

Absence of pseudogap in heavily overdoped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ from tunneling spectroscopy of break junctions

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Abstract. – We report tunneling spectroscopy of superconductor-insulator-superconductor break junctions on heavily overdoped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ with $T_c = 56$ K. At $T \ll T_c$, the junction conductances display well-defined quasiparticle peaks at $\pm 2\Delta$ and in some cases a Josephson current at zero bias. Gap values as small as $\Delta = 10.5$ meV have been observed leading to $2\Delta/kT_c$ near the BCS limit for $d_{x^2-y^2}$ pairing. Temperature dependence of the gap magnitude, $\Delta(T)$, follows the BCS relation and both the quasiparticle gap and Josephson current vanish for $T > T_c$. Above T_c , the tunneling conductance shows a flat background without any indication of a pseudogap near the Fermi level.

High-temperature superconductors (HTSs) have emerged with properties that are very different from those found in conventional superconductors. One such property of the underdoped phase is the presence of a pseudogap observed well above T_c , up to a characteristic temperature T^* , that has been detected by a number of experimental techniques, such as in-plane resistivity, angle-resolved photoemission spectroscopy (ARPES), specific heat, and NMR [1, 2]. One reason why the presence of the pseudogap garners a lot of attention is that it might be related to the phenomenon of superconductivity in HTSs. Numerous theories have been proposed to explain the origin of the pseudogap in the underdoped phase, such as pairing fluctuation theory, spin-charge separation scenario, phase fluctuations [3–6].

While it is generally accepted that the pseudogap is a property of the underdoped phase, it is still debatable whether it is also present in the overdoped region. Whereas T^* can be determined in various experiments (*e.g.*, a change in slope of in-plane resistivity *vs.* T) a corresponding direct measurement of a pseudogap in the density of states is not always available. For overdoped HTSs, none of the experimental techniques listed earlier have detected a

clear pseudogap [1]. Some results from scanning tunneling microscopy (STM) [7, 8] and planar tunnel [9] junctions have suggested that the pseudogap exists at all hole doping concentrations in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ (Bi-2212). In contrast, another STM study in the ab -plane of Bi-2212 does not show any gap-like feature in the overdoped phase [10]. These conflicting observations hinder our understanding of the pseudogap and demand further tunneling investigations of the overdoped phase of Bi-2212. One reason for the conflict might be that the doping level of Bi-2212 in the region of the junction is not the same as in the bulk. Miyakawa *et al.* [11] have demonstrated a clear relationship between the energy gap (Δ) and hole concentration and therefore the measured gap provides important information on the local doping level. Here we report measurements of heavily overdoped crystals ($T_c = 56$ K) that exhibit a Δ value as low as 10.5 meV, much smaller than the optimal doped value of 38 meV and clearly in the overdoped state.

The break junction method can be used to measure the superconductor-insulator-superconductor (SIS) tunneling conductance in conventional and HTSs. One advantage of this method is that the junction can be fabricated at low temperatures so that it is unexposed to air. The SIS tunneling spectroscopy is not only capable of measuring quasiparticle excitations but also the Cooper pairs in the form of the Josephson current, I_c , which is purely a superconducting phenomenon. This capability provides an important advantage since $I_c(T) \rightarrow 0$ defines the junction T_c . In other words, the phase coherence temperature can be identified by measuring the temperature dependence of the critical current.

Single crystals of Bi-2212 were grown by a self-flux technique in a strong thermal gradient to stabilize the direction of solidification. Overdoping was accomplished using stainless-steel cells sealed with the sample immersed in liquid oxygen [12]. The critical temperature of the samples was determined from the magnetization which showed sharp transitions. Tunneling measurements were done with a point contact apparatus [13]. The Bi-2212 crystal is cleaved along the ab -plane and mounted on a substrate so that the tip approaches along the c -axis. A novel method is used to form the SIS break junctions. A differential micrometer-driven Au tip approaches the sample at LHe temperature and superconductor-insulator-normal-metal (SIN) junctions are first formed. These conductances exhibit quasiparticle peaks at $eV \sim \pm\Delta$ and clearly show the standard dip and hump features above Δ in the occupied part of the DOS [7, 11]. Further increasing the force of the tip leads to an Ohmic contact (less than 10 ohm) with the crystal and a mechanical bond. A Bi-2212 piece is easily dislodged by cleaving along the double Bi-O layer. As a result of this process, an SIS junction forms between the Bi-2212 piece and the rest of the crystal. The SIS conductances show quasiparticle peaks at $\pm 2\Delta$ that are consistent with the Δ value obtained from the SIN junctions. Additionally, the SIS conductances also show a symmetric dip, hump and background, and also the presence of a Josephson current. The magnitude of this current generally decreases as the junction resistance increases.

