PHYSICAL REVIEW B VOLUME 50, NUMBER 13 1 OCTOBER 1994-I

Observation of Josephson self-coupling in $Nb/AlO_x/Nb$ tunnel junctions

M. Maezawa, M. Aoyagi, H. Nakagawa, I. Kurosawa, and S. Takada Electrotechnical Laboratory, 1-1-4 Umezono, Tsukuba, Ibaraki 305, Japan

(Received 31 May 1994)

We present the observation of Josephson self-coupling in $Nb/AlO_x/Nb$ tunnel junctions. In current-voltage characteristics, anomalous subharmonic gap structures were observed; steep current steps and peaks which were sensitive to the magnetic field appeared at the voltages of $2\Delta/3e$ and $2\Delta/5e$. The characteristics of these structures could not be explained by multiparticle tunneling and/or multiple Andreev reflection. We conclude that the structures at $2\Delta/3e$ and $2\Delta/5e$ are due to Josephson self-coupling.

In Josephson tunnel junctions, it is possible that tunneling characteristics are affected by the electromagnetic field due to its own ac Josephson effect. This effect is known as Jo-'sephson self-coupling $(JSC)^{1,2}$ JSC is directly derived from the Werthamer's equation which is based on the tunneling Hamiltonian method and describes the basic behavior of tunnel junctions. ' JSC occurs at voltages determined by $eV + nhv = 2\Delta$, where nhv is the Josephson photon energy and Δ is the superconducting gap energy. This effect should be present in a Josephson tunnel junction with sufficiently low capacitance or high current density.

The main results of JSC are the appearance of current steps and peaks at voltages corresponding to $2\Delta/(2n+1)e$, where n is an integer. JSC as well as other two theories of multiparticle tunneling $(MPT)^3$ and multiple Andreev reflection $(MAR)^{4,5}$ have been considered as possible explanations for the current steps and peaks appearing at $2\Delta/ne$ ($n=2,3,4,...$); namely, the subharmonic gap structure (SGS).⁶⁻¹³ However, it is rather interesting that in previous experimental reports of SGS, several important features required by the JSC model are absent. To our knowledge there are few reports on the direct observation of JSC so $\tilde{\tan}$.¹¹⁻¹³

In this paper, we present the observation of Josephson self-coupling in $Nb/AlO_r/Nb$ junctions. Anomalous SGS was observed in these junctions. Steep current steps and peaks which emerged at odd submultiples of the gap voltage $2\Delta/e$ showed a magnetic-field dependence similar to the critical current I_c , while weak structures at even submultiples, which were also observed, were insensitive to magnetic fields. Moreover, step height and peak intensity at $2\Delta/3e$ were larger than those of $2\Delta/2e$ without the external magnetic field. These behaviors are different from the ordi-
nary SGS studies^{6–13} reported so far. We demonstrate tha only JSC can explain these anomalous odd series consistently. To our knowledge this is the first report on clear observation of JSC.

Subharmonic gap structure is directly predicted by JSC 'theory or by Werthamer's equation, $\frac{1}{2}$ while multipartic tunneling and multiple Andreev reflection also can explain SGS. In addition these three could arise simultaneously. Therefore it is difficult to make clear whether JSC really occurs or not, even if SOS is clearly observed. However, following criteria allow one to distinguish JSC experimentally from the two other mechanisms MPT and MAR.

(1) The magnetic-field dependence. JSC predicts both pair-current and quasiparticle-current contributions to ' SGS ,^{1,2} whereas MPT and MAR predict only quasiparticlecurrent contributions to SGS. The pair current is sensitive to the external magnetic field and the quasiparticle current is insensitive. If SGS is caused by JSC, the step height of SGS should show a magnetic-field dependence.¹⁰ For small rectangular junctions with uniform distribution of current density, the magnetic-field dependence of the height of the paircurrent step should be similar to the Josephson critical entent, i.e., a Fraunhofer pattern.¹⁴ If SGS is caused by MPT and/or MAR, the step height of SGS should be insensitive to magnetic field.

gnetic field.
(2) The line shape of SGS.^{2,10,13} The current peaks witl negative differential resistance can be explained only by the JSC picture, while the current steps can be explained by any of the three.

