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Intrinsic and structural isotope effects in iron-based superconductors

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The currently available results of the isotope effect on the superconducting transition temperature T_c in Fe-based high-temperature superconductors (HTSs) are highly controversial. The values of the Fe isotope effect exponent α_{Fe} for various families of Fe-based HTS were found to be as well positive, as negative, or even be exceedingly larger than the BCS value $\alpha_{\mathrm{BCS}} \equiv 0.5$. Here we emphasize that the Fe isotope substitution causes small structural modifications which, in turn, affect T_c. Upon correcting the isotope effect exponent for these structural effects, an almost unique value of $\alpha \sim 0.35-0.4$ is observed for at least three different families of Fe-based HTS.

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The isotope effect on the transition temperature T_c traditionally plays an important role in identifying the superconducting pairing mechanism. As a rule, an impact of the isotope substitution and, consequently, an involvement of the lattice degrees of freedom in formation of the supercarriers are determined by comparing the isotope effect exponent $\alpha = -(\Delta T_c/T_c)/(\Delta M/M)$ (M is the atomic mass) with the universal value $\alpha_{\mathrm{BCS}} \equiv 0.5$ as predicted within the framework of BCS theory of electron-phonon-mediated superconductivity.

In conventional phonon-mediated superconductors such as simple metals, alloys, etc., α, typically, ranges from 0.2 to 0.5, see, e.g., Ref. 1, and references therein. The only exceptions are Ru and Zr exhibiting zero isotope effect and PdH(D) with $\alpha_{\mathrm{H(D)}} = -0.25$.[2] The negative isotope effect of PdH(D) is explained, however, by the presence of strong lattice anharmonicity caused by the double-well potential in the proton (deuteron) bond distribution.[3] This was confirmed by neutron scattering data where the large zero-point motion of H in comparison with that of deuterium results in 20% change in the lattice force constants.[4] A similar finding exists in organic superconductors where the H(D) isotope effect changes sign as compared, e.g., to ^{34}S, ^{13}C, and ^{15}N isotope replacements, see Ref. 5, and references therein. Again, an unusually strong anharmonic lattice dynamics are attributed to this observation.[5,6] The cuprate high-temperature superconductors (HTSs) are characterized by a vanishingly small but positive isotope effect exponent in optimally doped compounds which increases in a monotonic way upon decreasing doping for almost all cuprate families[7–13] with the exception of the doped $La_{2-x}Sr_xCuO_4$ system, where at 1/8th an anomaly appears,[14,15] most likely caused by the low-temperature tetragonal phase transition. For the optimally doped cuprate HTS the smallest value of the oxygen-isotope exponent $\alpha_{\mathrm{O}} \simeq 0.02$ was obtained for $YBa_2Cu_3O_{7-\delta}$ and $Bi_2Sr_2Ca_2Cu_3O_{10+\delta}$ while it reaches $\alpha_{\mathrm{O}} \simeq 0.25$ for $Bi_2Sr_{1.6}La_{0.4}CuO_{6+\delta}$.[7–9,12,13] In addition, it was demonstrated that in underdoped materials α_{O} exceeds substantially the BCS limit $\alpha_{\mathrm{BCS}} \equiv 0.5$.[8,13,15] It is important to note here that the values of both, the oxygen and the copper isotope exponents in cuprate HTS are *always* positive. Similar tendencies, with the only few above-mentioned exceptions, are realized in a case of conventional phonon-mediated superconductors.

