Observation of the Bloch Oscillations in an Ultrasmall Josephson Junction

L. S. Kuzmin^{(1),(2)} and D. B. Haviland⁽¹⁾

⁽¹⁾Department of Physics, Chalmers University of Technology, S-412 96 Göteborg, Sweden ⁽²⁾Laboratory of Cryoelectronics, Physics Department, Moscow State University, Moscow 119 899 GSP, U.S.S.R. (Received 25 March 1991; revised manuscript received 25 September 1991)

We have studied the low-temperature behavior of lead-alloy Josephson tunnel junctions with area $S \approx 0.01 \ \mu m^2$, isolated from their electromagnetic environment by high-resistance metallic resistors inserted into the current and voltage leads. Under irradiation with microwaves frequencies, f=3.5-10GHz, the dc differential resistance dV/dI, as a function of the dc current I, showed peaks at $I = \pm 2ef$. This effect, and other observations, arises due to the periodic electrical recharging of the junction by discrete Cooper pairs, and can be explained by the "orthodox" theory of Bloch oscillations.

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After a few years of intensive theoretical and experimental research it is now well established that a considerable time and/or space correlation of the single-electrontunneling (SET) events does exist in ultrasmall junctions, provided that they are well shielded from the shunting effect of their high-frequency electrodynamic environment (for a recent review, see, e.g., Ref. [1]). In a single junction biased by a dc current I, the correlation takes the form of periodic SET oscillations of frequency f_{SET} =I/e.

Oscillations with a similar frequency-to-current relation

$$f_B = I/2e , \qquad (1)$$

although of a fundamentally very different character, were predicted [2] to take place in Josephson junctions due to correlated tunneling of Cooper pairs. This phenomenon (named Bloch oscillations) is of quite general importance to physics [1]. Experimental studies of the correlated tunneling were, however, considerably hindered by the shunting effect of the high-frequency environment coupled to the junction via unavoidable current and voltage leads [1]. In this uncertain situation the very existence of continuous-wave Bloch oscillations was repeatedly questioned (see, e.g., Ref. [3]).

If the Bloch oscillations do really exist, one can observe this effect by applying an ac drive of frequency f (typically in the microwave range) to the junction. In this case, complete phase locking of the oscillations by harmonics of the ac drive would result in voltage steps in the dc current-voltage (I-V) characteristic of the junction, located at quantized levels of the dc current [2]

$$I = I_n = n2ef, \quad n = \pm 1, \pm 2, \dots$$
 (2)

These steps are the quantum-dual analog [1] of the wellknown Shapiro steps in larger Josephson junctions. In practice, the phase locking can hardly be perfect (at least due to nonvanishing thermal fluctuations and singleelectron quasiparticle tunneling [4-6]) and the voltage steps are not horizontal. Nevertheless, one can expect to observe more or less sharp singularities at the current values given by Eq. (2).

To our knowledge, the only claim to observing these singularities has come from studies [7] of granular tin films near the percolation threshold, where a single (critical) Josephson weak link between the grains could be screened from the environment by other junctions in the percolation path. However, a further analysis of the I-Vcurves (which had been heavily contaminated by noise) has shown that the claim cannot be justified [8].

The purpose of this Letter is to report preliminary experimental [1] results of a study of single Josephson junctions screened from a low-impedance environment by special high-resistance resistors [1,6,9]. We believe that the data obtained do give convincing evidence of the voltage steps (2) and hence of the Bloch oscillations.

Lead-alloy (CrPbAuIn/PbAu) junctions were fabricated by the usual shadow-mask evaporation technique (see, e.g., Ref. [10]). The suspended mask was formed in double-layer resist (bottom layer PMMA/PMAA and top layer PMMA) using electron-beam writing. The junctions had a thermal-oxidation barrier and a nominal area of $0.1 \times 0.1 \ \mu m^2$. The Cr thin-film resistive leads and the superconducting tunnel junction were made in one vacuum cycle by evaporation from three different angles with a shift of 0.3 μ m between successive angles. To achieve a smaller grain size of the Pb alloy, we used multilayer evaporation (up to ten layers in each electrode) at oxygen pressure near 1×10^{-5} Torr. The component materials (Cr, Pb, Au, and In) were evaporated onto an unoxidized silicon substrate at temperatures close to 0°C. A schematic of the four-point measurement scheme is shown in the inset in Fig. 1.

The sample was placed in a dilution refrigerator and cooled to 60-70 mK, where it was measured with wellshielded, battery-operated, analog electronics. The dV/dI measurements were made with two lock-in amplifiers (one regulating a feedback circuit for constant excitation dI) at 130 Hz. Measurements were made in magnetic fields of up to 2.8 kG, however, the exact value of the field at the junction point is not known at this stage of our experiments due to a complicated geometry. Microwave



FIG. 1. The general shape of the dc I-V curve of the junction (curve A), and a blowup of its central part (curve B) showing the current I_m where the I-V curve jumps to the quasiparticle branch. Inset: The layout of our junction-plus-resistors circuit and the four-point dc measurement scheme.

excitation could be fed to the junction via a microcoax line with a helium-cooled 20-dB attenuator and 100-pF chip capacitors.

