Extracting the diffusivity ratio from point contact Andreev reflection spectroscopy and upper critical field measurements in MgB₂

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Abstract. The diffusivity ratio η, which measures the relative intraband scattering in the π and σ bands in MgB₂ has been determined by fitting the Hc²(T) at T ∼ Tc and by Point Contact Andreev Reflection. We find a satisfactory agreement between the values for η obtained by both methods for c-axis orientated MgB₂ thin films. Point contact Andreev Reflection was then applied to bulk MgB₂ containing Mg vacancies. Spectra obtained in zero field indicate a distribution of the two gaps ∆σ,π but no merging of the values with increased magnesium deficiency. Spectra fitted as a function of field are consistent with an increase in π intraband scattering with increasing magnesium deficiency. Measurement of the point contact Andreev reflection spectra as a function of temperature revealed features not immediately expected from current theoretical models.

Much progress has been made in understanding the inter and intraband scattering mechanisms in the multiband superconductor MgB₂ [1]. Point contact Andreev reflection spectroscopy (PCAR) in particular has proven to be a very useful tool to probe a large variety of the physical properties of MgB₂ [1, 2, 3, 4]. The multi-gap nature of the material was confirmed [5], interband scattering quantified [1, 3] and the intraband scattering, via the diffusivity ratio, η, estimated [4, 2]. This last parameter is the focus of this paper, as it has been predicted to be responsible not just for the behaviour of the upper critical field, Hc₂, as a function of temperature [6], but also for the zero bias mixed state density of states (DOS) [7, 8, 9].

Gurevich [6] has shown that the Hc₂(T) dependence is intricately tied to the behaviour of η. In this work, we define η as Dσ/Dπ where Di are the diffusivities in the σ, π bands¹. The distinct upturn at low temperatures in Hc₂, predicted to occur with dirtier π bands, (η >> 1) has indeed been observed in thin film samples [10, 11]. In these studies the entire Hc₂(T) curve

¹ In some publications [6, 10], the definition of η is the inverse of that used here. We keep with the notation used in previous PCAR [4, 7] studies to try to minimise confusion.
was fitted to the expressions given in reference [6] to obtain \( \eta \). An independent measure of \( \eta \) can be obtained from the field dependent PCAR spectra [2, 4]. With PCAR, the conductance spectra obtained by using a point contact tip of Au against the MgB\(_2\) sample are fitted, as a function of field, to the two channel adaptation of the BTK model used generally to fit the data [12, 13]. To our knowledge, a direct comparison has not been undertaken on the same samples of the values of \( \eta \) obtained by fitting \( H_{c2}(T) \) data to the model of reference [6] and the two channel fitting of the PCAR data. In order to test this, we compare results of the \( \eta \) determination from PCAR on a well orientated thin film and that obtained by fitting the \( H_{c2}(T) \) at \( T \sim T_c \) of the same film. The PCAR technique was then applied to the measurement of magnesium deficient bulks to determine the effect of magnesium stoichiometry on inter and intra band scattering rates at 4.2 K and as a function of temperature.

The film used in the study was prepared by HPCVD technique onto an Al\(_2\)O\(_3\) substrate as described extensively elsewhere, [14, 15], the film had a \( T_c \) of 39 K. The bulk samples used in this study were made by conventional solid state reaction technique as described in [16, 17]. Two samples were compared, “x=1.0” and “x=1.5”. The values of Mg:B before annealing. Due to the vapour pressure of Mg, the magnesium is expected to evaporate during sample preparation [18], the \( x \) values are therefore nominal. We note that it is difficult to quantify the resultant magnesium level in the samples by XRD and therefore assume that both samples are magnesium deficient, with the \( x = 1.0 \) sample the more Mg deficient. X-ray diffraction indicated that the sample \( x=1.0 \) contained some MgB\(_4\) indicative of a Mg-poor reaction environment.

