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Lattice dynamics in the itinerant helical magnet MnSi

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Abstract. The phonon dispersion relations in MnSi were measured using inelastic neutron scattering. At the same time, calculations of the phonon spectra of MnSi were carried out in the framework of density functional theory in the local density and the generalized gradient approximations. The calculations match most of the phonon modes in the frequency range up to 40 meV. The phonon density of states obtained on the basis of the phonon dispersion was used to estimate the lattice contribution to the specific heat of MnSi.

MnSi is an itinerant magnet with a helical spin order below $T_c \sim 28.8K$. Its transport, magnetic and thermodynamic properties reveal giant anomalies at temperatures centered at $\sim 30K$ that is slightly above the magnetic phase transition. The magnetic phase transition at $28.8K$ is signified by sharp peaks in heat capacity, thermal expansion coefficient and temperature derivative of resistivity in a temperature interval less than 0.1K. Elastic constants, controlling propagation of longitudinal waves display significant softening at the temperature interval, corresponding the mentioned anomalies, and discontinuities at $\sim 28.8K$ [1, 2, 3, 4, 5, 6, 7]. These observations, indicating a strong electron-phonon coupling in MnSi, call for a search of specific features in the lattice dynamics of MnSi.

In this paper, we report first measurements of the phonon dispersion in MnSi, performed by making use of the inelastic neutron scattering (INS) technique. The experimental and theoretical data allowed us to calculate the phonon density of state (PhDOS) and subsequently estimate a lattice contribution to the specific heat in MnSi.

Measurements were performed on two samples: both were nearly perfect single crystals grown by the Bridgman method and having a very small mosaic spread. One had a volume of 2cm$^3$ and the other of 7.5cm$^3$. Further details of crystal characterization are given in Refs. [5, 6]. MnSi has the cubic B20-type crystal structure, space group $P2_13$ with four manganese and four silicon atoms in the unit cell that gives rise the 24 phonon modes making a lattice dynamics study highly challenging.

The INS measurements were carried out on the triple-axis spectrometer IN8 at the Institute Laue Langevin (ILL), Grenoble and on the 1T1 triple axis spectrometer at the Laboratoire Léon
Brillouin, Saclay. The single crystals were aligned in the [1 10]/[001] scattering plane in order to measure phonons with the reduced wavevector \( q \) along the main crystallographic directions [001], [110] and [111]. The measurements were done at \( T = 15K, 30K \) and \( 150K \). The samples were placed in a conventional closed cycle refrigerator. Most of the INS data were collected around the zone centers (1,1,4), (2,2,2) and (1,1,1) using constant-Q scans in the transverse, longitudinal or mixed polarizations at the relatively low and medium energy transfer up to a maximum value of \( \sim 40\text{meV} \).

**Figure 1.** (Color online) Measured (circles) and calculated (solid lines) phonon frequencies in MnSi at \( T = 15K \): The lines correspond to the GGA calculations at the experimental value of the lattice spacing with optimized atomic positions.

The phonon dispersion curves along the main crystallographic direction evaluated from these data are shown in Fig. 1.

The phonon dispersion measured at \( 15K, 30K \) and \( 150K \) does not show any significant changes within the accuracy of the INS data. The only noticeable influence of temperature on the INS spectra is caused by the conventional Bose temperature factor for the phonon intensity. The slight changes of the phonon linewidth observed on heating lie well within experimental error. Note that no anomalous phonon mode has been observed.

The density-functional calculations were carried out within the framework of the local density approximation (LDA) and the generalized gradient approximation (GGA) using a mixed-basis pseudopotential method [8, 9, 10]. For Mn the 3s and 3p semicore states were treated as valence states. Plane waves up to a kinetic energy of 22 Ry were augmented by local functions of s, p, and d type at the Mn sites.

For both LDA and GGA, we have first performed a full optimization of the structure by minimizing the total energy with respect to the lattice parameter and the internal parameters characterizing the positions of the Mn and Si atoms in the unit cell. While the optimized internal parameters agree well with experimental data, we found for both LDA and GGA an overbinding of about 3% and 1.5%, respectively, of the lattice constant. The resulting phonon spectra turned out to be too hard when compared with the measured ones. Because for the analysis of the specific heat data we require a sufficiently accurate phonon model, we also performed additional calculations using the experimental room-temperature lattice constant of \( a = 4.5598\text{Å} \), but still
optimizing the internal parameters. Additional spin-polarized calculations demonstrate that the phonon spectra are little influenced by a spin order.

All model calculations show qualitatively similar dispersion curves, which consist of weakly dispersive optic bands grouped into three sets of bands in the middle part of the spectrum, and three high-frequency bands containing mainly Si vibrations. Quantitatively, however, the spectra based on experimental (LDAexp and GGAexp) and optimized (LDAopt and GGAopt) lattice constants were different from each other regarding both acoustic and optic phonon dispersion curves. This is exemplified in Fig. 2 where the phonon density of states (PhDOS) for the two LDA models is compared. Clearly, using the optimized lattice constant results in significantly higher frequencies for practically all optic modes.

