Role of interband scattering in neutron irradiated MgB_2 thin films by scanning tunneling spectroscopy measurements

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A series of MgB_2 thin films systematically disordered by neutron irradiation have been studied by scanning tunneling spectroscopy. The c-axis orientation of the films allowed a reliable determination of the local density of states of the π band. With increasing disorder, the conductance peak moves towards higher voltages and becomes lower and broader, indicating a monotonic increase of the π gap and of the broadening parameter. These results are discussed in the framework of two-band superconductivity.

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The discovery of two gap superconductivity in MgB_2 (Refs. 1 and 2) has renewed the interest in this mechanism, which has been theoretically predicted since the 1950s. This peculiar feature occurs in MgB_2 because of the presence of two sets of bands which cross the Fermi level. The larger gap ($\Delta_\sigma \sim 7$ meV) is related to the σ bands, originated by p_{xy} B orbitals, strongly interacting with phonons, and characterized by a bidimensional Fermi surface; the smaller gap ($\Delta_\pi \sim 2$ meV) is related to the π bands originated by p_z B orbitals and having an isotropic Fermi surface. Two-band models predict that nonmagnetic scattering causes pair breaking as the magnetic one does in a one-band superconductor;[3] this is a unique effect of interband scattering, because mixing σ and π Cooper pairs causes a complete isotropization of the entire Fermi surface. In the strong interband scattering limit, the two gaps should merge to one when the critical temperature (T_c) drops to the isotropic value of about 20–25 K.[4,5] To verify these predictions, several efforts have been made to evaluate the energy gaps in samples where defects were introduced by different techniques such as substitutions [Al (Refs. 6–8) or C (Refs. 9–12)], irradiation,[13,14] or in films grown naturally disordered.[15] Due to the possible simultaneous occurrence of several sources of disorder, quantitative evidence of the role of interband scattering has not yet been given. In particular, substitution may induce extrinsic effects related to charge doping, structural instability, and inhomogeneous distribution of impurities. However, it is widely accepted that the Δ_π value is weakly affected by disorder while Δ_σ decreases linearly with T_c, and recently the merging of the gap has been observed in irradiated samples at a critical temperature (11 K) lower than the predicted one.[14]

In this paper, we report on scanning tunneling spectroscopy (STS) measurements on c-axis-oriented thin films where the defects were systematically introduced by neutron irradiation. Neutron irradiation introduces neither charge doping nor electronic or structural instabilities; furthermore, in thin films it guarantees a uniform distribution of defects in the entire sample.[16] STS directly probes the local density of states (LDOS), whose broadening can be analyzed in connection with scattering mechanisms induced by irradiation.

MgB_2 thin films (2000 Å thick) were grown at the Pennsylvania State University by hybrid physical chemical vapor deposition (HPCVD) on 5×5 mm^2 SiC substrates. Details about the samples fabrication and their basic characterization can be found in Ref. 17. Neutron irradiation was carried out at the spallation neutron source SINQ of Paul Scherrer Institut in Villigen. Each film was sealed under high vacuum (10^{-5} mbar) in a small quartz ampoule just after deposition; quartz tubes were kept in the oven at 600 °C for 4 days before we put the film inside, in order to minimize the presence of impurities. This setup allowed us to perform irradiation also for long times without compromising samples quality. Several samples (samples IRR10, IRR30, IRR35, and IRR40 in the following) have been irradiated at thermal neutron fluences ranging from 6.4×10^{15} to 9.5×10^{18} cm^{-2}. A description of the damage mechanism, as well as detailed structural, transport, and magnetic characterization, was reported elsewhere.[16] Low-temperature STS experiments were performed in a cryogenic system able to operate at variable temperature and in magnetic field, using a PtIr homemade tip. The films were mounted on a scanning tunneling microscope scanning head in inert helium atmosphere to preserve the surface quality.

Table I reports the main features of the measured samples: as the fluence increases, the T_c value changes from 41 K to 14.5 K and the residual resistivity ρ_0 varies by two orders of magnitude. The superconducting transitions of the four irradiated films are reported in Fig. 1. The amplitude of the transition is extremely small in less irradiated samples (less than 0.5 K) and increases in IRR40.

