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Dispersion relations and density of states of the collective oscillations of the electronic dipoles in high-T_c superconductors

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Previously we have suggested the collective oscillations of the crystal-field-generated electronic dipoles on oxygen ions as a possible excitation responsible for the electron-electron (or hole-hole) pairing mechanism in high-T_c superconductivity. These self-sustained collective dipole excitations are expected to exist in the high-T_c superconductors. We have investigated these excitations by performing first-principles calculations of the dispersion relations and the density of states associated with these excitations in several high-T_c superconductors (La_2CuO_4, $YBa_2Cu_3O_{7-\delta}$, $Bi_2Sr_2CuO_6$, $Bi_2Sr_2CaCu_2O_8$, $Tl_2Ba_2CaCu_2O_8$, and $Tl_2Ba_2Ca_2Cu_3O_{10}$) including the cases of a single dipole and those of a linear and a planar system of dipoles in a uniform field as illustrative examples. The absence of inversion symmetry at the oxygen sites and the high polarizability of the atoms such as oxygens are found to be necessary for the existence of these quantized collective dipole excitations. Our calculations show good consistency between the existence of superconductivity and the presence of such collective dipole oscillations in the conducting planes of the high-T_c superconductor crystals, as expected by our earlier explanation of the variation in T_c versus δ in $YBa_2Cu_3O_{7-\delta}$ by the same mechanism. The associated frequencies characteristic of these excitations in the conducting planes in all high-T_c superconductors are found to be of the order of 10^{14} Hz as predicted previously. Furthermore, the variation of these characteristic excitation frequencies in $YBa_2Cu_3O_{7-\delta}$ as a function of the oxygen-stoichiometry parameter δ has been found to agree with our previous deductions. The density of states of these collective dipole excitations show broader bandwidth, unlike the case of excitons. The present mechanism is likely to be a good candidate to explain many observed features of the high-T_c superconductors, particularly the optical properties. For the $YBa_2Cu_3O_{7-\delta}$ system, our calculated density of states predicts two optical absorption peaks, one at 0.36 eV and another at 2.5 eV, which compare very well with the observed peaks close to 0.37 and 2.5 eV, respectively. Similarly for the $La_{2-x}Ba_xCuO_4$ system, our predicted peaks are at 0.17 and 1 eV, which are also in agreement with the observed peaks around 0.18 and 1.3 eV, respectively. The existence of similar peaks due to these collective dipole excitations in the other superconductors has been predicted. The optical absorption peaks at frequencies around 1×10^{14} Hz with broad bandwidth are expected to be observed in all high-T_c oxide superconductors. The possibility of explaining other important properties via this collective dipole mechanism, such as the pressure effect and the isotope effect observed in the high-T_c superconductors, has been discussed.

I. INTRODUCTION

In this paper we have made first-principles calculations of excitation frequencies of self-sustained collective oscillations (dipoles) of crystal-field-generated electronic dipoles in several high-T_c superconductors, and the associated dispersion relations and density of states. These oscillations are the same as those discussed previously.[1-3] The dipole-mediated electron-electron (or hole-hole) pairing mechanism[1-3] has been found to explain the observed variation in T_c as a function of the oxygen-stoichiometry parameter δ in $YBa_2Cu_3O_{7-\delta}$ superconductors. It is possible that this dipole-mediated mechanism is responsible for the occurrence of superconductivity in high-T_c superconductors and thus warrants special attention for understanding the range of frequencies, dispersion relations, and density of states of dipole excitations in these systems.

The structural properties, such as crystal symmetry, oxygen vacancies, and lattice distortions, play a crucial role in high-T_c superconductors. For instance, an undoped La_2CuO_4 crystal with no distortion does not show superconductivity unless the Sr (or Ba) atoms that substitute for the La atoms produce lattice distortions.[4] In such systems, experimentally one finds that the doping produces a periodic tilting of the CuO_2 planar structure, thereby producing periodic distortions.[4-6] In the pure $YBa_2Cu_3O_7$ (without vacancy) superconductor, there exists a lack of inversion symmetry, particularly at the oxygen sites in the CuO_2 planes. The oxygen deficiency in $YBa_2Cu_3O_{7-\delta}$ further affects its superconductive character.[1-3,7-11] The undistorted $Bi_2Sr_2CuO_6$ crystal shows no high-T_c superconductivity.[12,13] Crystals such as $Bi_2Sr_2CaCu_2O_8$, $Tl_2Ba_2CaCu_2O_8$, and their families[14-19] with multiple CuO_2 planes show high-T_c superconductive behavior similar to what one finds in pure $YBa_2Cu_3O_7$ (without vacancy). Another class of superconductor, $Ba_{0.6}K_{0.4}BiO_3$ (without Cu atoms), has a simple-cubic perovskite structure, which tends to show vacancy effects.[20] Furthermore, the recently discovered superconductor Nd_2CuO_4 requires either doping with tetravalent thorium (or cerium) atoms or the introduction of oxygen vacancies in order to exhibit superconductivity.[21,22] In brief, the distortion effects of the substituting atoms or vacancies have a direct bearing on the occurrence of superconductivity not only for the single-layered superconducting crystals such as Nd_2CuO_4, La_2CuO_4, $Bi_2Sr_2CuO_6$, $Tl_2Ba_2CuO_6$, $TlBa_2CuO_5$, etc., but also for the multilayered superconducting crystals such as $YBa_2Cu_3O_{7-\delta}$, $Bi_2Sr_2CaCu_2O_8$, $Tl_2Ba_2CaCu_2O_8$, $Tl_2Ba_2Ca_2Cu_3O_{10}$, etc.

The broken symmetry, lattice distortion, and oxygen vacancies lead to local polarization for highly polarizable ions. For instance, the absence of the inversion symmetry at an oxygen site in multilayered high-T_c superconductors causes the electronic polarization of oxygens. Our calculations show that the electronic distortions due to doping and vacancies in single-layered systems are smaller than those in multilayered systems, since in the multilayered systems the lack of inversion symmetry and thereby polarizations exist naturally even without dopants or vacancies. Also, the doping effects on the superconductivity are further complicated by the amount of vacancies and extent of substitutions. It is also well known that an oxygen ion (O^{2-}) is stable only in a crystal because of its high electronic polarizability. The high electronic polarizability implies that the valence electrons of the ions adjust easily by deformation confirming to the local symmetry to minimize the energy as a first dominant effect, which is to be compared against the next dominant effect arising from the displacement of the ions as a whole in the crystals as discussed earlier.[1] Similar concepts may easily be extended to cover other polarizable ions, such as S^{2-}, Cl^-, and F^-.

Once induced dipoles are present in a system due to crystalline electric fields, they oscillate in a self-sustained and collective manner as will be demonstrated by our present calculations. If some dynamic perturbations occur on the crystal-field-generated electronic dipoles due to thermally fluctuated electrons (or holes) or even due to the zero-point motions at absolute zero temperature, they change the dipoles with time, in general, in magnitude and direction and thereby provide the time-dependent changing electric fields on the other dipoles. These dipoles, influenced by the changing electric fields, in turn generate self-consistently the changing electric fields on the other dipoles, thus forming a system of dipoles oscillating collectively. In order to achieve long-range collective oscillations, one needs to have well-arranged dipoles on a periodic lattice, even with distortions or vacancies, together with a non-inversion-symmetry point at polarizable ions. Clearly, such electronic dipolar oscillations are different[1] from the case of phonons where the ionic vibrations are involved. We shall see in the following that the electronic dipolar oscillations are generated and self-sustained by the crystal fields naturally. In Sec. II we present the theory relevant to the self-consistent calculations of the crystal-field-generated electronic dipoles in a crystal and give results of our calculations in $YBa_2Cu_3O_{7-\delta}$, Bi-Ca-Sr-Cu-O, Tl-Ca-Ba-Cu-O, and La_2CuO_4 (undistorted and distorted due to substitutions). In Sec. III the theory and, wherever possible, calculations of the dispersion relations and density of states of these quantized collective dipole excitations are given for various cases: (i) single dipole in an electric field, (ii) dipole chain generated by a uniform electric field, (iii) planar system of dipoles with dipoles generated by a uniform electric field, (iv) $YBa_2Cu_3O_{7-\delta}$, (v) La_2CuO_4 (undistorted as well as distorted), and (vi) other systems such as $Bi_2Sr_2CuO_6$, $Ba_2Sr_2CaCu_2O_8$, $Tl_2Ba_2CaCu_2O_8$, and $Tl_2Ba_2Ca_2Cu_3O_{10}$. Their interaction with electrons are discussed in Sec. IV. Finally, the discussion and conclusion are given in Sec. V, with special emphasis on the explanation of the relevant observed optical absorption in various superconducting systems by these collective excitations and the possible predictions.

II. CRYSTAL-FIELD-GENERATED ELECTRONIC DIPOLES

We have found that the crystal fields that can induce dipoles on highly polarizable ions are present on the oxygen-ion sites in the high-T_c superconductors. We can determine the local electric field $\mathbf{E}_i$ acting on the ith ion as

$$\mathbf{E}_i = \mathbf{E}_i^m + \sum_j (\tilde{F}_{ij}) \cdot \mathbf{p}_j \ , \tag{1}$$

where $\mathbf{p}_j$ is the electronic dipole of the jth ion; $\mathbf{E}_i^m$ is the electric field at the ith ion produced by the monopoles (ionic charges) on the surrounding ions, and $(\tilde{F}_{ij}) \cdot \mathbf{p}_j$ represents the electric field at the ith site due to a dipole $\mathbf{p}_j$ at the jth site. $\mathbf{E}_i^m$, in general, is not zero where a sublattice or lattice point does not have inversion symmetry and expressed as

$$\mathbf{E}_i^m = \sum_k \frac{q_k^*}{r_{ik}^3} \mathbf{r}_{ik} \ , \tag{2}$$

where q_k^* is the (effective) charge (monopole) of the kth ion; $\mathbf{r}_{ik}$ is the position vector directed from the kth ion to the ith ion; $(\tilde{F}_{ij})$ are the Lorentz tensors,

$$(\tilde{F}_{ij}) = \begin{vmatrix} \dfrac{3x^2-r^2}{r^5} & \dfrac{3xy}{r^5} & \dfrac{3xz}{r^5} \\ \dfrac{3yx}{r^5} & \dfrac{3y^2-r^2}{r^5} & \dfrac{3yz}{r^5} \\ \dfrac{3zx}{r^5} & \dfrac{3zy}{r^5} & \dfrac{3z^2-r^2}{r^5} \end{vmatrix} \ , \tag{3}$$

where $\mathbf{r}$ stands for $\mathbf{r}_{ij}$ and x for x_{ij}, etc. The matrix elements of the Lorentz tensors are usually calculated under the assumption that the dipoles on the polarized ions act as point dipoles. This assumption is justified when the electronic overlap matrix elements between the neighboring ions are small. We note that the high-T_c superconductors with which we are concerned are, to a great extent, ionic in character as discussed previously,[1] particularly, in the case of $YBa_2Cu_3O_{7-\delta}$ by analyzing the sharp peaks in the band structure and the observed low carrier density. This also justifies that the screening effects are small as far as the calculations of the dipoles are concerned.

If the ith ion has an electronic polarizability α_i, the induced electric-dipole moment $\mathbf{p}_i$ due to the electric field $\mathbf{E}_i$ is

$$\mathbf{p}_i = \alpha_i \mathbf{E}_i \ . \tag{4}$$

From Eqs. (1) and (4), one obtains

$$\mathbf{p}_i = \alpha_i \left[\mathbf{E}_i^m + \sum_j (\tilde{F}_{ij}) \cdot \mathbf{p}_j \right] . \tag{5}$$

For the evaluation of $\mathbf{E}_i^m$ and the summations involving the Lorentz tensors $(\tilde{F}_{ij})$ one may utilize direct-lattice summation techniques.[24] For numerical calculations one correctly uses the value of the dipole at a sublattice in a unit cell the same as the dipole at an equivalent sublattice in another unit cell for a system with equivalent unit cells (as in periodic systems). This condition is valid even for thin films or polycrystalline high-T_c superconductors assuming a finite periodic domain for which the calculations are made.[23] In those cases one may replace the index i by $(l|\kappa)$, where l is an index for a unit cell and κ is a sublattice in a unit cell. With this notation the position vector of a sublattice site κ in the lth unit cell, measured from a given origin, is

$$\mathbf{r}(l|\kappa) = \mathbf{r}(l) + \mathbf{r}(\kappa) \ .$$

Thus, rewriting Eq. (5), we get

$$p_{\lambda\kappa} = \alpha_\kappa \left[E^m_{\lambda\kappa} + \sum_{\lambda'\kappa'} g_{\lambda\lambda'\kappa\kappa'} p_{\lambda'\kappa'} \right] , \tag{6}$$

where

$$g_{\lambda\lambda'\kappa\kappa'} = \sum_{l-l'} \left[\frac{3 r_\lambda r_{\lambda'} - r^2 \delta_{\lambda\lambda'}}{r^5} \right] ,$$

where λ describes the Cartesian components of the relevant quantities and

$$\mathbf{r} = \mathbf{r}(l|\kappa) - \mathbf{r}(l'|\kappa') \ .$$

Equation (6) gives a set of linear equations that can be solved self-consistently[24] to yield the values of the λth dipole components at the sublattice sites κ, namely, $p_{\lambda\kappa}$. For the calculations of the lattice sums we find that the convergent lattice sums for our system can be obtained if one takes the contribution from the sites lying within a radius of 70–300 Å.

