Josephson Effect in MgB_2 Break Junctions

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(Received 11 May 2001; published 9 August 2001)

We present the first observation of the dc and ac Josephson effect in MgB_2 break junctions. The junctions, obtained at 4.2 K in high-quality, high-density polycrystalline metallic MgB_2 samples, show a nonhysteretic dc Josephson effect. By irradiating the junctions with microwaves we observe clear Shapiro steps spaced by the ideal ΔV value. The temperature dependence of the dc Josephson current and the dependence of the height of the steps on the microwave power are obtained. These results directly prove the existence of pairs with charge $2e$ in MgB_2 and give evidence of the superconductor-normal metal-superconductor weak link character of these junctions.

DOI: 10.1103/PhysRevLett.87.097001 PACS numbers: 74.50.+r, 74.70.Ad, 74.80.Fp

The discovery of superconductivity at 39 K in magnesium diboride [1] has raised great excitement in the scientific community. Apart from the possibility of finding new simple compounds with a higher critical temperature, a great interest arises from the possible applications of this new superconductor. Among the others we can mention the realization of superconducting cavities for particle accelerators working at the liquid helium temperature and superconducting electronics with good performances at a temperature ($\sim$10–15 K) accessible to cryocoolers. These goals require the realization of high-quality thin films and junctions of MgB_2 but also the knowledge of the fundamental superconducting properties of the material. Josephson junctions are the heart of most of the superconducting electronic devices and, therefore, the knowledge of their dc and ac properties and of their temperature dependence is crucial. On the other hand, the Josephson effect is a *direct probe* for the existence of pairs and a precise instrument for the study of the surface properties of superconductors. From the I-V characteristics of Josephson contacts the superconducting gap Δ can also be estimated. Many papers have recently appeared in the literature concerning the determination of Δ from tunneling [2–5], Andreev reflection [6,7], angle resolved photoemission specroscopy (ARPES) [8,9], specific heat [10], and Raman [11] measurements. The results are still controversial: Δ ranges between $\sim$2 and $\sim$7 meV in the different experiments but its temperature dependence seems to approximately follow the BCS behavior [6,7]. Various authors have reported the possible presence of a depressed superconducting layer on the MgB_2 surface [2].

In the present Letter we report the first (to our knowledge) observation of the dc and ac Josephson effect in MgB_2 junctions obtained by using the break-junction technique. The measured I-V curves show values of the product I_cR_N (where I_c is the critical current and R_N is the normal resistance) which are smaller than the value predicted by the BCS theory, and a reduced T_c with respect to the bulk value. We determined the temperature dependency of the Josephson critical current by fitting the experimental I-V curves with the resistively shunted junction (RSJ) model. The results give evidence of the superconductor–normal metal–superconductor long weak-link nature of the Josephson break junctions. Finally, by applying to the junctions a microwave (MW) excitation, we observed clear Shapiro steps and we were able to study the dependency of their height on the normalized MW voltage.

The starting MgB_2 polycrystal bulk material, of cylindrical shape, was obtained by reaction sintering of elemental B and Mg for 3 h at 950 °C in a sealed stainless steel container, lined with Nb foil. Details about the preparation technique will be given elsewhere [12]. The resulting highly dense (density $= 2.40$ g/cm^3) MgB_2 cylinders had a metallic appearance and a very high hardness. By using a fine, 0.15 mm thick diamond circular saw we cut from the bulk material small thin plates ($\sim 2 \times 1$ mm^2) with a thickness of 0.2–0.5 mm. The polished surfaces of these plates presented a shiny metallic aspect and, when observed by a metallographic microscope, they showed large (up to 50–60 μm) single-crystal-like grains, with dark mirrorlike surface, embedded in a more amorphous metallic-like background. The superconducting properties of these samples were studied by ac susceptibility (field equal to 0.1 G at 10 kHz) and resistivity (four-probe ac technique with frequency equal to 133 Hz) measurements. The main graph of Fig. 1 shows the resistivity of our MgB_2 samples, while in the upper and lower insets the real part of the susceptibility and an enlarged view of the resistive transition are presented, respectively. Both the measurements give a critical temperature (onset) $T_c = 38.8$ K. The transition

