Evidence of strong electron-phonon interaction in superconducting MgB$_2$ from electron tunneling.

A. I. D’yachenko, V. Yu. Tarenkov, A. V. Abal’oshev, and S. J. Lewandowski
Instytut Fizyki Polskiej Akademii Nauk, Al. Lotników 32, 02-668 Warszawa, Poland

We report tunneling measurements of the electron-phonon (e-ph) interaction in superconducting MgB$_2$ using the MgB$_2$-I-Nb junctions, where I stands for insulator. The phonon structure in tunneling density of states in MgB$_2$ clearly indicates strong e-ph coupling for the E$_{2g}$ in-plane boron phonons in a narrow range around 60 meV. The Eliashberg spectral function $\alpha^2(\omega)F(\omega)$ reconstructed from the tunneling data, exhibits significant additional contribution into e-ph interaction from other vibrations such as acoustic ($\sim 38$ meV) and optical ($\sim 90$ meV) bands. Our results are in reasonable agreement with neutron scattering experiments, and also to some data of Raman and infrared spectroscopy.

PACS numbers: 74.25.Kc, 74.50.+r, 74.70.-b

The recent discovery of superconductivity near 40K in MgB$_2$, stimulated a renewed interest in the pairing mechanism of cuprate superconductors. It is generally believed that understanding superconductivity in the simple binary MgB$_2$ compound should be much easier than in the high-T$_c$ superconductors. Theoretical ab initio calculations show that dominant contribution to Eliashberg function in MgB$_2$ comes from the Raman-active E$_{2g}$ phonons near the Brillouin zone center at $\sim 75$ meV. This conclusion is supported by Raman studies, which indicate a mode at $\sim 600 - 620$ cm$^{-1}$ ($\sim 74 - 77$ meV) with a very large width $\sim 200$ cm$^{-1}$ (25 meV), usually related to anharmonic effects. However, there has been no consensus about the energy and role of the E$_{2g}$ mode, because this mode has not been identified unambiguously by inelastic neutron scattering or in Raman experiments.

Tunneling spectroscopy can provide a direct measure of the Eliashberg electron-phonon spectral function $\alpha^2(\omega)F(\omega)$, which can reveal the importance of different phonon modes in the superconductivity of MgB$_2$. Here F(\omega) is phonon density of states and $\alpha(\omega)$ is an effective electron-phonon coupling function for phonons of energy $\hbar\omega$. Tunneling conductivity $\sigma = dI/dV$ gives direct information about the density of states $N(\omega) \propto \text{Re}\{\omega/\omega^2 - \Delta(\omega)^2\}^{1/2}$ related to quasi-particle excitations in the superconductor, where $\Delta(\omega)$ is a complex and energy dependent gap function. $\alpha^2(\omega)F(\omega)$ can be obtained by the inversion of the Eliashberg equations for $\Delta(\omega)$. The method is based on the determination of $\Delta(\omega)$, therefore it non-sensitive to the presence of non-superconducting impurity phases in MgB$_2$. It can give comparable features in Raman and infrared spectra at energy range typical for the superconducting MgB$_2$.

We report here high resolution superconducting tunneling spectroscopy measurements of the phonon structure of MgB$_2$. We used tunnel point-contact junctions MgB$_2$-I-MgB$_2$, where I is a natural tunnel barrier. We found that the second derivative of tunnel current $d^2I/dV^2$ exhibits features, which coincide with peaks in the phonon density of states (DOS) $F(\omega)$ obtained from neutron scattering, Raman and infrared spectroscopy. Numerical treatment of the experimental data indicates that the observed phonon structures between 55 - 65 meV and near 90 meV are very strongly coupled to the electrons.

