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The angular dependence of the upper critical field H_{c2} of MgB_2 single crystals is studied at various temperatures by means of specific-heat and transport measurements in magnetic fields up to 17 T. Clear deviations from Ginzburg–Landau behavior are observed at all temperatures and are explained by two-band effects. The angular dependence and temperature dependence of the deviations are in qualitative agreement with theoretical predictions based on band-structure calculations. Quantitative agreement is obtained with an interband coupling slightly stronger than the calculated one, enabling band-structure anisotropies and interband coupling strength to be experimentally estimated. This provides a pathway to the study of disorder and doping effects in MgB_2.

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The emergence of new theoretical works with close experimental connections has significantly deepened the understanding of the properties of magnesium diboride (MgB_2). Despite the fact that the superconducting properties of MgB_2 with its fairly simple atomic structure were just recently discovered,[1] this phonon mediated *s*-wave superconductor has already been the subject of intense and numerous studies[2] due to its exotic properties arising from a complex, disconnected, multiband Fermi surface. Band-structure calculations have demonstrated that the Fermi surface is composed of pairs of three-dimensional π bands and quasi-two-dimensional (2D) σ bands.[3] This effective two-band structure has been confirmed by de Haas–van Alphen (dHvA) measurements[4] and angle-resolved photoemission spectroscopy.[5]

The superconducting properties of the two sets of bands are quite different, due to the low overlap of the orthogonal σ- and π-band wave functions. The superconducting gap ranges from 1.5 to 3.5 meV on the π bands and from 5.5 to 8 meV on the strongly superconducting σ bands.[6] This double-gap nature has been verified by tunneling experiments,[7–9] heat capacity measurements,[10] Raman,[11] and point contact spectroscopy.[12,13]

Theoretically, two-band superconductivity has a history starting well before MgB_2.[14–16] Through theoretical advances, a fairly unified picture has emerged with predictions that can be experimentally substantiated.[6,17–26] One of the salient predictions associated with a pronounced two-band effect is a difference between the coherence length anisotropy $\gamma_\xi = \xi_{ab}/\xi_c$ (Refs. 19–22) and the penetration depth anisotropy $\gamma_\lambda = \lambda_c/\lambda_{ab}$,[23,24] both of which become temperature dependent with opposite tendencies. For MgB_2, a strong decrease of $\gamma_\xi = H_{c2}^{ab}/H_{c2}^{c}$ from $\gamma_\xi(0) \sim 5$ to $\gamma_\xi(T_c) \approx 2$ is found experimentally,[27–32] while controversy remains about the experimental temperature dependence of γ_λ.[31–33]

In this Brief Report we present evidence of clear deviations of the angular dependence of H_{c2} from the anisotropic Ginzburg–Landau (GL) description. The $H_{c2}(T,\theta)$ transition of MgB_2 single crystals was determined from resistivity measurements and specific heat with excellent agreement between the two. With a slight adjustment of some of the parameters supplied by band-structure calculations, good quantitative agreement is found between theory[22] and experiment, yielding fundamental estimates of band-structure anisotropies and the interband coupling strength.

Several MgB_2 crystals with typical dimensions $50-250\ \mu$m were obtained through a high-pressure heat treatment of a mixture of Mg and B in excess Mg as described elsewhere.[34] The crystals had T_c values of 34–36 K, residual resistivity ratios (RRR) ~ 3.5, and a $H_{c2}^{c}(0) \approx 3.5$ T. Transport measurements (sample *A*) were performed using standard ac techniques at 23 Hz. For the specific-heat measurements, each crystal was mounted on top of a flattened 12.7 μm chromel/constantan thermocouple junction. Small temperature oscillations of the sample were induced by either a resistive heater wire (sample *B*, Argonne and NHMFL) or by modulating the temperature of the copper base[35] (sample *C*, Grenoble).

