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Effects of spin structures on phonons in $BaFe_2As_2$

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The dependences of phonons on the spin structures of $BaFe_2As_2$ were studied using first-principles supercell approach. We considered the nonmagnetic, checkerboard, stripe, and spin-density-wave structures as well as a spin structure labeled as STR17. We find that STR17 is lower in energy than the stripe. It is seen the ~20 meV missing peak at the generalized phonon density-of-states and the measured specific heat are well reproduced when the magnetic effects are accounted. We see phonon energy gaps for all the four magnetic structures but not for the nonmagnetic structure. © *2010 American Institute of Physics*. [doi:10.1063/1.3464166]

The interplay among conduction electron, magnetism, and phonon plays the key role in understanding the mechanism of superconductivity.[1–3] For $BaFe_2As_2$, it has recently been shown that consideration of magnetism leads to much better agreements with experiments for almost all properties, including the lattice parameters, the arsenide position, and the generalized phonon density-of-states (GDOS) for which the observed ~20 meV peak, that is completely lost in a nonmagnetic (NM) calculation, is recovered by the magnetic calculations.[4,5]

In this paper, we present the calculated phonons and thereafter thermodynamic properties of four spin structures and one NM structure of the iron-based superconductor parent compound $BaFe_2As_2$. Figures 1(a)–1(e) illustrate the spin distributions of these structures, i.e., the ordered spin-density-wave (SDW),[5] STR17 (labeled by us), stripe,[6,7] checkerboard,[6,7] and NM (Ref. 8) structures, respectively.

The calculations were performed employing the projector-augmented wave method[9,10] within the generalized gradient approximation (GGA) of Perdew–Burke–Ernzerhof (Ref. 11) implemented in the VASP package.[9,10] A plane wave cutoff of 348 eV is used. For each of the five structures, total supercell energies were calculated at seven different volumes (in the vicinity of the equilibrium volume of the SDW structure) by a volume step of 6%. At each volume for each of the five structures, both the cell shape and the internal parameters of each atom were fully relaxed to find the geometry that led to the lowest energy, the best atomic coordinates, and the local magnetic moments (for the four magnetic structures out of the five structures) of the Fe atoms.

Table I lists the calculated equilibrium lattice properties and total energies for the five structures. It is seen that the SDW structure is indeed the ground state, in agreement with experiments.[12,13] The calculated volumes for the magnetic states are larger than the measured value by 2%, which is typically acceptable considering the overall overestimation of the GGA functional.[14] It is seen the calculated crystallographic properties with the magnetic structure are much better than those calculated with the NM structure when compared with experiments.

The most notable results are those for STR17. Its energy is even lower than that of the stripe which is a prototype of many recent theoretical models.[7,8,15] By energy, both STR17 and stripe structures are rather close to the SDW structure (SDW is the 0 K ground state, see Table I). Therefore, at around the $BaFe_2As_2$ SDW transition temperature of 140 K the STR17 and stripe structures should be substantially thermally occupied.

We then present our calculations for phonons. We employed the supercell approach with a supercell containing ~80 atoms created using the ATAT code.[16] We have followed Zbiri *et al.*[17] to calculate the GDOS, i.e., GDOS $=\Sigma_i(\sigma_i/M_i)\mathrm{pDOS}_i$ where σ_i, M_i, and pDOS_i represent, respectively, the atomic scattering cross section,[18] the atomic mass, and the partial phonon density-of-states with $i=\{\mathrm{Fe},\mathrm{As},\mathrm{Ba}\}$. To get smooth GDOS, the phonon frequencies

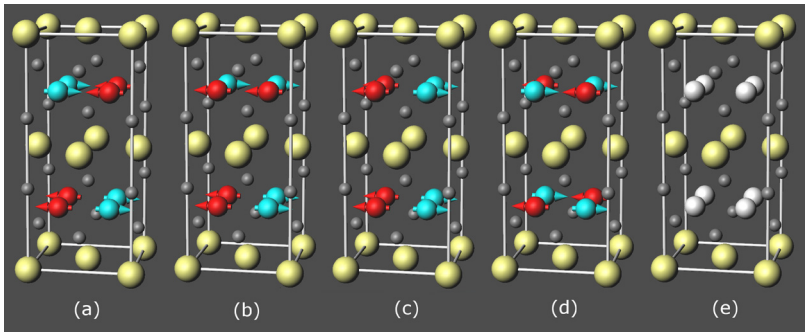

FIG. 1. (Color online) The used magnetic structures of $BaFe_2As_2$: (a) SDW; (b) STR17; (c) stripe; (d) checkerboard; and (e) NM. Big balls (golden): Ba; small balls (grey): As; medium balls (red) with arrows pointing left: Fe with spin up; medium balls (cyan) with arrows pointing right: Fe with spin down. Fe atoms are shown as medium balls (white) for NM case.

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TABLE I. The calculated 0 K static properties of $BaFe_2As_2$.

	a (Å)	*b* (Å)	*c* (Å)	z (As)	Volume[a] (Å^3)	Energy[a] (meV)
			Orthorhombic phase			
Stripe	5.69188	5.61057	12.9055	0.34996	412.13	−489.97
SDW	5.70409	5.61116	12.8853	0.35033	412.41	−516.18
Expt.[b]	5.61587	5.57125	12.9428	0.35406	404.95	
			Tetragonal phase			
NM[c]	5.62035	5.62035	12.6187	0.34511	398.60	0
Checkerboard[c]	5.65263	5.65263	12.7353	0.34729	406.92	−122.65
STR17	5.66196	5.66196	12.8742	0.35004	412.72	−497.29
Expt.[c,d]	5.59607	5.59607	12.9685	0.35406	406.12	

[a]In unit of 4-formula cell (20 atoms) and the total energy of the NM structure is taken as the reference.
[b]Measured at 5 K by Huang *et al.*[26]
[c]Lattice parameter *a* has been multiplied with $\sqrt{2}$ for the convenience of comparison with the orthorhombic phase.
[d]Measured at 175 K by Huang *et al.*[26]

have been calculated using 80 000 *k* points in the **q** space.

