Isotope effect and the role of phonons in the iron-based superconductors

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(Received 7 December 2008; revised manuscript received 24 January 2009; published 10 March 2009)

We studied the isotope effect of phonon in the Fe-based superconductors using a phenomenological two band model for the sign-changing s-wave ($\pm s$-wave) state. Within this mean-field model, we showed that the large isotope effect is not inconsistent with the $\pm s$-wave pairing state and its high transition temperature. In principle, a large phonon isotope coefficient α implies a large phonon coupling constant. However, the asymmetric density of states between two bands substantially enhances the value of α, so that a moderate value of the phonon coupling constant ($\lambda_{ph} \approx 0.4$) can produce a very large value of $\alpha(\approx 0.4)$ as well as a high transition temperature together with an antiferromagnet-induced interaction.

DOI: 10.1103/PhysRevB.79.092503 PACS number(s): 74.20.Mn, 74.20.Rp, 74.25.Kc

Recent discovery of the Fe-based superconductors (SCs) by Kamihara *et al.*[1,2] greatly spurred the research activity of unconventional superconductivity. Regarding the pairing mechanism and symmetry of these different superconducting (S) materials, there are already numerous experimental and theoretical investigations. Since the first theoretical proposal of the $\pm s$-wave state by Mazin *et al.*,[3] as a best pairing state in the Fe-based SCs, several subsequent theoretical studies[4–7] supported this idea. Experiments such as angle resolved photoemission spectroscopy (ARPES) (Ref. 8) and penetration depth measurements[9] unanimously indicate a full gap around the Fermi surfaces (FSs) consistent with an s-wave gap state. Then the nuclear-spin-lattice relaxation rate $1/T_1$,[10] which is seemingly consistent with a nodal gap state such as a d-wave gap, provided a strong evidence of the sign-changing nature of the gaps on different bands, so that actually strengthened the case of the $\pm s$-wave state.[5,11,12]

As to the pairing glue, most researchers at the moment tend to believe an electronic origin rather than a phonon origin paring.[4–7,13] In particular, an antiferromagnetic (AFM) correlation-induced interaction appears to be the most natural pairing glue in view of the common spin-density wave (SDW) instability at around ~150 K and the overall phase diagram with doping in this series of Fe pnictides.[14] It is shown by several authors that the AFM-induced potential, when combined with the unique band structure (or the FS topology) of the Fe pnictides, naturally leads to the $\pm s$-wave state as the best pairing solution.[4–7]

However, Liu *et al.*[15] recently measured isotope effect on T_c with a substitution of ^{56}Fe by ^{54}Fe in $Ba_{1-x}K_xFe_2As_2$ and reported an unexpectedly large isotope coefficient $\alpha(\sim 0.4)$. This observation, if confirmed, is drastically perpendicular to the current line of thought for the pairing mechanism of the Fe-based SCs. Theoretical investigations about the electron-phonon coupling in these materials are yet only a few. Boeri *et al.*[16] calculated a very weak electron-phonon coupling constant ($\lambda_{ph} < 0.2$)—the value averaged over the Brillouin Zone (BZ). Eschrig[17] argued that the proper electron-phonon coupling is not the averaged coupling constant but one of a particular phonon mode, i.e., in-plane Fe-breathing mode, which may have a large electron-phonon coupling constant.

Besides theoretical investigations, the experimental fact is that all Fe pnictides, either RFeAsO (R=La, Ce, and Nd) or AKFe_2As_2 (A=Ba and Sr) compounds, display the structural instability from tetragonal to orthorhombic symmetry and it always occurs at temperatures very near the SDW transition temperatures.[18] It implies, at least, two things. (1) The lattice degrees of freedom (structural instability) and the spin degrees of freedom (magnetic instability) are not independent; (2) this structural instability is not a Jahn-Teller-type instability of the Fe 3d electrons but is closely related with the metallicity of the Fe 3d electrons.[13] Therefore, the issue of the electron-phonon coupling in the Fe pnictides needs to be further investigated.

