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Josephson effect in high-T_c superconductivity*†

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Abstract. In this report we present the most important results of our recent analysis (Kupriyanov and Likharev 1990) of the Josephson effect in both the natural (intergrain) and artificial junctions using high-T_c superconductors (HTS). A comparison of the experimental data with the BCS-based theories of the Josephson effect in various tunnel-junction-type and weak-link-type structures has been carried out. The main conclusion is that the data presently available do not enable one to either confirm or reject the theories, and thus to reveal possible deviations of the real microscopic mechanism of the high-T_c superconductivity from the BCS mechanism. We suggest several experiments which would be more fruitful for this purpose, as well as for finding ways of reproducible fabrication of practically useful Josephson junctions.

Keywords. Josephson effect; intergrain; artificial junction.

1. Introduction

The Josephson effect was reliably observed (Tsai *et al* 1987a) in the high-T_c superconductors shortly after the discovery of these new materials. The observations included all classical features of the effect, including the dc supercurrent within some range $-I_c < I < +I_c$, Josephson-Shapiro current steps at quantized voltages

$$V_n = n(\hbar\omega/2e), n = \pm 1, \pm 2, \ldots, \tag{1}$$

arising at microwave irradiation, and flux quantization of the magnetic flux in superconducting loops closed by the junctions, with the usual period $\Phi_0 = h/2e$. Moreover, periodic oscillation of the current step heights as functions of the microwave power, observed in most junctions, testifies to a single-valued and quasi-sinusoidal relationship between the supercurrent I_s and the Josephson phase difference φ. All these observations imply that the high-T_c superconductivity is due to the usual singlet-state Cooper pairs.

In order to help distinguishing specific pairing mechanisms (see, the review of Chakraverty *et al* 1989), it would be desirable to carry out a more quantitative comparison of the data with predictions of at least the existing theories of the Josephson effect based on the standard BCS model. What follows is a brief description of our attempt (Kupriyanov and Likharev 1990) to carry out such a comparison. The reader will see that the result of the analysis are somewhat inconclusive, the main reason being a complex and irreproducible structure of most high-T_c Josephson junctions studied up to now.

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2. High-T_c superconductor surfaces and interfaces

2.1 *Surfaces*

Complex surface layer is typical for all high-T_c superconducting materials, although its structure is highly dependent on the particular way of their synthesis. In *ex-situ* technologies (involving post-synthesis annealing) even a short exposure of the sample to air before the annealing leads to formation of a relatively thick (nearly $3 \div 5$ nm) dielectric layers (Thomas and Labib 1987; Kumar *et al* 1988; van Veen *et al* 1988; Nefedov *et al* 1989) of BaO and $BaCO_3$. Using *in-situ* technologies this effect can be avoided using high rates of the annealing temperature increase (Talvacchio 1989; Talvacchio *et al* 1989) (> 10°C/s). Irrespective of the technology, the pure YBaCuO surface is, however, metastable, and gradually loses oxygen via diffusion at any temperature above (List *et al* 1988) ~ 20 K.

2.2 *HTS/metal interfaces*

Practically all metals react with the high-T_c oxides forming semiconductor layers with a thickness typically within the range $2 \div 5$ nm at the HTS/metal interface (Meyer *et al* 1988). Transparency of such a layer for conduction electrons is very low (boundary resistance R_s in the range (Takeuchi *et al* 1987; Ekin *et al* 1988a, b; Suzuki *et al* 1988; Talvacchio 1989) from 10^{-2} to 10^{-4} ohm-cm^2) and prevents any Josephson coupling (see e.g. Blamire *et al* 1987; Iguchi *et al* 1987; Katon *et al* 1987a, b; Naito *et al* 1987; Fornel *et al* 1988; Gijs *et al* 1988).

Notable exceptions are gold and silver which do not form oxides reacting with the HTS material components (Gao *et al* 1988; Laubschat *et al* 1988; Oshima *et al* 1988; Wagener *et al* 1988; Meyer *et al* 1989; Weschke *et al* 1989). As a result, the specific resistance of the HTS/Au and HTS/Ag boundaries can be reduced to $\sim 10^{-10}$ Ohm-cm^{-2} using *in-situ* technologies (Gavaler *et al* 1988).

