

Nanoscopic stripe-like inhomogeneities and optical conductivity of doped cuprates

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Abstract

We propose a new approach to theoretical description of doped cuprate like

$La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_{6+x}$, assuming phase separation and treating it as inhomogeneous composite material, containing the dielectric and metallic stripe-like nanoparticles. The formalism of effective medium theory is then applied for calculation of dielectric permittivity, optical and EELS spectra of $La_{2-x}Sr_xCuO_4$ with x varying in a wide range. Reasonable semi-quantitative agreement with experiment has been obtained even for the simplest version of the theory. The model was found able to reproduce all essential features of optical conductivity $\sigma(\omega)$ and transmittance both for thin films (M. Suzuki, Phys. Rev. **B 39**, 2321 (1989)) and bulk single-crystalline samples (S. Uchida *et al.* Phys. Rev. **B 43**, 7942 (1991)). Substantial difference in spectral and doping dependence of optical absorption for the thin-film and bulk samples is easily explained if only to assume different shape of metallic and dielectric regions in both materials. New peaks in $\sigma(\omega)$ and absorption spectra, that emerge in the midinfrared range near 0.5 and 1.5 eV upon doping are attributed to geometrical (Mie's) resonances. Overall, we point out that all main peculiarities of the doping effect on optical and EELS spectra for cuprates including the spectral changes accompanying the insulator-to-metal transition can be explained rather prosaically by recognizing that the doping results in emergence of nanoscopic metallic stripe-like droplets.

Key words: High- T_c cuprates. Inhomogeneity. Effective medium.

1 Introduction

High- T_c superconductivity and other unconventional properties of doped quasi-2D cuprates remain a challenging and hot debated problem. As usual, the active CuO_2 planes in cuprates are considered as homogeneous systems, starting from one-, three- or multiband Hubbard models. Often only in-plane

$Cu(3d) - O(2p\sigma)$ basis set is taken into account and the well defined Zhang-Rice singlet ground state for doped holes is presumed. In frames of such theories the dependence of various properties of copper oxides on doping are related to the rearrangements of Hubbard bands.

However there are clear experimental evidences in favour of intrinsic and generic inhomogeneous nature of the systems under consideration with well developed static and dynamic spatial inhomogeneities [1]. Nonisovalent chemical substitution in insulating cuprates like $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_{6+x}$ results in an increase of the energy of the parent phase and creates proper conditions for its competing with other, possibly metallic phases capable to provide an effective screening of the charge inhomogeneity potential.

At the beginning (nucleation regime) a new phase appears in form of a somewhat like a metallic droplet in insulating matrix. The parent phase and droplets of new phase may coexist in the phase separation regime. At different stages of the "chemical" phase separation regime we deal with the isolated droplets, with the percolation effect and finally with a complete removal of the parent phase [2]. This viewpoint on the phase separation phenomena in cuprates is generally compatible with the pioneer ideas by Emery and Kivelson [3] and some other model approaches. So, on the basis of the neutron scattering data Egami [4] conjectured an appearance of a nano-scale heterogeneous structure which is composed of a plenty mobile-carrier existing region of metallic conductivity and semi-localized scarce carrier region with antiferromagnetic spin ordering.

At present, there are numerous experimental indications on the phase separation and nanoscopic stripe-like textures in doped cuprates. Hence when considering the phenomena with a relevant characteristic length, e.g. optical absorption in infrared ($\lambda \simeq 0.5 - 2 \mu m$), doped cuprates can be roughly regarded as a binary granular medium, composed of insulating and conducting regions and described in frames of the formalism of an *effective medium* (EM) theory.

2 Physical properties of composites: effective medium approach

In what concerns the optical properties the classical problem of EM is known since long and is related to the calculation of some property of a composite system (typically, the dielectric permittivity ε_{eff}), those of pure components and their volume fractions been given. Few versions of the theory have been proposed [5]. Many of them were shown to follow from one integral equation [6] and have inherent shortcoming, assuming the concentration of one of components to be small. Among them the Maxwell-Garnett theory is one of the

most popular ones for its simplicity. More sophisticated version of the theory was given by Ping Sheng [7]; it has been proven valid in entire range of concentration of components.

