Half-integral constant voltage steps in high-$T_c$ grain boundary junctions

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A novel effect from microwave radiation near 9.3 GHz applied to high-$T_c$ YBa$_2$Cu$_3$O$_{7.5}$ single grain boundary junctions was observed. In addition to the usual Shapiro steps resulting from the ac Josephson effect, constant voltage steps with voltages halfway between the voltages of the Shapiro steps were present. The widths of these "half-integral" steps were measured as a function of microwave power, and the influence of a magnetic field was investigated. From previous results on high-$T_c$ grain boundary junctions and a comparison of the results presented here with single- and multiple-junction effects in low-$T_c$ materials, we conclude that the half-integral steps are likely to be a result of grain boundaries being composed of multiple junctions in parallel.

Among the many types of Josephson junctions of high-$T_c$ superconductors, much research has focused on grain boundary junctions. Not only are the properties of grain boundaries relevant for an understanding of bulk polycrystalline samples, but also these junctions are a possible means for achieving technological applications of high-$T_c$ superconductors. In particular, the bi-epitaxial technique for producing single grain boundary junctions of YBa$_2$Cu$_3$O$_{7.5}$ has yielded junctions with many of the necessary properties for these applications. While studying the response of these junctions to microwave radiation, we observed a new and interesting effect, namely constant voltage steps in the current-voltage ($I$-$V$) curves at exactly half the voltages of the Shapiro steps expected from the ac Josephson effect. We refer to these anomalous constant voltage steps as "half-integral" steps.

The bi-epitaxial YBa$_2$Cu$_3$O$_{7.5}$ grain boundary junctions used in this study were prepared using a technique described in detail elsewhere. One single chip contained several junctions, with widths from 30 to 50 μm. Individual chips were placed in an X-band wave guide, pre-cooled in liquid nitrogen, and cooled in a liquid helium bath. With no applied microwave radiation, all the junctions had behavior typical of the resistively-shunted-junction (RSJ) model, with no excess current or hysteresis and no noticeable rounding from noise or reactive elements.

When microwave radiation is applied to a Josephson junction, constant voltage, or Shapiro, steps due to the ac Josephson effect appear in the $I$-$V$ curve at voltages $V$ given by

$$V = nV_I / K_f,$$

where $n$ is an integer and indexes the step number, $n$ is the frequency, and $K_f = 2e/h = 0.4835979 \text{ GHz} / \mu \text{V}$ is the Josephson constant. Here, we refer to these steps as integral steps. When microwaves with frequency near 9.3 GHz were applied to the junctions studied here, these integral steps were clearly visible, in some cases for step numbers as high as 120, indicating a very strong response of the junctions to the microwaves. In addition to the integral steps, there were instances where steps appeared at voltages for which $n$ is half-integral. An illustration of this is given in Fig. 1, which shows the $I$-$V$ curve of a 50 μm wide junction at 4.2 K in an applied microwave field of 9.311 GHz. At this frequency, from Eq. (1), constant voltage steps for integral $n$ are separated by 19.25 μV. These are clearly visible in Fig. 1, and are labeled accordingly. In addition, remarkably distinct and constant voltage steps for half-integral $n$ are also present. Detailed observations of $I$-$V$ curves with clearly defined half-integral steps revealed no additional step structure.

Half-integral steps were observed on all three junctions on one chip, but not on a 50 μm wide junction on another chip. The only obvious differences between the junctions on different chips were that those with half-integral steps had lower normal-state resistances and higher critical currents.

![Figure 1](image-url)
Fig. 2, which shows three distinct behaviors of the half-integral steps. The steps are indexed by \( n \), and the microwave frequency \( \nu \), critical current \( I_c \), normal-state resistance \( R_n \), and normalized frequency \( \Omega \) are given for each behavior.

Also, the occurrence of half-integral steps depended on the cool-down history, suggesting that trapped magnetic flux is important. This inference was corroborated by the effect of a small (< 5 Oe) applied magnetic field, which modulated the critical current in a complicated but reproducible manner.