2.3 *HTS/ dielectric interfaces*

To our knowledge, virtually all dielectric materials react chemically with the high-T_c superconductors, mainly producing barium salt layers at the interfaces (Williams and Chaudhury 1988). Even the most promising substrate materials like $SrTiO_3$, MgO, ZrO_2 do form such interlayers at temperatures (Cima *et al* 1988; Koinuma *et al* 1988; Nakajima *et al* 1988; Cheung and Ruckenstein *et al* 1989; Ren *et al* 1989) above ~ 700°C.

3. HTS/LTS Josephson junction

The above facts enable one to explain an extreme irreproducibility of the basic parameters (the critical current I_c and normal resistance R_N) of the point-contact HTS/LTS junctions which were extensively studied at the first stage of the HTS research (Kita *et al* 1987; Kuznik *et al* 1987; Mc Grath *et al* 1987; Tsai *et al* 1987a; Yamashita *et al*

1987; Yang 1987; Andersen *et al* 1988; Barone *et al* 1988; Eidelloth *et al* 1988; Imai *et al* 1988, 1989; Kuznik *et al* 1988; Nishino *et al* 1988; Shiping *et al* 1988; Nakayama and Okabe 1989). Moreover, the product $V_c = I_c R_N$ (which is much more stable than I_c in traditional LTS/LTS junctions) varies within a broad range ($\sim 0{\cdot}03 \div 1{\cdot}0$ mV).

It is remarkable that this range falls well below the value

$$V_{CO} \approx \frac{\Delta'(T)}{e} ln\{4\Delta(T)/\Delta'(T)\} \approx 5\,\mathrm{mV} \tag{2}$$

allowed by the BCS-based theories for the "perfect" Josephson junctions (Ambegaokar and Baratoff 1963; Kulik and Omel'yanchuk 1975, 1978) with $T_c \approx 100$ K and $T_c' \approx 10$ K. Several attempts to fabricate more well-defined SIS' (tunnel) junctions (Barone *et al* 1988; Camerlingo *et al* 1988; Inone *et al* 1988; Nakayama *et al* 1988, 1989; Tsai *et al* 1989) and SNS' junctions with gold interlayers (Akoh *et al* 1988, 1989) did not yield larger $V_c (0{\cdot}6\,\mu\mathrm{V} \leqslant V_c \leqslant 0{\cdot}3\,\mathrm{mV}$ at $T = 4{\cdot}2$ K).

Another common feature of all studied HTS/LTS junctions is a considerable ($I_{ex} \approx I_c$) excess current I_{ex} defined as

$$I_{ex} = I(V) - V/R_N|_{I \gg I_c}. \tag{3}$$

This feature is typical for weak-link-type structures with their metallic conductivity (Likharev 1979).

Unfortunately most experiments with HTS/LTS junctions were oriented to mere a demonstration of the Josephson effect. The data vital for a more quantitative discussion (including temperature dependencies of I_c, I_{ex} and R_N, as well as magnetic-field dependence of I_c) were not recorded (or just not published).

4. HTS/HTS Josephson junctions

The last remark is valid as well for most experiments with the HTS/HTS junctions, despite the fact that a larger variety of the junction types was studied.

4.1 *Point-contact junctions*

These traditional junctions are typically formed after the surfaces brought in contact had been exposed to air, so that relatively thick dielectric layers had been formed on them. Thus the contacts reveal critical currents (de Waele *et al* 1988; Niemeyer *et al* 1987; Ryhanen and Seppa 1989; Nakane *et al* 1987; Komatzu *et al* 1987; Olsson *et al* 1987) (typically very low ones) only at high pressure. Their V_c's are rather low ($\leqslant 1$ mV), i.e. much lower than the maximum BCS value