Since the discovery of superconductivity in Fe-based compounds few attempts to measure the isotope effect on T_c in these materials were made. Currently we are aware of four papers reporting, however, rather contradictory results.[16–19] Liu *et al.*[16] and Khasanov *et al.*[19] have found a *positive* Fe isotope effect (Fe-IE) exponent α_{Fe} for $Ba_{0.6}K_{0.4}Fe_2As_2$, $SmFeAsO_{0.85}F_{0.15}$, and $FeSe_{1-x}$ with the corresponding values $\alpha_{\mathrm{Fe}} = 0.34(3)$, 0.37(3), and 0.81(15), respectively. Note that $\alpha_{\mathrm{Fe}} = 0.81(15)$ for $FeSe_{1-x}$ exceeds grossly the BCS value. In the other two studies Shirage *et al.*[17] have reported a *negative* $\alpha_{\mathrm{Fe}} = -0.18(3)$ and $-0.024(15)$ for $Ba_{0.6}K_{0.4}Fe_2As_2$ and $SmFeAsO_{1-y}$,[18] respectively. These controversial results are unlikely to stem from different pairing mechanisms to be realized in different Fe-based superconductors. Especially, in the case of $Ba_{0.6}K_{0.4}Fe_2As_2$, nominally identical samples were isotope replaced with one exhibiting a positive[16] and the other a negative isotope exponent.[17] Note, that the sign reversed isotope exponent seen by Shirage *et al.*[17,18] was attributed to multiband superconductivity with different pairing channels, namely, a phononic one and an antiferromagnetic (AF) fluctuation dominated one.[20] However, as shown by Bussmann-Holder and Keller,[21] the model presented in Ref. 20 and its extensions cannot give rise to *any* sign reversed isotope effect even if both pairing channels stem from AF fluctuations.

In the present study we demonstrate that the very controversial results for α_{Fe} are caused by small structural changes occurring simultaneously with the Fe isotope exchange. As such, we decompose the Fe-IE exponent into one related to the structural changes $\alpha_{\mathrm{Fe}}^{\mathrm{str}}$ and the genuine (intrinsic) one $\alpha_{\mathrm{Fe}}^{\mathrm{int}}$ to arrive at

$$\alpha_{\mathrm{Fe}} = \alpha_{\mathrm{Fe}}^{\mathrm{int}} + \alpha_{\mathrm{Fe}}^{\mathrm{str}}. \tag{1}$$

By comparing the c-axis lattice constants for the pairs of isotopically substituted samples we observe that $\alpha_{\mathrm{Fe}}^{\mathrm{str}}$ is negative for $Ba_{0.6}K_{0.4}Fe_2As_2$ and $SmFeAsO_{1-y}$ studied by Shirage *et al.* in Refs. 17 and 18, positive for $FeSe_{1-x}$ from Ref. 19 and close to 0 for $Ba_{0.6}K_{0.4}Fe_2As_2$ and $SmFeAsO_{0.85}F_{0.15}$ measured in Ref. 16. By taking into account the sign of $\alpha_{\mathrm{Fe}}^{\mathrm{str}}$

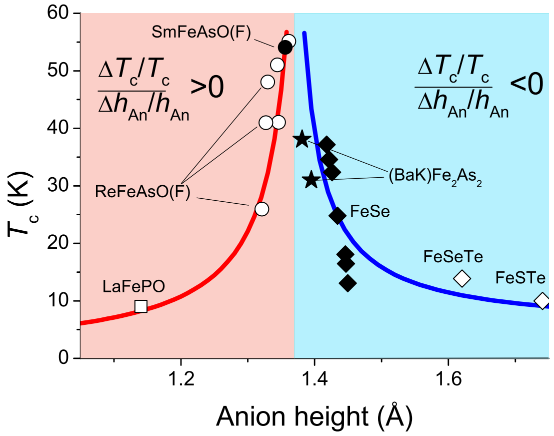

FIG. 1. (Color online) Dependence of the superconducting transition temperature T_c on the height of the anion atom (h_{An}, $An = \mathrm{As, Se, P}$) for various families of Fe-based HTS, after Mizuguchi *et al.* (Ref. 22). The closed symbols represent the samples which are relevant for the present study. The lines are guide for the eyes. Shaded areas on the left (right) side of the figure represent the regions of T_c vs h_{An} diagram where T_c increases (decreases) with increasing h_{An}.

we arrive at the conclusion that $\alpha_{\mathrm{Fe}}^{\mathrm{int}}$ is positive for all so far studied Fe-based HTS.

