

**Phonon thermal conductivity in doped  $\text{La}_2\text{CuO}_4$ : Relevant scattering mechanisms**C. Hess,<sup>1,\*</sup> B. Büchner,<sup>2</sup> U. Ammerahl,<sup>3</sup> and A. Revcolevschi<sup>3</sup><sup>1</sup>*Département de Physique de la Matière Condensée, Université de Genève, Genève, Switzerland*<sup>2</sup>*Physikalisches Institut, RWTH-Aachen, 52056 Aachen, Germany*<sup>3</sup>*Laboratoire de Physico-Chimie, Université Paris-Sud, 91405 Orsay, France*

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Results of in-plane and out-of-plane thermal-conductivity measurements on  $\text{La}_{1.8-x}\text{Eu}_{0.2}\text{Sr}_x\text{CuO}_4$  ( $0 \leq x \leq 0.2$ ) single crystals are presented. The most characteristic features of the temperature dependence are a pronounced phonon peak at low temperatures and a steplike anomaly at  $T_{LT}$ , i.e., at the transition to the low-temperature-tetragonal phase (LTT phase), which gradually decrease with increasing Sr content. Comparison of these findings with the thermal conductivity of  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  and  $\text{La}_2\text{NiO}_4$  clearly reveals that in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  the most effective mechanism for phonon scattering is impurity scattering (dopants), as well as scattering by soft phonons that are associated with the lattice instability in the low-temperature-orthorhombic phase (LTO phase). There is no evidence that stripe correlations play a major role in suppressing the phonon peak in the thermal conductivity of  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ .

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**I. INTRODUCTION**

The segregation of spins and holes into stripelike arrangements appears to be a common feature of doped Mott insulators.<sup>1-5</sup> These so-called *stripe correlations* seem to be of particular importance in understanding the electronic phase diagram of high-temperature superconductors, where a competition between a static stripe phase and the superconducting phase is widely discussed.<sup>6-8</sup> Such a competition is expected to be reflected by the dynamics of stripes, i.e., static stripes should reduce the superconducting order parameter while stripes should be fluctuating in the presence of fully developed superconductivity. Indeed, there is growing experimental evidence for such a scenario.<sup>2,9-12</sup> While many experiments give evidence towards static stripes of holes and spins,<sup>2,13,14</sup> signatures of stripe fluctuations currently only comprise magnetic correlations.<sup>15-17</sup> A direct observation of fluctuating charge stripes, however, is still missing. A promising alternative approach to study stripe dynamics involves the phonon heat transport which is an indirect probing method. Since charge stripes lead to lattice distortions, a sensitivity of the phonon heat transport to the dynamics as well as the degree of periodicity of stripes can be expected.

The thermal conductivity  $\kappa$  of doped  $\text{La}_2\text{CuO}_4$  has repeatedly been the subject of experimental research, yet no detailed understanding of the rather complicated and strong changes of the temperature dependence of  $\kappa$  upon partial substituting Sr and/or small rare earths (RE) like Nd or Eu for La has been achieved. For example, in the antiferromagnetic insulators  $\text{La}_{2-y}(\text{RE})_y\text{CuO}_4$ , a huge peak at room temperature which arises due to magnetic heat transport is found in the thermal conductivity parallel to the  $\text{CuO}_2$  planes ( $\kappa_{ab}$ ), while the thermal conductivity perpendicular to the  $\text{CuO}_2$  planes ( $\kappa_c$ ) is purely phononic without a high-temperature peak.<sup>18</sup> Similarly intriguing is a strong suppression of the phononic low-temperature peak in both  $\kappa_{ab}$  and  $\kappa_c$ , which is found in the superconducting doping levels of  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ .<sup>19</sup> There is a seeming correlation between this suppression and superconductivity because a phononic

low-temperature peak reappears in overdoped, nonsuperconducting  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  as well as in  $\text{La}_{2-x-y}(\text{RE})_y\text{Sr}_x\text{CuO}_4$ , where superconductivity is suppressed in favor of static stripe order.<sup>9,10,20</sup> Baberski *et al.*<sup>20</sup> qualitatively explained these observations based on the idea that in superconducting compounds fluctuating stripes provide a new scattering channel for phonons. In recent studies by Sun *et al.*, such a scattering channel plays an important role in the data interpretation.<sup>21-23</sup>

There is clear-cut evidence that in isostructural stripe ordering  $\text{La}_{2-x}\text{Sr}_x\text{NiO}_4$ , the phonon thermal conductivity  $\kappa_{\text{ph}}$  is closely correlated with both the dynamics and the periodicity of stripes. While  $\kappa_{\text{ph}}$  is almost unaffected in the presence of static and long-range ordered stripes, it is strongly suppressed as soon as the stripes become disordered or dynamic.<sup>24-26</sup> Apparently, in these compounds the thermal conductivity is indeed a probe for stripe correlations. One might question, however, whether this is true also in the cuprates for two reasons. First, the electron-phonon coupling in the nickelates is much stronger than in the cuprates.<sup>27-32</sup> Therefore, the effect of stripes on  $\kappa_{\text{ph}}$  can be expected to be much smaller in the cuprates. Second, from a structural point of view the situation in the nickelates is much simpler than in the cuprates. In the latter, a structural instability exists and as a consequence a number of structural phase transitions occur as a function of temperature as well as of Sr and RE content.<sup>9</sup> Since the structural instability involves soft phonon modes,<sup>33-36</sup> enhanced scattering of the heat carrying phonons is likely.

In this paper we reinvestigate the phonon thermal conductivity  $\kappa_{\text{ph}}$  of doped  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  and present new experimental results on Eu-doped single crystals. The single crystalline data allow us to investigate the anisotropy of the  $\kappa$  tensor and provide more precise absolute values of  $\kappa$ . In previous measurements on polycrystals,<sup>20,37</sup> this information was not available. It is, however, necessary in order to judge the strength and therefore the importance of various scattering mechanisms for phonons. The analysis of our data yields compelling arguments that both impurities (dopants) and soft

phonons, which are associated with the lattice instability in these compounds, strongly scatter phonons and therefore must not be neglected in the data interpretation. Data on  $\text{La}_2\text{NiO}_4$  corroborate this conclusion and allow us to qualitatively understand  $\kappa_{\text{ph}}$  of  $\text{La}_{2-x-y}(\text{RE})_y\text{Sr}_x\text{CuO}_4$  in a wide doping range. In particular, it is not necessary to include a stripe induced scattering channel.