A. Dipoles in $YBa_2Cu_3O_{7-\delta}$

The dipoles on high-T_c superconductor $YBa_2Cu_3O_{7-\delta}$ depend strongly on the oxygen-deficiency parameter δ. Since there are 14 sublattices in a unit cell of $YBa_2Cu_3O_{7-\delta}$, we will have, in general, 42 linear equations [Eq. (6)] to solve simultaneously to obtain dipoles in the system.

Our results of the self-consistent calculations of the dipoles in $YBa_2Cu_3O_7$ (without vacancies) have been summarized in Table I, where we list the various sublattice atoms[7] in a unit cell of $YBa_2Cu_3O_7$ along with the corresponding Pauling's ionic polarizabilities,[25] the possible effective ionic charges q_κ^*, the coordinates $x_\kappa, y_\kappa, z_\kappa$, the sublattice occupancy factor f_κ, the calculated values of the electric fields $E_{z\kappa}^m$ due to monopoles and the calculated dipoles $p_{z\kappa}$. The electric-field components due to dipoles at various sublattices have not been given as they are too many to be listed here. The listed $p_{z\kappa}$ are the self-consistent solutions of Eq. (6) containing the effects of monopoles as well as dipoles in the system. One notes that the directions of the static dipoles and the electric fields are only along the $\hat{\mathbf{z}}$ direction due to the crystal symmetry where the x,y,z axes are taken along the crystallographic a,b,c axes, respectively. The total polariza-

TABLE I. List of sublattice atoms, Pauling's polarizabilities α_κ (in units of Å^3), effective charge of ions q_κ^* (in units of e), the sublattice coordinates x_κ, y_κ, and z_κ (in units of a, b,, and c, respectively), sublattice occupancies f_κ, the calculated electric fields $E_{z\kappa}^m$ (in units of $e^2/\text{Å}^2$) at the sublattice sites due to the ionic charges and the calculated electronic dipoles $p_{z\kappa}$ (in units of e Å) at various sites in $YBa_2Cu_3O_7$. Lattice constants (Ref. 7) used for the calculations are $a = 3.823$ Å, $b = 3.886$ Å, and $c = 11.680$ Å.

Sublattice atom (κ)	α_κ	q_κ^*	x_κ	y_κ	z_κ	f_κ	$E_{z\kappa}^m$	$p_{z\kappa}$
O(4)	3.88	−2	0.0	0.0	0.8416	1	−0.526	−1.384
Cu(2)	0.1	2	0.0	0.0	0.6444	1	−0.027	−0.025
Cu(2)	0.1	2	0.0	0.0	0.3556	1	0.027	0.025
O(4)	3.88	−2	0.0	0.0	0.1584	1	0.526	1.384
Cu(1)	0.1	3	0.0	0.0	0.0	1	0.0	0.0
O(3)[a]	3.88	−2	0.0	0.5	0.6211	1	0.076	0.187
O(3)[a]	3.88	−2	0.0	0.5	0.3789	1	−0.076	−0.187
O(2)[a]	3.88	−2	0.5	0.0	0.6227	1	0.054	0.046
O(2)[a]	3.88	−2	0.5	0.0	0.3773	1	−0.054	−0.046
O(1)	3.88	−2	0.0	0.5	0.0	1	0.0	0.0
Ba	1.55	2	0.5	0.5	0.8157	1	−0.214	0.202
Y	0.55	3	0.5	0.5	0.5	1	0.0	0.0
Ba	1.55	2	0.5	0.5	0.1843	1	0.214	−0.202
O(5)	3.88	−2	0.5	0.0	0.0	0	0.0	0.0

[a] Oxygen atoms in the CuO_2 plane.

TABLE II. Fractional occupancies f, the calculated dipoles p_z (in units of e Å) in $YBa_2Cu_3O_{7-\delta}$ system at various quenching temperatures (T_q) corresponding to different oxygen-stoichiometry parameters δ. The subscript κ designating the sublattice atoms in various quantities has been suppressed for simplicity. (Details of structural data are given in Ref. 11.)

	$T_q=605$ °C $\delta=0.29$		$T_q=650$ °C $\delta=0.4$		$T_q=705$ °C $\delta=0.46$		$T_q=755$ °C $\delta=0.56$		$T_q=775$ °C $\delta=0.54$		$T_q=805$ °C $\delta=0.55$	
Atom	f	p_z	f	p_z	f	p_z	f	p_z	f	p_z	f	p_z
O(1)	0.68	0.0	0.61	0.0	0.53	0.0	0.46	0.0	0.43	0.0	0.40	0.0
O(2)[a]	1.0	0.060	1.0	0.021	1.0	0.014	1.0	0.026	1.0	0.006	1.0	0.018
O(3)[a]	1.0	0.080	1.0	0.024	1.0	0.042	1.0	0.037	1.0	0.034	1.0	0.049
O(4)	1.99	1.632	1.97	1.727	1.96	1.758	1.91	1.792	1.92	1.829	1.93	1.826
O(5)	0.04	0.0	0.02	0.0	0.05	0.0	0.07	0.0	0.11	0.0	0.12	0.0

[a] Oxygen atoms in the CuO_2 plane.

tion of a unit cell is found to be zero, and the dipoles on the equivalent sites related by mirror symmetry in a unit cell are of opposite signs as expected. Thus the macroscopic ferroelectric character is absent in this system, consistent with experimental observations. The dipoles at O(4) sites are very strong compared with the dipoles at O(3) sites in the CuO_2 plane. The magnitude of the dipoles at an O(3) site is comparable to the molecular dipoles of water (0.4e Å, where e is the magnitude of the charge of an electron).

For the calculations of the dipoles in $YBa_2Cu_3O_{7-\delta}$ for $\delta \neq 0$ we have performed calculations for two different structural models simply because the exact vacancy distributions in this system are not yet known.

In the first case, we adopt the crystallographic structural data of $YBa_2Cu_3O_7$ as given by Jorgensen *et al.*[7] and change the fractional occupancies of the O(1) and O(5) sites in the Cu-O chains by means of $\delta = 1 - f_1 - f_2$, where f_1 and f_2 are the fractional occupancies of the O(1) and O(5) sites, respectively, which incorporates all the observed facts in Ref. 7. We found by our self-consistent calculations that the variation of dipoles at an O(3) site with respect to δ could be represented perfectly well by a straight line described by $p = p_0(1 - \delta/\delta_c)$ with $p_0 = 0.187e$ Å and $\delta_c = 0.77$, and the dipoles at an O(2) site are much smaller in magnitude compared with the

TABLE III. List of sublattice atoms (Ref. 14), Pauling's polarizabilities α (in units of Å^3), effective charges of ions q^* (in units of e), the sublattice coordinates x, y, and z (in units of a, b, and c, respectively), occupancies f, and the calculated electronic dipoles p_z (in units of e Å) in undistorted $Bi_2Sr_2CuO_6$. The subscript κ in various quantities has been suppressed for simplicity. Lattice constants used for calculations are $a = 3.8097$ Å, $b = 3.8097$ Å, and $c = 24.607$ Å.

Sublattice atom	α	q^*	x	y	z	f	p_z
Cu	0.1	2	0.0	0.0	0.0	1	0.0
O	3.88	−2	0.0	0.0	0.092	1	−0.763
Bi	0.5	3	0.0	0.0	0.183	1	−0.111
O	3.88	−2	0.0	0.0	0.313	1	1.057
Sr	0.86	2	0.0	0.0	0.428	1	0.138
Sr	0.86	2	0.0	0.0	0.572	1	−0.138
O	3.88	−2	0.0	0.0	0.687	1	−1.057
Bi	0.5	3	0.0	0.0	0.817	1	0.111
O	3.88	−2	0.0	0.0	0.908	1	0.763
O[a]	3.88	−2	0.0	0.5	0.0	1	0.0
O[a]	3.88	−2	0.0	0.5	0.5	1	0.0
O[a]	3.88	−2	0.5	0.0	0.0	1	0.0
O[a]	3.88	−2	0.5	0.0	0.5	1	0.0
Sr	0.86	3	0.5	0.5	0.072	1	0.010
O	3.88	−2	0.5	0.5	0.187	1	0.787
Bi	0.5	3	0.5	0.5	0.317	1	0.324
O	3.88	−2	0.5	0.5	0.408	1	1.920
Cu	0.1	2	0.5	0.5	0.5	1	0.0
O	3.88	−2	0.5	0.5	0.592	1	−1.920
Bi	0.5	3	0.5	0.5	0.683	1	−0.324
O	3.88	−2	0.5	0.5	0.813	1	−0.787
Sr	0.86	3	0.5	0.5	0.928	1	−0.010

[a] Oxygen atoms in the CuO_2 plane.

dipole at an O(3) site. Previously[1] we have used these results to explain the observed T_c as a function of δ in this system.

In the second case, we have taken the structural data as determined by Rietveld refinement of neutron powder diffraction data given by Kwok *et al.*[11] where the positions of Cu(2), O(2), O(3), O(4), and Ba, the fractional occupancies of oxygens at O(1), O(4), and O(5), and the lattice constants change as δ changes. The calculated values of the dipoles along with the fractional occupancies at oxygen sites are summarized in Table II for different δ values and the associated quenching temperatures T_q (in units of °C). The polarizabilities and possible ionic charges in $YBa_2Cu_3O_{7-\delta}$ system are taken to be the same as in Table I. From Table II it is evident that the dipoles at O(2) and O(3) sites in CuO_2 planes decrease in magnitude as δ increases with the exception that near $\delta=0.4$ the $p_{z\kappa}$ values do not change in a regular way, which may be due to the uncertainties in the assumed values of the structural data and in the ordering of the oxygen vacancies. In addition, one finds that the dipoles on O(2) and O(3) sites in the CuO_2 planes in this case are comparable in magnitude, which warrant further investigations as to how this changes the superconductive properties, particularly, the T_c values.

B. Dipoles in Bi-Ca-Sr-Cu-O and Tl-Ca-Ba-Cu-O systems

Another class of high-T_c superconductors has been discovered which constitutes the two families of compounds based on Bi-Ca-Sr-Cu-O (Refs. 14–16) and Tl-Ca-Ba-Cu-O (Refs. 17–19).

In pure undistorted (without lattice distortions, vacancies or impurities) single-layered crystals $Bi_2Sr_2CuO_6$,[14] $Tl_2Ba_2CuO_6$, and $TlBa_2CuO_5$,[19] the oxygen sites in the CuO_2 planes contain inversion symmetry,[13] and, consequently, there are no oxygen dipoles in the planes, as is also clear from our calculations (Table III) of the dipoles in one of these crystals, namely, $Bi_2Sr_2CuO_6$. Because of the absence of dipoles in the conducting planes of these systems, the superconductivity is not expected to occur due to the collective dipole mechanism. Indeed, these systems are not found to be high-T_c superconductors. On the other hand, the multilayered pure crystals

TABLE IV. List of sublattice atoms (Ref. 16), Pauling's polarizabilities α (in units of Å^3), effective charges of ions q^* (in units of e), the sublattice coordinates x, y, and z (in units of a, b, and c, respectively), occupancies f, and the calculated electronic dipoles p_z (in units of e Å) in $Bi_2Sr_2CaCu_2O_8$. The sublattice atom subscript κ has been suppressed in various quantities. Lattice constants used for calculations are $a=3.83$ Å, $b=3.83$ Å, and $c=30.89$ Å.

Sublattice atom	α	q^*	x	y	z	f	p_z
Ca	0.47	2	0.0	0.0	0.0	1	0.0
Sr	0.86	2	0.0	0.0	0.1091	1	−0.19
Bi	0.5	3	0.0	0.0	0.3011	1	0.17
O	3.88	−2	0.0	0.0	0.38	1	1.33
Cu	0.1	2	0.0	0.0	0.4457	1	0.0
Cu	0.1	2	0.0	0.0	0.5543	1	0.0
O	3.88	−2	0.0	0.0	0.62	1	−1.33
Bi	0.5	3	0.0	0.0	0.6989	1	−0.17
Sr	0.86	2	0.0	0.0	0.8909	1	0.19
O[a]	3.88	−2	0.0	0.5	0.051	1	−0.15
O	3.88	−2	0.0	0.0	0.198	1	−0.77
O[a]	3.88	−2	0.0	0.5	0.449	1	0.15
O[a]	3.88	−2	0.0	0.5	0.551	1	−0.15
O	3.88	−2	0.0	0.0	0.802	1	0.77
O[a]	3.88	−2	0.0	0.5	0.949	1	0.15
Cu	0.1	2	0.5	0.5	0.0543	1	0.0
O	3.88	−2	0.5	0.5	0.12	1	−1.33
Bi	0.5	3	0.5	0.5	0.1989	1	−0.17
O	3.88	−2	0.5	0.5	0.302	1	0.77
Sr	0.86	2	0.5	0.5	0.3909	1	0.19
Ca	0.47	2	0.5	0.5	0.5	1	0.0
Sr	0.86	2	0.5	0.5	0.6091	1	−0.19
O	3.88	−2	0.5	0.5	0.698	1	−0.77
Bi	0.5	3	0.5	0.5	0.8011	1	0.17
O	3.88	−2	0.5	0.5	0.88	1	1.33
Cu	0.1	2	0.5	0.5	0.9457	1	0.0
O[a]	3.88	−2	0.5	0.0	0.051	1	−0.15
O[a]	3.88	−2	0.5	0.0	0.449	1	0.15
O[a]	3.88	−2	0.5	0.0	0.551	1	−0.15
O[a]	3.88	−2	0.5	0.0	0.949	1	0.15

[a] Oxygen atoms in the CuO_2 plane.