Figure 1(a) shows the temperature dependence of the tunneling conductance for junction # 1 on the heavily overdoped Bi-2212. The 4.2 K data show a tunneling conductance of a Bi-2212 break junction with sharp quasiparticle peaks at $\pm 2\Delta$, a weak Josephson current peak at zero bias, and the dip and hump features above $\pm 2\Delta$. One of the distinct features of this junction is the relatively flat background. This is different than the typical SIS conductances found from optimally doped [14] and overdoped [15] Bi-2212 that show a decreasing background up to ± 350 meV. For SIS junctions at low temperatures, the thermal smearing is minimal and the quasiparticle peaks can be used to estimate the energy gap size, which is 14 meV for the 4.2 K spectra in fig. 1(a), giving a coupling ratio $2\Delta/kT_c \sim 5.8$. A set of temperature-dependent data for another SIS break junction (junction # 2) of Bi-2212 is shown in fig. 1(b). Although the junction resistance in fig. 1(b) is approximately the same

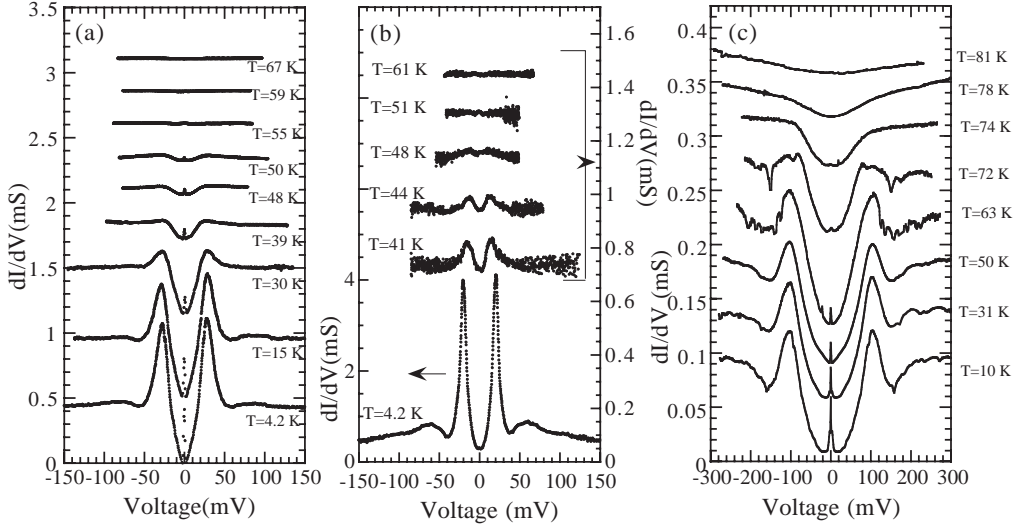


Fig. 1 – The temperature evolution of the tunneling conductance for overdoped Bi-2212 with bulk $T_c = 56$ K, (a) junction # 1 and (b) junction # 2; (c) underdoped Bi-2212 with bulk $T_c = 77$ K [11]. The tunneling conductances (except for 4.2 K) are shifted upward for clarity. Note that the scale at higher temperatures is different for (b).

as in fig. 1(a), we do not observe the Josephson current. This discrepancy may be due to a slightly different junction orientation or possibly due to the smaller energy gap which is estimated from the 4.2 K data to be 10.5 meV. To the best of our knowledge fig. 1(b) shows the smallest gap ever found in the Bi-2212 system. The energy gap magnitude found from tunneling is consistent with that observed in recent ARPES measurements around $(\pi, 0)$ on similar crystals [16]. Using the bulk T_c , the coupling ratio $2\Delta/kT_c$ is 4.35, which is very close to the BCS mean field $d_{x^2-y^2}$ -wave (d -wave) prediction of 4.28. The decrease in gap size and T_c with increasing hole doping in the overdoped phase is consistent with recent experiments [11, 14, 17] and the phase diagram of HTSs.