The structure of this paper is as follows. After a brief description of the experimental details in Sec. II, we review previous results on the thermal conductivity of doped  $\text{La}_2\text{CuO}_4$  in Sec. III, before we proceed to the presentation of our new experimental results on  $\text{La}_{1.8-x}\text{Eu}_{0.2}\text{Sr}_x\text{CuO}_4$  in Sec. IV. Our main results will be discussed in Sec. V.

## II. EXPERIMENT

We have prepared single crystals of  $\text{La}_{1.8-x}\text{Eu}_{0.2}\text{Sr}_x\text{CuO}_4$  ( $x=0, 0.08, 0.15, 0.2$ ) as well as of  $\text{La}_2\text{NiO}_4$  utilizing the traveling solvent floating zone technique.  $\kappa$  of these crystals was measured as a function of temperature  $T$ . We used a standard steady-state method on pieces cut along the principal axes with a typical length of 2 mm along the measuring direction and of about 0.5 mm for the two other directions. The thermal gradient was determined by measuring the temperature difference  $\Delta T$  between the junctions of a differential Au/Fe-Chromel thermocouple. The junctions of this thermocouple have been glued onto the sample using GE varnish.<sup>38</sup>  $\Delta T$  varied between 0.5% and 2% of the absolute temperature, which has been stabilized for each data point. Errors due to radiation loss, which could occur at higher temperatures, are avoided in our experimental setup. Stoichiometric oxygen contents in  $\text{La}_{1.8}\text{Eu}_{0.2}\text{CuO}_4$  and  $\text{La}_2\text{NiO}_4$  were achieved by annealing in high vacuum and  $\text{CO}/\text{CO}_2$  atmosphere, respectively.

## III. PREVIOUS RESULTS

Prior to discussing our new experimental results, we review previous results<sup>19,20</sup> on the striking doping dependence of the thermal conductivity of doped  $\text{La}_2\text{CuO}_4$ . At first, we concentrate on the low-temperature phononic peak in the thermal conductivity of  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ . As is evident from the lower panel of Fig. 1, which reproduces  $\kappa_c$  of  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  ( $x=0, 0.1, 0.15, 0.2, 0.3$ ) as published by Nakamura *et al.*,<sup>19</sup> this peak evolves nonmonotonically with increasing Sr content. A well pronounced phononic low- $T$  peak in  $\kappa_c$  is only present at  $x=0$  and  $x=0.3$ , whereas at intermediate doping levels ( $0.1 \leq x \leq 0.2$ ) a peak structure is hardly identifiable. Note that the material is a superconductor in this doping range whereas it is insulating and metallic at  $x=0$  and  $x=0.3$ , respectively.

Baberski *et al.* have pointed out that the suppressed low- $T$  peak at intermediate Sr doping reappears upon RE doping, provided that this doping induces the so-called LTT phase (low-temperature-tetragonal) and thereby suppresses superconductivity.<sup>20</sup> This is illustrated in the upper panel of Fig. 1, where  $\kappa$  of polycrystalline Pr- and Nd-doped  $\text{La}_{2-x-y}(\text{RE})_y\text{Sr}_x\text{CuO}_4$  at the finite Sr content  $x=0.12$  is shown. The Pr-doped compound does not undergo the tran-

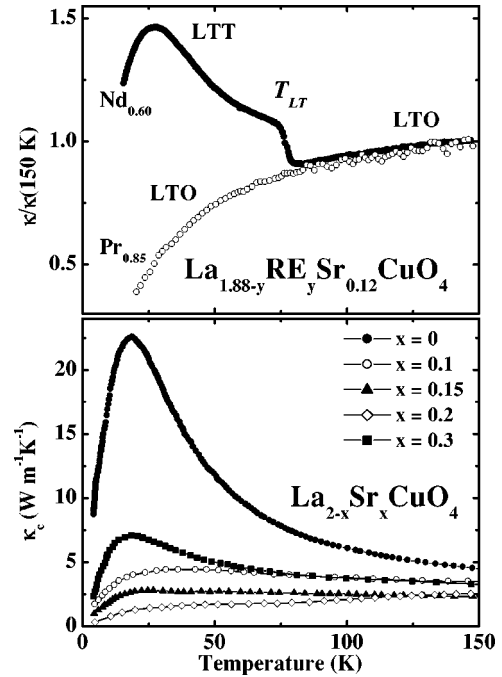


FIG. 1. Bottom: Temperature dependence of  $\kappa_c$  of  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  for  $x=0, 0.1, 0.15, 0.2$ , and  $0.3$ . Data are reproduced from Ref. 19. Top: Thermal conductivity of Pr ( $y=0.85$ ) and Nd ( $y=0.6$ ) doped  $\text{La}_{1.88-y}(\text{RE})_y\text{Sr}_{0.12}\text{CuO}_4$  polycrystals normalized to  $\kappa(150\text{ K})$  as a function of temperature. Data are taken from Ref. 20.

sition to the LTT phase. Like  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ , it remains in the so-called LTO phase (low-temperature-orthorhombic) and is a superconductor.<sup>39</sup> Its thermal conductivity monotonically decreases with decreasing  $T$  and hence is very similar to the aforementioned findings by Nakamura *et al.* for  $\kappa$  of  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  at  $0.1 \leq x \leq 0.2$ . The situation is completely different in the nonsuperconducting, Nd-doped compound, which is in the LTT phase below  $T_{LT} \approx 80\text{ K}$ . Here,  $\kappa_{\text{ph}}$  abruptly enhances at  $T_{LT}$  and exhibits a well pronounced phononic peak around 25 K.

The doping dependence of  $\kappa$  described above must be attributed to strong changes in the phonon thermal conductivity  $\kappa_{\text{ph}}$  of this material. Nonphononic contributions to  $\kappa$  can be excluded in the out-of-plane direction, since the electrical conductivity<sup>40,41</sup> and the magnetic coupling<sup>42</sup> along the  $c$  axis are too small to allow significant electronic and magnetic thermal conduction. Concomitantly, the doping dependence of the low- $T$  peak is evident along all crystal directions<sup>19</sup> (cf. also the data below).

