## Magnetic-field dependence of the c-axis plasma edge of $La_{1.85}Sr_{0.15}CuO_4$

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We investigated the influence of a magnetic field on the c-axis reflectance edge of single-crystalline La<sub>1.85</sub>Sr<sub>0.15</sub>CuO<sub>4</sub>. We used fields up to 25 T oriented parallel to the superconducting layers. In contrast to theoretical predictions we find that the screened plasma frequency  $\omega_{ps}$  itself is basically field independent. Also at  $\omega_{ps}$  there is no change in the absorption. However, there is a small but significant field-dependent renormalization of the reflectance edge just below  $\omega_{ps}$ , which indicates an enhanced dissipation in the vortex state. The data are discussed using several recent models for the interaction between electromagnetic radiation and vortices in a layered superconductor.

A surprising feature of the cuprate high- $T_c$  superconductors is that the c-axis interlayer optical conductivity is highly unusual. In the normal state the conductivity is completely phonon dominated with only a marginal contribution of free carrier transport. The onset of coherent transport in the superconducting state is marked by the sudden appearance of a clear low-frequency plasma edge in the reflectance spectra. In single crystals first observed for La<sub>2-x</sub>Sr<sub>x</sub>CuO<sub>4</sub> (Ref. 1) and later for YBa2Cu3O6.7 (Ref. 2) and for YBa2Cu4O8, this low-frequency plasma mode seems to be a universal feature in the highly anisotropic layered high- $T_c$  superconduct-

Although the observation of this plasma edge was completely unexpected and has no parallel in ordinary isotropic superconductors, its physical origin is rather simple and is related to the well-known Josephson plasmon in classical tunnel junctions. In these systems one can characterize the plasmon as a propagating electromagnetic mode coupled to local oscillations of the phase of the complex superconductive order parameter.4

As was shown previously, the study of this low-frequency mode can give detailed information about the effective coupling between the CuO<sub>2</sub> layers, the intrinsic critical current, and residual dissipation within the gap. Although the effect itself does not depend on the specific mechanism for superconductivity it does give relevant constraints on input parameters for various models.

In this paper we report on an experimental investigation of the magnetic-field dependence of the c-axis reflectance of single-crystal La<sub>1.85</sub>Sr<sub>0.15</sub>CuO<sub>4</sub>. Related measurements have been reported before on small particles of La<sub>2-x</sub>Sr<sub>x</sub>CuO<sub>4</sub> albeit with a smaller Sr content.5

The fact that there is a plasma resonance in a frequency region where there is little or no damping allows a detailed study of the electromagnetic interaction with the vortices introduced by a magnetic field. As we will show, this spectroscopic investigation of the vortex dynamics provides specific constraints on the nature of the vortices, the pinning potential, and vortex viscosity. This information can also be derived from transport measurements. However, a full spectral analysis provides self-consistency checks on model assumptions and is not hampered by extrinsic problems such as local heating, self-field effects, edge problems, etc.

In the following, we will first describe the experimental details and present the basic results. We will discuss the data in the context of recent theoretical models.

We used various techniques to measure the reflectance of the ac-oriented sample surface of large La<sub>1.85</sub>Sr<sub>0.15</sub>CuO<sub>4</sub>  $(T_c = 34 \text{ K})$  single crystals grown by the traveling solvent floating zone method. Polarized measurements at zero magnetic field were done with a Bruker IFS113v Fourier Transform spectrometer at temperatures between 10 and 300 K. The wide band reflectivity at 1.2 K in magnetic fields up to 17.5 T was measured with the same instrument coupled to a special superconducting magnet system with <sup>3</sup>He cooled bolometers. The magnetic field was in all experiments parallel to the crystal b axis.