With this motivation, in this Brief Report, we studied the isotope effect and the role of phonon using a phenomenological model for the $\pm s$-wave pairing state. The details of the model can be found in Ref. 5 and here we briefly sketch the essential ingredients for our purpose. The model consists of two bands: one hole band centered around Γ point and one-electron band centered around M point in the reduced BZ scheme, and it has a phenomenological pairing interaction induced from an AFM correlation, hence peaking around (π, π) momentum exchange. In this Brief Report, we add a phonon interaction to this model. Because our purpose of this Brief Report is to study the isotope effect of phonon when the total interaction gives rise to the $\pm s$-wave pairing state, we assume only a general condition of the phonon interaction and vary the basic parameters of the phonon interaction such as the coupling strength λ_{ph} and the characteristic phonon frequency ω_{ph}.

The Hamiltonian is written as

$$
\begin{aligned}
H = & \sum_{k\sigma} \varepsilon_h(k) h^\dagger_{k\sigma} h_{k\sigma} + \sum_{k\sigma} \varepsilon_e(k) e^\dagger_{k\sigma} e_{k\sigma} \\
& + \sum_{kk'\uparrow\downarrow} V_{\mathrm{AFM}}(k,k') h^\dagger_{k\uparrow} h^\dagger_{-k\downarrow} h_{k'\downarrow} h_{-k'\uparrow} \\
& + \sum_{kk'\uparrow\downarrow} V_{\mathrm{AFM}}(k,k') e^\dagger_{k\uparrow} e^\dagger_{-k\downarrow} e_{k'\downarrow} e_{-k'\uparrow} \\
& + \sum_{kk'\uparrow\downarrow} V_{\mathrm{AFM}}(k,k') h^\dagger_{k\uparrow} h^\dagger_{-k\downarrow} e_{k'\downarrow} e_{-k'\uparrow} \\
& + \sum_{kk'\uparrow\downarrow} V_{\mathrm{AFM}}(k,k') e^\dagger_{k\uparrow} e^\dagger_{-k\downarrow} h_{k'\downarrow} h_{-k'\uparrow} \\
& + \sum_{kk'\uparrow\downarrow} V_{\mathrm{ph}}(k,k') h^\dagger_{k\uparrow} h^\dagger_{-k\downarrow} h_{k'\downarrow} h_{-k'\uparrow}
\end{aligned}
$$

$$+\sum_{kk'\uparrow\downarrow} V_{\mathrm{ph}}(k,k')e^{\dagger}_{k\uparrow}e^{\dagger}_{-k\downarrow}e_{k'\downarrow}e_{-k'\uparrow}, \quad (1)$$

where $\varepsilon_{h,e}(k)$ are the dispersions of the hole band and electron band, respectively, representing two main bands in the Fe pnictides. The details of the dispersions are not important for our purpose except the density of states (DOS) of each band, N_h (hole band) and N_e (electron band), respectively. $h^{\dagger}_{k\sigma}$ and $e^{\dagger}_{k\sigma}$ are the electron creation operators on the hole and electron bands, respectively. As mentioned previously, $V_{\mathrm{AFM}}(k,k')$ is the AFM-induced pairing potential, which is all repulsive in momentum space, and $V_{\mathrm{ph}}(k,k')$ is the phonon interaction, which is all attractive in momentum space.

The characteristics of the interactions to promote the $\pm s$-wave gap solution are $V_{\mathrm{AFM}}(k,k')$, peaking around (π,π) momentum exchange, which should have a stronger interband interaction than the intraband one, and $V_{\mathrm{ph}}(k,k')$, being stronger for small momentum exchange, which should have a stronger intraband interaction than the interband one. The latter requirement for the phonon interaction is already included in the Hamiltonian by not including the interband terms such as $V_{\mathrm{ph}}(k,k')e^{\dagger}_{k\uparrow}e^{\dagger}_{-k\downarrow}h_{k'\downarrow}h_{-k'\uparrow}$ and its Hermitian conjugate. This assumption of the phonon interaction is indeed the property of the main phonons in Fe pnictides.[16] If the phonon interaction was absolutely momentum independent, it would have a null effect for the $\pm s$-wave pairing.