Figure 1 shows the transitions from resistivity (top) and specific heat (bottom) as a function of angle at $T = 27.5$ K and $T = 25.0$ K, respectively. The resistive transitions were measured at a relatively high current density to suppress the effects of surface superconductivity at the well-shaped crystal surfaces, as discussed in Ref. 36. Restrictions on the current to avoid heating from the Ohmic contacts limited the transport measurements to temperatures $T/T_c > 0.7$. The value of $H_{c2}(\theta)$ was determined through a linear extrapolation of the steep drop to zero resistivity as shown by the

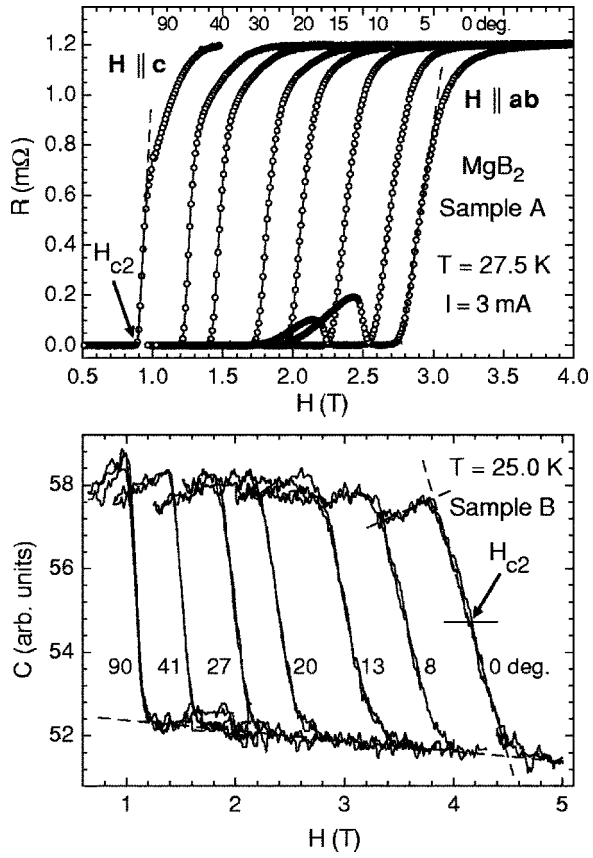

FIG. 1. Top, resistive H_{c2} transition at $T=27.5$ K as a function of magnetic field H and angle θ from the basal plane. Bottom, specific heat signature at $T=25.0$ K for sample B.

dashed lines in the top panel.[36] The appearance of the peak effect just below H_{c2} for some angles is evident in the figure. The thermodynamic signature of H_{c2} was defined from the midpoint of the specific heat transitions, as illustrated in the bottom panel. The choice of definition was checked not to be significant. It is interesting to note that the specific heat step height is fairly independent of the field direction. This is in agreement with GL theory, where the step height should scale with $T(\mathrm{d}H_c/\mathrm{d}T)^2$, where H_c is the (isotropic) thermodynamic critical field. Possible deviations from a constant step height arising from two-band effects are too small to be resolved in the current data due to uncertainties in the experimental method.

Clear deviations from an anisotropic GL description are, however, seen in the angular dependence of H_{c2}. In Fig. 2, the $H_{c2}(\theta)$ curves are shown for two selected temperatures together with corresponding fits to the effective mass description $H_{c2}^{GL}(\theta)=H_{c2}^{ab}/(\cos^2\theta+\gamma_\xi^2\sin^2\theta)^{1/2}$. The relative deviations are fairly small at 12.1 K as compared to 25.0 K. They are nevertheless clearly discernible at all temperatures and are reproducible between different measuring setups, samples, and methods. Resistive measurements by Eltsev *et al.* displayed similar deviations but were not analyzed in detail.[37] Deviations were also reported at 33 K using torque measurements.[38] The latter, however, suffer from the inability to measure H_{c2} along the symmetry axes. Deviations from GL behavior have also been observed on thin films.[39]