The zero-field measurements in both setups were in good agreement. This allowed us to deconvolute the unpolarized magnetic-field data to obtain the c-axis spectra, where we assumed that the flat a-axis reflectivity is relatively insensitive to the applied field. This assumption is quite reasonable in view of the in-plane reflectivity data of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> films in similar fields. In principle, the a- and c-axis response will be mixed due to the Hall field. However, from previous measurements on the ab response there are no reasons to assume that the magnetic field will induce any features in the a-axis reflectivity. The spectral position and shape of the plasma edge will be exclusively determined by the c-axis response. Therefore, any Hall field mixing is expected to give a small featureless renormalization of the reflectivity within the experimental noise.

Figure 1 shows the spectroscopic results at 1.2 K at various fields up to 17.5 T. The spectra were taken in three separate runs, always with increasing field, and were averaged afterwards. A hysteresis corresponding to a remanent magnetic field of about 2 T is found in the spectra when after each run the applied field is lowered to zero again. We ob12 050

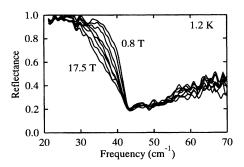


FIG. 1. Reflectance spectra at 1.2 K for magnetic fields of 0.8, 2.5, 5, 7.5, 10, 12.5, 15, and 17.5 T.

serve a clear renormalization of the reflectance edge which increases with field. In contrast, the position of the reflectance minimum at 43 cm<sup>-1</sup> does not shift appreciably to lower frequencies and its depth is also rather field insensitive. From these data a few qualitative observations are obvious: In contrast to theoretical predictions, there is no appreciable field induced shift of the plasma frequency [or zero crossing in the real part of the dielectric function  $\epsilon_1(\omega)$ ]. This shows that in this field range and for this geometry (B||b) pair-breaking effects can be neglected and the interplane coupling remains largely unaffected. A second qualitative observation is that there is no clear reduction of the reflectivity at low frequencies, as would be expected if the vortices could freely move. This illustrates that the vortices are rather efficiently pinned and cannot be described in terms of ideal Josephson vortices. Instead, their character is Abrikosov-like.

This is consistent with the observed hysteresis. Using Bean's model and an average sample diameter of 4 mm, we find that the hysteresis corresponds to a critical current of  $1.6 \times 10^9$  A/m², which is somewhat larger than but in reasonable agreement with magnetization measurements on similar samples. It is important to note the difference between this pinning limited transport critical current density and the intrinsic critical current density which is limited by the coupling between the layers. It is this last critical current which we measure in the infrared. (In a simple Josephson approximation the observed plasma frequency is equivalent with an intrinsic upper limit for  $j_c$  of  $8 \times 10^9$  A/m².)

We will come back to a more detailed analysis in the last part of this paper.

In a second experiment we measured the temperature-dependent microwave and far infrared reflectivity in a hybrid Bitter/superconductor magnet at fields up to 25 T using both an optically pumped molecular gas far-infrared (FIR) laser and a tunable millimeter wave-vector network analyzer with heterodyne detection. In this case the magnetic-field-dependent reflectance of the sample is measured at various monochromatic energies as a function of temperature. Typical curves, taken at 6.5 and 17.5 cm<sup>-1</sup>, are shown in Figs. 2(a) and 2(c), respectively, at four different fields between 0 and 25 T. The pronounced reflectance drop upon heating is caused by the plasma edge shifting below the frequency of the applied radiation. After applying the magnetic field, the reflectance edge shifts only slightly to a lower temperature and the main effect of the field is an increase of the transition

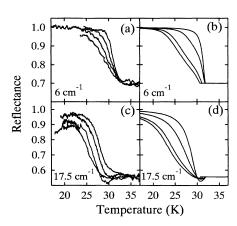


FIG. 2. Measured reflectance as a function of temperature for (a)  $\omega = 6$  and (c) 17.5 cm<sup>-1</sup> at (from right to left) B=0, 8, 20, and 25 T. Corresponding calculations based on the model by CC are shown in (b) respectively (d).

width. This again illustrates that even at 25 T there is no appreciable pair-breaking reduction of both  $T_c$  or  $\omega_p$ . Although it is known that the low-temperature upper critical field  $B_{c2}$  for the present orientation is quite high, it is surprising that the plasma frequency, which is a measure of the interplanar coupling strength, is hardly affected even at elevated temperatures.