Figure 2 is a comparison between the four magnetic structures and the NM structure for the GDOS. The major observations are as follows:

(i) Clear phonon energy gaps, located at ~24 meV, are seen for all the magnetic structures except the NM structure.
(ii) The experimentally observed peak[17,19,20] at ~20 meV that was missed in the previous calculations[17,19] is reproduced by the present calculation for all the magnetic structures but not for the NM structure.

The existence of phonon energy gaps (at ~20 and 28 meV) are observed in the calculation of Zbiri *et al.*[17] who refers the gap as pseudogap. In comparison, we note that the recent phonon calculation with magnetic structure by Yildirim[4] reproduces the ~20 meV experimental peak. The recent measurement and *ab initio* calculation considering magnetism by Hahn *et al.*[21] also show some phonon branches with frequencies ~20 meV for $CaFe_2As_2$.

Figure 3 are our calculated values for specific heat for all the five structures together the measured data.[22–24] We have accounted for the thermal electronic contribution following a previous work.[25] Again, it is seen that the measured specific heats are reproduced very well with the magnetic structures but not with the NM structure. We also deduce Debye temperature by fitting the calculated constant volume lattice contribution to the specific heat with Debye formula.[1] We see that the calculated Debye temperature is ~220 K at low temperature while ~320 K at high temperature.

In summary, the effects of different spin structures on the phonon-related properties of the iron-based superconductor parent compound $BaFe_2As_2$ were studied with first-principles method. The main findings are that as long as the magnetism is considered, the experimental data, including the lattice parameters, the arsenide internal position, the generalized phonon DOS, and the specific heat are all well reproduced.

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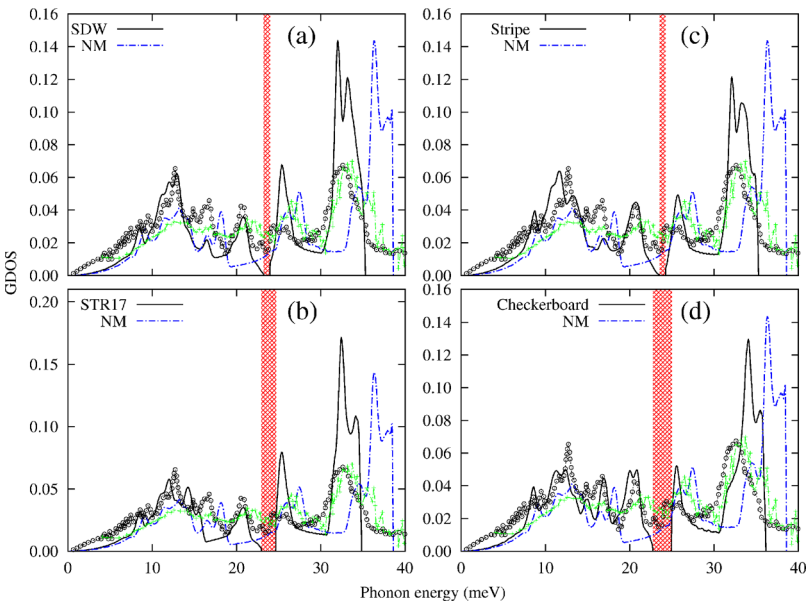

FIG. 2. (Color online) The calculated GDOS for $BaFe_2As_2$ for the four magnetic structures [solid lines (black) in (a) SDW; (b) STR17; (c) stripe; and (d) checkerboard] in comparison to that for the NM structure [dotted-dashed line (blue)] together the measured GDOS by Zbiri *et al.* (Ref. 17) [small open circles connected by dotted line (black)] and Mittal *et al.* (Ref. 19) [small plus signs connected by dotted-dashed line (green)]. The netted shadows (red) show the calculated phonon energy gap for the corresponding magnetic structures.

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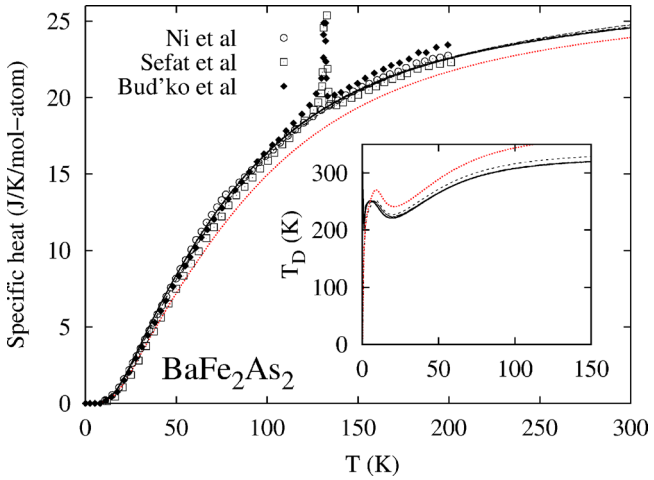

FIG. 3. (Color online) The calculated specific heats [overlapped lines (black)] for $BaFe_2As_2$ for the four magnetic structures in comparison to that for the NM structure [dotted line (red)] together the measured data by Ni *et al.* (Ref. 23) (open circles), Sefat *et al.* (Ref. 24) (open squares), and Bud'ko *et al.* (Ref. 22) (solid diamonds). The inset shows the deduced Debye temperature by fitting the calculated lattice contribution to the specific heat with Debye formula (Ref. 1).

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