Figure 1 shows the dc I-V curve of one particular sample discussed here, measured at T = 60 mK. In zero magnetic field, it had a dc Josephson-current-like branch with a vanishing dc voltage. Application of a magnetic field, however, not only suppressed the height of this branch, I_{m} , but also induced a distinguishable voltage step near the origin (Fig. 2). The differential resistance, dV/dI, shows this step clearly as a peak at I=0 [see the rightmost trace of Fig. 3(b)].

When microwaves were applied the central peak was



FIG. 2. The effect of the magnetic field on the dc I-V curve at temperature T=70 mK. The curves are offset along the V axis for clarity.



FIG. 3. (a) dc I-V curve with and without microwave irradiation at f = 4.0 GHz. Measurements of the dynamic resistance dV/dI (horizontal axis) vs the dc current I (vertical axis) for (b) several intermediate values of the microwave power at frequency f = 4.09 GHz, and (c) several values of f at certain power levels. Two traces of each curve were recorded to show the noise level, and the curves are displaced along the horizontal axis for clarity. The arrows mark the theoretical prediction $I = \pm 2ef$. An external magnetic field of 2 kG was applied.

suppressed [Figs. 3(a) and 3(b)], and, simultaneously, two symmetric side peaks appeared within a range of microwave power [Fig. 3(b)]. The peaks were rather broad $(\Delta I \approx 1 \text{ nA})$, and their center positions on the dc current axis could be only measured to a few tens percent error. Within this accuracy, and our frequency range (3.5-10 GHz), the peak positions were independent of the power as shown in Fig. 3(b), and corresponded to Eq. (2) [Figs. 3(b), 3(c), and 4].

All the observed features find a ready qualitative explanation within the framework of the present-day "orthodox" theory of correlated tunneling of Cooper pairs [1], although a quantitative comparison is difficult to carry out because at this stage of our work there is a significant uncertainty in important parameters of the theory.

The Josephson coupling energy E_J was maximum at zero magnetic field and could be calculated from the



FIG. 4. The dc current *I*, which is the half-distance between the microwave-induced peaks of dV/dI, plotted as a function of the applied frequency *f*. Points show experimental data, while the line corresponds to the theoretical prediction I = 2ef.

Ambegaokar-Baratoff theory to be close to 400 μ eV, using the measured values of the normal resistance of the junction $R_n = 8.5 \text{ k}\Omega$, and the superconducting energy gap $2\Delta/e = 2.1 \pm 0.1 \text{ mV}$. At the magnetic fields corresponding to the appearance of the voltage steps (0.7 kG), however, E_J could be only crudely estimated as being of the order of 200 μ eV.

For estimation of the elementary charging energy $E_C = e^2/2C$, we could use either the junction capacitance value $C^{(1)} \approx 4.8 \times 10^{-16}$ F calculated from the nominal area $S = 0.01 \ \mu\text{m}^2$ and the specific capacitance $C_S = 4.8 \ \mu\text{F/cm}^2$ deduced from large-junction measurements or calculate [1] E_C from the asymptotic voltage offset ΔV of the dc *I-V* curve $E_C^{(2)} = e\Delta V/2$. ΔV was determined by careful extrapolation of the linear part of the *I-V* curve from voltage greater than $2\Delta/e$, but less than barrier suppression voltages (~20 mV). These two values, $E_C^{(1)} \approx 170 \ \mu\text{eV}$ and $E_C^{(2)} \approx 350 \ \mu\text{eV}$, respectively, differ considerably for reasons which are not yet understood.

Thus we could only conclude that the ratio E_J/E_C , which is important for the theory, is of the order of unity. This fact did not make the analysis easy, because in this range the theory [1] makes very few quantitative predictions. On the other hand, exactly this range is optimal for the realization of the Bloch oscillations [9,11].