Metal clusters and small metallic particles embedded in a dielectric matrix have been extensively studied in the past years. One of the most interesting features of these systems is an appearance of the morphological (geometrical), or Mie resonances [8] related to the excitation of surface plasmon modes in nanoparticle when it scatters the light wave of appropriate frequency. The dispersion law of surface eigenmodes in particles explicitly depends on its geometric shape. Rigorous theory of this effect was first derived by Mie for the case of a spherical particle [8]. In principle, infinite number of geometric resonances should be observed, that correspond to dipolar, quadrupolar etc. surface eigenmodes. However, only dipole contribution is practically accounted. This is valid provided that the particle is placed in homogeneous field, i.e. that the size of the particle is small compared with the wavelength. The frequency of geometric resonance is then easily obtained as the one at which the polarizability of small particle diverges. For the case of spherical particle embedded in the matrix with dielectric permittivity ε_d this leads to the equation:

$$\varepsilon(\omega)_{part.} + 2\varepsilon_d = 0, \quad (1)$$

whence the resonance frequency is

$$\omega_r = \frac{\omega_p}{\sqrt{1 + 2\varepsilon_d}}, \quad (2)$$

if the Drude's expression is adopted for the particle permittivity and ε_d is assumed to be constant. In the case of arbitrary ellipsoid there are three different principal values of polarizability and the last formula generalizes to

$$\omega_r^i = \omega_p \sqrt{\frac{L_i}{\varepsilon_d - L_i(\varepsilon_d - 1)}}, \quad i = 1, 2, 3, \quad (3)$$

where L_i are three (shape-dependent) depolarization factors. In both expressions ω_p is the plasma frequency.

Below we outline in brief some underlying ideas of the EM theory having in mind its further application to doped cuprates. The medium is assumed to be composed of ellipsoidal particles of two kinds. Recalling the lamellar structure of copper oxides we assumed the particles to be randomly oriented in **ab** plane, one of their principal axes being aligned in **c** direction. When one addresses the microstructure of such composite two cases may occur, when

either of components form the grain and other - the coating. The essential of the theory consists in treating the structure of material as a weighted average of these two conformations in proportion, that depends on volume fraction of components. This has an advantage in correctly displaying the percolation threshold. Final expression which determines an effective dielectric function has a form [7]:

$$f D(\varepsilon_{eff}, \varepsilon_1, \varepsilon_2, p) + (1 - f) D(\varepsilon_{eff}, \varepsilon_2, \varepsilon_1, 1 - p) = 0. \quad (4)$$

Here, the quantity $D(\varepsilon_{eff}, \varepsilon_1, \varepsilon_2, p)$ is the orientationally averaged dipole moment of a kind-I particle coated by kind-II particles, embedded in effective medium, f and $(1 - f)$ are relative probabilities of occurrence for two kinds of microstructure of the composite. Rough estimate yields f in the form: $f = v_1 / (v_1 + v_2)$, $v_1 = (1 - \sqrt[3]{p})^3$, $v_2 = (1 - \sqrt[3]{1 - p})^3$, where p is the volume fraction of component I. Analytical derivation of corresponding expression needs the grain and coating to be confocal and leads to the result [9]:

$$D(\varepsilon_{eff}, \varepsilon_1, \varepsilon_2, p) = \frac{1}{2} \sum_{i=1,2} \frac{p(\varepsilon_1 - \varepsilon_2)\varepsilon_2 + ((L_i^{in} - pL_i^{out})(\varepsilon_1 - \varepsilon_2) + \varepsilon_2)(\varepsilon_2 - \varepsilon_{eff})}{pL_i^{in}(\varepsilon_1 - \varepsilon_2)\varepsilon_2 + ((L_i^{in} - pL_i^{out})(\varepsilon_1 - \varepsilon_2) + \varepsilon_2)(\varepsilon_{eff} - L_i^{out}(\varepsilon_{eff} - \varepsilon_2))}, \quad (5)$$

where $L_i^{in, out}$ are two in-plane depolarization factors of inner and outer ellipsoids, respectively. These are defined as follows:

$$L_i = \frac{a_1 a_2 a_3}{2} \int_0^\infty \frac{dt}{(t + a_i^2) \sqrt{\sum_{k=1}^3 (t + a_k^2)}}, \quad (6)$$

and can be easily re-expressed in terms of ratios between the semi-axes of ellipsoid.

Despite the shape anisotropy of nanoscopic inclusions the dielectric functions for both media I and II together with the effective medium are assumed to be isotropic for a sake of simplicity. This assumption of optically isotropic media implies only a semi-quantitative description of such an anisotropic system as quasi-2D cuprate.

