needed to appropriately discriminate between these possibilities in the present case.

We have investigated by inelastic neutron scattering the magnetic excitations in the field-driven ordered phase of $TlCuCl_3$. Our main result is the evidence of a gapless mode emerging at the field-induced magnetic Bragg point. The gapless mode is accompanied by two renormalized Zeeman modes at higher energies. We find almost complete agreement between experimental observations and theoretical predictions that are based on a BEC theory. However, this theory does not involve a dilute gas cooled below the de Broglie temperature, but rather dimer states split at high magnetic fields. We hope that our approach to the study of a fundamental quantum effect like BEC will enable further theoretical and experimental work. □

Methods

The inelastic neutron scattering investigations up to $H = 14$ T were performed on the three-axis spectrometer V2 at the Hahn-Meitner-Institut, Germany. Preliminary measurements up to $H = 6$ T were performed on the three-axis spectrometers IN14 at the Institut Laue-Langevin, France, and on TASP at the spallation neutron source SINQ, Switzerland. All instruments featured initial and final energy selection by pyrolytic graphite (0 0 2) crystals. The measurements were performed at fixed final energy, with a cooled beryllium filter in front of the analyser set at 4.7 meV. Standard instrumental configurations with 40′-open-open and 60′-60′-60′ horizontal collimation were adopted. The typical elastic energy resolution was 0.2 meV (full-width at half-maximum). Superconducting cryomagnets with split coils were adopted on all instruments. The cooling of the sample in the cryomagnets was achieved by ^{4}He flow through a root pump ($T = 1.5$ K) and by a ^{3}He–^{4}He dilution insert system ($T = 50$ mK), producing the data set presented in the text. Further details of the sample environment will be provided elsewhere.

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The origin of multiple superconducting gaps in MgB_2

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Magnesium diboride, MgB_2, has the highest transition temperature ($T_c = 39$ K) of the known metallic superconductors[1]. Whether the anomalously high T_c can be described within the conventional BCS (Bardeen–Cooper–Schrieffer) framework[2] has been debated. The key to understanding superconductivity lies with the 'superconducting energy gap' associated with the formation of the superconducting pairs. Recently, the existence of two kinds of superconducting gaps in MgB_2 has been suggested by several experiments[3–9]; this is in contrast to both conventional and high-T_c superconductors. A clear demonstration of two gaps has not yet been made because the previous experiments lacked the ability to resolve the momentum of the superconducting electrons. Here we report direct experimental evidence for the two-band superconductivity in MgB_2, by separately observing the superconducting gaps of the σ and π bands (as well as a surface band). The gaps have distinctly different sizes, which unambiguously establishes MgB_2 as a two-gap superconductor[10,11].

Soon after the discovery of MgB_2, many experimental studies indicated that MgB_2 should be basically classified as a conventional phonon-mediated BCS superconductor[12–16]. Recently, however, its deviation from the simple BCS framework has been inferred from several experiments using high-quality samples[3–9,17]. The most striking deviation is the complex superconducting order parameter, referred to as the 'multi-component superconducting gap' or 'multi-gap', indicating that two or more superconducting gaps with different sizes develop simultaneously at T_c between the occupied and unoccupied electronic states[10,11]. This multi-gap

behaviour has been analysed by several models[10–13,18–20] based on the different role of σ and π bands[10,11,18] or surface and bulk[12,13,19], or the strongly anisotropic coupling to lattice vibrations (phonons)[20].

Of these, the two-band model[10,11] may be theoretically described as follows: electrons in the σ bands are strongly coupled to phonons confined within the honeycombed boron layer and give rise to a large gap, whereas a relatively small gap opens in the π band due to the weak electron–phonon coupling. There has been no direct experimental evidence, however, that establishes the two different gaps originating in the σ and π bands. Previous experiments such as the specific-heat measurements[3,4], and Raman[6,7] and tunnelling[8,9] spectroscopies provide the information averaged over all the momentum space and therefore could not separate the different bands. In contrast, the present high-resolution angle-resolved photoemission spectroscopy (ARPES) experiment has succeeded in resolving the σ and π bands and directly observing the superconducting gaps.

Figure 1a shows ARPES spectra of MgB_2 measured along the ΓKM(AHL) direction (see inset to Fig. 1a) with 28-eV photons at 45 K. We find several band dispersions which cross the Fermi level (E_F) in this momentum region. To visualize the band dispersion more clearly, we plot the ARPES intensity as a function of wave vector and binding energy in Fig. 1b. We find three distinct band dispersions. The first is a large electron-like band which crosses E_F near the K(H) point. The second is a hole-like dispersion centred at Γ(A), crossing E_F near Γ(A). The third is a small electron-like pocket centred at Γ(A).

This result is consistent with the previous ARPES report[21]. According to the band calculation[22], the first and second bands are attributed to the boron $2p_\pi$ and $2p_\sigma$ bands, respectively. The rather broad feature of the π band reflects its three-dimensional nature, while the sharp σ band shows the strong two-dimensional character. The third band, which is not seen in the band calculation, has been ascribed to a surface state[21]. The observed clear difference in the E_F-crossing point among the three bands enables us to perform precise ARPES measurements on the superconducting gaps of different bands separately.

