Dipolon-plasmon interaction in high-T_c superconducting materials

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(Received 6 November 1995; revised manuscript received 29 January 1996)

In this article interaction between the dipolon waves and the longitudinal plasma waves has been studied adopting a self-consistent field approach. The combined dispersion relation has been obtained which shows that both the dipolon waves and the plasma waves are modified. We discuss our results in relevance to free-electron plasma peaks observed at ~1.1 eV in inelastic electron scattering and optical spectroscopy measurements in high-temperature superconductors. Our calculations for $YBa_2Cu_3O_7$ show that the plasma frequency decreases whereas the slope of the plasmon dispersion curve at long wavelength increases because of interaction with dipolon excitations, consistent with the experimental result due to Romberg *et al.* Our investigation has made it possible to propose that the experimentally observed peak at ~1.1 eV may be due to plasmons interacting with dipolon excitations in high-temperature superconductors. [S0163-1829(96)02737-3]

I. INTRODUCTION

Since the discovery of high-temperature superconductivity by Bednorz and Müller[1] extensive experimental and theoretical research has been performed to study the properties of high-temperature superconductors. Experimental information as to the electronic structure and excitations has been derived from various techniques such as optical reflectivity measurements,[2–9] photoelectron spectroscopy,[10–12] electron-energy-loss spectroscopy,[13–18] x-ray-absorption spectroscopy,[19,20] inverse photoemission,[21,22] Raman measurements,[23] etc.

Several authors have directly observed the free-electron plasma peak at about 1.1 eV in inelastic electron-scattering experiments[13–18] and optical reflectivity measurements[2–9] in several high-temperature superconductors. In inelastic electron-scattering transmission measurements Tarrio and Schnatterly[15] have observed a 1.1 eV free-electron plasma peak in $YBa_2Cu_3O_{7-\delta}$ but they could not determine the origin of this peak. Romberg *et al.*[9] have investigated electronic excitations with polarization parallel to the CuO_2 *planes* in single crystalline $YBa_2Cu_3O_7$ by optical reflectance and by high-energy electron-loss spectroscopy in transmission in the energy range from 50 meV to 6 eV and 0.2 to 150 eV, respectively. In their observed spectra the plasma edge near 1 eV was clearly visible in the metallic samples. They observed the plasma oscillations of energy $\varepsilon_{\mathrm{pl}}(q)$ as a function of the wave number q which may be expressed as

$$\varepsilon_{\mathrm{pl}}(q)=\varepsilon_{\mathrm{pl}}(0)+Aq^2 \tag{1}$$

with $\varepsilon_{\mathrm{pl}}(0)=1.4$ eV which is smaller than the Drude plasma frequency ω_p (=2.75 eV) and $A\simeq3.3$ eV Å^2 which is about three times the expected value of $A\simeq1.0$ eV Å^2 for the plasma oscillations. From the analysis of their results they also deduced that the effective mass associated with the plasmons is near the free-electron mass. Furthermore, the plasma oscillations disappeared in the semiconducting samples. A similar situation exists in other investigations as regards the origin of this peak.[2–8,13–14,16–18] From the detailed form of the observed frequency and temperature dependence of the conductivity in $YBa_2Cu_3O_{7-\delta}$ Thomas *et al.*[5] have predicted that the electrons interact strongly with some (unknown) spectrum of excitations to produce this (1.1 eV) peak.

Recently Lee and Sharma[24] have calculated the dispersion relations and density of states in several high-temperature superconductors for the dipolon excitations which are the quantized self-sustained collective oscillations of the crystal-field-generated electronic dipoles of the polarizable (oxygen) ions and obey Bose statistics. These calculations establish theoretically the existance of dipolon excitations in high-temperature superconductors. The dipolons have been suggested by these authors[25] as the possible mediators for the carrier-carrier pairing mechanism in high-temperature superconductors. In addition, the experimentally observed broad peaks at 2.5 and 0.36 eV in $YBa_2Cu_3O_{7-\delta}$,[3,26] the observed broad peaks at 1.0 and 0.17 eV in $La_{2-x}Ba_xCuO_4$,[27,28] and the observed peak in $HgBa_2CuO_{4+\delta}$ at about 195 cm^{-1},[23] which agree with our corresponding calculated dipolon excitations,[24,25,29] provide further validity for the existence of dipolon excitations, particularly when no other acceptable explanation has yet been available for the existence of such peaks.[30] This conclusion is further strengthened by the fact that the dipolon theory provides a possible explanation not only for T_c values in the $HgBa_2CuO_{4+\delta}$ high-temperature superconductors[29] but also for the T_c and the variation of T_c as a function of the oxygen-stoichiometry parameter δ in $YBa_2Cu_3O_{7-\delta}$,[24,25] and for the variation of T_c with the hydrostatic pressure, by first-principles without fitting with any parameters, in $YBa_2Cu_3O_{6.93}$ and $YBa_2Cu_3O_{6.60}$.[31]

In Refs. 24 and 25 it has been emphasized that the crystal fields induce dipole moments on the polarizable ions. By self-consistent treatment one obtains a static electric field $\mathbf{E}_0(\mathbf{r}_i)$ and an induced dipole moment $\mathbf{p}_{0i}$ which are related by the polarizability α_i of the ion at the site $\mathbf{r}_i$ as

$$\mathbf{p}_{0i}=\alpha_i\mathbf{E}_0(\mathbf{r}_i). \tag{2}$$

These induced dipoles have been shown in Ref. 24 to have self-sustained collective oscillations, *dipolons*, which involve fluctuations in the alignments of the dipoles. The dipolon frequency ω_d of oscillations in high-temperature su-

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perconductors turns out to be $\sim 10^{14}$ Hz which is much larger than the Debye frequencies and smaller than the plasmon frequencies in high-temperature superconductors. Since the plasma oscillations are purely electrical in nature and dipolons produce electrical oscillating fields, they are expected to interact.

