The interplay of electron doping and chemical pressure in Ba(Fe$_{1-y}$Co$_y$)$_2$(As$_{1-x}$P$_x$)$_2$

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Abstract – The effects of internal chemical pressure on electron-doped iron arsenide superconductors are studied in the series Ba(Fe$_{1-y}$Co$_y$)$_2$(As$_{1-x}$P$_x$)$_2$. Combinations of both dopants induce superconductivity also in areas where only one would not suffice, and can move the system into an overdoped state, while the highest critical temperature is not raised above 31 K in BaFe$_2$(As$_{1-x}$P$_x$)$_2$. The $T_c(x,y)$ phase diagram contradicts the creation of holes by P-substitution, but supports the finding that P-substitution is charge neutral. Chemical and physical pressure act similarly in Ba(Fe$_{1-y}$Co$_y$)$_2$(As$_{1-x}$P$_x$)$_2$, but our data reveal that the most important control parameter is the length of the Fe-As bond and not the unit cell volume. This emphasizes that differences between chemical and physical pressure, which manifest themselves as the non-linear reduction of the Fe-As distance in (As$_{1-x}$P$_x$)$_2$, are strongly linked to the superconducting properties also in the Co-doped compounds.

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Introduction. – The phase diagrams of copper-oxide, heavy-fermion, and iron-arsenide superconductors imply the occurrence of unconventional superconductivity in the proximity of a magnetic quantum critical point [1–4]. In copper oxides, solely chemical doping can destabilize the antiferromagnetic ground state in favor of the superconducting one, while in heavy-fermion and iron-based materials doping as well as pressure can induce superconductivity. Instead of applying external pressure, the volume reduction can also be achieved by the substitution of elements with those of smaller sizes, which is often referred to as chemical or internal pressure.

Virtually all possible options of suppressing the spin-density-wave (SDW) state have been realized with the iron arsenide BaFe$_2$As$_2$ [5] as parent compound. Hole doping by substitution of K for Ba [6], electron doping by substitution of Co for Fe [7], applying physical pressure [8], and finally by internal pressure through replacing smaller P-atoms for As atoms [9]. We have proposed a more sophisticated approach to chemical pressure in BaFe$_2$(As$_{1-x}$P$_x$)$_2$ by showing that the length of the Fe-As bond is a gauge of the magnetic moment, which is gradually suppressed while doping with phosphorus because the width of the 3d-bands increase [10]. On the other hand we have recently studied Ba$_{1-x}$K$_x$(Fe$_{0.93}$Co$_{0.07}$)$_2$As$_2$ and were able to compensate electron and hole doping, strongly suggesting that the charge of the (FeAs)$_3$ layer viz. band filling is likewise an important control parameter with respect to suppression of the SDW and emerging of superconductivity [11].

In this paper we study the interplay between electron doping by cobalt and the effect of internal chemical pressure by phosphorus substitution in the series Ba(Fe$_{1-y}$Co$_y$)$_2$(As$_{1-x}$P$_x$)$_2$. We present a phase diagram and analyze the mutual effects of both modifications with respect to inducing or suppressing of superconductivity. Chemical and physical pressure effects on the critical temperature are discussed in terms of the Fe-As bond length as essential control parameter.

Experimental methods. – Fe$_{1-y}$Co$_y$As$_{1-x}$P$_x$ was prepared by heating stoichiometric mixtures of iron, cobalt, arsenic and phosphorus at 973 K for 10 h in sealed silica ampoules under an atmosphere of purified argon, followed by homogenizing and sintering two times at 1123 K for 30 h. Stoichiometric amounts of Ba were added and the mixtures were heated to 943 K for 10 h.
in alumina crucibles. The products were homogenized and sintered at 1273 K and 1373 K, respectively, for another 30 h. The dc-resistivities were measured with pellets which had been cold pressed and sintered at 1273 K for 10 h. Bulk superconductivity was confirmed by ac-susceptibility measurements, and the superconducting transition temperatures were extracted by fitting the steepest descent with a line and using the point of intersection with the normal-state susceptibility as \( T_c \) (onset).