3 Optical spectroscopy of doped cuprates: experiment and theoretical models

Optical properties of HTSC cuprates have been the subject of numerous experimental and theoretical investigations. One of the first systematic studies of the doping dependence of the optical reflectance and transmittance for *epitaxial films* of $La_{2-x}Sr_xCuO_4$ deposited on $SrTiO_3$ substrate were reported in [10]. The most dramatic change with increasing x was observed near 1.5 eV. The author speculated that the emergence and near proportional strengthening of the additional absorption centered at 1.5 eV (see Fig. 4) might be related to a dramatic change in the valence band structure. However, the careful measurements of optical reflectivity spectra for *bulk single crystals* of $La_{2-x}Sr_xCuO_4$ with x varying in wide range, which covers the whole phase diagram of the material, performed by Uchida *et al.* [11], reveal only a relatively weak feature near 1.5 eV which becomes apparent only for $x > 0.10$. The authors suppose that the 1.5 eV feature is extrinsic in origin, and may be related, for instance, to some undetectable amount of oxygen vacancies.

Main effect of doping in $La_{2-x}Sr_xCuO_4$ and other cuprates is associated with the gradual shift of the spectral weight from the high-energy part of the spectrum to the low-energy part with formation of the prominent midinfrared (MIR) region absorption band and pronounced Drude-like peak. This spectral shift is particularly emphasized by the presence of the *isosbestic* point at which $\sigma(\omega)$ is invariant against x [11].

The authors [10,11] state that simple band theory cannot describe the low-energy part of the spectrum within CuO_2 planes, whereas it nicely mimics the higher-energy excitations. For $x > 0.05$ the former is clearly separated into a Drude-type peak at $\omega = 0$ and a broad band centered in the mid-IR region.

One of the puzzling features of the reflectivity spectra in $La_{2-x}Sr_xCuO_4$ and other cuprates is that the position of the reflectivity edge is almost unchanged against dopant concentration and eventually becomes a plasma edge of the Drude carriers in the overdoped region. In frames of a conventional approach it means that the edge frequency, or a screened plasma frequency, has nearly constant magnitude that rules out the simple relation $n \sim x$, which means that the edge is associated straightforwardly with the doped holes. Instead, it is plausible to suppose that $n \sim (1 - x)$ [12]. Comparative analysis of the reflectivity spectra of various cuprates [12] shows that the plasma frequency of the materials with a nearly half-filled band is almost independent of x for small doping but dependent on the average spacing of the CuO_2 planes. This conclusion clearly contradicts to the results based on the $t - J$ model [13] which show a rather strong dependence of plasma frequency ω_p on hole concentration, especially at small doping.

Extremely lightly oxygen doped crystals La_2CuO_{4+y} grown by two different methods (top-seeded and float-zone) [14] exhibit different low-temperature optical absorption spectra from 0.1 to 1.5 eV photon energy, albeit at room temperature the spectra are nearly identical both each other and those for slightly "chemically" doped $La_{2-x}Sr_xCuO_4$ [11] and "photo-doped" La_2CuO_4 [15]. In other words, in all cases, irrespective of the "external" cause, we deal with the same intrinsic mechanism of optical absorption.

Electron-hole optical excitations for the doped cuprate have been calculated by Wagner *et al.* [16] via exact diagonalization on finite clusters for a 2D multiorbital Hubbard model. Doping with one and two holes for 12-site lattices introduces a shift of absorption weight from the high-energy side to the low-energy one, in general agreement with experiment. Three additional features can clearly be identified on the low-energy side: first, the Drude peak with weight $\propto x$, second, a small feature caused by internal transitions in the Zhang-Rice singlet manifold which may be related to the experimentally observed midinfrared absorption. Finally, a strong absorption structure caused by the transitions from the Zhang-Rice singlet states into the upper Hubbard band was detected and assigned to experimental spectral weight found in the conductivity measurements between the Drude and the charge transfer peaks.

The optical conductivity has been calculated using the Hubbard and $t - J$ models by exact diagonalization on small clusters [17]. These studies predict that at low hole density, in addition to a Drude peak with a spectral weight proportional to doping there is a MIR band centered at photon energy $\sim 2J$, which should be rather broad and presumably caused by hole coupling to spin excitations. The high energy ($\geq 4J$) of the MIR bands as observed experimentally is in clear contradiction with such a theory.