In crystalline materials, the low- $T$  peak in  $\kappa_{\text{ph}}$  is very sensitive to scattering of phonons. Generally, the height of this peak reduces with increasing phonon scattering rates. One important mechanism in this regard is scattering of phonons by impurities. In alloyed compounds like doped  $\text{La}_2\text{CuO}_4$ , this phonon-impurity scattering is induced by nonuniform ions on one lattice site. Upon alloying, this should lead to a gradual reduction of the phonon peak.<sup>43</sup> Even though phonon-defect scattering inevitably must be present in this material, it is not a scattering mechanism

which solely dominates  $\kappa_{\text{ph}}$ , because the phonon peak evolves nonmonotonically upon Sr doping and even reappears at additional RE doping.

Another scattering mechanism for phonons in this material is suggested by the abrupt change of  $\kappa_{\text{ph}}$  at the structural phase transition, which occurs in the Nd-doped compound. It appears that phonons are scattered stronger in the LTO phase. This interpretation is confirmed by neutron-scattering studies on Nd-doped  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  at  $T_{LT}$ . The acoustic phonon line width abruptly decreases at the transition from the LTO phase into the LTT phase, signaling a proportional decrease of scattering processes.<sup>44</sup> Anomalous phonon thermal conductivity in the vicinity of structural phase transitions is well known from a number of perovskite oxides as, for example,  $\text{SrTiO}_3$  and  $\text{KTaO}_3$ . There, a suppression of  $\kappa_{\text{ph}}$  is caused by enhanced scattering of acoustic phonon modes due to their energetic degeneracy with soft optical phonon modes.<sup>45,46</sup> Indeed, soft phonon branches do exist in the LTO phase of doped  $\text{La}_2\text{CuO}_4$ ,<sup>33-36</sup> which could cause a suppression of  $\kappa_{\text{ph}}$  (in the following this scattering mechanism will briefly be named ‘soft-phonon scattering’). The change of  $\kappa_{\text{ph}}$  at  $T_{LT}$  then would follow from the discontinuous hardening of the soft phonon branch in the LTT phase<sup>35,36</sup> and an associated reduced scattering rate of acoustic phonons.<sup>47</sup>

Since all compounds with a suppressed phonon peak are in the LTO phase at low  $T$ , soft-phonon scattering could be important for understanding the suppression of the peak as well.<sup>49</sup> There is, however, not a one to one correlation between the LTO phase, i.e., the possible presence of soft phonon scattering, and the suppression of the peak. This is most obvious in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ , because the soft phonon properties only slightly change for  $x \leq 0.2$ , whereas the phonon peak is maximum for  $\text{La}_2\text{CuO}_4$ .

Baberski *et al.* have noticed that both impurity scattering and soft-phonon scattering separately do not allow to understand the suppression of the phonon peak. They therefore suggested that the suppression of the phononic peak could be correlated with the superconducting properties, since the peak suppression occurs whenever the material is superconducting.<sup>20</sup> In their model, they proposed that stripes couple to phonons and thereby cause an unconventional scattering mechanism for phonons depending on the dynamics of stripes. A suppression of the phonon peak then is the consequence of fluctuating stripes, which are present in the superconducting compounds, while  $\kappa_{\text{ph}}$  exhibits a usual phonon peak when stripes are static or even absent in the nonsuperconducting cases. There, static stripes are only present in the LTT phase. Therefore, instead of being caused by an abrupt softening of optical phonon modes, the jump of  $\kappa_{\text{ph}}$  at  $T_{LT}$  could just as well originate from a change of the stripe dynamics from static (LTT) to fluctuating (LTO). Even though a consistent interpretation of the data discussed afore is possible within such a model, its validity is questionable because the actual role of impurity- and soft-phonon scattering remains unclear.

#### IV. NEW RESULTS

In order to elucidate the importance of this conventional scattering mechanisms we now turn to our measurements on

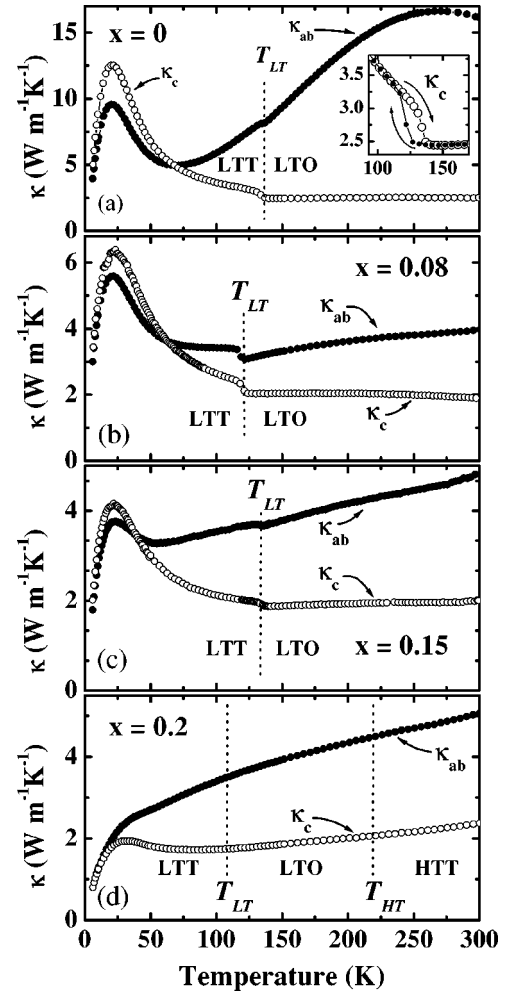


FIG. 2. Thermal conductivity of  $\text{La}_{1.8-x}\text{Eu}_{0.2}\text{Sr}_x\text{CuO}_4$  [ $x=0, 0.08, 0.15, 0.2$ , panels (a) to (d)] along the  $c$  axis ( $\kappa_c$ , open circles) and parallel to the  $ab$  planes ( $\kappa_{ab}$ , closed circles) as a function of temperature. Inset:  $\kappa_c$  for  $x=0$  near  $T_{LT}$  at cooling (full circles) and heating (open circles).

single crystals, which provide profound information in this regard.