$Bi_2Sr_2CaCu_2O_8$, $Tl_2Ba_2CaCu_2O_8$, $Bi_2Sr_2Ca_2Cu_3O_{10}$, and $Tl_2Ba_2Ca_2Cu_3O_{10}$ do not have inversion symmetry[15,16] at the oxygen sites in the CuO_2 planes and thereby possess induced dipoles. We have made self-consistent calculations of the induced dipoles in the systems $Bi_2Sr_2CaCu_2O_8$, $Tl_2Ba_2CaCu_2O_8$, and $Tl_2Ba_2Ca_2Cu_3O_{10}$, and the results are presented in Tables IV, V, and VI, respectively. Similar calculations for $Bi_2Sr_2Ca_2Cu_3O_{10}$ have not been made because of the lack of availability of the relevant crystal-structure data at present. The calculated dipoles in these systems are somewhat approximate because of the uncertainties in the available structural data. However, they are very informative as to the superconducting properties of these systems. Since these systems have induced dipoles in the conducting planes, they are expected to be the high-T_c superconductors under the present collective dipole mechanism, consistent with experimental observations. These substances form good examples to demonstrate the consistency between the presence of dipoles at the oxygen sites in the conducting planes and the existence of high-T_c superconductivity.

Though the undoped $Bi_2Sr_2CuO_6$ is not a high-T_c superconductor, the doping by atoms such as Ca makes it a high-T_c superconductor because the Ca dopants produce the lattice distortions and thereby generate (though small) dipoles at the oxygen sites in the CuO_2 planes, which are then responsible for the superconductivity found in the $Bi_2Sr_xCa_{1-x}CuO_6$ system according to the collective dipole mechanism. Since the magnitudes of the dipoles produced by the distortion due to impurity Ca atoms are small, the T_c values in these systems are expected to be low, which remains to be verified experimentally.

The high-T_c superconductor $Tl_2Ba_2Ca_2Cu_3O_{10}$ referred to above belongs to a triple-layered system and contains a high number, namely, 38, of sublattice atoms per unit cell. The positions of the sublattices determined by experiments are somewhat uncertain compared with the single-or double-layered crystals. For our calculations presented in Table VI we have assumed that the central

TABLE V. List of sublattice atoms (Ref. 17), Pauling's polarizabilities α (in units of Å^3), charges of ions q^* (in units of e), the sublattice coordinates x, y, and z (in units of a, b, and c, respectively), occupancies f, and the calculated electronic dipoles p_z (in units of e Å) in $Tl_2Ba_2CaCu_2O_8$ suppressing the subscript κ in various quantities. Lattice constants used for calculations are $a=3.855$ Å, $b=3.855$ Å, and $c=29.32$ Å.

Sublattice atom	α	q^*	x	y	z	f	p_z
Ca	0.47	2	0.0	0.0	0.0	1	0.
Ba	1.55	2	0.0	0.0	0.1218	1	−0.13
Tl	0.5	3	0.0	0.0	0.2864	1	−0.09
O	3.88	−2	0.0	0.0	0.3539	1	1.43
Cu	0.1	2	0.0	0.0	0.446	1	0.0
Cu	0.1	2	0.0	0.0	0.554	1	0.0
O	3.88	−2	0.0	0.0	0.6461	1	−1.43
Tl	0.5	3	0.0	0.0	0.7136	1	0.09
Ba	1.55	2	0.0	0.0	0.8782	1	0.13
O[a]	3.88	−2	0.0	0.5	0.0531	1	−0.09
O	3.88	−2	0.0	0.0	0.2185	1	−1.50
O[a]	3.88	−2	0.0	0.5	0.4469	1	0.09
O[a]	3.88	−2	0.0	0.5	0.5531	1	−0.09
O	3.88	−2	0.0	0.0	0.7815	1	1.50
O[a]	3.88	−2	0.0	0.5	0.9469	1	0.09
Cu	0.1	2	0.5	0.5	0.054	1	0.0
O	3.88	−2	0.5	0.5	0.1461	1	−1.43
Tl	0.5	3	0.5	0.5	0.2136	1	0.09
O	3.88	−2	0.5	0.5	0.2815	1	1.50
Ba	1.55	2	0.5	0.5	0.3782	1	0.13
Ca	0.47	2	0.5	0.5	0.5	1	0.0
Ba	1.55	2	0.5	0.5	0.6218	1	−0.13
O	3.88	−2	0.5	0.5	0.7185	1	−1.50
Tl	0.5	3	0.5	0.5	0.7864	1	−0.09
O	3.88	−2	0.5	0.5	0.8539	1	1.43
Cu	0.1	2	0.5	0.5	0.946	1	0.0
O[a]	3.88	−2	0.5	0.0	0.0531	1	−0.09
O[a]	3.88	−2	0.5	0.0	0.4469	1	0.09
O[a]	3.88	−2	0.5	0.0	0.5531	1	−0.09
O[a]	3.88	−2	0.5	0.0	0.9469	1	0.09

[a] Oxygen atoms in the CuO_2 plane.

layer (in the three consecutive CuO_2 layers) is the undistorted one, which, in fact, may not be true and requires further consideration in improved calculations.

C. Dipoles in La_2CuO_4

The calculated dipoles in an undistorted La_2CuO_4 crystal[5] (structure similar to tetragonal K_2NiF_4) are summarized in Table VII. Perusal of Table VII indicates that there are no dipoles present at the oxygen sites in the CuO_2 planes of this undistorted system. Thus, superconductivity is expected to be absent in this system, consistent with experimental observations.[5] On the other hand, experiments indicate that the superconductivity in $La_{2-x}Sr_xCuO_{4-\delta}$, $La_{2-x}Ba_xCuO_{4-\delta}$, $La_{2-x}Na_xCuO_{4-\delta}$, and $La_{2-x}Ca_xCuO_{4-\delta}$ occurs in their distorted K_2NiF_4 phase.[6] The partial substitution by Sr, Ba, Na, or Ca of La in La_2CuO_4 stabilizes the tetragonal K_2NiF_4 structure with distortion and produces also the doping effects on its electronic states. In the distorted phase, the dipoles on O(1) sites (in the CuO_2 planes) are not zero due to the lack of inversion symmetry at these sites, and thus the system is expected to be a superconductor, consistent with observations.[6] We will also see later that the dipoles on O(1) sites give rise to collective excitation frequencies, which are necessary for the pres-

TABLE VI. List of sublattice atoms (Ref. 17), Pauling's polarizabilities α (in units of Å^3), charges of ions q^* (in units of e), the sublattice coordinates x, y, and z (in units of a, b, and c, respectively), occupancies f, and the calculated electronic dipoles p_z (in units of e Å) in $Tl_2Ba_2Ca_2Cu_3O_{10}$ suppressing the subscript κ in various quantities. Lattice constants used for calculations are $a = 3.85$ Å, $b = 3.85$ Å, and $c = 35.88$ Å.

Sublattice atom	α	q^*	x	y	z	f	p_z
O[a]	3.88	−2	0.0	0.5	0.0	1	0.0
O[a]	3.88	−2	0.0	0.5	0.0875	1	0.17
O[a]	3.88	−2	0.0	0.5	0.4125	1	0.17
O[a]	3.88	−2	0.0	0.5	0.5	1	0.
O[a]	3.88	−2	0.0	0.5	0.5875	1	−0.17
O[a]	3.88	−2	0.0	0.5	0.9125	1	−0.17
Ca	0.47	2	0.0	0.0	0.0463	1	0.0
Ba	1.55	2	0.0	0.0	0.1448	1	−0.42
O	3.88	−2	0.0	0.0	0.2276	1	−2.70
Tl	1.54	3	0.0	0.0	0.2799	1	−0.53
O	3.88	−2	0.0	0.0	0.3412	1	1.06
Cu	0.1	2	0.0	0.0	0.4104	1	0.0
Cu	0.1	2	0.0	0.0	0.5	1	0.0
Cu	0.1	2	0.0	0.0	0.5896	1	0.0
O	3.88	−2	0.0	0.0	0.6588	1	−1.06
Tl	1.54	3	0.0	0.0	0.7201	1	0.53
O	3.88	−2	0.0	0.0	0.7724	1	2.70
Ba	1.55	2	0.0	0.0	0.8552	1	0.42
Ca	0.47	2	0.0	0.0	0.9537	1	0.0
Cu	0.1	2	0.5	0.5	0.0	1	0.0
Cu	0.1	2	0.5	0.5	0.0896	1	0.0
O	3.88	−2	0.5	0.5	0.1588	1	−1.06
Tl	1.54	3	0.5	0.5	0.2201	1	0.53
O	3.88	−2	0.5	0.5	0.2724	1	2.70
Ba	1.55	2	0.5	0.5	0.3552	1	0.42
Ca	0.47	2	0.5	0.5	0.4537	1	0.0
Ca	0.47	2	0.5	0.5	0.5463	1	0.0
Ba	1.55	2	0.5	0.5	0.6448	1	−0.42
O	3.88	−2	0.5	0.5	0.7276	1	−2.70
Tl	1.54	3	0.5	0.5	0.7799	1	−0.53
O	3.88	−2	0.5	0.5	0.8412	1	1.06
Cu	0.1	2	0.5	0.5	0.9104	1	0.0
O[a]	3.88	−2	0.5	0.0	0.0	1	0.0
O[a]	3.88	−2	0.5	0.0	0.0875	1	−0.17
O[a]	3.88	−2	0.5	0.0	0.4125	1	0.17
O[a]	3.88	−2	0.5	0.0	0.5	1	0.0
O[a]	3.88	−2	0.5	0.0	0.5875	1	−0.17
O[a]	3.88	−2	0.5	0.0	0.9125	1	0.17

[a] Oxygen atoms in the CuO_2 plane.

TABLE VII. List of sublattice atoms (Ref. 5), Pauling's polarizabilities α (in units of Å^3), charges of ions q^* (in units of e), the sublattice coordinates x, y, z (in units of a, b, and c, respectively), occupancies f, and the calculated electronic dipoles p_z (in units of e Å) in La_2CuO_4 (undistorted) where the subscript κ has been suppressed for simplicity. Lattice constants used for calculations are $a=3.7817$ Å, $b=3.7817$ Å, and $c=13.2487$ Å.

Sublattice atom	α	q^*	x	y	z	f	p_z
O(2)	3.88	−2	0.0	0.0	0.8176	1	0.417
La	1.04	3	0.0	0.0	0.63925	1	0.110
La	1.04	3	0.0	0.0	0.36075	1	−0.110
O(2)	3.88	−2	0.0	0.0	0.1824	1	−0.417
Cu	0.1	2	0.0	0.0	0.0	1	0.0
La	1.04	3	0.5	0.5	0.86075	1	−0.110
O(2)	3.88	−2	0.5	0.5	0.6824	1	−0.417
Cu	0.1	2	0.5	0.5	0.5	1	0.0
O(2)	3.88	−2	0.5	0.5	0.3176	1	0.417
La	1.04	3	0.5	0.5	0.13925	1	0.110
O(1)[a]	3.88	−2	0.0	0.5	0.5	1	0.0
O(1)[a]	3.88	−2	0.5	0.0	0.5	1	0.0
O(1)[a]	3.88	−2	0.0	0.5	0.0	1	0.0
O(1)[a]	3.88	−2	0.5	0.0	0.0	1	0.0

[a] Oxygen atoms in the CuO_2 plane.

ence of superconducticity under the dipole mechanism and will be discussed in Sec. V with respect to the observed optical absorptions in these superconductors.

