Photoemission results of intermetallic superconductors: Nb_3Al and MgB_2

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Abstract

We study the superconducting electronic structures of Nb_3Al and MgB_2 using high-resolution spectroscopy. The obtained spectrum of Nb_3Al measured below T_c shows clear opening of the superconducting gap with a sharp pile up in the density of states and a shift of the leading edge. In addition, the spectrum shows a peak–dip–hump line shape expected from the strong-coupling theory. On the other hand, for MgB_2, the superconducting-state spectrum measured at 5.4 K shows a coherent peak with a shoulder structure, in sharp contrast to that expected from a single isotropic gap. The superconducting spectral shape of MgB_2 can be explained in terms of a multicomponent gap. © 2002 Elsevier Science Ltd. All rights reserved.

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1. Introduction

Recently, because of the advances in energy resolution and sample cooling technique of photoemission spectroscopy, we have been able to study the energy gap of a superconductor with a relatively small transition temperature (T_c). For examples, the superconducting gaps of C_{60} based superconductors [1] and an A15 compound V_3Si [2] have been successfully measured with photoemission spectroscopy. Furthermore, in a strong electron–phonon coupling superconductor Pb, a peak–dip–hump line shape as well as the superconducting gap has been clearly observed [3] to justify the photoemission results of high temperature superconductors, where the similar peak–dip–hump structure has been reported [4,5]. An anisotropy of the superconducting gap of Ni borocarbides have been discussed from the change of leading edge line shapes as a function of impurity doping [6].

In this paper, we extend the photoemission study on the superconducting gap to intermetallic compounds: Nb_3Al [7] and MgB_2 [8]. For Nb_3Al, we study the superconducting gap as well as phonon-induced structures. Since Nb_3Al has been

classified into a moderate- to strong-coupling superconductor, we try to see whether the strong-coupling spectral function can be observed even for compounds using photoemission spectroscopy. The obtained spectrum shows a superconducting gap with a peak–dip–hump line shape expected from the strong-coupling theory. For MgB_2, which is a newly discovered intermetallic superconductor with T_c as high as 40 K [8], we study the superconducting electronic structures responsible for such a high T_c. The observed superconducting-state spectrum at 5.4 K shows a coherent peak with a shoulder structure, in sharp contrast to that expected from single isotropic-gap opening. From analyses using single isotropic and anisotropic Dynes functions and two Dynes functions, we find that the weighted sum of the two Dynes functions can reproduce the obtained spectral shape very well. This indicates that MgB_2 has a multicomponent gap.

2. Experiment

Polycrystalline Nb_3Al samples were prepared by arc-melting, while high-density MgB_2 polycrystalline samples were prepared with a high-pressure synthesis [9].
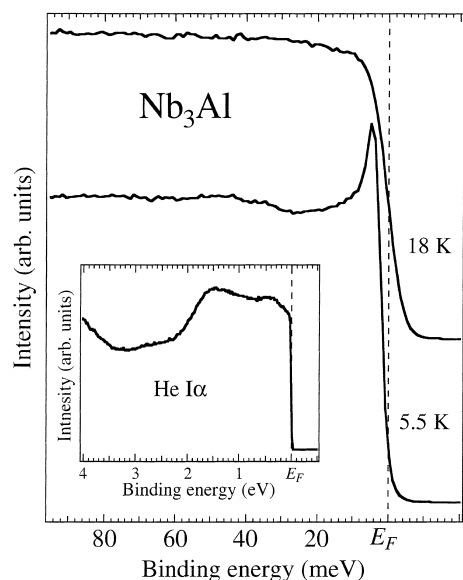
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Fig. 1. High-resolution photoemission spectra of polycrystalline Nb$_3$Al (T_c = 17.6 K) measured at superconducting state (5.5 K) and normal state (18 K) using the He Iα resonance line. Inset shows the valence band spectrum of Nb$_3$Al.

Magnetization measurements exhibited sharp transitions (midpoint) at 17.6 K for Nb$_3$Al and 36.5 K for MgB$_2$.