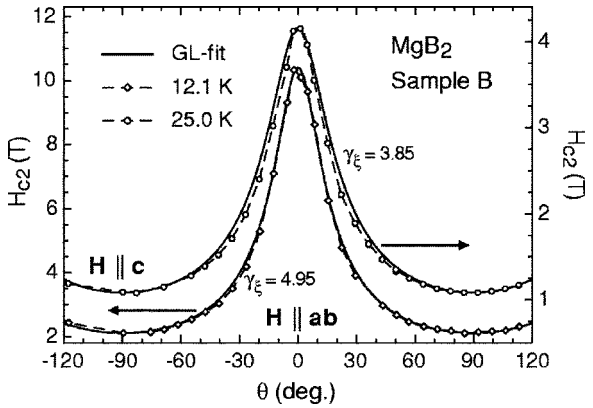

FIG. 2. Angular dependence of the upper critical field at 12.1 K and 25.0 K. The solid lines correspond to the GL theory. Small, but clear and consistent deviations from the anisotropic, effective-mass description are seen.

The deviations from GL behavior are illuminated by plotting the ratio $\mathcal{A}=[H_{c2}(\theta)/H_{c2}^{\mathrm{GL}}(\theta)]^2$ as a function of $\cos^2\theta$ as shown in Fig. 3. When the field is directed along the c axis or within the basal plane there are no deviations, since the experimental H_{c2}^{ab} and H_{c2}^{c} were used as parameters for the GL-fit ($\mathcal{A}=1$) at each temperature. The shape of the deviations as a function of angle is similar for all temperatures, with a maximum amplitude at around $\theta=20°-30°$, i.e., for $\cos^2\theta\sim 0.9$.

The maximum amplitude of $\mathcal{A}(\theta)$ is plotted as a function of temperature in Fig. 4 (top). Good agreement is found between the data on all the samples, illustrating the fundamental and consistent nature of the deviations. The amplitude is relatively small at low temperatures and reaches a maximum slightly below T_c. By comparing the temperature dependence of $\mathcal{A}_{max}$ with that of γ_ξ (bottom panel) one can see that the maximum of $\mathcal{A}_{max}(T)$ occurs at intermediate values of γ_ξ.

The general experimental features of $H_{c2}(T,\theta)$ can be excellently described by the recent theory of the angular depen-

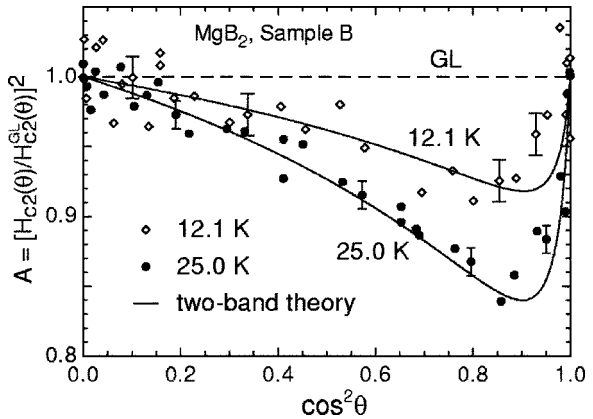

FIG. 3. Ratio $\mathcal{A}=[H_{c2}(\theta)/H_{c2}^{GL}(\theta)]^2$ as a function of $\cos^2\theta$ for the two temperatures of Fig. 2. The dashed line $\mathcal{A}=1$ corresponds to the GL description with a separate anisotropy parameter $\gamma_\xi=H_{c2}^{ab}/H_{c2}^{c}$ for each temperature as illustrated in Fig. 2. Solid curves are given by two-band theory with best-fit parameters as discussed in the text.