In distorted La_2CuO_4 [$La_{2-x}(Sr,Ba,Na,Ca)_xCuO_{4-\delta}$] the atomic positions in the CuO_2 planes are slightly shifted[4,5] relative to their positions in the undistorted one. Because the exact positions of the substituting cations such as Sr, Ba, Na, or Ca are not available, we have taken the distorted lattice structure of $La_2CuO_{3.94}$ so as to conform with the tilting of the CuO_2 planes by about 5°.[4,5] This model structure and the calculated dipoles based on it are summarized in Table VIII. The dipoles on O(1) sites in the CuO_2 planes are now about 0.06e Å, and other dipoles are changed only slightly compared with the results in the undistorted system. Because of these finite dipoles on O(1) sites, the superconductivity is expected to appear under the collective dipole mechanism in distorted $La_{2-x}(Sr,Ba,Na,Ca)_xCuO_{4-\delta}$, consistent with observations.[4,5,26,27] Distorted $Ba_{0.6}K_{0.4}O_3$ (Ref. 20) and $Nd_{2-x}(Ce,Sr)_xCuO_{4-\delta}$ (Ref. 22) crystals are also expect-

TABLE VIII. List of sublattice atoms (Refs. 4 and 5), Pauling's polarizabilities α (in units of Å^3), charges of ions q^* (in units of e), the sublattice coordinates x, y, and z (in units of a, b, and c, respectively), occupancies f, and the calculated electronic dipole p_z (in units of e Å) in the distorted $La_2CuO_{3.94}$ model structure where the subscript κ has been suppressed for simplicity. Lattice constants used for calculations are $a=3.7817$ Å, $b=3.7817$ Å, and $c=13.2487$ Å with $z_0=0.0436$. In the last four rows the upper (lower) signs correspond to each other showing the tilting of the CuO_2 planes appropriate to the distorted structure.

Sublattice atom	α	q^*	x	y	z	f	p_z
O(2)	3.88	−2	0.0	0.0	0.8176	0.97	0.376
La	1.04	3	0.0	0.0	0.63925	1.0	0.147
La	1.04	3	0.0	0.0	0.36075	1.0	−0.147
O(2)	3.88	−2	0.0	0.0	0.1824	0.97	−0.376
Cu	0.1	2	0.0	0.0	0.0	1.0	0.0
La	1.04	3	0.5	0.5	0.86075	1.0	−0.156
O(2)	3.88	−2	0.5	0.5	0.6824	1.0	−0.422
Cu	0.1	2	0.5	0.5	0.5	1.0	0.0
O(2)	3.88	−2	0.5	0.5	0.3176	1.0	0.422
La	1.04	3	0.5	0.5	0.13925	1.0	0.156
O(1)[a]	3.88	−2	0.0	± 0.5	$0.5 \pm z_0$	1.0	± 0.05
O(1)[a]	3.88	−2	∓ 0.5	0.0	$0.5 \pm z_0$	1.0	± 0.05
O(1)[a]	3.88	−2	0.0	∓ 0.5	$\pm z_0$	1.0	± 0.06
O(1)[a]	3.88	−2	± 0.5	0.0	$\pm z_0$	1.0	± 0.06

[a] Oxygen atoms in the CuO_2 plane.

ed to display similar behavior. Doped La_2NiO_4, because of its similarity to doped La_2CuO_4, is expected to possess electronic properties similar to those of the latter. However, the Ni^{2+} ions, being magnetic, are expected to suppress the superconductivity in this substance.

As for the pressure effect on the dipoles in distorted La_2CuO_4 $[La_{2-x}(Sr,Ba,Na,Ca)_xCuO_{4-\delta}]$ we expect the values of the induced dipoles to change with pressure significantly in this system compared with the results in multilayered systems where dipoles are generated by the lack of inversion symmetry. This is consistent with experimental observations,[28–34] where the single-layered crystal has been found to have a much larger pressure effect on the superconducting properties compared with the effect on the multilayered systems.

Since the dipoles produced by lattice distortion or oxygen vacancies in a single-layered crystal are smaller than the dipoles in the multilayered superconductors, one expects lower T_c values in the single-layered system with doping and vacancies according to the present collective dipole mechanism.

III. DISPERSION RELATIONS AND DENSITY OF STATES OF COLLECTIVE DIPOLE EXCITATIONS

At all temperatures, whether at the absolute zero as a result of zero-point motions or at finite temperatures as a result of thermal fluctuations and interactions with thermally fluctuated electrons (or possibly holes), electronic dipoles in a high-T_c superconductor oscillate about their equilibrium positions. We shall consider these oscillations self-consistently here under the harmonic approximation. In this paper, we shall also ignore, as a first approximation as clarified earlier,[1] the motion of (heavy) nuclei and core electrons and find that the characteristic collective dipole excitation frequencies are much higher than the phonon frequencies. In the following, we shall obtain first the frequency of the oscillations of a single dipole in an electric field corresponding to a local electric field at a sublattice in a crystal for simplicity and then the dispersion relations of collective dipole excitations for a linear and a planar system of dipoles generated by uniform electric field and for various high-T_c superconducting systems.

A. A single dipole in an electric field

Let us first consider a single induced electric dipole at a given site aligned in a crystal due to the local crystal electric field. Let it be fixed at its heavy (nuclear) end and released at the other end so that it is free to oscillate if the dipole is disturbed in alignment or if the electric field fluctuates. The resulting torque on the dipole makes the dipole oscillate about its equilibrium position. The frequency in this case for small oscillations comes out to be

$$\omega \cong \left[\frac{|\mathbf{p}|\cdot|\mathbf{E}|}{I}\right]^{1/2} = \left[\frac{|\mathbf{p}|^2}{\alpha I}\right]^{1/2} ,$$

where I is assumed to be the effective moment of inertia of the oscillating dipole. $|\mathbf{p}|$ and $|\mathbf{E}|$ are absolute values of the dipole moment and the local electric field, respectively, with $p=\alpha E$ where α is the polarizability. For an ion such as O^{2-} in a crystal, we take appropriately $p=0.2e$ Å, $\alpha=3.88$ Å^3, and $I=\frac{2}{5}(8m_e)R^2$, with the radius of the oxygen ion[25] $R=\alpha^{1/3}$ to obtain $\omega=0.92\times10^{14}$ Hz, an expected value[1] for the oscillations of a single dipole, many of such dipolar-coupled oscillations forming collective waves as will be clear in the following.

B. The dynamic matrix of collective dipole excitations

The crystal electric field at oxygen sites induces electronic dipoles on oxygens. To investigate their self-sustained oscillations we write the ith dipole as

$$\mathbf{p}_i(\mathbf{q}|t)=\mathbf{p}_i(0)+\Delta\mathbf{p}_i(\mathbf{q}|t) , \tag{7}$$

where $\mathbf{p}_i(0)$ is the static dipole at the ith site and $\Delta\mathbf{p}_i(\mathbf{q}|t)$ is the time-dependent oscillatory part with $\mathbf{q}$ as the wave vector of the propagating wave. In the following we will assume that the magnitude of the change in the dipole, $|\Delta\mathbf{p}|$, is much smaller than the magnitude of the dipole p itself ($|\Delta\mathbf{p}|\ll p$). Then Eq. (1) (assuming now $\mathbf{p}_j$ to be time dependent) in conjunction with Eq. (7) may be rewritten as

$$\mathbf{E}_i(\mathbf{q}|t)=\mathbf{E}_i(0)+\sum_j(\tilde{F}_{ij})\cdot\Delta\mathbf{p}_j(\mathbf{q}|t) , \tag{8}$$

where $\mathbf{E}_i(0)$ is the static electric field and is given by $\mathbf{E}_i$ in Eq. (1) due to the (static) monopoles and the static dipoles as considered in Sec. II. The total electric field $\mathbf{E}_i(\mathbf{q}|t)$ gives rise to a torque $\tau_i(\mathbf{q}|t)$ on the dipole $\mathbf{p}_i(\mathbf{q}|t)$ according to

$$\tau_i(\mathbf{q}|t)=\mathbf{p}_i(\mathbf{q}|t)\times\mathbf{E}_i(\mathbf{q}|t) . \tag{9}$$

In a first-order approximation,

$$\tilde{I}_i\cdot\frac{\partial^2\boldsymbol{\theta}_i}{\partial t^2}\cong\mathbf{p}_i(0)\times\left[-\frac{\Delta\mathbf{p}_i(\mathbf{q}|t)}{\alpha_i}+\sum_j(\tilde{F}_{ij})\cdot\Delta\mathbf{p}_j(\mathbf{q}|t)\right] , \tag{10}$$

where we have taken

$$\tau_i(\mathbf{q}|t)=\tilde{I}_i\cdot\frac{\partial^2\boldsymbol{\theta}_i}{\partial t^2} \tag{11}$$

with $\boldsymbol{\theta}_i$ defined as

$$\boldsymbol{\theta}_i\approx\frac{\mathbf{p}_i(0)\times\Delta\mathbf{p}_i(\mathbf{q}|t)}{|\mathbf{p}_i(0)|^2} , \tag{12}$$

and $\tilde{I}_i$ is the moment of inertia tensor associated with the electronic charge distribution $\rho_e(\mathbf{r})$ at the ith site with its $\lambda\lambda'$ components given by

$$(\tilde{I}_i)_{\lambda\lambda'}=m_e\int\rho_e(\mathbf{r})(r^2\delta_{\lambda\lambda'}-r_\lambda r_{\lambda'})d^3r , \tag{13}$$

where m_e is the mass of an electron.

In the cases under consideration the directions of the static dipole $\mathbf{p}_i(0)$ are along $\pm\hat{\mathbf{z}}$, and thus Eq. (10) yields

$$\frac{\partial^2 \Delta p_{xi}(\mathbf{q}|t)}{\partial t^2} = \frac{p_i^2(0)}{I_i}\left[-\frac{1}{\alpha_i}\Delta p_{xi}(\mathbf{q}|t) + \sum_j \left[\frac{3x_{ij}^2 - r_{ij}^2}{r_{ij}^5}\right]\Delta p_{xj}(\mathbf{q}|t) + \sum_j \left[\frac{3x_{ij}y_{ij}}{r_{ij}^5}\right]\Delta p_{yj}(\mathbf{q}|t)\right] , \tag{14}$$

$$\frac{\partial^2 \Delta p_{yi}(\mathbf{q}|t)}{\partial t^2} = \frac{p_i^2(0)}{I_i}\left[-\frac{1}{\alpha_i}\Delta p_{yi}(\mathbf{q}|t) + \sum_j \left[\frac{3y_{ij}x_{ij}}{r_{ij}^5}\right]\Delta p_{xj}(\mathbf{q}|t) + \sum_j \left[\frac{3y_{ij}^2 - r_{ij}^2}{r_{ij}^5}\right]\Delta p_{yj}(\mathbf{q}|t)\right] , \tag{15}$$

where $I_i = (\tilde{I}_i)_{xx} = (\tilde{I}_i)_{yy}$. It is clear that the dynamics of the polarization along the $\hat{\mathbf{z}}$ direction do not play any role, which is due to the fact that the quadratic and higher-order terms in $\Delta\mathbf{p}_j(\mathbf{q}|t)$ have been neglected in arriving at the present results. Following a general treatment of collective oscillations[35] with i standing for $(l|\kappa)$ and λ designating polarizations along x and y,

$$\Delta p_{\lambda(l|\kappa)}(\mathbf{q}|t) = \frac{1}{2}\frac{A(\mathbf{q})|p_\kappa(0)|}{\sqrt{NI_\kappa}}[\xi_\lambda(\mathbf{q}|\kappa)e^{i[\mathbf{q}\cdot\mathbf{r}(l|\kappa) - \omega_\lambda(\mathbf{q})t]} + \text{c.c.}] , \tag{16}$$

where $A(\mathbf{q})$ is the amplitude assumed to be real, and $\xi_\lambda(\mathbf{q}|\kappa)$ is the λth component of the unit polarization vector $\hat{\boldsymbol{\xi}}(\mathbf{q}|\kappa)$ of the collective wave; $|p_\kappa(0)|/\sqrt{NI_\kappa}$ acts as a renormalization factor with N being the number of oscillating dipoles in a crystal under consideration; c.c. stands for the complex conjugate of the first term. Equation (16) describes a plane wave propagating with wave vector $\mathbf{q}$ which, in conjunction with Eqs. (14) and (15), yields

$$\omega_\lambda^2(\mathbf{q})\xi_\lambda(\mathbf{q}|\kappa) = \sum_{\lambda'\kappa'} D_{\lambda\lambda'}(\mathbf{q}|\kappa\kappa')\xi_{\lambda'}(\mathbf{q}|\kappa') , \tag{17}$$

where $D_{\lambda\lambda'}(\mathbf{q}|\kappa\kappa')$ is the dynamic matrix given by

$$D_{\lambda\lambda'}(\mathbf{q}|\kappa\kappa') = \frac{|p_\kappa(0)||p_{\kappa'}(0)|}{\sqrt{I_\kappa I_{\kappa'}}} e^{-i\mathbf{q}\cdot[\mathbf{r}(\kappa) - \mathbf{r}(\kappa')]} \begin{cases} \dfrac{1}{\alpha_\kappa}\delta_{\lambda\lambda'}, & (l|\kappa) = (l'|\kappa') \\ -\displaystyle\sum_{l-l'} \left[\frac{3r_\lambda r_{\lambda'} - r^2\delta_{\lambda\lambda'}}{r^5}\right] e^{-i\mathbf{q}\cdot[\mathbf{r}(l) - \mathbf{r}(l')]}, & (l|\kappa) \neq (l'|\kappa') \end{cases} \tag{18}$$

with λ (and λ')$\equiv x,y$ and, in general, $\mathbf{r}(\kappa) = \mathbf{r}(0|\kappa)$ and $\mathbf{r}(l) = \mathbf{r}(l|0)$. Equation (17) represents a set of $2n$ linear homogeneous equations in $2n$ unknown amplitudes $\xi_\lambda(\mathbf{q}|\kappa)$ for a given wave vector $\mathbf{q}$, where n is the total number of sublattices in a unit cell.

