We report inelastic neutron scattering measurements of the phonon density–of–states in Mg$^{11}$B$_2$, which has a superconducting transition at 39.2 K. The acoustic phonons extend in energy to 36 meV, and there are highly dispersive optic branches peaking at 54, 78, 89 and 97 meV. A simple Born-von Kármán model is able to reproduce the mode energies, and provides an estimate of the electron-phonon coupling of $\lambda \sim 0.9$. Furthermore, the estimated boron and magnesium contributions to the isotope effect are in qualitative agreement with experiment. The data confirm that a conventional phonon mechanism, with moderately strong electron-phonon coupling, can explain the observed superconductivity.

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There has already been rapid progress in our understanding of the properties of MgB$_2$, since the first announcement of superconductivity with a critical temperature of 39K [1]. Although some have speculated that the high value of $T_c$ must require a novel mechanism for superconductivity [2], initial experimental reports suggest that a conventional phonon mechanism is sufficient. Both the observation of the isotope effect [3,4] and a simple BCS form for the energy gap measured by point-contact tunneling [5] are consistent with phonon-mediated superconductivity, with the high transition temperature made possible by the high frequency of boron vibrational modes. Initial estimates of phonon frequencies, from electronic structure calculations [7,8], predict that they are consistent with the observed critical temperature, although An et al [6] and Kong et al [8] predict that only a few modes have significant electron-phonon coupling.

The strength and frequency-dependence of the electron-phonon coupling is determined by measurements of both the electron-phonon spectral density, $\alpha^2(\omega)F(\omega)$, and the bare phonon density–of–states (PDOS), $F(\omega)$. $\alpha^2(\omega)F(\omega)$ can be derived by inversion of tunneling data, while $F(\omega)$ is derived from inelastic neutron scattering measurements. In systems where both sets of data are available, $\alpha(\omega)$ usually varies smoothly with frequency so the bare phonon density–of–states provides useful estimates of the frequency moments that determine $T_c$ [6,9].

We report measurements of the phonon density–of–states on an isotopically enriched polycrystalline sample of Mg$^{11}$B$_2$ [10] using time-of-flight neutron spectroscopy at the Intense Pulsed Neutron Source, Argonne National Laboratory. They confirm that boron vibrational frequencies extend as high as 100 meV. The acoustic modes peak at 36 meV, while there are dispersive optic branches at 54, 78, 89 and 97 meV, in good agreement with predictions based on electronic structure calculations [7,8]. A simple Born-von Kármán (BvK) model, which reproduces the energies of the main spectral features, is used to determine the logarithmic frequency moments of the PDOS, which provides a preliminary estimate of the electron-phonon coupling, $\lambda \sim 0.9$ (assuming $\mu^* = 0.1$). We also predict the scale of PDOS shifts with isotopic mass, providing estimates of the magnesium and boron contributions to the isotope effect, although the values depend on the definition of the PDOS used. Given the uncertainties in this analysis, the agreement with experiment is satisfactory, and allows us to conclude that the superconductivity is consistent with a conventional phonon mechanism with moderately strong electron-phonon coupling.

The MgB$_2$ sample was synthesized from a high purity, 3mm diameter Mg rod and isotopic $^{11}$B (Eagle Picher, 98.46 atomic % $^{11}$B). The Mg rod was cut into pieces about 4mm long and mixed with the 200 mesh $^{11}$B powder. The reaction was done under moderate pressure (50 bars) of UHP Argon at 850°C. At this temperature the gas-solid reaction was complete in about one hour. The sample (approximately 20 g) was contained in a 30 ml tantalum crucible with a closely fitting cover. There was surface reaction between the Ta crucible and the reactants at the synthesis temperature but the products of this reaction could be mechanically removed. X-ray diffraction showed some small, unidentified impurity peaks in the resultant powder, but the impurity concentration is small and should have no effect on the PDOS measurements.

The neutron scattering experiments were performed on the Low Resolution Medium Energy Chopper Spectrometer (LRMECS), using incident energies of 200 meV, 130 meV, and 50 meV. LRMECS has continuous detector coverage with scattering angles from 2.4° to 117.5° so that a broad range of wavevector transfers are measured simultaneously, e.g. 2 to 12 Å$^{-1}$ at an energy transfer of 60 meV. This is extremely important in accurate determinations of the PDOS when the scattering is dominantly coherent, as it is in Mg$^{11}$B$_2$. The coherent (incoherent) scattering cross sections are 3.63 barns (0.08 barns) and 5.56 barns (0.21 barns) for Mg and $^{11}$B re-
of the optic phonon branches. We have also performed measurements at 30K, 50K, and 100K. The only statistically significant difference between these temperatures is a small suppression of the peak at 54 meV below $T_c$. In a polycrystalline average, this is a very small effect, but it could still represent a significant change in an individual phonon branch. Interestingly, this is the mode that An and Pickett predict to have the strongest electron-phonon coupling [7], but such conclusions need to await single crystal phonon dispersion measurements.