In the present paper we present the results of our investigation of the interaction of dipolons with the plasma oscillations. Some scattered parts of our results have already been presented previously[32] in a somewhat different context.[33] In our treatment the dipolon and plasmon motions are coupled by means of the total fluctuating electric field which is the sum of the electric fields produced by plasmons and dipolon excitations. We find that both the plasmons and the dipolons are mutually affected.

In the next section we present the relevant theory and consider two cases which deal with the dipolons on a linear lattice and on a planer lattice. The calculations and discussion appropriate to high-temperature superconducting materials, in particular, the $YBa_2Cu_3O_7$ system are presented in Sec. III followed by the conclusion in Sec. IV.

II. THEORY

We consider that the crystal under investigation is comprised of dipoles on the lattice sites embedded in a distribution of free electrons. The dipoles on the lattice are generated by the crystalline electric fields and oscillate in the system to constitute dipolons whereas the free-electron oscillations give rise to solid-state plasmons. In the following we derive formulas for the frequency and dispersion relations for the coupled dipolon-plasmon interaction.

In order to be devoid of any confusion[32,33] we start with the most basic equations. The continuity equation for the electrons is

$$\frac{\partial \varrho}{\partial t}+\boldsymbol{\nabla}\cdot\mathbf{j}=0, \tag{3}$$

which, with the electron charge density ϱ

$$\varrho=n(-e) \tag{4}$$

and the electron current density $\mathbf{j}$

$$\mathbf{j}=n(-e)\mathbf{v}, \tag{5}$$

n being the electron number density, yields

$$\frac{\partial n}{\partial t}+\boldsymbol{\nabla}\cdot(n\mathbf{v})=0. \tag{6}$$

The force equation corresponding to the motion of an electron of mass m and velocity $\mathbf{v}$ is given by

$$\frac{\partial \mathbf{v}}{\partial t}=\frac{(-e)\mathbf{E}}{m}-\frac{\beta^2\boldsymbol{\nabla}n}{n}, \tag{7}$$

where $\mathbf{E}$ is the net electric field experienced by the electron and the parameter β is defined as

$$\beta=\sqrt{0.6}\,v_F \tag{8}$$

with v_F as the Fermi velocity.

Equation (6) leads to

$$\frac{\partial^2 n}{\partial t^2}+\boldsymbol{\nabla}\cdot\left(\frac{\partial}{\partial t}\,(n\mathbf{v})\right)=0, \tag{9}$$

which with Eq. (7) gives

$$\frac{\partial^2 n}{\partial t^2}+\boldsymbol{\nabla}\cdot\left(\frac{\partial n}{\partial t}\,\mathbf{v}\right)-\frac{e}{m}\,\boldsymbol{\nabla}\cdot(n\mathbf{E})-\beta^2\boldsymbol{\nabla}\cdot\left(\frac{\boldsymbol{\nabla}n}{n}\right)=0. \tag{10}$$

Separating the static and dynamic parts we write

$$n=n_0+n_1 \tag{11}$$

and

$$\mathbf{E}=\mathbf{E}_0+\mathbf{E}_1, \tag{12}$$

where the subscript 0 designates the static part and the subscript 1, the dynamic part.

Retaining only the first-order terms in Eq. (10) one obtains the second-order space- and time-dependent differential equation

$$\frac{\partial^2 n_1}{\partial t^2}=\frac{n_0 e}{m}\,\boldsymbol{\nabla}\cdot\mathbf{E}_1+\frac{n_1 e}{m}\,\boldsymbol{\nabla}\cdot\mathbf{E}_0+\beta^2\boldsymbol{\nabla}^2 n_1. \tag{13}$$

As for the meaning of the approximation adopted in obtaining Eq. (13) and its validity area it must be mentioned that Eq. (13) is valid when the fluctuations n_1 and $\mathbf{E}_1$ are much smaller than the corresponding static values n_0 and $\mathbf{E}_0$, respectively. In other words, Eq. (13) describes the spatial and temporal variations of n_1 when $n_1 \ll n_0$ in the presence of the static field $\mathbf{E}_0$ and the spatial and time varying field $\mathbf{E}_1$ when $E_1 \ll E_0$.

The importance of the term $(n_1 e/m)\boldsymbol{\nabla}\cdot\mathbf{E}_0$ appearing in Eq. (13) has been emphasized previously.[32,33] For a general situation this term should not, *a priori*, be assumed to vanish even though for a uniform plasma it is zero.

The dipolon equation of motion is given by[24]

$$I_i\,\frac{\partial^2}{\partial t^2}\left[\frac{\mathbf{p}_{0i}\times\mathbf{p}_i}{p_{0i}^2}\right]=\mathbf{p}_i\times\mathbf{E}(\mathbf{r}_i), \tag{14}$$

where I_i is the moment of inertia tensor of the electron charge distribution at the ith site, and, in terms of static ($\mathbf{p}_{0i}$) and dynamic ($\mathbf{p}_{1i}$) parts, the dipole moment $\mathbf{p}_i$ is

$$\mathbf{p}_i=\mathbf{p}_{0i}+\mathbf{p}_{1i}. \tag{15}$$

Retaining only the first-order terms in Eq. (14) we have

$$I_i\,\frac{\partial^2}{\partial t^2}\left[\frac{\mathbf{p}_{0i}\times\mathbf{p}_{1i}}{p_{0i}^2}\right]=\mathbf{p}_{0i}\times\mathbf{E}_1(\mathbf{r}_i)+\mathbf{p}_{1i}\times\mathbf{E}_0(\mathbf{r}_i). \tag{16}$$

Further, the net electric field $\mathbf{E}$ is

$$\mathbf{E}=\mathbf{E}_{\rm mp}+\mathbf{E}_{\rm dip}+\mathbf{E}_{\rm pl}, \tag{17}$$

which is the sum of the electric field due to monopoles $\mathbf{E}_{\rm mp}$, the electric field due to dipoles $\mathbf{E}_{\rm dip}$, and the electric field due to plasma $\mathbf{E}_{\rm pl}$.

