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Field-induced quantum breakdown of superconductivity in magnesium diboride

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Abstract

The quantum breakdown of superconductivity (QBS) is the reverse, comprehensive approach to the appearance of superconductivity. A quantum phase transition from superconducting to insulating states tuned by using nonthermal parameters is of fundamental importance to understanding the superconducting (SC) phase but also to practical applications of SC materials. However, the mechanism of the transition to a nonzero resistive state deep in the SC state is still under debate. Here, we report a systematic study of MgB₂ bilayers with different thickness ratios for undamaged and damaged layers fabricated by low-energy iron-ion irradiation. The field-induced QBS is discovered at a critical field of 3.2 Tesla ($=H_c$), where the quantum percolation model best explains the scaling of the magnetoresistance near H_c . As the thickness of the undamaged layer is increased, strikingly, superconductivity is recovered from the insulating state associated with the QBS, showing that destruction of quantum phase coherence among Cooper electron pairs is the origin of the QBS.

Introduction

Disorder in materials is undesirable because it prevents investigations of the intrinsic properties of the material. In correlated superconductors, however, disorder can be useful in manipulating superconducting properties. Even though the superconducting (SC) transition temperature (T_c) is decreased with a moderate level of disorder, the current-carrying capacity in high magnetic fields can be improved because disordered regions prevent vortex creep^{1,2}. Strong disorder, however, breaks the coherence of SC electron pairs and gives rise to a change from the SC ground state to an insulating or non-SC metallic state at the quantum breakdown of superconductivity (QBS)^{3–14}. As disorder increases, superconducting islands may appear in the destroyed background of non-SC regions owing to inhomogeneous suppression of superconductivity^{3–6,11}. Because the presence of SC islands and

the phase coherence among them are important to the emergence of superconductivity, numerous experimental and theoretical efforts have been expended to understand the role of disorder in the SC state^{3–14}. However, systematic control of disorder in correlated superconductors has been technically difficult.

Ion beam irradiation is one approach that provides engineered disorder in SC materials. For example, low-energy ion irradiation produces atomic lattice displacements in crystalline materials because elastic scattering between incident ions and atoms in the target material is dominant^{15–17}. In addition, the tunability and simplicity of this technique make the localization of Cooper pairs in disordered SC systems easier to control. The quasi-two-dimensional superconductor MgB₂ is an ideal example for this study because of its high T_c of 40 K and the possibility of localizing Cooper pairs near the QBS in disordered MgB₂^{18,19}.

In this work, we report magnetic-field-induced quantum breakdown of superconductivity in MgB₂ thin films via irradiation with 140-keV Fe-ion beams. The metallic characteristics of MgB₂ were significantly suppressed after

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low-energy ion irradiation. When subjected to a magnetic field, the irradiated MgB_2 films revealed QBS at a critical field H_c of 3.2 Tesla. The superconductor-to-insulator transition (SIT) within the SC state indicates that SC islands are formed in the MgB_2 film due to irradiation and that phase coherence between them is destroyed by the applied magnetic field. When the irradiated MgB_2 layer (S_D) is in contact with a pristine, undamaged MgB_2 layer (S_S), the SIT-like behavior disappears. Here, the two layers comprise a S_D/S_S bilayer. Suppressed superconductivity in the damaged S_D layer was gradually restored to that of its pristine state as the thickness of the S_S was increased, revealing a giant superconducting proximity effect (GSPE). These discoveries suggest that SC islands embedded in the normal matrix are the origin of GSPE and QBS in magnesium diboride bilayers and should provide valuable insights into the development of SC junctions and their applications.

Materials and methods

MgB_2 thin films were fabricated using a hybrid physical-chemical vapor deposition (HPCVD) method, which is an effective technique for fabricating high-quality MgB_2 thin films^{20,21}. For Fe-ion irradiation, *c*-axis-oriented MgB_2 thin films with various total thicknesses (*t*) of 215 (MB215nm), 440 (MB440nm), and 600 nm (MB600nm) were fabricated on *c*-cut Al_2O_3 substrates at a growth temperature of 680 °C, a pressure of 100 Torr and flow rates for H_2 and B_2H_6 of 100 and 50 sccm, respectively. The fabrication and quality of the films were described in detail in previous studies^{20,22}.