In a multicomponent system, the inelastic neutron scattering law in polycrystalline samples is given by

$$S(Q, \omega) = \sum_{i=Mg,B} \sigma_i \frac{hQ^2}{2M_i} \exp(-2W_i) \frac{G_i(\omega)}{\omega} [n(\omega) + 1]$$

where $\sigma_i$ and $M_i$ are the neutron scattering cross section and atomic mass of the $i^{th}$ atom and $n(\omega) = [\exp(\hbar\omega/k_BT) - 1]^{-1}$ is the Bose population factor. The generalized PDOS, $G(\omega) = \sum_i G_i(\omega)$, where $G_i(\omega)$ is defined as

$$G_i(\omega) = \frac{1}{3N} \sum_{jq} |\epsilon_i(j, q)|^2 \delta[\omega - \omega(j, q)]$$

which, in turn, defines the Debye-Waller factor, $W_i(Q)$ through

$$W_i(Q) = \frac{hQ^2}{2M_i} \int_0^\infty \frac{G_i(\omega)}{\omega} [2n(\omega) + 1]$$

$\omega(j, q)$ and $\epsilon_i(j, q)$ are the frequencies and eigenvectors, respectively, of the phonon modes. Because the Mg and B partial contributions are weighted by $\sigma/M$, the measured spectrum does not represent the true PDOS, although the stronger cross section and lighter mass of the boron atoms ensure that they make the dominant contribution.

We have analyzed the data in terms of a simple BvK model, that is sufficient to describe the principal features. MgB$_2$ has a hexagonal structure with the lattice parameters $a = 3.082 \text{ \AA}$ and $c = 3.515 \text{ \AA}$ [4]. The lattice is composed of a honeycomb of B atoms separated by hexagonal Mg layers, leading to comparisons with graphite intercalation compounds [5]. The nearest-neighbor distances are 3.082 and 3.515 Å for Mg-Mg pairs in the basal plane and along the $c$-axis, respectively, 2.501 Å for Mg-B pairs, and 1.780 and 3.515 Å for B-B pairs in the basal plane and along $c$-axis, respectively.

Assuming central forces, each atom-atom interaction can be represented by two parameters, the longitudinal and transverse force constants. We have included only nearest-neighbor interactions between Mg-Mg and B-B atoms in the basal plane and along the $c$-axis, and between Mg-B pairs in neighboring layers. The results of the calculation, which was performed using the

![Graph](image)

FIG. 1. (a) The generalized phonon density–of–states, GДOS = $\sum_{i=Mg,B} \sigma_i \exp(-2W_i)G_i(\omega)/M_i$, determined from the inelastic neutron scattering spectrum of MgB$_2$ measured on LRMECS at 8 K with an incident neutron energy 130 meV integrated over wavevectors from 9 to 12 Å$^{-1}$. The solid line is the estimated multiphonon contribution. (b) The calculated GДOS based on the BvK model described in the text convoluted with the resolution function of the spectrometer. The dashed lines represent the projections of the partial contributions to GДOS; the in-plane boron modes (long-dashed), out-of-plane boron modes (dotted), in-plane magnesium modes (dash-dotted), and out-of-plane magnesium modes (dash-double-dotted).

spectively. Unless the scattering is purely incoherent, it is necessary to average the measured neutron scattering over a large volume of reciprocal space in order for the resulting measurements to reflect the true phonon density–of–states.

The neutron data taken with an incident energy of 200 meV shows that the PDOS extends up to 100 meV. Above that energy, the scattering only comprises a broad multiphonon tail. Figure 1 shows data taken at 130 meV at 8K. The peak in the acoustic PDOS occurs at about 36 meV, while there are broad optic bands at 54, 78, 89 and 97 meV. The spectrometer resolution decreases with energy transfer from 5.9 meV at $\hbar\omega = 40$ meV to 3.4 meV at $\hbar\omega = 90$ meV. The large width of these features is not just a resolution effect, but arises from dispersion

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to provide good estimates of the frequency moments of the model to be refined, but the present analysis is sufficient for the purpose. Complete measurements of the frequencies of the main peaks in the PDOS are also plotted.

It is possible to reproduce the frequencies of the peaks in the PDOS with only six force constants. These are the longitudinal and transverse in-plane B-B interactions (90 and 7.5 N/m respectively), the longitudinal out-of-plane B-B interaction (20 N/m), the longitudinal in-plane and out-of-plane Mg-Mg interactions (both 0.6 N/m) and the longitudinal and transverse Mg-B interactions (70 and 2 N/m respectively). The remaining transverse nearest-neighbor interactions had a negligible effect on the mode frequencies.

The frequencies of the main peaks in the PDOS are well-represented by this model, but it is clear that the widths of the peaks are not, i.e., we have underestimated the dispersion of the optic branches. It would be possible to improve the agreement by the introduction of additional parameters representing longer-range interactions, but the results of such an analysis would be meaningless at the present time. Complete measurements of the phonon dispersions on single crystals would allow the model to be refined, but the present analysis is sufficient to provide good estimates of the frequency moments of the PDOS that determine the phonon contribution to $T_c$.