Later on Lorenzana and Yu [18] calculated the optical conductivity of $La_{2-x}Sr_xCuO_4$ in the inhomogeneous Hartree-Fock plus random-phase approximation using the $p - d$ model with parameters taken from first-principle calculations. The same as for Wagner *et al.* [16] CT band peaks now at $1.7t_{pd} \approx 2.7$ eV. The precursor of the MIR band is identified with transitions from the localized states on Cu to the extended states above the Fermi level. Despite the authors [18] state close agreement with experiments regarding the peak positions and relative intensities for zero and small doping, their model calculations fail to properly describe the overall evolution of spectra with dielectric-to-metal transition.

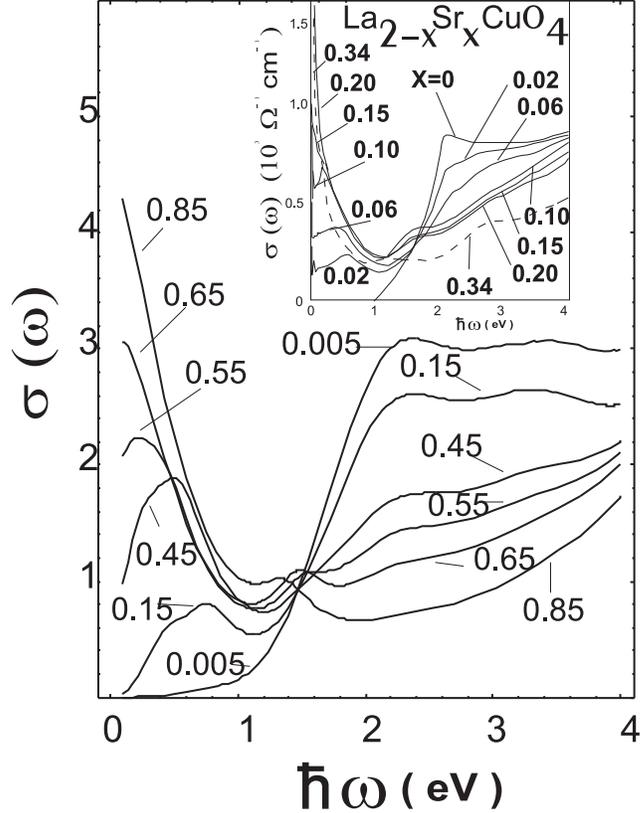


Fig. 1. Calculated optical conductivity for the model bulk single-crystalline $La_{2-x}Sr_xCuO_4$ system at different volume fractions of metallic phase. Inset: measured optical conductivity [11] for $La_{2-x}Sr_xCuO_4$ at different x .

4 The effective-medium theory of optical conductivity in doped cuprates

We have applied the formalism of effective medium theory [7] in order to reproduce some tendencies in evolution of optical spectra of high- T_c cuprates upon doping. One should note that the optical conductivity and its doping dependence are considered to be one of the most important and informative characteristics of the doped cuprates. In particular, we have attempted to describe the experimental data on doping dependence of optical conductivity in $La_{2-x}Sr_xCuO_4$ [10,11] in framework of the EM model. It should be emphasized that in our case the EM theory is nothing but a phenomenological approach to the description of the complex spatially non-uniform rearrangements in the electronic structure of doped cuprate. The concept of binary composite merely reflects the fact that upon doping some fraction of electrons remains localized in the regions of parent insulating phase while other electrons becomes nearly metallic inside regions embedded into parent insulating matrix.

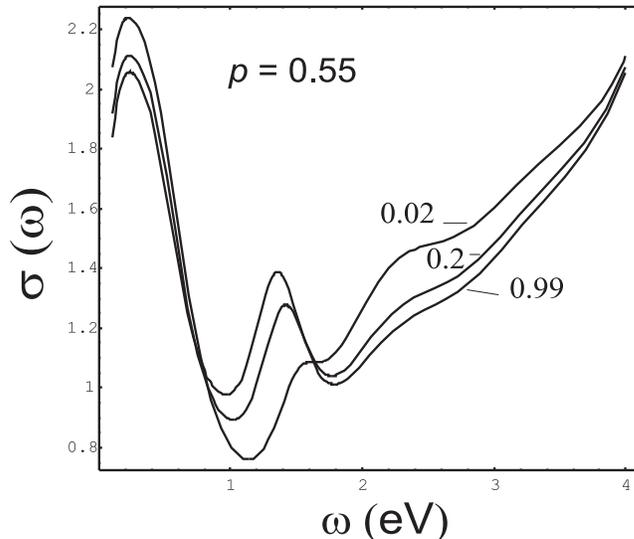


Fig. 2. Optical conductivity for the model $La_{2-x}Sr_xCuO_4$ system at different shape of inclusions (lesser number corresponds to more plate particles embedded in **ab**-plane).