From Eqs. (12) and (17) we have

$$\mathbf{E}_0=\mathbf{E}_{0,\rm mp}+\mathbf{E}_{0,\rm dip}+\mathbf{E}_{0,\rm pl} \tag{18}$$

and

$$\mathbf{E}_1=\mathbf{E}_{1,\mathrm{dip}}+\mathbf{E}_{1,\mathrm{pl}} \tag{19}$$

where[24]

$$\mathbf{E}_{1,\mathrm{dip}}=\sum_i \widetilde{F}_{ij}\cdot\mathbf{p}_{1j} \tag{20}$$

with $(\widetilde{F}_{ij})$ as the Lorentz tensor

$$(\widetilde{F}_{ij})=\begin{vmatrix} \frac{3x^2-r^2}{r^5} & \frac{3xy}{r^5} & \frac{3xz}{r^5} \\ \frac{3yx}{r^5} & \frac{3y^2-r^2}{r^5} & \frac{3yz}{r^5} \\ \frac{3zx}{r^5} & \frac{3zy}{r^5} & \frac{3z^2-r^2}{r^5} \end{vmatrix} \tag{21}$$

and

$$\mathbf{E}_{1,\mathrm{pl}}(\mathbf{r}_i,t)=\int \frac{(-e)n_1(\mathbf{r},t)(\mathbf{r}_i-\mathbf{r})}{|\mathbf{r}_i-\mathbf{r}|^2}\,d^3r, \tag{22}$$

which satisfies

$$\nabla\cdot\mathbf{E}_{1,\mathrm{pl}}=-4\pi e n_1. \tag{23}$$

From Eqs. (19), (20), and (23) one writes

$$\nabla\cdot\mathbf{E}_1=-4\pi e n_1+\nabla\cdot\sum_j \widetilde{F}_{ij}\cdot\mathbf{p}_{1j}. \tag{24}$$

The dynamic parts may be expressed in terms of the propagating wave with wave vector $\mathbf{q}$ and frequency ω as

$$\mathbf{E}_1(\mathbf{r},t)=\mathbf{E}_1^0\exp[i(\mathbf{q}\cdot\mathbf{r}-\omega t)], \tag{25}$$

$$n_1(\mathbf{r},t)=n_1^0\exp[i(\mathbf{q}\cdot\mathbf{r}-\omega t)], \tag{26}$$

$$\mathbf{p}_1(\mathbf{r},t)=\mathbf{p}_1^0\exp[i(\mathbf{q}\cdot\mathbf{r}-\omega t)], \tag{27}$$

where, in general, $\mathbf{q}$, $\mathbf{E}_1^0$ and $\mathbf{p}_1^0$ are in arbitrary directions.

Considering the variations given by Eqs. (25)–(27) in Eqs. (13), (16), and (24) and assuming that all lattice sites are identical so that p_{0i}, I_i, and α_i are independent of i, we obtain, after dropping the subscript i in p_{0i}, I_i, and α_i, the secular equation for the dipolon-plasmon coupled oscillations

$$\det\begin{vmatrix} \omega^2-\beta^2q^2+\frac{e}{m}\nabla\cdot\mathbf{E}_0 & \frac{in_0e\mathbf{q}\cdot\hat{E}_1^0}{m} & 0 \\ 0 & \alpha\omega_0^2 & \omega^2-\omega_0^2 \\ -4\pi e & -i\mathbf{q}\cdot\hat{E}_1^0 & i\mathbf{q}\cdot\Sigma_j\widetilde{F}_{ij}\cdot\hat{p}_1^0\exp[i(\mathbf{q}\cdot\mathbf{r}_j)] \end{vmatrix}=0 \tag{28}$$

assuming, for simplification, that $\hat{E}_1^0$ (unit vector along $\mathbf{E}_1^0$) is parallel to $\hat{p}_1^0$ (unit vector along $\mathbf{p}_1^0$). In Eq. (28)

$$\omega_0^2=p_0^2/\alpha I \tag{29}$$

and p_0 is the magnitude of the static dipoles assumed to be the same on all the lattice sites.

In Eq. (28) $\nabla\cdot\mathbf{E}_0$ is to be evaluated at the site of the dipole under consideration. For longitudinal wave the solution of Eq. (28) yields the dipolon-plasmon coupled frequency ω as

$$\omega^2=\frac{1}{2}\left[\left(\omega_1^2-\frac{e}{m}\nabla\cdot\mathbf{E}_0+\omega_2^2\right)\pm\left\{\left(\omega_1^2-\frac{e}{m}\nabla\cdot\mathbf{E}_0-\omega_2^2\right)^2-4\omega_0^2S\omega_p^2\right\}^{1/2}\right], \tag{30}$$

where ω_1 is the frequency of the noninteracting plasmon wave given by

$$\omega_1^2=\omega_p^2+\beta^2q^2 \tag{31}$$

and ω_2 is the frequency of the noninteracting dipolon wave given by

$$\omega_2^2=\omega_0^2(1-S) \tag{32}$$

with

$$\omega_p^2=\frac{4\pi n_0e^2}{m}, \tag{33}$$

which corresponds to the Drude value of the plasma frequency, and

$$S=\alpha\sum_j \hat{q}\cdot\widetilde{F}_{ij}\cdot\hat{p}_1^0\exp[i(\mathbf{q}\cdot\mathbf{r}_j)] \tag{34}$$

subject to the condition that $\hat{q}$ (unit vector along $\mathbf{q}$) is in the direction of $\hat{E}_1^0$.