In Fig. 2, we present the complete data set of  $\kappa$  of  $\text{La}_{1.8-x}\text{Eu}_{0.2}\text{Sr}_x\text{CuO}_4$  ( $x=0, 0.08, 0.15, 0.2$ ) measured along the  $c$  axis ( $\kappa_c$ ) and parallel to the  $ab$  planes ( $\kappa_{ab}$ ) in the temperature range 7–300 K. All compounds are in the LTT phase at  $T < T_{LT}$ , with a variation of  $T_{LT}$  between about 110 K and 130 K. For  $T > T_{LT}$  and  $x \leq 0.15$ , the structure is LTO in the investigated temperature range. At  $x=0.2$ , the compound undergoes a further structural phase transition from the LTO into the so-called HTT phase (high-temperature-tetragonal) at  $T_{HT} \approx 220$  K.<sup>10</sup>

Here  $\kappa_c$  of  $\text{La}_{1.8}\text{Eu}_{0.2}\text{CuO}_4$  [cf. panel (a) of Fig. 2] exhibits a low-temperature peak around 20 K with a falling edge that continuously extends to  $T_{LT} \approx 130$  K. A jumplike decrease occurs at  $T_{LT}$ , followed by a constant  $\kappa_c$  up to room temperature. The inset of Fig. 2 depicts a cooling and heating curve of  $\kappa_c$  in the area of the jump. A clear hysteresis being a characteristic feature of first-order phase transitions is evident.



For  $T \lesssim 50$  K, the thermal conductivity along the  $\text{CuO}_2$  planes,  $\kappa_{ab}$ , is comparable to  $\kappa_c$ . The peak centered around 20 K is slightly smaller; we find  $\kappa_c^{\text{max}}/\kappa_{ab}^{\text{max}} \approx 1.3$ , which is similar to the findings in nondoped  $\text{La}_2\text{CuO}_4$  and isostructural  $\text{La}_{5/3}\text{Sr}_{1/3}\text{NiO}_4$ .<sup>19,24</sup> For  $T \gtrsim 50$  K, the temperature dependencies of  $\kappa_{ab}$  and  $\kappa_c$  differ completely. With rising temperature,  $\kappa_{ab}$  strongly increases and evolves into a broad peak at room temperature. A steplike anomaly is present at  $T_{LT}$ , which is in a similar way hysteretic as the anomaly in  $\kappa_c$  (not shown).

The panels (b) and (c) of Fig. 2 show  $\kappa_{ab}$  and  $\kappa_c$  of  $\text{La}_{1.8-x}\text{Eu}_{0.2}\text{Sr}_x\text{CuO}_4$  for  $x=0.08$  and  $x=0.15$ , respectively. For both compounds  $\kappa_{ab}$  and  $\kappa_c$  exhibit a low temperature peak, where the peak of  $\kappa_{ab}$  is again slightly smaller than that of  $\kappa_c$ . The temperature dependence of  $\kappa_c$  is very similar to that of  $\kappa_c$  of  $\text{La}_{1.8}\text{Eu}_{0.2}\text{CuO}_4$  in the whole temperature range. As in the case at  $x=0$ ,  $\kappa_{ab}$  deviates from the qualitative  $T$  dependence of  $\kappa_c$  above  $\sim 50$  K and evolves into an increase with rising temperature. A steplike anomaly at  $T_{LT}$  exists in each case, but no high-temperature peak as in  $\text{La}_{1.8}\text{Eu}_{0.2}\text{CuO}_4$  is present.

A comparable anisotropy as in the previous compounds is also evident at  $x=0.2$  [cf. panel (d) of Fig. 2], but the low-temperature peak in  $\kappa_c$  is almost completely suppressed and  $\kappa_c$  slightly increases for  $T \gtrsim 75$  K with nearly constant positive slope. The low-temperature peak is even absent in  $\kappa_{ab}$ . Here,  $\kappa_{ab}(T)$  monotonically increases upon heating in the entire temperature range. At  $T_{LT} \approx 110$  K no anomaly is present, neither in  $\kappa_{ab}$  nor in  $\kappa_c$ . Apparently, the transition at  $T_{HT}$  also causes no anomaly in the thermal conductivity.

## V. DISCUSSION

### A. Anisotropy

As mentioned before,  $\kappa_c$  is purely phononic, since electronic and magnetic contributions can be ruled out for heat transport along the  $c$  axis. In order to understand the anisotropies of  $\kappa$  for  $T \gtrsim 50$  K, it is reasonable to distinguish the cases  $x=0$  and  $x>0$ . It has been shown in Ref. 18 that in insulating, antiferromagnetic  $\text{La}_{1.8}\text{Eu}_{0.2}\text{CuO}_4$  the high-temperature peak of  $\kappa_{ab}$  must be explained by magnetic contributions. Upon Sr doping, the doped holes destroy the magnetic order, which leads to a strong suppression of the high-temperature peak (cf. Fig. 2). Simultaneously, the material becomes electrically conducting within the  $ab$  planes.<sup>18,19</sup> Therefore, the much smaller, but apparently still existing anisotropy in the Sr-doped compounds is due to electronic, rather than magnetic, contributions to  $\kappa_{ab}$ . Indeed, an estimation using the Wiedemann-Franz law yields electronic contributions of the same order of magnitude as the observed anisotropy for  $\kappa_{ab}$  and negligible electronic contributions for  $\kappa_c$ . The magnetic and electronic contributions to  $\kappa_{ab}$  become important for  $T \gtrsim 50$  K. Below this temperature,  $\kappa_{ab}$  can be considered to be primarily phononic with a similar magnitude of  $\kappa_{ph}$  as  $\kappa_c$ .<sup>52</sup> This is nicely confirmed by the very similar temperature dependence of  $\kappa_{ab}$  and  $\kappa_c$  in this temperature range, as shown in Fig. 3. It is remarkable that for all compounds the phonon peak of  $\kappa_c$  is slightly larger