Transforming Eq. (16) into the second quantization notation one obtains

$$\Delta p_{\lambda(l|\kappa)}(\mathbf{q}|t) = |p_\kappa(0)|\left[\frac{\hbar}{2I_\kappa N\omega_\lambda(\mathbf{q})}\right]^{1/2} \xi_\lambda(\mathbf{q}|\kappa)(b_{-\mathbf{q}\lambda}^\dagger + b_{\mathbf{q}\lambda})e^{i\mathbf{q}\cdot\mathbf{r}(l|\kappa)} , \tag{19}$$

where $b^\dagger$ and b are the quantized-collective-dipole excitation creation and annihilation operators (time dependent).

From Eq. (17) one notes that the collective excitation frequencies are proportional to the magnitude of the dipoles. One finds the eigenfrequencies and eigenvectors of these excitations by diagonalizing the dynamic matrix $D_{\lambda\lambda'}(\mathbf{q}|\kappa\kappa')$, which may then be analyzed to determine whether collective dipole excitations in a given system are indeed feasible.

C. Density of states of the collective dipole excitations

The associated density of states $F(\omega)$ can be defined as

$$F(\omega) = \sum_{\lambda\mathbf{q}} \delta(\omega - \omega_\lambda(\mathbf{q})) , \tag{20}$$

where $\omega_\lambda(\mathbf{q})$ are the solutions of Eq. (17). By definition, $F(\omega)$ should satisfy

$$\int_0^{\omega_{\max}} F(\omega)d\omega = 2N_d N_c , \tag{21}$$

where N_d is the number of dipoles in a unit cell and N_c is the number of cells in a crystal assuming the normalization of $F(\omega)$ within a unit cell for two polarizations along x and y in the present case.

D. Dipolar chain with interacting dipoles generated by a uniform electric field

Suppose that there are equivalent equidistant dipoles of magnitude p arranged in a lattice along the x direction with lattice constant a. The dipoles are assumed to be due to the presence of a uniform electric field E in the z direction so that $\mathbf{p} = \alpha\mathbf{E}$. In this case, Eq. (17) yields the dispersion relation in terms of the wave vector of magnitude q_x for the longitudinal mode as

$$\omega_x^2 = \frac{p^2}{I}\left[\frac{1}{\alpha} - \sum_{n=1}^{\infty}\frac{2}{(na)^3}2\cos(q_x na)\right] \tag{22}$$

and the dispersion relation for the transverse mode as

$$\omega_y^2 = \frac{p^2}{I}\left[\frac{1}{\alpha} + \sum_{n=1}^{\infty}\frac{1}{(na)^3}2\cos(q_x na)\right] . \tag{23}$$

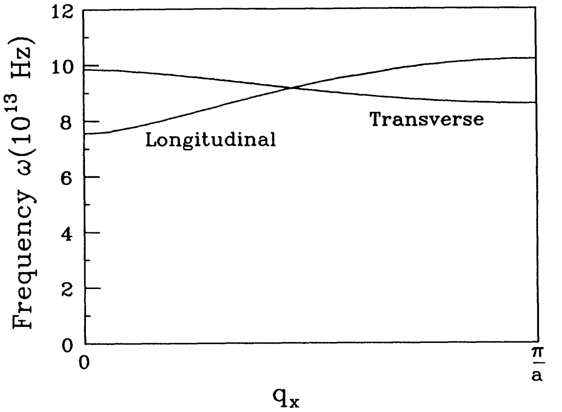

FIG. 1. Calculated dispersion relations of collective dipole excitations in one-dimensional dipole chain.

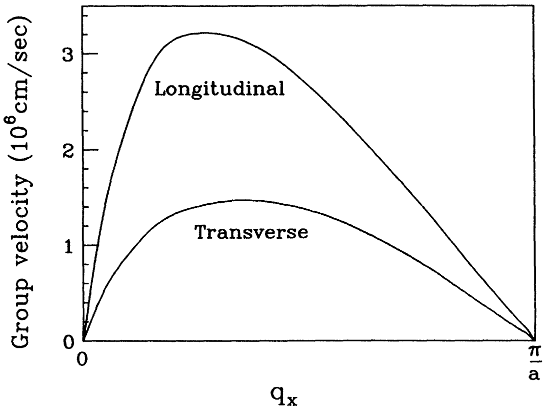

FIG. 2. Calculated group velocities of collective dipole excitations in one-dimensional dipole chain.

The first term of Eq. (22) as well as of Eq. (23) corresponds to the contribution as if it arises from the oscillations of the independent dipoles and the remaining terms are the results of the interaction between the dipoles giving rise to the collective oscillations of the system as a whole. For estimating I, since we assume that the $1s$ core electrons of an oxygen ion are not shifted when dipoles are induced, we employ the atomic wave functions[35] of $2s$ and $2p$ electrons of the oxygen ion O^{2-} to obtain $I=5.2\times10^{-43}$ g cm^2. Taking appropriately $p=0.2$ Å, $\alpha=3.88$ Å^3, and $a=3.85$ Å, valid for high-T_c compounds, the estimated frequencies from Eqs. (22) and (23) lie in the range 2500 cm^{-1} (0.75×10^{14} Hz) to 3400 cm^{-1} (1.02×10^{14} Hz). It is also important to note that slight readjustments of the values of α and a in Eqs. (22) and (23) cover the range of values from 0 to $\omega_{\max}$ with $\omega_{\max}\sim10^{14}$ Hz, a result of prime importance for the discussion of the collective dipole mechanism for high-T_c superconductivity. The calculated dispersion relations for this one-dimensional dipole chain are shown in Fig. 1. The calculated group velocities $(\partial\omega_\lambda/\partial q_x)$ in this case have been depicted in Fig. 2 where we note the presence of the asymmetries in the group velocity of collective dipole excitations, which are due to the long-range interaction between the dipoles indicating that the large limits in the summation in Eqs. (22) and (23) are necessary for the accurate results.

It is evident that these oscillating dipoles are analogous to a system of rotating spins (magnon) if viewed as combinations of the longitudinal and transverse modes. These results also lead to the suggestion that, if lattice distortions or defects are present in a system, they should at least be arranged approximately periodically for dipolar oscillations to be feasible.

E. Dipolar plane with interacting dipoles generated by a uniform electric field

In this case there are equivalent equidistant dipoles of magnitude p arranged in a planar lattice in the x-y plane with lattice constants a and b. The dipoles are due to a uniform electric field $\mathbf{E}$ in the $\hat{\mathbf{z}}$ direction so that $\mathbf{p}=\alpha\mathbf{E}$. Here, Eq. (17) yields two dispersion relations in terms of wave vector $\mathbf{q}$ for two different modes of vibrations with frequencies given by

$$\begin{pmatrix} \frac{p^2}{I}\left\{\frac{1}{\alpha}-\sum_{n=0}^{\infty}\sum_{m=0}^{\infty}(1-\delta_{n,0}\delta_{m,0})\times\left[\frac{2n^2a^2-m^2b^2}{(n^2a^2+m^2b^2)^{5/2}}\right](2-\delta_{n,0})\times(2-\delta_{m,0})\cos(q_xna)\cos(q_ymb)\right\}-\omega^2 & \frac{p^2}{I}\left\{\sum_{n=0}^{\infty}\sum_{m=0}^{\infty}\left[\frac{3namb}{(n^2a^2+m^2b^2)^{5/2}}\right]\times4\sin(q_xna)\sin(q_ymb)\right\} \\ \frac{p^2}{I}\left\{\sum_{n=0}^{\infty}\sum_{m=0}^{\infty}\left[\frac{3namb}{(n^2a^2+m^2b^2)^{5/2}}\right]4\sin(q_xna)\sin(q_ymb)\right\} & \frac{p^2}{I}\left\{\frac{1}{\alpha}-\sum_{n=0}^{\infty}\sum_{m=0}^{\infty}(1-\delta_{n,0}\delta_{m,0})\times\left[\frac{2m^2b^2-n^2a^2}{(n^2a^2+m^2b^2)^{5/2}}\right](2-\delta_{n,0})\times(2-\delta_{m,0})\cos(q_xna)\cos(q_ymb)\right\}-\omega^2 \end{pmatrix}\begin{pmatrix}\xi_x\\ \xi_y\end{pmatrix}=\begin{pmatrix}0\\0\end{pmatrix}. \quad (24)$$

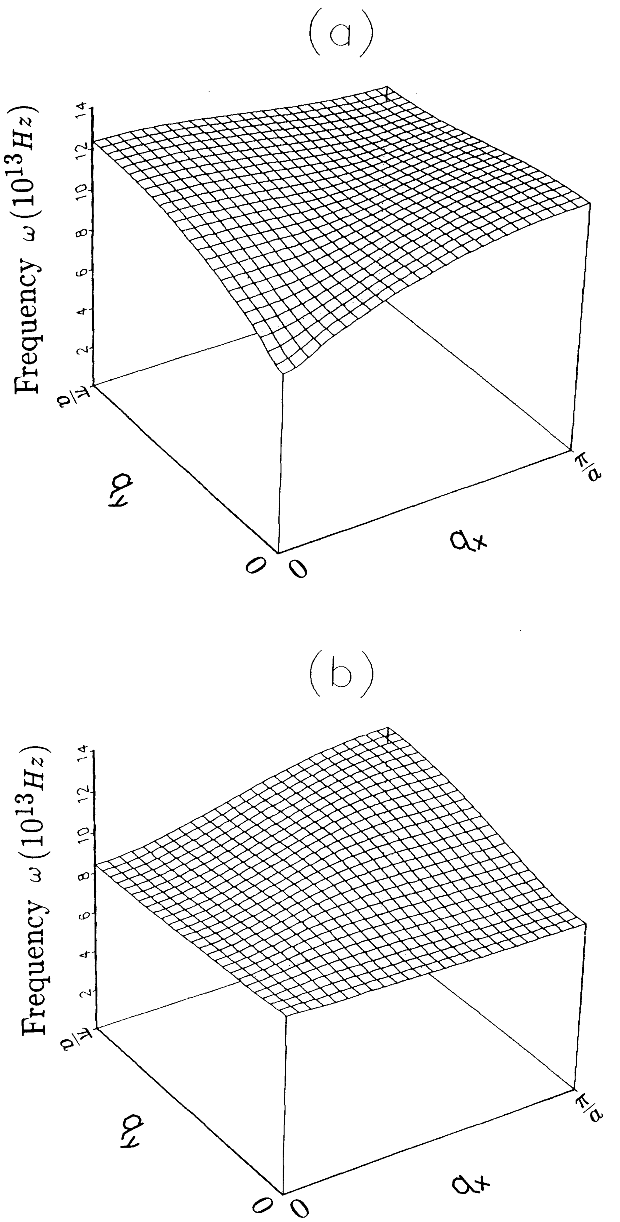

FIG. 3. Calculated dispersion relations of collective dipole excitations in two-dimensional dipole plane (a) for ω_+ and (b) for ω_-.

Again, assuming appropriately $p=0.2$ Å, $I=5.2\times10^{-43}$ g cm^2, $\alpha=3.88$ Å^3, and $a=b=3.85$ Å, Eq. (24) yields the solutions $\omega_+(q_x,q_y),\omega_-(q_x,q_y)$ as the dispersion relation of collective dipole excitations in this case for the two modes that are shown in Figs. 3(a) and 3(b), respectively.

F. $YBa_2Cu_3O_{7-\delta}$

In the $YBa_2Cu_3O_{7-\delta}$ system, there are three pairs of oxygen dipoles, members of each pair being of equal strength but pointing in opposite directions (Table I). In the present calculations we have neglected the oscillations of the dipoles of Ba atoms, since the associated moment of inertia is relatively large, which makes the relevant frequency of oscillations negligibly small. Each pair of dipoles produces four branches in the dispersion relations corresponding to the symmetric and antisymmetric modes for each polarization. Thus the modes of the oscillations are (1) longitudinal symmetric, (2) longitudinal antisymmetric, (3) transverse symmetric, and (4) transverse antisymmetric. The calculated dispersion relations and the density of states $F(\omega)$ in $YBa_2Cu_3O_7$ are shown in Figs. 4(a) and 4(b), respectively.

