Eur. Phys. J. B **21**, 465–467 (2001)

THE EUROPEAN PHYSICAL JOURNAL B

EDP Sciences

Phonon density-of-states in the new superconductor MgB_2

E.S. Clementyev[1], K. Conder[1], A. Furrer[1,a], and I.L. Sashin[2]

[1] Laboratory for Neutron Scattering, ETH Zurich & Paul Scherrer Institute, 5232 Villigen PSI, Switzerland
[2] FLNP, Joint Institute for Nuclear Research, 141980 Dubna, Russia

Received 19 March 2001

Abstract. We provide here experimental data on the phonon density-of-states of MgB_2 obtained by the inelastic neutron scattering technique. The measurements were performed for the natural boron-based magnesium diboride with use of a time-of-flight neutron spectrometer. Several phonon bands were observed in the phonon spectrum at energies of about 33, 55, 82 and 99 meV. We show that the cut-off energy of the density-of-states occurs at around 105 meV which is much higher than expected so far from heat-capacity data and partially explains the high T_c value observed for MgB_2. The characteristic phonon energies are indicative of an intermediate coupling regime in this compound. We conclude that a much needed neutron experiment aimed at the study of the isotopic effect in the phonon density-of-states of MgB_2 is conceivable.

PACS. 74.70.-b Superconducting materials (excluding high-T_c compounds) – 74.25.Kc Phonons – 78.70.Nx Neutron inelastic scattering

Rapid Note

Recently, superconductivity at $T_c \approx 39$ K was observed in magnesium diboride MgB_2 by Nagamatsu *et al.* [1]. The transition temperature is surprisingly high for a non-cuprate material and much above the limit expected for a classical superconducting compound. The observation of a boron isotope effect on T_c suggests a phonon-mediated pairing mechanism [2]. In the Bardeen-Cooper-Schrieffer (BCS) theory [3] T_c scales with the Debye energy when the effective attractive interaction between electrons is due to phonons. A detailed picture of the phonon density-of-states is therefore highly desirable. This was the motivation for our experimental study. Inelastic neutron scattering (INS) is a most powerful tool to probe the characteristic energies of lattice vibrations.

MgB_2 crystallizes in the hexagonal AlB_2-type structure [4] in which the B ions constitute graphite-like sheets in the form of primitive honeycomb lattices separated by hexagonal layers of Mg as illustrated in Figure 1. It was concluded from band structure calculations [5] that the charge carriers reside in the boron planes which are metallic due to the covalent B-B bonds. Thus there is a structural similarity to the high-T_c cuprate superconductors in which the charge carriers are also located in the CuO_2 planes. It is well known that the phonon energies of atoms in a two-dimensional network are enhanced compared to a three-dimensional one. For example, the phonon density-of-states of (two-dimensional) graphite extends to 198 meV [6], whereas for (three-dimensional) diamond the phonon cut-off energy is reduced to 164 meV [7].

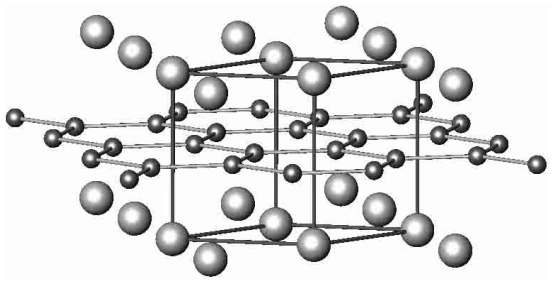

Fig. 1. Crystal structure of MgB_2. Magnesium and boron are drawn as big and small spheres, respectively. The structure can be considered as a sequence of alternating graphite-like layers of hexagonal symmetry. The boron atoms are six-fold coordinated and situated inside trigonal prisms of the magnesium atoms.