(3) The position of the structure.^{1,2} The JSC model derived from Werthamer's equation predicts only odd series of ~GS, while MPT and MAR predict both even and odd series. If one intends to attribute even series of SGS to JSC, additional processes, such as pair breaking in superconduction
electrodec $1,2,15$ should be considered to in addition to If electrodes, $1,2,15$ should be considered to in addition to JSC model.

The above three features are reasonable consequences of JSC and Werthamer's equation and are unlike those of ordireported until now. Although many SGS studies $6-13$ reported until now. Although many demonstrations of SGS have been reported, none of them satisfies all of the above three features.

The investigated tunnel junctions consisting of $Nb/AlO_r/Nb$ trilayer were fabricated by a method described before¹⁶ for 5 \times 5 μ m², 3 \times 3 μ m², and 2 \times 2 μ m² size and by CLIP method¹⁷ for 2×2 μ m², 1.5×1.5 μ m², 1.2×1.2 μ m², 1×1 μ m², and 0.9×0.9 μ m² size. The thickness of Al normal layer was less than 6 nm for all junctions. The critical-current density J_c of the junctions was in the range of 2.1 kA/cm² to 33 kA/cm². The current-voltage characteristic measurements were carried out under current biased condition. Measured characteristic parameters of typical junctions are given in Table I.

Current-voltage characteristics of $J_c = 11$ kA/cm² junction at 4.2 K with and without the external magnetic field parallel to the junction are shown in Fig. 1.A steep current step and a smooth one appear at the voltages corresponding to $2\Delta/3e$ and $2\Delta/2e$, respectively. The step at $2\Delta/3e$ was

TABLE I. Measured characteristic parameters of typical junctions. J_c is critical-current density under zero magnetic field. R is normal resistance. $2\Delta/e$ is gap voltage; the average voltage of current rising point and shoulder in $I-V$ characteristic. V_m is quality parameter, $I_c(0)R_{2mV}$. $RC/\tau_g = 2\Delta RC/h$, where h is Planck's constant and C is junction capacitance; we assume $C=6 \mu$ F/cm² for Nb/AlO_x/Nb junction. $I_s(0)/I_c(0)$ is step height at $2\Delta/3e$ normalized by critical current $I_c(0)$. All parameters were obtained at 4.2 K. J_c and R are the average for junctions of the same wafer and other parameters are for individual junction.

	J_{c}	R	$2\Delta/e$	V_m		Sample (kA/cm ²) ($\mu\Omega$ cm ²) (mV) (mV) RC/τ_g $I_s(0)/I_c(0)$
a	2.1	0.68	2.7	30.0	2.67	0.0041
b	7.1	0.21	2.8	36.5	0.86	0.035
$\mathbf c$	11	0.14	2.7	10.5	0.55	0.048
d	14	0.096	2.75	7.5	0.39	0.055
e	16	0.095	2.7	6.7	0.39	0.052
f	24	0.079	2.7	3.8	0.31	
g	33	0.050	2.6	2.7	0.19	

sensitive to the external magnetic field, while the step at $2\Delta/2e$ was insensitive. This SGS was observed independent of the size of junction and wiring pattern of junction. When dc Josephson current was suppressed by applying the external magnetic field, the SGS like those reported in Refs. 6, 7, and 10 was clearly observed (the lower curve in Fig. 1).This ordinary SOS was observed in all measured junctions and became more noticeable with increasing J_c .

Contrary to the ordinary SGS, the step at $2\Delta/3e$ was larger in height and steeper in slope than that at $2\Delta/2e$ without the external magnetic field (the upper curve in Fig. 1). This anomalous structure at $2\Delta/3e$ was observed in the junctions with J_c = 2.1 kA/cm² to 16 kA/cm². The structure at $2\Delta/3e$ appeared more clearly with increasing J_c (see Table I). But the structure at $2\Delta/3e$ could not be observed in junctions with $J_c>16$ kA/cm², since these still higher J_c junc-

FIG. 1. Current-voltage characteristics of a 3×3 μ m² junction with J_c = 11 kA/cm² at 4.2 K. The upper curve was obtained without the external magnetic field and the lower was with magnetic field of 14 mT. The field was applied parallel to junction and was corresponding to the fourth minimum of I_c . Vertical scale: 50 μ A/ div., horizontal scale: 500 μ V/div.

