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Matthiessen's rule in MgB_2: Resistivity and T_c as a function of point defect concentration

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We present the results of a systematic study of the temperature-dependent resistivity and of T_c of a single crystal MgB_2 sample as a function of point defect concentration. We have found linear, albeit weak, correlation between the decreasing superconducting critical temperature and the residual resistivity and no variation of the slope of the $\rho(T)$ curve at high temperature. These findings reinforce the already existing picture of *s*-wave pairing for the superconductivity. The interband scattering is low despite increasing disorder. Somewhat surprisingly, Matthiessen's rule is followed even in this two-band metal.

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Magnesium diboride has been extensively studied since the discovery of superconductivity at 40 K in it. There is a wealth of experimental results ranging from tunneling spectroscopy[1] to specific heat measurements,[2] which are very coherently explained in a model with two distinct superconducting gaps, forming in the σ and the π bands in this material. While the pairing mechanism itself is well explained by BCS theory,[3] the material has a number of unusual properties. One of these is the relation between the resistivity and the superconducting transition temperature.

Mazin *et al.*,[4] by comparing a large number of measurements, made two observations: the absence of correlation between the residual resistivity and the critical temperature, on one hand, and a correlation of the high temperature slope of the resistivity curves and the residual resistivity, on the other hand. In order to reconcile these findings, they suggest weak interband scattering, and weak scattering in the σ band as compared to the π band. One consequence of the model is that the high temperature slope of the resistivity curves depends on the residual resistivity, which would be the violation of Matthiessen's rule.

However, the experimental situation is not very clear. Results obtained on thin films, single crystals, ceramics, and doped boron wires had to be compared. Other authors, by studying a somewhat different subset of experimental data, arrive at the conclusion that most of the inconsistencies of the resistivity measurements are extrinsic due to secondary phases, scattering on the grain boundaries, etc.[5]

One way to avoid this problem is to introduce disorder by atomic substitution. It is possible to substitute both the Mg and the B sites with elements such as Zn, Si, Li, Ni, Fe, Al, C, Co, and Mn.[15–17] Although in this case the sample quality is well controlled, besides introducing disorder, the substitutions dope the material, thus changing the electronic system as well.

More recently, several authors have performed measurements on irradiated samples in order to obviate the problem of comparing distinct samples. Wang *et al.*[6] and later Putti *et al.*[7] have used neutron irradiation to create point defects in polycrystalline samples. It is known that neutron irradiation is producing cascades of atomic displacements resulting in clustering of defects. This has induced almost a factor of 100 change in the residual resistivity, and the authors found a linear variation of T_c with it. Specific heat measurements have indicated a change from two-gap to single-gap superconductivity when T_c was suppressed below 20 K.[7]

Our goal was to study the relation between residual resistivity, temperature-dependent resistivity, critical temperature, and defect concentration in the very same single crystal. In our measurements, we used high-quality samples grown by the cubic anvil technique.[8] The resistivity was measured by a four-point method. The contacts were obtained by sputtering gold on the sample, and the wires were glued with silver paste. We introduced homogeneously distributed point defects by fast electron irradiation. The irradiation was performed at the Laboratoire des Solides Irradiés, Ecole Polytechnique in Palaiseau. 2.5 MeV electrons interact rather weakly with the material, and they mostly create interstitial and/or vacancy pairs by head-on collisions with the nuclei. It is considered that these defects do not carry magnetic moment. The displaced atoms are mobile at high temperature, and they can be annealed or they can form clusters of defects. In order to avoid this, we carried out the irradiation at 20 K in a liquid H_2 cryostat. In this way, defect clustering can be avoided and the induced number of point defects is a linear function of the received electron fluence. The electron flux was limited to $3\times10^{14}\ e/\mathrm{cm}^2$ to avoid heating.

Irradiation was interrupted regularly, and the resistivity was measured between 20 and 50 K. This range is reasonably wide to allow the determination of T_c and the residual resistivity, but the temperature is low enough to avoid defect recombination. After a few such cycles, the sample was heated up to room temperature and cooled down again. This has allowed us to compare the temperature dependence of the resistivity in the whole 20–290 K range. In this way, the same single crystal sample with increasing defect concentration could be used, avoiding any artifacts which are inevitable if measurements on different samples are compared.