For simplicity of the analysis but without the loss of generality, we only need the FS-averaged interactions: for the AFM-induced interactions such as $V^{he}_{\mathrm{AFM}}=\ll V_{\mathrm{AFM}}(k_h,k_e)\gg_{k_h,k_e}$, $V^{hh}_{\mathrm{AFM}}=\ll V_{\mathrm{AFM}}(k_h,k'_h)\gg_{k_h,k'_h}$, etc., and similarly for the phonon interactions such as $V^{hh}_{\mathrm{ph}}=-\ll V_{\mathrm{ph}}(k_h,k'_h)\gg_{k_h,k'_h}$ and $V^{ee}_{\mathrm{ph}}=-\ll V_{\mathrm{ph}}(k_e,k'_e)\gg_{k_e,k'_e}$. Notice that, in these definitions, we absorbed the signs of the interactions and therefore all V^{ab}_{AFM} and V^{ab}_{ph} are positive values. Also assuming the $\pm s$-wave solution, we fix the signs of the s-wave gaps as $\Delta_h=|\Delta_h|$ on the hole band and $\Delta_e=-|\Delta_e|$ on the electron band, respectively. The coupled T_c equations are written as

$$\Delta_h=-[V^{hh}_{\mathrm{AFM}}N_h\chi^{\mathrm{AFM}}-V^{hh}_{\mathrm{ph}}N_h\chi^{\mathrm{ph}}]\Delta_h+[V^{he}_{\mathrm{AFM}}N_e\chi^{\mathrm{AFM}}]\Delta_e, \quad (2)$$

$$\Delta_e=-[V^{ee}_{\mathrm{AFM}}N_e\chi^{\mathrm{AFM}}-V^{ee}_{\mathrm{ph}}N_e\chi^{\mathrm{ph}}]\Delta_e+[V^{eh}_{\mathrm{AFM}}N_h\chi^{\mathrm{AFM}}]\Delta_h. \quad (3)$$

The pair susceptibilities χ^{AFM} and χ^{ph} at $T=T_c$ are defined as

$$\chi(T_c)^{\mathrm{AFM,ph}}=\int_0^{\omega_{\mathrm{AFM,ph}}}\frac{d\xi}{\xi}\tanh\left[\frac{\xi}{2T_c}\right]\approx\ln\left[\frac{1.14\omega_{\mathrm{AFM,ph}}}{T_c}\right], \quad (4)$$

where ω_{AFM} and ω_{ph} are the cut-off frequencies of the AFM fluctuations and phonon, respectively. The second expression is the well-known BCS approximation valid only when $\omega_{\mathrm{AFM,ph}}\gg T_c$, otherwise the first expression should be numerically calculated. Equations (2) and (3) with Eq. (4) constitute the T_c equation of the $\pm s$-wave state of the two band model, including phonon pairing interaction as well as the AFM-induced pairing interaction. Before we show the numerical results, we can analyze a simple case and gain general insight about the model.

In the case (we call it "symmetric case") that $N_h=N_e$ and $V^{ee}_{\mathrm{AFM}}=V^{hh}_{\mathrm{AFM}}$ and $V^{ee}_{\mathrm{ph}}=V^{hh}_{\mathrm{ph}}$ ($V^{he}_{\mathrm{AFM}}=V^{eh}_{\mathrm{AFM}}$ is always true), we define the dimensionless pairing constants as follows:

$$\lambda^{\mathrm{inter}}_{\mathrm{AFM}}=N_hV^{he}_{\mathrm{AFM}}=N_eV^{eh}_{\mathrm{AFM}}, \quad (5)$$

$$\lambda^{\mathrm{intra}}_{\mathrm{AFM}}=N_hV^{hh}_{\mathrm{AFM}}=N_eV^{ee}_{\mathrm{AFM}}, \quad (6)$$

$$\lambda_{\mathrm{ph}}=N_hV^{hh}_{\mathrm{ph}}=N_eV^{ee}_{\mathrm{ph}}. \quad (7)$$