It is reasonable to conjecture, that different electronic states of La_2CuO_4 do not contribute equally to this process. In particular, we simply assumed that CT excitation band(s) peaked near 2.5 eV is involved in such a "metallization" while the states at higher energy remain rigid. These ideas can easily account for the behaviour of optical conductivity of $La_{2-x}Sr_xCuO_4$: upon doping the spectral weight rapidly transfers from the CT band(s) to a narrow band of extended states, while the high-energy tails converge beyond ~ 4.5 eV for different x .

The imaginary part of ε for the parent undoped cuprate La_2CuO_4 was taken from [11] and [19]. The spectrum of $\text{Im}\varepsilon$ of La_2CuO_4 we used in our calculations was modeled by a superposition of two CT bands fitted by a sum of two Gaussians centered at 2.0 and 3.0 eV, and the "rigid" high-energy contribution fitted by a single Lorentzian centered at 6.0 eV. Only the first CT term is involved in EMT computations, its spectral weight being partly substituted by that of new metallic-like phase. As for the real part of dielectric function, near dispersionless value close to $\varepsilon = 4$ has been chosen. To model the dielectric function of the metallic phase we used the simplest Drude form

$$\varepsilon(\omega) = \varepsilon_\infty \left(1 - \frac{\omega_p^2}{\omega(\omega + i\gamma)} \right), \quad (7)$$

where ω_p and γ are the plasma frequency and the electron scattering rate, respectively. Together with ε_∞ these are regarded as adjustable parameters which magnitude does not depend on doping, frequency, and temperature. The results presented below were obtained with the values: $\omega_p = 1.65$ eV,

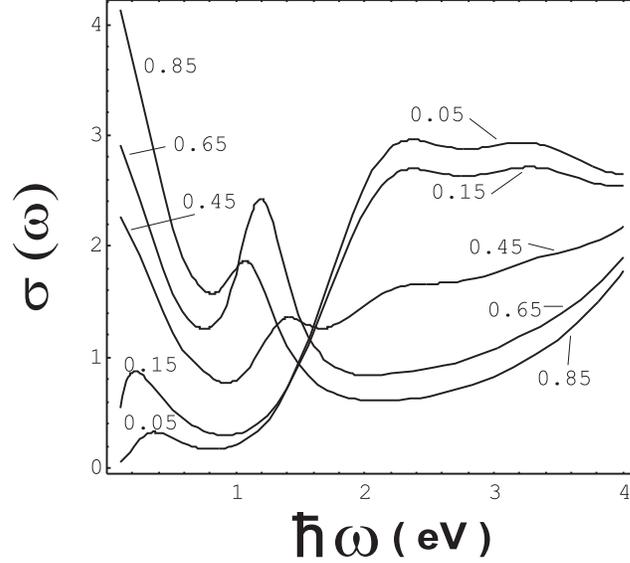


Fig. 3. Calculated optical conductivity for the thin film $La_{2-x}Sr_xCuO_4$ sample at different volume fractions of metallic phase: effect of the modified particle shape (see text).

$\gamma = 0.6$ eV, $\varepsilon_\infty = 1.0$, which once tuned, were retained unchanged against the doping level.

Another set of free parameters stand for the microtexture of the sample under investigation. Here we take into account numerous manifestations of stripe-like textures in doped cuprates. We denote the in-plane semi-axes ratio for particles of both kinds as α and that of out-of-plane and major in-plane semi-axes as β . These geometrical parameters enter the theory through the depolarization factors (6) and were found to affect strongly the dielectric function of doped cuprate.

Calculated spectra of optical conductivity $\sigma(\omega)$ for different level of doping x are shown in Fig. 1 together with experimental curves from [11]. The numbers near the curves stand for the fractions of metal phase p , which are functions of x . The geometrical parameters $\alpha_1 = 0.3$, $\beta_1 = 0.09$ and $\alpha_2 = 1.0$, $\beta_2 = 0.05$ were adopted for metallic and insulating grains, respectively. This implies the effective metallic nanoparticle to be stripe-shaped (elongated in **ab** - plane and nearly plate in the **c** - direction) and dielectric ones to be oblate ellipsoid-like round pellets (equal in-plane semi-axes and much smaller out-of-plane semi-axis). Spectral weight was found to transfer dramatically from CT band to a low-energy band at 0.7 eV in the range $p \sim 0.3$ that corresponds to $x \sim 0.06$. Further increase of metal fraction results in full removal of 2.2 eV feature of parent compound and emergence of a new weak peak at ~ 1.5 eV for large enough p , really seen in the heavily doped compositions ($x \sim 0.15$). Simultaneously, the 0.7 eV band shifts to lower energies and gradually transforms into the Drude-like peak. Many doped cuprates demonstrate isosbestic be-

