# Quantitative evidence for spin-fluctuation-mediated higher-harmonic *d*-wave components: Hole- and electron-doped cuprates

David Parker<sup>1,2,3</sup> and Alexander V. Balatsky<sup>4</sup>

<sup>1</sup>Max-Planck Institute for the Physics of Complex Systems, Nöthnitzer Strasse 38, D-01187 Dresden, Germany <sup>2</sup>Max-Planck Institute for the Chemical Physics of Solids, Nöthnitzer Strasse 40, D-01187 Dresden, Germany

<sup>3</sup>U.S. Naval Research Laboratory, 4555 Overlook Avenue SW, Washington, DC 20375, USA

<sup>4</sup>Theoretical Division, MS-B262, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA

(Received 24 July 2008; published 1 December 2008)

Experimental evidence for higher harmonics in the cuprate *d*-wave gap function  $\cos(2\phi)$  or  $\cos(k_x) - \cos(k_y)$  has been slowly mounting. Here we analyze all of the data that have been generated in this field for both hole-doped and electron-doped cuprates, demonstrating a preponderance of these symmetry-allowed terms. We then show that the simple spin-fluctuation pairing mechanism can reproduce the detailed angular dependence of the gap. We also present an analysis of pseudogap higher-harmonic symmetry based on the picture of the Fermi arcs as being due to lifetime broadening.

DOI: 10.1103/PhysRevB.78.214502

PACS number(s): 74.72.Hs, 74.20.Rp

## I. INTRODUCTION

In the mid-to-late 1990s, predominant *d*-wave  $(d_{x^2-y^2})$  symmetry was established in the hole-doped cuprates<sup>1-4</sup> following a long controversy. Later work has established that at least some of the electron-doped cuprates have predominant *d*-wave symmetry, although with an interesting, unusual nonmonotonic gap function based on the Raman data of Blumberg<sup>5</sup> and of Matsui.<sup>6</sup>

As is well known, the square symmetry of the CuO<sub>2</sub> plane permits gap basis functions obeying the same symmetry properties (vanishing along  $k_x = k_y$  and related directions, and even parity and antisymmetric around the nodal lines) but of higher angular momentum, such as  $\cos(6\phi)$  or  $\cos(10\phi)$ . In general, all these gap functions belong to the same irreducible representation of the rotation group  $C_{4V}$  applicable to the cuprates (ignoring orthorhombic distortions as observed in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> (YBCO), so there is no physical reason for them to be excluded, either at zero or finite temperature. Reasonable gap functions, of course, will require that the expansion coefficients of the higher-harmonic basis functions decrease rapidly with increasing angular momentum.

The first cuprate study indicating the possibility of higher harmonics was performed by Mesot *et al.*,<sup>7</sup> who examined via angle-resolved photoemission spectroscopy (ARPES) a number of Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>8</sub> (Bi-2212) samples, both overdoped and underdoped, with maximum gap values ranging from 25 to 45 meV. This group found that the overdoped samples contained essentially no higher harmonics, with the weighting of the second harmonic  $\cos(6\phi)$  term varying between 0% and 4% [in units of the  $\cos(2\phi)$  fraction], but a substantially larger fraction—as large as 12%—on the underdoped side. These underdoped higher-harmonic terms resulted in a slope of the gap near the node approximately half that of a pure  $\cos(2\phi)$  gap function and were attributed to an increasing importance of long-range interactions with underdoping as one nears the antiferromagnetic insulating state.

Higher-harmonic structure in the hole-doped cuprates was next examined by Borisenko *et al.*,<sup>8</sup> who measured via ARPES the gap in the presence of bilayer splitting in under-

doped ( $T_c$ =77 K) (Pb,Bi)-2212. This work found a much larger proportion of the cos(6 $\phi$ ) harmonic, at 27%. The data were analyzed using the leading edge gap method for determination of the gap, which does introduce some uncertainty as this method is largely a qualitative measure of the gap. This analysis was largely focused on a comparative determination of gap symmetry on the bilayer-split Bi-2212 Fermi surfaces so that the 27% figure should be considered only as an estimate of the higher-harmonic content.

More recently, Terashima *et al.*<sup>9</sup> studied optimally doped  $La_{1.85}Sr_{0.15}CuO_4$  (LSCO) via ARPES. They found a gap function which deviated significantly from the pure  $cos(2\phi)$  form but did not interpret these data in terms of higher harmonics because a simple fit with the additional  $cos(6\phi)$  term was found to be unsatisfactory. These data were then interpreted in terms of a two-gap picture, as in Ref. 10. We show in Sec. II that these data are in fact consistent with a higher-harmonic picture when the effect of still higher-order terms in the expansion is included.