Our motivation to separate the isotope coefficient into the above-mentioned two components, see Eq. (1), stems from the fact that superconductivity in these compounds is intimately related to small structural changes as reported in various works. As an example, we mention the strong nonlinear dependence of the superconducting transition temperature on the anion atom height (h_{An}, An=As, P, S, or Se) with a sharp maximum of T_c at $h_{An} \simeq 1.38$ Å, see Mizuguchi *et al.*[22] and Fig. 1. The influence of the Fe isotope substitution on the crystal structure, on the other hand, was considered by Granath *et al.*[23] based on the results of Raman studies of $CaFeAsO_{1-x}$ and $NdFeAsO_{1-x}$ and further confirmed by Khasanov *et al.*[19] in neutron powder-diffraction experiments on ^{54}Fe to ^{56}Fe substituted $FeSe_{1-x}$.

The c-axis lattice constants for the pairs of Fe isotope substituted samples $Ba_{0.6}K_{0.4}Fe_2As_2$, $SmFeAsO_{0.85}F_{0.15}$, $SmFeAsO_{1-y}$, and $FeSe_{1-x}$ studied in Refs. 16–19, are summarized in Table I. The choice of the c-axis lattice constant as the relevant quantity in deriving the structural isotope effect might appear to be rather arbitrary since T_c is influenced by all structural details, namely, tetrahedral angle, a-axis lattice constant, internal bond lengths, etc. However, the c-axis lattice constant provides a very sensitive probe since its compression (expansion) is directly accompanied by the corresponding variation in the distance from the Fe planes to the above (below) lying anions which, in turn, is a well characterized property for many Fe-based compounds.[22] A survey of the literature shows that the proportionality between the anion atom height and the c-axis lattice constant indeed holds for $FeSe_{1-x}$, (BaK)Fe_2As_2, and SmFeAsO(F) families of Fe-based HTS considered in the present study.[24–26] From Table I it is obvious that in $Ba_{0.6}K_{0.4}Fe_2As_2$ and $SmFeAsO_{0.85}F_{0.15}$ (Ref. 16) the c-axis constants are the same within the experimental error for both isotopically substituted sets of the samples.[27] In $FeSe_{1-x}$ (Ref. 19) the c-axis constant is larger while in $Ba_{0.6}K_{0.4}Fe_2As_2$ (Ref. 17) it is smaller for the sample with the heavier Fe isotope. In $SmFeAsO_{1-y}$, studied by Shirage *et al.*,[18] both c-axis lattice constants seem to coincide within the experimental resolution. However, since the difference between them is 1.5 times larger than one standard deviation, it is conceivable to attribute an increase in the c-axis lattice constant in $SmFeAsO_{1-y}$ with the heavier Fe isotope.

The use of the empirical T_c vs h_{An} relation from Ref. 22 combined with the intrinsic relation of the proportionality between the c-axis constant and the anion atom height ($\Delta c \propto \Delta h_{An}$, see Refs. 24–26) enables us to determine the sign of the structurally related shift of T_c induced by isotopic exchange. By defining the shift of a given quantity X as $\Delta X/X = ({}^{\mathrm{light}}\mathrm{Fe}X - {}^{\mathrm{heavy}}\mathrm{Fe}X)/{}^{\mathrm{heavy}}\mathrm{Fe}X$ and following Mizuguchi *et al.*,[22] see also Fig. 1, the sign of $(\Delta T_c/T_c)/(\Delta h_{An}/h_{An})$ is positive for SmFeAsO(F) as well as for various Fe-based HTS belonging to *Re*FeAsO(F) family (Re=Nd,Ce,La) and negative for (BaK)Fe_2As_2 and $FeSe_{1-x}$. Consequently the change in the c-axis constant caused by Fe isotope substitution as presented in Table I results in an additional structurally related shift of T_c being positive for $FeSe_{1-x}$,[19] negative for $Ba_{0.6}K_{0.4}Fe_2As_2$ and $SmFeAsO_{1-y}$,[17,18] and close to 0 for $Ba_{0.6}K_{0.4}Fe_2As_2$ and $SmFeAsO_{0.85}F_{0.15}$.[16] It is rather remarkable that the corresponding "structural" Fe-IE exponents $\alpha_{\mathrm{Fe}}^{\mathrm{str}}$ would lead to the shift of genuine (intrinsic) $\alpha_{\mathrm{Fe}}^{\mathrm{int}}$ in the direction of 0.35–0.4, see Fig. 2.

