar to that discussed in section 2, but for the AF on the square lattice, we found that the energy of the Nagaev ferron with square shape (see the green droplet shape on figure 4) is

$$E_p = 2\sqrt{\frac{h^2J}{2ma^2}}$$

Depending on the value J or m this energy may be higher or lower than the energy of the domain loop with trapped hole on the square lattice. This balance also depends strongly on the contribution from the hole motion transverse to the domain wall. Therefore, although the domain loop with the trapped hole is a locally stable state, the ground state of a single hole will be either this loop or the Nagaev magnetic polaron (see section 3). The energy of the transverse motion decreases even more when more than one hole is trapped by the loop.

The evolution of the domain wall is completely determined by the creation of kink and anti-kink pairs as well as by the number of holes trapped by the loop. The loop size may decrease or increase depending mostly on kink–anti-kink generation and on the motion of the next doped hole and on the number of trapped holes. To create a single such loop requires a lot of energy. It is the self-trapping of holes that makes this generation possible. A loop created without holes usually decays via step-by-step generation of kink–anti-kink pairs and their subsequent motion. On the other hand when a hole is attached to a domain wall, the size of the domain wall is stabilized. The collapse of the domain wall will increase the kinetic energy

of the trapped holes. In fact, the most optimal energy situation is when the loop traps many holes. Then each hole may be a constant source of kink–anti-kink pairs arising at the start of each even or odd lap when the particular hole moves around the loop. The scattering of the holes on the kinks decreases the life time of the hole quasi-particles.

Considering those holes sitting on the kinks as free spinless fermions we may estimate the kinetic energy of these M holes trapped by the loop as

$$E_K = \frac{h^2k_F{}^2}{2m}$$

For motion of the hole along the domain wall we may consider a one-dimensional band with Fermi momentum

$$k_F = 2\pi\frac{M}{aL}$$

To be precise, we have to consider the motion of the hole along the sides and around the corners, scattering on the kinks, and any transverse motion. However to a first approximation, to make some simple energy estimates at the conceptual level, let us neglect these contributions, assuming that all motion of the trapped hole is coherent and one-dimensional.

The exchange energy needed to create a domain loop with M trapped holes is equal to $J\cdot(L–M)$. The energy associated with Coulomb repulsion between holes we estimate in electrostatic approximation assuming that the holes are equidistantly distributed along the domain loop (see figure 7). Then the Coulomb energy for $M = 4$ holes is given by

$$E_l = \frac{2\varepsilon_l}{L}, \text{where } \varepsilon_l = (2 + 4\sqrt{2})e^2/(\varepsilon_0 a).$$

and the total energy per single hole is

$$E = \frac{h^2 4^2}{2ma^2L^2} + \frac{JL}{4} + \frac{2\varepsilon_l}{L} - J \tag{14}$$

One sees that this expression has an absolute minimum with respect to L. Minimization with respect to L gives the optimal size of this domain wall loop. If the parameter ε_0 is large, as in the case of metallic oxides, the last term may be neglected. Then the size of the loop is

$$L = 4\sqrt[3]{\frac{h^2}{ma^2J}}$$

From this equation we see that for many-particle self-trapping (here, the value is $M = 4$) the size of the loop increases by $M = 4$ times, while the loop energy per single
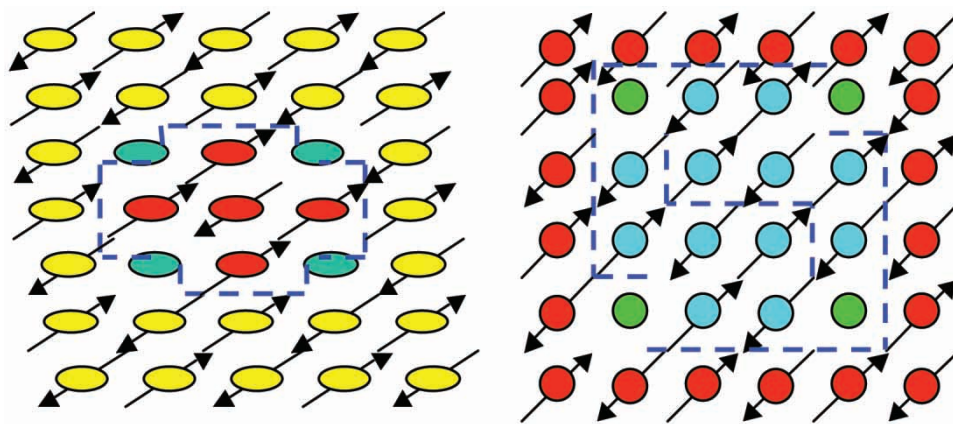
Figure 7. Domain wall loops with four holes self-trapped by the loop.

hole remains the same: $E(M = 4) \equiv E(M = 1)$. This indicates the strong degeneracy of the loop states, a degeneracy which we have already noticed in our consideration of different string states. Thus, with the Coulomb energy taken into account, the energies of e-molecules associated with different numbers of particles are very close to each other. Therefore all types of such clusters with 2, 3, and, in general, any number of particles, may co-exist with each other (see figure 7).