As the temperature increases towards T_c , the tunneling conductances of fig. 1(a) and fig. 1(b) show several notable changes. Most importantly, all traces of a superconducting gap, or any other type of gap, have disappeared for temperatures above the bulk T_c . Additionally, the magnitude of the Josephson current peak in fig. 1(a) diminishes until it vanishes around T_c . This is further evidence that the junction T_c is the same as the bulk value. Both figures show no presence of any depression above T_c such as is observed in underdoped Bi-2212 [7, 11]. Note the increased sensitivity of the vertical scale in fig. 1(b) for the higher-temperature data. For comparison, we show some previously published data on underdoped Bi-2212 [11] in fig. 1(c). The superconducting gap is washed out around the bulk $T_c = 77$ K of the crystal but a weak depression in the conductance remains which is consistent with a pseudogap. The absence of any pseudogap in our present measurements is consistent with recent ARPES studies [16] on the same type of samples. In that study, the midpoint of the spectral weight peak shifted to the Fermi level at T_c , showing no evidence of the pseudogap that is seen in optimally doped or underdoped Bi-2212 [1]. Thus, there are now two measurements which show that the pseudogap disappears in heavily overdoped Bi-2212.

Considering other studies, STM measurements of overdoped Bi-2212 with $T_c \sim 74$ K have

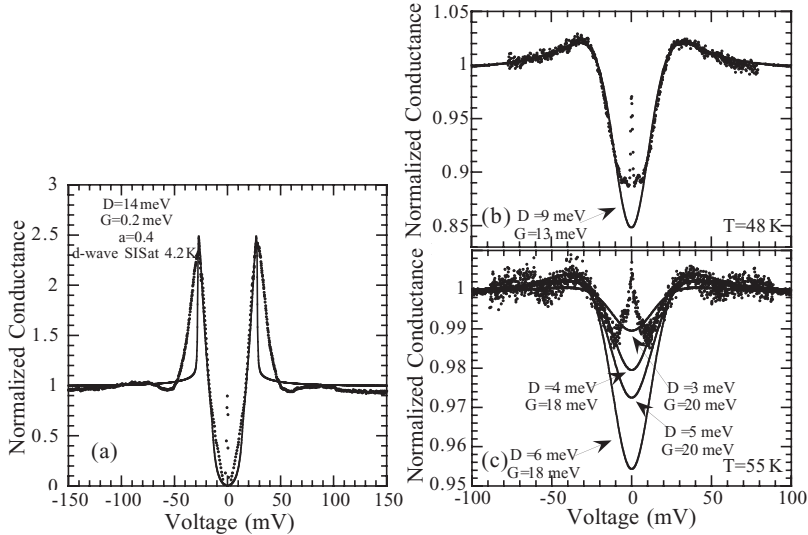


Fig. 2 – (a) Normalized conductance of SIS junction (dots), which is given in fig. 1(a). The data corresponds to 4.2 K and normalized by a constant. The solid line is a SIS-generated d -wave fit which includes the smearing factor Γ and a directionality function $f(\theta)$. (b) and (c) Normalized conductance of SIS junctions (dots) for 48 K and 55 K, which is given in fig. 1(a). The solid line in (b) is SIS-generated pure d -wave fit which only includes the smearing factor Γ . The solid lines in (c) show different attempts to fit the data.

reported the observation of a pseudogap above T_c [7]. Since STM is a local probe, there is a possibility that it does not reflect the bulk T_c of the sample and therefore it is important to consider the energy gap magnitude. Both refs. [7] and [8] present energy gaps in the range 35–40 meV in addition to the pseudogap behavior. This gap magnitude is consistent with optimally doped or slightly overdoped samples and thus the doping level at the junction is not well established. In a more recent study, SIS break junctions of underdoped and overdoped Bi-2212 with $T_c = 82$ K were fabricated using an STM by M. Oda *et al.* [18]. Both sets of data exhibited a pseudogap above T_c ; however, the overdoped tunneling conductance showed a smaller gap which fills in at a much lower temperature than underdoped and thus T^* is closer to T_c . Taken together the tunneling studies have established that the pseudogap persists with overdoping down to $T_c = 82$ K, but is absent when $T_c = 56$ K.