Then, the T_c equations are simplified as

$$\Delta_h(1+\lambda^{\mathrm{intra}}_{\mathrm{AFM}}\chi^{\mathrm{AFM}}-\lambda_{\mathrm{ph}}\chi^{\mathrm{ph}})=\lambda^{\mathrm{inter}}_{\mathrm{AFM}}\chi^{\mathrm{AFM}}\Delta_e,$$

$$\Delta_e(1+\lambda^{\mathrm{intra}}_{\mathrm{AFM}}\chi^{\mathrm{AFM}}-\lambda_{\mathrm{ph}}\chi^{\mathrm{ph}})=\lambda^{\mathrm{inter}}_{\mathrm{AFM}}\chi^{\mathrm{AFM}}\Delta_h. \quad (8)$$

The above T_c equations can be solved analytically with the BCS approximation of the pair susceptibilities χ^{AFM} and χ^{ph} [the second expression of Eq. (4)] as

$$T_c=1.14\omega_{\mathrm{AFM}}^{\tilde{\lambda}_M}\cdot\omega_{\mathrm{ph}}^{\tilde{\lambda}_{\mathrm{ph}}}\cdot\exp^{-1/\lambda_{\mathrm{tot}}}, \quad (9)$$

where

$$\lambda_{\mathrm{tot}}=(\lambda^{\mathrm{inter}}_{\mathrm{AFM}}-\lambda^{\mathrm{intra}}_{\mathrm{AFM}})+\lambda_{\mathrm{ph}}, \quad (10)$$

$$\tilde{\lambda}_M=(\lambda^{\mathrm{inter}}_{\mathrm{AFM}}-\lambda^{\mathrm{intra}}_{\mathrm{AFM}})/\lambda_{\mathrm{tot}}, \quad (11)$$

$$\tilde{\lambda}_{\mathrm{ph}}=\lambda_{\mathrm{ph}}/\lambda_{\mathrm{tot}}. \quad (12)$$

Equations (9) and (10) show that the AFM interaction and the phonon interaction are additive in the exponential form so that T_c can be dramatically boosted even with a small value of λ_{ph}.[19] We can also easily read the phonon isotope coefficient α from Eq. (9) as

$$\alpha=\frac{1}{2}\frac{d\ln T_c}{d\ln\omega_{\mathrm{ph}}}=0.5\times\frac{\lambda_{\mathrm{ph}}}{(\lambda^{\mathrm{inter}}_{\mathrm{AFM}}-\lambda^{\mathrm{intra}}_{\mathrm{AFM}})+\lambda_{\mathrm{ph}}}. \quad (13)$$

This result conforms with a physical insight, i.e., a large α value means a relatively large phonon coupling compared to the total AFM pairing interaction $\lambda^{\mathrm{eff\text{-}tot}}_{\mathrm{AFM}}=(\lambda^{\mathrm{inter}}_{\mathrm{AFM}}-\lambda^{\mathrm{intra}}_{\mathrm{AFM}})$. However, Eq. (9) also shows that a large α value does not necessarily mean that the large phonon contribution to the pairing energetics when ω_{ph} is much smaller than ω_{AFM}.

For the general cases where $N_h\neq N_e$ and $V^{ee}_{\mathrm{AFM}}\neq V^{hh}_{\mathrm{AFM}}$ and $V^{ee}_{\mathrm{ph}}\neq V^{hh}_{\mathrm{ph}}$ ("nonsymmetric case"), it is not possible to find an analytic solution of the T_c equation. However, an inspection suggests us to generalize Eq. (9) to the nonsymmetric case. Equations (9) and (13) can be used as good approximations for T_c and α with the definitions of the effective dimensionless coupling constants as follows:[20]

$$\lambda^{\mathrm{inter}}_{\mathrm{AFM}}=\sqrt{N_hN_eV^{he}_{\mathrm{AFM}}V^{eh}_{\mathrm{AFM}}}, \quad (14)$$

$$\lambda^{\mathrm{intra}}_{\mathrm{AFM}}=\sqrt{N_hN_eV^{hh}_{\mathrm{AFM}}V^{ee}_{\mathrm{AFM}}}, \quad (15)$$

$$\lambda_{\mathrm{ph}}=\sqrt{N_hN_eV^{hh}_{\mathrm{ph}}V^{ee}_{\mathrm{ph}}}. \quad (16)$$