The first high-resolution ARPES work on YBCO was performed recently by Nakayama *et al.*,<sup>11</sup> who found a gap that essentially followed the  $cos(2\phi)$  form with no  $6\phi$  component. This sample was estimated to be slightly overdoped, with p=0.175, consistent with Mesot's earlier data.<sup>7</sup> Also recently,<sup>12</sup> Yoshida reported ARPES measurements on optimally and underdoped LSCO, again interpreted in terms of a two-gap picture.

Regarding higher-harmonic gap symmetry in the electrondoped cuprates, the relevant works are those of Blumberg<sup>5</sup> and Matsui.<sup>6</sup> Blumberg performed Raman scattering on Nd<sub>2-x</sub>Ce<sub>x</sub>CuO<sub>4</sub> and found evidence for a nonmonotonic *d*-wave gap function, with a gap maximum not at  $\phi=0$  but at  $\phi=\pi/6$ , much closer to the node at  $\pi/4$ . This spot is believed to correspond to the "hot spot" (the intersection of the Fermi surface with the antiferromagnetic Brillouin-zone boundary) and is thus strong evidence for spin-fluctuation mediated pairing. Matsui found a similar result via ARPES measurements on Pr<sub>0.89</sub>LaCe<sub>0.11</sub>CuO<sub>4</sub>, with the best fit gap function  $\Delta(\phi)=1.43\cos(2\phi)-0.43\cos(6\phi)$ ; here the negative 6 $\phi$  component produces the nonmonotonicity. Recent tunneling work by Dagan *et al.*<sup>13</sup> also presented evidence for a nonmonotonic *d*-wave order parameter in the electron-doped cuprates, with measurements on  $Pr_{2-x}Ce_xCuO_{4-\delta}$  for various Ce dopings. These data were best fit by a nonmonotonic *d*-wave order parameter, with an angle of maximum  $\Delta$  ranging from 16° to 23°, relative to the antinodal direction.

The several data sets described above lead to the conclusion that with the exception of overdoped hole-doped cuprates, higher harmonics are a fairly robust property of experimental cuprate *d*-wave gap functions. In Sec. II we show how these harmonics arise naturally within a spin-fluctuation mediated pairing scenario.

## **II. MODEL**

In this section we briefly present the spin-fluctuation pairing model; detailed descriptions of this model are contained in Refs. 14 and 15. The spin-fluctuation pairing mechanism as proposed by Monthoux *et al.*<sup>14</sup> is as follows:

$$H_{\rm int} = \frac{1}{V} \sum_{\mathbf{q}} g(\mathbf{q}) s(\mathbf{q}) \cdot S(-\mathbf{q}), \qquad (1)$$

where

$$\mathbf{s}(\mathbf{q}) = \frac{1}{2} \sum_{\alpha,\beta,\mathbf{k}} \psi^{\dagger}_{\alpha,\mathbf{k}+\mathbf{q}} \sigma_{\alpha,\beta} \psi_{\mathbf{k},\beta}.$$
 (2)

Here **S** is the spin-fluctuation operator, while  $\sigma_{\alpha,\beta}$  is the quasiparticle Pauli spin operator, with  $\psi^{\dagger}$ , the usual quasiparticle creation operator, and  $g(\mathbf{q})$ , a (momentum dependent in general) coupling constant. The correlation function of **S**, representing propagation of a spin fluctuation, is empirically modeled by the susceptibility,

$$\chi(\mathbf{k} - \mathbf{k}', \omega) = \frac{1}{\xi^{-2} + (\mathbf{k} - \mathbf{k}' - \mathbf{Q})^2 - i\frac{\xi^{-2}\omega}{\omega_{c}}},$$
(3)

$$=\frac{\chi_0}{1+\xi^2(\mathbf{k}-\mathbf{k}'-\mathbf{Q})^2-i\frac{\omega}{\omega_{\rm sf}}}.$$
 (4)

Here  $\xi$  is the antiferromagnetic correlation length,  $\omega_{\rm sf}$  is the spin-fluctuation frequency,  $\chi_0$  is the long-wavelength limit of the susceptibility ( $\propto \xi^2$ ), and the ordering vector  $\mathbf{Q} = (\pm \frac{\pi}{a}, \pm \frac{\pi}{a})$ .

