Electronic structure and spin dynamics of ACo$_2$As$_2$  
($A=$Ba, Sr, Ca)

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Abstract

The electronic structures, charge and spin dynamics of the cobalt pnictide compounds ACo$_2$As$_2$ ($A=$Ba, Sr, Ca) in the paramagnetic state are investigated by using density functional theory combined with dynamical mean-field theory. In contrast to their iron counterparts, these cobalt pnictide compounds have three-dimensional electronic structures and strong ferromagnetic low-energy spin excitations. The Co 3$d$ $e_g$ orbitals dominate the electronic states around the Fermi level and have stronger electronic correlation strength than the Co 3$d$ $t_{2g}$ orbitals. The overall electronic correlation strength is much weaker than that in the iron arsenides; however, the most strongly correlated Co 3$d$ $x^2$-$y^2$ orbital, especially in CaCo$_2$As$_2$, has electronic correlation strength comparable to Fe 3$d$ $t_{2g}$ orbitals in iron arsenides. ACo$_2$As$_2$ ($A=$Ba, Sr, Ca) shows similar electronic structures where a conduction band of primarily Co 3$d$ $x^2$-$y^2$ orbital character is close to a Van Hove singularity around the Brillouin-zone corner, which promotes ferromagnetic low-energy spin excitations. Originated from its increased nearest-neighbor Co-Co distance and significantly reduced As height from the Co plane, the strong electronic correlation strength and close proximity to the Van Hove singularity of the Co 3$d$ $x^2$-$y^2$ orbital in CaCo$_2$As$_2$ is responsible for its unique A-type antiferromagnetic order observed in experiments. In comparison, despite substantial ferromagnetic low-energy spin excitations, BaCo$_2$As$_2$ and SrCo$_2$As$_2$ remain paramagnetic down to very low temperature because the Co 3$d$ $x^2$-$y^2$ orbital has weaker electronic correlation strength and is further away from the Van Hove singularity.
I. Introduction

The discovery of superconductivity at 26 K in La(O$_{1-x}$F$_x$)FeAs (x = 0.05 - 0.12) [1] creates tremendous activity in the scientific community. So far, AFe$_2$As$_2$ (A=Ba, Sr, Ca) 122 systems have been extensively studied because of the availability of large single crystals [2-6]. They all exhibit simultaneous structural and stripe antiferromagnetic (AFM) transition at the Neel temperature of 143 K (BaFe$_2$As$_2$) [7], 203 K (SrFe$_2$As$_2$) [8] and 167 K (CaFe$_2$As$_2$) [9]. The cobalt arsenides ACo$_2$As$_2$ (A= Ba, Sr, Ca) with a full substitution of Fe by Co have received increased attention recently due to their close relationship to the parent compounds of the AFe$_2$As$_2$ superconductor family.