The third key parameter is the damping coefficient $\alpha(\omega) \equiv \operatorname{Re} Y(\omega) R_Q$, where R_Q is $h/4e^2 \cong 6.5 \text{ k}\Omega$, the quantum unit of resistance, and $\operatorname{Re} Y(\omega)$ is the active conductance of the electrodynamic environment as seen by the junction [12]. Most theoretical calculations were carried out with the assumption that $\operatorname{Re} Y(\omega) = G$ =const, while in our case it is essentially frequency dependent. As $\omega \rightarrow 0$, ReY(ω) equals just $1/R_l$, where R_l is the dc resistance of each lead ($R_l = 95 \text{ k}\Omega$ for the particular sample discussed here). However, because of the considerable stray capacitance C_l of the resistors and their relatively large length $(l = 28 \ \mu m)$, at high frequencies Re $Y(\omega)$ increases. At $\omega > 1/R_lC_l$ it can be crudely estimated as $(\omega C_l/2R_l)^{1/2}$. To our knowledge, the effect of such a frequency dispersion on the Bloch oscillations has not yet been studied, however, we can guess that the most important frequencies for the existence of the effect are of the order E_I/h . With our estimate of E_I , this gives $\omega^{(1)} \approx 3 \times 10^{11} \text{ s}^{-1}, \text{ Re}Y^{(1)} \approx (20 \text{ k}\Omega)^{-1}, \text{ and } \alpha \approx 0.3$ (we have used a calculated value $C_l/l = 0.6 \times 10^{-16}$ F/ μ m). According to the theory [1], we should observe the Bloch oscillations for this value of α , although their linewidth should be considerably broadened by quantum fluctuations, even as $T \rightarrow 0$. In the experiment, however, the most important reason of the broadening should be the thermal fluctuations which yield the linewidth

$$\Delta f = 2\Gamma_{\omega}/2\pi, \quad \Gamma_{\omega} = (\pi/e)^2 \operatorname{Re} Y^{(2)} k_B T , \qquad (3)$$

where $\operatorname{Re} Y^{(2)}$ should be evaluated at relatively low frequencies $(0 < \omega < \Gamma_{\omega})$. For our sample, at T = 70 mK, this yields $\Delta f \approx 1.2$ GHz (i.e., $\Delta I \approx 0.4$ nA in dc current 2892 units). From Fig. 3 we see that the observed ΔI is somewhat larger (≈ 1.0 nA). This discrepancy can be readily attributed to an additional broadening of the oscillation linewidth by a finite rf current used to measure the derivative, and some additional low-frequency noise caused by heating of the resistors due to a hot-electron effect [13]. Analyzing the temperature dependence of the I-V curves, we can experimentally determine an upper limit of this noise as an effective temperature of about 100 mK. This effective temperature would result in a linewidth of $\Delta I \approx 0.6$ nA.

Note that an alternative explanation of the ac-induced peaks as the second-number SET-oscillations step is excluded, first, by a complete absence of the first number steps of this type (at I=ef) and, second, by a very large $\omega R_{qp}C$ product (>10), where $R_{qp} \approx 10^6 \Omega$ is the effective quasiparticle resistance of the junction at these voltages (the SET oscillations can have a noticeable amplitude only at $\omega R_{qp}C < 0.1$).

We may also rule out the possibility that the observed features in dV/dI under microwave irradiation are due to the Josephson-Shapiro steps, which would appear as a "dip" in dV/dI. Figures 3(a) and 3(b) show that the features in dV/dI observed under microwave irradiation occur on the Josephson-current-like branch of the *I-V* curve, which has the slope $\sim 0.3 \text{ k}\Omega \ll R_n$. Examining the traces labeled -10 dB of Fig. 3 (f = 4.0 GHz), we see a dip located at $I_{\text{dip}} = \pm 2.2 \text{ nA}$ [Fig. 3(b)] or $V_{\text{dip}} = \pm 0.66 \ \mu\text{V}$ [using Fig. 3(a)]. The voltage position of this dip is completely wrong for Josephson-Shapiro steps, which would occur at $V_{\text{step}} = \pm 8.3 \ \mu\text{V}$ for f = 4.0 GHz.

The central peak of the dV/dI, i.e., the voltage step ΔV in the *I-V* curves (Fig. 2), appears in the theory as the Coulomb blockade of the Cooper-pair tunneling, and has also been seen in similar junctions [14,15]. In our case it is somewhat smeared out by fluctuations, but its qualitative behavior (a rapid growth with the increase of magnetic field which suppresses E_J) is in accord with the theory [1] which yields $V_I \propto \exp[-(8E_J/E_C)^{1/2}]$ at E_J > E_C .

Finally, we can identify the top of the Josephsoncurrent-like branch [current I_m in Fig. 1(a)] with the crossover between the Bloch oscillations and Zener tunneling [1]. Substituting our parameters to the tightbinding and weak-binding formulas [1] for the crossover current, we get the values 50 and 9 nA, respectively. The experimental value of 11 nA is within this bracket.

In conclusion, we believe that the observed microwave-induced singularities of the Josephson-junction dynamic resistance at the dc current values $I = \pm 2ef$ give convincing evidence of the existence of continuous-wave Bloch oscillations. Other features of the observed dc I-Vcurves, including the voltage peaks near the origin, can be qualitatively explained by the existing orthodox theory of the correlated tunneling. A more exact comparison is, however, hindered by the absence of quantitative theoretical predictions for our parameter region $(E_J \approx E_C)$, and by a lack of means to measure relevant parameters of the system independently. A more accurate comparison should be the main goal of future experiments.

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