With increasing doping, the number of loops in the anti-ferromagnetic insulator increases and a percolation may occur. The loops will coalesce and intersect each other, creating a net. With further increase of the doping the net may be ordered into a stripe phase. Our finding of these new elementary excitations (a loop of the domain wall with trapped holes) may explain many puzzles existing in the physics of under-doped cuprates, such as a metallic conductivity, enormously strong Hall effect of the under-doped cuprates at high temperatures, photo-emission data, origin of the pseudo-gap as a localization of holes in these loops, time reversal symmetry breaking as a persistent current in these loops and many others, which have been observed in numerous experiments and which are primarily based on investigation of spin textures with the use of Nuclear Magnetic Resonance (NMR) and muon Spin Rotation (μSR) measurements (see, for example, [69] and references therein). The NMR and μSR studies of spin and hole texture in $La_{2-x}Sr_xCuO_4$ in the doping range $0 < x < 0.12$ have shown that the static and dynamic properties of this compound are intimately related to the generation of domain walls associated with the micro-segregation of holes. In addition, the electron–phonon interaction helps to stabilize these loops.

Note that in this section we have discussed the formation of e-molecules (or hole segregation) due to a magnetic interaction only, while in real systems in most cases there is a strong electron–phonon interaction present.

7. Summary, visible consequences and speculations

Thus, for metallic and magnetic oxides (cuprates, nickelates and manganites) with high values of dielectric constant, screening and electron–phonon coupling are strong. Consequently, e-molecules (strings, loops and others) arise in these materials and are very important in explaining many exotic properties, such as microscopic electronic phase separation and the formation of stripes [35–57].

Indeed, the concept of e-molecules yields fundamentally new insights into a wide range of materials. The creation of such molecules in perovskite manganites would account for the microscopic phase separation observed recently in $La_{5/8-y}Pr_yCa_{3/8}MnO_3$ [43] and many other materials (for details, see, the review [70]). In these experimental works it was clearly concluded that there was percolative transport through submicrometre-scale two-phase mixtures. This could be associated with the types of electronic molecules responsible for the colossal magnetoresistance behaviour observed in these materials, where the e-molecules may be distributed in a disordered way or ordered into the static charged stripes observed, for example, in [42–44]. Thus the phenomenon of the formation of these static charged stripes can be related to the ordering of e-molecules or, more precisely, to the stripe instability associated with the formation of e-molecules.

Detailed comparisons of the theories for multi-electron self-trapping with experimental data obtained for a broad range of manganite materials have been made in [71]. There, measurements of the temperature dependence of resistivity at different values of magnetic field have been analysed. It was concluded that the theory of multi-electron self-trapping or the formation of magnetic droplets is consistent with existing experimental data. The magnetic droplets are similar to the single polaron bubble discussed in section 3 and have a diameter of about 7–8 times the lattice constant, taken as $d = 3.9$ Å for all types of material.

Each magnetic droplet must contain from 50 to 100 particles, which can be electrons or holes depending on the type of material under consideration.

Due to the fact that electronic masses are smaller than atomic masses the electronic molecules have much smaller masses than atomic molecules and in solids they may therefore be very mobile. Also due to their small masses they are very much quantum objects. Therefore, the shape of these e-molecules fluctuates and changes over time. The existence of these quantum molecules in cuprate oxides, like YBCO, LaCuO and others, results in the formation of dynamical stripes or string glass formations as discussed extensively in [46–57]. It is probable that the quantum fluctuational dynamics associated with the quantum nature of electronic molecules gives rise to the mysterious dynamics of the stripes observed in these materials. Reference [72] gives an extensive analysis of experimental data for high-temperature superconducting cuprates. In [72], experimental results from X-ray absorption fine structure (XAFS), inelastic neutron scattering (INS), Raman spectra, infrared absorption spectra and time-resolved femtosecond spectra were analysed. Their analysis supports the scenario in which the pseudogap-associated temperature T^* is related to the formation of 'polaronic filamentaries' (strings), which were described in this paper, and were originally predicted in [35–37]. At temperatures $T < T^*$ there is a visible change in the optical spectra (~ 1.5 eV), which is very close to the estimate made in section 5 for the value of the confining potential well $E_p \sim 2.5$ eV (the polaron shift).