FIG. 2. The position of the structure V_s as a function of the gap voltage $2\Delta(T)/e$ for a $J_c=11$ kA/cm² junction. A solid line presents one-third of $2\Delta(T)/e$. The value of $\Delta(T)$ was varied by means of changing temperature.

tions switched back to zero voltage state at $V > 2\Delta/3e$ under zero magnetic field.

We observed the structure at $2\Delta/3e$ in junctions of the same size with different wiring pattern and in junctions of various size with the same wiring pattern. Therefore, any Josephson resonance mechanism proposed until now, e.g., Fiske step, 18 zero-field step, 19 and step due to external resonator, 20 is ruled out as an explanation for the observed structure.

Figure 2 shows the relationship between position corresponding to the $2\Delta/3e$ step and gap voltage $2\Delta(T)/e$ of a junction with $J_c=11$ kA/cm². The position of $2\Delta/3e$ step was proportional to the gap voltage $2\Delta(T)/e$ at a rate of 1/3. This figure shows that the steep step is not a resonance step but a subharmonic gap structure.

The line shapes of structure at $2\Delta/3e$ were steep steps in most cases. But the junctions with $J_c=7.1$ kA/cm² showed clear current peak at $2\Delta/3e$ with negative differential resistance, as shown in Fig. 3. The current peak cannot be ex-

FIG. 3. Current-voltage characteristics of a 5×5 μ m² junction with J_c = 7.1 kA/cm² at 4.2 K. The upper curve was obtained without magnetic field and the lower was with magnetic field of 21 mT. The field was parallel to the junction and corresponding to the tenth minimum of I_c ; this rather large field was necessary in order to suppress the first Fiske step emerging near 1.5 mV. Vertical scale: 20 μ A/div., horizontal scale: 500 μ V/div.

FIG. 4. Magnetic-field dependence of the current step height I_s at $2\Delta/3e$ (solid circles) and Josephson critical current I_c (open circles) of a $3 \times 3 \mu m^2$ junction with $J_c = 11$ kA/cm². The magnetic field B was applied parallel to the junction. In this case the structure at $2\Delta/3e$ showed step figure. I_s and I_c are normalized by the values $I_s(0)$ and $I_c(0)$ at zero magnetic field, respectively. A solid line is the theoretical dependence, $\sin(\pi B/B_0)/(\pi B/B_0)$ where B_0 is the field corresponding to the first minimum of I_c .

plained by MPT and MAR.

The magnetic-field dependence of step height at $2\Delta/3e$ of a J_c = 11 kA/cm² junction is plotted in Fig. 4. At the current minimum, a small bump was observed at $2\Delta/3e$ (see the lower curve in Fig. 1). We defined the zero of the step height as this minimum current. It is found that the magnetic-field dependence of the $2\Delta/3e$ step is similar to Josephson critical current I_c of the junction. This magnetic-field dependence corresponds to the prediction of JSC for the small rectangular junction and is never explained by MPT and MAR. Position of the step was not shifted in voltage scale by applying the magnetic field.

For some junctions, temperature dependence of the SGS was measured over the range of 1.6–4.2 K. Distinguishable change in magnitude and position of SGS was not observed with decreasing temperature except for the decrease of background leak current.

Almost all measured junctions switched back to zero voltage state at $V \ge 2\Delta/3e$ (as shown in Figs. 1 and 4) without the external magnetic field. The existence of higher-order structures in these junctions are unknown without the magnetic field. As shown in Fig. 5, however, in case of $J_c=2.1$ $kA/cm²$, some junctions showed a current step at voltage corresponding to $2\Delta/5e$ as well as $2\Delta/3e$ while neither current step nor current peak was found at $2\Delta/4e$. The normalized step heights $I_s/I_c(0)$ at $2\Delta/5e$ and at $2\Delta/3e$ in Fig. 5 are 0.0046 and 0.0041, respectively. The step at $2\Delta/5e$ was suppressed by magnetic field in the same way as that of $2\Delta/3e$ and critical current. We tried to measure quantitatively the magnetic-field dependence of these structures appearing in the J_c = 2.1 kA/cm² junction but could not obtain accurate data since these structures are so small. MPT and MAR predict both even and odd series of SGS and monotonous decrease of step height at $2\Delta/ne$ with increasing n.^{6,7} The emergence of a step at $2\Delta/5e$ without a step at $2\Delta/4e$ strongly suggests that the step at $2\Delta/5e$ is caused by JSC.