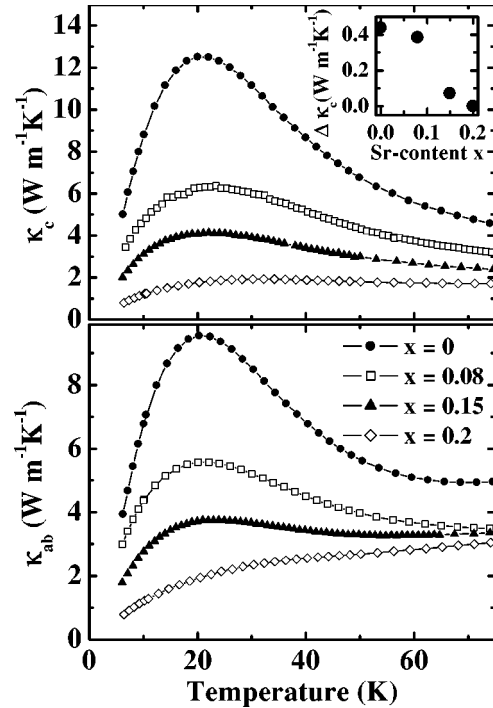


FIG. 3. Doping dependence of low-temperature peak in the thermal conductivity of  $\text{La}_{1.8-x}\text{Eu}_{0.2}\text{Sr}_x\text{CuO}_4$  along the  $c$  axis ( $\kappa_c$ , top) and parallel to the  $ab$  planes ( $\kappa_{ab}$ , bottom). Inset: Doping dependence of the jump size  $\Delta\kappa_c$  at  $T_{LT}$ .

than that of  $\kappa_{ab}$  as long as a peak is clearly resolved in both quantities. A possible explanation could be related to slightly different velocities of the acoustic phonons along the  $ab$  and  $c$  directions.<sup>53</sup>

### B. Phononic peak and jump at $T_{LT}$

The development of the low-temperature peak of  $\kappa_{ph}$  upon doping can be viewed in Fig. 3. Obviously, the peak size is gradually reduced as the Sr content increases. A similar result is found for the size of the jump  $\Delta\kappa_c$  at  $T_{LT}$ , which is shown as a function of doping in the inset of Fig. 3. While the gradual reduction of the peak size can straightforwardly be explained by phonon-impurity scattering, the reduction of the jump at  $T_{LT}$  requires further comments. First of all, we stress the presence of a jump in insulating  $\text{La}_{1.8}\text{Eu}_{0.2}\text{CuO}_4$ . It clearly proves that it is caused by soft-phonon scattering in the LTO phase, which therefore has to be regarded as an important scattering channel for phonons indeed. When this scattering channel becomes active in the LTO phase, the relative importance of phonon-impurity scattering is reduced. The gradual reduction of the jump size  $\Delta\kappa_c$  with increasing Sr content may therefore be attributed to doping induced phonon-impurity scattering as well. However, we should note that the structural differences between the LTT and LTO phases diminish with increasing Sr content. Therefore, apart from phonon-impurity scattering, structural reasons play a role in the suppression of the jump at  $T_{LT}$ . This is consistent with the observation by Baberski *et al.*, that the jump disappears, whenever the tilting angle of the  $\text{CuO}_6$  octahedra becomes lower than a critical value.<sup>20</sup>

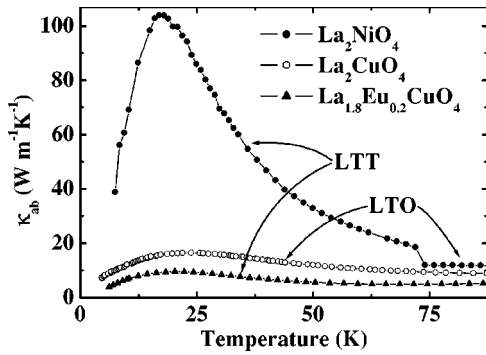


FIG. 4. Temperature dependence of  $\kappa_{ab}$  of  $\text{La}_2\text{NiO}_4$  in comparison with  $\kappa_{ab}$  of  $\text{La}_2\text{CuO}_4$  and  $\text{La}_{1.8}\text{Eu}_{0.2}\text{CuO}_4$ . The data for  $\text{La}_2\text{CuO}_4$  are reproduced from Ref. 19.

It is now very instructive to consider  $\kappa_{\text{ph}}$  of pure  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  (cf. Fig. 1) for comparison. Apparently, doping Sr into  $\text{La}_2\text{CuO}_4$  suppresses the phonon peak much more effectively than in the Eu-doped counterpart. On one hand the phonon peak of nondoped  $\text{La}_2\text{CuO}_4$  is by a factor of about 2 larger than in nondoped  $\text{La}_{1.8}\text{Eu}_{0.2}\text{CuO}_4$ . On the other hand the phonon peaks in  $\text{La}_{1.8-x}\text{Eu}_{0.2}\text{Sr}_x\text{CuO}_4$  are clearly better developed for finite Sr contents with higher maximum values as in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ . One has therefore to conclude that a further scattering mechanism exists in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  which becomes more important upon doping and which is absent or at least much weaker in the Eu-doped compounds. In this case, this mechanism must cause a more effective scattering than the surely present phonon-impurity scattering induced by the Eu ions. One plausible candidate (besides scattering due to dynamic stripes) for such a mechanism is soft-phonon scattering connected with the structural instability of the LTO phase since the soft-phonon energies decrease further upon Sr doping<sup>54</sup> and hence enhance soft-phonon scattering.

In order to judge the relevance of soft-phonon scattering a measure for its strength with respect to phonon-impurity scattering is necessary. Such a measure could be achieved, for example, by comparison with a compound where neither a lattice instability nor doped impurities are present. Though being no cuprate, isostructural  $\text{La}_2\text{NiO}_4$  is yet a well suited candidate perfectly fulfilling these requirements. The phonon spectra of  $\text{La}_2\text{CuO}_4$  and  $\text{La}_2\text{NiO}_4$  are almost identical<sup>55,56</sup> because the atomic masses of Ni and Cu are very similar. At low temperatures ( $T \leq 73$  K), this compound is in the LTT phase and hence a structural instability does not exist.<sup>57</sup> Moreover, no phonon-impurity scattering is present in this nondoped compound.