This result also supports the suggestion that the origin of high-temperature superconductivity in doped cuprates may also be related to the strings, namely, to the vibronic modes associated with these strings [73]. The polaronic tunnelling frequency deduced in [72] associated with the tunnelling transition in a double well potential is nothing but the frequency of electronic vibrons introduced in [73] (the longitudinal stretching vibrational mode of the strings, which may naturally lead to tunnelling in a double well potential discussed in [72]). This frequency has anomalous behaviour in the vicinity of the critical temperature of the superconducting phase transition (see, figure 2 in [72]), indicative of an intimate relationship between polaronic strings and the origin of superconductivity in these compounds. Due to the small electronic masses the vibronic modes of these e-molecules have higher frequencies (electrons move faster than atoms) than in normal molecules and therefore they may play an important role in the Cooper pairing mechanism. Due to the high frequencies of such modes, Cooper pairing may be more efficient and therefore the resulting superconducting critical temperature could be very high.

Recently, it was shown [74] that the stability of e-molecules in the sea of free fermions increases provided that the fermion density decreases and becomes smaller than some critical value, which is smaller than in conventional metals. Indeed, in the underdoped cuprates the holes density is lower than in conventional metals and the polaronic strings will be stable there. They will also remain stable with decreasing temperature when the compound becomes superconducting. The coexistence of charge carriers and associated polaronic distortions with superconductivity are implied in several recent experiments [75]. On the other hand, at such transitions into the superconducting state, strings will also have an opposite, parasitic influence on high-temperature superconductivity. Strings are charged objects and, therefore, they create an inhomogeneous potential in which the superconducting order parameter is embedded. This will obviously lead to a modulation of the order parameter, and the superconducting gap will form a mesoscopic and nanoscopic structure that will be associated with the shape, size and distribution of these e-molecules, which are probably polaronic strings. For example, in the local area of the string the gap will be smaller, i.e. partially suppressed, while in the area between the strings the gap will remain nearly the same or it will be even larger. Indeed such modulations of the superconducting gap have recently been observed in superconducting $Bi_2Sr_2CaCu_2O_{8+\delta}$ compound (Bi-2212) [76,77] using high resolution scanning tunnelling microscopy (STM) measurements. Moreover, by applying the Fourier transform to the atomic-scale spatial modulations in Bi-2212 density of states they obtained elements of the Fermi-surface and energy gap in agreement with photoemission experiments [78–80]. They found consistency between numerous sets of dispersing modulations and the quasiparticle interference model and that the momentum-space structure of the unoccupied states is similar to that of the occupied states. This shows that no additional order parameter besides the superconducting one is required and that the copper oxide quasiparticles are similar to Landau particle–hole quasiparticles, which exist in conventional superconductors. This is completely consistent with the co-existence of e-molecules and free fermions. Again in complete agreement with theoretical results presented in this and previous papers, they found that near the energy gap maximum, the modulations become strong and commensurate with the crystal, and that they are bounded by nanometre-scale domains, which, presumably, are associated with the strings. All these results support the scenario of a nearly conventional superconductor arising due to the existence of strings dissolved in Fermi liquid [73]. Thus, the e-molecules may play an important role in the origin of the high-temperature superconductivity observed in cuprates [73].

It is also important to note that the existence of strings is more favourable for low-dimensional systems, such as quasi-two-dimensional or quasi-one-dimensional solids. The point is that in this case, suppression of the kinetic

energy of electron and holes in the transverse direction with respect to the orientation of the string, needed for string confinement, is more easily reached. Therefore, strings must occur in materials having, for example, a chain structure. Indeed such types of compound exist among cuprates and are known as YBCO (or, more precisely, $YBa_2Cu_3O_{6+x}$). Its crystal structure consists of alternating planes having quasi-two-dimensional and quasi-one-dimensional character. Each quasi-one-dimensional plane consists of parallel Cu–O chains. Thus, if strings exist in this material they will first arise on the Cu–O chains, because there the kinetic energy loss arising at the formation of strings is minimal. Indeed, such strings have been seen in recent scanning tunnelling spectroscopy measurements of the Cu–O chain plane in YBCO, which show a 25 meV gap in the local density of states (LDOS) filled by numerous intragap peaks (resonances) [81].

The LDOS gives information about the energy spectrum and about the spatial distribution of the localized states. The intense peaks in LDOS spectra could be associated with strings formed on these chains. The strings here can be considered as localized objects, which are formed from 'small' polarons. To be consistent with the experiment we must assume that these polarons have small or intermediate radius. The experiment [81,82] indicates that the effective size of such polarons is equal to the wavelength of the string modulation, $\lambda = 1.4$ nm. The number of particles of which the string consists or the length of the strings varies.