The investigated samples were prepared by compacting commercially available MgB$_2$ powder (Alfa Aesar, purity 98%) at high pressure ($P > 5$ GPa) into thin rectangular bars of about $1 \times 0.6 \times 0.08$ mm$^3$. Details of the sample preparation are described elsewhere. The samples revealed a superconductive transition temperature (onset) at $T_c = 38$ K and a high critical cur-
rent density $J_c > 10^7 \text{A/cm}^2$ at $T = 4.2$ K, considerably larger than the current densities employed in the tunneling measurements. Therefore, current depairing had no effect on the phonon-mediated region of tunneling data. The point-contact junctions were obtained by pressing Nb wire ($\phi \sim 50\mu$m) into the surface of the sample. The tunnel characteristics were measured using a standard four probe lock-in technique.

In a number of Andreev S-N-S and tunnel S-I-S experiments at $T = 4.2$ K we observed two gaps $\Delta$, the smaller gap value of about 3 meV, and a larger gap close to 7 meV, both results in agreement other measurements \cite{22}. In tunnel S-I-S junctions we observed usually only the smaller gap with s-wave symmetry. Let us observe that the smaller $\sim 3$ meV and the larger $\sim 7$ meV gaps are associated with the 3d parts and quasi-2D sheets of the Fermi surface, respectively \cite{3}. The experimental detection of the single small gap is possible in the case of tunneling perpendicular to the honeycomb boron planes. Due to the strong directionality of the tunnelling experiments, the probability of normal injection is maximal and electronic properties are mainly probed along the direction perpendicular to the junction surface, i.e. in our case normal to the born planes.

At high bias voltages ($> 20$ meV) the $dI/dV(V)$ data reveal a structure (Fig.1), which in the case of conventional superconductors is characteristic of phonon effects \cite{23}. To investigate this structure in more detail, we used the second derivative of the tunneling current $d^2I/dV^2$, obtained by standard harmonic detection, as well as by straightforward numerical differentiation of the tunneling conductivity $dI/dV$. Both methods give analogous results. In these spectra, we observed sometimes peaks at $\sim 17$ meV and $\sim 28$ meV, which are not intrinsic to clean MgB$_2$. The structure near 28 meV most likely arises from a thin pure Mg layer in proximity with the superconducting MgB$_2$ (analogous structures have been observed in Ag-I-Mg/Nb junctions \cite{24}). For this reason, we excluded these singularities from further considerations, especially as they do not affect the subsequent treatment of the raw data.

The plot $d^2I/dV^2(V)$, shown in Fig. 2, at $eV > 30$ meV directly reflects the electron-phonon spectral function $\alpha^2(\omega)F(\omega)$ of MgB$_2$; in particular the dips in $d^2I/dV^2$ correspond to peaks in $\alpha^2(\omega)F(\omega)$. Peaks in $F(\omega)$ should be reproduced in $\alpha^2(\omega)F(\omega)$ at approximately the same energy, even if the coupling factor $\alpha^2(\omega)$ is strongly energy dependent. The $dI/dV$ and $d^2I/dV^2$ data were used to obtain the normalized tunneling conductance $\sigma/\sigma_N$, where $\sigma_N$ is a normal-state (background) conductance, and the tunneling density of states $N(\omega)$; the relevant numerical method is given in \cite{23}.

The normalized density of states $n(\omega) = N(\omega)/N_{BCS}(\omega) - 1$, proportional to the smoothed $dI/dV$ curve, is shown in Fig. 3. Here $N_{BCS}(\omega)$ is the broadened BCS density of states (Dynes function).