$$V_{CO} \approx \Delta(T)/e \approx 30\,\mathrm{mV}. \tag{4}$$

4.2 *Bulk junctions*

Josephson junctions are naturally formed between the grains of the high-T_c ceramics (Hatano *et al* 1989; Cui *et al* 1987; Sugishita *et al* 1987; Wu *et al* 1987; Higashino *et al* 1987; Shen *et al* 1989; Changxin *et al* 1987; Yang *et al* 1989; Shablo *et al* 1988; Li *et al* 1988; Robbes *et al* 1989; Song *et al* 1989; Kataria *et al* 1988; Akimov *et al* 1989; Hauser *et al* 1987; Nakane *et al* 1987; Golovashkin *et al* 1989; Ono *et al* 1989; Gergis *et al* 1988; Higashino *et al* 1989; Wiener-Avnear *et al* 1989; Hilton *et al* 1989; White *et al* 1988; Katon *et al* 1988; Tanabe *et al* 1987; Lin *et al* 1988; Iguchi *et al* 1987; Yuan *et al* 1988; Kita *et al* 1989; Wen *et al* 1989; Wang *et al* 1989; Matsuda *et al* 1989; Yamashita *et al* 1989; Yamashita *et al* 1988; Noge *et al* 1989; Hauser *et al* 1989; Takeuchi *et al* 1988; Chaudhari *et al* 1988; Dimos *et al* 1988; Maunhart *et al* 1988; Koch *et al* 1989; Vedeneev *et al* 1989). In order to single out and study such a junction, one can use a bulk sample with a mechanically-formed constriction with its width W and length L of the order of the grain size a, so that the current is concentrated in a single junction while the other junctions remain in their superconducting state (Yamashita *et al* 1989; Yamashita *et al* 1988; Noge *et al* 1989; Hauser *et al* 1989; Takeuchi *et al* 1988; Chaudhari *et al* 1988; Dimos *et al* 1988; Maunhart *et al* 1988; Koch *et al* 1989; Vedeneev *et al* 1989). (Note that if W and L are much larger than a, many juctions with random parameters are involved to the sample dynamics, and it is virtually hopeless to extract a meaningful information from the data (Hatano *et al* 1989; Cui *et al* 1987; Sugishita *et al* 1987; Wu *et al* 1987; Higashino *et al* 1987; Shen *et al* 1989; Changxin *et al* 1987; Yang *et al* 1989; Shablo *et al* 1988; Li *et al* 1988; Robbes *et al* 1989; Song *et al* 1989; Kataria *et al* 1988; Akimov *et al* 1989; Hauser *et al* 1987; Nakane *et al* 1987; Golovashkin *et al* 1989; Ono *et al* 1989; Gergis *et al* 1988; Higashino *et al* 1989; Wiener-Avnear *et al* 1989; Hilton *et al* 1989; White *et al* 1988; Katon *et al* 1988; Tanabe *et al* 1987; Lin *et al* 1988; Iguchi *et al* 1987; Yuan *et al* 1988; Kita *et al* 1989; Wen *et al* 1989; Wang *et al* 1989; Matsuda *et al* 1989). Intergrain boundaries are typically cleaner than the surface. As a consequence, somewhat higher values of V_c (up to ~ 1 mV at $T = 77$ K and ~ 3 mV at $T = 4{\cdot}2$ K) have been registered (Li *et al* 1988; Gergis *et al* 1988; Wen *et al* 1989).

4.3 *Break junctions*

Another way to form a Josephson junction from a bulk high-T_c superconducting sample is to produce a tiny crack in it (see e.g. Tsai *et al* 1987b; Moreland *et al* 1987a, b). These cracks usually follow the intergrain boundaries, and thus the basic properties of these "break junctions" are close to those of the best bulk junctions (with W, $L \leqslant a$).

A notable exception is the break junctions formed in monocrystallic samples (Aminov *et al* 1989), which exhibit extremely high values of V_c: from $5 \div 10$ mV for YBaCuO to ~ 20 mV for BiSrCaCuO and TlBaCaCuO (at $T = 4{\cdot}2$ K). These values are only slightly less than those given by the BCS equation (4) for "perfect" Josephson junctions.

Nevertheless, these structures are as irreproducible as all point contacts and intergrain junctions.

4.4 *Tunnel junctions*

In order to get something reproducible, several attempts have been made (Shiota *et al* 1989; Kominami *et al* 1989) to form HTS/HTS structures with artificial tunnel barriers; none of those attempts, however, has lead to a non-vanishing critical current.

4.5 *SNS structures*

We are familiar with only two successful attempts to fabricate potentially reproducible HTS/HTS junctions of the SNS type. The SNS-sandwich junctions with 5-nm-thick Ag interlayer (Moreland *et al* 1989) have exhibited V_c of the order of 1 mV, while the SNS microbridges (Schwarts *et al* 1989) with YBaCuO banks and $\sim$ 1-μm-long Au span had much lower V_c ($\sim 3{\cdot}5\,\mu$V at 4·2 K). Unfortunately, no $I_c(T)$ and $I_c(H)$ dependences were reported for these junctions. It makes a reliable identification of the data with theoretical predictions hardly possible.

5. Comparison with theoretical models

Despite suggestions of several new mechanisms of the high-T_c superconductivity (see, e.g. Chakraverty *et al* 1989 for their review), none of them can claim to provide a ready explanation of all peculiarities of the new materials. This is why we have restricted ourselves to comparison of the data available with the BCS-based theories.