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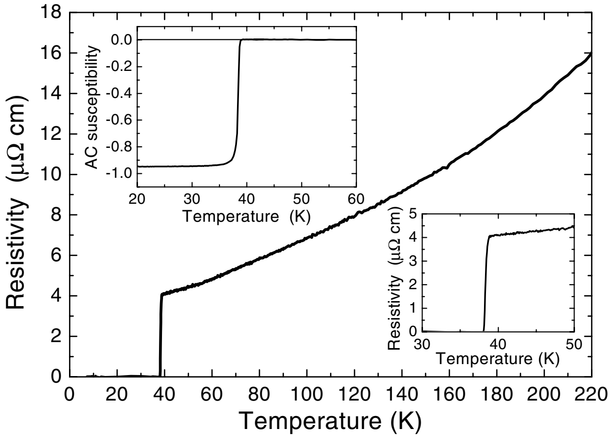

FIG. 1. Temperature dependency of the resistivity of a MgB_2 sample; upper inset: temperature dependency of the real part of the ac susceptibility of the same sample; lower inset: enlarged view of the resistive transition.

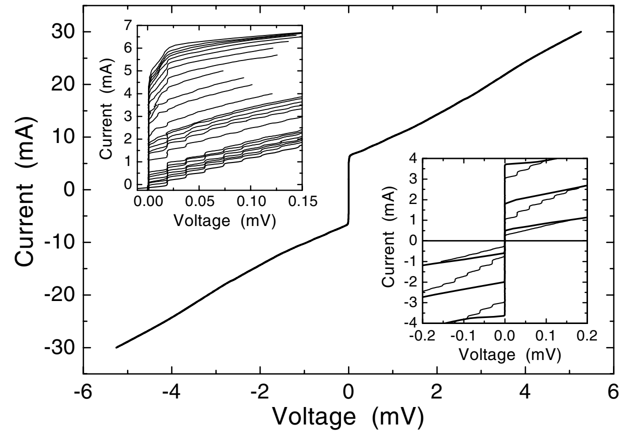

FIG. 2. Current vs voltage characteristic of a MgB_2 break junction at 5.28 K. In the upper inset, the I-V curves of the same junction for increasing rf powers (from zero to 20 mW going from top to bottom) of an applied microwave radiation at 8.843 GHz are shown. The lower inset shows the I-V curves of three other junctions with and without microwave excitation at 14.58 GHz (thin and thick solid lines, respectively).

width $\Delta T_c = 0.5$ K was determined from the full width at half maximum of the derivative of the real part of the susceptibility. The rather low value of the residual resistivity ($\rho_0 \approx 4\ \mu\Omega$ cm) and the sharpness of the resistive and diamagnetic transitions prove the high quality of the samples.

The thin MgB_2 plates were successively cut with the diamond saw into small parallelepiped samples ($\sim 0.5 \times 0.5 \times 2\ \mathrm{mm}^3$) which were then fixed to an insulated elastic support by means of epossidic glue. The current and voltage contacts on the samples were obtained by means of Ag paste and showed a contact resistance of the order of 0.2–1 Ω. By breaking the sample at 4.2 K and then carefully adjusting the pressure between the two parts by means of a tip which bends the elastic sample holder [13], we were able to obtain reproducible and stable nonhysteretic Josephson junctions with normal resistances between $\sim$ 0.1 and 11 Ω. The I-V curves were recorded by injecting a current in the junction (Keithley 220 current source) and measuring the voltage drop at its ends (HP 3457A digital voltmeter).

An example of the low-temperature I-V characteristics is shown in Fig. 2 (main graph). In this case at $T = 5.28$ K we have $I_c \approx 7$ mA and $R_N \approx 0.15\ \Omega$ from which a product $I_cR_N \approx 1$ mV is obtained. Notice that for an ideal, pin-hole free S-I-S planar junction with critical temperature equal to the bulk T_c ($\simeq 39$ K) the BCS theory predicts instead $I_cR_N \approx 9.3$ mV at low temperature. This disagreement is not surprising, since a break junction should be thought of as being made up of a large number of weak links, each with its own critical current density and I_cR_N product. As a consequence, every value we report in the present Letter should be considered as an effective average and should be more appropriately compared to the estimates provided by the theories which expressly describe the behavior of short and long weak links. This is exactly what we will do in the following discussion of I_c vs T results.