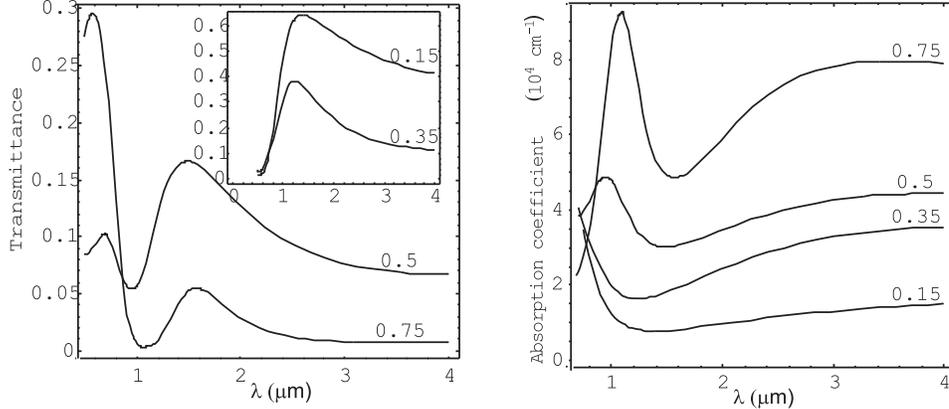


Fig. 4. Calculated absorption coefficient (right panel) and transmittance (left panel) for the thin film $La_{2-x}Sr_xCuO_4$ sample vs. wavelength at different volume fractions of metallic phase.

haviour of optical conductivity, particularly well pronounced e.g. in T' phase of $Nd_{2-x}Ce_xCuO_4$: $\sigma(\omega, x)$ cross the same point for various x . It is seen that our model theory remarkably well reproduces this feature.

Overall agreement of simple EM theory with experimental data [11] is rather impressive despite such oversimplified assumptions as invariable value of shape and size of grains, plasma frequency, relaxation rate and other quantities regardless the doping level. At the same time one observe clear shortcomings in what concerns the detailed quantitative description of the low-frequency ($\hbar\omega \leq \gamma$) spectral range and the percolation phenomenon. The EM theory yields the percolation threshold volume fraction $p_c \sim 0.6$ that seems too high. The more detailed description of the low-frequency spectral range in frames of the EM theory first of all implies the account of the size and shape distribution of metallic/dielectric grains along with the distribution of quantities like ω_p and γ . We have identified the low-energy peak and 1.5 eV feature with geometric Mie resonances as they coincide with divergencies in some terms of polarizability of composite (nullification of denominator in (5)). This is a quite novel type of resonant absorption, inherent for composite materials, which is not originated straightforwardly from either electronic transitions. The strong influence of the grain shape on the 1.5 eV feature is fairly well illustrated in Fig. 2 which demonstrates the calculated optical conductivity $\sigma(\omega)$ given the metallic volume fraction $p = 0.55$ at different values of parameter β which defines the ratio of the out-of-plane and in-plane semi-axes of dielectric grain.

The prominent ~ 1.5 eV peak is substantially more pronounced in optical absorption spectra reported by Suzuki [10] for thin-film samples. We show that good agreement with both sets of the experimentals [10,11] could be obtained merely adjusting the "geometric", or shape parameters of metallic and insulating droplets without modifying any underlying physics of the model.