Fe ions with an energy of 140 keV were used to irradiate MgB_2 thin films at the Korea Multi-purpose Accelerator Complex (KOMAC) at room temperature. Samples MB215nm, MB440nm, and MB600nm were placed together in a sample holder for irradiation. The mean projected ion range (R_p) and the thickness t_D of the irradiated MgB_2 thin films were simulated with Monte Carlo simulations. The Stopping and Range of Ions in Matter (SRIM) (The projected range of Fe ions and the damage events in the MgB_2 thin film were calculated using the SRIM software (<http://www.srim.org/>), a MgB_2 target density of 2.57 g/cm³ and averaged displacement threshold energy values of 20 eV (Mg) and 46 eV (B) were used²³. As the total damage and the damage profile depend on dose level for the same incident energy, different dose levels were used to obtain various t_D values ranging from 126 to 203 nm (see Figs. S1 and S2 in SI).

Changes in the *c*-axis lattice constants of MgB_2 layers damaged by Fe-ion irradiation were investigated by X-ray diffraction (θ -2 θ scan) before and after irradiation. Values of T_c for the MgB_2 bilayers before and after irradiation were determined by using the temperature dependence of electrical resistivity (ρ), as obtained using a Physical

Property Measurement System (PPMS 9 T, Quantum Design). Bulk superconductivity was evaluated by using the zero-field-cooled (ZFC) dc magnetization (*M*) obtained with a Magnetic Property Measurement System (MPMS 5 T, Quantum Design). A standard four-probe method was used for electrical-resistivity measurements. Measurements of the magnetic field dependence of resistivities of the MB215nm bilayers with $\gamma_t = 0.12$ and 0.06 were performed using the PPMS. The temperature dependences of the zero-bias conductances (ZBCs) of pristine MB600nm and the bilayer with $\gamma_t = 1.96$ were measured in various magnetic fields by soft point-contact spectroscopy (SPCS) in the PPMS (14 T, Quantum Design). Soft-point contacts on the surface of MB600nm were made with a gold wire (diameter: 30 μm) imbedded in a drop of Ag paint at the end nm. The total contact diameter of the Ag paint on the film surface was ~50–100 μm , where thousands of parallel nanoscale junctions were assumed to exist between individual Ag particles and the film surface for SPCS²⁴.

Results and discussion

Figure 1a schematically illustrates the effects on a pristine crystal caused by low-energy iron-ion irradiation. Because elastic scattering of the incident ions by nuclei in the materials is dominant for low-energy ion irradiation, lattice displacements, together with the formation of vacancies and interstitials, take place in the irradiated crystal, leading to changes in SC critical properties^{15–17}. To probe tunable SC properties by introducing lattice disorder, we used low-energy iron-ion irradiation to fabricate MgB_2 bilayers composed of S_D (damaged MgB_2 layer) and S_S (undamaged MgB_2 layer), as depicted in Fig. 1b. The superconductivity of S_D was destroyed by the disorder produced from ion irradiation, whereas the superconductivity of S_S was maintained as that of the pristine state because it was unaffected by irradiation.

Figure 1c shows a representative enlarged view near the (002) peaks of X-ray diffraction (XRD) patterns for MgB_2 with a thickness of 600 nm (see Fig. S3 in SI). The (002) peak splitting indicates the separation of films into two layers after ion irradiation, and the peak position for the irradiated part of the film shifted to a smaller angle as the dose of irradiating Fe ions was increased; this corresponded to an increase in the *c*-axis lattice constant, as presented in Fig. 1d. The appearance of a secondary peak at a lower angle reflects the formation of MgB_2 bilayers owing to the separation of damaged and undamaged MgB_2 layers. All MgB_2 films with thicknesses of 215, 440, and 600 nm, which are identified as MB215nm, MB440nm, and MB600nm, respectively, show the same dose dependences of the *c*-axis lattice constant and changes in the *c*-axis lattice constant Δc , which are plotted as the left and right ordinates of Fig. 1d, respectively. The

Δc values calculated from the position of the second (2nd) peak as a function of the dose are similar for all samples regardless of the thicknesses of the films, showing that the damaged MgB₂ layers in all the films have similar degrees of disorder produced by irradiation.

Figure 2a–d contains plots of the electrical resistivities (ρ) of MgB₂ films with a total thickness of 215 nm as a function of temperature for the pristine layer and the bilayers (S_D/S_S) with $\gamma_t = 0.54$, 0.30, and 0.06, respectively (see Fig. S4 in SI). Here, γ_t is the ratio ($\gamma_t = t_S/t_D$) between the thickness t_S of S_S and thickness t_D of S_D layers, and the value of $\rho(T)$ is normalized by the resistivity value (ρ_n) at the onset temperature of the SC transition for each sample. The $\rho(T)$ for pristine MB215nm decreased with decreasing temperature, exhibiting metallic behavior. As the thickness ratio γ_t decreased, the metallic characteristic was suppressed owing to an increase in the relative thickness of disordered layer S_D . For the bilayer with $\gamma_t = 0.06$, $\rho(T)$ reached a minimum near 112 K and increased with further decreases in temperature, exhibiting

insulating behavior. Below 40 K, $\rho(T)$ began to decrease because of the SC islands formed in the irradiated films.

