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Free-charge-carrier plasmons in $Ba_{1-x}K_xBiO_3$: A close relation to cuprate superconductors

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Broad-range reflectance and ellipsometric spectra of high-quality epitaxial superconducting $Ba_{1-x}K_xBiO_3$ thin films are reported. From these data, accurate spectral functions of $Ba_{1-x}K_xBiO_3$ have been obtained. They are found to be quite similar to the in-plane spectra of the superconducting cuprates; in particular, the frequency dependence of the reflectance is nearly linear, while the dielectric loss function is nearly quadratic below the plasma edge in all of these compounds. It is therefore unlikely that acoustic plasmons or magnetic fluctuations are the cause of these anomalous spectral features.

I. INTRODUCTION

Since high-temperature superconductivity in cuprates was discovered five years ago,[1] a lot of effort has been devoted to unraveling of its physical origins. For this reason, low-energy excitations in these materials have been investigated extensively and by a variety of methods.[2] Our approach[3] has been to attempt accurate determination of the complex dielectric function by utilizing a combination of two techniques, spectroellipsometry and nearly-normal-incidence reflectance measurements over a broad frequency region. When samples of good optical quality—such as large-area, cleaved single crystals or high-quality epitaxial thin films—were available, it was possible to obtain spectral functions that we believe are accurate to within 2 to 3 percent, in the range from few meV to 5–6 eV, i.e., from far-infrared to ultraviolet. So far, we have investigated $La_{1.85}Sr_{0.15}CuO_4$, $YBa_2Cu_3O_7$, $Bi_2Sr_2CaCu_2O_8$, and $Tl_2Ba_2Ca_2Cu_3O_{10}$ in this way.[3]

Several types of excitations can be identified unambiguously in these spectra. First, we see free-charge carriers, which make all these materials almost perfectly reflective at low frequencies. Second, phonons are certainly present also, although in the best samples hardly any can be seen if the infrared spectra are taken with the light beam perpendicular to the CuO_2 planes. The reason is simply that the strongly polar infrared-active in-plane optical modes are virtually completely screened by the free carriers; indeed, optical phonons are very visible in infrared spectra at other polarizations, as well as in Raman spectra and in inelastic neutron diffraction data.[2] Finally, in each of these compounds there is clearly an optical-plasmon mode, at roughly 1 eV, which comes from the free-charge carriers. We believe that this is not controversial.

Many other types of low-energy excitations have also been postulated to exist in cuprate superconductors, and even to play an essential role in the high-T_c superconductivity phenomenon itself.[4] Among them, acoustic plasmons[5] have a somewhat favorable position insofar that they should be expected to occur, from rather general theoretical arguments,[6] irrespective of the nature (e.g., the spin character) of the charge carriers, as long as the material can be viewed as a layered conductor. Given that in $La_{1.85}Sr_{0.15}CuO_4$, $Bi_2Sr_2CaCu_2O_8$, and $Tl_2Ba_2Ca_2Cu_3O_{10}$ the *c*- vs *ab*-axis resistivity ratio is[7] $\rho_\perp/\rho_\parallel = 10^5-10^6$ (which is not surpassed in any other quasi-two-dimensional metal known to us), and that the reflectivity for the light polarized with the electric field perpendicular to CuO_2 planes is very low and distinctly ionic-insulator-like[8], etc., it seems very likely that at least these compounds should belong to the above class. However, no direct experimental observation of acoustic plasmons in cuprates has been reported so far.

In our earlier work[3] we have noticed that in $La_{1.85}Sr_{0.15}CuO_4$, $YBa_2Cu_3O_7$, $Bi_2Sr_2CaCu_2O_8$, and $Tl_2Ba_2Ca_2Cu_3O_{10}$ the frequency dependence of the dielectric loss function was nearly exactly quadratic:

$$\mathrm{Im}(-1/\epsilon) = \alpha\omega^2 \tag{1}$$

for ω almost all the way up to the optical plasmon energy, which is unusual and not what one would expect in a normal (Drude) metal. We speculated that this peculiar spectral characteristic could be related to existence of acoustic plasmons in these layered cuprates. In view of the potential importance of the issue, it seems of interest to further test this proposal critically. The obvious first task is to investigate comparatively some other related materials.