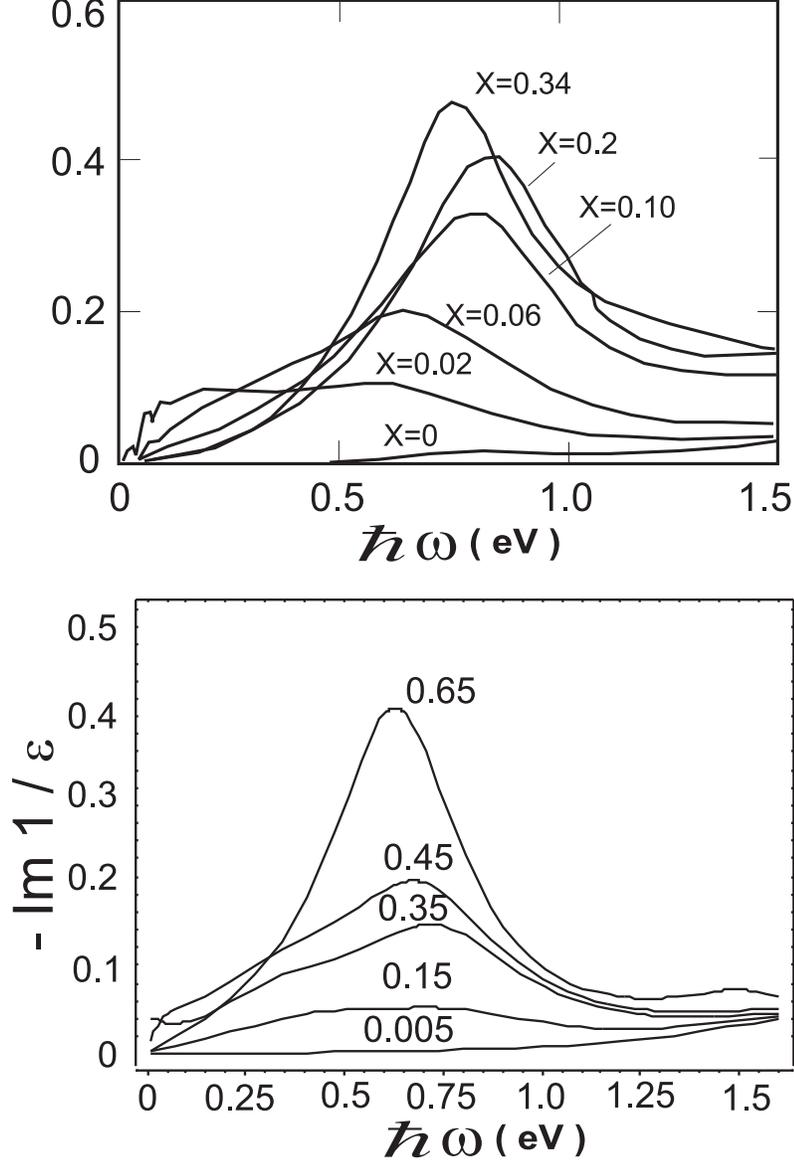


Fig. 5. EELS spectra of $La_{2-x}Sr_xCuO_4$ at $q = 0$: experiment [11] (upper panel) and the model EM theory (lower panel).

Calculated spectra of optical conductivity $\sigma(\omega)$, absorption $\alpha(\lambda)$ and transmittance $T(\lambda)$ for the $0.6 \mu\text{m}$ thin film samples of $La_{2-x}Sr_xCuO_4$ are presented in Fig. 3 and Fig. 4. Actually, all physical parameters were retained unvaried except of Drude's relaxation rate: $\gamma = 0.4 \text{ eV}$ was set instead of previous value 0.6 eV , but new values for shape parameters $\alpha_1 = 0.9$, $\beta_1 = 0.01$ and $\alpha_2 = 0.9$, $\beta_2 = 1.5$ are taken. This implies the oblate ellipsoid-like metallic grains and predominantly 3D character of dielectric regions. The most essential consequence of these changes is associated with pronounced development of 1.5 eV feature in optical conductivity. Accordingly, the $\alpha(\lambda)$ and $T(\lambda)$ spectra fit fairly well the experimental curves [10].