Figure 4 presents our result for phononic<sup>58</sup>  $\kappa_{ab}$  of electrically insulating  $\text{La}_2\text{NiO}_4$  in comparison with  $\kappa_{ab}$  of  $\text{La}_2\text{CuO}_4$  as measured by Nakamura *et al.* and with  $\kappa_{ab}$  of  $\text{La}_{1.8}\text{Eu}_{0.2}\text{CuO}_4$ . It is intriguing that the phonon peak of  $\text{La}_2\text{NiO}_4$  is about one order of magnitude larger than the peak in both  $\text{La}_2\text{CuO}_4$  and  $\text{La}_{1.8}\text{Eu}_{0.2}\text{CuO}_4$ . At  $T_{LT} \approx 73$  K, the structure changes from LTT to LTO, which also in this material causes a jumplike decrease in the thermal conduc-

tivity. Compared to  $\text{La}_{1.8}\text{Eu}_{0.2}\text{CuO}_4$  the jump size is larger by a factor of about 10. In the LTO phase,  $\kappa_{ab}$  is of the similar size as in  $\text{La}_2\text{CuO}_4$ .

It immediately follows from this observation that in  $\text{La}_2\text{CuO}_4$  as well as in  $\text{La}_{1.8}\text{Eu}_{0.2}\text{CuO}_4$ , the heat carrying phonons are subject to severe scattering at low temperatures. In both cases this scattering obviously suppresses the peak of  $\kappa_{\text{ph}}$  by about one order of magnitude. Translated to the strength of the two discussed scattering mechanisms, this means that referring to a non-impurity-doped LTT phase both, soft-phonon scattering and phonon-impurity scattering independently of each other reduce the peak of  $\kappa_{\text{ph}}$  by almost the same amount.

This conclusion now allows us to understand the doping dependence of the phonon peak, when a combination of both, phonon-impurity and soft-phonon scattering is considered. In  $\text{La}_{1.8-x}\text{Eu}_{0.2}\text{Sr}_x\text{CuO}_4$ , where the thermal conductivity is already strongly influenced by the Eu ions, Sr doping simply further increases the phonon-impurity scattering rate. This consequently leads to a gradual reduction of the phonon peak with increasing Sr content. Soft-phonon scattering is not relevant here.

In  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  with the same Sr content than a Eu-doped counterpart, the phonon-impurity scattering rate is slightly reduced since no Eu ions are present. Yet, the resulting phonon thermal conductivity is somewhat smaller. Hence, the effect of a slightly reduced phonon-impurity scattering rate must be overcompensated by soft-phonon scattering. This is indeed reasonable since it is a qualitatively different scattering mechanism whose importance upon Sr doping grows since thereby the relevant soft modes soften further.<sup>50</sup>

The reappearance of the phonon peak in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  at high Sr concentrations as  $x = 0.3$  can now be understood as a natural consequence of the doping dependence of  $T_{HT}$ . For  $x \geq 0.22$ , the LTO phase disappears in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  and the structure remains in the HTT phase for all temperatures.<sup>59</sup> Hence, for  $x = 0.3$  soft-phonon scattering associated with the LTO phase is not active for such overdoped compounds. The result of such decreased phonon scattering is the reappearance of the phonon peak.

It is necessary to mention that the growing density of holes as the Sr content is increased in principle could be a further source of scattering and therefore contribute to the reduction of  $\kappa_{\text{ph}}$  and in particular the phonon peak via phonon-electron scattering. However, since the transition to superconductivity in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  causes no significant anomaly in  $\kappa_c$  at  $T_c$  (cf. Fig. 1), this scattering mechanism is usually considered to be unimportant in this material and its RE-doped relatives.<sup>20</sup> Yet if this scattering channel is active in this material no inconsistency to our interpretation arises because in this case the phonon-impurity scattering induced by Sr ions can be regarded simply as slightly more effective than the phonon-impurity scattering induced by Eu ions.

### C. Stripes as a scattering mechanism for phonons?

The above discussion of  $\kappa_{\text{ph}}$  of the insulating materials  $\text{La}_2\text{CuO}_4$ ,  $\text{La}_{1.8}\text{Eu}_{0.2}\text{CuO}_4$ , and  $\text{La}_2\text{NiO}_4$  provides unam-

biguous evidence that phonon impurity and soft-phonon scattering are important scattering mechanisms for phonons in doped  $\text{La}_2\text{CuO}_4$ . Since the major doping dependencies of the low- $T$  peak can be explained based on these mechanisms without any problems, there is no compelling reason to incorporate a stripe-induced scattering channel in the data interpretation. It is unlikely though that stripes in doped  $\text{La}_2\text{CuO}_4$  have no effect at all on  $\kappa_{\text{ph}}$  because the aforementioned stripe-induced phonon scattering in the nickelates<sup>24,25</sup> is an unambiguous physical fact. However, it appears extremely difficult to prove the existence and to study the strength of purely stripe-induced scattering in doped  $\text{La}_2\text{CuO}_4$  via  $\kappa_{\text{ph}}$ . This is not only because phonon impurity and soft-phonon scattering are already dominating  $\kappa_{\text{ph}}$  in insulating  $\text{La}_2\text{CuO}_4$ ; due to the intimate relation of LTT phase and stripe dynamics, scattering on fluctuating stripes in the Sr-doped compounds could be viewed as an altered but already existing soft-phonon scattering, i.e., from this point of view these two scattering mechanisms are conceptually indistinguishable.

#### D. High temperature increase of $\kappa_{\text{ph}}$

For completeness, we briefly mention the unusual temperature dependence of  $\kappa_c$ , which is evident at higher temperatures  $T \gtrsim 100$  K. Instead of a decrease as  $\sim T^{-1}$ , which at elevated temperatures is usually expected for thermal con-

ductivity by *acoustic* phonons,  $\kappa_c$  is almost temperature independent or even increases with rising  $T$ . As has been shown in Ref. 60, this strong deviation from the usual behavior arises due to thermal conduction by dispersive *optical* phonons in addition to the usual contribution by acoustic phonons.