Fig. 2 shows the resulting bare PDOS, $F(\omega)$ defined as

$$F(\omega) = \frac{1}{N} \sum_{j\mathbf{q}} \delta[\omega - \omega(j, \mathbf{q})]$$  

This is similar to equation (2) but does not include the vibrational eigenvectors. In a Bravais lattice, the two quantities are equivalent, but in multicomponent systems with very different atomic masses, $G(\omega)$ will tend to weight the PDOS to higher frequencies because of the smaller atomic displacements of the heavier atoms.

We now discuss the implications of this data for understanding the superconducting transition temperature. This may be estimated using the Allen–Dynes equation [13]

$$k_B T_c = \frac{\hbar \omega_{\text{ln}}}{1.2} \exp \left[ -\frac{1.04(1 + \lambda)}{\lambda - \mu^*(1 + 0.62\lambda)} \right]$$  

where $\lambda$ is the electron-phonon coupling, $\mu^*$ is the Coulomb repulsion, and $\omega_{\text{ln}}$ is the logarithmic phonon average defined in equation 2.16 of ref. [13]. $\omega_{\text{ln}}$ is an average over $\alpha^2(\omega)/F(\omega)$, so we need to make the assumption that $\alpha(\omega)$ is approximately energy-independent, in which case it cancels from the equation. However, we have a choice of two functions to use. It is more conventional to use the bare PDOS, $F(\omega)$, given by equation (2). However, this function fails to take into account the size of the atomic displacements of the different phonon modes; they are included in $G(\omega)$. The electron-phonon matrix elements contain the product of the mode eigenvectors and the gradient of the crystal potential, so it is likely that $G(\omega)$ represents a better approximation to $\alpha^2(\omega)/F(\omega)$. In fact, the choice of $F(\omega)$ or $G(\omega)$ only affects significantly the calculation of the isotope effect, which we discuss below.

The Allen-Dynes equation can be inverted to estimate the value of $\lambda$ for a given value of $T_c$ and $\mu^*$. From integrating over the model calculation of $G(\omega)$, we estimate that $\omega_{\text{ln}}$ is 57.9 meV [53.8 meV when determined from $F(\omega)$]. Fig. 3 shows calculations of $T_c$ using three typical values of $\mu^*$ compared to the measured value of Mg$^{11}$B$_2$. This shows that the measured value of $T_c$ is consistent with a phonon mechanism with values of $\lambda$ in the range 0.6 to 1.2. $\lambda = 0.9$ at the most frequently cited value of $\mu^* = 0.1$. $\omega_{\text{ln}}$ nearly coincides with the energy of the E$_{2g}$ mode that An and Pickett predict will dominate the electron-phonon coupling [3]. It is impossible to determine, on the basis of our measurements, whether the entire phonon spectrum is involved in the superconductivity or only a subset that occurs close to the average energy.

Finally, we can use the BvK model to estimate the isotope effect. The dotted line in Fig. 2 shows a calculation of the PDOS in Mg$^{10}$B$_2$, illustrating the significant energy shifts in all the high frequency boron modes. Substituting 26Mg for 24Mg produces much smaller shifts. In the absence of strong coupling effects, the total isotope effect is given by the sum of the partial isotope effects, $\beta_i = \delta \ln M_i = \delta \ln M_i / \delta \ln M_i$ and should always equal 0.5 if $\mu^* = 0$ [9]. More accurate predictions suggest that it can fall below this value when $\lambda$ becomes small, but not at the value of 0.9 that we estimate. If we use $G(\omega)$ to estimate...
FIG. 3. Variation of the superconducting transition temperature with $\lambda$ using the Allen-Dynes equation and the value of the logarithmic phonon average, $\omega_{\ln} = 57.9$ meV, determined from the generalized PDOS. Three values of the Coulomb repulsion, $\mu^* = 0.0$ (dashed line), 0.1 (solid line), and 0.2 (dotted line), are plotted. The dash-dotted line is the measured value of $T_c$ in Mg$_{11}$B$_2$.

$\beta_B$ and $\beta_{Mg}$, we obtain values of 0.42 and 0.08 respectively, whereas using $F(\omega)$, we obtain values of 0.28 and 0.22, respectively. Reported value of $\beta_B$ are in the range 0.28(3) to 0.30(2) \cite{Bud'ko_2001, Hinks_2001}. Hinks et al have measured an average value of $\beta_{Mg} = 0.018(8)$ \cite{Hinks_2001}. The experimental values are slightly lower than our calculations but the difference may not be significant given the limitations of our analysis.

In conclusion, we have measured the phonon density–of–states in Mg$_{11}$B$_2$, and used the data to produce a simple BvK force-constant model of the vibrational spectra. Our analysis allows us to conclude that the measured values of the transition temperature and the isotope effect are consistent with a conventional phonon mechanism for the superconductivity with moderately strong electron-phonon coupling. It will be extremely useful to compare our data with tunneling measurements of the electron-phonon spectral density, which determine whether the whole phonon spectrum is involved in the superconductivity, or only a few specific modes.

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