The dependence on the thickness ratio γ_t of the SC volume fraction of the MgB₂ bilayers is presented in Fig. 2e–h, which shows the zero-field-cooled (ZFC) and field-cooled (FC) dc magnetizations measured at 5 Oe for all samples (see Fig. S5 in SI). The SC volume fraction of pristine MB215nm at 2 K was assumed to be 100%, while that of bilayers with different values of γ_t was estimated relative to pristine MB215nm. As γ_t decreased, the SC volume fraction decreased, and the SC transition width of the ZFC $M(T)$ curve broadened because the magnetic field easily penetrated into disordered MgB₂ SC regions. When the value of the thickness ratio γ_t was 0.06, interestingly, the ZFC and the FC $M(T)$ for the bilayer showed clear separation at quite high temperatures even though the SC volume fraction was < 0.1%. Taken together with $\rho(T)$, the small volume fraction of $M(T)$ resulting in the film with $\gamma_t = 0.06$ is suggestive of the formation of local SC regions in the damaged MgB₂ layer.

Superconductivity in disordered SC systems is not suppressed homogeneously, but disorder-induced inhomogeneity does occur, and local SC regions can be formed. The magnetic field, one of the nonthermal control parameters that can introduce a quantum phase transition at zero Kelvin, is expected to be effective in suppressing phase coherence between local SC regions, i.e., SC islands, thus driving quantum breakdown of superconductivity in disordered SC thin films^{3–6,11–14,25–28}. Figure 3a, b presents $\rho(T)$ of MgB₂ bilayers with a total thickness ($t = t_D + t_S$) of 215 nm for $\gamma_t = 0.12$ and 0.06 , respectively, with several magnetic fields applied perpendicular to the film plane. Evidence for a magnetic-field-induced QBS was observed for both bilayers, wherein $\rho(T)$ increased with decreasing temperature below the SC transition temperature, T_c . At 0 Tesla, the resistivity gradually decreased with decreasing temperature owing to the superconducting transition in SC islands and the weak correlation between SC islands for $T < T_c$. As the applied magnetic field was increased, the rate of decrease in $\rho(T)$ became weaker because of the suppression of phase coherence between SC islands. At 3 T, $\rho(T)$ was almost constant, showing a plateau behavior. With a further increase in the field, $\rho(T)$ increased with decreasing temperature, reflecting insulating behavior due to the destruction of interisland coupling (see Fig. S6 in SI). The magnetic field dependences of the resistivities are plotted for various temperatures in the insets of Fig. 3c, d. A crossover from an SC state to an insulating-like state is observed at the critical field (H_c) near 3.2 T for both bilayers, indicating that the SC islands were weakly

coupled for $0 \leq H \leq H_c$ but electrically isolated for $H_c < H \leq H_{c2}$.

The main panels of Fig. 3c, d reveal scaling of the resistivity as a function of $|H - H_c|/T^{1/z\nu}$ on a semilogarithmic scale. The dynamic exponent z is determined by a characteristic energy $\Omega \propto \xi^{-z}$, where the SC correlation length $\xi(H) \propto |H - H_c|^{-\nu}$. The best scaling was observed when the exponent product $z\nu$ was $7/3$, which is consistent with quantum percolation, indicating that quantum breakdown of superconductivity at H_c occurred owing to the destruction of phase coherence between SC islands in disordered MgB₂ films^{3,12,13,29} (see Fig. S7 for classical percolation results in SI). We note that the critical resistance at the crossing point of magnetoresistance H_c was considerably smaller than the predicted quantum resistance ($R_Q = h/4e^2 = 6.45$ k Ω) at the quantum critical point^{3,5,29,30}, indicating the possibility of an anomalous metallic phase in disordered MgB₂ thin films^{3,10,30–35}. A large charge carrier density and fermionic (unpaired electrons) excitations have been proposed for the origin of unusual metallic behavior and small critical sheet resistance^{33–35}. The fact that MgB₂ has a relatively large charge carrier density³⁶ indicates that the anomalous metallic phase in disordered MgB₂ thin films could be associated with contributions from a large number of unpaired electrons to the background conduction bath. However, further studies are required to understand the QBS in quasi-2D MgB₂.