We measured ARPES spectra in the vicinity of E_F at three different points on the Fermi surfaces (points A, B and C in Fig. 1b) at two temperatures below and above T_c (17 K and 45 K). The results are shown in Fig. 2. The ARPES spectrum from the σ band shows a remarkable temperature dependence which cannot be explained by the simple temperature effect due to the Fermi–Dirac function. The midpoint of the leading edge in the spectrum at 17 K is not at E_F but is shifted by about 2 meV toward the higher binding energy, while the leading-edge midpoint at 45 K is located at E_F. This clearly indicates that a superconducting gap opens at 17 K in the σ band. We observe a small piling-up effect of spectral weight around 7–15 meV in the 17 K spectrum, which indicates the emergence of a superconducting coherent peak below T_c, as observed in other superconductors[23]. We find it surprising that a similar gap-opening behaviour is observed in the surface band as seen in Fig. 2. This point will be discussed later.

In contrast to the σ and surface bands, the π band does not show a clear gap-opening behaviour. The shift of the midpoint of the leading edge at 17 K is much less (0–1 meV) and no clear piling-up effect around 10 meV is seen, suggesting that no or a very small superconducting gap opens in the π band. This reveals the clear difference in the contribution to the superconductivity among three bands, supporting the multi-band superconductivity scenario in MgB_2 (refs 10, 11).

To estimate the gap size (Δ), we numerically fitted the spectrum using the BCS spectral function with an integrated-type background representing the incoherent part of the spectral function and inelastically scattered electrons, multiplied by the Fermi–Dirac function and convoluted with the energy resolution, as has been employed in high-T_c copper oxides[24]. As shown in Fig. 2, ARPES