A natural question might be whether there is any change in the pairing symmetry associated with the loss of a pseudogap. One may assume that increasing hole doping makes the Bi-2212 normal-state more Fermi-liquid-like, leading possibly to an s -wave BCS superconductor. There are ARPES observations that suggest s -wave BCS behavior in the heavily overdoped phase [19]. However, the tunneling subgap conductance shapes at low temperatures in figs. 1(a) and (b) are not consistent with the s -wave BCS theory prediction which is supposed to be flat near zero bias. Here what we observed is that the d -wave-like subgap structure persists at the $T_c = 56$ K overdoped Bi-2212. Figure 2 shows the normalized tunneling conductance of SIS break junction # 1 at 4.2 K (dots) which is given in fig. 1(a). Since the tunneling conductance is flat above T_c , the data have been normalized by a constant. The fit is generated using a d -wave gap function, $\Delta = \Delta_0 \cos(2\theta)$, in the BCS DOS, $N_s(E, \theta) = (E - i\Gamma)/\sqrt{(E - i\Gamma)^2 - \Delta(\theta)^2}$. Here, Γ is a smearing parameter to account for

quasiparticle lifetime. The SIS conductance is calculated by

$$\frac{dI}{dV} = c \frac{d}{dV} \iint f(\theta) N_s(E, \theta) N_s(E + eV, \theta) [F(E) - F(E + eV)] dE d\theta,$$

where c is a proportionality constant. $F(E)$ is the Fermi function. The formula also includes a directionality function $f(\theta) = 1 + \alpha \cos(4\theta)$ which corresponds to a preferred tunneling along the d -wave lobes. Here α is the directionality strength, which is assumed temperature independent ($\alpha = 0.4$) [15]. The fit (solid line in fig. 2(a)) shows good agreement in the subgap region with the experimental data. Thus there appears to be no change of symmetry away from d -wave. However for $|eV| > 2\Delta$ the data deviate from the BCS d -wave fit, exhibiting broader peak widths and the dip feature. In general, these features are compensating which allows conservation of states; however, the data in fig. 2(a) lead to an integrated area out to 150 meV which is about 6% above the BCS d -wave fit. We attribute the discrepancy to the inability to accurately know the background conductance at 4.2 K. The broad peaks and dip feature are observed throughout the doping range and have been attributed to strong-coupling effects [20].

The weak temperature dependence of quasiparticle peak position has been observed in the bare STM tunneling conductance on underdoped, optimally doped and overdoped Bi-2212 [7]. Analysis of the underdoped data has been carried out by Franz and Millis [21] using the classical phase fluctuation model. What they find in their model fit to the tunneling conductance of underdoped Bi-2212 data is that the gap is decreasing until $3/4$ of T_c , and sharply increases near T_c . This unusual effect seems to be tied to the presence of a pseudogap. In the present study, we show that the gap magnitude for heavily overdoped Bi-2212 shows no anomalies near T_c but continues to decrease and appears to close near the bulk T_c value. Figures 2(b) and (c) shows the d -wave fit for $T_c = 48$ K and 55 K. The tunneling conductance in the fig. 2(b) displays peak values near 30 meV, however, the best fit for fig. 2(b) corresponds to $\Delta = 9$ meV and $\Gamma = 13$ meV, so the energy gap is smaller than obtained value at 4.2 K. For 55 K, we plot fits using different Δ and Γ values in fig. 2(c) to prove that the gap is very small around T_c . The data are in good agreement with a fit using $\Delta = 4$ meV and $\Gamma = 18$ meV. These results show the energy gap magnitude reduces with increasing temperature as indicated in fig. 3 and appears to close at the bulk T_c .