#### VI. SUMMARY

In summary, we have presented experimental results on the thermal conductivity of  $\text{La}_{1.8-x}\text{Eu}_{0.2}\text{Sr}_x\text{CuO}_4$  for a wide doping range of Sr. The analysis of our data suggests that in this material, phonons are strongly scattered on the doped impurities, i.e., Sr and Eu, as well as on soft phonons that are present in the LTO phase of this material. Comparison of our data with the thermal conductivity of  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  and isostructural  $\text{La}_2\text{NiO}_4$  leads to the conclusion that these scattering mechanisms are most relevant in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  as well. In particular, in contrast to other studies,<sup>20-22</sup> there remains no direct evidence that the stripe correlations cause a relevant scattering channel in doped  $\text{La}_2\text{CuO}_4$ .

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<sup>1</sup>J.M. Tranquada, D.J. Buttrey, V. Sachan, and J.E. Lorenzo, Phys. Rev. Lett. **73**, 1003 (1994).

<sup>2</sup>J.M. Tranquada, B.J. Sternlieb, J.D. Axe, Y. Nakamura, and S. Uchida, Nature (London) **375**, 561 (1995).

<sup>3</sup>J.M. Tranquada, J.E. Lorenzo, D.J. Buttrey, and V. Sachan, Phys. Rev. B **52**, 3581 (1995).

<sup>4</sup>S.-H. Lee and S.-W. Cheong, Phys. Rev. Lett. **79**, 2514 (1997).

<sup>5</sup>S. Mori, C.H. Chen, and S.-W. Cheong, Nature (London) **392**, 473 (1998).

<sup>6</sup>S.R. White and D.J. Scalapino, Phys. Rev. B **60**, 753 (1999).

<sup>7</sup>S. Chakravarty, R.B. Laughlin, D.K. Morr, and C. Nayak, Phys. Rev. B **63**, 094503 (2001).

<sup>8</sup>J. Zaanen, Nature (London) **422**, 569 (2003).

<sup>9</sup>B. Büchner, M. Breuer, A. Freimuth, and A.P. Kampf, Phys. Rev. Lett. **73**, 1841 (1994).

<sup>10</sup>H.-H. Klauss, W. Wagener, M. Hillberg, W. Kopmann, H. Walf, F.J. Litterst, M. Hücker, and B. Büchner, Phys. Rev. Lett. **85**, 4590 (2000).

<sup>11</sup>B. Lake *et al.*, Science **291**, 1759 (2001).

<sup>12</sup>B. Lake *et al.*, Nature (London) **415**, 299 (2002).

<sup>13</sup>J.M. Tranquada, J.D. Axe, N. Ichikawa, A.R. Moodenbaugh, Y. Nakamura, and S. Uchida, Phys. Rev. Lett. **78**, 338 (1997).

<sup>14</sup>J.M. Tranquada, N. Ichikawa, and S. Uchida, Phys. Rev. B **59**, 14 712 (1999).

<sup>15</sup>S.-W. Cheong, G. Aeppli, T.E. Mason, H. Mook, S.M. Hayden, P.C. Canfield, Z. Fisk, K.N. Clausen, and J.L. Martinez, Phys. Rev. Lett. **67**, 1791 (1991).

<sup>16</sup>K. Yamada, C.H. Lee, K. Kurahashi, J. Wada, S. Wakimoto, S. Ueki, H. Kimura, Y. Endoh, S. Hosoya, G. Shirane, R.J. Birge-

neau, M. Greven, M.A. Kastner, and Y.J. Kim, Phys. Rev. B **57**, 6165 (1998).

<sup>17</sup>H.A. Mook, P. Dai, S.M. Hayden, G. Aeppli, T.G. Perring, and F. Doğan, Nature (London) **395**, 580 (1998).

<sup>18</sup>C. Hess, B. Büchner, U. Ammerahl, L. Colonescu, F. Heidrich-Meisner, W. Brenig, and A. Revcolevschi, Phys. Rev. Lett. **90**, 197002 (2003).

<sup>19</sup>Y. Nakamura, S. Uchida, T. Kimura, N. Motohira, K. Kishio, K. Kitazawa, T. Arima, and Y. Tokura, Physica C **185-189**, 1409 (1991).

<sup>20</sup>O. Baberski, A. Lang, O. Maldonado, M. Hücker, B. Büchner, and A. Freimuth, Europhys. Lett. **44**, 335 (1998).

<sup>21</sup>X.F. Sun, J. Takeya, S. Komiya, and Y. Ando, Phys. Rev. B **67**, 104503 (2003).

<sup>22</sup>X.F. Sun, S. Komiya, and Y. Ando, Phys. Rev. B **67**, 184512 (2003).

<sup>23</sup>X.F. Sun, Y. Kurita, T. Suzuki, S. Komiya, and Y. Ando, cond-mat/0308263 (unpublished).

<sup>24</sup>C. Hess, B. Büchner, M. Hücker, R. Gross, and S.-W. Cheong, Phys. Rev. B **59**, 10 397 (1999).

<sup>25</sup>D. Cassel, C. Hess, B. Büchner, M. Hücker, R. Gross, O. Friedt, and S.-W. Cheong, J. Low Temp. Phys. **117**, 1083 (1999).

<sup>26</sup>S.-H. Lee, J.M. Tranquada, K. Yamada, D.J. Buttrey, Q. Li, and S.-W. Cheong, Phys. Rev. Lett. **88**, 126401 (2002).

<sup>27</sup>H. Eisaki, S. Uchida, T. Mizokawa, H. Namatame, A. Fujimori, J. van Elp, P. Kuiper, G.A. Sawatzky, S. Hosoya, and H. Katayama-Yoshida, Phys. Rev. B **45**, 12 513 (1992).

<sup>28</sup>V.I. Anisimov, M.A. Korotin, J. Zaanen, and O.K. Andersen, Phys. Rev. Lett. **68**, 345 (1992).

<sup>29</sup>X.-X. Bi and P.C. Eklund, Phys. Rev. Lett. **70**, 2625 (1993).