Figure 2 shows the typical normalized tunneling conductance STS spectra for neutron irradiated films at different fluences. All the spectra were collected through a standard lock-in technique and were acquired (at $T=4.2$ K) by stabilizing the feedback loop with a tunnel current of 100 pA and a bias voltage of 20 mV. In the figure, each curve represents a single STS spectrum per film, representative of all the

TABLE I. Main properties of the measured irradiated films. The resistivity values have been normalized following the criterion proposed by Rowell in Ref. 18 [$\rho_0=\rho_{expt}\Delta\rho_{theor}/\Delta\rho$, with $\Delta\rho=\rho(300\text{ K})-\rho(42\text{ K})$] to $\Delta\rho=7.6\ \mu\Omega$ cm. The transition widths are defined as the amplitude between 10% and 90% of normal-state resistivity.

Sample	Neutron fluence (cm^{-2})	T_c (K)	ΔT_c (K)	ρ_0 ($\mu\Omega$ cm)
IRR10	6.4×10^{15}	41.0	0.1	0.9
IRR30	7.7×10^{17}	36.1	0.4	12
IRR35	3.0×10^{18}	22.2	1.9	36
IRR40	9.5×10^{18}	14.5	4	55

curves collected on the surface. The STS spectra on each sample were reproducible from point to point on the sample surface, and they overlapped when normalized by changing the tunnel resistance. This assures that we were measuring in the pure tunneling regime and guarantees a reduced role of possible contaminated surface layers. STS measurements were performed only when such conditions were achieved. A spectrum on a nonirradiated film (T_c=41 K, from Ref. 15), measured in the same conditions, is also reported as a reference.

The plot reveals a clear monotone trend of the spectra. As T_c decreases, with increasing neutron fluences, the zero-bias conductance (ZBC) increases and the coherence superconductivity peaks shift to higher voltages and appear less pronounced. This behavior is more clearly shown in the inset to Fig. 2. Only one-peak spectra were found in all the samples. Because in c-axis-oriented films the current is injected parallel to the c axis, the π-band contribution is dominant in the tunneling spectra as compared to the σ-band contribution.[19] Therefore, we identify the observed peaks as corresponding to the π gap, Δ_π.

The increase of the ZBC is likely due to a broadening of the superconducting LDOS, as a consequence of the increasing disorder, which introduces subgap states and smears the LDOS divergence. The displacement of the peaks towards higher energies could be ascribed mainly to two effects: the LDOS broadening, supported by the simultaneous decrease in the peak height, and an intrinsic change of the Δ_π value as disorder increases and T_c decreases.

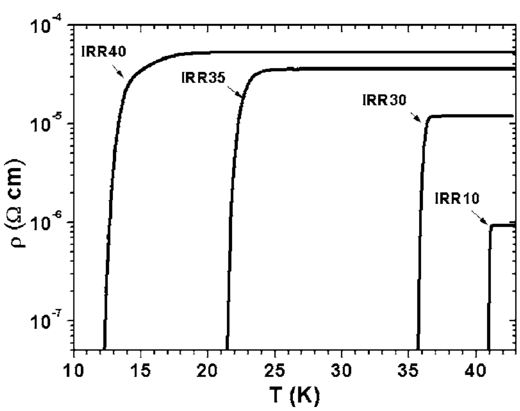

FIG. 1. Superconducting transitions for the four measured irradiated samples.

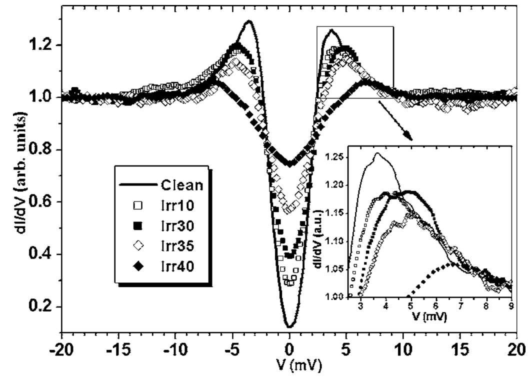

FIG. 2. STS spectra, at T=4.2 K, on the irradiated measured MgB_2 films. Measurement parameters: tunnel current, 100 pA; bias voltage, 20 mV. A spectrum on a clean sample, from Ref. 15, is also reported as reference.