In the following we will give a more quantitative discussion of the results in terms of existing theoretical models.

As described previously,4 the zero-field reflection edge can be fitted suitably in a Josephson coupled layer model, where the screened plasma frequency squared  $\omega_{ps}^2$  is proportional to the critical current density  $j_c$ , which in turn is related to the order parameter  $\Delta$  by the Ambegaokar-Baratoff expression for a Josephson tunnel junction.8 A similar Lawrence-Doniach-type model has been suggested in Ref. 9. The electromagnetic interaction of the induced currents with the vortex lattice due to the Lorentz force was treated explicitly in the work of Brandt, 10 Coffey and Clem 11 (CC), and more recently by Tachiki, Koyama, and Takahashi<sup>12</sup> (TKT). In these models, the interaction with the vortices is treated phenomenologically by introducing an effective pinning force constant  $\kappa_n$ , vortex viscosity  $\eta$ , and vortex mass M. In the absence of pinning one expects an enhanced microwave absorption due to dissipation in the cores of moving vortices. In the Bardeen-Stephen<sup>13</sup> approximation one can relate the vortex viscosity to the normal-state resistivity multiplied by the density and size of the vortex cores. It is not evident that the mass of the vortices can be neglected at FIR frequencies. Nevertheless, in the analysis presented here we took M = 0. Taking a finite value for M only adds an extra fitting parameter and does not change the main conclusions of this analysis.

A drawback of these models is that they are based on the semiphenomenological London approximation for the ac response of the superconductor with a two-fluid description of the quasiparticle contribution to the complex conductivity. As a result, none of the models is strictly applicable near the plasma edge. This is due to the fact that in the London approximation dielectric displacement currents are neglected. However, the essential cause of the plasma edge is that the

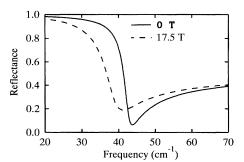


FIG. 3. Calculation using the TKT model of the effect of vortex motion on the plasma edge at 17.5 T (dashed line) together with a zero-field calculation (solid line).

inductive current carried by the condensate is balanced by the out-of-phase dielectric current of the bound charges, leading to a zero crossing of  $\epsilon_1(\omega)$ . The unique aspect of the high- $T_c$  superconductors is that the low c-axis conductivity leads to a plasma edge below the gap frequency. The fact that there is almost no damping due to dissipative currents ( $\epsilon_2$  is small) below the gap leads to the pronounced plasmon effects that are observed. It is important to note that it is the total current that drives the vortex motion by the Lorentz force and not just the free carrier supercurrent. This general argument shows that, at the zero crossing in  $\epsilon_1$ , the electromagnetic coupling to the vortices vanishes (except for the small resistive currents due to excited quasiparticles). Thus, in contrast to theoretical predictions, we expect basically no reflectance change near the plasma frequency due to the presence of vortices. The experimental observation that the reflectance minimum is independent of the field as shown in Fig. 1 nicely confirms this point.

A shift of the plasma frequency would, however, be possible due to a pair-breaking reduction of the superconductive order parameter. For the present orientation, the upper critical field is very high (above 100 T). Using standard theory for quasi-two-dimensional superconducting layers in a parallel field this would lead to a depression of the plasma frequency of 0.4 cm<sup>-1</sup> at the maximum field. This is within the experimental accuracy of the data.

For a more quantitative comparison with theoretical predictions we calculated the reflectance using the dielectric function of TKT:

$$\frac{\epsilon(\omega)}{\epsilon_0} = \epsilon_c \left( 1 - \frac{\omega_{p0}^2 - \omega_{pn}^2}{\omega(\omega + i0^+)} - \frac{\omega_{pn}^2}{\omega(\omega + i\gamma)} \right) \times \left[ 1 + \frac{\Phi_0}{\mu_0 \lambda^2(0)} \frac{B}{\kappa_p - i\omega\eta - M\omega^2} \right]^{-1}.$$
(1)