Except for the cuprates, $Ba_{1-x}K_xBiO_3$ with $x \approx 0.4$ is the oxide with the highest known T_c, exceeding 30 K at the optimal doping level.[9] This material has some remarkable similarities with the copper oxide superconductors; for example, it also has a very high ratio of T_c vs the density of states.[10] The most notable difference–apart from the absence of magnetism–is that $Ba_{1-x}K_xBiO_3$ is cubic. Hence, while the layered-electron-gas model[6] may be relevant for cuprates, certainly it is not applicable to $Ba_{1-x}K_xBiO_3$. So it seems opportune to compare and contrast their plasmon spectra, as revealed from the respective dielectric loss functions, which is our principal task in this paper.

Several optical studies of $Ba_{1-x}K_xBiO_3$ and the related $BaPb_xBi_{1-x}O_3$ compounds have already been reported.[11,12] Nevertheless, we feel that there may be some interest in trying to compare bismuthates and cuprates on an equal and rigorous level. Since large $Ba_{1-x}K_xBiO_3$ single crystals seem to be difficult to make homogeneously, we have chosen to study large-area, highly reflective oriented thin films.[13] (These films also tend to show better transport properties, e.g., a lower resistivity.) Further improvement comes from the method; in addition to the broad-frequency-range reflectance measurements, we also report ellipsometric spectra of $Ba_{1-x}K_xBiO_3$. Combining these two techniques, the spectral functions can be determined much more accurately; in particular, we can eliminate the substantial error which is brought in by extrapolating arbitrarily the reflectance outside the experimentally accessible range in order to employ the Kramers-Kronig transformations.

II. EXPERIMENT

A. Sample preparation and characterization

In this paper we will present the spectroscopic data for two $Ba_{1-x}K_xBiO_3$ films, each of which showed good optical quality–homogeneous appearance, high specular reflectance, and low diffuse reflectance. The films were grown on (1 0 0) MgO substrates by molecular-beam epitaxy, using techniques described previously.[13] Film *A* was 7600 Å thick with composition $Ba_{0.54}K_{0.46}BiO_x$, according to Rutherford backscattering spectroscopy measurements, and was grown at 325±25 °C. Film *B* was 3700 Å thick and had composition $Ba_{0.36}K_{0.60}Bi_{1.04}O_x$. Both films were nucleated for 3 min at 560±20 °C, which should result in an initial 100-Å layer of $BaBi_2O_y$. Electrical measurements of the films indicated zero-resistance transition temperatures of 23.2 K for film *A* and 20.6 K for film *B*, with transition widths of 1.5 and 2.5 K, respectively. The normal-state characteristics of the films were representative of $Ba_{1-x}K_xBiO_3$ films; both films were weakly metallic below 100 K. Film *B* was more conductive than film *A* by a factor of 4 at room temperature.

B. Spectroscopy

Far-infrared reflectance spectra were recorded using a Bio-Rad FTS-40V Fourier-transform infrared (FTIR) spectrometer. For mid-infrared reflectance spectra, we used a Bio-Rad FTS-40 FTIR spectrometer as well as a Spectra-Tech IR-PLAN Microscope coupled to that spectrometer. The microscope was equipped with a dedicated liquid-nitrogen-cooled Hg-Cd-Te detector and it provided spatial resolution better than 100 μm. Thus we were able to test the film uniformity on this length scale by moving the spot across the film surface and verifying that no significant variations in the spectra were observed. Near-infrared through near-ultraviolet reflectance spectra were recorded on a Perkin-Elmer Lambda-9 double-beam, double-monochromator spectrophotometer. The diffuse reflectance spectra were recorded utilizing a Perkin-Elmer 60-mm Integrating Sphere coupled to the Perkin-Elmer Lambda-9 spectrophotometer. A small diffuse reflectance component was taken as an indicator of the optical quality of the film and the smoothness of the surface. Ellipsometric measurements were performed on a Model 445A15 Rudolph Instruments Automatic Spectro-Ellipsometer. In the spectral regions where they overlap, a good agreement was found between the spectra taken on different spectrometers.