TABLE I. Summary of Fe isotope effect studies for $FeSe_{1-x}$ (Ref. 19), $SmFeAsO_{0.85}F_{0.15}$ and $Ba_{0.6}K_{0.4}Fe_2As_2$ (Ref. 16), $Ba_{0.6}K_{0.4}Fe_2As_2$ (Ref. 17), and $SmFeAsO_{1-x}$ (Ref. 18). The parameters are: T_c—superconducting transition temperature for the sample with the natural Fe isotope (${}^{\mathrm{nat}}$Fe); α_{Fe}-Fe isotope effect exponent; c—the c-axis lattice constant for the sample with the lighter (${}^{\mathrm{light}}$Fe) and the heavier (${}^{\mathrm{heavy}}$Fe) Fe isotope; $\Delta c/c$—the relative shift of the c-axis constant caused by the Fe isotope substitution; $\alpha_{\mathrm{Fe}}^{\mathrm{str}}$ and $\alpha_{\mathrm{Fe}}^{\mathrm{int}}$—the structural and the intrinsic contributions to α_{Fe}. See text for details.

Sample	Reference	T_c(${}^{\mathrm{nat}}$Fe) (K)	α_{Fe}	c axis (${}^{\mathrm{light}}$Fe) (Å)	c axis (${}^{\mathrm{heavy}}$Fe) (Å)	$\Delta c/c$	$\alpha_{\mathrm{Fe}}^{\mathrm{str}}$	$\alpha_{\mathrm{Fe}}^{\mathrm{int}}$
$FeSe_{1-x}$	19	8.21(4)	0.81(15)	5.48683(9)	5.48787(9)	>0	$\simeq 0.4$	$\simeq 0.4$
$Ba_{0.6}K_{0.4}Fe_2As_2$	16	37.30(2)	0.37(3)	13.289(7)	13.288(7)	~ 0	~ 0	~ 0.35
$Ba_{0.6}K_{0.4}Fe_2As_2$	17	37.78(2)	−0.18(3)	13.313(1)	13.310(1)	<0	~ -0.5	~ 0.35
$SmFeAsO_{0.85}F_{0.15}$	16	41.40(2)	0.34(3)	8.490(2)	8.491(2)	~ 0	~ 0	~ 0.35
$SmFeAsO_{1-y}$	18	54.02(13)	−0.024(15)	8.4428(8)	8.4440(8)	$\gtrsim 0$	<0	

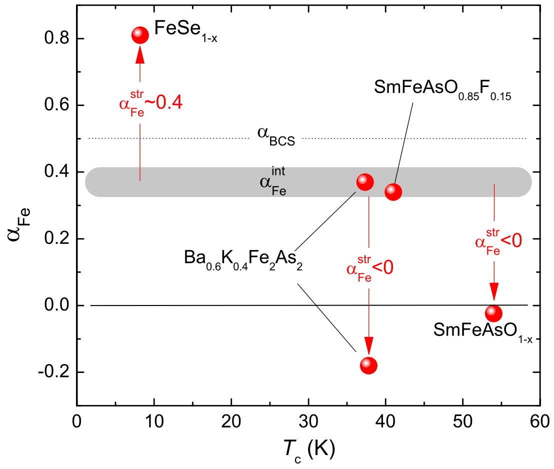

FIG. 2. (Color online) Fe isotope effect exponent α_{Fe} as a function of the superconducting transition temperature T_c for the samples considered in the present study: FeSe$_{1-x}$ (Ref. 19), Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ and SmFeAsO$_{0.85}$F$_{0.15}$ (Ref. 16), Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ (Ref. 17), and SmFeAsO$_{1-x}$ (Ref. 18). Arrows indicate the direction of the shift from the "intrinsic" Fe-isotope effect exponent α_{Fe}^{int} $\sim$0.35–0.4 caused by the structural effects. $\alpha_{BCS}\equiv 0.5$ is the BCS value for electron-phonon-mediated superconductivity. See text for details

Note that the above-mentioned discussion allows only to determine the sign of the structurally related isotope effect but not the absolute value. The reasons are the following. First, the relative change in the c-axis constant is proportional but not identical to the one of h_{An}. As an example, ^{56}Fe to ^{54}Fe isotope substitution in FeSe$_{1-x}$ leads to an increase in the c-axis constant by approximately 0.02% while the change of the Se height amounts to $\simeq$0.22%, see Ref. 19. Second, the height of the anion atom is clearly not the only parameter which is crucial for T_c of Fe-based HTS as already mentioned above. However, the lack of a consistent structural characterization limits this study to a single parameter which was emphasized to be of uppermost relevance to T_c.