Photoemission measurements were performed on a spectrometer built using a GAMMADATA-SCIENTA SES2002 electron analyzer and a high-flux discharging lamp with a toroidal grating monochromator. The total energy resolution (analyzer and light) used the He Iα (21.218 eV) resonance line was set to $\sim$4 meV. The sample temperatures were measured using a silicon-diode sensor
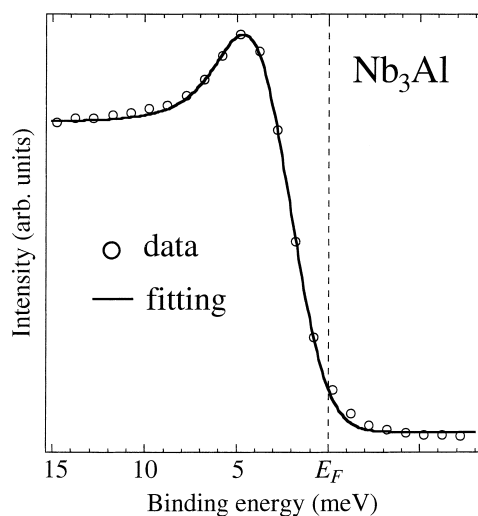

Fig. 2. Representative result of Dynes function analysis (curve) with Δ = 4.0 meV and Γ = 0.005 meV together with the superconducting spectrum at 5.5 K (open circles).

mounted just close to it. The base pressure of the spectrometer was better than 5×10^{-11} Torr. Samples were fractured in situ to obtain clean surfaces. We did not see any spectral changes within the measurements, indicating the observed spectra are reflecting intrinsic electronic structures. Fermi energy of samples was referenced to that of a gold film evaporated onto the sample substrate and its accuracy is estimated to be better than 0.2 meV.

3. Results and discussion

Fig. 1 shows high-resolution photoemission spectra of Nb$_3$Al measured at 5.5 K (superconducting state) and 18.0 K (normal state) using the He Iα resonance line, together with the total valence band spectrum (inset). The valence band spectrum, which has a dominant Nb 4d character [10], shows three main structures around 0.5, 1.5 and 2.7 eV with the two structures near the Fermi level (E_F) having higher intensity. Those structures have not been observed clearly in a previous report [11] most probably due to difference in procedure to get fresh surfaces; scraping or fracturing. For the normal state spectrum near E_F measured at 18 K, we observe a nearly flat density of states (DOS) in the regions at least up till 90 meV with a Fermi edge structure, where the leading edge mid point is nearly located at E_F. In contrast, once we cool the samples below T_c, the spectral shape changes drastically. We see a sharp peak just below E_F followed by a dip and a hump. To see the superconducting gap more clearly, we enlarge the spectrum at 5.5 K as shown in Fig. 2 with open circles. Now it is clear that the spectrum has a peak at 4.5 meV and a shift of the leading edge, indicating opening of the superconducting gap. To estimate the gap value, we use the Dynes function [12] to fit the experimental spectrum. The Dynes function is multiplied by the Fermi function of 5.5 K and convolved with a gaussian with a full width at half maximum of the known energy resolution. In Fig. 2, we show a representative fitting result with a gap value Δ = 3.0 meV and a thermal broadening parameter Γ = 0.005 meV (curve), which reproduces the experimental result very well. The obtained Δ = 3.0 meV corresponds to a reduced gap value of 4, classifying Nb$_3$Al into a medium- to strong-coupling superconductor, consistent with the previous thermodynamic and tunneling measurements [7]. In addition, the small value of Γ may imply that the superconducting gap of Nb$_3$Al is nearly isotropic.

As for the structures beyond the superconducting coherent peak, we see a dip structure around 20 meV and a hump structure around 40 meV. The energy scale of these features are surely larger than those observed in Pb, where a dip and a hump are observed around 10 and 20 meV, respectively. This relates to the fact that the width of phonon DOS in Nb$_3$Al ($\sim$40 meV) is larger in Pb ($\sim$10 meV). The shape of the peak–dip–hump structure is consistent to that obtained by
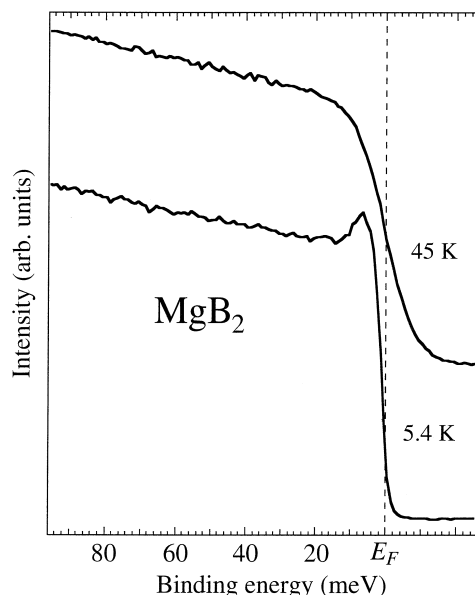
Fig. 3. High-resolution photoemission spectra of polycrystalline MgB$_2$ (T_c = 36.5 K) measured at superconducting state (5.4 K) and normal state (45 K) using the He Iα resonance line.