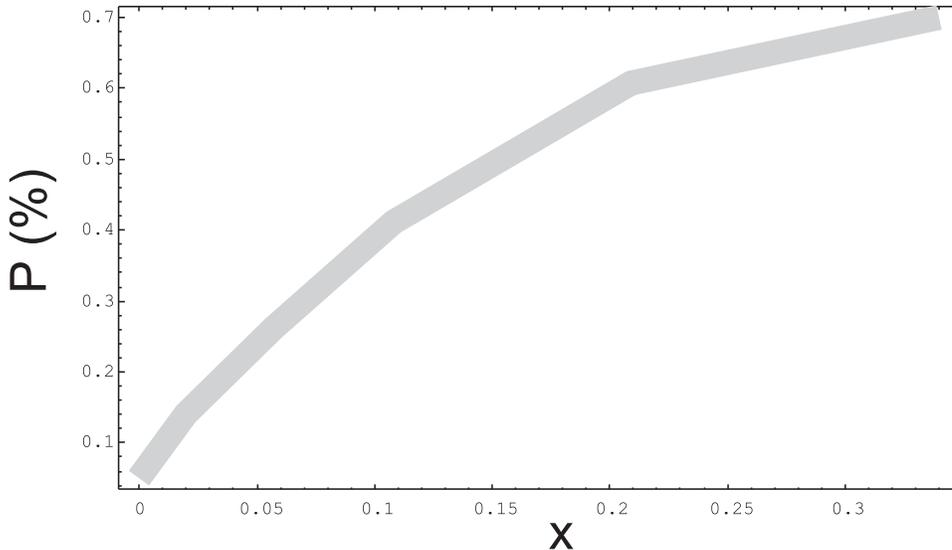


Fig. 6. Approximate dependence of the metal volume fraction on Sr content in $La_{2-x}Sr_xCuO_4$.

Real part of optical conductivity addressed above relates to imaginary part of dielectric permittivity. As an important additional check of validity of our simple model we address the calculation of the electron energy dependent loss function $\text{Im}(-1/\epsilon(\omega))$ which is determined both by real and imaginary parts of $\epsilon(\omega)$. The Figure 5 presents a calculated spectral dependence of the electron energy loss function for a model $La_{2-x}Sr_xCuO_4$ system given the same model parameters used above for description of the optical conductivity for the bulk samples (see Fig. 1). Comparison with experimental data [11] shows that our simple model allows correctly describe all features of the EELS spectra, in particular, a non-monotonous doping dependence of the peak in the loss function. It should be noted that in contrast with conventional approach both reflectivity edge and the main peak in the loss function in frames of EM theory do not relate straightforwardly to the bare plasma frequency ω_p .

We assumed through the article that the doping level x (say, strontium content) can be simulated by an appropriate tuning of metal volume fraction p in the composite. It is, however, desirable to establish the relation between these quantities in more accurate manner. For the case of optical conductivity in bulk single-crystalline $La_{2-x}Sr_xCuO_4$ samples [11] this can be done e.g. through the calculation relative decrease of the 2.2 eV peak with doping, then adjusting p to keep the same proportion in theoretical graph. The Fig. 6 illustrates the relation $p(x)$ obtained in this way. The saturation behaviour, discernible for large enough x can be attributed to the percolation phenomenon.

5 Conclusion

In conclusion, we have proposed a new approach to theoretical description of doped cuprates, treating these as inhomogeneous composite materials, containing the dielectric and metallic stripe-like nanoparticles. The formalism of effective medium theory is then applied for calculation of dielectric permittivity and optical spectra of $La_{2-x}Sr_xCuO_4$ with x varying in a wide range. Reasonable quantitative agreement with experiment has been obtained. The model in its simplest form was found able to reproduce all essential features of the transmittance [10], optical conductivity $\sigma(\omega)$, and EELS spectra [11]. Substantial difference in spectral and doping dependence of optical conductivity for thin-film [10] and bulk samples [11] is easily explained if to only assume different shape of metallic and dielectric regions in both materials. New peaks in $\sigma(\omega)$ and absorption spectra, that emerge in the midinfrared range upon doping are attributed to geometrical (Mie's) resonances. Overall, the model theory allows to properly describe the evolution of optical spectra at the percolative insulator-to-metal transition.

Finally, we would like to note that an occurrence of the phase separation with percolative nature of the insulator-to-metal transition is accompanied by many specific features which in many cases mask the real electronic structure and can lead to the erroneous conclusions. In particular, making use of either experimental data as a trump in favor of either mechanism of the high- T_c superconductivity should be made with some caution, if a phase homogeneity of the samples under examination is questionable. In this connection one may mention a long-standing discussion regarding the nature of the midinfrared bands in doped cuprates.

Further extension of the model developed here should include the size and shape distribution of metallic-like inclusions, temperature effects, as well as effects of different external factors such as pressure, isotopic substitution, photo-doping. Of course, this semi-empirical model needs in more detailed microscopic reasoning.

Acknowledgements

We are grateful to N. Loshkareva, Yu. Sukhorukov, K. Kugel, P. Horsch for discussions. One of the authors (A.S.M.) acknowledges stimulating discussion with N. DelFatti. The research described in this publication was made possible in part by Award No.REC-005 of the U.S. Civilian Research & Development Foundation for the Independent States of the Former Soviet Union (CRDF). The authors acknowledge a partial support from the Russian Ministry of Ed-

ucation, grant E00-3.4-280.

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