The analysis of the structural data together with the dependence of T_c on Se height in FeSe$_{1-x}$, as well as on Se(Te)-Fe-Se(Te) angle and the a-axis constant in FeSe$_{1-y}$Te$_y$ for $y\leq 0.5$ admits to extract the "structural" Fe isotope effect exponent $\alpha_{Fe}^{str}\simeq 0.4$ for ^{56}Fe to ^{54}Fe substituted FeSe$_{1-x}$ samples.[19] The absence of precise structural data complicates the analysis as outlined in Refs. 16–18. However, a zero, within the experimental accuracy, Fe isotope shift of the c-axis lattice constant for Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ as reported by Liu *et al.*[16] is a clear indication that no structural effect is present for this particular set of the samples. Consequently, the negative isotope effect exponent $\alpha_{Fe}\simeq -0.18$ obtained for nominally identically doped Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ by Shirage *et al.*[17] stems from summing both effects, i.e., $-0.18(\alpha_{Fe})=0.35(\alpha_{Fe}^{int})-0.53(\alpha_{Fe}^{str})$, see Eq. (1). It is important to recognize that for SmFeAsO(F) a similar analysis is impossible since samples with different doping levels (different T_c's, see Table I and Fig. 2) were studied in Refs. 16 and 18.

Recently Bussmann-Holder *et al.*[28] investigated a multiple gap scenario of superconductivity in Fe-based HTS with the aim to search for possible sources of the isotope effect on T_c. Typical phonon-mediated scenarios were contrasted to polaronic effects and found to have very different impacts on the isotope effect. While phonon mediated superconductivity slightly suppresses the isotope effect as compared to the BCS value $\alpha_{BCS}\equiv 0.5$, polaronic effects can largely enhance it. The scenario of electron-phonon-mediated superconductivity within the dominant gap channel predicts a T_c independent isotope effect with the α value being slightly smaller than 0.5 thus agreeing rather well with that observed for FeSe$_{1-x}$,[19] Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$,[16,17] and SmFeAsO$_{0.85}$F$_{0.15}$.[16] Indeed, for these particular samples, which belong to three different families of Fe-based HTS and have T_c's ranging from 8 to 44 K, the "intrinsic" Fe isotope exponent is almost constant with $\alpha_{Fe}^{int}\sim 0.35-0.4$, see Table I and Fig. 2. As such, the independent on T_c value of α_{Fe}^{int} would suggest $\alpha_{Fe}^{str}\sim -0.4$ for SmFeAsO$_{1-x}$ studied by Shirage *et al.*[18]

To conclude, the currently available Fe isotope effect data on the superconducting transition temperature T_c for various Fe-based HTS were reanalyzed by separating the measured Fe-IE exponent α_{Fe} into a structural and an intrinsic (unrelated to the structural changes) component. Accounting for the empirical relation between T_c and the anion atom height h_{An} (Ref. 22) we have demonstrated that the structural contribution to the Fe-IE exponent is negative for Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ and SmFeAsO$_{1-x}$ studied by Shirage *et al.*,[17,18] positive for FeSe$_{1-x}$,[19] and close to 0 for SmFeAsO$_{0.85}$F$_{0.15}$ and Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ measured by Liu *et al.*[16] By taking such corrections into account we infer that the value of the genuine Fe-IE exponent is close to $\alpha_{Fe}^{int}\sim 0.35-0.4$ for compounds belonging to at least three different families of Fe-based HTS. We are convinced that the analysis presented in our Brief Report helps in clarifying the existing controversy on the isotope effect in Fe-based superconductors.

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