In a weak-coupling BCS framework, the above susceptibility leads to the following gap equation (we neglect the imaginary part of the susceptibility, as in Monthoux *et al.*<sup>14</sup>):

$$\Delta(\mathbf{k}) = \int g \frac{d^2 \mathbf{k}'}{(2\pi)^2} \frac{\operatorname{Re}[\chi(\mathbf{k} - \mathbf{k}', E_k)] \operatorname{tanh}(E_{k'}/2T) \Delta(\mathbf{k}')}{2E_{k'}}.$$
(5)

Here  $E_{k'} = \sqrt{(\varepsilon_{k'} - \mu)^2 + \Delta^2(\mathbf{k}')}$ ,  $\mu$  is the chemical potential, and  $\varepsilon_{k'}$  is the quasiparticle dispersion which we take as

$$\varepsilon_{k'} = -2t(\cos k'_x + \cos k'_y) + 4t' \cos k'_x \cos k'_y -2t''(\cos 2k'_x + \cos 2k'_y),$$
(6)

with  $t \equiv 1$ , t' = 0.22, t'' = 0.1. The values assumed for t' and t'' were chosen for consistency with the previous work,<sup>15</sup> but are also within the range of fitted experimental parameters for several cuprates, as published in Ref. 16. It should be emphasized that the results of this paper do not significantly depend upon the choice of these parameters, so long as some basic features [such as the generally  $(\pi, \pi)$ -centered Fermi surface in hole-doped materials and motion of the hot spots along the zone diagonal with underdoping] are respected. In physical units we take t as 0.35 eV for the hole-doped cuprates and 0.13 eV for the electron doped,<sup>17</sup> and the coupling constant g as approximately 1.1 eV for the hole-doped materials and 1.6 eV for the electron doped.

In the previous work<sup>15</sup> this gap equation was solved in a quasi-two-dimensional framework by assuming that  $\Delta(\mathbf{k})$  depended only on the Fermi-surface angle, not on the distance from the Fermi surface; in this work we use a fully twodimensional calculation, solving for the gap everywhere in the Brillouin zone and then extracting the angular dependence of the gap on the Fermi surface. For these calculations, unlike in Ref. 15, we assume a finite spin-fluctuation frequency consistent with more microscopic modeling<sup>17</sup> and recent experiment.<sup>18</sup> For the hole-doped cuprates we take  $\omega_{sf}$ =0.05t, and for the electron-doped materials we take  $\omega_{\rm sf}$ =0.04t. These finite spin-fluctuation frequencies play an important role in reducing the gap anisotropy; in the previous work<sup>15</sup> gap functions which were too flat in the nodal region (relative to experiment) were generated. Even with this modification, it was found necessary to use rather short correlation lengths to reproduce the detailed angular form of the gap since, as shown in Ref. 15, this form is rather less sensitive to correlation length than in a one-dimensional approximation.<sup>19</sup> For the calculations here  $\xi/a$  (a is the lattice spacing) was taken as 3.3 for the hole-doped cuprates and 3.0 for the electron-doped materials.

The values of  $\omega_{\rm sf}$  and  $\xi/a$  chosen for the cuprates were selected based upon available experimental data,<sup>18,20</sup> as well as values extracted from more microscopic theoretical modeling.<sup>17</sup> The hole-doped  $\omega_{\rm sf}$  of 0.05*t* compares reasonably well with the optimally doped value of 0.09*t* (this value also decreases with underdoping) calculated by use of the FLEX Eliashberg calculation.<sup>17</sup> Published hole-doped data<sup>20</sup> on the momentum-space half-width of inelastic neutronscattering excitations (the inverse of the correlation length) give values between 0.12 and 0.26 Å<sup>-1</sup>, corresponding to  $\xi/a$  between 1 and 2, but this value increases with underdoping so that the value of 3.3 is a reasonable one.

Regarding the electron-doped cuprates, available inelastic neutron-scattering data<sup>18</sup> indicate  $\chi''$  (imaginary part of the spin susceptibility) maxima for energies between 2 and 6 meV, depending on doping. In a simple Ornstein-Zernicke spin-fluctuation model, these maxima correspond to the spinfluctuation frequency, and accordingly we have taken  $\omega_{sf}$  as  $0.04t \approx 5$  meV. With respect to the value of  $\xi/a$ , these data indicated linewidths of 0.02-0.08 Å, corresponding to  $\xi/a$ 



FIG. 1. (Color online) Normalized gap amplitudes from several data sets, as indicated, are compared with theoretical normalized gap amplitudes assuming spin-fluctuation pairing. Insets: unnormalized data and fits.

values of 12 (electron doping x=0.09) and 3 (x=0.18), at an energy of 12 meV. The modeled doping level for the electron-doped cuprates was approximately 0.18 so the value of 3 is a fair choice.

#### **III. RESULTS AND DISCUSSION**

Depicted in Fig. 1 is the main result of this paper. We show in Fig. 1(a) the several gap function data sets on hole-doped cuprates along with our theoretical result, and in Fig. 1(b) the corresponding electron-doped result with the parameters as indicated. We note excellent agreement for both hole-doped and electron-doped cuprates.