Powder diffraction data were measured using a Huber G670 diffractometer with Co-\( \text{K}_\alpha \) or Cu-\( \text{K}_\alpha \) radiation. Phase homogeneity and structural parameters were determined by Rietveld refinements with the TOPAS package [12] using the fundamental parameter approach with an empirical 2θ-dependent intensity correction for Guinier geometry. The Fe:Co ratios were fixed and the As:P ratios refined. Both Co and P contents were checked by EDX measurements for most samples, revealing variations by no more than 1% for the Co content and by no more than 2–4% for the P content. In a few samples small percentages of Fe2P were observed, while others contained traces of an unknown foreign phase with main reflection at 2θ \( \approx 28^\circ \), which was also reported to occur in similar samples [13].

**Results and discussion.** – Figure 1 shows the variation of the lattice parameters \( a \) and \( c \) with increasing phosphorus substitution for phosphorus-only–doped samples (black) [13] and samples with additional 3% (blue) or 7% cobalt (red). For all Co doping levels both lattice parameters decrease with increasing phosphorus substitution, as expected because of the smaller radius of phosphorus atoms.

Co doping seems to decrease both lattice parameters slightly, but the effect is minimal compared to the decrease due to phosphorus substitution. Thus, all structural parameters are dominated by the phosphorus content and hardly influenced by Co doping.

Although Co doping causes no significant structural changes, distinct effects on the physical properties occur. Figure 2 depicts resistivity vs. temperature plots for the series \( \text{Ba(Fe}_{1-y}\text{Co}_y)\text{As}_{1-x}\text{P}_x \) with \( y = 0.03 \) and \( x = 0.07–0.40 \) while fig. 3 presents the results of ac-susceptibility measurements confirming bulk superconductivity. \( y = 0.03 \) is just outside the superconducting dome of \( \text{Ba(Fe}_{1-y}\text{Co}_y)\text{As}_{1-x}\text{P}_x \). Consequently, a superconducting volume fraction of only 3% and a \( T_c \) (onset) \( = 3 \) K was observed for \( y = 0.0315 \) Co, while bulk superconductivity with \( T_c = 7 \) K was found for \( y = 0.0366 \) Co [14]. Substituting only \( x = 0.07 \) phosphorus for arsenic increases \( T_c \) to 14 K. EDX measurements reveal a Co concentration of \( y = 0.0322 \) in this sample, but even if bulk superconductivity could already be possible at this doping level, the transition temperature observed here is much higher than for Co doping alone.

Further substitution of arsenic by phosphorus quickly rises \( T_c \) to a maximum of 28 K at \( x \approx 0.2 \) as shown in
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Fig. 4: (Colour on-line) Ba(Fe$_{0.97}$Co$_{0.03}$)$_2$(As$_{1-x}$P$_x$)$_2$: temperature dependency of lattice parameters $a$ and $b$ (pm), for $x = 0.07$ and 0.13 orthorhombic distortion is observed.

Fig. 5: (Colour on-line) ac-susceptibility of Ba(Fe$_{0.95}$Co$_{0.05}$)$_2$(As$_{1-x}$P$_x$)$_2$ with $x = 0.16$, 0.23 and 0.36; insert: relative resistance $\rho/\rho_{300}$.

Fig. 6: Ba(Fe$_{0.93}$Co$_{0.07}$)$_2$(As$_{1-x}$P$_x$)$_2$: normalized electrical resistances $\rho/\rho_{300}$ for $x = 0$–0.45.

Fig. 7: (Colour on-line) ac-susceptibility of Ba(Fe$_{0.93}$Co$_{0.07}$)$_2$(As$_{1-x}$P$_x$)$_2$ with $x = 0.08$–0.45.

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We consider the additional P-doping of Ba(Fe$_{1-x}$Co$_x$)$_2$ (As$_{1-y}$P$_y$)$_2$, data points in black, $T_c$ is colour-coded: $T_c \approx 30$ K (dark red), $T_c \approx 10$ K (yellow), non-superconducting (blue); data for $x = 0$ and $y = 0$ are from [16] and [9].