Figure 3. (Color online) Temperature dependence of the electronic and magnetic contribution to the heat capacity of MnSi divided by temperature. 1 - experimental values of $C_p/T$ [5, 6]; 2 - electronic and magnetic contributions obtained from 1 by subtraction of the lattice part.

The best match with our neutron experiment was obtained for the models based on the experimental lattice constant (see Fig. 1). Both LDA and GGA calculations lead to very similar results. The main difference is a slightly stiffer spectrum for the GGA model, which ranges up to 55meV while for LDA the high-frequency cut-off is at 52.5meV. However, the measured low energy phonon data do not allow discriminating between the two models.

Another way to judge the accuracy of the models is a comparison of the experimentally determined and calculated sound velocities. The sound velocities of MnSi were measured in the course of the study of elastic properties of MnSi with an accuracy better than 1% [7]. As is seen in Table 1, the velocities of the longitudinal waves are too high for both LDA and GGA calculations based on the fully optimized structures.

The calculated phonon density of states in MnSi allows to estimate the lattice contribution to the total heat capacity of MnSi. The GGA-exp model has been selected for the estimation of the phonon contribution to the heat capacity of MnSi material base on results of present neutron experiments. Note that the elastic continuous media approximation works purely above $\sim 10K$ in case of MnSi and can not be used to calculate the lattice heat capacity of MnSi in the required temperature range.

The curve 1 in Fig. 3 displays the experimental heat capacity of MnSi divided by temperature $C_p/T$. The corresponding peak is related to the magnetic phase transition in MnSi (see details in Ref. [5, 6]). After subtraction of the phonon contribution from the curve 1 one gets the electronic and magnetic contributions (curve 2).

The both curves cross the ordinate axis at the same value of $C_p/T = \gamma = 0.0365$ joule/moleK$^2$, which implies that the subtraction does not influence the linear term in the heat capacity [6]. On the other hand this procedure unveils a nontrivial feature in behavior of $C_p/T$. As is seen in Fig. 3 magnitudes of $C_p/T$ for $T > T_c$ drop below of the corresponding
value at zero temperature. This could be treated as a consequence of evolution of the electron effective mass in MnSi with temperature. However, published experimental data show that the temperature dependence of resistivity of MnSi can be described by a single expression $\rho = \rho_0 + a T^{2+\delta}$ in the temperature range $0 – 28K$ [12,13]. That certainly indicates a lack of noticeable change of the electron effective mass in the corresponding interval of temperatures. However the same resistivity measurements show a quite different situation at temperatures above $T_c$, where the resistivity tends to saturate and most likely pointing to the suppression of spin fluctuations. So perhaps our observation support earlier claims of existence of a reduction above $T_c$.

The phonon dispersions have been measured in MnSi by means of inelastic neutron scattering experiments. The ab initio model calculations of the lattice dynamics of MnSi are in good agreement with the experimental data. The phonon density of states of MnSi was obtained on the basis of calculations and experiments that allowed an estimation of the phonon contribution to the specific heat of MnSi. So far no detectable temperature effects in the phonon spectra of MnSi at temperatures above and below the phase transition temperature $T_c = 29K$ were found within the experimental error limits.

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Table 1. Comparison of the sound velocity in MnSi obtained from the DFT calculations with results from ultra sound experiments at 6.5K [7].

| Propagation and polarization | LDAopt | LDAexp | GGAopt | GGAexp | Experiment |
|-----------------------------|--------|--------|--------|--------|-------------|
| Longitudinal waves (x 10^5 cm/s) |        |        |        |        |             |
| $V_{[100]} = |C_{11}/\rho|^{1/2}$ | 8.33   | 6.98   | 7.01   | 7.17   | 7.40        |
| $V_{[110]} = [1/2(C_{11} + C_{12} + 2C_{44})/\rho]^{1/2}$ | 8.78   | 7.45   | 8.12   | 7.33   | 7.49        |
| $V_{[111]} = [1/3(C_{11} + 2C_{12} + 4C_{44})/\rho]^{1/2}$ | 8.92   | 7.59   | 8.25   | 7.80   | 7.52        |
| Transverse waves (x 10^5 cm/s) |        |        |        |        |             |
| $V_{[100][001]} = |C_{44}/\rho|^{1/2}$ | 5.32   | 4.72   | 4.93   | 4.71   | 4.64        |
| $V_{[110][001]} = |C_{44}/\rho|^{1/2}$ | 5.22   | 4.65   | 4.87   | 3.89   | 4.61        |
| $V_{[111][001]} = [1/2(C_{11} - C_{12})/\rho]^{1/2}$ | 4.52   | 3.91   | 4.15   | 4.66   | 4.46        |
| $V_{[111][111]} = [1/3(C_{11} - C_{12} + 2C_{44})/\rho]^{1/2}$ | 4.77   | 4.20   | 4.39   | 4.15   | 4.53        |

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