The hole-doped data sets, as previously indicated, show substantial variation, likely as a result of material-specific band structure, renormalization, and doping effects, and we note that our theoretical result fits well near the average. Another possible reason for the variation is a difference in the microscopic parameters  $\xi/a$  and  $\omega_{\rm sf}$ . The electron-doped results are reasonably well fit by the theory; due to the narrow range of electron-doped superconducting dopings and materials, and fewer data sets, these results are more consistent. The electron-doped results of Blumberg<sup>5</sup> can be fit even more closely if one changes the chemical potential so that the hot spots move further toward the node. However this also results in a substantial reduction in absolute gap magnitude. In this regime an extended s-wave solution may be energetically favored, since in a d-wave scenario as the hot spots move toward the zone diagonal  $(k_x = k_y)$  the gap maximum (generally found at or near the hot spots) is forced nearer to the node, thus diminishing the gap amplitude.<sup>21</sup> We therefore ascribe the observed *d*-wave nature of the order parameter to strong-coupling effects not included in our weak-coupling formalism. All the plots present scaled gap functions, as well as absolute gap values (insets). These absolute gap value results are of roughly the same size as the experimental results. We do not attempt to fit the experimentally observed magnitude of the gap for each data set theoretically. We note that with the exception of the Nakayama<sup>11</sup> YBCO data, the theory captures the gap slope near the node very well. This is done without explicit recourse to a "second energy scale" although the spin-fluctuation frequency is an additional energy scale (and in the hole-doped case of the approximate magnitude of the gap). As indicated in Fig. 1(a), the hole-doped data are generally doped in the range from optimal to underdoped; we have not included Mesot's optimally or overdoped data, which show smaller harmonic content than presented here.

The data indicate a substantial discrepancy between the data of Borisenko<sup>8</sup> and Mesot,<sup>7</sup> despite the fact that both these samples are underdoped and in the Bi-2212 family [as indicated, the data of Borisenko are on a (Pb-Bi)-2212 sample]. The discrepancy is further heightened by the fact that  $T_c$ 's for these two samples are nearly identical (75 vs 77 K). It is possible that the lead doping in Borisenko's sample affects the gap symmetry, but in the previous work<sup>15</sup> we found that impurity scattering, at least in the Born and unitary limits, had little effect on gap symmetry. Since the reason for the discrepancy is unclear and both sets of data appear to be accepted by the scientific community, we have simply attempted to fit the majority of the data, which are closer to that of Borisenko.

Based on the dispersion assumed above and the Luttinger sum rule, the hole-doped cuprates are modeled at a hole doping of 0.30 and the electron-doped cuprates at an electron doping of 0.17. The hole doping, in particular, may appear somewhat anomalous given that it corresponds to a severely overdoped regime, while most of the data presented are for optimally to underdoped cuprates. However, in this theoretical formalism the relevant factor affecting the gap symmetry is the location of the hot spot, where the antiferromagnetic Brillouin zone crosses the Fermi surface. It is at this hot spot that the pairing potential is maximized and the gap function maximum generally occurs at this point. At a particular doping, this hot spot lies exactly at the Van Hove singularity at  $(\pi, \pi)$ . Recently, Storey *et al.*<sup>22</sup> have presented evidence of a universal doping for this point in the range of x=0.23. Using the Luttinger sum rule, with our dispersion this crossing occurs 0.07 holes/plaquette away from this point. This places the hole doping at x=0.16 or optimal doping.

One natural question to ask is the evolution of the gap function shape with doping. In particular, is it possible to reproduce the more nearly  $\cos(2\phi)$  shape of the overdoped hole-doped compounds within this formalism, and how should the gap shape change with increasing underdoping? Figure 1(a) attempts to fit the majority of the data with a single-gap function, but it is plausible that significant changes in gap symmetry with doping may occur.

Regarding the first question, it turns out that it is very difficult within this formalism to obtain a gap function significantly closer to the pure  $\cos(2\phi)$  form. Changes in correlation length, as shown in Ref. 15, have only a small impact on gap symmetry so that even substantially shorter correlation lengths do not reproduce the pure  $\cos(2\phi)$  form observed on the overdoped side, and in addition severely reduce the overall (unscaled) gap amplitude.

As for the change in the shape of the gap with underdoping, from a theoretical perspective there are two main factors at work: the change in correlation length and spin-fluctuation frequency with underdoping and the motion of the hot spots away from the antinodal direction. As one underdopes,  $\xi$ generally increases while  $\omega_{sf}$  decreases. These two behaviors have the opposite effect on gap amplitude, so that the net gap amplitude effect is unclear. Regarding the gap symmetry, a decrease in spin-fluctuation frequency tends to make the gap less anisotropic, as this frequency cuts out the pairing interaction beyond a given range in quasiparticle energy  $(=\sqrt{\varepsilon_k^2 + \Delta_k^2})$ . The net effect is that regions which would otherwise have large gap amplitudes have these amplitudes substantially reduced, while near-nodal areas do not, thus rendering the gap less anisotropic. As mentioned previously, the hot-spot motion tends to move the region of maximum gap away from the antinode.