Measurements of the PCAR as a function of field allows us to fit the spectra in order to extract a parameter \( n_\pi \) that is a measure of the number of normal state conducting channels within the point contact. The measurement and fitting procedures have been fully described in references [2, 4, 12, 17]. It has been shown [4] that this parameter \( n_\pi \) can be directly related to the zero bias density of states (DOS) of the \( \pi \) band, \( N_\pi \), as used in references [7, 8]. Koshelev and Golubov have shown that the behaviour of \( N_{\pi,\sigma} \) is determined by the diffusivity ratio \( \eta \). A comparison of the \( n_\pi \) from the PCAR with the predictions of Koshelev and Golubov, therefore allows the determination of \( \eta \). By fitting the PCAR data of a \( c \)-axis orientated MgB\(_2\) thin film, as described in [4], a value of \( \eta = 0.5 \) was obtained. The \( H_{c2}(T) \) was determined resistively for the same film and is shown in figure 1. Due to the limitations of the magnet used for these measurements, \( H_{c2} \) values greater than 8T could not be quantified. Our data is therefore necessarily restricted to \( T \sim T_c \). The inset to figure 1 shows the anisotropy \( \gamma = H_{ab}/H_c \) as a function of temperature. The relatively strong dependence of \( \gamma \) allows us to be confident that, within the Gurevich model, \( \eta < 1 \). This fact gives us a consistency check for the values of \( \eta \) obtained from either PCAR or fitting to the \( H_{c2}(T) \). The behaviour of the \( \gamma(T) \) immediately shows that the scattering in the \( \sigma \) bands is stronger in these samples than the scattering in the \( \pi \) bands, the opposite of what has been observed in films grown by HPCVD onto SiC [11]. We stress that this observation does not come from fitting any data, but from the behaviour of the \( \gamma(T) \). In order to be more quantitative about the value of \( \eta \) determined from fitting to the model of Gurevich [6], we can fit the \( H_{c2}(T) \) at \( T \sim T_c \) at temperatures close to \( T_c \), based on the model of Gurevich [6], Putti et al. [19] have shown that the gradient of the \( H_{c2}(T) \) is given by

\[
\frac{dB_{c2,ab}}{dT}_{T_c} \sim \frac{8\phi_0 k_B}{\pi^2 \hbar} \frac{1}{a_1 D_\sigma / \gamma + a_2 D_\pi}
\]

where \( \phi_0 \) is the magnetic flux quantum, \( D_\sigma \) are the diffusivities in the \( \pi \) and \( \sigma \) bands and \( a_1 \) and \( a_2 \) are proportional to the coupling constants, \( \lambda_{i,j} \). For pure MgB\(_2\), \( a_{1,2} \), have been calculated [6, 19, 20] to be \( a_1 = 1.93 \) and \( a_2 = 0.07 \). Following the method set out in reference [19] we find that \( \eta = 0.10 \pm 0.06 \). While this value is smaller than that found by PCAR, we note that the values are of the same order of magnitude and consistent considering particularly that \( \eta \) varies...
over several orders of magnitude [6, 7, 10, 11, 21, 22]. Further comparison of the two methods would be possible were the entire $H_{c2}(T)$ curve obtained and fitted.

Having explored the consistency of the extracted value of $\eta$ determined by two independent methods (and validating the point contact method), we turn our attention to the Mg deficient bulk samples. We have recently made a study of these samples at 4.2 K [17] but here, extend that study to explore the behaviour of the diffusivity ratio as the temperature is increased. Note, that it is not possible to extract such information by fitting the $H_{c2}(T)$ curve. To summarise our previous findings, a series of point contact spectra were obtained at 4.2 K as a function of field using an Au tip as described previously [4]. In zero applied field, both samples showed a distribution of observed gap values, but no evidence was found for the gradual closing of the two gaps; the separation between $\Delta_\pi$ and $\Delta_\sigma$ bands was similar in both samples. The increased magnesium deficiency did not therefore increase the interband scattering between the $\pi$ and $\sigma$ bands. Interband scattering is associated with suppressed $T_c$ in the MgB$_2$ [1, 3, 23]. The similarity in the $T_c$s obtained for the samples (x=1.0, 37.5K; x=1.5, 38K) is therefore consistent with the PCAR data. On the other hand, the magnesium deficient samples did show an enhanced low temperature $H_{c2}$ value [16, 17]. In order to investigate this, the spectra, as a function of field, were fitted to the two channel model as previously described in references [2, 4, 12, 17]. Although strictly the model of Koshelev and Golubov [7, 8] is only valid for fields applied parallel to the c-axis, we and others have shown that the model can be applied to bulk samples as the $\pi$ band is relatively isotropic at low fields [2, 4, 17]. Although strictly the model of Koshelev and Golubov [7, 8] is only valid for fields applied parallel to the c-axis, we and others have shown that the model can be applied to bulk samples as the $\pi$ band is relatively isotropic at low fields [2, 4, 17]. As the relative scattering of the $\pi$ band increases, that is, for higher values of $\eta$, the gradient of the $N_\pi$ DOS becomes shallower with field. The $x=1.5$ sample has a steeper gradient of $n_\pi(h)$ at lower fields, indicative of a lower value of $\eta$. This result suggests that the effect of introducing magnesium vacancies into the MgB$_2$ structure is to selectively increase the scattering in the $\pi$ band.