By applying to this junction a microwave excitation at $\nu = 8.843$ GHz, clear Shapiro steps spaced by the theoretical value $\Delta V = h\nu/2e \approx 18.3\ \mu$V were observed, whose heights modulated at the change of the MW power. These current steps were observed at voltages up to 1.1 mV, and for some values of the MW power submultiples Shapiro steps at $V = nh\nu/4e$ were also present. The curves reported in the upper inset of Fig. 2 were obtained in the same junction of the main graph, by varying the MW power from zero up to the maximum power available from our source (20 mW). The presence of the steps and their spacing in voltage are a direct proof of the Josephson nature of the zero-bias current. By varying the contact and changing the samples we obtained various I-V curves with the same features of that shown in the main graph of Fig. 2. In the lower inset of the same figure, for example, the characteristics at 4.2 K of three different junctions both with and without MW excitation at 14.58 GHz are presented. The shape of all these curves (when no rf signal is applied) is similar to that observed in superconductor–normal metal–superconductor (S-N-S) junctions [14–16] and close to the prediction of the standard RSJ model for a current-biased junction and in the presence of thermal fluctuations. We will discuss this point in the following.

All the results reported so far were obtained in junctions having $R_N \lesssim 1\ \Omega$ and a high critical current ($I_c \sim 0.6$–8 mA). In these junctions the I_cR_N value remains small (0.3–1.7 mV) and all the I-V curves present the same features (including the ac Josephson effect) as those shown in Fig. 2. We also obtained a second kind of junctions, having $R_N > 1\ \Omega$ (up to $\sim$11 Ω), and a smaller zero-bias current ($I_c \sim 0.3$–2.5 mA). In these junctions the product I_cR_N reaches the largest values (3.2–3.8 mV) and the I-V characteristics are still nonhysteretic, but present some differences with respect to those of the first kind. In particular, an evident knee is present at $|V| \approx 4-5$ mV, that we interpret as being due to the quasiparticle tunneling features of the junction. Some traces of this knee (always in the same position) are also present in most of the I-V curves of the first kind at low temperature (see Fig. 2). From the local peaks of the junction's dynamic conductance dI/dV due to these knees, an approximate value of the low temperature gap $\Delta \sim 1.7-2$ meV can be obtained. Unfortunately, applying the MW excitation to the junctions with higher I_cR_N causes the total cancellation of the zero-bias current, without producing any sign of Shapiro steps. This result proves the non-Josephson nature of the zero-bias current in the higher-resistance junctions. In the present paper we analyze only the I-V curves of the junctions showing both the dc and ac Josephson effect.

The mechanical stability of the junctions during thermal cycling was usually high. As a consequence, we were able to determine the temperature dependency of the I-V curves in most of the contacts. Figure 3(a) shows this dependency between $T = 5.28$ K and the *critical temperature of the junction* $T_c^j \approx 26-27$ K for the same contact already shown at low T in Fig. 2. In this figure, for clarity, only some of the measured curves are presented. To give an idea of the contact stability, we also report in Fig. 3(a) the I-V characteristic measured at 5.4 K after the heating of the break junction up to 40 K (dashed line). Very small differences are present with respect to the curve measured before the thermal cycle.

Even though, as we will see, the intrinsic nature of our junctions is related to the presence of S-N-S weak links, as a first approximation we can compare the I-V curves with the prediction of the RSJ model [16] for current-biased junctions with a very small capacitance and in the presence of thermal fluctuations. The result of this comparison is shown in the inset of Fig. 3(a) for the I-V curve at $T = 5.28$ K (open circles are experimental data while the solid line is the result of the RSJ model). By applying this approach to all the data shown in Fig. 3(a) we determined the I_c vs T dependency that is presented in Fig. 3(b). The expanded view around $T_c^j = 26.4$ K reported in the inset of the same figure (solid circles) clearly shows that, at $T \approx T_c^j$, $I_c \propto (T_c^j - T)^2$. This is a sign of the S-N-S nature of the contact in our break junctions. A comparison with the predictions of the de Gennes theory for the proximity effect in metal barrier junctions [16] gives good results at $22\text{ K} < T < T_c^j$ but completely fails at low temperature.