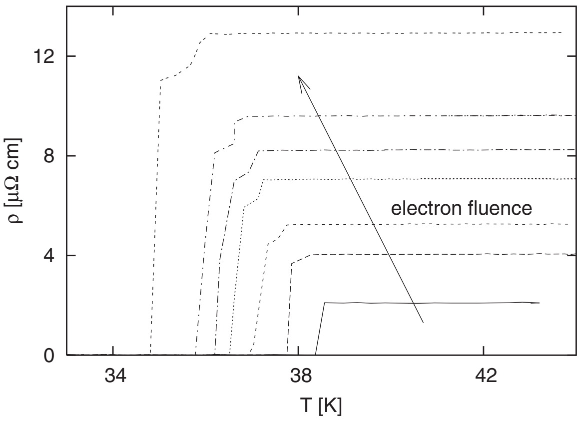

FIG. 1. Resistivity of MgB_2 versus temperature at different electron fluences.

Figure 1 shows $\rho(T)$ curves in the 20–50 K range. Already at the lowest applied fluence, one can observe a doubling of the superconducting transition. By themselves, both transitions are sharp. It is very likely that the phase with higher T_c is due to areas under the contacts, partially protected from radiation. This is why we used the lower temperature transition to determine the residual resistivity and T_c. The residual resistivity was found to vary linearly with electron fluence (see inset on Fig. 2), which is a proof that defects were created independently and that they do not interact. The main panel of Fig. 2 shows T_c as a function of the residual resistivity. This again is a linear function of the electron fluence, the slope of which can give information about the nature of the defects, and when compared to superconductors with known pairing symmetry, it can also give a hint on the pairing symmetry of the gap.

In order to put this result in context, in Fig. 3, we plot the scaled transition temperature and the scaled residual resistivity together with similar curves of superconductors with various order parameters (V_3Si, *s* wave;[9] Sr_2RuO_4, *p* wave;[10] and $YBa_2Cu_3O_7$, *d* wave[11]). In all these compounds, it is considered that the residual resistivity was increased by nonmagnetic defects. The decrease of T_c with increasing residual resistivity indicates pair breaking by the induced scattering centers. According to the theorem of Anderson,[12] in a dirty superconductor with *s*-wave symmetry, nonmagnetic impurities do not reduce the transition temperature, as they do not introduce time reversal symmetry breaking. In a two-band superconductor, the situation is, however, different: interband scattering breaks the time reversal symmetry and should decrease T_c.[13]

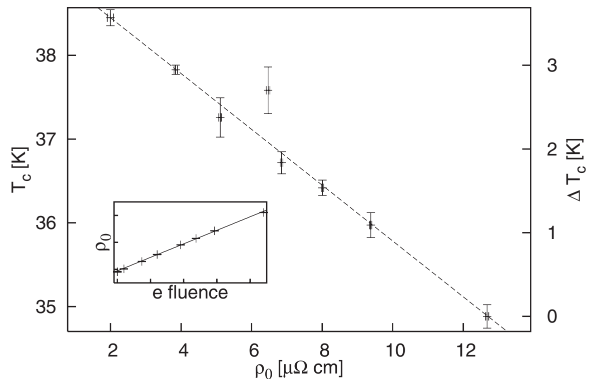

FIG. 2. The critical temperature versus residual resistivity in MgB_2. The inset shows the variation of the residual resistance with electron fluence.

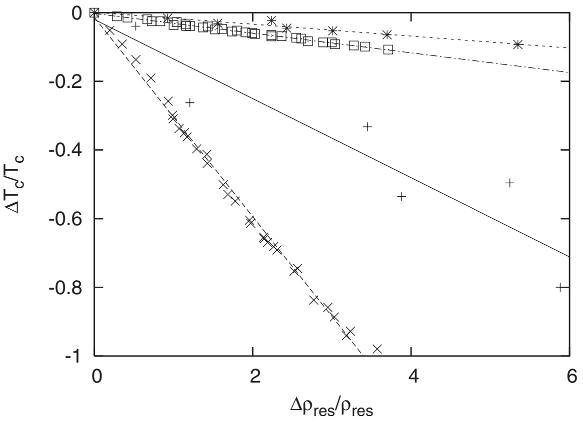

FIG. 3. Normalized critical temperature versus normalized residual resistivity of superconductors with different order parameter symmetries (in order of steepness of the curve: MgB_2, *; V_3Si, ⊡; Sr_2RuO_4, +; $YBa_2Cu_3O_7$, ×). Lines are guides for the eyes.