Complicating any attempt to model theoretically the change in gap shape with underdoping is that the several data sets do not show any obvious trend with underdoping; of the three data sets in Fig. 1(a) which are best fit by the model calculation, two are optimally doped and one is underdoped, while the underdoped data of Mesot are much closer to the pure  $\cos(2\phi)$  form of the slightly overdoped sample. This is at odds with Mesot's pioneering work,<sup>7</sup> which found a general trend of increasing harmonic content with underdoping. Differences in sample preparation may have an effect, but as yet the general trend of gap symmetry with underdoping apart from the motion of the gap maximum, reflected in the divergent behavior in Figs. 1(a) and 1(b)] is not completely clear. For this reason we have focused on the existence of the higher harmonics and leave the subject of doping dependence for future study.

A point of interest in the theoretical formalism is that gap symmetry, surprisingly, depends to a small extent on *the coupling constant g*. Naively, one would expect this value to simply affect the overall scale of the gap function, not its detailed shape, but as shown in the Appendix, this is not true.

#### IV. FERMI ARCS AND PSEUDOGAP HIGHER-HARMONIC GAP SYMMETRY

The Fermi arcs<sup>23–25</sup> present one unusual aspect of cuprate physics, in which ARPES measurements of the pseudogap state show a vanishing gap magnitude at certain momenta (as measured by the peak-to-peak separation in the photoemission spectra). These arcs have a length which depends quasilinearly on the ratio  $T/T^*$ , where  $T^*$  is the temperature for the opening of the pseudogap. Within a single-gap picture (in which the pseudogap corresponds to a precursor d-wave pairing state), it has been noted by several authors<sup>24-27</sup> that this can be explained as arising from inelastic quasiparticle lifetime effects. If the quasiparticle scattering rate  $\Gamma > \sqrt{3}\Delta(\phi)$ , then the ARPES  $k_F$  energy distribution curve will exhibit zero gap for quasiparticle momenta, with momenta in direction  $\phi$ . Storey *et al.*,<sup>22</sup> in particular, noted that if the quasiparticle lifetime is taken as linearly proportional to temperature, one reproduces rather well the experimental data, as indicated in Fig. 2(a).

Recently, Chubukov *et al.*<sup>25</sup> completed an Eliashberg-type calculation of the effects of inelastic scattering upon the Fermi arcs and found an arc-length which increased quasilinearly with temperature, as shown in Fig. 2(a). As is evident, however, at low temperatures the arc length is not well fit by this microscopic calculation, and as observed in Ref. 25 a gap function flatter nearer the nodes would result in a better fit to the data.

To this end we have undertaken a simple calculation which *extracts* the gap symmetry in the pseudogap phase from the experimental length of the Fermi arcs<sup>23</sup> and the results of the Eliashberg theoretical calculation. Our first observation is that the length *L* of the Fermi arcs, in radians, is well modeled by  $L = \frac{\sin^{-1}[t(\equiv T/T^*)]}{2}$ , as shown in Fig. 2(a). The inelastic-scattering rate  $\Gamma(T)$  is of the form  $\frac{\alpha\Omega_0}{\sinh(\Omega_0/T)}$ , with  $T^*$ taken as 300 K and the mode energy  $\Omega_0$  taken as 28 meV  $\approx 300$  K. Substituting  $t=T/T^*$ , we then have  $\Gamma(t)$  $= \frac{\alpha\Omega_0}{\sinh(\Omega_0/tT^*)} = \frac{\alpha\Omega_0}{\sinh(1/t)}$ . The condition determining the angle  $\phi$ at which the Fermi arcs begin is that  $\Gamma(t) = \sqrt{3}\Delta(\phi)$ . Now, from the previous empirical relationship we have  $t = \sin(2L)$ . The relationship between *L* and the angle  $\phi$  where the Fermi arc begins is  $L = \pi/4 - \phi$ , so that we have, for the condition determining the length of the Fermi arcs,  $t = \cos(2\phi)$ , so that

$$\Delta(\phi) = \frac{\alpha \Omega_0}{\sqrt{3} \sinh[1/\cos(2\phi)]},\tag{7}$$

$$\propto 1/\sinh(1/\cos(2\phi)). \tag{8}$$

The constant of proportionality is clearly  $\sinh(1)=1.1752$  so that we find



FIG. 2. (Color online) The Fermi arc data, the results of Ref. 25, and our empirical fit are presented in panel (a). Note that the theoretical calculation of Storey (Ref. 24), in which the quasiparticle scattering rate  $\Gamma(T)$  is assumed to be linear in temperature, is essentially identical with the empirical curve  $L = [1/2 \sin^{-1}(T/T^*)]$ . Panel (b): the pseudogap symmetry extracted from the first plot, along with  $\cos(2\phi)$  and the theoretical curve from the model hole-doped calculation.