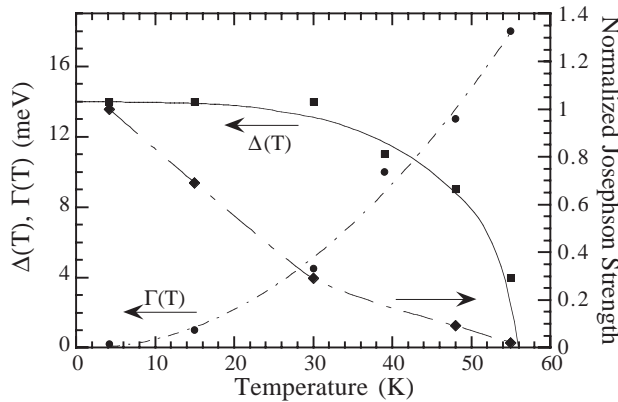


Fig. 3 – Graph obtained from analyzing fig. 1(a) temperature dependence of superconducting gap $\Delta(T)$ (squares), quasiparticle scattering rate $\Gamma(T)$ (circles), and Josephson strength, $I_c R_n$ (diamonds) normalized by $I_c R_n$ (4.2 K). Here, the Josephson current I_c is estimated from the peak in conductance at zero bias. The full curve represents the BCS superconducting gap $\Delta(T)$.

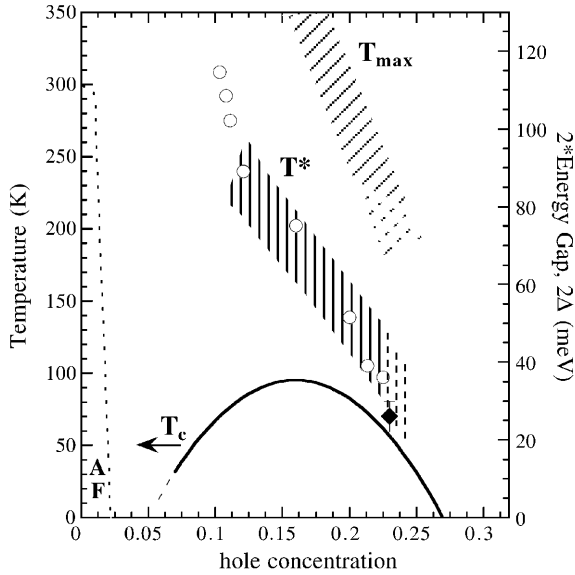


Fig. 4 – The temperature and 2Δ vs. hole concentration for Bi-2212. The dashed area is the measured T^* by different experimental techniques (see ref. [18]). The circles are our data that were published before [11]. The diamond is the present work.

Figure 3 presents the various parameters derived from fig. 1(a). The solid line corresponds to the temperature dependence of s -wave BCS superconductor energy gap which is similar to the d -wave BCS [22] behavior. Filled squares are energy gap magnitude, filled circles are Γ . The T -dependence of the gap magnitude follows the BCS prediction, although the increased smearing due to the scattering rate Γ prevents exact determination of the gap magnitude around T_c .

As discussed before, one of the advantages of SIS tunnel junctions is the possible Josephson current which can be used to determine T_c of the superconductor. Even if there is a pairing above T_c , such as preformed pairs [5], any Josephson current that might persist above T_c , would be extremely weak. Therefore, we use the tunneling conductance peak (a more sensitive measurement) to obtain the Josephson strength $I_c R_n$. In fig. 3, diamonds correspond to normalized Josephson strength which vanishes around bulk T_c of the crystal, the same temperature where the quasiparticle gap vanished. These two measurements point to the absence of superconducting pairing and Cooper pairs above the bulk T_c of heavily overdoped Bi-2212.

We now combine in fig. 4 these results along with our previously published data and the phase diagram for Bi-2212 as suggested in ref. [18]. Figure 4 shows that our energy gap values lie within the experimentally determined region of T^* over a wide doping range. Here T^* is obtained from various experimental techniques including SIS break junctions using STM [18]. Extrapolating T^* to the overdoped phase suggests that T_c and T^* are very close and might even merge in the overdoped region. This might be the reason that we do not observe a pseudogap in our tunneling studies. The implication of the doping dependence of superconducting energy gap and T^* is that they are intimately related. The simplest notion is that T^* represents the mean-field temperature for pairing, whereas T_c defines when long-range phase coherence is established. However, if the two temperatures were distinct over the entire doping range, then that would suggest that T^* corresponds to physics distinct from

superconductivity. Our present result suggests that there is a particular doping value where the two temperature scales merge and this provides further support for pseudogap models based on superconducting fluctuations [4, 7].