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{fig2.png}
\caption{Second derivative $d^2I/dV^2$ plots of a MgB$_{2}$-I-Nb junction. The minima in the curve correspond to peaks in Eliashberg function $\alpha^2(\omega)F(\omega)$. Voltage origin is at $\Delta_{MgB_2} + \Delta_{Nb}$.}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{fig3.png}
\caption{The tunneling density of states of MgB$_{2}$, $n(\omega)$, normalized to the BCS density of states. Experimental data are plotted as a solid line; dashed line shows $n(\omega)$ calculated from Eliashberg function $\alpha^2(\omega)F(\omega)$.}
\end{figure}

The $dI/dV$ curve displays a well-known behavior due to the phonon mode reflection in tunneling S-I-S conduction \cite{13, 16}, namely, sharp drops of $dI/dV$ near the phonon emission thresholds, to which correspond dips in $d^2I/dV^2$. We can rule out very weak inelastic process of phonon-assisted tunneling, because such process increases the tunneling conductance near the peaks in $F(\omega)$ and gives corresponding peaks in $d^2I/dV^2$ \cite{23}. The common feature of all measured $d^2I/dV^2$ curves is the reproducibility of dip positions at $\sim 40$, $55 \div 65$ and $90 \div 92$ meV, measured from the gap sum.
$\Delta_{N_{gB_2}} + \Delta_{N_6}$ (Fig.2). There is a close correlation in the peak positions in our $d^2 I/dV^2$ data and those in the generalized phonon density of states (GPDOS) as measured by inelastic neutron scattering data [13, 14]. The first tunneling peak near 40 meV arises from acoustic modes of MgB$_2$. The well-resolved tunneling structures at $\sim 60$ meV and $\sim 90$ meV are associated with optical vibration of the born atoms. As it is well known, in the vicinity of peaks in $\alpha^2(\omega)F(\omega)$, the gap function $\Delta(\omega)$ has a strong energy dependence and large $\text{Im}\{\Delta(\omega)\}$ [15]. At $h\omega \gg |\Delta|$ tunneling density $N(\omega) = 1 + (1/2)\text{Re}\{\Delta^2(\omega)/\omega^2\}$, so the phonon structure in $N(\omega)$ is weighted by the factor $(\delta\Delta/\omega)^2$. In the strong coupling limit (the McMillan’s electron-phonon coupling constant $\lambda$ of the order of 1) variation of $\Delta(\omega)$, $\delta\Delta \sim \Delta$, and the deviation in $N(\omega)$ is of the order $1 - 2\%$ (Fig.3). Even without detailed calculations one can see that since the amplitude of tunneling structure at 40 meV is roughly one half of that at 60 meV (c.f. Fig.2) and the optical phonon energy is 1.5 times higher, the actual electronic coupling to the phonons at 60 meV may be as much as $(1.5)^2 \sim 3$ times greater than that for acoustic phonons at $\sim 40$ meV. On the other hand, it is evident, that the optical modes at $\sim 90$ meV give approximately the same contribution than that of modes at 60 meV. Such conclusion, obtained directly from the tunneling data, is independent of the anisotropy of the energy gap $\Delta(k)$ and agrees well with similar estimations based upon Raman and infrared spectra measurements [13, 14].