The structure of Ba(Fe$_{1-y}$Co$_y$)$_2$(As$_{1-x}$P$_x$)$_2$ is dominated by the chemical pressure generated by phosphorus doping, therefore the comparison to physical pressure suggests itself. Several studies with Ba(Fe$_{1-y}$Co$_y$)$_2$As$_2$ report the increase of $T_c$ under pressure in underdoped samples and mostly decrease for optimally doped ones [15, 21–23]. Especially uniaxial pressure seems to have a big influence, for example in Ba(Fe$_{0.92}$Co$_{0.08}$)$_2$As$_2$, where $T_c$ rises slightly with pressure under hydrostatic conditions, while it is quickly diminished if pressure is applied along the c-axis [23] in agreement with the uniaxial pressure dependencies calculated from thermal expansion and specific heat data, with a positive value for $dT_c/dp_a$ and $dT_c/dp_c = -7$ K/GPa [24]. In the underdoped case the uniaxial pressure derivatives suggest increasing $T_c$ with c-axis pressure [25]. As phosphorus substitution intrinsically provides an uniaxial component [26], our data should agree better with studies under less hydrostatic pressure conditions. For comparison, we have related the phosphorus content to physical pressure via the cell volume, which gives 1 GPa $\approx 13.12$% phosphorus (data for (As$_{1-x}$P$_x$)$_2$ from [13], and for pressurized BaFe$_2$As$_2$ from [27], ratio of changes for $c$ and $a$ similar for both data sets). Plots of $T_c$ vs. phosphorus content (filled symbols) or physical pressure (open symbols and dashed lines) for different Co doping are displayed in fig. 9(a) ($y = 0$ (black), $y \approx 0.03$ (blue) and $y \approx 0.07$ (red)).

The trend for each Co doping level is the same both for chemical and physical pressure, however, physical pressure produces broader superconducting domes that are shifted to higher pressures. It appears as if chemical pressure is more efficient in pushing the system to the maximum critical temperature $T_{c,\text{max}}$, emphasized by the large pressure difference of $\approx 3$ GPa between BaFe$_2$As$_2$ under physical pressure and BaFe$_2$((As$_{1-x}$P$_x$)$_2$.

Considering details of the crystal structure helps to understand these discrepancies. While the lattice parameters $a$ and $c$ vary similarly under chemical and physical pressure, significant differences are found in the reduction of the Fe-As bond lengths. Figure 10 depicts the changes in the normalized Fe-As distances in BaFe$_2$(As$_{1-x}$P$_x$)$_2$ (red) [10] and BaFe$_2$As$_2$ under pressure (black) [27, 28]. While the Fe-As bonds in BaFe$_2$As$_2$ decrease linearly with increasing pressure, the Fe-As bonds in BaFe$_2$(As$_{1-x}$P$_x$)$_2$ contract rather strongly at low phosphorus doping levels and then converge [10].

The shortening of the Fe-As bond length is known to be connected with the suppression of the SDW state in BaFe$_2$(As$_{1-x}$P$_x$)$_2$, thus chemical and physical pressure may result in similar states for similar Fe-As distances. For example, in BaFe$_2$As$_2$ the Fe-As distance is reduced by $\approx 1.35\%$ at $\approx 5.5$ GPa, where $T_{c,\text{max}}$ is observed. An equal reduction of the bond length is found for $x \approx 0.3$ in BaFe$_2$(As$_{1-x}$P$_x$)$_2$, again close to the maximum of the

Fig. 8: (Colour on-line) Phase diagram of Ba(Fe$_{1-x}$Co$_x$)$_2$(As$_{1-y}$P$_y$)$_2$, data points in black, $T_c$ is colour-coded: $T_c \approx 30$ K (dark red), $T_c \approx 10$ K (yellow), non-superconducting (blue); data for $x = 0$ and $y = 0$ are from [16] and [9].

Components of the CDW transition is similar to what has been found by thermal expansion measurements [17, 18].

Recent ARPES experiments revealed inconsistent results with respect to the effect of phosphorus substitution. Ye et al. suggested that P-doping in BaFe$_2$(As$_{1-x}$P$_x$)$_2$ generates 0.3 holes per iron when one arsenic is replaced by phosphorus [19]. We have recently shown that hole and electron doping can be compensated in Ba$_{1-x}$K$_x$(Fe$_{1-y}$Co$_y$)$_2$As$_2$, where the non-superconducting magnetic state is recovered along the line $2x = y$ [11]. If we accept that P-substitution generates 0.3 holes at $x = 0.5$, these should be compensated by Co doping along the dashed line in fig. 8, but no indication of such a compensation is discernible. On the other hand, an ARPES study by Thirupathaiah et al. on EuFe$_2$(As$_{1-x}$P$_x$)$_2$ [20] concluded that P-substitution changes the electronic structure in non-rigid-band-like manner, while Co-substitution shifts the Fermi-energy due to increased band filling. Thus, our results are in agreement with the latter and support the finding that P-substitution is charge neutral.