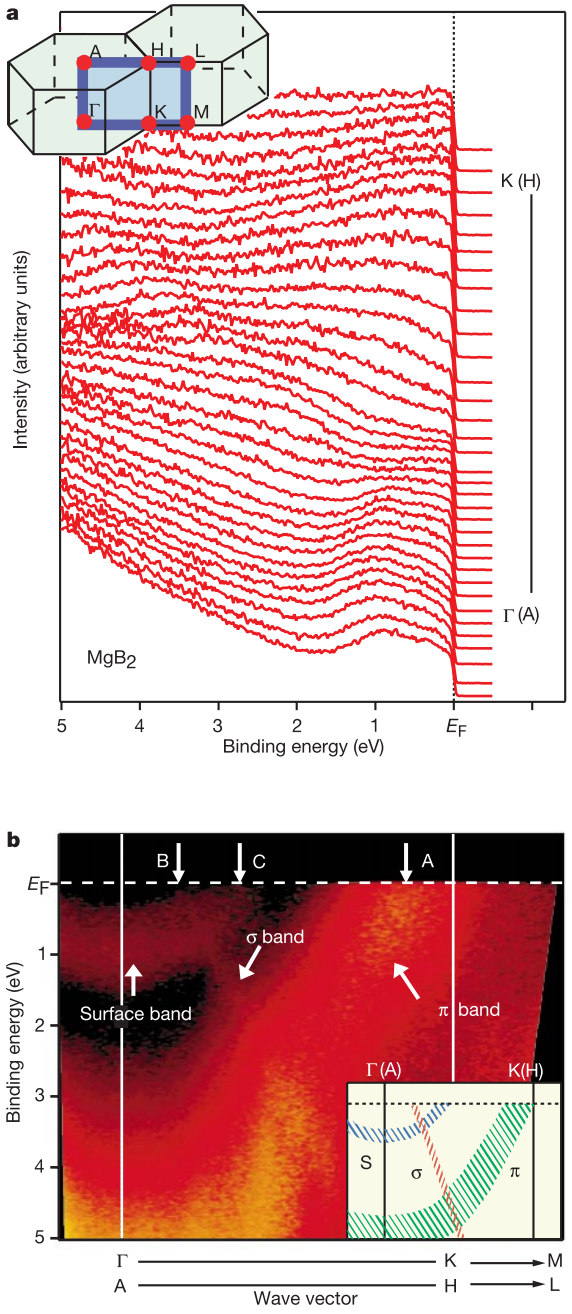

Figure 1 Experimental band structure near E_F of MgB_2 obtained by ARPES. **a**, ARPES spectra of MgB_2 measured at 45 K along the ΓKM(AHL) direction in the Brillouin zone (inset to **a**) with 28-eV photons. **b**, ARPES-intensity plot of MgB_2 as a function of wave vector and binding energy. White arrows A, B and C indicate the location of Fermi-level crossing points for the π, σ and surface bands. Inset to **b** shows the schematic view of the band dispersion; S indicates surface. Single crystals of MgB_2 were grown in a closed stainless steel tube filled with argon gas. The tube was heated up to 1,200 °C and slowly cooled down to room temperature in 12 h. Details of sample preparation are described elsewhere[26]. Magnetization measurement confirmed that crystals exhibit superconductivity at 38 K with a transition width of $\Delta T_c = 0.8$ K. The typical size of crystals used in ARPES measurements is about $100 \times 100 \times 10\,\mu m^3$. ARPES measurements were performed using a SCIENTA SES-2002 spectrometer at the undulator 4 m-NIM beam line at the Synchrotron Radiation Center, Wisconsin. The energy and angular (momentum) resolution were set to 10 meV and 0.3° (0.01 $Å^{-1}$), respectively. We obtained a clean surface for ARPES measurements by cleaving a single crystal *in situ* along the (0001) plane in an ultrahigh vacuum (better than 2×10^{-10} torr). The crystal orientation was determined by Laue X-ray diffraction before the ARPES measurement. Polarization vector of photons was set parallel to the ΓKM(AHL) direction.

spectra at 17 K for the σ, π and surface bands are satisfactorily fitted with gap sizes of $\Delta = 6.5 \pm 0.5$ meV, 1.5 ± 0.5 meV, and 6.0 ± 0.5 meV, respectively, with broadening factor of 1.0 meV for the σ and surface bands and 0.5 meV for the π band. A slight deviation above E_F observed for the σ and surface bands may be due to a normal metallic region at the surface[25].

From the viewpoint of gap size, we consider that the present ARPES experiment has observed 'two' gaps in MgB_2 because the size is almost the same between the σ and surface bands. Thus far, many experiments, in particular those using tunnelling spectroscopy, have reported the multi-gap feature in MgB_2 and the reported gap sizes are roughly categorized into two groups: small and large superconducting gaps. The small gap has a value of 1–4 meV and the large gap has a value of 5.5–10 meV. The present ARPES result shown above seems to be consistent with these previous reports.

The two-gap nature in MgB_2 reported by previous studies has been interpreted by several models: (1) the simultaneous observation of the genuine bulk superconductivity and the weakened superconductivity in the surface layer[12,13]; (2) the proximity effect in a thin metallic surface layer[19]; (3) the strongly anisotropic coupling to phonons[20]; (4) the contribution from the σ and π bands, which give the large and small gaps, respectively[10,11]; and (5) as for model (4) but with opposite band contibutions, that is, the π band produces a large gap[18].

It is clear from the present ARPES results that the σ and the surface bands have a large gap of 6–7 meV whereas the π band shows a small gap of 1–2 meV. Taking into account that the superconductivity of MgB_2 is bulk in nature[1,3], we conclude that the σ band is dominant in the superconductivity of MgB_2 with a stronger coupling to phonons, while the π band is less important to the superconductivity with a much weaker coupling. This unambiguously indicates that the two-band model[10,11] describes the superconductivity of MgB_2 most appropriately.

We also conclude from the present ARPES results that the large gap observed by surface-sensitive techniques such as tunnelling spectroscopy may be due to both the σ and the surface bands, because the gap size is almost the same for the two bands. A large variation in the size of the large gap may stem from the various surface conditions, depending on the surface preparation method. Although why the surface band exhibits a similar gap size to the σ band is unclear at present, the proximity of the two bands in the momentum and energy spaces as seen in Fig. 1 may account for it, because the interaction between the surface and σ bands is expected to be larger than that between the surface and π bands. □

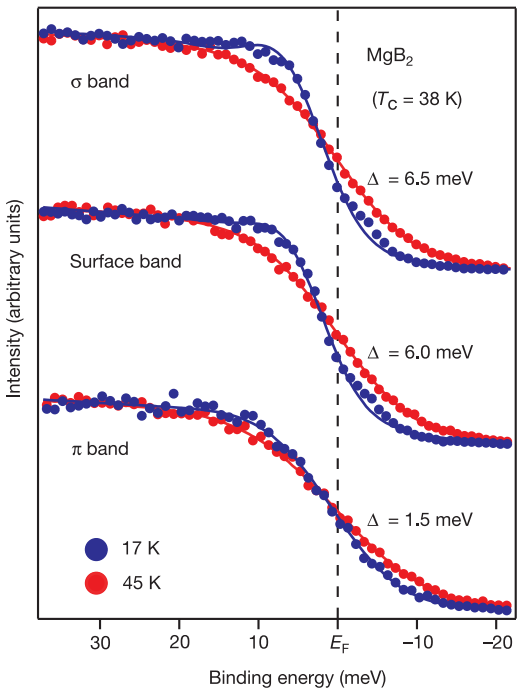

Figure 2 Temperature dependence of ARPES spectra near E_F of MgB_2. Spectra are measured at three points A, B and C in Fig. 1b, which correspond to the π, σ and surface bands, respectively. Measurements were done at 17 K (blue circles) and 45 K (red circles). Spectra are normalized by the area under the curve from 50 meV above E_F to 200 meV below E_F. Blue and red lines on the spectra show the results of numerical fitting at 17 K and 45 K, respectively. The reproducibility of experimental data has been confirmed by measuring the spectra with the cycling temperature of the sample across T_c.

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