An exact determination of $\alpha^2(\omega)F(\omega)$ requires full inversion of the Eliashberg equations. We used normalized conductivity $\sigma/\sigma_N$ for self-consistent calculation of the spectral function $\alpha^2(\omega)F(\omega)$ with a standard inversion program [15], which allows to take into account the already mentioned possible presence of a thin Mg layer on the surface of MgB$_2$, in which reduced superconductivity is induced by proximity effect. The gap edge region below 26 meV was cut off to avoid a divergence of DOS. The obtained "experimental" function $\alpha^2(\omega)F(\omega)$ is shown in Fig.4 together with theoretical ab initio phonon density of states $F_i(\omega)$ combined with neutron scattering [16]. In isotropic limit for $\lambda = 0.9$ and $\mu^* = 0.1$ Eliashberg function yields $T_c \approx 38$ K, and $|\Delta| = 6.5$ meV. Neutron GPDOS [13, 19] differs in its detailed shape from $F_i(\omega)$, but peaks in $F_i(\omega)$, GPDOS and our $\alpha^2(\omega)F(\omega)$ all occur at the same energies. They all show pronounced minima at 40 $\div$ 50 meV and maxima at 37, 62 and 90 $\div$ 92 meV. The peak in $\alpha^2(\omega)F(\omega)$, responsible for acoustic bands ($\sim 38$ meV), is strongly suppressed, but does not disappear completely, as in the theoretical calculations [1]. We do not see also the prevailing amplitude of one mode at 60 $- 70$ meV, as predicted for the theoretical function $\alpha^2(\omega)F_i(\omega)$ [1, 16], although our experimental data indicate two bands of optical modes corresponding primarily to the born motions at $\sim 60$ and $\sim 90$ meV. For comparison, in Fig.4(b) the calculated phonon dispersion curves are shown along the high-symmetry directions of the Brillouin zone [19]. As seen, the E$_{1u}$ and A$_{2u}$ infrared active modes give noticeable contribution to the $\alpha^2(\omega)F(\omega)$ at 40 and 50 meV, respectively. They have been also observed in infrared absorption experiments at 333 cm$^{-1}$ (41 meV) and 387 cm$^{-1}$ (48 meV) [14]. Optical studies in the c-axis oriented superconducting MgB$_2$ films show two strong phonon peaks at 380 cm$^{-1}$ (48 meV) and 480 cm$^{-1}$ (60 meV) [19]. The peak in the "experimental" $\alpha^2(\omega)F(\omega)$ at 60 $\div 65$ meV arises mainly from the E$_{2g}$ phonon modes with wave vector along the $\Gamma$–A direction [Fig.4(b)]. This phonon mode is Raman-active. In most investigations the Raman scattering in MgB$_2$ revealed a broad peak near 620 cm$^{-1}$ (78 meV) but, Meletov et al...
have observed a peak near $\sim590$ cm$^{-1}$ (73 meV). They investigated ceramic MgB$_2$ samples using a micro-Raman system, which rendered possible the identification of small crystalline grains of MgB$_2$, whose Raman spectra differ drastically from those of MgO or metallic Mg inclusions. After homogenisation of ceramic samples induced by high pressure (> 5 GPa) the $E_{2g}$ phonon mode was observed at 500 – 550 cm$^{-1}$ (62 – 68 meV) [2] and second-order Raman signal from the acoustic phonon mechanism of superconductivity in MgB$_2$ (Fig.4).

In summary, we have obtained high resolution tunneling spectra of MgB$_2$ with $T_c = 38$ K. The tunneling density of states deviated from the standard Dynes function above 30 meV in a way, which is characteristic of phonon effects. The second derivative curve displays dips at energies, which correspond to peaks in the phonon density of states measured by neutron scattering an also to some data of Raman and infrared spectroscopy. The spectral function $\alpha^2(\omega)F(\omega)$ has been reconstructed from tunneling data. The relatively big structure in tunneling spectra confirms strong coupling of electrons to optical $E_{2g}$ phonon modes at $\sim60$ meV. However, other vibrations such as acoustic ($\sim38$ meV) and optical ($\sim90$ meV) bands are also important and give noticeable peaks in the tunneling spectra. Therefore, the predominance of the $E_{2g}$ phonon mode near the $\Gamma$ point in the electron-phonon mechanism of superconductivity in MgB$_2$ may be questioned.

This work was supported by Polish Government (KBN) Grant No PBZ-KBN-013/T08/19.

---

* permanent address: Donetsk Physico-Technical Institute, Ukrainian National Academy of Sciences, R.Luxemburg St. 72, 340114 Donetsk, Ukraine.