C. Computation of spectral functions

In Ref. 3(a) an algorithm was introduced which allows one to determine the complex index of refraction $n+ik$ from the normal-incidence reflectance spectrum measured over a broad range of frequencies (in our case from a few meV to about 5–6 eV) and the pseudodielectric function $\langle\epsilon\rangle = \langle\epsilon_1\rangle + i\langle\epsilon_2\rangle$ as determined from ellipsometric spectra in some interval $\omega_L < \omega < \omega_H$ (here $\hbar\omega_L = 1.3$ eV and $\hbar\omega_H = 4.5$ eV).

First, one assumes that $R(\omega) = R_H = \text{const.}$ for $\omega \geq \omega_H$, and utilizes the Kramers-Kronig transformation to compute the auxiliary phase $\theta_{\text{uncorr}}(\omega)$. Next, from $\langle\epsilon_1\rangle + i\langle\epsilon_2\rangle$, one calculates $\theta_{\text{ellips}}(\omega)$, for $\omega_L < \omega < \omega_H$. Then one finds the difference $\Delta\theta(\omega) = \theta_{\text{ellips}}(\omega) - \theta_{\text{uncorr}}(\omega)$, which can be proved to be an analytic function of frequency, and furthermore expandable into a series which contains only odd powers. So we fit this difference by a polynomial $P(\omega) = a_1\omega + a_3\omega^3 + \cdots + a_{2r+1}\omega^{2r+1}$ (with r typically between 10 and 20). The corrected phase $\theta_{\text{corr}}(\omega) = \theta_{\text{uncorr}}(\omega) + P(\omega)$ is obtained next and utilized to compute the real and the imaginary part of the complex index of refraction, $n = (1-R)/(1+R-2\sqrt{R}\cos\theta)$ and $k = 2\sqrt{R}\sin\theta/(1+R-2\sqrt{R}\cos\theta)$. From these values, one can calculate $\epsilon_1 = n^2 - k^2$ and $\epsilon_2 = 2nk$, the dielectric loss function $\text{Im}(-1/\epsilon) = \epsilon_2/(\epsilon_1^2 + \epsilon_2^2)$, and any other spectral function of interest.

In principle, this method is very accurate and one can realistically expect the resulting spectral functions to be correct to within 2 to 3 percent. In the present case, the major sources of error are the following: some (small) divergence of the beam and its departure from the normal incidence; a small diffuse component due to some surface imperfections; a small discrepancy between the spectra taken on different instruments or when certain optical components were changed; and finally some transparency of the films at the frequencies close to the plasma edge, where optical penetration depth increases dramatically. (The last issue was partially taken into account by means of multilayer modeling; the optical constants of the substrate are known.) Overall, the error is believed to be less than 5% over the whole spectral range covered. As it will be clear from what follows, sample-to-sample differences in the spectra which originate from stoichiometry variations, which in turn result in charge-carrier density variations, by far exceed this level anyway. Our conclusions will be restricted to statements which we believe are valid generally and which should be affected neither by the experimental errors nor by such sample-to-sample variations.