5.1 *"Perfect" junction models*

Josephson junctions with sufficiently small spacing of their superconducting electrodes exhibit the "perfect" behaviour with maximum value of V_c and nearly sinusoidal $I_s(\varphi)$ relationship (Likharev 1979). For tunnel junctions, such behaviour is described by the Ambegaokar-Baratoff-Werthamer-Larkin-Ovchinnikov theory (Ambegaokar and Baratoff 1963; Larkin and Ovchinnikov 1966; Werthamer 1966) (AB), while short metallic-conducting weak links obey one of the Kulik-Omeliyanchuk-Artemehko-Volkov-Zaitsev theories (Kulik and Omel'yanchuk 1975, 1978; Artemehko *et al* 1979a, b; Zaitsev 1980, 1984; Zaitsev and Ovsyannikov 1989) (KO-1, KO-2). Absolute values of V_c given by these theories are close to each other (see equation (4)), but vanishing I_{ex} are predicted for the tunnel junctions, while $I_{ex} \approx I_c$ for weak links.

The only experimental results comparable with the perfect Josephson effect theories are those obtained for the monocrystalline break junctions (Aminov *et al* 1989). Nevertheless, some peculiarities of the junctions, including temperature dependencies of R_N and I_{ex} (in particular, negative values of I_{ex} registered for the junctions at $T \ll T_c$) cannot be explained within this framework.

Taking into account the information mentioned in § 2, it is natural that more complex models for both the tunnel junctions and weak links should be used to interpret these (as well as other) observations.

5.2 *More complex weak link models*

The first factor not appreciated by the KO models of a weak link is its nonvanishing length $L \geqslant \xi(T)$. The simplest theory taking this factor into account (Likchrev 1976) enables one to describe the smallness of V_c, provided that the interlayer material is a normal metal. This model, however, does not enable one to describe high values ($R_N \geqslant 10^{-7} \div 10^{-8}\,\Omega\mathrm{cm}^2$) of the normal resistance of most junctions.

A further complication of the models concerns a more realistic description of the proximity effect at the SN boundaries of SNS junctions. Calculations (Ivanov *et al* 1981; Kupriyanov and Lukichev 1982, 1988; Golubov *et al* 1983; Kupriyanov 1989a, b) show that the effect is dependent of two-dimensionless parameters

$$\gamma = \rho_S \xi_S^* / \rho_N \xi_N^* \quad \gamma_B = R_B / \rho_N \xi_N^*, \tag{5}$$

where $\rho_{N,S}$ and $\xi_{N,S}^*$ are the normal-state resistivities of junction materials, and their coherence lengths, while R_B is the specific resistance of the SN boundary. An increase of any of γ, γ_B leads to a decrease of V_c, while the normal resistance of the junction is mainly influenced by γ_B. Independent measurements of the parameters involved in (5) enable one to make the following estimates for the typical YBaCuO/Au(Ag)/YBaCuO structures: $\gamma_B \approx 30 \div 1000, 1 \ll \gamma \ll \gamma_B$.

With these values, the theory would be consistent with the data on experimental SNS structures (Moreland *et al* 1989; Schwarts *et al* 1989), provided that their effective areas were much less than those implied by their physical dimensions. Unfortunately, this fact could be only confirmed by the (missing) $I_c(H)$ dependence, so that no convincing conclusion can be made on this point now.

Concerning the intergrain junctions, the model could also describe the observed low values of V_c, but it implies a much more steep rise of V_c at $T \to 0$ than that observed in experiments (Yamashita *et al* 1988, 1989; Noge *et al* 1989; Hauser *et al* 1989; Takeuchi *et al* 1988; Chaudhari *et al* 1988; Dimos *et al* 1988; Maunhart *et al* 1988; Koch *et al* 1989; Vedeneev *et al* 1989). This discrepancy could be removed by an assumption that a temperature independent pair-breaking takes place in the junctions. Electron scattering on uncompensated spins of Cu^{+2} ions can be one of such mechanisms (Bulaevskii *et al* 1977, 1978).

Thus the experimental data for the intergrain HTS/HTS junctions do allow a semi-quantitative interpretation within some BCS-based weak-link models.