In summary, we have performed break junction tunneling spectroscopy on heavily overdoped Bi-2212. The tunneling conductance displays a flat background above T_c without any indication of pseudogap near the Fermi level. In addition, the energy gap magnitude reaches a value as low as $2\Delta/kT_c = 4.35$, very close to the BCS value for d -wave superconductors. The quasiparticle gap appears to close at the same temperature that the Josephson current disappears. Taken together, the results suggest a merging of the temperature scales for pairing and phase coherence. This occurs at a region of hole concentration where the strong-coupling ratio approaches the BCS limit. This gives further support for the ideas that the pseudogap associated with T^* is due to some type of precursor superconductivity.

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REFERENCES

- [1] DING H. *et al.*, *Nature (London)*, **382** (1996) 51; LOESER A. G. *et al.*, *Science*, **273** (1996) 325.
- [2] LORAM J. *et al.*, *Phys. Rev. Lett.*, **71** (1993) 1740; WARREN W. W. *et al.*, *Phys. Rev. Lett.*, **62** (1989) 1193.
- [3] CHEN Q., KOSZTIN I., JANKO B. and LEVIN K., *Phys. Rev. Lett.*, **81** (1998) 4708.
- [4] LEE P. A and WEN X.-G., *Phys. Rev. Lett.*, **78** (1997) 4111.
- [5] EMERY V. and KIVELSON S. A., *Nature (London)*, **374** (1995) 434.
- [6] TIMUSK T. and STATT B., *Rep. Prog. Phys.*, **62** (1999) 61.
- [7] RENNER CH., REVAZ B., GENOUD J.-Y., KADOWAKI K. and FISHER O., *Phys. Rev. Lett.*, **80** (1998) 149.
- [8] MATSUDA A., SUGITA S. and WATANABE T., *Phys. Rev. B*, **60** (1999) 1377.
- [9] TAO H. J., LU F. and WOLF E. L., *Physica C*, **282-287** (1997) 1507.
- [10] GUPTA, A. K. and NG K.-W., *Phys. Rev. B*, **58** (1998) R8901.
- [11] MIYAKAWA N., ZASADZINSKI J. F., OZYUZER L., GUPTASARMA P., HINKS D. G., KENDZIORA C. and GRAY K. E., *Phys. Rev. Lett.*, **83** (1999) 1018.
- [12] KENDZIORA C., KELLEY R. J., SKELTON E. and ONELLION M., *Physica C*, **257** (1996) 74.
- [13] OZYUZER L., ZASADZINSKI J. F. and GRAY K. E., *Cryogenics*, **38** (1998) 911.
- [14] DEWILDE Y., MIYAKAWA N., GUPTASARMA P., IAVARONE M., OZYUZER L., ZASADZINSKI J. F., ROMANO P., HINKS D. G., KENDZIORA C., CRABTREE G. W. and GRAY K. E., *Phys. Rev. Lett.*, **80** (1998) 153.
- [15] OZYUZER L., ZASADZINSKI J. F., KENDZIORA C. and GRAY K. E., *Phys. Rev. B*, **61** (2000) 3629.
- [16] YUSOF Z., WELLS B. O., VALLA T., FEDOROV A. V., JOHNSON P. D., LI Q., KENDZIORA C., JIAN S. and HINKS D. G., cond-mat/0104367.
- [17] RENNER C., REVAZ B., GENOUD J.-Y. and FISHER O., *J. Low Temp. Phys.*, **105** (1996) 1083.
- [18] ODA M., DIPASUPIL R. M., MOMONO N. and IDO M., submitted to *Phys. Rev. Lett.*
- [19] KELLEY R. J., QUITMANN C., ONELLION M., BERGER H., ALMERAS P., and MARGARITONDO G., *Science*, **271** (1996) 1255.
- [20] ZASADZINSKI J. F., OZYUZER L., MIYAKAWA N., GRAY K. E., HINKS D. G. and KENDZIORA C., *Phys. Rev. Lett.*, **87** (2001) 067005.
- [21] FRANZ M. and MILLIS A. J., *Phys. Rev. B*, **58** (1998) 14572.
- [22] WON H. and MAKI K., *Phys. Rev. B*, **49** (1994) 1397.