By varying the critical current with the applied magnetic field, we reached a general conclusion that half-integral steps occur near minima in the critical current.

The current widths of the constant voltage steps were measured as a function of microwave power for a 50 \( \mu \)m wide junction with the same orientation in the wave guide and with respect to the applied magnetic field, but with different cool-down histories. The results are displayed in Fig. 2, which shows three distinct behaviors of the half-integral steps. They are shown in decreasing order of RSJ-like behavior of the integral steps and increasing width of the half-integral steps, with the behavior shown in Fig. 2(b) being the most commonly observed. For each behavior, the frequency \( \nu \) of the microwaves and the critical current \( I_c \), normal-state resistance \( R_n \), and normalized frequency \( \Omega = \nu I_c R_n K_f \) of the junction are given. The applied magnetic field was varied to obtain an \( I_c \) at which half-integral steps occurred, while \( R_n \) of this junction increased slowly due to aging. The voltages of the steps are indexed by \( n \), from Eq. (1), solid circles are data points, and lines are guides to the eye. The microwave amplitude is somewhat arbitrary since the power coupled into the junction is a small, unknown fraction of the measured power. Only one type of behavior was possible for a given cool-down history, probably because the small applied magnetic field had less of an effect than the field from the trapped flux, and the microwave frequency had no effect on the behavior, at least in the range 8.5 to 10.5 GHz. The \( I-V \) curve shown in Fig. 1 corresponds to a microwave amplitude of 1.75 (\( \mu \)W)\(^{1/2} \) in Fig. 2(b).

For the smallest half-integral step widths, shown in Fig. 2(a), the integral steps have nearly the behavior predicted by the RSJ model, with, for example, the maximum (minimum) of the \( n = 0 \) and 2 steps occurring at the same power as the minimum (maximum) of the \( n = 1 \) step. However, the widths of the integral steps never go completely to zero, and the half-integral steps have a complicated dependence on microwave power, with an alternation between the \( n = 1/2 \) and 3/2 steps. For intermediate half-integral step widths, shown in Fig. 2(b), the integral steps are less like those of the RSJ model. As above, the widths of the integral steps never go to zero, and also there are two minimum widths for the integral steps, although the wider minima are probably a result of the half-integral steps. In contrast to Fig. 2(a), here at higher power the half-integral steps, \( n = 1/2 \) and 3/2, have maxima at the same microwave power. For the largest half-integral step widths, shown in Fig. 2(c), the behavior of the widths of the integral steps is not at all like that given by the RSJ model. For example, the maximum width of the \( n = 1 \) step occurs at a maximum in the width of the \( n = 0 \) step. However, the widths of the integral steps do go completely to zero. As for the half-integral steps, the widths do not have maxima and minima at the same power, in agreement with those shown in Fig. 2(a) but in contrast to those in Fig. 2(b). Finally, at magnetic fields for which no half-integral steps occurred, the dependence of the integral steps on microwave power was that predicted by the RSJ model.

What is responsible for these half-integral steps? Several possibilities can be rejected quickly and are discussed first. Other, more plausible possibilities are divided into single- and multiple-junction effects and discussed later.

An obvious possibility is a subharmonic of the primary microwave frequency, but the cut-off frequency of the X-band wave guide is 6.55 GHz. Single-electron effects, such as phonon- and photon-assisted tunneling, would have a factor of \( e \), rather than \( 2e \), in the denominator of Eq. (1), resulting in voltages twice those of the integral steps. Finally, effects from "long" junctions are ruled out because in such junctions the voltages of steps due to flux flow are inversely proportional to the width of the junction, whereas the voltages observed here were always given by Eq. (1) to within 200 ppm (parts per million) regardless of the width of the junction.