tunneling measurements using thin films [13]. Thus, the present results show that the photoemission spectroscopy is reliable and powerful technique to investigate superconducting electronic structures even for compounds.
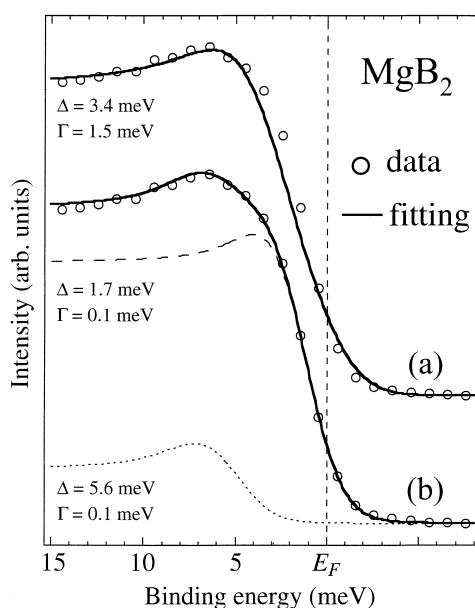

Fig. 4. Representative fitting result using: (a) a single isotropic Dynes function (curve) with Δ = 3.4 meV and Γ = 1.5 meV and (b) weighted sum of two Dynes functions with Δ = 1.7 meV and Γ = 0.1 meV (smaller gap, broken lines) and with Δ = 5.6 meV and Γ = 0.1 meV (larger gap, dotted lines). Open circles are the superconducting spectrum of MgB$_2$ measured at 5.4 K.

Next topic is on the superconducting electronic structure of MgB$_2$, which is a high T_c intermetallic superconductor. In Fig. 3, we show high-resolution photoemission spectra of MgB$_2$ measured at 5.4 K (superconducting state) and 45 K (normal state). While normal-state spectrum is characterized by a Fermi edge like structure, the superconducting-state spectrum shows a peak and a shift of the leading edge, indicative of opening of the superconducting gap. An enlargement of the near-E_F region has been shown in Fig. 4 with open circles, together with fitting results (curve). We find, besides the peak around 7 meV, a weak shoulder structure around 3 meV. These features could not be clearly distinguished by a recent high-resolution photoemission study [14] maybe due to the difference in sample quality and/or preparation to get fresh surfaces; scraping or fracturing. The unusual spectral features are more easily seen if one compare the superconducting spectrum of MgB$_2$ with that of Nb$_3$Al. While the leading edge of Nb$_3$Al is very sharp and actually resolution limited, that of MgB$_2$ is broad. Further, the leading edge mid point of MgB$_2$ is not so large compared to that of Nb$_3$Al in spite of the large difference in T_c. This indicates that the superconducting gap of MgB$_2$ is not a simple isotropic one.

To get further insight into the superconducting gap, we tried to fit using single Dynes functions with isotropic and anisotropic gaps. For the anisotropic gap, we used proposed anisotropic gap functions from the group theory [15,16]. We find single Dynes functions with both isotropic gap and anisotropic gap could not reproduce the spectral shape with any sets of parameters, especially for the shoulder structure. In Fig. 4(a), we only show a representative result using a single isotropic Dynes function with Δ = 3.4 meV and Γ = 1.5 meV. Therefore, assuming the shoulder structure to result from another gap, we use two Dynes functions and find weighted sum of them can reproduce the spectral very well as shown in Fig. 4(b), where broken and dotted lines represent two single Dynes functions with Δ = 1.7 meV and Γ = 0.1 meV as well as Δ = 5.6 meV and Γ = 0.1 meV. We make the intensity of the Dynes function with the smaller gap five times larger than that for the larger gap. From the earlier analysis, we come to know that the superconducting gap is neither a simple isotropic gap nor a simple anisotropic one, but rather consists of two dominant components. This is consistent with results reported from several groups using different experimental techniques [17].