The observed anomalous behaviors of odd series of subharmonic gap structure are never predicted by multiparticle tunneling and/or multiple Andreev reflection. In other words, only the Josephson self-coupling can explain the magnetic-

FIG. 5. Current-voltage characteristics of a 5×5 μ m² junction with $J_c=2.1$ kA/cm² at 4.2 K. The upper curve was obtained without magnetic field and the lower was with magnetic field of 20 mT which was parallel to the junction and corresponding to the tenth minimum of I_c . Vertical scale: 5 μ A/div., horizontal scale: 500 μ V/div.

field dependence, current peak with negative differential resistance and the amplitude of the structure. The observed current-voltage characteristics contained two kinds of SGS; anomalous SGS caused by JSC and ordinary SGS.

Deviation of the magnetic-field dependence from JSC theory (in Fig. 4), which becomes pronounced in low current region, may be due to suppression of Josephson current by external noise.

According to the results of numerical calculation of Werthamer's equation, 15 the condition of emergence of the structures due to JSC is sufficiently low RC/τ_g value of the junction, where R is normal resistance junction, C is capacitance of the junction and $\tau_g=h/2\Delta$. As shown in Table I, the RC/τ_g value of our junctions which showed the structure at $2\Delta/3e$ was in the range 0.4–3 for $C=6$ μ F/cm². These RC/τ_g values suggest the observed structures are due to JSC.

The above condition of emergence of SGS due to JSC, low RC/τ_g value, implies a high critical-current density of the junction. Moreover low subgap leakage is required, since large leakage causes switching back to zero voltage state at large currents or at large voltages and smears the characteristic structures. Generally, it is difficult to fabricate junctions with both high J_c and low leakage.²¹ This is a possible reason why JSC has not been observed before. Note that JSC does not require large tunneling matrix element $|T|^2$; the assumption of patchy barrier^{8,9} or pinhole defects^{4,10} is no necessary.

We do not know the cause of the ordinary SGS observed in our experiment. To clarify this problem further study is required.

In conclusion, we have observed Josephson self-coupling in $Nb/AlO_x/Nb$ tunnel junctions. These junctions showed anomalies in odd series of subharmonic gap structure. At voltages $2\Delta/3e$ and $2\Delta/5e$, steep current steps and current peaks emerged. Contrary to ordinary subharmonic-gapstructure observations, these steps and peaks showed a

magnetic-field dependence that was quite similar to the Josephson critical current I_c . Even series of subharmonic gap structure which were also observed were insensitive to the magnetic field and showed rather smooth current steps. The

most likely explanation of the anomalous odd series observed here is Josephson self-coupling.

We wish to thank H. Akoh for valuable discussion and his critical reading and S. Kohjiro for valuable discussion.