A temperature behavior of the critical current very similar to our experimental data in the whole temperature range is instead predicted by the theory of weak links with S-N-S structure, which was derived by solving the Usadel equations for a one-dimensional structure with electrodes in equilibrium and a zero critical temperature in the weak link material [17,18]. The I_c vs T theoretical curve is similar to the experimental one shown in Fig. 3(b) for values of the parameter $L/\xi_N(T_c)$ between 6 and 10. Here L is the length of the normal weak link and $\xi_N(T_c)$ is the decay

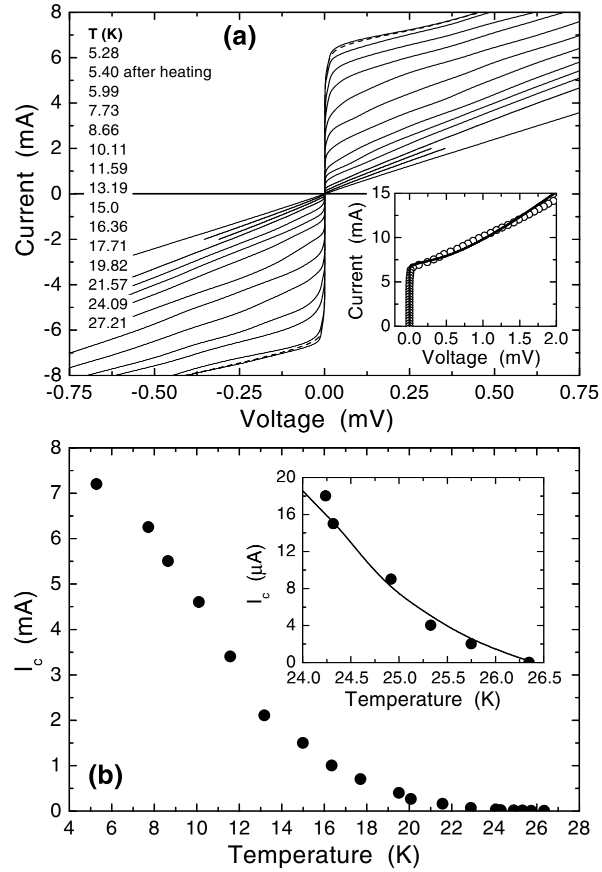

FIG. 3. (a) Temperature dependency of the I-V curves of the same contact shown at low T in Fig. 2. The inset shows the RSJ fit (solid line) of the data at 5.28 K (open circles). (b) Temperature dependency of the critical current and low-temperature fit by the model for S-N-S weak link structures (solid line in the inset).

length in the weak link material at the critical temperature of the banks. The solid line in the inset of Fig. 3(b) represents the theoretical curve predicted by this model at $T \approx T_c^j$ which best agrees with our experimental data. It was calculated by using $T_c = T_c^j = 26.4$ K and the corresponding BCS gap $\Delta = 4$ meV, and adjusting the value of $L/\xi_N N(T_c)$ to get the best fit [obtained for $L/\xi_N(T_c) = 9.9$] . Nevertheless, other values of the parameters, ranging between $\Delta = 1.7$ meV, $L/\xi_N(T_c) = 7.85$, and $\Delta = 4.2$ meV, $L/\xi_N(T_c) = 10$, give theoretical curves in good agreement with the experimental data at $T > 0.9\, T_c^j$. In this framework, the S-N-S long weak-link nature of the junction [with $L/\xi_N(T_c) = 8 \div 9$] and the reduced T_c^j also explain the value of the product $I_cR_N \approx 1$ mV we measured at low temperature ($\sim 0.2T_c$).

The present theory for S-N-S weak link structures is valid only for a superconductor in the dirty limit. This condition is not fulfilled in the *bulk* MgB_2 since $\ell = 600$ Å and $\xi \approx 50$ Å [19]. Nevertheless, we argue that the conditions for the dirty limit can be fulfilled, at least at low temperature, in the region of our contact: the low T_c^j with respect to the bulk T_c of MgB_2 indicates a local depression of the superconducting properties in the bank material with a possible increase of disorder and a reduction of ℓ in the weak link region. At least two explanations are possible for this low T_c^j value: (i) an intrinsic layer with depressed superconducting properties at the surface of the MgB_2 grains; (ii) an extrinsic effect of damaging maybe due to the great mechanical stress produced in the region where the break has occurred. On the other hand, we have clear evidence of a reduction of T_c^j from $\approx$33 K to $\approx$26 K after repeated thermal cycling of the same junction between 4.2 K and room temperature, even though very good Josephson characteristics are still present a few days after breaking.