- <sup>30</sup>C.H. Chen, S.-W. Cheong, and A.S. Cooper, Phys. Rev. Lett. **71**, 2461 (1993).
- <sup>31</sup>J. Zaanen and P.B. Littlewood, Phys. Rev. B **50**, 7222 (1994).
- <sup>32</sup>S.-W. Cheong, H.Y. Hwang, C.H. Chen, B. Batlogg, L.W. Rupp, and S.A. Carter, Phys. Rev. B **49**, 7088 (1994).
- <sup>33</sup>R.J. Birgeneau, C.Y. Chen, D.R. Gabbe, H.P. Jenssen, M.A. Kastner, C.J. Peters, P.J. Picone, T. Thio, T.R. Thurston, H.L. Tuller, J.D. Axe, P. Böni, and G. Shirane, Phys. Rev. Lett. **59**, 1329 (1987).
- <sup>34</sup>L. Pintschovius, Festkörperprobleme **30**, 183 (1990).
- <sup>35</sup>J.L. Martínez, M.T. Fernández-Díaz, J. Rodríguez-Carvajal, and P. Odier, Phys. Rev. B **43**, 13 766 (1991).
- <sup>36</sup>B. Keimer, R.J. Birgeneau, A. Cassanho, Y. Endoh, M. Greven, M.A. Kastner, and G. Shirane, Z. Phys. B: Condens. Matter **91**, 373 (1993).
- <sup>37</sup>M. Sera, M. Maki, M. Hiroi, and N. Kobayashi, J. Phys. Soc. Jpn. **66**, 765 (1997).
- <sup>38</sup>IMI 7031 Insulating Varnish.
- <sup>39</sup>W. Schäfer, M. Breuer, G. Bauer, A. Freimuth, N. Knauf, B. Roden, W. Schlabitz, and B. Büchner, Phys. Rev. B **49**, 9248 (1994).
- <sup>40</sup>Y. Nakamura and S. Uchida, Phys. Rev. B **46**, 5841 (1992).
- <sup>41</sup>Y. Nakamura and S. Uchida, Phys. Rev. B **47**, 8369 (1993).
- <sup>42</sup>T. Thio, T.R. Thurston, N.W. Preyer, P.J. Picone, M.A. Kastner, H.P. Jenssen, D.R. Gabbe, C.Y. Chen, R.J. Birgeneau, and A. Aharony, Phys. Rev. B **38**, 905 (1988).
- <sup>43</sup>R. Berman and J. Brock, Proc. R. Soc. London, Ser. A **289**, 46 (1965).
- <sup>44</sup>L. Pintschovius and W. Reichardt, in *Neutron Scattering in Layered Copper-Oxide Superconductors*, edited by A. Furrer (Kluwer Academic, Dordrecht, 1998), pp. 165–224.
- <sup>45</sup>E.F. Steigmeier, Phys. Rev. **168**, 523 (1968).
- <sup>46</sup>H.H. Barret and M.G. Holland, Phys. Rev. B **2**, 3441 (1970).
- <sup>47</sup>We should note that Sera *et al.* explain the change of  $\kappa_{\text{ph}}$  at  $T_{LT}$  by an observed change of the velocity of sound  $v_s$  at  $T_{LT}$  (Ref. 37). The actual changes of  $v_s$  are, however, far too small ( $\sim 1\%$ ) (Ref. 48) to account for the much larger changes of  $\kappa_{\text{ph}}$ . We therefore regard these changes of  $v_s$  at  $T_{LT}$  as a further accompanying phenomenon of the structural phase transition.
- <sup>48</sup>J. Yamada, M. Sera, M. Sato, T. Takayama, M. Takata, and M. Sakata, J. Phys. Soc. Jpn. **63**, 2314 (1994).
- <sup>49</sup>Note that the reported degeneracy of energy of acoustic and soft optical phonons is  $\sim 10$  meV (Ref. 33). Hence, the effect of soft-phonon scattering on  $\kappa_{\text{ph}}$  should be strongest around  $\sim 25$  K, i.e., in the vicinity of the phononic peak, since acoustic phonons with energy  $\sim 4k_B T$  contribute most significantly to  $\kappa_{\text{ph}}$  (Ref. 51). Apart from this lowest-order scattering, three-phonon processes and processes of higher order involving much softer phonon modes close to the Brillouin zone center with energies between 2 meV and 5 meV (Refs. 33,36,50) are very likely.
- <sup>50</sup>M. Braden, W. Schnelle, W. Schwarz, N. Pyka, G. Heger, Z. Fisk, K. Gamayunov, I. Tanaka, and H. Kojima, Z. Phys. B: Condens. Matter **94**, 29 (1994).
- <sup>51</sup>R. Berman, *Thermal Conduction in Solids* (Clarendon Press, Oxford, 1976).
- <sup>52</sup>Note that the elastic constants of this material are only weakly anisotropic (Ref. 53).
- <sup>53</sup>L. Pintschovius, N. Pyka, W. Reichardt, A.Y. Rumiantsev, N.L. Mitrofanov, A.S. Ivanov, G. Collin, and P. Bourges, Physica C **185-189**, 156 (1991).
- <sup>54</sup>The low- $T$  energy of the soft mode at the zone center decreases from 2.44 meV ( $x=0$ ) to 1.82 meV ( $x=0.13$ ) (Ref. 50).
- <sup>55</sup>L. Pintschovius, J.M. Bassat, P. Odier, F. Gervais, G. Chevrier, W. Reichardt, and F. Gompf, Phys. Rev. B **40**, 2229 (1989).
- <sup>56</sup>L. Pintschovius, W. Reichardt, M. Braden, G. Dhalenne, and A. Revcolevschi, Phys. Rev. B **64**, 094510 (2001).
- <sup>57</sup>J. Rodríguez-Carvajal, J.L. Martínez, J. Pannetier, and R. Saez-Puche, Phys. Rev. B **38**, 7148 (1988).
- <sup>58</sup>We note that similar, as in  $\text{La}_2\text{CuO}_4$ , magnetic contributions might be present for  $T \geq 100$  K.
- <sup>59</sup>H. Takagi, R.J. Cava, M. Marezio, B. Batlogg, J.J. Krajewski, W.F. Peck, P. Bordet, and D.E. Cox, Phys. Rev. Lett. **68**, 3777 (1992).
- <sup>60</sup>C. Hess and B. Büchner, cond-mat/0304248 (unpublished).