5.3 *More complex tunnel junction models*

The first group of possible factors leading to lower V_c's is related to possible thin normal layers formed near the tunnel barrier (resulting in a SNINS structure (Golubov *et al* 1984; Golubov and Kupriyanov 1989a, b; Aslamazov and Fistul 1982; Fistul and Tartakovskii 1988)). A degree of suppression of V_c is dependent of parameters γ and γ_B defined by (5), and the normal layer thickness. Calculations show, however, that neither combinations of these parameters enable one to describe the low values of V_c together with the slow variation of V_c with temperature at $T \ll T_c$.

Another possible mechanism of the suppression of V_c is the resonant tunnelling of electrons via localized states inside the tunnel barrier of the junction, combined with the thermally-activated hopping via these states (Aslamazov and Fistul 1982; Fistul and Tartakovskii 1988; Larkin and Matveev 1987; Glazman and Matveev 1988, 1989). Our analysis shows that the data got for intergrain Josephson junctions can be fit by this theory as well, at least in a semi-quantitative way.

This duality of the possible interpretations of the data is not so surprising after all: the physics of the electron transfer via localized states of a large concentration is quite similar to that through a very dirty normal metal, so that two classes of theories approach each other qualitatively for such a system (unfortunately, no quantitative link between them has been developed yet).

6. Toward the quantitative understanding of the Josephson effect in high-T_c superconductivity

Even if the existing theories of the Josephson effect are extended to merge inside the just mentioned range of interest, the irreproducibility of point contacts and intergrain junctions would hardly enable one to extract much fundamental information from the theory-vs-experiment comparison. The only hope for such information is promised by special reproducible structures. We can see at least two families of such structures which would allow a gradual increase of our quantitative understanding of the Josephson effect in the high-T_c superconductors, and hence of the superconductivity itself.

6.1 *Weak-link structures*

The first step in this way would be an *in-situ* fabrication and detailed studies of reproducible interface between HTS and either gold or silver. Note that the interfaces with high boundary transparency ($R_B \leqslant 10^{-10}$ ohm cm^2, i.e. $\gamma_B \leqslant 10$) are alone of a real interest here.

The second step would be a fabrication of SNIN junctions using the SN structures studied at the first step. Measurements of the dI/dV as a function V for the structures would enable one to restore the density of states in the normal layer and thus determine parameter γ_B. (Note that already this step would allow one to detect possible deviations from the BCS theory).

At the third step, the external normal metal could be replaced by a classical superconductor (say, aluminum), and V_c of the resulting SNIS' junction could be measured as a function of the normal metal thickness $L \approx \xi_N^*$ and temperature T (accompanying I_c-vs-H measurements and structural studies are also crucial here for a control of the real geometry and homogeneity of the structures). Independent determination of the basic parameters (γ_B, γ and ξ_N^*) could be carried out using SNS' structures, although their reduction from the data is somewhat impeded here by nontrivial properties of the NS' boundary which should be characterized by its own γ_B and γ.

Lastly, one should study the HTS/HTS junctions of the SNS type. The simplest (mechanical) way to form such junctions from bilayer SN structures (Moreland *et al* 1989) can hardly give reproducible results, so that one should find other ways. Presently a consequent expitaxial growth of YBaCuO and PrBaCuO layers seems quite feasible (Soderholm and Goodman 1989; Poppe *et al* 1989) although undoped praseodim compound is a semiconductor rather than a normal metal. The SNS junctions could be very important both from the fundamental point of view (e.g. for a search for possible non-singlet pairing), and for various applications of the Josephson junctions in the superconductor electronics (Likharev 1989) (the BCS-based estimates show that values of V_c up to 1 mV at 77 K are quite feasible).

6.2 *Tunnel junctions*

The tunnel (SIS) junctions seem to be less important for applications (due to their low plasma frequencies) (Likharev 1989), but can give a more direct information on the high-T_c superconductivity (both from dI/dV-vs-V and V_c-vs-T dependences) because parameters of a perfect tunnel barrier determine nothing more than the junction resistance. Such junctions, however, seem more difficult for fabrication than the SNS structures, because here one faces hard problems mentioned in § 2. Presumably, extensite structural and chemical studies will be necessary before a proper material for the tunnel barrier, and a way of the junction fabrication, are found.

7. Conclusion

Despite a somewhat pessimistic view on the experimental results accumulated in this field up to present, we believe that getting a more valuable information is quite feasible in a near future. However, this progress would be impossible without the use of the very modern technologies for fabrication of reproducible thin film structures, and without a careful comparison of the data with results of the advanced theories of the Josephson effect, based on relatively complex models of the junction.

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