In the BCS theory the superconducting transition temperature is given by the semiempirical McMillan formula [8]

$$T_c = \frac{\hbar\omega_D}{1.45\,k_B} \exp\left[-\frac{1.04(1+\lambda)}{\lambda - \mu_c^*(1+0.62\,\lambda)}\right],$$

where ω_D is the Debye frequency, λ the effective electron-phonon coupling constant, and μ_c^* the repulsive Coulomb interaction parameter which is of the order of 0.15 for

[a] e-mail: albert.furrer@psi.ch

all superconductors, *i.e.*, T_c can be raised by increasing the parameters ω_D and λ. The arrangement of the boron atoms in a two-dimensional network is obviously beneficial to achieve a high value of ω_D in MgB_2 as demonstrated in the present work.

Commercially available MgB_2 (Alfa) was used in this work. The phase purity was at least 96%. As checked with X-ray diffraction the primary phase of the material was MgB_2 with some traces of MgO. Magnetic measurements (SQUID magnetometer) showed the onset of superconductivity at 38 K in good agreement with Nagamatsu *et al.* [1].

The INS measurements were carried out at room temperature with use of the time-of- flight spectrometer KDSOG-M of inverted geometry located at the IBR-2 pulsed reactor in Dubna. The final energy was fixed at $E_f = 4.9$ meV. A pyrolitic graphite analyzer was used in combination with a beryllium filter to suppress higher-order contaminations. The energy resolution was 5 to 8% of the incoming energy. Data were taken at scattering angles ranging from 80 to 140 degrees. Two detector arrays located symmetrically with respect to the incoming beam were used in reflection scattering geometry. The neutron beam size was 14×16 cm^2. Natural boron is a mixture of two isotopes: ^{11}B (80% abundance) and ^{10}B (20%) with absorption cross-sections for thermal neutrons of 0.006 and 3835 barns, respectively. The latter value makes INS experiments on the ^{10}B-rich sample very difficult. Natural boron has an absorption cross-section of 767 barns (2200 m/s neutrons). To increase the counting rate and partially compensate for the very strong neutron absorption by natural boron a sample with a huge surface area of more than 300 cm^2 was prepared. The MgB_2 powder was disposed into two envelopes made from Al foils. These envelopes were placed on two Cd plates perpendicular to the direction of the outgoing beam. The mass of the MgB_2 powder sample was 60 g. In order to determine the instrument background and the contribution from the Al phonon density-of-states, the Cd plate and the Al foil on the Cd substrate were measured without the MgB_2 sample using the same experimental conditions. Multiphonon corrections based on two- and three-phonon scattering [9] were carried out self-consistently and subtracted from the raw data. The Debye-Waller attenuation was obtained in the isotropic "single-atom lattice" approximation, and the raw data were corrected accordingly.

Figure 2 shows the generalized phonon density-of-states (GDOS) of MgB_2 determined from the neutron spectra measured at room temperature. The data taken at different scattering angles (80 to 140 degrees) were averaged to get a good approximation of the incoherent neutron scattering function. The momentum transfer was large (up to 7 Å^{-1}) for high energy transfers providing a good average over several Brillouin zones. As no correction for the energy dependence of neutron absorption was made, the relative intensities of peaks must be treated with caution. Several phonon bands show up in the GDOS. In particular, there are maxima at about $E = 33$ (narrow spectral feature), 55 (broad peak), 82 and 99 meV. The positions of the latter two peaks are very approximate because of a rather big width in energy of the time-of-flight channels for high energies. The observed phonon bands are in reasonable agreement with the zone-center phonon modes estimated by the LAPW method [5]. For example, the peak at $E \sim 82$ meV must be related to the boron B_{1g} and E_{2g} modes according to [5]. Our experiment covers the entire energy range of lattice vibrations in MgB_2. We estimate the cut-off energy of the GDOS to be about 105 meV. The cut-off energy obtained in the present work is considerably higher than extrapolated from heat-capacity data (65 meV [2], 69 meV [10]). The GDOS shown in Figure 2 reflects the partial phonon density-of-states of both magnesium and boron. The lighter boron atoms contribute mainly to the high frequency part of the phonon spectrum.