Measurements in magnetic field perpendicular to the films surface were also performed. Figure 3 reports the ZBC as a function of the applied field for some samples. The plot shows that the most irradiated samples are much less sensitive to the magnetic field, as already observed in differently disordered samples.[15] This result will be discussed in detail elsewhere.

To get further information on the evolution of LDOS as a function of irradiation-induced disorder, a quantitative analysis can be developed. As a first approximation, we fitted the measured dI/dV spectra through a simple (single-gap) BCS model with a phenomenological Dynes parameter Γ_D.[20] In fact, when only the π band gives a significant contribution to the tunnel current, the two-band calculation for MgB_2 approaches a simple one-band BCS model,[19] with Δ_π and Γ_D as fitting parameters. The Γ_D parameter includes all the measured broadening on the superconducting LDOS, due to intrinsic, pair-breaking, effects, but also to extrinsic effects such as external noise. The results of the best fit curves and fitting parameters are shown in Fig. 4.

The good agreement between the fits and data in Fig. 4 confirms the adequacy of the one-band model. Although the high Γ_D value estimated for the lowest-T_c sample seriously affects the quantitative reliability of the simple model, it fur-

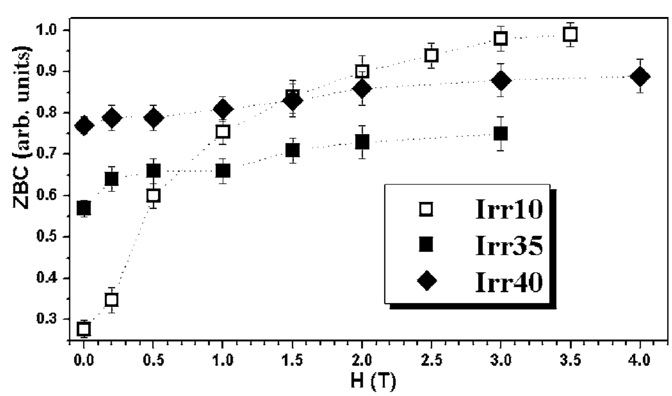

FIG. 3. Zero-bias conductance as a function of the applied magnetic field (perpendicular to the surface) for samples Irr10, Irr35, and Irr40.

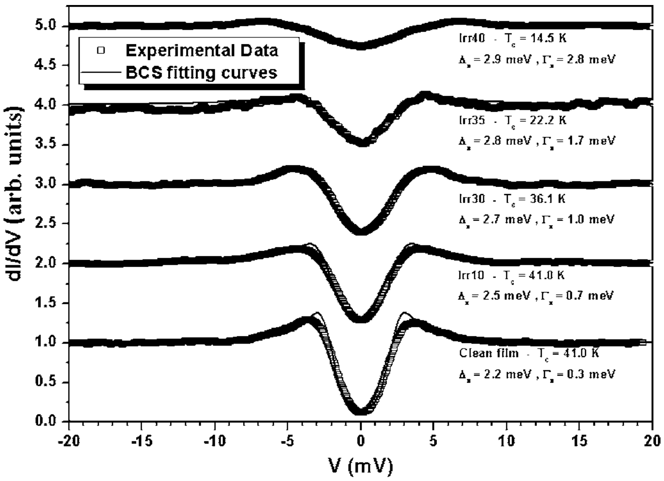

FIG. 4. STS spectra, at T=4.2 K, on the irradiated measured MgB_2 films; fitting curves, evaluated by a BCS one-gap model, are superimposed to the experimental data. Plots are vertically shifted for clarity.

nishes anyway an indication of the film behavior. The use of a two-band model, with the estimated σ gap from the specific heat data on bulk samples irradiated through the same technique,[14] does not change the extracted π-gap value substantially, but introduces more free parameters, making the overall analysis less straightforward and clear.