Finally, in Figs. 4(a) and 4(b) we present in greater detail the ac Josephson effect results. Figure 4(a) shows a set of I-V curves measured in a junction with low resistance after irradiation with microwaves at $\nu = 14.58$ GHz and various powers. We observed up to 9–10 vertical Shapiro steps spaced by the ideal value $\Delta V = 30.2\ \mu$V. The magnitude of the supercurrent ($n = 0$) and the heights of the first three steps ($n = 1, 2, 3$) as functions of the normalized rf voltage are shown in Fig. 4(b). The frequency of the applied microwave gives a normalized frequency $\Omega = h\nu/2eR_N I_c = 0.19$. The experimental curves shown in Fig. 4(b) are in good qualitative agreement with the results of the standard RSJ current biased model of the irradiated junction [16].

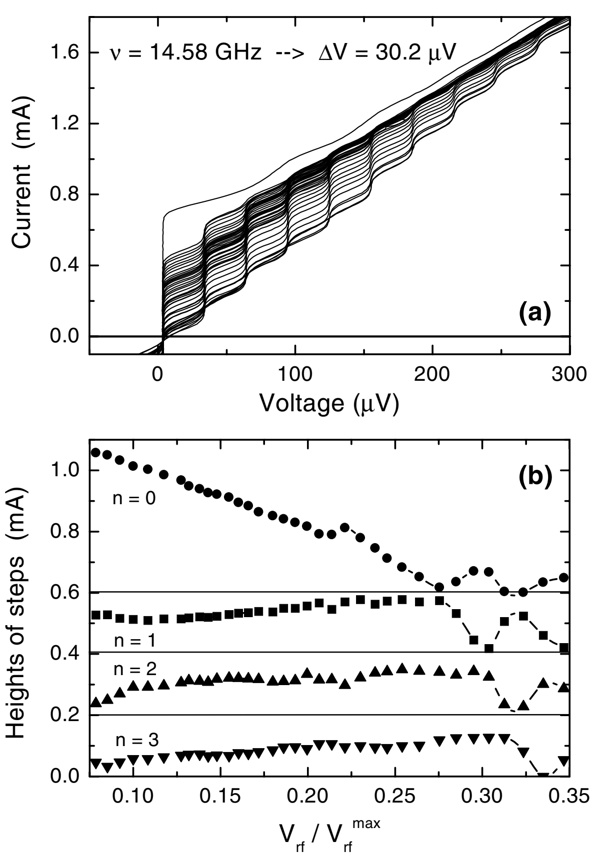

FIG. 4. (a) Shapiro steps of a break junction in the presence of external microwave radiation at $\nu = 14.58$ GHz at different power levels. The upper curve is at zero MW power; (b) magnitude of the supercurrent and the first three current steps as a function of the normalized rf voltage. The zero of the curves for $n = 0, 1, 2$ has been shifted for clarity.

In conclusion, the first experimental evidence of the Josephson effect in MgB_2 is given by using the break-junction technique. This result is a direct proof of the existence of pairs with charge $2e$ in this new metallic superconductor. The product $I_c R_N$ of our junctions is in many cases comparable to the value obtained by the present Nb-based junction technology (up to $\approx$2 mV), and critical currents up to 8 mA are observed. The temperature dependency of the critical current gives evidence for a S-N-S long weak link nature of the junctions and for a depressed T_c in the region of the contact.

We thank A. Barone, A. Bianconi, and A. Perali for useful discussions, and D. Andreone for MW instrumentation. One of the authors (V. A. S.) acknowledges partial support by the Russian Foundation for Basic Research (Grant No. 99-02-17877) and by the Russian Ministry of Science and Technical Policy within the program "Actual Problems of Condensed Matter Physics" (Grant No. 96001).

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