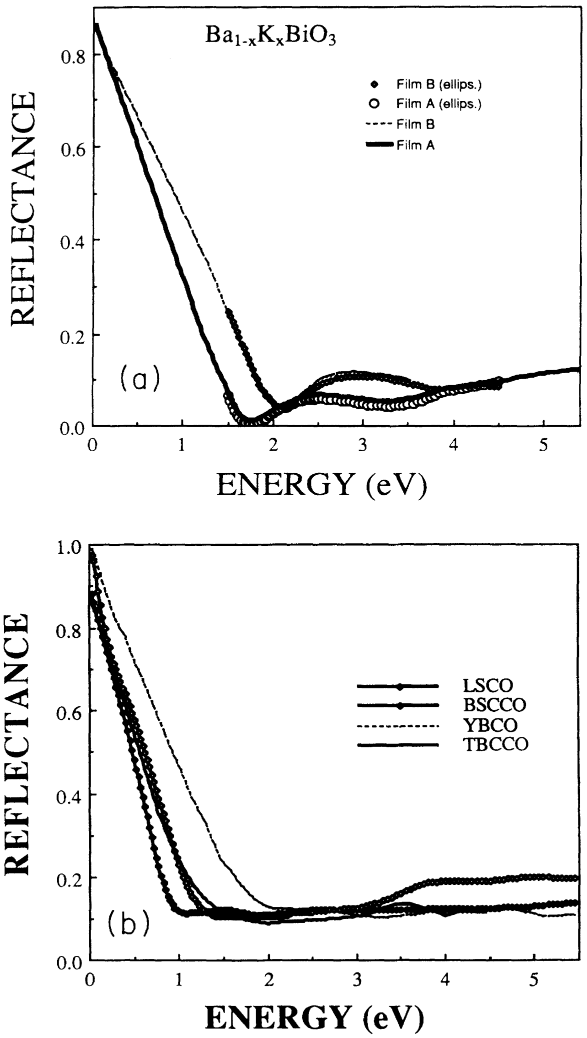

FIG. 1. Broad-range specular reflectance spectra of (a) the two $Ba_{1-x}K_xBiO_3$ films, measured directly (thick solid line, film *A*; thin dashed line: film *B*) and calculated from the ellipsometric spectra (film *A*, open circles; film *B*, open diamonds) and (b) $La_{1.85}Sr_{0.15}CuO_4$(LSCO), $YBa_2Cu_3O_7$(YBCO), $Bi_2Sr_2CaCu_2O_8$(BSCCO), and $Tl_2Ba_2Ca_2Cu_3O_{10}$(TBCCO).

III. RESULTS

In Fig. 1(a) we present the specular reflectance spectrum (full line), taken at nearly normal incidence, of the "best" $Ba_{1-x}K_xBiO_3$ film *A*. The reflectance computed by utilizing the pseudodielectric functions determined from the ellipsometric spectra of the same film is also plotted (open circles) to show that the agreement is indeed satisfactory. The corresponding spectra for film *B* are also displayed (dashed line) to illustrate what may be typical sample-to-sample variations in superconducting $Ba_{1-x}K_xBiO_3$ films. The spectra of the two films are similar except that the plasma edge in film *B* is higher, indicating a higher carrier density. This observation correlates well with the measured dc resistivities of the two films; film *B* was indeed more conducting (at room temperature, it had $\rho=0.9$ mΩ cm as compared to $\rho=4$ mΩ cm in film *A*). On the other hand, the plasma edge in film *A* is sharper; it also had a sharper resistive transition. For comparison, in Fig. 1(b) we have replotted the corresponding spectra of $La_{1.85}Sr_{0.15}CuO_4$, $YBa_2Cu_3O_7$, $Bi_2Sr_2CaCu_2O_8$, and $Tl_2Ba_2Ca_2Cu_3O_{10}$, after Ref. 3.

In Fig. 2 we have plotted the real and the imaginary parts of the complex dielectric function calculated according to the algorithm expounded above, for both films *A* and *B*. They agree with the corresponding pseudodielectric functions as determined from spectroellipsometric measurements (not shown) to within a percent or so.

In Fig. 3(a) we have shown the dielectric loss function $\mathrm{Im}(-1/\epsilon)$ of the same two $Ba_{1-x}K_xBiO_3$ films, while the analogous data for the four cuprate compounds are reproduced in Fig. 3(b).

Analyzing these data, one notices the striking overall similarity of the reflectance spectra of the cuprates and the bismuthate, once they are doped close to the "op-