Concerning calculations for $YBa_2Cu_3O_{7-\delta}$ $(\delta\neq0)$ we have used two different structural models as mentioned in the preceding section. The dispersion relations using the first model are shown in Fig. 5, corresponding to the oscillations of dipoles which lie in the CuO_2 planes. The calculated dispersion relations of the collective dipole excitations clearly show that the characteristic frequencies in CuO_2 planes decrease for increasing δ, consistent with our previous results.[1] The maximum allowable frequencies $f_{\max}$ in the CuO_2 planes are listed in Table IX for easy reference. Previously,[1] we have used the relation

$$\omega_d(\delta)\simeq\omega_d^0(1-g\delta) \tag{25}$$

for the characteristic frequency derived by maximizing the binding energy of a collective-dipole-excitation-mediated electron-electron pair to explain the T_c variations versus δ in $YBa_2Cu_3O_{7-\delta}$, where $\omega_d^0=1\times10^{14}$ Hz and $g=1.187$ (as calculated from the derived relation[1] for g). Here, in comparison, by assuming that the maximum frequencies represent the characteristic frequencies we find that the same relation [Eq. (25)] is still valid with $\omega_d^0=1.3\times10^{14}$ Hz and $g=1.26$ $(0\leq\delta\leq0.4)$, very close to the previous results.[1]

The second structural model reproduces (not shown here) the overall shapes of the dispersion curves of the first structure model with the changes in frequencies occurring due to the uncertainties in the assumed values of the structural data and in the ordering of oxygen vacancies. The calculated maximum frequencies of the collective dipole excitations in CuO_2 planes for different δ values from this second structure model are listed in Table X.

The collective dipole frequencies corresponding to the dipoles at the O(4) site have a tendency to increase slightly as δ increases owing mainly to the increase in the mag-

TABLE IX. The calculated maximum collective dipole excitation frequencies in the CuO_2 planes of $YBa_2Cu_3O_{7-\delta}$ for various δ values using the first structural model described in the text.

δ	0.0	0.28	0.337	0.383	0.434	0.5
$f_{\max}$ (10^{13} Hz)	13.188	8.344	7.364	6.664	6.580	6.860

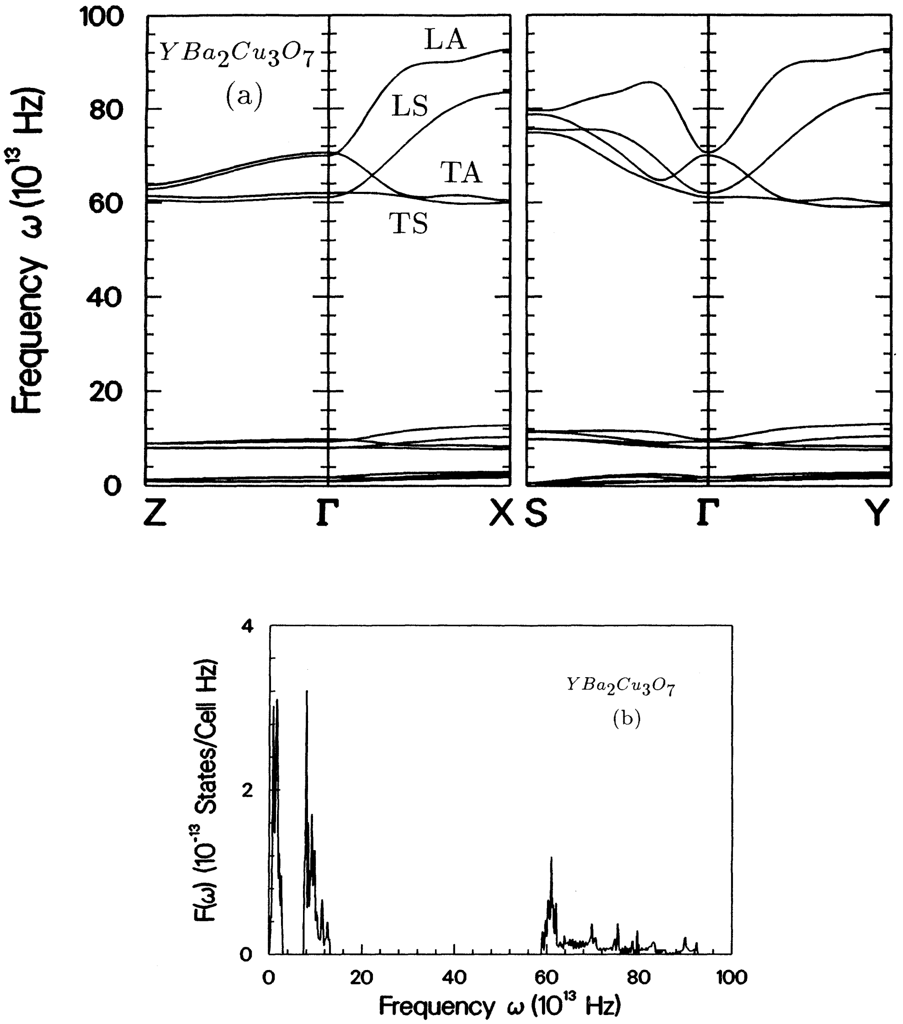

FIG. 4. (a) Calculated dispersion relations for collective dipole excitations in $YBa_2Cu_3O_7$, where the magnitudes of the wave vectors are measured from $\Gamma(0,0,0)$ to $X(\pi/a,0,0)$, $Y(0,\pi/b,0)$, $Z(0,0,\pi/c)$, and $S(\pi/a,\ \pi/b,\ 0)$, respectively; (b) calculated density of states $F(\omega)$ of collective dipole excitations in $YBa_2Cu_3O_7$.

nitude of these dipoles.

As for confirming the predicted frequencies experimentally in $YBa_2Cu_3O_{7-\delta}$, we suggest that one may use a light beam parallel to the c axis ($\mathbf{E}\perp c$), and change the polarization and the intensity of light to observe absorption peaks (with broad bandwidth) at the predicted frequencies. The absorption peak due to the collective dipole excitations involving O(4) sites should appear around 2.5 eV with the broad bandwidth as much as 1.6 eV, and should shift to the high-energy side as δ increases. The absorption peaks due to the oscillations of dipoles in the CuO_2 planes should be observed around 0.36 eV with a bandwidth as broad as 0.2 eV and should shift to the lower-frequency side as δ increases. One more low-energy peak at 0.08 eV with a bandwidth as broad as 0.15 eV has been predicted to exist by our calculated density of states, which shifts to higher energy as δ increases and tends to merge with the peak at 0.36 eV, thereby making it somewhat difficult to be resolved experimentally.

Furthermore, because the collective dipole excitation frequencies depend on the magnitudes of dipoles involved, it is more difficult to observe low-frequency absorption peaks than the high-frequency ones. For this

TABLE X. The calculated maximum collective dipole excitation frequencies in the CuO_2 planes of $YBa_2Cu_3O_{7-\delta}$ for various δ values using the second structural model described in the text.

δ	0.29	0.4	0.46	0.56	0.54	0.55
$f_{\max}$ (10^{13} Hz)	5.964	1.876	2.968	2.576	2.324	3.444

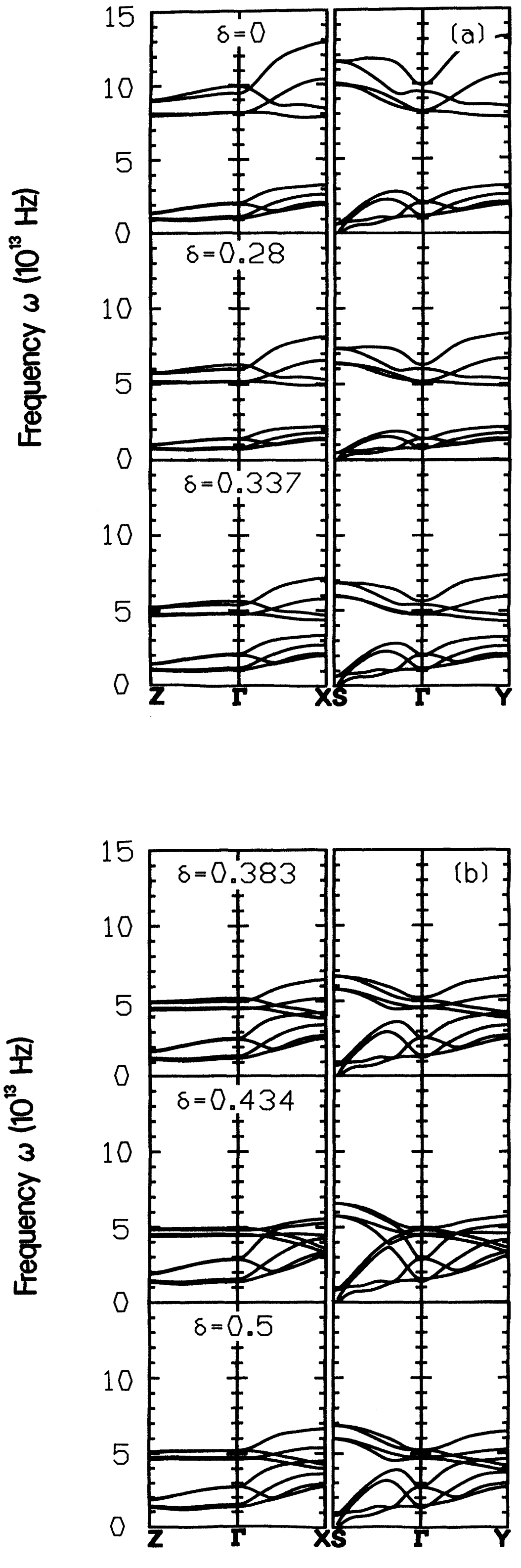

FIG. 5. Calculated dispersion relations of collective dipole excitations in the CuO_2 planes in $YBa_2Cu_3O_{7-\delta}$ system for various δ values where the relative positions of the wave vector are the same as in Fig. 4(a) using the first model described in the text (the higher-frequency branches have not been shown).

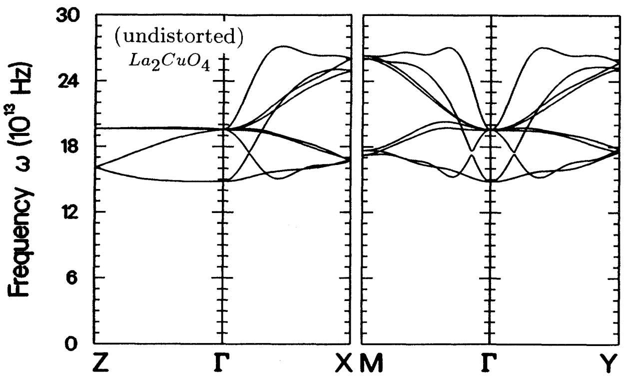

FIG. 6. Calculated dispersion relations of collective dipole excitations in La_2CuO_4 (undistorted) where the magnitude of wave vectors are measured from Γ(0,0,0) to $X(\pi/a,0,0)$, $Y(\pi/2a,\pi/a,0)$, $Z(0,0,\pi/c)$, and $M(\pi/a,\pi/a,0)$, respectively.

reason, one may be required to use a light beam of carefully adjusted power. In addition, for the peaks at high frequencies one must exercise caution as they may be missed because of the overlap with the absorption peaks due to the electronic transitions between the (electronic) bands of the system.

The peak positions that we have predicted here are likely to change slightly because of the approximate average oxygen occupancy factors used in our calculations. The changes in the polarizabilities[25] are not expected to lead to drastic changes of these values as our calculations indicate that such changes bring about only small changes in the magnitudes of the frequencies.

G. La_2CuO_4

We have also calculated the dispersion relations of the collective dipole excitations in undistorted La_2CuO_4 (nonsuperconductor) as shown in Fig. 6. Clearly there are no dispersion relations of collective dipole excitations found near or below 1×10^{14} Hz essential for the presence of high-T_c superconductivity, because of the absence of dipoles in the CuO_2 plane in the undistorted system. However, the frequencies due to dipoles at O(2) sites (not

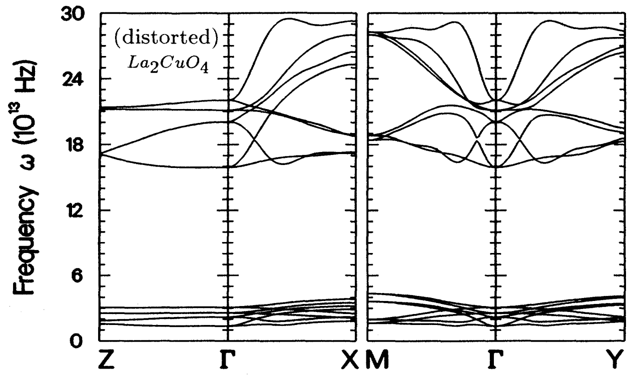

FIG. 7. Calculated dispersion relations of collective dipole excitations in distorted La_2CuO_4 with the same notation as in Fig. 6.

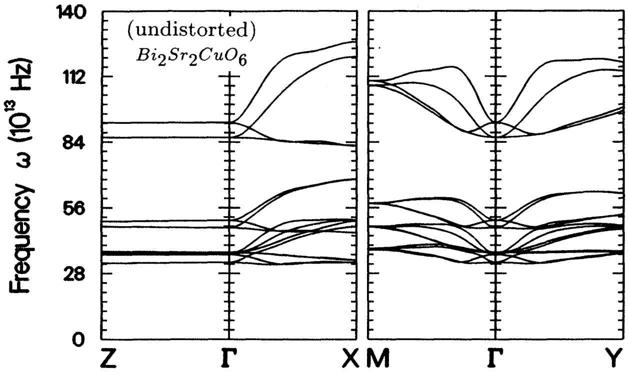

FIG. 8. Calculated dispersion relations of collective dipole excitations in $Bi_2Sr_2CuO_6$ (undistorted) where the magnitude of wave vectors are measured from $\Gamma(0,0,0)$ to $X(\pi/a,0,0)$, $Y(\pi/2a,\pi/a,0)$, $Z(0,0,\pi/c)$, and $M(\pi/a,\pi/a,0)$, respectively.