Figure 5 shows Δ_π values as a function of T_c measured from our STS spectra. With decreasing T_c, Δ_π slightly increases. Δ_σ and Δ_π evaluated by specific heat measurements on neutron-irradiated polycrystalline samples[13,14] are also plotted for comparison. Differences between gap values estimated by the two techniques are expected because the specific heat is a bulk property, while STS probes locally the sample surface. On the other hand, STS gives a direct estimation of the energy gap, which is extracted by specific heat data only through a model-dependent analysis. These reasons

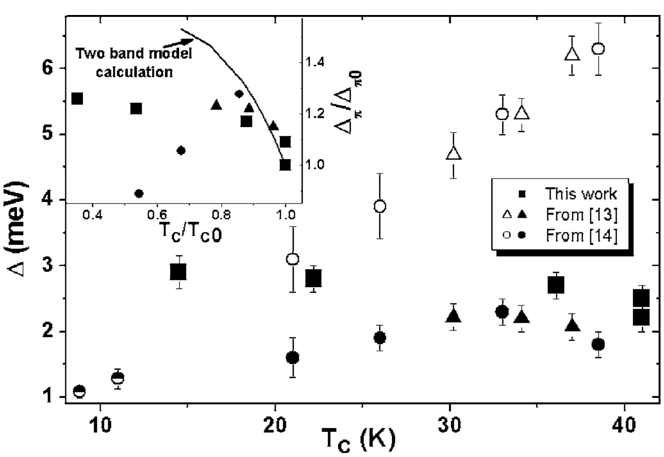

FIG. 5. Δ_π values evaluated by STS spectra as a function of T_c (squares). For comparison, Δ_π and Δ_σ evaluated by specific heat measurements on neutron-irradiated polycrystalline samples (triangles, from Ref. 13, and circles, from Ref. 14) are also reported. In the inset, $\Delta_\pi/\Delta_{\pi 0}$ vs T_c/T_{c0} is plotted together with a theoretical calculation (Ref. 21).

can account for the slightly different Δ_π values obtained by the two techniques for the lightly irradiated samples.

Larger discrepancies are instead present in heavily irradiated thin films (T_c=22 and 14.5 K), for which the extracted Δ_π values from the STS spectra are close to Δ_σ estimated by the specific heat. It is worth noting that these samples have T_c values for which the merging of the gap is expected, even if the merging has been unambiguously seen at lower temperatures.[14] In such disordered samples, interband scattering must be strong, producing a mixing of σ and π Cooper pairs, which largely modifies the density of states.[21] In the STS technique, only quasiparticles with electron momentum parallel to the tunnel direction—i.e., perpendicular to the film surface—contribute to the tunnel current. Therefore, for heavily irradiated c-axis-oriented thin films it is not possible to distinguish whether the single-gap superconductivity is established or we are approaching it. In both cases the extracted gap could be no more related only to the π band, and this circumstance can be also partially responsible for the observed discrepancies.

The increase of Δ_π is predicted by the two-band model for increasing interband scattering.[3,21] Up to now, an experimental proof of this effect has been lacking due to the difficulty in introducing disorder in a controlled way without simultaneous doping effects. With the reliable gap data from the neutron irradiated samples a comparison with the theory is possible. In the inset to Fig. 5, $\Delta_\pi/\Delta_{\pi 0}$ data as a function of T_c/T_{c0} from this work and from Refs. 13 and 14 are shown, where $\Delta_{\pi 0}$ and T_{c0} are the π gap and the T_c values of the unirradiated samples, respectively. The solid line represents the theoretical prediction of the two-band model for increasing interband scattering.[21] For T_c/T_{c0} ranging from 1 to 0.85 ($\Delta T_c = T_{c0} - T_c \sim 6$ K), all the three data series show a similar increase of $\Delta_\pi/\Delta_{\pi 0}$, in agreement with the theoretical curve. This suggests that for low levels of disorder the main mechanism of T_c reduction is the pair breaking due to interband scattering. In the regime of small interband scattering, the scattering rate Γ_{inter} can be calculated as[22] $\Delta T_c/T_{c0} = 0.2\Gamma_{inter}/k_B T_{c0}$; with ΔT_c=6 K, we estimate $\Gamma_{inter} \sim 2.5$ meV.