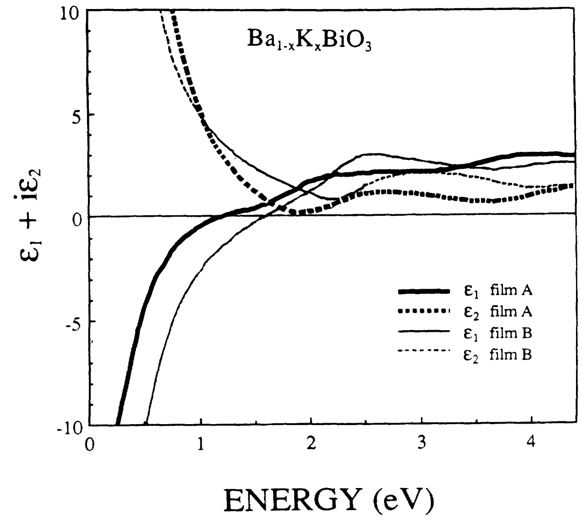

FIG. 2. The calculated real and imaginary part of the complex dielectric function $\epsilon=\epsilon_1+i\epsilon_2$ of $Ba_{1-x}K_xBiO_3$ film *A* (thick lines) and film *B* (thin lines).

timal" level which corresponds to the highest T_c achievable in the material considered. All these spectra are nearly linear in frequency:

$$R(\omega)\approx A-B\omega \qquad (2)$$

[where A and B are constants, $A\approx 1$ and $B/\hbar \approx 1\ (\mathrm{eV})^{-1}$] below the plasma edge, i.e., for $\omega<\omega_p$. Consequently, all the corresponding dielectric loss functions are remarkably similar to one another as well; in other words, the $Ba_{1-x}K_xBiO_3$ films indeed also exhibit the quadratic frequency dependence as given by Eq. (1).

The key feature to be noticed in Fig. 2 is the occurrence of the zero of $\epsilon_1(\omega)$ at about 1.2 eV in film A. For this film, one can see in Fig. 3(a) that there is a pronounced peak of the dielectric loss function at about 1.4 eV, i.e., at nearly the same energy. The same is true for film B, except that both the zero of $\epsilon_1(\omega)$ and the peak of $\mathrm{Im}(-1/\epsilon)$ are located at a somewhat higher energy, about 1.6 and 1.75 eV, respectively. These are typical signatures of the optical plasmons which originate from free-charge carriers. From Fig. 3(b), we see that similar peaks occur in each of the four cuprates considered; in these compounds, the dependence of the peak position on momentum transfer has also been established by means of electron-energy-loss spectroscopy,[14] and that enabled unambiguous assignment of this spectral feature to the optical plasmon. Hence we can state with some certainty that also in $Ba_{1-x}K_xBiO_3$ with $x\approx 0.4$ there is a free-carrier optical plasmon at about 1.4–1.8 eV.