Turning now to single-junction effects, early experiments on the ac Josephson effect in weak link junctions observed non-integral steps in the \( I-V \) curves. It is entirely reasonable to view grain boundary junctions as weak links, rather than tunnel junctions, because of their \( I-V \) curves. In general, non-integral steps occur in weak links when a harmonic of the Josephson frequency coincides with a harmonic of the applied frequency and the two phases lock. Harmonics can be produced by either a non-sinusoidal current-phase relation or the inclusion of reactive elements in the RSJ model. The addition of a capacitance, \( C \), results in the definition of a dimensionless parameter \( \beta_c = 2\pi K H_c R_n C \). With no microwave radiation, the \( I-V \) curve is hysteresic for \( \beta_c > 1.3 \).
The published results on non-integral steps from single-junction effects in low-$T_c$ materials, however, are not in agreement with our results. Specifically, we observed only half-integral steps, while the published results have steps for fractional values of $n$ other than one-half. Also, non-integral steps seem to occur only for values of $\beta \geq 1$ or $\Omega \geq 1$, or both,\(^7\) while $\beta < 1$ for our junctions since they are not hysteretic and $\Omega < 1$ from Fig. 2. Finally, the dependences of the step widths on microwave power do not agree, either in the relative widths of the steps or in the powers at which minima occur.\(^7\)\(^8\)

As for multiple-junction effects, evidence is increasing that $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ grain boundary junctions are heterogeneous. This comes from microstructural features observed in grain boundaries,\(^10\) as well as measurements of the critical current as a function of magnetic field suggesting that a network of microbridges occupies a small portion of the total grain boundary.\(^11\)

Assuming that grain boundary junctions are composed of a network of smaller junctions, one possibility is a two-dimensional (2D) array of junctions. Non-integral steps have been observed in such arrays when a current flows parallel to the plane of the array and a magnetic field is applied perpendicular to this plane.\(^2\) These non-integral steps are explained by the field-induced vortex superlattice slipping perpendicularly to the applied current,\(^13\) in synchrony with the rf drive current.\(^13\) The suggestive properties of 2D arrays relevant to our results are the occurrence of non-integral steps and the necessity of a magnetic field. However, if grain boundary junctions are actually 2D arrays of junctions, the plane of the array would be in the plane of the junction, meaning the current flow would be perpendicular to this plane. However, disordering could modify the direction of current flow, and there is evidence that the plane of these grain boundary junctions are inclined by as much as $10^\circ$ from the normal of the substrate.\(^14\) Another point of disagreement is that in 2D arrays the voltages of the non-integral steps are not constant.\(^12\)

The other possible arrangement of multiple junctions in grain boundaries is a one-dimensional (1D) array, with a number of junctions in parallel. Recent experimental and theoretical results by Vanneste et al.\(^15\) on two Nb-based junctions in parallel (a SQUID) with applied magnetic and microwave fields show consistent similarities with our results. They observed distinct, constant voltage half-integral steps in $I$-$V$ curves, and the pattern of these curves was periodic as a function of magnetic field. By using the RSJ model and including capacitive and inductive elements, they were able to reproduce their experimental results and found that the half-integral steps are a result of a flip-flop between two fluxoid states of the SQUID synchronized to the microwave field. There are several other striking similarities between our results and those of Vanneste et al. Their junctions were characterized by $\beta = 0.86$ and $\Omega = 0.21$, values which are consistent with those of our junctions. Moreover, they found that half-integral steps occur as a function of magnetic field, which agrees with our suggestion that trapped magnetic flux is important, and that the maximum widths of the half-integral steps occurred at a magnetic field for which the critical current was a minimum. This last point is in complete agreement with our results.

It is reasonable to consider $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ grain boundary junctions to be composed of several weak links, and the half-integral steps we observe are most likely caused by multiple junctions in parallel. Efforts are now underway to more fully investigate such an arrangement of junctions to determine how the values of inductance and capacitance and the number of junctions influence the occurrence and behavior of half-integral steps. Also, additional experiments are needed to correlate the occurrence of half-integral steps with junction parameters such as $I_c$ and $R_p$.

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