$$\Delta(\phi) = \frac{1.1752}{\sinh[1/\cos(2\phi)]} \tag{9}$$

for the gap symmetry in the pseudogap state. We have plotted this function, along with  $\cos(2\phi)$  and the gap function determined for the superconducting state, in Fig. 2(b). Significant differences in symmetry between the empirical pseudogap symmetry and predicted superconducting state symmetry are apparent, with the pseudogap approaching the  $\cos(2\phi)$  shape much more closely than the theoretical curve. It should be mentioned that differences of this order exist in the various superconducting data sets, so one should not yet conclude that the pseudogap and superconducting state have different higher-harmonic content. Nevertheless, for reference we present here a table of the leading terms (Table I), as determined by the fits to ARPES and Raman data, in the gap functions of the cuprates. The gap function along the Fermisurface angle  $\phi$  can be expanded as

$$\Delta(\phi) = A_0 \cos(2\phi) + A_1 \cos(6\phi) + \cdots . \tag{10}$$

The superconducting (SC) results are determined by the theoretical spin-fluctuation pairing calculation fit to the data, while the pseudogap result is a result of the empirical fit to

TABLE I. Higher harmonic gap function content.

Coefficients of $\cos(n\phi)$ terms			
Phase	$2\phi$	$6\phi$	$10\phi$
Hole doped, SC	0.732	0.211	0.052
Electron doped, SC	1.024	-0.373	-0.019
Hole doped, PG	0.913	0.128	-0.058

the Fermi arc data using the Eliashberg model. We note that the electron-doped values have a large negative value for the  $\cos(6\phi)$  term, which results from the nonmonotonic nature of the order parameter.<sup>5,6</sup> We also note that the pseudogap  $6\phi$ term is substantially smaller than the hole-doped superconducting, which results from its more  $\cos(2\phi)$ -like dependence for small  $\phi$ , as depicted in Fig. 2. The 10 $\phi$  terms are generally smaller than the  $6\phi$ , as is physically reasonable, and vary in sign, depending on the detailed form of the gap function. A simple analysis of the experimental error bars from the data which these fits have aimed to duplicate suggests that all of the coefficients, with the possible exception of the electron-doped  $\cos(10\phi)$  term, are statistically significant, with error bars of approximately 0.04 for the  $\cos(6\phi)$ term and 0.015 for the  $\cos(10\phi)$  term. The rapid decrease in these terms with higher order, however, means that for practical purposes terms of higher order than depicted here are likely to be statistically insignificant and of little interest.

#### V. COMMENT: ONE GAP VS TWO GAPS

The fits of the ARPES and Raman data for the superconducting state lead to the suggestion that the superconducting gap as observed in these measurements *is* the gap and that there is only one gap. However, if that is the case the superconducting and pseudogap should have the same shape, and our fits indicate substantial differences. These fits are of course dependent on the Eliashberg model, which assumed an Einstein bosonic mode at 28 meV. However, the model was based on the empirical ARPES data and seems reasonable. Further experimental and theoretical study will clearly be necessary to resolve this important issue. At present we are not aware of evidence that allows one to rule out the possibility of two distinct gaps.

#### **VI. CONCLUSION**

In this paper we have summarized the evidence from a substantial body of data, including Raman, ARPES, and tunneling measurements, indicating the presence of higher harmonics to the fundamental  $\cos(2\phi)$  term of the superconducting and pseudogap order parameters. We have presented good theoretical fits to these data based on a spin-fluctuation pairing mechanism with reasonable values of spin-fluctuation frequencies and correlation lengths for the superconducting data, and an empirical fit to the pseudogap pairing symmetry based on Fermi arc-length measurements. We comment on the implications of our results for the one-gap-two-gap controversy.

## ACKNOWLEDGMENTS

This work was supported by the U.S. DOE (A.V.B.).

#### APPENDIX

In this section we show explicitly that within a weakcoupling approach the gap function symmetry is not scale invariant—i.e., changes in the coupling constant g affect the shape of the gap function as well as its overall size.