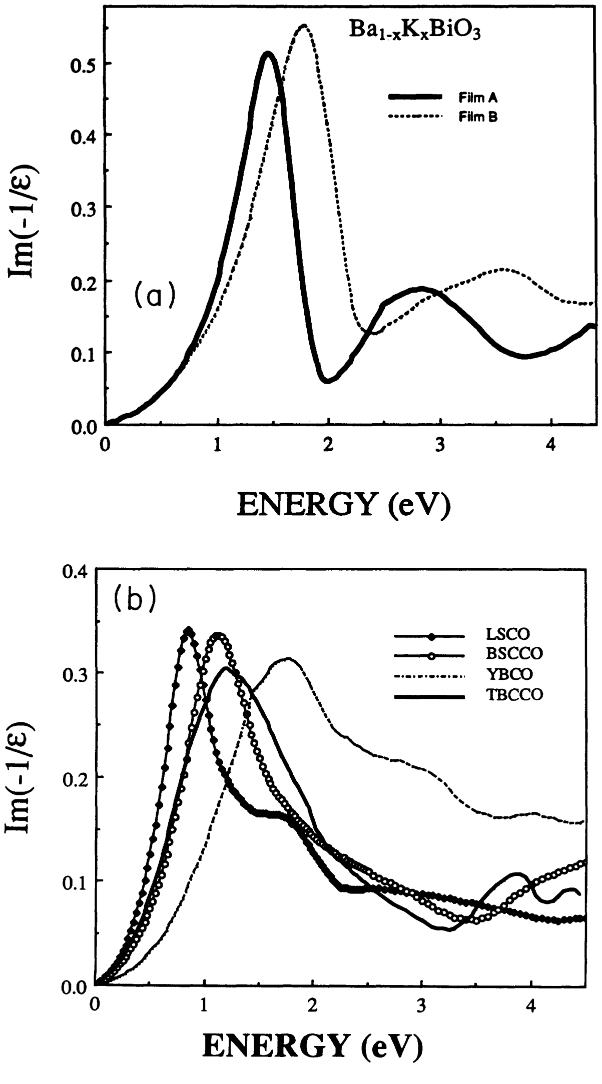

FIG. 3. The calculated dielectric loss functions of (a) the two $Ba_{1-x}K_xBiO_3$ films, and (b) $La_{1.85}Sr_{0.15}CuO_4$, $YBa_2Cu_3O_7$, $Bi_2Sr_2CaCu_2O_8$, and $Tl_2Ba_2Ca_2Cu_3O_{10}$.

From the spectra shown in Figs. 1–3, we can determine the *bare* plasma frequency, which is defined by $\omega_p^2=4\pi Ne^2/m^*$ (where N is the carrier density and m^* is the optical effective mass) using the sum rule:

$$\int_0^{\omega_1}\mathrm{Re}\,\sigma(\omega)d\omega=\omega_p^2/8\ , \qquad (3)$$

where $\sigma(\omega)$ is the optical conductivity and ω_1 is a cutoff frequency which should be chosen high enough to include the entire free-carrier response, but low enough to exclude interband transitions. With this motivation, in Fig. 4 we plot the quantity

$$\Omega(\omega_1)=\left[8\hbar^2\int_0^{\omega_1}\mathrm{Re}\,\sigma(\omega)d\omega\right]^{1/2}.$$

Apparently, in the vicinity of the plasma edge this function is virtually horizontal, having the value of about 3.05 eV. This evaluation is much more accurate here, because of the sharper plasma edge, than in the cuprates. Utilizing $\hbar\omega_p\approx 3$ eV and the measured high-frequency dielectric constant, one would predict the optical plasmon to occur near the *screened* plasma frequency $\tilde{\omega}_p\approx\omega_p/\sqrt{\epsilon_\infty}=1.6$ eV in film A – which is indeed quite close to the actual position of the peak in Fig. 2. Analogously, for film B we get $\hbar\omega_p=3.4$ eV and $\hbar\omega_p\approx 1.8$ eV.

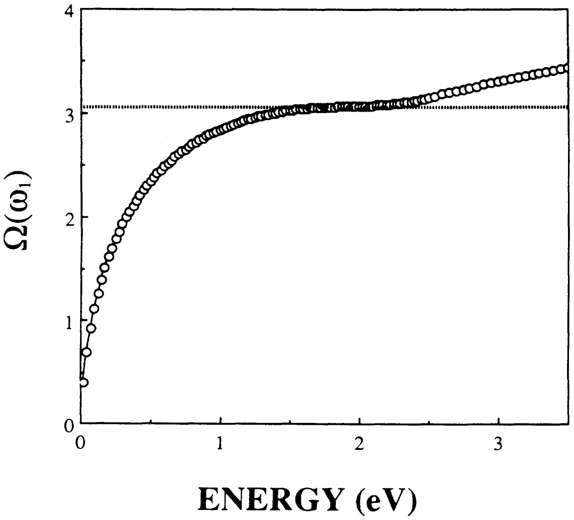

FIG. 4. The f-sum rule function $\Omega(\omega_1)$ for the $Ba_{1-x}K_xBiO_3$ film A.

IV. DISCUSSION AND CONCLUSIONS

First, the great similarity of the reflectance and other spectral functions of $Ba_{1-x}K_xBiO_3$ and the cuprates, including specifically the anomalous dependences given by Eqs. (1) and (2), suggests that one and the same physical mechanism is involved. Since acoustic plasmons are not expected to occur in $Ba_{1-x}K_xBiO_3$, which is cubic, they probably do *not* give rise to these anomalies in cuprates, either. (On the other hand, this does *not* imply that acoustic plasmons are absent in cuprates, but merely that their contribution to the loss function is not dominant.) The same is true for magnetic fluctuations, as well.