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superconducting dome. Figure 9(b) shows a plot of $T_c$ vs. the changes in the normalized Fe-As distance for the data depicted in fig. 9(a), where a correlation between phosphorus content and physical pressure with the Fe-As distances was done by using the dashed lines of fig. 10. This shows very good agreement between BaFe$_2$As$_2$ and (As$_{1-x}$P$_x$)$_2$, indicating that, regardless of physical or chemical pressure, the Fe-As bond length is the crucial factor that controls $T_c$ if no Co doping is present.

For the Co-underdoped sample ($y = 0.03$) the maximal $T_c$ values for chemical and physical pressure coincide at the same value $-\Delta d/d = 0.005$ (green banded dome in fig. 9(b) only if we anticipate that the bond shortening with phosphorus substitution is linear in both cases. This assumes that the steeper decrease of $-\Delta d/d$ at lower phosphorus concentrations (fig. 10) is due to the suppression of the magnetism, which may already be weakened in the case of Co doping. If we alternatively anticipate that the bond length curve for chemical pressure is equally valid for the Co-doped sample, the maximum $T_c$ for chemical pressure is shifted to $-\Delta d/d \approx 0.01$ (blue filled dome in fig. 9(b)), but the shape of the dome is now more similar to that of physical pressure. A shift of the $T_{c,max}$ value for chemical pressure (filled blue) to higher $-\Delta d/d$ relative to $T_{c,max}$ for physical pressure (dashed blue) is expected due to the slightly lower Co concentrations (chemical pressure: $y = 0.03$, physical pressure $y = 0.038$). However, the observed shift is bigger than estimated by assuming a linear dependency $-\Delta d/d(T_{c,max})$, but a more concrete analysis is still not possible due to the lack of sufficiently precise structural data.

**Conclusion.** – We have studied the effects of simultaneous doping with cobalt and phosphorus in Ba(Fe$_{1-y}$Co$_y$)$_2$(As$_{1-x}$P$_x$)$_2$ and presented a comprehensive phase diagram. Superconductivity can be induced by a combination of both dopants in such areas where the content of only one would not suffice, but a combination of both can also drive the system from the optimally to an overdoped state. The maximum $T_c$ is not in the Co plus P doped area of the phase diagram, but occurs in BaFe$_2$(As$_{1-x}$P$_x$)$_2$ at $x = 0.33$. Our $T_c(x,y)$ phase diagram contradicts the creation of holes by P-substitution, because we find no compensation of these holes by Co doping as in Ba$_{1-x}$K$_x$(Fe$_{1-y}$Co$_y$)$_2$As$_2$. Thus, our results support the finding that P-substitution is charge neutral. Chemical and physical pressure act similarly in Ba(Fe$_{1-y}$Co$_y$)$_2$As$_2$, but our data strongly suggest that the most important parameter is the length of the Fe-As bond and not the unit cell volume. This shows that differences between chemical and physical pressure that manifest oneself as the non-linear reduction of the Fe-As distance in BaFe$_2$(As$_{1-x}$P$_x$)$_2$ are strongly linked to the superconducting properties also in the Co-doped materials.

Fig. 9: (Colour on-line) (a) Comparison of the effects of physical pressure on Ba(Fe$_{1-y}$Co$_y$)$_2$As$_2$ and chemical pressure in Ba(Fe$_{1-y}$Co$_y$)$_2$(As$_{1-x}$P$_x$)$_2$; continuous lines, filled symbols and areas: Ba(Fe$_{1-y}$Co$_y$)$_2$(As$_{1-x}$P$_x$)$_2$ with $y = 0$ (black), $y = 0.03$ (blue) and $y = 0.07$ (red). Open symbols and dashed lines: Ba(Fe$_{1-y}$Co$_y$)$_2$As$_2$, physical pressure: $y = 0$ (black) [29], $y = 0.038$ (blue) and $y = 0.074$ (red) [21]. (b) Plot of the same data relative to the change of the Fe-As distances taken from fig. 10. The green banded dome results if the linear decrease of the bond lengths from fig. 10 is used.

Fig. 10: (Colour on-line) Changes in the normalized Fe-As bond length in BaFe$_2$(As$_{1-x}$P$_x$)$_2$ [10] (red) and BaFe$_2$As$_2$ under pressure (black) [27,28], lines are a guide to the eye, chemical pressure related to physical pressure via cell volume.
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