We begin with the T=0 weak-coupling gap equation

$$\Delta(\mathbf{k}) = -\sum_{\mathbf{k}'} \alpha V_{k,k'} \frac{\Delta_{\mathbf{k}'}}{2E_{k'}}.$$
 (A1)

Here  $E_{k'} = \sqrt{\xi_{k'}^2 + \Delta^2(\mathbf{k}')}$  is the standard BCS quasiparticle energy and  $\alpha$  is a constant. Now we consider the effect of

changing  $V_{k,k'}$  by a constant factor, resulting in a different gap equation

$$\Delta(\mathbf{k},\beta) = -\sum_{\mathbf{k}'} \beta V_{k,k'} \frac{\Delta(\mathbf{k}',\beta)}{2E_{k',\beta}}.$$
 (A2)

If the gap function is to be scale invariant, we must have  $\Delta(\mathbf{k}, \beta) = f(\alpha, \beta)\Delta(\mathbf{k})$ . Substituting this in the second equation above and canceling common factors of  $f(\alpha, \beta)$ , one finds the following gap equation:

$$\Delta(\mathbf{k}) = -\sum_{\mathbf{k}'} \beta V_{k,k'} \frac{\Delta(\mathbf{k}')}{2\sqrt{\xi_{k'}^2 + f^2(\alpha,\beta)\Delta^2(\mathbf{k}')}}.$$
 (A3)

This gap equation is equivalent to the first gap equation if and only if

$$\frac{\alpha}{\sqrt{\xi_{k'}^2 + \Delta^2(\mathbf{k}')}} = \frac{\beta}{\sqrt{\xi_{k'}^2 + f^2(\alpha, \beta)\Delta^2(\mathbf{k}')}},$$
 (A4)

which is easily shown to imply that  $\frac{\xi_{k'}}{\Delta(\mathbf{k}')}$  is a constant, which is physically unreasonable, thus completing the proof. We note that in the special case where  $V_{\mathbf{k},\mathbf{k}'}$  is a separable potential, i.e.,  $V(k) \propto g(k)g(k')$ , the shape of the gap function is of course unaffected by scale changes.

- <sup>1</sup>D. A. Brawner and H. R. Ott, Phys. Rev. B **50**, 6530 (1994); **53**, 8249 (1996); A. Mathai, Y. Gim, R. C. Black, A. Amar, and F. C. Wellstood, Phys. Rev. Lett. **74**, 4523 (1995); C. C. Tsuei, J. R. Kirtley, C. C. Chi, Lock See Yu-Jahnes, A. Gupta, T. Shaw, J. Z. Sun, and M. B. Ketchen, *ibid.* **73**, 593 (1994).
- <sup>2</sup>C. C. Tsuei, J. R. Kirtley, M. Rupp, J. Z. Sun, A. Gupta, M. B. Ketchen, C. A. Wang, Z. F. Ren, J. H. Wang, and M. Bhushan, Science **271**, 329 (1996); J. R. Kirtley, C. C. Tsuei, J. Z. Sun, C. C. Chi, Lock See Yu-Jahnes, A. Gupta, M. Rupp, and B. Ketchen, Nature (London) **373**, 225 (1995); W. N. Hardy, D. A. Bonn, D. C. Morgan, Ruixing Liang, and Kuan Zhang, Phys. Rev. Lett. **70**, 3999 (1993); W. N. Hardy, S. Kamal, D. A. Bonn, K. Zhang, R. Liang, E. Klein, D. C. Morgan, and D. J. Baar, Physica B **197**, 609 (1994).
- <sup>3</sup>K. A. Moler, D. J. Baar, J. S. Urbach, Ruixing Liang, W. N. Hardy, and A. Kapitulnik, Phys. Rev. Lett. **73**, 2744 (1994); C. Renner and O. Fischer, Phys. Rev. B **51**, 9208 (1995); M. Oda, C. Manabe, and M. Ido, *ibid.* **53**, 2253 (1996); T. E. Mason, G. Aeppli, and H. A. Mook, Phys. Rev. Lett. **68**, 1414 (1992); T. E. Mason, G. Aeppli, S. M. Hayden, A. P. Ramirez, and H. A. Mook, *ibid.* **71**, 919 (1993).
- <sup>4</sup>J. Y. T. Wei, N.-C. Yeh, D. F. Garrigus, and M. Strasik, Phys. Rev. Lett. **81**, 2542 (1998); G. Deutscher, Rev. Mod. Phys. **77**, 109 (2005).
- <sup>5</sup>G. Blumberg, A. Koitzsch, A. Gozar, B. S. Dennis, C. A. Kendziora, P. Fournier, and R. L. Greene, Phys. Rev. Lett. **88**,

107002 (2002).