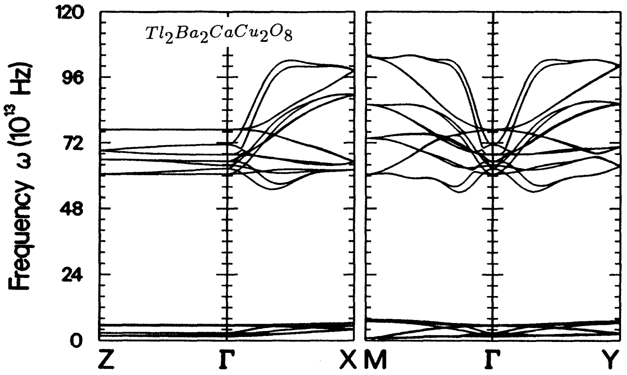

FIG. 10. Calculated dispersion relations of collective dipole excitations in $Tl_2Ba_2CaCu_2O_8$ with the same notation as in Fig. 8.

in the CuO_2 plane) that do not play a part in superconductivity are found to be around 1 eV with as broad a bandwidth as 0.4 eV. On the other hand, the distorted $La_2CuO_{3.94}$ exhibits the collective dipole excitation frequencies in the CuO_2 plane around 0.2 eV with a bandwidth as broad as 0.17 eV from our calculations (Fig. 7) using the distorted structural model mentioned above. One notes that, as expected, the collective dipole excitation frequencies around 1 eV due to the dipoles on O(2) sites in this class of substances are present in both nonsuperconductor and superconductor crystals. Our calculated dispersion relations of the collective dipole excitations in the distorted La_2CuO_4 are shown in Fig. 7.

H. Other systems

The dispersion relations of collective dipole excitations for $Bi_2Sr_2CuO_6$ (undistorted), $Bi_2Sr_2CaCu_2O_8$, $Tl_2Ba_2CaCu_2O_8$, and $Tl_2Ba_2Ca_2Cu_3O_{10}$ are given in Figs. 8, 9, 10, and 11, respectively. As clearly seen in Fig. 8, the dispersion relations corresponding to collective dipole oscillations in the CuO_2 planes in $Bi_2Sr_2CuO_6$ (undistorted) do not appear simply because there are no induced dipoles in the CuO_2 plane. Owing to this, the undistorted $Bi_2Sr_2CuO_6$ is not expected to be a superconductor according to the present model, an experimental fact. However, we find collective dipole excitation frequencies due to the oxygens at other sites, which happen to lie in the high-frequency range. On the other hand, the calculated dispersion relations of collective dipole oscillations in CuO_2 planes for $Bi_2Sr_2CaCu_2O_8$, $Tl_2Ba_2CaCu_2O_8$, and $Tl_2Ba_2Ca_2Cu_3O_{10}$ are found to fall in the expected frequency range, and, hence, these systems are expected to be superconductors under the present mechanism, consistent with experimental observations. Also, major peaks with broad bandwidth in these cases appear at the same frequencies as in $YBa_2Cu_3O_{7-\delta}$ systems.

IV. INTERACTION OF COLLECTIVE DIPOLE EXCITATION WITH ELECTRONS

A dipole $\mathbf{p}(\mathbf{R}|t)$ at the location $\mathbf{R}$ interacts with an electron at the position $\mathbf{r}$ with the potential given by

$$V_{pe}(\mathbf{r}-\mathbf{R})=(-e)\frac{\mathbf{p}(\mathbf{R}|t)\cdot(\mathbf{r}-\mathbf{R})}{\epsilon|\mathbf{r}-\mathbf{R}|^3} \tag{26}$$

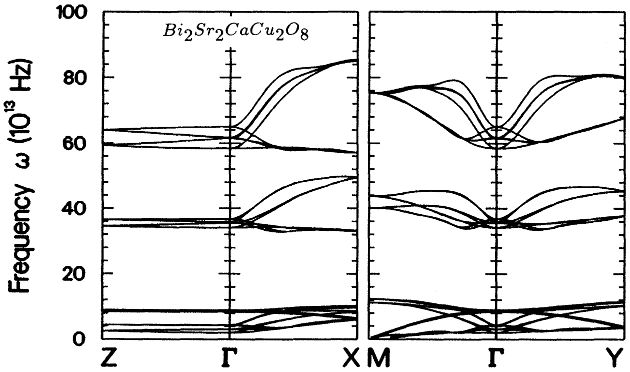

FIG. 9. Calculated dispersion relations of collective dipole excitations in $Bi_2Sr_2CaCu_2O_8$ with the same notation as in Fig. 8.

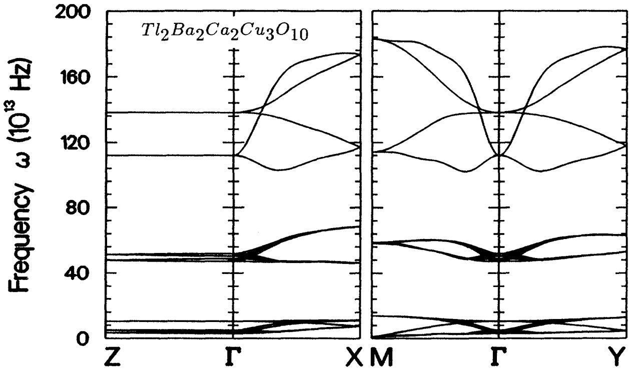

FIG. 11. Calculated dispersion relations of collective dipole excitations in $Tl_2Ba_2Ca_2Cu_3O_{10}$ with the same notation as in Fig. 8.

with ϵ as a screening factor (introduced for general treatment) and

$$\mathbf{p}(\mathbf{R}|t)=\mathbf{p}(\mathbf{R}|0)+\Delta\mathbf{p}(\mathbf{R}|t) ,$$

where $\mathbf{p}(\mathbf{R}|0)$ is the static part and $\Delta\mathbf{p}(\mathbf{R}|t)$ as the time-dependent part so that

$$V_{pe}(\mathbf{r}-\mathbf{R})\cong\mathbf{p}(\mathbf{R}|0)\cdot\nabla_{\mathbf{r}}\left[\frac{e}{\epsilon|\mathbf{r}-\mathbf{R}|}\right]+\Delta\mathbf{p}(\mathbf{R}|t)\cdot\nabla_{\mathbf{r}}\left[\frac{e}{\epsilon|\mathbf{r}-\mathbf{R}|}\right] . \tag{27}$$

The first term on the right-hand side of Eq. (27) is independent of time, which just provides the correction on the electronic bands, and the second term is the dynamic one, which, in conjunction with Eq. (19), in the second-quantized notations leads to the interaction Hamiltonian

$$\mathcal{H}_{\text{ed}}=\sum_{\mathbf{k}\sigma\mathbf{k}'\sigma'\mathbf{q}\lambda} h_{\mathbf{k}\sigma\mathbf{k}'\sigma'\mathbf{q}\lambda}(b^{\dagger}_{-\mathbf{q}\lambda}+b_{\mathbf{q}\lambda})C^{\dagger}_{\mathbf{k}'\sigma'}C_{\mathbf{k}\sigma} , \tag{28}$$

where $C^{\dagger}_{\mathbf{k}\sigma}$ and $C_{\mathbf{k}\sigma}$ are the creation and annihilation operators for an electron with wave vector $\mathbf{k}$ and spin σ, and $b^{\dagger}_{\mathbf{q}\lambda}$ and $b_{\mathbf{q}\lambda}$ are the creation and annihilation operators for a collective dipole quantum with wave vector $\mathbf{q}$ and polarization index λ and

$$h_{\mathbf{k}\sigma\mathbf{k}'\sigma'\mathbf{q}\lambda}=i|p_{\kappa}(0)|\left[\frac{\hbar}{2I_{\kappa}N\omega_{\lambda}(\mathbf{q})}\right]^{1/2}[\xi_{\mathbf{q}\lambda}\cdot(\mathbf{k}-\mathbf{k}')]\times v_d^{\mathbf{k},\mathbf{k}'}\delta_{\mathbf{k}'-\mathbf{k},\mathbf{q}+\mathbf{G}} , \tag{29}$$

where $\mathbf{G}$ is a reciprocal vector, and

$$v_d^{\mathbf{k},\mathbf{k}'}=\int d^3r u^{\dagger}_{\mathbf{k}',\sigma'}(\mathbf{r})e^{-i\mathbf{k}'\cdot\mathbf{r}}\frac{e}{\epsilon|\mathbf{r}-\mathbf{R}|}u_{\mathbf{k},\sigma}(\mathbf{r})e^{i\mathbf{k}\cdot\mathbf{r}} \tag{30}$$

with $u_{\mathbf{k},\sigma}(\mathbf{r})e^{i\mathbf{k}\cdot\mathbf{r}}$, in general, being the Bloch state of the electron in the system.

Expression (28) leads to the electron-electron interaction V_{ee} via virtual exchange of collective-dipole-excitation quanta as

$$V_{ee}=\sum_{\mathbf{k}\sigma\mathbf{k}'\sigma'} V_{\mathbf{k}\mathbf{k}'}C^{\dagger}_{\mathbf{k}'\sigma'}C^{\dagger}_{\mathbf{k}\sigma}C_{\mathbf{k}\sigma}C_{\mathbf{k}'\sigma'} , \tag{31}$$

where

$$V_{\mathbf{k}\mathbf{k}'}=\sum_{\lambda}\frac{|h_{\mathbf{k}\sigma\mathbf{k}'\sigma'\mathbf{q}\lambda}|^2\hbar\omega_{\lambda}(\mathbf{q})}{\{(\epsilon_{\mathbf{k}}-\epsilon_{\mathbf{k}'})^2-[\hbar\omega_{\lambda}(\mathbf{q})]^2\}} . \tag{32}$$

$V_{\mathbf{k}\mathbf{k}'}$ is attractive in the range $|\epsilon_{\mathbf{k}}-\epsilon_{\mathbf{k}'}|<\hbar\omega_{\lambda}(\mathbf{q}),\epsilon_{\mathbf{k}}$ being the electron energy. If we wish to apply the BCS approximation, we need to make $V_{\mathbf{k}\mathbf{k}'}$ independent of $\mathbf{k},\mathbf{k}'$ (or $\mathbf{q}$ since $\mathbf{k}'-\mathbf{k}=\mathbf{q}$ for $\mathbf{G}=0$). The calculations of T_c for the $YBa_2Cu_3O_{7-\delta}$ system as a function of the oxygen-stoichiometry parameter δ based on this electron-electron interaction under the BCS approximation reproduces the results obtained previously,[1] where the dipolar oscillations were assumed to be of a very general form.

The calculations of T_c and gap energy using the integral equations[37,38] and the density of states of these collective dipole excitations including Coulomb repulsion between the electrons are in progress and will be reported elsewhere.

V. DISCUSSION AND CONCLUSION

The main aim of this work is to establish the feasibility of self-sustained collective dipole excitations in high-T_c superconductors and prove that their characteristic excitation frequencies are $\sim 10^{14}$ Hz. We have demonstrated above how to calculate the dipoles and collective dipole excitation frequencies in the high-T_c superconductors. There remains the possibility of improving these calculations when more exact structural information and probabilities of the site occupancies by the oxygens become available. Further, the present calculations are not expected to change drastically on changing parameters such as charges, polarizabilities, the moment of inertia of dipoles, etc., within their expected physical limits.

As for the experimental results which are observed in the mid-infrared absorption spectra and which are expected to be relevant to the collective dipole excitations in high-T_c superconductors, we discuss them briefly as follows.

Schlesinger, Collins, and Shafer[39] have observed experimentally that the reflectivities with the broad bandwidth just below $\simeq 5000\ \text{cm}^{-1}$ (0.62 eV) increase gradually as a function of x in $La_{2-x}Sr_xCuO_4$. Herr *et al.*[40] have found two broad infrared peaks at 0.44 and 1.3 eV. Similar results have been observed by Geserich, Sheiber, and Renker,[41] Etemad *et al.*,[42] and Orenstein and co-workers.[43,44] Ohbayashi *et al.*[45] have found a 1400–1430-cm^{-1} (0.18-eV) peak for x lying between 0.1 and 0.2 in $(La_{1-x}Ba_x)_2CuO_4$ and no peak for $x=0$ (when the substance is a nonsuperconductor). Interestingly, the peak at around 1.3 eV appears for both a superconductor[40] and a nonsuperconductor[46,47] in this class of substances.

Kamarás and co-workers[48,49] have claimed that the broad peaks that they observed at 0.37 and 2.5 eV in $YBa_2Cu_3O_{6.9}$ (no peak observed at 0.37 eV in the $YBa_2Cu_3O_{6.2}$ nonsuperconductor) may be due to excitons. However, Bozovic *et al.*[50] criticized the conclusion of Kamarás and co-workers as to the excitonic peaks based on the fact that the observed peak happened to be broad in contrast to the excitonic peaks, which are usually narrow. Furthermore, the observed peaks show little temperature dependence in Y-Ba-Cu-O superconducting films, unlike that of excitons.