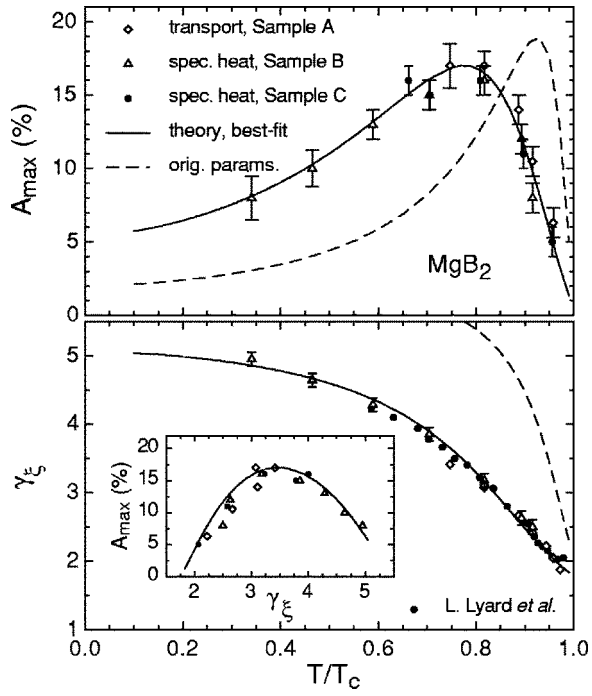

FIG. 4. Top, maximum deviations of $\mathcal{A}(\theta)$ from the GL theory as a function of reduced temperature. The dashed curve is taken from Ref. 22. Bottom, temperature dependence of the experimental anisotropy γ_ξ. Solid circles are taken from Ref. 29. Other symbols are as above. The inset shows $\mathcal{A}_{max}$ as a function of anisotropy, illustrating maximum deviations at intermediate values of γ_ξ where both bands contribute equally. Parameters for the theoretical curves are given in Table I.

dence of dirty two-band superconductors.[22] The two-band theory requires as input (i) band anisotropies γ_σ and γ_π, (ii) a ratio of the diffusion constants in the two bands, e.g., $r_x=\mathcal{D}_{\pi,x}/\mathcal{D}_{\sigma,x}$, and (iii) the matrix of effective coupling constants $\Lambda_{\alpha\beta}=\lambda_{\alpha\beta}-\mu^*_{\alpha\beta}$, where $\lambda_{\alpha\beta}$ are the electron-phonon coupling constants and $\mu^*_{\alpha\beta}$ are the Coulomb pseudopotentials (α and β are indices for the σ and π bands). The theoretical dependencies of $\mathcal{A}(T)$ and $\gamma_\xi(T)$ obtained by using coupling constants and anisotropies as supplied by band-structure calculations[18,25] are illustrated by the dashed curves in Fig. 4. It is clear that, while qualitatively similar, the theoretical curves are displaced closer to T_c and the predicted anisotropy is higher than the experimental one.

To find the set of parameter values that would best describe the data, we allowed the parameters γ_σ, γ_π, r_x, $\Lambda_{\sigma\sigma}$, and $\Lambda_{\pi\pi}$ to vary freely. The off-diagonal Λ-parameters were calculated from the constraints arising from the value of T_c (Ref. 20) and the requirement of detailed balance, $\Lambda_{\sigma\pi}/\Lambda_{\pi\sigma}=N_\sigma/N_\pi$, where N_α is the partial density of states. It is useful to know that $\gamma_\xi(0)\approx\gamma_\sigma$ and that the overall change in anisotropy can be estimated as $\gamma_\xi(0)/\gamma_\xi(T_c)\approx\sqrt{1+S_{12}r_z}$, where $S_{12}\approx\Lambda_{\sigma\pi}\Lambda_{\pi\sigma}/(\Lambda_{\sigma\sigma}-\Lambda_{\pi\pi})^2$ is the reduced interband coupling strength and $r_z=r_x(\gamma_\sigma/\gamma_\pi)^2$.[22] The optimum fit parameters are listed in Table I. The corresponding fit curves, included in Figs. 3 and 4, give an excellent account of all the data. We note that calculations[14,20] using the obtained parameters give a reasonable gap anisotropy, $\Delta_\pi/\Delta_\sigma\approx0.38$ and that the parameter value $1/r_x\approx0.23$ is in good agreement with the observation of an enlarged vortex core.[26,40]

TABLE I. Parameters used in the theoretical computations.