For $T_c/T_{c0} < 0.85$ the experimental data do not agree; the $\Delta_\pi/\Delta_{\pi 0}$ values of this work are nearly constant and those from the specific heat on bulk samples decrease with decreasing T_c. In both cases, the data remain below the theoretical curve, which continues to increase. Thus for these levels of disorder other mechanisms cause the decrease of Δ_π and consequently the suppression of the T_c.

In Fig. 6 the estimated Dynes broadening parameter Γ_D is reported as a function of ρ_0. It increases linearly with ρ_0 from 0.3 to 2.8 meV. This linear correlation seems to suggest that quasiparticle relaxation processes, which affect ρ_0, could enhance pair-breaking mechanisms which influence Γ_D. A linear increase of Γ_D with resistivity was first observed and discussed in Ref. 23, which suggested that finite-lifetime effects due to inelastic electron-electron scattering vary linearly with resistivity, being enhanced by disorder. In a two-gap superconductor, in principle both intraband and interband events can contribute to the observed scattering processes, which cannot be sorted out from the present data. However, the intraband relaxation rate Γ_{intra} can be roughly

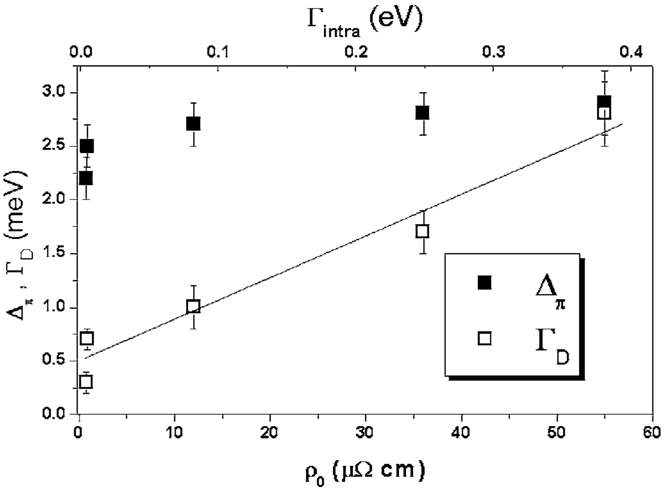

FIG. 6. Δ_π and Γ_D values estimated by tunnel spectra as a function or residual resistivity ρ_0 (lower scale). On the upper x scale, the intraband scattering Γ_{intra} has been plotted, estimated as in Ref. 24 in the hypothesis of equal relaxation rates in σ and π bands. The solid line is only a guide for the eyes.

estimated by ρ_0 assuming equal intraband relaxation rates in σ and π bands.[24] It ranges from 0.1 to 0.4 eV (see upper scale in Fig. 6), much higher than Γ_D. Interestingly Γ_D values are close to Γ_{inter}. For example, the sample with T_c=36 K has $\Gamma_{inter}\sim 1.6$ meV and Γ_D=1 meV. We believe that this agreement is not fortuitous. Indeed, in a two-gap superconductor the interband scattering is expected to cause pair breaking as scattering with magnetic impurities does in single-gap superconductors; therefore, it is reasonable that the interband scattering can strongly influence the broadening of the tunnel LDOS of the π band. Furthermore, this framework implies a linear dependence between interband and intraband scattering rates (suggested by the linear dependence of Γ_D on ρ_0, as discussed above) as both mechanisms scale with the defect density, which increases with the neutron fluence. Since the extrapolated low-resistivity Γ_D value is different from zero, part of the LDOS broadening should be ascribed to extrinsic factors such as finite noise temperature or presence of a contaminated surface layer.

In conclusion, we measured, by STS, neutron-irradiated MgB_2 thin films grown by HPCVD. From tunnel spectra analysis we estimated the behavior of Δ_π and Γ_D as a function of the introduced disorder. At a low level of disorder (T_c lowered from 40 to 36 K) Δ_π increases slightly as an effect of interband scattering, in quantitative agreement with the two-band theory. Furthermore, by comparing the behavior of the Γ_D parameter with resistivity we were also able to discuss the scattering mechanisms and reasonably infer the role of the interband scattering in determining the broadening of the measured density of states.

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