In the first order in S we have

$$\omega^2\simeq\begin{cases}\left(\omega_1^2-\frac{e}{m}\nabla\cdot\mathbf{E}_0\right)-\frac{\omega_0^2S\omega_p^2}{[\omega_1^2-(e/m)\nabla\cdot\mathbf{E}_0-\omega_2^2]}, \\ \omega_2^2+\frac{\omega_0^2S\omega_p^2}{[\omega_1^2-(e/m)\nabla\cdot\mathbf{E}_0-\omega_2^2]}.\end{cases} \tag{35}$$

Assuming that

$$\omega_1^2\simeq\omega_p^2\gg\omega_0^2 \tag{36}$$

and that

$$\left|\frac{e}{m}\,\nabla\cdot\mathbf{E}_0\right|\sim\omega_p^2, \tag{37}$$

Eq. (35) yields

$$\omega^2\simeq\begin{cases}\left(\omega_1^2-\dfrac{e}{m}\,\nabla\cdot\mathbf{E}_0\right)-\dfrac{\omega_0^2S\omega_p^2}{[\omega_p^2-(e/m)\nabla\cdot\mathbf{E}_0]},\\ \omega_2^2+\dfrac{\omega_0^2S\omega_p^2}{[\omega_p^2-(e/m)\nabla\cdot\mathbf{E}_0]}.\end{cases} \tag{38}$$

For numerical evaluations, first, one estimates S from Eqs. (21) and (34) in a given situation and then uses Eq. (30) [or Eq. (38)] to obtain the frequency ω of the dipolon-plasmon coupled oscillations knowing from Eqs. (31) and (32) the uncoupled frequencies for the plasmon and dipolon oscillations. In the following we give explicit expressions useful for the calculations of the dipolon-plasmon interaction for a linear and a planar system of dipoles.

A. Linear lattice of dipoles

Let there be equivalent equidistant dipoles $\mathbf{p}_0$ arranged in a linear lattice along the x direction with lattice constant a and orientation of the (static) dipoles perpendicular to the x axis. Equation (34) then gives S, for a longitudinal wave propagating along the x axis,

$$S=\alpha\sum_{n\geq 1}\frac{2}{n^3a^3}\times 2\cos(qna), \tag{39}$$

which may be used in Eqs. (30) [or Eq. (38)] to calculate ω, the frequency of the coupled oscillations in this case. The uncoupled dipolon frequencies obtained from Eqs. (32) and (39) revives the corresponding result given in Ref. 24.

B. Planar lattice of dipoles

Next we consider a planar lattice of equivalent static dipoles $\mathbf{p}_0$ oriented along the z axis and arranged in the x-y plane with lattice constants a and b. For a longitudinal wave propagating along the x axis with wave number q, S may be written down from Eq. (34) explicitly as

$$S=\alpha\sum_{n\geq 0}\sum_{m\geq 0}(1-\delta_{n,0}\delta_{m,0})\left[\frac{2n^2a^2-m^2b^2}{(n^2a^2+m^2b^2)^{5/2}}\right]\times(2-\delta_{n,0})(2-\delta_{m,0})\cos(qna). \tag{40}$$

Equation (40) may be used along with Eqs. (29)–(33) to calculate the frequency ω for the coupled oscillations for the longitudinal wave propagating along the x axis.

As for investigating the behavior of the coupled oscillations in the *limit* $q\to 0$ we expand $\cos(qna)$ in Eq. (40) in powers of q and retain terms up to second order in q to write

$$S\simeq S_0-\frac{1}{2}\,S_1q^2, \tag{41}$$

where S_0 and S_1 are given by

$$S_0=\alpha\sum_{n\geq 0}\sum_{m\geq 0}(1-\delta_{n,0}\delta_{m,0})\left[\frac{2n^2a^2-m^2b^2}{(n^2a^2+m^2b^2)^{5/2}}\right]\times(2-\delta_{n,0})(2-\delta_{m,0}) \tag{42}$$

and

$$S_1=\alpha\sum_{n\geq 0}\sum_{m\geq 0}(1-\delta_{n,0}\delta_{m,0})n^2a^2\left[\frac{2n^2a^2-m^2b^2}{(n^2a^2+m^2b^2)^{5/2}}\right]\times(2-\delta_{n,0})(2-\delta_{m,0}). \tag{43}$$

Next, from Eq. (38) we obtain

$$\omega^2\simeq\begin{cases}\left\{\omega_p^2-\dfrac{e}{m}\,\nabla\cdot\mathbf{E}_0-\dfrac{\omega_0^2S_0\omega_p^2}{[\omega_p^2-(e/m)\nabla\cdot\mathbf{E}_0]}\right\}+\left\{\beta^2+\dfrac{\omega_0^2S_1\omega_p^2}{2[\omega_p^2-(e/m)\nabla\cdot\mathbf{E}_0]}\right\}q^2,\\ \omega_0^2\left\{1-\left(1-\dfrac{\omega_p^2}{[\omega_p^2-(e/m)\nabla\cdot\mathbf{E}_0]}\right)S_0\right\}-\left\{\dfrac{\omega_0^2S_1\omega_p^2}{2[\omega_p^2-(e/m)\nabla\cdot\mathbf{E}_0]}\right\}q^2,\end{cases} \tag{44}$$

which yields up to second order in q the coupled dipolon-plasmon frequencies

$$\omega\simeq\begin{cases}\left\{\omega_p^2-\dfrac{e}{m}\,\nabla\cdot\mathbf{E}_0-\dfrac{\omega_0^2S_0\omega_p^2}{[\omega_p^2-(e/m)\nabla\cdot\mathbf{E}_0]}\right\}^{1/2}+\dfrac{1}{2}\left\{\omega_p^2-\dfrac{e}{m}\,\nabla\cdot\mathbf{E}_0-\dfrac{\omega_0^2S_0\omega_p^2}{[\omega_p^2-(e/m)\nabla\cdot\mathbf{E}_0]}\right\}^{-1/2}\left(\beta^2+\dfrac{\omega_0^2S_1\omega_p^2}{2[\omega_p^2-(e/m)\nabla\cdot\mathbf{E}_0]}\right)q^2,\\ \omega_0\left\{1-\left(1-\dfrac{\omega_p^2}{[\omega_p^2-(e/m)\nabla\cdot\mathbf{E}_0]}\right)S_0\right\}^{1/2}-\dfrac{1}{2}\left\{1-\left(1-\dfrac{\omega_p^2}{[\omega_p^2-(e/m)\nabla\cdot\mathbf{E}_0]}\right)S_0\right\}^{-1/2}\left(\dfrac{\omega_0S_1\omega_p^2}{2[\omega_p^2-(e/m)\nabla\cdot\mathbf{E}_0]}\right)q^2.\end{cases} \tag{45}$$