In previous papers [35–37] we have indicated that the strings are associated with a very complicated energy landscape, which consists of many very closely (in energy space) located energy minima. These minima correspond to strings having different sizes or consisting of different numbers of lowest energy states corresponding to the most popular string, of size equal to the observed correlation length $l_0 = 4$ nm. This string consists of $l_0/\lambda \cong 3$ particles. Because the energy minima are very close to each other there will exist also strings having different sizes. In the experiment [81] the conductance associated with tunnelling of a single electron to and from the string was measured. According to the Franck–Condon principle during such a tunnelling event the electronic structure of the string will not be changed and all relaxation processes will start after the tunnelling event. Therefore the string, if it exists, will be completely visualized in the measurements of the LDOS presented in [81,82].

However, in this experiment string-like resonances were seen not only in the low energy region of the gap but all around the gap. The primary reason for this is the existence of vibronic modes or quantized vibration of the strings. We also have to stress that the observed resonances may be associated not only with the strings in the ground state but also with vibronic satellites. On each string there are vibronic excitations: these vibrons may accompany each tunnelling event; i.e. at the electron or hole tunneling from the STM tip to the sample there will be one or two vibrons emitted or absorbed. Therefore, besides the main resonances associated with the string in the ground state, the measurements [81] should show single vibron or, slightly less probable, two vibron satellites. Of course the intensity of the satellite peaks will be significantly lower than the intensity of the main peaks. A similar phenomenon of satellite resonances is well known in exciton physics. Many exciton absorption spectra have optical phonon satellites similar to the vibron satellites that can arise during measurement of the LDOS. Optical phonon satellites could have arisen during the measurements of LDOS made in [81,82].

There is another reason for displacement of the resonance energy position. Due to the fact that oxygen vacancies are mostly localized in the Cu–O chains they are very inhomogeneous. Therefore, in addition to the self-organized potential, strings are located in the random potential associated with these vacancies. Due to such extra potential the position of the ground state energy of the string as well as the string-vibron satellites may be shifted both up and down the energy scale.

Now let us estimate the energy of the vibron arising on a string following [73]. Knowing the polaron diameter $2R = 1.4$ nm one may estimate the polaron shift as $E_p = Ry/R^2 \sim 70$ meV. Then with the use of the polaron shift the energy of the longitudinal stretching vibron will be given by $\omega = E_p/N_0$, where N_0 is the number of particle in the string. With the use of this estimate we find that the most optimal strings, consisting of three particles in the vibron satellite peak, should arise at the energy 23 meV higher than the position of the main resonance. For four-particle strings, satellites will be separated by vibron energy equal to 17 meV; for five-particle strings the satellites arise at an energy equal to 14 meV, and so on. It is important also to note that we have made the estimation only for one type of vibron, and that there are other types of vibron that are able to make a contribution to LDOS. However, to make more precise estimates of the vibronic energies we have to use more sophisticated models. Thus we may conclude that the multiple set of resonances observed in [81,82] may be ascribed to the tunnelling conductance through the strings arising on Cu–O chains and their vibronic and optical phonon satellites.

The intensity of the peaks of these resonances enables separation of the main and satellite resonances. The differential conductance associated with the satellite peaks should be significantly weaker. The strings and their satellites may be visualized in animations [82] derived from the experimental measurements of LDOS. A complete set of spectroscopic maps obtained at high spatial resolution for different window sizes has been presented. Each frame

is the conductance at a fixed energy (written in green in the upper corner of the movie frame). The field of view is 25 nm, 50 nm and 100 nm in size for the three animations, respectively. All three animations were taken at the same location on the surface of $YBa_2Cu_3O_{6+x}$. The data show the same phenomena arising inside the gap (from about -25 mV to $+25$ mV): there are localized regions with large conductance and with sharply defined energies. There is space–energy correlation of these regions, which is apparent as the animation progresses. The participation of strings together with their vibronic excitations and optical phonons in the differential tunnelling conductance is very natural and therefore strings are primary candidates for an explanation of these amazing experimental observations of different resonances in LDOS [81,82].

There is another type of string, which was directly observed in luminescence spectra in different quasi-one-dimensional materials [83–85]. These strings are created by the illumination of light and are called exciton strings. There can be just a few or many excitons self-trapped along the chains of different organic materials. The exciton is a neutral object and, therefore, Coulomb repulsion plays a less important role in the structure of such strings. The exciton strings may play a very important role in structural photoinduced phase transitions, creating, effectively, nucleons of a new phase [83–85]. The nucleation of excitonic strings has also been seen in cryocrystals, in particular in solid Xe [86,87]. There the excitons are self-trapped on three atomic complexes.

Finally, the finding of domain loop excitation with self-trapped holes is fundamentally important, because this completes the picture of elementary excitations in doped anti-ferromagnetic solids. With the use of these e-molecule loops one can describe how the anti-ferromagnetic state may disappear with increasing doping, and how a metal-insulator transition may arise.

Acknowledgements

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