The effect of temperature on the point contact spectra is shown in the inset to figure 2. As the temperature is increased the peaks due to $\Delta_{\pi,\sigma}$ move to lower energy while the weighting between the conductance from the $\pi$ and $\sigma$ band conductances decreases such that the contribution from
the \( \sigma \) band decreases. The stability of the contact is indicated by the fact that the data in the inset to figure 2 is the raw conductance data, it is not normalised. The behaviour of the \( \Delta_\pi(T) \) is shown in the main panel to figure 2 and shows typical behaviour for \( \Delta_\pi \). The behaviour of \( n_\pi(h) \) as a function of temperature for the \( x=1.5 \) sample is shown in figure 3. The hysteresis observed in the \( n_\pi(h) \) at 4.2K (marked with arrows on the figure) has previously been attributed to vortex pinning behaviour \[24\]. As an aside, such hysteresis in the PCAR data due to vortex pinning has not been previously noted. With increasing temperature there is a marked decrease in both the gradient of the \( n_\pi(h) \) and its apparent saturation. A naïve interpretation of the model of references \[7, 8\] would imply that \( \eta \) increases substantially with temperature. That is, figure 3 appears to imply that the \( \pi \) band becomes increasingly dirty with respect to the \( \sigma \) as the temperature is increased from 4.2 K to 20 K. However the model we have used does not explicitly take into account the increase in the zero field quasiparticle density of states that occurs as the temperature is increased. This is the likely explanation for the depressed saturation value of \( n_\pi(h) \) as the temperature is increased. The decrease in the zero field gap value with increasing temperature, figure 2, certainly reflects the same phenomenon. Consequently we also examine how the field dependence of the \( \Delta_\pi \) varies as a function of temperature (inset to figure 3). There may be subtle differences between the two normalised \( \Delta_\pi(h) \) curves, but the error bars are extremely large so it is difficult to say anything quantitative. Recently, the behaviour of the zero bias DOS for the \( \sigma \) and \( \pi \) bands as a function of temperature has been studied by Tanaka et al \[9\]. In this model, the decrease of the \( \pi \) band contribution to the current density at distances far from the vortex core decreases with increasing temperature similarly to the decrease in \( n_\pi \) observed here and is therefore an alternative explanation for the results. However, Tanaka et al, used a model of a ballistic \( \sigma \) band inducing superconductivity in a diffusive \( \pi \) band. It is therefore questionable whether this model is applicable to bulk samples in which \( \eta \) has been shown to be less than 1 \[17\]. To understand the temperature dependence of the PCAR data in these bulk samples would therefore require further theoretical development.

In summary we have shown that the values of \( \eta \) obtained by fitting the PCAR data as a function of field are qualitatively consistent with that extracted from fitting the \( H_{c2} \) at \( T \sim T_c \).
Figure 3. The $n_\pi$ as a function of reduced field at $\he{4.2}$, $\he{10}$ and $\he{20}$. The $H_{c2}$ values taken to normalise the data were obtained from reference [16]. Lines are guides for the eye. Inset shows the $\Delta_\pi(h)$ normalised to its zero field value at $4.2$ K ($\he{4.2}$) and $20$ K ($\he{20}$).

For a thin film with a well defined c-axis orientation the value of $\eta$ obtained from the PCAR data was $\eta = 0.5$, while the value of $\eta$ obtained from fitting the gradient of $H_{c2}$ at $T\sim T_c$ was $\eta=0.1 \pm 0.06$ assuming a value of $\gamma = 4$. Point contact data on magnesium deficient bulk samples at zero field indicated that magnesium vacancies do not increase the interband scattering in the MgB$_2$ samples and that the $\pi$ intraband scattering increases with increasing magnesium deficiency. The temperature dependence results of the $n_\pi(h)$ in bulk samples are certainly interesting and suggest that further theoretical and experimental development would be a worth while extension of this present work.

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