To obtain results for ω for a longitudinal wave propagating along the y axis in the planar system one uses Eqs. (29)–(33) [or Eq. (45) as required] with the corresponding values of S, S_0, and S_1 which in this case may be obtained directly from Eqs. (40), (42), and (43), respectively, on interchanging a and b.

As for obtaining the frequencies of the coupled oscillations for a general direction of propagation of the longitudinal wave in the x-y plane the problem becomes more intricate since one needs to solve Eqs. (13), (16), and (24) for general direction of polarization. The frequencies and the polarization vectors for the noninteracting dipolon wave are required to be obtained from[24]

$$\begin{vmatrix} \omega_0^2[1-S_{x,x}(q_x,q_y)]-\omega^2 & \omega_0^2 S_{x,y}(q_x,q_y) \\ \omega_0^2 S_{y,x}(q_x,q_y) & \omega_0^2[1-S_{y,y}(q_x,q_y)]-\omega^2 \end{vmatrix} \begin{vmatrix} p_{1x}^0 \\ p_{1y}^0 \end{vmatrix} = \begin{vmatrix} 0 \\ 0 \end{vmatrix}, \tag{46}$$

where p_{1x}^0 and p_{1y}^0 are the Cartesian components of the polarization vector $\mathbf{p}_1^0$ and

$$S_{x,x}(q_x,q_y)=\alpha \sum_{n\geqslant 0}\sum_{m\geqslant 0}(1-\delta_{n,0}\delta_{m,0})\left[\frac{2n^2a^2-m^2b^2}{(n^2a^2+m^2b^2)^{5/2}}\right] \times(2-\delta_{n,0})(2-\delta_{m,0})\cos(q_x na)\cos(q_y mb), \tag{47}$$

$$S_{x,y}(q_x,q_y)=S_{y,x}(q_x,q_y)=\alpha \sum_{n\geqslant 0}\sum_{m\geqslant 0}\left[\frac{3namb}{(n^2a^2+m^2b^2)^{5/2}}\right] \times 4\sin(q_x na)\sin(q_y mb), \tag{48}$$

and

$$S_{y,y}(q_x,q_y)=\alpha \sum_{n\geqslant 0}\sum_{m\geqslant 0}(1-\delta_{n,0}\delta_{m,0})\left[\frac{2m^2b^2-n^2a^2}{(n^2a^2+m^2b^2)^{5/2}}\right] \times(2-\delta_{n,0})(2-\delta_{m,0})\cos(q_x na)\cos(q_y mb), \tag{49}$$

For a particular case of $a \simeq b$, the above equations show that pure longitudinal wave propagating along the [110] direction are sustained by the system. In this case, the parameter S to be used in Eqs. (30), (32), and (38) is

$$S=S_{x,x}\left(\frac{q}{\sqrt{2}},\frac{q}{\sqrt{2}}\right)+S_{x,y}\left(\frac{q}{\sqrt{2}},\frac{q}{\sqrt{2}}\right), \tag{50}$$

where $S_{x,x}(q/\sqrt{2},q/\sqrt{2})$ and $S_{x,y}(q/\sqrt{2},q/\sqrt{2})$ are obtained from Eqs. (47) and (48), respectively, by substituting $q_x=q_y=q/\sqrt{2}$. Equation (50) may be derived from Eq. (46) in conjunction with Eq. (32); it is also possible to obtain it directly from Eqs. (21) and (34) by taking appropriately both $\hat{q}$ and $\hat{p}_1^0$ along the [110] direction. Just for completeness of presentation we verify that $\mathbf{E}_1$ [Eq. (19)], as required, is along $\mathbf{q}$ since $\mathbf{E}_{1,\text{dip}}$ [Eqs. (20) and (21)] comes out to be in the [110] direction and $\mathbf{E}_{1,\text{pl}}$ is taken in the direction of $\mathbf{E}_{1,\text{dip}}$ for plasmons to interact with dipolons.

III. CALCULATIONS AND DISCUSSION

Here we present calculations of the plasma frequencies interacting with the dipolon excitations. As we shall see the plasma frequency is reduced and the slope A [Eq. (1)] in the variation of plasma frequency as a function of q^2 is increased, consistent with the experimental observation.

In the previous section we have presented for the case of planar lattice of dipoles the expressions which lead to the modification of the plasmon as well as the dipolon frequencies and dispersion relations as a result of the dipolon-plasmon interaction. These expressions are relevant to the investigation of the dipolon-plasmon interaction in high-temperature superconductors as the induced dipoles and associated dipolons corresponding to the oxygen ions [particularly, the O(3) ions in the $YBa_2Cu_3O_7$ system] lie in a plane (the CuO_2 plane).