The results for zero and 17.5 T are shown in Fig. 3, where we used as basic parameters  $\kappa_p = 2500$  Pa,  $\lambda(0) = 6.8$   $\mu$ m,  $\omega_{p0} = 42.5$  cm<sup>-1</sup>,  $\omega_{pn} = 15$  cm<sup>-1</sup>,  $\gamma = 20$  cm<sup>-1</sup>, and  $\epsilon_c = 30$ . The vortex viscosity  $\eta$  was calculated from the Bardeen-Stephen relation using  $\rho_n(T) = 0.001$  25  $\Omega$  m. In this parameter set  $\kappa_p$  is the basic unknown. All other parameters are either determined directly by the experiment or taken from independent measurements on similar samples. For the present parameter range the TKT result is identical to

that of the earlier work by CC. Although the general agreement between theory and experiment is not too bad, an essential disagreement is the fact that the experimental plasma frequency is not field dependent as predicted. This is due to the London approximation that is used in the model, as already mentioned. We generalized the theoretical expressions to include displacement currents of the bound charges, and indeed find a much better agreement near the reflectance minimum. However, at present we are not able to reproduce the detailed shape of the observed reflectance edge. In particular, for the same parameters as above we find only a very small deviation from the zero-field curve. If we assume a much lower pinning force constant we find a somewhat better agreement near the edge, but a serious discrepancy at low frequencies arises.

In the present context it is not possible to give a full description of this generalized model and details will be published separately. We conclude that the qualitative agreement of the data with flux flow models is somewhat fortuitous and more theoretical work is needed to decide if the renormalization of the edge can be understood in terms of electromagnetic interaction with flux motion. Also it is not clear whether collective pinning and vortex deformation need to be incorporated. However, it is clear that pinning is substantial and a description in terms of pure Josephson vortices for magnetic fields parallel to the planes is not appropriate.

This brings us to another possible cause for the reflectance changes in a magnetic field: the vortex cores. In a generalized Abrikosov vortex model it is plausible to assume that the coupling between adjacent planes is no longer uniform and is locally depressed near the vortex cores. Within the assumption of local electrodynamics, this will lead to a local suppression of the plasma frequency and a broadening of the reflectance edge. Despite the small characteristic dimensions of the vortex core this influence can be quite substantial. This is because the large anisotropy in the coherence length implies a large elliptical eccentricity of the vortices and thus a large depolarization factor. Unfortunately, an effective medium model for a continuously varying dielectric function cannot be solved analytically. A preliminary numerical discrete approximation shows that accounting for the local depression of  $\omega_{ps}$  can indeed have an influence on the reflectivity close to the observed one.

Finally, we turn to the analysis of the high-temperature monochromatic data. At these elevated temperatures one may expect that vortices become more mobile and thermally activated (Brownian) motion becomes possible. To account for the thermal effects, we will use the CC approach. The basic expression for the dielectric function  $\epsilon_{\rm CC}(\omega)$  which includes both vortex motion and thermally activated creep effects is

$$\frac{\epsilon_{\text{CC}}(\omega)}{\epsilon_0} = \epsilon_c - \frac{c^2}{\omega^2} \frac{1 + 2i\lambda^2(B, T)\delta_{\text{nf}}^{-2}(\omega, B, T)}{\lambda^2(B, T) - (i/2)\tilde{\delta}_{\text{vc}}^2(\omega, B, T)}$$
(2)

in which  $\delta_{\rm nf}(\omega,B,T)$  and  $\tilde{\delta}_{\rm vc}(\omega,B,T)$  are, respectively, the normal fluid skin depth and the complex effective skin depth due to vortex motion and creep. We replaced some of the two fluid expressions used by CC by their analogs in the Josephson picture. From the relation  $\lambda = c/\omega_p$  follows for the

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temperature-dependent (Josephson) penetration depth  $\lambda(T)$  in the absence of pair-breaking effects,

$$\lambda^{2}(T) = \lambda^{2}(0) \frac{\Delta(0)}{\Delta(T)} \frac{1}{\tanh[\Delta(T)/2k_{B}T]}.$$
 (3)

The normal fraction is accordingly replaced by  $f(T) = 1 - \lambda^2(0)/\lambda^2(T)$ . For magnetic fields parallel to the planes, we assumed that  $B_{c2}^2(T) = B_{c2}^2(1-t^2)/(1+t^2)$  with  $t = T/T_c$  the reduced temperature.