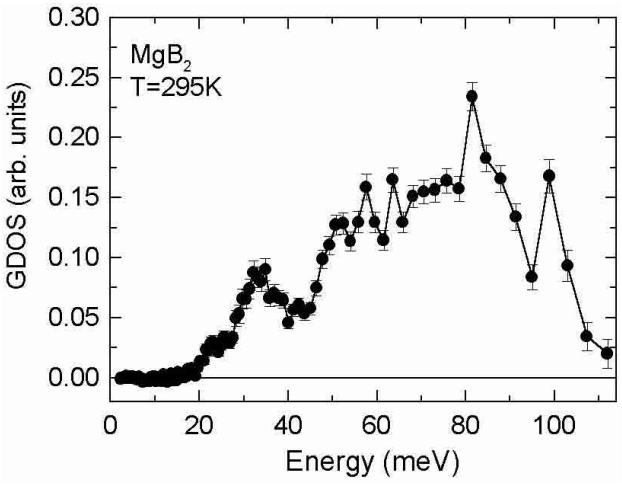

Fig. 2. Generalized phonon density-of-states (GDOS) in MgB_2 at $T = 295$ K. The data are corrected for multiphonon scattering, Debye-Waller attenuation and background contributions as explained in the text. The line connects the experimental points.

After the characteristic phonon energy $\hbar\omega_D$ is established one can easily derive the effective electron-phonon coupling parameter λ from the McMillan formula. Thus the present controversy whether the superconductivity of MgB_2 is governed by a weak ($\lambda \ll 1$), intermediate ($\lambda \approx 1$), or strong ($\lambda \gg 1$) coupling regime can be solved. We found $\lambda \approx 1$ (for $\hbar\omega_D \approx 80$–105 meV), therefore, MgB_2 has to be placed unambiguously into the intermediate coupling regime. This is in contradiction to the conclusions from an NMR study [11] which places MgB_2 into the strong coupling regime.

Another important message from our measurements is the possibility to collect neutron data with reasonable statistics on ^{10}B-rich MgB_2 samples. We believe that an INS experiment on a $Mg^{10}B_2$ isotopic sample is conceivable. Such a measurement taken along with a relatively easy experiment on a $Mg^{11}B_2$ sample will provide crucial information on the role played by phonons in superconductivity of the magnesium diboride.

We are thankful to A. Shengelaya for his help in the sample characterization.

References

1. J. Nagamatsu, N. Nakagawa, T. Muranaka, Y. Zenitani, J. Akimitsu, Nature **410**, 63 (2001).
2. S.L. Bud'ko, G. Lapertot, C. Petrovic, C.E. Cunningham, N. Anderson, P.C. Canfield, Phys. Rev. Lett. **86**, 1877 (2001), cond-mat/0101463.
3. J. Bardeen, L.N. Cooper, J.R. Schrieffer, Phys. Rev. **108**, 1175 (1957).
4. H.J. Goldschmidt, *Interstitial Alloys* (Butterworth, 1967), p. 427.
5. J. Kortus, I.I. Mazin, K.D. Belashchenko, V.P. Antropov, L.L. Boyer, Phys. Rev. Lett. **86**, 4656 (2001), cond-mat/0101446.
6. R.M. Nicklow, N. Wakabayashi, H.G. Smith, Phys. Rev. B **5**, 4951 (1972).
7. J.L. Warren, J.L. Yarnell, G. Dolling, R.A. Cowley, Phys. Rev. **158**, 805 (1967).
8. W.L. McMillan, Phys. Rev. **167**, 331 (1968).
9. A. Sjoelander, Ark. Fys. **14**, 315 (1958).
10. R.K. Kremer, B.J. Gibson, K. Ahn, cond-mat/0102432.
11. H. Kotegawa, K. Ishida, Y. Kitaoka, T. Muranaka, J. Akimitsu, cond-mat/0102334.