Next, we need information as to the values of the various parameters required for our calculations. Our self-consistent calculations[24] in $YBa_2Cu_3O_7$ yield the value of the static dipoles on O(3) ions as $p_0=0.19\ e$ Å; the estimated value of the moment of inertia is $I=5.4\times10^{-43}$ g cm^2. As for the polarizability of oxygen we use the Pauling's polarizability α=3.88 Å^3.[34,35]

The lattice constants for $YBa_2Cu_3O_7$ are $a=3.823$ Å and $b=3.886$ Å as given by Jorgensen *et al.*[36] The value of the Fermi velocity has been estimated by Nücker *et al.*[13] as $v_F=0.6\times10^8$ cm/s. The free-electron density is found to be $n_0=5.4\times10^{21}$ cm^{-3} as given by Tarrio and Schnatterly.[15] This value of n_0 gives the Drude plasma frequency $\omega_p=2.73$ eV. As for the value of $\nabla\cdot\mathbf{E}_0$, in absence of any known procedure for its estimation in the present situation, we have taken it equal to $4\pi|e|4.2\times10^{21}$ cm^{-3} which corresponds to the associated effective charge density 4.2×10^{21} $|e|$ cm^{-3}. There is no *a priori* reason for taking this value but it, as will be seen in the following, gives the correct estimates for the plasma energy and the slope of the dispersion relation; it corresponds to a single charge $|e|$ distributed inside the sphere of radius equal to the cell dimension a (3.823 Å).

Knowing the values of the parameters as mentioned above it is straightforward to calculate the coupled dipolon-plasmon frequencies ω as a function of $\mathbf{q}$ from the expressions derived in the previous section. First, we calculate the longitudinal wave propagating along the crystallographic a direction in the a-b CuO_2 *planes*. From expressions (29) and (33) we find $\omega_0=1.05\times10^{14}$ Hz and $\omega_p=2.73$ eV. Next,

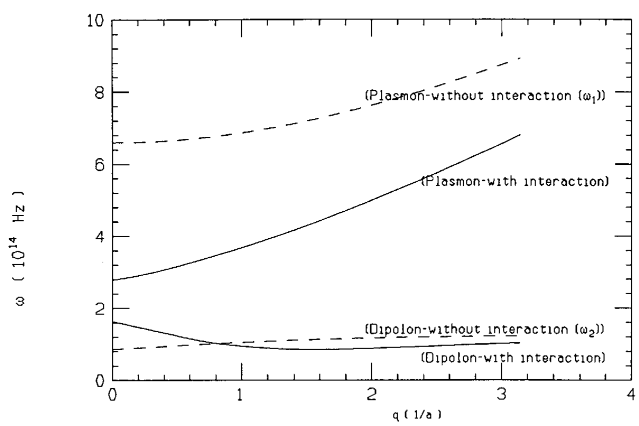

FIG. 1. Depicts the calculated values of the dipolon-plasmon frequencies (in units of 10^{14} Hz) as a function of wave number q (in units of $1/a$) for longitudinal waves propagating along the [100] direction in the $YBa_2Cu_3O_7$ high-temperature superconductor. Dashed lines show the noninteracting plasmon (ω_1) and noninteracting dipolon (ω_2) frequencies whereas the solid lines show the interacting plasmon and dipolon frequencies.

from Eqs. (31), (32), and (40) we calculate the noninteracting plasma wave frequency ω_1 and the noninteracting dipolon wave frequency ω_2 as a function of q which have been shown in Fig. 1 by dashed lines with frequencies in units of 10^{14} Hz and q in units of $1/a$. For $q=0$, as it is clear from Fig. 1, the noninteracting plasma frequency is $\omega_1=\omega_p=6.60\times10^{14}$ Hz (2.73 eV), the Drude plasma frequency, whereas the noninteracting dipolon frequency $\omega_2=0.857\times10^{14}$ Hz (0.354 eV). One notes in Fig. 1 that both the noninteracting plasma wave frequency ω_1 and the noninteracting dipolon wave frequency ω_2 increase with q.

Further, we use Eq. (30) in conjunction with Eqs. (31)–(33) and Eq. (40) to calculate the coupled dipolon-plasmon frequencies as a function of q which have been shown in Fig. 1 by solid lines. Perusal of Fig. 1 reveals that at $q=0$, the interacting plasma frequency is 2.78×10^{14} Hz (1.15 eV) and the interacting dipolon frequency is 1.63×10^{14} Hz (0.675 eV). Furthermore, the interacting plasma wave frequency increases with q whereas the interacting dipolon wave frequency decreases with q. Thus the plasma wave frequency has decreased to 1.15 eV from its Drude value 2.73 eV as a result of interactions with dipolons. Our calculated value (1.15 eV) of the plasma frequency agrees with the experimental value observed in various high-temperature superconductors.[2–9,13–18]

Comparing the slopes of the interacting plasma wave frequency (solid line—with interaction—in Fig. 1) with the noninteracting plasma wave frequency ω_1 (dashed line—without interaction—in Fig. 1) as a function of q, Fig. 1 shows that the slope of the plasma wave frequency has increased with interaction with dipolons, consistent with the experimental observations.[9]

In order to derive quantitative information about the slopes of the plasma dispersion relations near *limit* $q\rightarrow0$ we first write from Eq. (31) for the noninteracting plasma wave

$$\omega_1=\omega_p+A^{(0)}q^2, \tag{51}$$

where the slope $A^{(0)}$ is

$$A^{(0)}=\frac{\beta^2}{2\omega_p}=\frac{3v_F^2}{10\omega_p}. \tag{52}$$

Employing the values of v_F and ω_p as given above we obtain $A^{(0)}=1.7$ eV Å^2 which is slightly higher than the value $A^{(0)}=1.0$ eV Å^2 deduced by incorporating the effect of oxygen chains.[9]

On the other hand, from Eq. (45) the corresponding slope (which we denote by A) for interacting plasma wave is

$$A=\frac{1}{2}\left\{\omega_p^2-\frac{e}{m}\,\nabla\cdot\mathbf{E}_0-\frac{\omega_0^2S_0\omega_p^2}{[\omega_p^2-(e/m)\nabla\cdot\mathbf{E}_0]}\right\}^{-1/2}\times\left(\beta^2+\frac{\omega_0^2S_1\omega_p^2}{2[\omega_p^2-(e/m)\nabla\cdot\mathbf{E}_0]}\right). \tag{53}$$

In the nearest-neighbor approximation one deduces from Eqs. (42) and (43) that

$$S_0=\alpha\left(\frac{4}{a^3}-\frac{4}{b^3}\right) \tag{54}$$

and

$$S_1=\frac{4\alpha}{a}. \tag{55}$$

Therefore, from Eq. (53)

$$A=\frac{\beta^2}{2[\omega_p^2-(e/m)\nabla\cdot\mathbf{E}_0]^{1/2}}+\frac{\omega_0^2\alpha\omega_p^2}{a[\omega_p^2-(e/m)\nabla\cdot\mathbf{E}_0]^{3/2}}, \tag{56}$$

where we have taken $S_0\simeq0$ from Eq. (54) as $a\simeq b$ for the present system under consideration.