In the following, we will show the numerical results directly obtained from Eqs. (2) and (3) and compare them with the

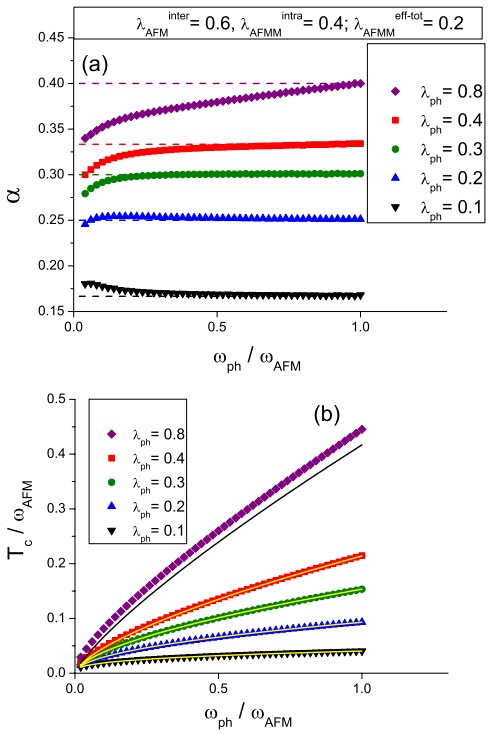

FIG. 1. (Color online) Symmetric case ($N_h/N_e=1.0$). (a) Isotope coefficients α as a function of the phonon cut-off frequency $\omega_{\rm ph}/\omega_{\rm AFM}$ for different phonon coupling constants $\lambda_{\rm ph}=0.1$, 0.2, 0.3, 0.4, and 0.8. (b) Normalized T_c vs $\omega_{\rm ph}/\omega_{\rm AFM}$ for the same parameters. Symbols are numerical calculations and lines are the results of Eqs. (9) and (13).

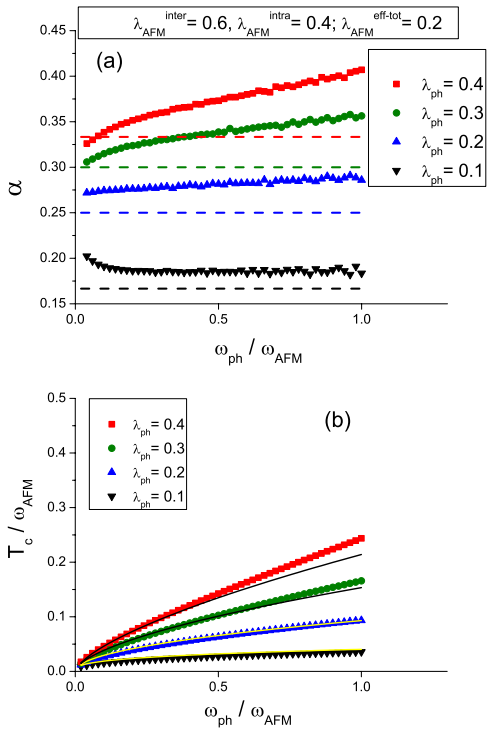

FIG. 2. (Color online) Nonsymmetric case ($N_h/N_e=3.0$). (a) Isotope coefficients α as a function of the phonon cut-off frequency $\omega_{\rm ph}/\omega_{\rm AFM}$ for different phonon coupling constants $\lambda_{\rm ph}=0.1$, 0.2, 0.3, and 0.4. (b) Normalized T_c vs $\omega_{\rm ph}/\omega_{\rm AFM}$ for the same parameters. Symbols are numerical calculations and lines are the results of Eqs. (9) and (13).

analytic formulas (9) and (13) both for the symmetric and nonsymmetric cases.