[1] Y. Kong, O.V. Dolgov, O. Jepsen, and O.K. Andersen, Phys. Rev. B 64, 20501 (2001).
[2] J.M. An and W.E. Pickett, Phys. Rev. Lett. 86, 4366 (2001).
[3] K.-P. Bohnen, R. Heid, and B. Renker, Phys. Rev. Lett. 86, 5771 (2001).
[4] Amy Y. Liu, I.I. Mazin, and Jens Kortus, Phys. Rev. Lett. 87, 087005 (2001).
[5] J. Kortus, I.I. Mazin, K.D. Belashchenko, V.P. Antropov, and L.L. Boyer, Phys. Rev. Lett. 86, 4656 (2001).
[6] A.F. Goncharov, V.V. Struzhkin, E. Gregoranz, Jingzhu Hu, R.J. Hemley, Ho-kwang Mao, G. Lapertot, S.L. Bud’ko, and P.C. Canfield, Phys. Rev. B 64, 100509 (2001).
[7] J. Hlinka, I. Gregora, J. Pokorný, A. Plecenik, P. Kúš, L. Satrapinsky, Š. Beňačka, Phys. Rev. B 64, 140503 (2001).
[8] K. Kunc, L. Loa, K. Syassen, R.K. Kremer, K. Ahn, cond-mat/0105402.
[9] X.K. Chen, M.J. Konstantinović, J.C. Irwin, D.D. Lawrie, and J.P. Francke, Phys. Rev. Lett. 87, 157002 (2001).
[10] P.M. Rabilo, S. Bahr, and C. Thomsen, Phys. Stat. Sol. B 226, R9 (2001).
[11] V.V. Struzhkin, A.F. Goncharov, R.J. Hemley, H.K. Mao, G. Lapertot, S.L. Bud’ko, and P.C. Canfield, cond-mat/0106576.
[12] K.P. Meletov, J. Arvanitidis, M.P. Kulakov, N.N. Kolesnikov, and G.A. Kouroukis, cond-mat/0110511.
[13] J.J. Tu, G.L. Carr, V. Perebeinos, C.C. Homes, M. Strongin, P.B. Allen, W.N. Kang, E.M. Choi, H.J. Kim, and Sung-Ik Lee, Phys. Rev. Lett. 87, 277001 (2001).
[14] C.S. Sundar, A. Bharathi, M. Premila, T.N. Sairam, S. Kalavathi, G.L.N. Reddy, V.S. Sastri, Y. Hariharan, and T.S. Radhakrishnan, cond-mat/0104354.
[15] J.R. Schrieffer, D.J. Scalapino, and J.W. Wilkins, Phys. Rev. Lett. 10, 336 (1963).
[16] W.L. McMillan and J.M. Rowell, Phys. Rev. Lett. 14, 108 (1965).
[17] A.A. Galkin, A.I. D'yachenko, and V.M. Svistunov, Sov. Phys. JETP 39, 1115 (1974).
[18] R. Osborn, E.A. Goremychkin, A.I. Kolesnikov, and D.G. Hinks, Phys. Rev. Lett. 87, 017005 (2001).
[19] T. Yildirim, O. Güseren, J.W. Lynn, C.M. Brown, T.J. Udovic, Q. Huang, N. Rogado, K.A. Regan, M.A. Hayward, J.S. Slusky, T. He, M.K. Haas, P. Khalifah, K. Inumaru, and R.J. Cava, Phys. Rev. Lett. 87, 037001 (2001).
[20] E.S. Clementyev, K. Conder, A. Furrer, and I.L. Sashin, Eur. Phys. J. B 21, 465 (2001).
[21] A.I. D'yachenko, V.Yu. Tarenkov, R. Szmyczak, A.V. Abal'oshev, I.S. Abal'osheva, S.J. Lewandowski, and L. Leonyuk, Phys. Rev. B 61, 1500 (2000).
[22] C. Buzea and T. Yamashita, Supercond. Sci. Technol. 14, R115 (2001).
[23] E. Wolf, Principles of Electron Tunneling Spectroscopy (Oxford University Press, New York, 1985).
[24] D.M. Burnell and E.L. Wolf, Phys. Lett. A 90, 471 (1982).
[25] J.G. Adler, Solid State Commun. 7, 1635 (1969).