One can see that despite the two-band nature of the superconductivity, the decrease of T_c is in MgB_2 is even less steep than that observed in V_3Si, a conventional *s*-wave superconductor; this hints the *s*-wave pairing in this material. It also seems to rule out the possibility that at these defect concentrations, increased interband scattering could be made responsible for a stronger decrease of T_c.[14]

In order to check the stability of the electron irradiation induced defect, we warmed up our sample to room temperature (Fig. 4) and cooled it down again. This has allowed us to compare the temperature dependence of the resistivity in the whole temperature range and check Matthiessen's rule. This

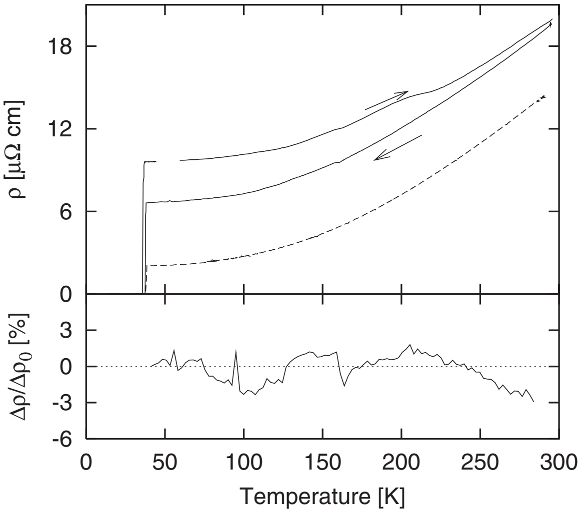

FIG. 4. (Upper) Resistivity versus temperature before and after irradiation. The arrows show the direction of the temperature sweep. (Lower) The difference between the irradiated and the nonirradiated curve as a function of temperature.

rule states that resistivities coming from different mechanisms are superadditive, that is, $\rho \geqslant \rho_1 + \rho_2$. The equality is only satisfied for certain special cases, but in reality, it holds with sufficient precision for simple metals.

It is also well known that impurity scattering is essentially temperature independent. This means that resistivity curves $\rho(T)$ of a simple metal which differs only by impurity scattering are parallel lines. Magnesium diboride is not a simple metal; however, it has two bands, with very little scattering between the two, which results in two almost independent conducting channels.[4] In this case the resistivity can be written as

$$\frac{1}{\rho} = \frac{1}{\rho_0^{(\pi)} + \rho^{(\pi)}(T)} + \frac{1}{\rho_0^{(\sigma)} + \rho^{(\sigma)}(T)}.$$

This form does not allow the separation of a temperature-independent term unless one of the temperature-dependent terms is very large, in which case, that particular channel is switched off. This can happen if the electron-phonon coupling is much stronger in one of the two bands.

The upper part of Fig. 4 shows the $\rho(T)$ curves. It is clear that some of the defects are annealed at room temperature, presumably in the Mg sublattice, but there are still some that remain. The lower part of the figure shows the difference between the partly annealed and the nonirradiated $\rho(T)$ curve. Despite a factor of 3 change in the residual resistivity, within the precision of the measurement, the difference between the two curves is temperature independent; thus, Matthiessen's rule seems to be followed. These two findings can be reconciled with two-band conduction if we assume that point defects do not increase interband scattering, and intraband scattering in the two bands is very different, independent of the impurity concentration.

In conclusion, we have measured the resistivity of a single crystal MgB_2 sample as a function of point defect concentration. We have found linear variation of the residual resistivity with electron fluence, showing the creation of homogeneously distributed point defects. T_c decreased linearly, but very weakly with electron fluence, suggesting s-wave pairing. A partly annealed irradiated sample, when compared to a nonirradiated one, has a $\rho(T)$ curve which is only shifted by a temperature-independent constant in agreement with Matthiessen's rule. This confirms that interband scattering does not play an important role at these levels of irradiation.

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