These characteristics of observed infrared absorptions are also common features of other oxide superconductors, including $BaPb_xBi_{1-x}O_3$,[51] a non-Cu-oxide superconductor.

Our present calculations strongly indicate that the observed infrared absorption peaks are due to the collective dipole excitations owing to the fact that we have found consistency between the calculated collective dipole excitation frequencies for various superconductor crystals and the observed peaks in their optical spectra along with the existence of the high-T_c superconductivity and the presence of collective dipole excitations in the CuO_2

planes without any exceptions. The calculated characteristic collective excitation frequencies are of the right order of magnitude ($\sim 10^{14}$ Hz) as predicted previously[1] and, in particular, show the correct variation with δ in $YBa_2Cu_3O_{7-\delta}$ as deduced previously.[1] Moreover, the collective dipole excitation peaks (corresponding to dipoles not lying in the CuO_2 planes) calculated in the nonsuperconducting crystals also agree with the peaks observed in experiments in these systems.

Table XI lists the energy peaks deduced from the collective-dipole-excitation dispersion relations and the observed peaks in various superconductors along with the expected (according to the present mechanism) and observed high-T_c superconductivity behavior. We note that the energies of the peaks at 2.5 and 0.36 eV derived from our calculations of the dispersion relations in $YBa_2Cu_3O_{7-\delta}$ and the peaks at 1 and 0.17 eV in the $La_2(Sr,Ba)CuO_4$ system agree very well with the experimental peaks. For $YBa_2Cu_3O_{7-\delta}$ the calculations show the presence of one more peak at 0.08 eV, which has the tendency to merge with the peak at 0.36 eV as δ increases, and because of this it may be difficult to identify experimentally. We also find that the calculated peaks due to collective dipole excitations are always much broader than exciton peaks, as is clear from Figs. 4, 5, 6, 7, and 8 of the density of states and the calculated dispersion relations. The peaks will be even broader for forced collective dipole oscillations if induced by external sources in the experiments. Further, the infrared spectra in thin-film superconductors could be broader than in bulk samples because of the more dipole fluctuations expected, and the power of the light beam may overwhelm the strength of the dipole couplings.

Yang, Zhu, and Xu[52] pointed out the importance of broken symmetry and lattice distortions, which seem to cause the local polarizations to yield ferroelectricity as well as superconductivity. Our calculations show that some ferroelectric properties can be expected, but the net polarization should be zero owing to the unique dipole arrangement in a unit cell in these systems. Arguing against their view is the fact that the directions of the calculated static dipoles we have found are perpendicular to the CuO_2 planes.

In the present calculations the somewhat uncertain parameter is the oxygen electronic polarizability. We found that the dipole at O(3) sites in $YBa_2Cu_3O_7$ is $0.187e$ Å for Pauling's polarizability 3.88 $Å^3$, which becomes $0.13e$ Å for the Tassman, Kahn, and Shockley (TKS) polarizability 2.4 $Å^3$.[25] Although the magnitudes of the dipoles change significantly by changing polarizabilities, we find that the collective dipole excitation frequencies remain almost the same provided that all the physical parameters involved in the calculations are properly accounted for, for the changes in the polarizabilities. This is clear if one first notes that, because the electric fields that produce dipoles are calculated self-consistently, the magnitude of the dipoles is not directly proportional to the polarizability values. In fact, an increase in the polarizability effectively reduces the total electric field. Furthermore, the frequency of collective dipole oscillations involves the quantity $(p^2/\alpha I)^{1/2}$ where the moment of inertial $I \propto \alpha^{2/3}$. As a combined effect of all these, one finds almost no change in the calculated collective dipole excitation frequencies. We further emphasize that the polarizability of oxygen smaller than the TKS value is unacceptably small, in general.

There still exist some arguments as to what the correct values of the effective charges of the ions are that are crit-

TABLE XI. Lists of the energy peaks derived from the collective dipole excitation dispersion relations and the observed peaks in various superconductors along with the expected (according to the collective dipole mechanism) and observed high-T_c superconductivity (HTSC) behavior.

System	Theory Peaks in $F(\omega)$ (eV)	HTSC	Experiment Optical abs. (eV)	HTSC (T_c)
$Ba_{1-x}K_xBiO_4$		Yes		Yes (0-29.8 K)
La_2CuO_4 (undoped)	1.0	No	1.2	No (0 K)
$La_2CuO_{4-\delta}$ (distorted)	0.17, 1.0	Yes		
$La_{2-x}Ba_xCuO_4$	~0.17, 1.0	Yes	0.18, 1.3	Yes (0–40 K)
$La_{2-x}Sr_xCuO_4$	~0.17, 1.0	Yes	~0.5, 1.2	Yes (0–37.5 K)
$La_{2-x}Na_xCuO_4$	~0.17, 1.0	Yes		Yes (0–40 K)
$La_{2-x}Ca_xCuO_4$	~0.17, 1.0	Yes		Yes (0–18 K)
Nd_2CuO_4 (undoped)		No		No (0 K)
$Nd_{2-x}Ce_xCuO_4$		Yes		Yes (0–24 K)
$Nd_{2-x}Sr_xCuO_4$		Yes		Yes (0–28 K)
$Bi_2Sr_2CuO_6$ (undoped)	1.7, 3.75	No		No (≤ 6 K)
$Tl_2Ba_2CuO_6$ (undoped)		No		No[a] (0–20 K)
$TlBa_2CuO_5$ (undoped)		No		No (0 K)
$YBa_2Cu_3O_7$	0.08, 0.36, 2.5	Yes	0.37, 2.5	Yes (92 K)
$Bi_2Sr_2CaCu_2O_8$	~0.3, 1.7, 2.5–3.3	Yes		Yes (85 K)
$Tl_2Ba_2CaCu_2O_8$	~0.3, 2.5–4.7	Yes		Yes (105 K)
$Bi_2Sr_2Ca_2Cu_3O_{10}$		Yes		
$Tl_2Ba_2Ca_2Cu_3O_{10}$	~0.4, 2.0, 4.6–7.2	Yes		Yes (125 K)

[a] T_c is extremely sample dependent.

ical in many current theories of high-T_c superconductivity. The correct values of the effective charges of the ions if inferred through experiments are expected to improve our calculations but not to change our results drastically, since if one uses the simulated nonintegral effective charges of ions changed by up to 10%, one finds only small changes in the values of the dipoles and the relevant collective dipole excitation frequencies.

Also, the screening effect, if expected to be significant for a given system, should not be ignored for more accurate calculations. We have neglected this effect in the present calculations simply because the carrier density for the systems under investigation is low so that the distances between the electrons are as large as about 10 Å. If the carrier density is found to be high in certain cases, the screening effect can easily be incorporated in our collective dipole mechanism by the introduction of a screening factor.

As for the moment of inertia I of the dipoles, we have adopted two alternate ways for its estimation for all cases. One method is based on the use of the polarizability and the other on the calculations of the moment of inertia matrix elements employing Yamashita's atomic wave functions[36] of the oxygen ions. Both values turn out to be very close. The calculated value of I has been made use of in our present calculations.

The frequency of collective dipole excitations in $YBa_2Cu_3O_{7-\delta}$ as a function of δ decreases for increasing δ in our calculations. This can be understood in terms of the occupancies of oxygen ions at O(1) and O(5) sites, which become randomly occupied as the oxygen vacancies increase so that the magnitudes of dipoles in the CuO_2 planes not only become small but also fluctuate randomly. Consequently, the oscillations are effectively reduced in frequency and get damped. The intensity of absorption spectra or the density of states of these collective excitations associated with dipoles in the CuO_2 planes becomes broader as the fluctuations increase, and it ultimately disappears.

Under the proposed mechanism, it is not surprising to find the existence of high-T_c superconductivity in systems without having Cu atoms[20] or upon substitution of other highly polarizable atoms such as F^-, S^{2-}, and Cl^- (Ref. 53) instead of O^{2-} atoms. Non-Cu-oxide superconductors[20] such as $Ba_{0.6}K_{0.4}BiO_3$ ($T_c=29.8$ K), $Ba_{0.6}Rb_{0.4}BiO_x$ ($T_c=28.6$ K) and others[54] have already been discovered, and $Ba_2Sr_xCa_{1-x}CuO_6$ is expected to be a superconductor, which warrants investigation. Doped La_2NiO_4 is a superconductor under the present mechanism, but its superconductivity is expected to be suppressed by the magnetic ions Ni^{2+}. Also, the present mechanism predicts that the single-layered systems are lower-T_c superconductors relative to the multilayered systems as discussed earlier.

Now as to the question of obtaining higher-T_c superconductors, it is clear that first of all one needs to have a higher density of states of electrons at the Fermi level. Secondly, the dipoles in the conducting planes should be large in magnitude. The multilayered structures may help in this regard, since these structures have a lack of inversion symmetry at the oxygen sites in the conducting planes and have large and well-arranged dipoles. However, one must be cautious in that values of the dipoles that are too large may suppress T_c because of the strong static attraction between the electrons and the dipoles, which may lead to a bound-state system (the critical values of the dipole moment for obtaining a quantized bound state with an electron are estimated by Fox[55] as the lowest value $0.343e$ Å and the next lowest $2.064e$ Å, etc.).

The phonon contribution relative to the collective dipole contribution to high-T_c superconductivity is small, as indicated by the observed isotope effect.[56–61] The combined effect of both phonons and collective dipole excitations are required for further investigation, and relevant work is in progress. One notes that the phonon contribution may be relatively significant where the dipoles in the conducting planes are small as in the single-layered superconductors. This argument may explain why the isotope effect[56–61] for the single-layered crystals is larger than for the multilayered crystals and, in general, why it is larger in one case than in the other.

It is also interesting to discuss the pressure effect in the high-T_c superconductors. We have estimated the pressure effect by reducing the lattice constants due to pressure and thereby obtaining changes in the dipoles in $YBa_2Cu_3O_7$ crystals. The calculated results show that the magnitudes of the dipoles increase slowly at the O(4) site and remain constant at the oxygen sites O(2) and O(3) in the CuO_2 plane as the pressure increases. Since the T_c depends on the dipoles on the oxygen sites O(2) and O(3) in the conducting planes, this result yields that T_c does not change very much as the pressure changes, and this agrees very well with experimental observations.[32–34] Similar behavior is also expected for the multilayered structures considering the similarity in their structures and taking account of the number of oxygen sites with dipoles in the conducting planes for a given size of the crystal. However, for the single-layered superconductors of the La-Sr-Cu-O system, where the dipoles in the CuO_2 planes are generated by the lattice distortion due to the impurities, the system is very sensitive to pressure variations. Thus relatively we expect the changes in the T_c values per unit pressure in the single-layered superconductors to be much larger than in the multilayered superconductors, in agreement with experimental observations.[28–34]

Previous suggestions[62–64] as to the bosons required for the pairing of the carriers in high-T_c superconductors include collective valence-electron excitations in conjunction with the polarization effects of the various ions in the system that effectively give rise to the altered ionic plasma frequencies. These excitations do not require the lack of inversion symmetry at the polarizable ions in the system, as in the present paper, and, hence, are quite different from the collective dipole excitations presented here.

In short, we have calculated the collective dipole excitation frequencies, dispersion relations, and the density of states in various superconducting systems (including a single oscillating dipole and a linear and a planar system of oscillating dipoles) and found that the relevant collective dipole excitation frequencies are of the right order of magnitude ($\sim 10^{14}$ Hz) as predicted by the collective dipole mechanism[1–3] for the explanation of T_c versus δ in the superconductor $YBa_2Cu_3O_{7-\delta}$. The calculated density of states for collective dipole excitations provide an explanation for the observed optical absorption data in various high-T_c superconductors. In particular, the peaks at 2.5 and 0.36 eV in $YBa_2Cu_3O_{7-\delta}$ and the ones at 1 and 0.17 eV in $La_{2-x}Ba_xCuO_4$ system derived from the collective-dipole-excitation density of states agree with the optical observations in these systems. Furthermore, the optical absorption peaks observed in the nonsuperconducting crystals are identified to be due to the collective dipole excitations corresponding to the dipoles that do not lie in the CuO_2 planes (there are no dipoles in the CuO_2 planes in nonsuperconductors). The variation of the collective dipole excitation frequencies as a function of δ in $YBa_2Cu_3O_{7-\delta}$ also agrees with our previous deduction.[1] Various predictions as to the possibility of finding non-Cu-oxide superconductors (including La_2NiO_4) and ones containing the polarizable ions other than oxygen have been discussed. The crystalline electric fields are essential for the existence of high-T_c superconductivity in view of the collective dipole excitation mechanism as these fields generate electronic dipoles on highly polarizable atoms which, in turn, produce the collective excitations. The collective dipole excitation quanta act as mediators of pairing in high-T_c superconductivity. The well-arranged electronic dipoles and appropriate collective dipole excitation frequencies in high-T_c superconductor crystals are so special that we may associate specific collective dipole oscillations in the conducting planes with the observed high-T_c superconductivity.

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