Josephson effect in MgB$_2$ break junctions

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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 non-hysteretic DC Josephson effect. By irradiating the junctions with microwaves we observe clear Shapiro steps spaced by the ideal $\Delta V$ 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 prove the conventional nature of the pairs in MgB$_2$ and give evidence of the superconductor-normal metal-superconductor weak link character of these junctions.

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The discovery of superconductivity at 39 K in magnesium diboride 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 $\Delta$ can also be estimated. Many papers have recently appeared in literature concerning the determination of $\Delta$ from tunneling, Andreev reflection, angle resolved photoemission spectroscopy (ARPES), specific heat, and Raman measurements. The results are still controversial: $\Delta$ ranges between 2 and 7 meV in the different experiments but its temperature dependence seems to approximately follow the BCS behaviour. Various authors have reported the possible presence of a depressed superconducting layer on the MgB$_2$ surface.

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 $I_c R_N$ (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 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. 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 (2 x 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 $\mu$m) single-crystal-like grains, with dark specular 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 gauss 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 $T_c = 38.8$ K with a transition width $\Delta T_c = 0.5$ K as determined from the derivative of the real part of the susceptibility. The 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 ($0.5 \times 0.5 \times 2$ mm$^3$) which were then fixed to an insulated elastic support by means of eposidic 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 $\Omega$. 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 non-hysteretic Josephson junctions with normal resistances between $\sim 0.1$ and 11 $\Omega$. 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 $I_c R_N \approx 1$ $\text{mV}$ is obtained. This value is very small if compared to the value $I_c R_N = \frac{\pi \Delta/2}{\hbar} = 3.52 \times 9.3$ $\text{mV}$ predicted by the BCS theory for $T_c \approx 39$ K.

By applying to this junction a microwave (MW) excitation at $\nu = 8.843$ GHz, clear Shapiro steps spaced by the theoretical value $\Delta V = \hbar \nu/2e \approx 18.3$ $\mu$V were observed, whose heights modulated at the change of the MW power. These current steps were observed at voltages up to 1.1 $\text{mV}$, and for some values of the MW power sub-multiples Shapiro steps at $V = n\nu/4e$ 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 $\text{mW}$). 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, 15, 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 $I_c R_N$ value remains small (0.3-1.7 $\text{mV}$) and all the $I - V$ curves present the same features (included 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 $\sim 11$ $\Omega$) and a smaller zero-bias current ($I_c \sim 0.3 - 2.5$ mA). In these junctions the product $I_c R_N$ reaches the largest values (3.2 - 3.8 $\text{mV}$) and the $I - V$ characteristics are still non-hysteretic, but present some differences with respect to the previous ones.
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).

to those of the first kind. In particular an evident knee is present at $|V| \approx 4 - 5 \text{ mV}$, that we interpret as 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 $\Delta \approx 1.7 - 2 \text{ meV}$ can be obtained. Unfortunately, applying the MW excitation to the junctions with higher $I_c R_N$ 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 only analyze 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. Fig. 3(a) shows this dependency between $T = 5.28 \text{ K}$ and the critical temperature of the junction $T^c_c \approx 26 - 27 \text{ K}$ 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 [10] 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 $T = 5.28 \text{K}$ (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 $T^c_c = 26.4 \text{ K}$ reported in the inset of the same figure (solid circles) clearly shows that, at $T \approx T^c_c$, $I_c \propto (T^c_c - T)^2$. 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 [14] gives good results at $22 \text{ K} < T < T^c_c$ but completely fails at low temperature.

A temperature behaviour 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 [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 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 $T \approx T^c_c$ which best agrees with our experimental data. It was calculated by using $T^c_c = T^c_c = 26.4 \text{K}$ and the corresponding BCS gap $\Delta = 4 \text{ meV}$, and adjusting the value of $L/\xi_N(T_c)$ to get the best fit (obtained for $L/\xi_N(T_c) = 9.9$). Nevertheless, other values of the parameters, ranging between $\Delta = 1.7 \text{ meV}$, $L/\xi_N(T_c) = 7.85$ and $\Delta = 4.2 \text{ meV}$, $L/\xi_N(T_c) = 10$, give theoretical curves in good agreement with the experimental data at $T > 0.9 T^c_c$.

The present theory for SNS weak link structures is valid only for a superconductor in the dirty limit. This condition is not fulfilled in the bulk MgB$_2$ since $\ell = 600 \text{Ang}$ and $\xi \approx 50\text{A}$ [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 $T^c_c$ with respect to the bulk $T_c$ of MgB$_2$ indicates a local depression of the superconducting properties in the
Explanations are possible for this low reduction of bank material with a possible increase of disorder and a reduction of $\ell$ in the weak link region. At least two explanations are possible for this low $T_j^0$ 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 a clear evidence of a reduction of $T_j^0$ 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 Fig. 4(a) and (b) we present in greater detail the AC Josephson effect results. Fig. 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 $\Omega = h \nu / 2e R_N I_c = 0.19$. 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.[14]

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 prove for the existence of pairs with charge $2e$ in this new metallic superconductor. The product $I_c R_N$ of our junctions is small if compared to the BCS value but critical currents up to 8 mA were 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.

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