- <sup>6</sup>H. Matsui, K. Terashima, T. Sato, T. Takahashi, M. Fujita, and K. Yamada, Phys. Rev. Lett. **95**, 017003 (2005).
- <sup>7</sup>J. Mesot, M. R. Norman, H. Ding, M. Randeria, J. C. Campuzano, A. Paramekanti, H. M. Fretwell, A. Kaminski, T. Takeuchi, T. Yokoya, T. Sato, T. Takahashi, T. Mochiku, and K. Kadowaki, Phys. Rev. Lett. **83**, 840 (1999).
- <sup>8</sup>S. V. Borisenko, A. A. Kordyuk, T. K. Kim, S. Legner, K. A. Nenkov, M. Knupfer, M. S. Golden, J. Fink, H. Berger, and R. Follath, Phys. Rev. B **66**, 140509(R) (2002).
- <sup>9</sup>K. Terashima, H. Matsui, T. Sato, T. Takahashi, M. Kofu, and K. Hirota, Phys. Rev. Lett. **99**, 017003 (2007).
- <sup>10</sup>M. Le Tacon, A. Sacuto, A. Georges, G. Kotliar, Y. Gallais, D. Colson, and A. Forget, Nat. Phys. 2, 537 (2006).
- <sup>11</sup>K. Nakayama, T. Sato, K. Terashima, H. Matsui, T. Takahashi, M. Kubota, K. Ono, T. Nishizaki, Y. Takahashi, and N. Kobayashi, Phys. Rev. B **75**, 014513 (2007).
- <sup>12</sup> T. Yoshida, M. Hashimoto, K. Tanaka, A. Fujimori, Z.-X. Shen, Z. Hussain, M. Kubota, K. Ono, S. Komiya, Y. Ando, H. Eisaki, and S. Uchida, Data Presented at 2007 Spectroscopy of Novel Superconductors Conference, Sendai, Japan (unpublished).
- <sup>13</sup>Y. Dagan, R. Beck, and R. L. Greene, Phys. Rev. Lett. **99**, 147004 (2007).
- <sup>14</sup> P. Monthoux, A. V. Balatsky, and D. Pines, Phys. Rev. Lett. 67, 3448 (1991); Phys. Rev. B 46, 14803 (1992); P. Monthoux and D. Pines, Phys. Rev. Lett. 69, 961 (1992).

- <sup>15</sup>D. Parker, S. Haas, and A. V. Balatsky, Phys. Rev. B 76, 104503 (2007).
- <sup>16</sup> A. D. Palczewski, T. Kondo, R. Khasanov, N. N. Kolesnikov, A. V. Timonina, E. Rotenberg, T. Ohta, A. Bendounan, Y. Sassa, A. Fedorov, S. Pailhés, A. F. Santander-Syro, J. Chang, M. Shi, J. Mesot, H. M. Fretwell, and A. Kaminski, Phys. Rev. B **78**, 054523 (2008).
- <sup>17</sup>D. Manske, I. Eremin, and K. H. Bennemann, Phys. Rev. B 67, 134520 (2003).
- <sup>18</sup>M. Fujita, M. Matsuda, S.-H. Lee, M. Nakagawa, and K. Yamada, Phys. Rev. Lett. **101**, 107003 (2008).
- <sup>19</sup>Y. Bang, I. Martin, and A. V. Balatsky, Phys. Rev. B 66, 224501 (2002).
- <sup>20</sup>H. F. Fong, P. Bourges, Y. Sidis, L. P. Regnault, A. Ivanov, G. D. Gu, N. Koshizuka, and B. Keimer, Nature (London) **398**, 588 (1999).

- <sup>21</sup>V. A. Khodel, V. M. Yakovenko, M. V. Zverev, and H. Kang, Phys. Rev. B **69**, 144501 (2004).
- <sup>22</sup>J. G. Storey, J. L. Tallon, and G. V. M. Williams, Phys. Rev. B 76, 174522 (2007).
- <sup>23</sup>A. Kanigel, M. R. Norman, M. Randeria, U. Chatterjee, S. Souma, A. Kaminski, H. M. Fretwell, S. Rosenkranz, M. Shi, T. Sato, T. Takahashi, Z. Z. Li, H. Raffy, K. Kadowaki, D. Hinks, L. Ozyuzer, and J. C. Campuzano, Nat. Phys. **2**, 447 (2006).
- <sup>24</sup>J. G. Storey, J. L. Tallon, G. V. M. Williams, and J. W. Loram, Phys. Rev. B **76**, 060502(R) (2007).
- <sup>25</sup>A. V. Chubukov, M. R. Norman, A. J. Millis, and E. Abrahams, Phys. Rev. B **76**, 180501(R) (2007).
- <sup>26</sup> M. R. Norman, A. Kanigel, M. Randeria, U. Chatterjee, and J. C. Campuzano, Phys. Rev. B **76**, 174501 (2007).
- <sup>27</sup>S. Graser, P. J. Hirschfeld, and D. J. Scalapino, Phys. Rev. B 77, 184504 (2008).