We further perform the same fitting to the temperature dependent spectra in order to obtain the temperature dependence of the two gaps, assuming the intensity ratio to be constant. In Fig. 5, we plot the gap values as a function of temperature. Surprisingly, we find the both of the larger and smaller gaps close nearly the bulk transition temperature. This shows evidence that the smaller gap represents the bulk electronic structure. The temperature dependence of the smaller gap follows the BCS prediction very well. However, that of the larger gap deviates from the BCS prediction at higher temperature, whose behavior is similar
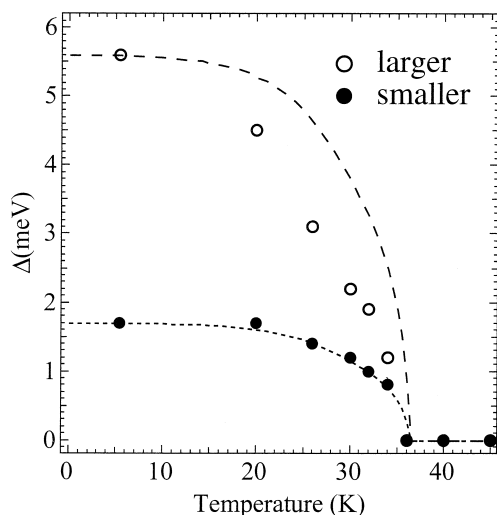
Fig. 5. Obtained values of the two gaps as a function of temperature. Dotted and broken lines are the BCS predictions for temperature-dependent gap using $\Delta(0) = 1.7$ and 5.6 meV, respectively.

to that obtained on MgB_2/Ag and MgB_2/In junctions [18]. From these analyses, the reduced gap values are 1.08 and 3.56 for smaller and larger gaps, respectively. It is worth noticing that the ratio of the two gaps at 5.4 K is nearly one-third, which is very close to the value predicted by recent band structure calculations [19]. In addition, we see a weak structure beyond the peak in the MgB_2 spectrum. We find the energy position of this structure is about 18 meV. Here we just note that a recent neutron scattering study has reported a phonon structure at the same energy [20].

4. Summary

High-resolution photoemission results on Nb_3Al show a peak−dip−hump structure characteristic of the strong-coupling spectral shape as well as the superconducting gap with the reduced gap value of ~ 4. These results characterize that the Nb_3Al we studied is a medium- to strong-coupling superconductor, and also establishes photoemission spectroscopy as a reliable technique to investigate superconducting electronic structures even for compounds. As for MgB_2, high-resolution photoemission results show

non-simple isotropic gap and numerical analyses tell us that superconducting gap of MgB_2 is most likely a multi-component gap. This result provides direct information on the superconducting electronic structure of MgB_2 which yields such a high T_c.

Acknowledgments

References

[1] R. Hesper, L.H. Tjeng, A. Heeres, G.A. Sawatzky, Phys. Rev. Lett. 85 (2000) 1970.

[2] F. Reinert, G. Nicolay, B. Eltner, D. Ehm, S. Schmidt, S. Hufner, U. Probst, E. Bucher, Phys. Rev. Lett. 85 (2000) 3930.

[3] A. Chainani, T. Yokoya, T. Kiss, S. Shin, Phys. Rev. Lett. 85 (2000) 1966.

[4] M.R. Norman, et al., Phys. Rev. Lett. 79 (1997) 3506.

[5] Z.-X. Shen, J.R. Schrieffer, Phys. Rev. Lett. 78 (1997) 1771.

[6] T. Yokoya, T. Kiss, T. Watanabe, S. Shin, M. Nohara, H. Takagi, T. Oguchi, Phys. Rev. Lett. 85 (2000) 4952.

[7] J.C. Carbotte, Rev. Mod. Phys. 62 (1990) 1027.

[8] J. Nagamatsu, N. Nakagawa, T. Muranaka, Y. Zentani, J. Akimitsu, Nature 410 (2001) 63.

[9] Y. Takano, et al., Appl. Phys. Lett. 78 (2001) 2941.

[10] W.E. Pickett, K.M. Ho, M.L. Cohen, Phys. Rev. B 19 (1979) 1734.

[11] M. Grioni, et al., Phys. Rev. B 43 (1991) 1216.

[12] R.C. Dynes, V. Narayanamurti, J.P. Garno, Phys. Rev. Lett. 41 (1965) 1509.

[13] J. Kwo, T.H. Geballe, Phys. Rev. B 23 (1981) 3230.

[14] T. Takahashi, T. Sato, S. Soma, T. Muranaka, J. Akimitsu, Phys. Rev. Lett. 86 (2001) 4915.

[15] P. Senor, C.-T. Chen, Phys. Rev. B 65 (2002) 012505.

[16] S. Haas, K. Maki, Phys. Rev. B 65 (2002) 020502.

[17] F. Bouquet, et al., Europhys. Lett. 56 (2001) 856.

[18] A. Plecenik, Ŝ. Beñaĉka, P. Kúš, M. Grajcar, Physica C 368 (2002) 251.

[19] A.Y. Liu, I.I. Mazin, J. Kortus, cond-mat/0103570.

[20] T.J. Sato, K. Shibata, Y. Takano, Phys. Rev. Lett. 87 (2001) 087005.