- ¹ N. R. Werthamer, Phys. Rev. 147, 255 (1966).
- 2 L. E. Hasselberg, M. T. Levinsen, and M. R. Samuelsen, Phys. Rev. B 9, 3757 (1974).
- 3 J. R. Schriffer and J. W. Wilkins, Phys. Rev. Lett. 10, 17 (1963).
- ⁴ T. M. Klapwijk, G. E. Blonder, and M. Tinkahm, Physica 109 & 110B, 1657 (1982).
- 5 G. B. Arnold, J. Low Temp. Phys. 68, 1 (1987).
- 6 J. M. Rowell and W. L. Feldman, Phys. Rev. 172, 393 91968).
- ${}^{7}E$. L. Wolf, Principles of Electron Tunneling Spectroscopy (Oxford University Press, New York, 1989), p. 132, and references therein.
- R. Cristiano, L. Frunzio, and R. Monaco, J. Supercond. 5, 451 (1992).
- 9 C. L. Foden, N. Rando, A. van Dordrecht, A. Peacock, J. Lumely, and C. Pereira, Phys. Rev. B 47, 3316 (1993).
- ¹⁰ A. W. Kleinsasser, R. E. Miller, W. H. Mallison, and G. B. Arnold, Phys. Rev. Lett. 72, 1738 (1994).
¹¹I. Giaever and H. R. Zeller, Phys. Rev. B 1, 4278 (1970).
-
- ¹²O. H. Soerensen, B. Kofoed, N. F. Pedersen, and S. Shapiro, Phys. Rev. B 9, 3746 (1974).
- ¹³ P. Mukhopadhyay, J. Phys. F 9, 903 (1978).
- ¹⁴ Under the uniform magnetic field B_y parallel to junction, dc current density of small Josephson tunneling junction with uniform current distribution derived by Werthamer's equation is $J_0(eV,x,y) = \text{Im}\Sigma_{k=-\infty}^{\infty} \{M_k M_k^* j_1[(1-2k)eV] + e^{i\alpha} M_{k+1} M_k\}$ $\times j_2[(1-2k)eV]$, $\alpha=2\pi(2\lambda+d)B_vx/\Phi_0$, where λ is London penetration depth of the superconductor, d is physical

barrier thickness and Φ_0 is magnetic flux quantum (Ref. 15). For the rectangular junction of dimensions l_x and l_{v} , whole dc current is

$$
I_0(eV) = \int \int dx \, dy J_0(eV, x, y) = \text{Im} \sum_{k=-\infty}^{\infty} l_x l_y \{M_k M_k^* \times j_1 [(1-2k)eV] + (\sin z/z)M_{k+1} M_k j_2 [(1-2k)eV] \},
$$

where $z = \pi(2\lambda + d)B_y l_x / \Phi_0$. From this equation, we know that the magnetic-field dependence of amplitude of SGS due to JSC is similar to that of critical current.

- ¹⁵D. G. McDonald, E. G. Johnson, and R. E. Harris, Phys. Rev. B 13, 1028 (1975).
- ¹⁶H. Nakagawa, G. Pepe, H. Akoh, L. Frunzio, R. Cristiano, E. Esposito, S. Pagano, G. Peluso, A. Barone, and S. Takada, Jpn. J. Appl. Phys. 32, 4535 (1993).
- ¹⁷M. Aoyagi, A. Shoji, S. Kosaka, F. Shinoki, and H. Hayakawa Advances in Cryogenic Engineering Materials (Plenum, New York, 1986), Vol. 32, p. 557.
- ¹⁸ M. D. Fiske, Rev. Mod. Phys. 36, 221 (1964).
- ¹⁹ J. T. Chen, T. F. Finnegan, and D. N. Langenberg, Physica 55, 413 (1971).
- 20 T. Lehnert, F. Schafer, and K. H. Gundlach, J. Appl. Phys. 74, 1403 (1993).
- ²¹ R. E. Miller, W. H. Mallison, A. W. Kleinsasser, K. A. Delin, and E. M. Macedo, Appl. Phys. Lett. 63, 1423 (1993).

FIG. 1. Current-voltage characteristics of a 3×3 μ m² junction with $J_c = 11$ kA/cm² at 4.2 K. The upper curve was obtained without the external magnetic field and the lower was with magnetic field of 14 mT. The field was applied parallel to junction and was corresponding to the fourth minimum of I_c . Vertical scale: 50 μ A/ div., horizontal scale: 500 μ V/div.

FIG. 3. Current-voltage characteristics of a 5×5 μ m² junction with J_c = 7.1 kA/cm² at 4.2 K. The upper curve was obtained without magnetic field and the lower was with magnetic field of 21 mT. The field was parallel to the junction and corresponding to the tenth minimum of I_c ; this rather large field was necessary in order to suppress the first Fiske step emerging near 1.5 mV. Vertical scale: 20 μ A/div., horizontal scale: 500 μ V/div.

FIG. 5. Current-voltage characteristics of a $5 \times 5 \ \mu m^2$ junction with J_c = 2.1 kA/cm² at 4.2 K. The upper curve was obtained without magnetic field and the lower was with magnetic field of 20 mT which was parallel to the junction and corresponding to the tenth minimum of I_c . Vertical scale: 5 μ A/div., horizontal scale: 500 μ V/div.