Using the values of the required parameters as given above the slope comes out to be, from Eq. (56), $A=3.7$ eV Å^2 which agrees with the slope $A=3.3(\pm0.9)$ eV Å^2 deduced experimentally by Romberg *et al.*[9] for the $YBa_2Cu_3O_7$ high-temperature superconductor. Interestingly, in the nearest-neighbor approximation for $q=0$ the interacting plasma wave frequency comes out to be 1.24 eV (which is slightly higher than 1.15 eV obtained above without nearest-neighbor approximation), closer to the observed value 1.24 eV.[9] As for the longitudinal wave propagating along the [010] direction, the results are the same as obtained above for the [100] direction since $a\simeq b$ in $YBa_2Cu_3O_7$ high-temperature superconductors.

We have also made calculations for the longitudinal wave propagating along the [110] direction in $YBa_2Cu_3O_7$ high-temperature superconductors using Eqs. (30) and (50). The calculated results in this case are not significantly different than what we have obtained for the case of the wave propagating along the [100] direction as given above and, hence are not presented here.

Perusal of Fig. 1 shows that plasmons and dipolons tend to come closer (i.e., they attract) by coupling. Why this is so can be understood easily if one notes that both systems, the dipolons and plasmons, are subjected to the same net electric field [see Eq. (17)], which is the sum of the electric fields due to dipolons and due to plasmons. It means that the dipolons are subjected to the extra field (due to plasmons) which acts as a forcing field on the dipolons tending them (the dipolons) to oscillate with frequency close to the forcing field (the plasmons). Similarly, the plasmons are subjected to the extra field (due to dipolons) which tends to force plasmons to oscillate with the forcing field due to dipolons. In other words, both systems have forced oscillations, one is forced by the other. Since the dipolon frequency is lower than the plasmon frequency the dipolon frequency increases whereas the plasmon frequency decreases. Thus the dipolons and plasmons appear to attract one another when they interact.

Since the existence of the dipolon excitations has been established by first-principles calculations,[24] without making any (unphysical) assumptions, it is likely that the dipolons should be the common feature of many materials—not just confined to the cuprates. It might be very well conceived that they would be commonly found in ferro/paraelectrics. In ferro/paraelectrics the concept of dipolon excitations has not yet been introduced. It is because of this that there has been difficulty in assigning correctly the broadbands observed by neutron-diffraction and infrared experiments. As for instance, the 2800 cm^{-1} broadband observed[37] in KH_2PO_4, by means of inelastic scattering of cold neutrons, was assigned to H vibrational modes; this assignment was found to be incorrect as determined by infrared reflectance measurements which established 540 cm^{-1} as the frequency of the H mode. We propose that the 2800 cm^{-1} broadband observed in KH_2PO_4 could be due to dipolon excitations. As for another instance, two broadbands $E=3200-3600$ cm^{-1} and $F=3400-3540$ cm^{-1} observed[38] in Rochelle salt in Raman spectra was tentatively assigned to the water molecule. To the knowledge of the author there has not yet been any appropriate explanation available for these broadbands which may possibly be due to dipolon excitations in this system. Thus there are possibilities that dipolon excitations exist also in ferro/paraelectrics. This will be worth investigating by making detailed calculations of the dipolon excitations in such systems.

To a reader the dipolons may be a curious concept and it may present a number of conceptual problem. Thus a reader may tend to understand it (since plasmon excitations are very well known) as a plasmonlike excitation involving polarizing of a core electron. Further, it could be thought over that the lowest energy absorption would involve an interband transition since an optical absorption requires scattering an electron to an unoccupied final state. Furthermore, a reader may think that at higher energies there will be a conventional plasmon, with the plasmon frequency determined by the total density of the core electrons. The above concept is not the correct one as far as the dipolons are concerned. This is because the dipolon frequencies involve the moment of inertia I [see Eqs. (29) and (32)] of the electron charge cloud which is polarized by the presence of the crystalline electric field. Further, the dipolons are the quantized cooperative oscillations or fluctuations, as explained in Ref. 24, of the polarized electron charge cloud and the quantized oscillations act as bosons. Thus an optical absorption merely excites a boson (the dipolon) to its higher quantized energy state just as a phonon is excited by energy absorption to its higher quantized energy state (even though the charge cloud on the atom is displaced along with the displacement of the nucleus in case of phonons). In fact, the dipolon excitations are not single-electron excitations and furthermore, they are not the interband electron transitions but they involve multielectron collective excitations and only the highly correlated multielectron theory can explain such (dipolon) excitations (this theory has not yet been developed). This also explains why the dipolon excitations are not like plasmons. This can be made more clear by emphasizing that the plasmons are longitudinal waves. That is, in the plasmons the electron charge displacements are in the direction of the electric field along which the plasmon wave propagates forming the longitudinal modes whereas in the dipolon excitations there is no such constraint.[24]

The above discussion also clarifies that it is not a simple matter to add the dipolon contribution to the absorption in the sum rule in optical absorption of a solid. Only the highly correlated multielectron theory, when developed, can give the answer to this problem. Though the modern band-structure calculations[39] can, in principle, calculate the electronic contribution to the optical properties of the solids they are not yet capable of giving the energy levels of highly correlated electrons in solids so as to obtain electronic excitations equivalent to what one obtains by superimposing the dipolon excitations on the present day electronic band-structure calculations.