Figure 1(a) shows the isotope coefficient α of the symmetric case ($N_h=N_e$). Symbols are the numerical results of Eqs. (2) and (3) and the lines are the results of Eq. (13). All the energy scales are normalized by the cut-off frequency of the AFM fluctuations $\omega_{\rm AFM}$ and we choose moderate strength of dimensionless coupling constants of the AFM-induced potential as $\lambda_{\rm AFM}^{\rm inter}=N_hV_{\rm AFM}^{he}=N_eV_{\rm AFM}^{eh}=0.6$ and $\lambda_{\rm AFM}^{\rm intra}=N_hV_{\rm AFM}^{hh}=N_eV_{\rm AFM}^{ee}=0.4$, which yields the effective total magnetic interaction $\lambda_{\rm AFM}^{\rm eff\text{-}tot}=0.2$. Figure 1(a) shows that the numerically calculated α (symbols) are in excellent agreement with the analytic results of Eq. (13) when $\omega_{\rm ph}\gg T_c$. When this condition is not fulfilled, deviations occur at low $\omega_{\rm ph}$ where the BCS approximation of the pair susceptibility becomes poor.

The results of Fig. 1(a) show that a large phonon isotope coefficient arises when the phonon coupling strength is much stronger than the magnetic coupling strength and it is in accord with a standard expectation. For example, in the symmetric case, in order to obtain $\alpha\approx0.4$ (the reported value by Liu *et al.*[15]), we need to have $\lambda_{\rm ph}=4\times\lambda_{\rm AFM}^{\rm eff\text{-}tot}$ which corresponds to the result of $\lambda_{\rm ph}=0.8$ (purple diamond symbols). If this is the case of reality *then the superconductivity of the Fe pnictides is a phonon-driven SC and the AFM fluctuations merely act as a tipping agent to introduce the* π phase between two bands widely separated in the BZ. It is not an impossible scenario, but we first need to find an evidence of such a strong phonon coupling in Fe pnictides. If confirmed, the current viewpoint about the pairing mechanism of the Fe-based SCs should completely be changed.

In Fig. 2 we show the results of the nonsymmetric case ($N_h=3N_e$).[21] We adjusted the interactions $V_{\rm AFM,ph}$ to keep the dimensionless coupling constants $\lambda_{\rm AFM}^{\rm inter}=0.6$ and $\lambda_{\rm AFM}^{\rm intra}=0.4$—the same values as in the symmetric case—according to the generalized formulas (14) and (15), which means that we chose $N_eV_{\rm AFM}^{eh}=0.346$, $N_hV_{\rm AFM}^{he}=1.039$ and $N_eV_{\rm AFM}^{ee}=0.231$, $N_hV_{\rm AFM}^{hh}=0.693$. The phonon coupling constants $\lambda_{\rm ph}=0.1,0.2,0.3,0.4$ are also the values according to Eq. (16) but keeping the ratio $N_hV_{\rm ph}^{hh}/N_eV_{\rm ph}^{ee}=3$. The behavior of α is very different compared to the symmetric case. (1) There are systematic deviations of the numerical (exact) results from the analytic (approximate) formula (13) and the numerically calculated values of α give substantially larger values than the ones of the analytic formula. As a result, a moderate value of $\lambda_{\rm ph}=0.4$ can cause a large isotope coefficient $\alpha\approx0.4$; (2) with increasing $\omega_{\rm ph}$, there is no saturation of α, in particular, with large phonon couplings. In contrast, the T_c values are rather similar to the symmetric case.