Parameter	Predicted value[a]	Experimental value
γ_π	0.82	1.02 ± 0.05
γ_σ	6.3	5.45 ± 0.10
$1/r_x=\mathcal{D}_{\sigma,x}/\mathcal{D}_{\pi,x}$	0.2	0.23 ± 0.02
$\begin{pmatrix}\Lambda_{\sigma\sigma} & \Lambda_{\sigma\pi}\\ \Lambda_{\pi\sigma} & \Lambda_{\pi\pi}\end{pmatrix}$	$\begin{pmatrix}0.81 & 0.115\\ 0.091 & 0.278\end{pmatrix}$	$\begin{pmatrix}0.695 & 0.177\\ 0.140 & 0.260\end{pmatrix}$

[a]As predicted by band-structure calculations. Λ values are taken from Ref. 25 and γ values are obtained from Ref. 18. The parameter $1/r_x=0.2$ was taken from the experimental observations of an enlarged vortex core (Ref. 26).

The most notable change of the parameters in Table I with respect to the band-structure results is the strong enhancement of the interband coupling, as reflected by an increase of S_{12} from 0.034 to 0.105. In addition, the σ-band anisotropy was found to be smaller and the π band almost isotropic. A probable explanation for the differences in parameter values is a slight, sample dependent variation of the band structure as seen, for example, in detailed comparisons between dHvA data and theoretical predictions.[4] A possible reason for the difference in interband coupling is also a theoretical overestimation of the off-diagonal Coulomb pseudopotentials (see discussion in Ref. 41) resulting in too low values of $\Lambda_{\sigma\pi}$ and $\Lambda_{\pi\sigma}$. Unfortunately, no independent, experimental probe of the off-diagonal coupling constants is available at present. The present theoretical analysis neglects interband scattering. However, by evaluating the corrections to the components of the upper critical fields arising from the inclusion of weak interband scattering, we conclude that it is unlikely that this scattering is responsible for differences between the calculated and experimental values (e.g., for the lower value of γ_σ).[42]

MgB_2 single crystals are usually described as fairly clean, with the σ band probably in the clean limit and the π band probably in the dirty limit.[4,40,43] The temperature dependence of γ_ξ measured on crystals similar to ours has recently been described successfully within the clean-limit formalism.[21] However, these calculations require modifications of coupling and band anisotropies from the predicted values similar to those found here. To our knowledge, a clean-limit calculation of the angular dependence of H_{c2} has not yet been presented. The theoretical analysis used here should be valid for the dirty π band in the entire temperature range and for a clean σ band at sufficiently high temperatures, where this band is described by GL theory. One could expect deviations from the present two-band theory at low temperatures arising from the Fermi surface structure of the clean σ band. This cannot be ruled out entirely, but a theoretical estimate of its importance has, to our knowledge, not yet been presented. Since our results show that the deviations decrease with decreasing temperature at low temperature, opposite to the expected single-band, clean-limit behavior, we believe that our experiments are accurately described within the present the-

oretical framework of two-band superconductivity. However, clean-limit effects and strong coupling corrections may, to some degree, alter the absolute values of the determined parameters.

In summary, we have studied the angular dependence and temperature dependence of the upper critical field of MgB_2 single crystals by means of heat capacity and transport measurements. Clear two-band effects are found in both $H_{c2}(\theta)$ and the temperature dependence of the upper critical-field anisotropy $\gamma_\xi(T)$. The experiments are well explained by the theory,[22] providing a deep understanding of the microscopic parameters describing the system. This work thus points out a pathway to the study of disorder and doping effects in MgB_2, with great implications for future applications.

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