The results, for the same parameters as in Fig. 3 and for  $B_{c2} = 120$  T,  $T_c = 32$  K, are shown in Figs. 2(b) and 2(d). U, the parameter which sets the energy scale for depinning is

taken as 1.5 eV T. Although there is no exact agreement, the general tendency of the magnetic-field effect is reproduced quite well.

In conclusion, we presented measurements of the c-axis reflectance in La<sub>1.85</sub>Sr<sub>0.15</sub>CuO<sub>4</sub> from 6 to 70 cm<sup>-1</sup> in fields up to 25 T. A small renormalization of the plasma edge is observed. No significant change in the plasma frequency nor in the absorption at the plasma frequency is found. This demonstrates that near the plasma edge one cannot neglect the displacement current due to bound charges as assumed in present models. The various results are fairly consistent and all imply a rather strong pinning of the vortices.

<sup>&</sup>lt;sup>1</sup>K. Tamasaku, Y. Nakamura, and S. Uchida, Phys. Rev. Lett. 69, 1455 (1992).

<sup>&</sup>lt;sup>2</sup>C.C. Homes, T. Timusk, R. Liang, D.A. Bonn, and W.N. Hardy, Phys. Rev. Lett. 71, 1645 (1993).

<sup>&</sup>lt;sup>3</sup>D.N. Basov, T. Timusk, B. Dabrowski, and J.D. Jorgensen, Phys. Rev. B **50**, 3511 (1994).

<sup>&</sup>lt;sup>4</sup> A.M. Gerrits, A. Wittlin, V.H.M. Duyn, A.A. Menovsky, J.J.M. Franse, and P.J.M. van Bentum, in proceedings M2S-HTSC IV, Grenoble, 1994 [Physica C **235-40**, 1117 (1994)].

<sup>&</sup>lt;sup>5</sup>T.W. Noh, S.G. Kaplan, and A.J. Sievers, Phys. Rev. B **41**, 307 (1990).

<sup>&</sup>lt;sup>6</sup> A.M. Gerrits, T.J.B.M. Janssen, A. Wittlin, N.Y. Chen, and P.J.M. van Bentum, in proceedings M2S-HTSC IV, Grenoble, 1994 [Physica C 235-40, 1115 (1994)].

<sup>&</sup>lt;sup>7</sup>K. Kishio, Y. Nakayama, N. Motohira, T. Noda, T. Kobayashi, K. Kitazawa, K. Yamafuji, I. Tanaka, and H. Kojima, Supercond. Sci. Technol. 5, S69 (1992).

<sup>&</sup>lt;sup>8</sup>V. Ambegaokar and A. Baratoff, Phys. Rev. Lett. **10**, 486 (1963); **11**, 104(E) (1963).

<sup>&</sup>lt;sup>9</sup>L.N. Bulaevskii, M. Zamora, D. Baeriswyl, H. Beck, and J.R. Clem, Phys. Rev. B 50, 12 831 (1994).

<sup>&</sup>lt;sup>10</sup>E.H. Brandt, Phys. Rev. Lett. **67**, 2219 (1991).

<sup>&</sup>lt;sup>11</sup>M.W. Coffey and J.R. Clem, Phys. Rev. Lett. 67, 386 (1991).

<sup>&</sup>lt;sup>12</sup> M. Tachiki, T. Koyama, and S. Takahashi, Phys. Rev. B **50**, 7065 (1994).

<sup>&</sup>lt;sup>13</sup>J. Bardeen and M.J. Stephen, Phys. Rev. **140**, A1197 (1965).

<sup>&</sup>lt;sup>14</sup>A.M. Gerrits and P.J.M. van Bentum (unpublished).