This seemingly inconsistent behavior between T_c and α can be most easily understood by comparison of the $\lambda_{\rm ph}=0.4$ results between the symmetric case (Fig. 1) and the nonsymmetric case (Fig. 2). α in the symmetric case quickly saturates to its maximum value [red square symbols in Fig. 1(a)] but α in the nonsymmetric case gradually increases with $\omega_{\rm ph}$ [red square symbols in Fig. 2(a)]. As a result, a large difference of the α value between the two cases is

possible when $\omega_{\rm ph}/\omega_{\rm AFM} \rightarrow 1$ (Ref. 22) while maintaining only slightly enhanced T_c [see red square symbols in Fig. 1(b) and in Fig. 2(b)]. Slightly enhanced T_c is due to the fact that the asymmetric phonon couplings on the hole and electron bands are slightly more advantageous for pairing than the equal phonon couplings on both bands when $\lambda_{\rm ph} = \sqrt{N_h N_e V_{\rm ph}^{hh} V_{\rm ph}^{ee}}$ is the same.

In summary, using a minimal two band model with both the AFM-induced interaction and the phonon interaction, which together yields the $\pm s$-wave gap pairing state, we studied the phonon isotope coefficient α for the symmetric and nonsymmetric cases. We confirmed that a large value of α indicates a large value of the phonon coupling constant $\lambda_{\rm ph}$ compared to the value of the AFM-induced coupling constant $\lambda_{\rm AFM}^{\text{eff-tot}}$. However, we found that the large asymmetric ratio (N_h/N_e) of DOSs between the hole and electron bands substantially enhances the α value compared to the symmetric band case. As a result, a relatively small value of the phonon coupling constant (say, $\lambda_{\rm ph}=0.4$ when $\lambda_{\rm AFM}^{\text{eff-tot}}=0.2$) can yield a very large isotope coefficient $\alpha=0.4$. Regardless of the symmetric or nonsymmetric cases, T_c is strongly enhanced by the addition of two pairing interactions $\lambda_{\rm ph}$ and $\lambda_{\rm AFM}^{\text{eff-tot}}$ in the exponential form. A similar result was obtained for the high-T_c cuprates SCs (Ref. 19) and possibly important role of phonon(s) discussed in this Brief Report is a very plausible scenario, in general, for the unconventional SCs. Further experimental study of the isotope effect in the Fe pnictide SCs is a pressing task. If the large isotope effect is confirmed, the importance of phonons for the unconventional S pairing in the correlated materials should be renewed.

Finally, we would like to make remarks about the validity and limitations regarding the quantitative calculations of T_c and α made in this Brief Report. A mean-field theory employed in this Brief Report is sufficient to demonstrate the principles: (1) the boost of T_c of the $\pm s$-wave pairing state with an anisotropic phonon and (2) a large enhancement of α with the asymmetric DOSs between bands. However, it is needless to say that we should go beyond a mean-field theory to calculate a quantitatively reliable T_c, including dynamics of phonons as well as the AFM fluctuations, and their strong-coupling effects. This is beyond the scope of the current Brief Report. Regarding the value of α, however, our calculations can have more meaning to make a direct connection with experiments, albeit with some reservations. Numerous studies[24] have shown that the anomalous DOS (such as Van Hove singularities), anharmonicity of phonon(s) dynamics, Coulomb pseudopotential, etc., are the direct sources for the deviation of the mean-field value of α. However, the large coupling constant itself does not yield the deviation from the mean field α (unless it is extremely large, say $\lambda > 2$). Therefore, unless the iron pnictides do not have such peculiar electronic properties such as anomalous DOS and anharmonic phonon(s), our calculations of α can serve as a useful guideline.

The author acknowledges useful discussions with S.-W. Cheong about the possible importance of phonons in Fe pnictides. This work is supported by the KOSEF through the Grant No. KRF-2007-521-C00081.

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[21] This choice of the DOS ratio $(N_h/N_e=3)$ is indeed a close value of the realistic band calculations of Ref. 3, which yields this ratio 2.6 (Ref. 5).
[22] A typical AFM-fluctuation frequency of the S pnictide $(Ba_{0.6}K_{0.4}Fe_2As_2)$ is measured by neutron experiments which is about 15 meV $\approx$ 180 K (Ref. 23) and a typical phonon frequency was theoretically estimated as a logarithmically averaged value $\omega_{\rm lg} \approx 200$ K (Ref. 16). They are comparable values to each other with some possible variations for different compounds and different dopings.
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