Figure 4a shows the γ_t dependence of the SC transition temperature ($T_{c,BL}$) of MgB₂ bilayers, where $T_{c,BL}$ was normalized by the T_c of the corresponding pristine MgB₂

thin film ($T_{c,\text{pri.}}$) (see Figs. S8 and S9 in SI). The $T_{c,\text{pri.}}$ values of MB215nm, MB440nm, and MB600nm were 39.3, 39.7, and 40 K, respectively. As t_s increased, $T_{c,\text{BL}}$ initially increased rapidly and saturated to $T_{c,\text{pri.}}$ even though the thickness of the damaged layer, t_D , was considerably larger than the coherence length of MgB_2 ($\xi_{\text{MgB}_2} \sim 7$ nm). The presence of proximity effects up to the surface of S_D was evidenced by SPCS (see Figs. S10 and S11 in SI). Although the behavior of $T_{c,\text{BL}}$ with respect to the thickness ratio χ_t was similar to results for the proximity effect in N/S bilayers, the length scale of the proximity effect in S_D/S_S was considerably larger than the value predicted using conventional theory^{37,38}. Here, the red solid line in Fig. 4a was obtained from the Werthamer theory in which the spatial variation in the BCS electron–electron interaction is considered³⁷.

The long-range proximity effect, the so-called giant superconducting proximity effect (GSPE), with an anomalously

large leakage distance for Cooper pairs was often observed between two superconductors composed of the same materials but with different T_c values^{39–44}. For example, when an optimally doped $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ (LSCO) layer with $T_c \approx 45$ K was in contact with an underdoped $\text{La}_2\text{CuO}_{4-d}$ (LCO) layer with $T_c \approx 25$ K, GSPE was observed at temperatures higher than the T_c of LCO⁴¹. Several scenarios, such as phase fluctuations, amplitude fluctuations, and proximity-induced interface superconductivity, were proposed to explain the origin of the GSPE^{43,45–48}. Figure 4b is a simple cartoon used to describe the GSPE in MgB_2 bilayers (S_D/S_S), and the unusual proximity length scale may be understood by the presence of spatially distributed SC islands in S_D . Phase coherence between the SC islands in S_D can be enhanced by leaking of Cooper pairs from S_S , sequentially forming strong Josephson-junction chains between the SC islands. The enhanced length scale from the proximity effect, in turn, gives rise to the suppression of the QBS in S_D ^{25,26,46}. These findings underscore

Fig. 4 Giant superconducting proximity effect in the MgB₂ bilayer. **a** The SC transition temperature ($T_{c,BL}$) of MgB₂ bilayers as a function of γ_t , where $T_{c,BL}$ is normalized to the T_c of the corresponding pristine MgB₂ thin film ($T_{c,pri.}$). Square, circular, and triangular symbols represent the three films with different thicknesses of 215 nm (MB215nm), 440 nm (MB440nm), and 600 nm (MB600nm), respectively. The thickness t_D of layer S_D (126–203 nm) was controlled by varying the dose of irradiated Fe ions. The red solid line is a fitting curve obtained by using the Werthamer theory that considers the spatial variation in the attractive electron–electron interaction. **b** Schematic view of the GSPE in the bilayer S_D/S_S . SC islands (SCI) in the S_D layer sequentially form Josephson-junction chains between the SC islands. Phase coherence between the SC islands is enhanced by Cooper pairs that leak from the S_S layer, which can lead to a power-law decay of the order parameter as a function of the distance from the S_D/S_S boundary, $\Delta(x) \propto 1/x$, rather than an exponential decay of $\Delta(x) \propto \exp(-x/\xi_D)$. Here, ξ_D is the leaking distance of the order parameter to the S_D layer from the boundary of the S_D and S_S layers. The regular array of SC islands is exaggerated for simplicity, and the spacing between them could be larger than the coherence length of MgB₂.

that SC islands formed in the normal matrix are the origin of the GSPE in the MgB₂ bilayers composed of a damaged layer (S_D) and an undamaged layer (S_S).

Conclusion

In conclusion, we observe field-induced quantum breakdown of superconductivity in disordered MgB₂ and a local pairing-induced GSPE in S_D/S_S MgB₂ bilayers

fabricated with low-energy ion irradiation. When an applied magnetic field is sufficiently high to break phase coherence among SC islands in S_D , QBS is observed at a critical field H_c , the scaling of which is consistent with that of the quantum percolation model. As the thickness of S_S is increased, the suppressed superconductivity of S_D is recovered to that of the pristine state, and the QBS does not occur, not even at fields larger than H_c . Taken together, these findings underpin the conclusion that local superconducting pairing is the origin of QBS and the GSPE in the MgB_2 bilayer.

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