To extract quantitative information from such spectra, two fitting schemes have been frequently employed recently. In the first approach, one utilizes a simple Drude model and varies ω_p, Γ, and ϵ_∞ to fit the measured reflectance or the optical conductivity. In this way, Sato *et al.*[11] obtained $\hbar\omega_p = 2.8$ eV, $\epsilon_\infty = 3$, and $\hbar\Gamma = 0.9$ eV. Alternatively, the absorption spectrum can be modeled by a sum of a much smaller Drude contribution and a strong mid-infrared Lorentz oscillator usually attributed to an interband transition or an exciton. Employing this picture, Schlesinger *et al.*[12] concluded that $\hbar\omega_p = 0.5$ eV, $\epsilon_\infty = 3$, $\hbar\Gamma = 0.25$ eV, and $\hbar\omega_0 = 0.6$ eV.

Given the sample-to-sample variations and other experimental uncertainties, fits to the (room-temperature) reflectance data provided by either of these models are reasonably good indeed. However, as we will argue below, neither of them provides a satisfactory physical picture, since each of them conflicts with some other experimental data.

To begin with, it has been amply demonstrated in cuprates[2(a)] that the simple Drude model provides an essentially wrong prediction for the temperature dependence of the absorption spectrum. Actually, that should not be surprising; the free-electron Drude model is not expected to be applicable when the damping Γ is so large that the mean free path of the carriers becomes comparable to the unit-cell length. Under such circumstances, one would not expect electron motion to be coherent or bandlike, but rather diffusive (or superdiffusive).

On the other hand, from the model which invokes a mid-infrared interband transition,[15] one would expect an optical plasmon to occur somewhere close to 0.2–0.3 eV, or even lower since one would expect that an interband transition would contribute to polarizability and enhance ϵ_∞. However, the actual optical plasmon frequency is 1.6–1.8 eV, i.e., more than five times higher. The corre-

So, the important message from the sum rules is that essentially entire infrared absorption in $Ba_{1-x}K_xBiO_3$ comes from the free-charge carriers. Their response exhausts the oscillator strength, and there is no room left for other strongly infrared-active absorption processes such as due to low-energy excitons or interband transitions. On the other hand, these charge carriers are *not* Drude-free-electron-like; rather, they look like composite particles with internal structure. A possible cause of this behavior could be polaron formation due, e.g., to strong coupling of charge carriers to polar optical phonon modes, which could lead to phonon emission and strong scattering at higher energies. However, it has been demonstrated explicitly[16] for several other microscopic or phenomenological models that they are capable of explaining the anomalous infrared absorption in oxide superconductors. The present study adds a constraint that such models have to be applicable to bismuthates as well, and in all likelihood this requirement rules out the models based on magnetic scattering.

Finally, let us point out that our remarks about similarities were restricted to the in-plane electron motion in cuprates. Indeed, the out-of-plane response in those compounds is very different;[8] the plasma edge in the direction perpendicular to CuO_2 planes is not observable at all in $La_{1-x}Sr_xCuO_4$, $Bi_2Sr_2CaCu_2O_8$, and $Tl_2Ba_2Ca_2Cu_3O_{10}$.

In conclusion, we have measured broad-range reflectance and ellipsometric spectra of high-quality epitaxial $Ba_{1-x}K_xBiO_3$ thin films. Combining these data, we have obtained accurate spectral functions of $Ba_{1-x}K_xBiO_3$ doped close to its optimal stoichiometry, and compared them to the corresponding spectra of several best-known superconducting cuprates which we have obtained earlier utilizing the same methodology. The comparison showed substantial similarity between the cuprates (in-plane response only) and the bismuthate. In particular, a nearly quadratic frequency dependence of the dielectric loss function and a pseudolinear frequency dependence of the reflectance, below the plasma edge, are seen in all of these compounds. That likely rules out acoustic plasmons and magnetic fluctuations as possible causes of these peculiar dependences. Finally, detailed sum-rule analysis showed that the strong infrared absorption in $Ba_{1-x}K_xBiO_3$ entirely comes from charge carriers, whose motion is strongly damped at higher energies.

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