Since many available experimental data are obtained for polycrystalline samples (particularly, the experimental data referred to here), it could be possible that some confusion may arise as regards whether the experimentally observed broad peaks which we have proposed to identify with the dipolon excitations really correspond to the CuO_2 *planes* or some excitations corresponding to the c axis in the systems. To remove this confusion we mention that Koch, Geserich, and Wolf[40] performed polarized optical reflection measurements on nontwinned domains of the (001) surface of $YBa_2Cu_3O_7$ single crystals in the energy range between 0.05 and 6.0 eV. In the energy range 1–3 eV their polarized reflectance spectra agree with those of previous investigators.[41] By fitting their observed results by means of Lorentz-Drude models and by Kramers-Kronig analysis they were able to separate the contributions of the CuO_2 *planes* and of the Cu-O chains to the electronic transport properties of $YBa_2Cu_3O_7$ crystals. They concluded that the conductivity in the CuO_2 *planes* is represented by a Drude term, superimposed by a broadband 0.58 eV Lorentz oscillator. This value agrees with our calculated[31] value (obtained without fitting with any parameter) of 0.62 eV (1.51×10^{14} Hz) of the dipolon excitation in a $YBa_2Cu_3O_{6.93}$ superconductor at zero hydrostatic pressure, which reduces to 0.44 eV for $YBa_2Cu_3O_{6.60}$ (at zero hydrostatic pressure). Consistently, this also explains (without fitting with any parameter), in conjunction with similar calculations for the hydrostatic pressure $P=0.310$ GPa, not only the observed T_c values but also the observed variations of T_c with hydrostatic pressure in $YBa_2Cu_3O_{6.93}$ as well as in $YBa_2Cu_3O_{6.60}$ systems and explains the variation of T_c with δ in $YBa_2Cu_3O_{7-\delta}$ (as also mentioned briefly in Sec. I).

Furthermore, Koch, Geserich and Wolf[40] have concluded that the 0.58 eV broadband is not due to the interband transitions as the interband transitions require energy greater than 1.7 eV. It is worth remarking that for the broadband 0.58 eV (or, in general, 0.5 eV broadband in cuprates) referred to above, different interpretations have been proposed by various authors which involve charge transfer or excitonic excitations,[3,4] inelastic-scattering process,[5] valence-conserving $d-d$ transitions within the e_g orbitals of the $Cu^{2+}(2)$ ions,[42] localized excitations associated with the b *axis* Cu-O chains,[43] etc. However, these interpretations have been proposed without performing realistic calculations on the relevant systems. On the other hand, our dipolon calculations[31] (as also mentioned above) which yield the value (obtained without fitting with any parameter) of 0.62 eV (1.51×10^{14} Hz) of the dipolon excitation in the $YBa_2Cu_3O_{6.93}$ superconductor at zero hydrostatic pressure, which reduces to 0.44 eV for $YBa_2Cu_3O_{6.60}$ (at zero hydrostatic pressure), are based on first-principles calculations and hence, are more reliable. It must be emphasized that these values[31] do not depend on the choice of the effective charge density which we have used in the present work to investigate the dipolon-plasmon interaction and thus the reliability of the broadband values obtained from dipolon calculations referred to above is justifiable.

Quite recently Sawada *et al.*[30] have made an attempt to explain the broad peaks in high-temperature superconductors in the framework of the $d-p$ model and have concluded that it is not possible to explain the anomalously broad peaks extended up to several thousands of cm^{-1} observed in the experiments. Some discussion as regards Eq. (13) is also warranted. This is a linearized equation containing only the first-order fluctuation terms and for a uniform plasma leads, in conjunction with Eqs. (23) and (26), to the plasmon dispersion relation as described by Eq. (31), a result which one obtains by means of microscopic models[44,45] for oscillations of quantum plasma involving electrostatic interactions. Further, in the limiting situation when the electrostatic effects vanish (limit $e^2 \rightarrow 0$), it leads also correctly to the usual dispersion relation $\omega \propto q$ for phonons in a gas.[45] This type of treatment has been used by several investigators[46] to study various physical properties in different situations.

The equations we have derived above for the dipolon-plasmon interaction are valid for a three-dimensional system as the equations which we have used here to describe the plasmons and the dipolons are appropriate to a three-dimensional system. This is particularly clear if one notes that the dispersion equations for plasmons in lower dimensions are different than those in three dimensions.[47]

In the present treatment we have not studied the anisotropy in the dipolon-plasmon coupled oscillations in the a-b plane. The anisotropy is expected from our theory if one considers the effect of the oxygen ions of the b chains in the system as has been emphasized in Refs. 6–8. Consideration of this in the theory involving dipolons due to all the oxygen ions, including those out of the CuO_2 *planes*, to understand the a-b plane anisotropy, makes the problem much more complicated and forms the subject of future investigations. A brief report of the present results has been presented earlier.[48]

IV. CONCLUSION

The equations of motion of dipolons and plasmons are coupled because both are subjected to the influence of the total electric field which is the sum of the electric fields created by dipolons and plasmons. Explicit expressions for the coupled dipolon-plasmon oscillation frequencies for a linear and planar system of dipoles have been derived. We have employed these expressions, particularly those obtained for a planar system of dipoles, to investigate the plasma wave propagating in the CuO_2 *planes* in high-temperature superconductors. Assuming a suitable value of the unknown parameter $\boldsymbol{\nabla}\cdot\mathbf{E}_0$ our estimates show that the plasmon frequency decreases and the slope of the dispersion curve at long wavelength increases as a result of interaction with dipolons, consistent with the experimentally observed values due to Romberg *et al.*[9] The present calculations of the coupled dipolon-plasmon frequency and the dispersion relation for long wavelength make it possible to propose that the dipolon excitations may be responsible, in the absence of any other explanation, for the experimentally observed $\sim$1.1 eV plasma peak.

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