The ACo$_2$As$_2$ compounds appear to exhibit very different behavior as compared to AFe$_2$As$_2$ (A=Ba, Sr, Ca). Experimental results show no structure phase transition and superconductivity down to 2 K for ACo$_2$As$_2$ (A=Ba, Sr, Ca) [10-12]. Neutron-diffraction measurements on SrCo$_2$As$_2$ show no evidence for long-range magnetic ordering above 2 K [11]. The isoelectronic BaCo$_2$As$_2$ exhibits the same feature, as suggested by magnetic susceptibility, electrical resistivity and specific-heat measurements [13]. In contrast to BaCo$_2$As$_2$ and SrCo$_2$As$_2$, CaCo$_2$As$_2$ undergoes an A-type AFM order below a sample-dependent Neel temperature of 52 - 76 K [14], where the magnetic moments of Fe atoms order ferromagnetically in the $ab$-plane and antiferromagnetically along the $c$-axis. Both the A-type AFM structure and the positive Weiss temperature in the Curie-Weiss law indicate that the dominant interactions in CaCo$_2$As$_2$ are ferromagnetic (FM) [10,14]. In SrCo$_2$As$_2$, nuclear magnetic resonance (NMR) measurements and inelastic neutron scattering suggest that both FM and stripe-type AFM fluctuations coexist, which is also supported by density functional theory (DFT) calculations, where the $q$-dependent static susceptibilities $\chi(q)$ at both the FM and stripe AFM in-plane wave vectors are enhanced [15,16]. Sefat et al. reported that the experimental measurements of magnetic, resistivity and thermal properties, combined with DFT calculations, show that BaCo$_2$As$_2$ is a highly renormalized paramagnet [13]. Moreover, NMR techniques indicate that both the Knight shift $\gamma_1$ and $1/T_1T$ increase toward $T = 0$ because of FM spin correlations in BaCo$_2$As$_2$. 
Furthermore, van Roekeghem et al. combined the screened exchange and dynamical mean-field-theory (SE + DDMFT) scheme to calculate the spectral function of BaCo$_2$As$_2$ and showed that the ferromagnetic instability is absent in this compound [17]. Nevertheless, extensive data analysis does not entirely rule out the possibility of AFM spin correlations [18]. Additionally, angle-resolved photoemission spectroscopy data and electronic structure calculations on BaCo$_2$As$_2$ [19,20] and SrCo$_2$As$_2$ [11] reveal a complex multiband Fermi surface without clear nesting features.

However, despite extensive research, it is still not clear why only CaCo$_2$As$_2$ has an A-type AFM transition while BaCo$_2$As$_2$ and SrCo$_2$As$_2$ exhibit paramagnetism down to 2 K. Furthermore, there is no consistent conclusion on which kind of magnetic fluctuation (AFM or FM) is dominant in ACo$_2$As$_2$ (A=Ba, Sr, Ca).

II. Computational details

In this paper, we use fully charge self-consistent density functional theory plus dynamical mean-field theory (DFT+DMFT) [21,22] to theoretically study the archetypical cobalt pnictide compounds ACo$_2$As$_2$ (A=Ba, Sr, Ca) in the paramagnetic (PM) state. The DFT part is based on the linearized augmented plane-wave method as implemented in WIEN2K [23]. We use the Perdew–Burke–Ernzerhof exchange correlation functional. Hubbard $U = 5.0$ eV and Hund’s coupling $J = 0.8$ eV are used in the calculations, consistent with previous calculations [24-26]. The formula $U(n - 1/2) - J(n - 1)/2$ ($n$ is the nominal occupation of Co 3$d$ electrons) is used to subtract the double counting. The impurity problem in DFT+DMFT calculations is solved using continuous time quantum Monte Carlo (CTQMC) method [27,28] at temperature $T = 72.5$ K. The dynamical spin structure factor is calculated using the method described in detail in Ref. [25]. We use the experimentally determined crystal structures including the internal atomic positions [10-12].

III. Results

After achieving full charge self-consistency, we compute the momentum resolved
spectra function (band structure), density of states (DOS), Fermi surface (FS), optical conductivity and spin excitation spectra. We find that the overall mass enhancement of the Co 3d electrons is much smaller in ACo$_2$As$_2$ ($A=$Ba, Sr, Ca) than that of Fe 3d electrons in BaFe$_2$As$_2$ and the Co 3d $e_g$ orbitals have larger mass enhancement than the $t_{2g}$ orbitals, in contrast to the Fe 3d orbitals in Fe-based compounds where the $t_{2g}$ orbitals have larger mass enhancement than the $e_g$ orbitals. The Co 3d $x^2$-$y^2$ orbital is the most strongly correlated orbital whereas the 3d $xy$ orbital is the least correlated orbital. The calculated dynamical spin structure factor shows that CaCo$_2$As$_2$ has strong ferromagnetic spin excitations which diverge at low energy and temperature and are responsible for the observed A-type AFM order in CaCo$_2$As$_2$. In contrast, although BaCo$_2$As$_2$ and SrCo$_2$As$_2$ also have dominate ferromagnetic spin excitations, the strength is much weaker than that in CaCo$_2$As$_2$. Therefore, they do not undergo a magnetic transition down to very low temperature and remain paramagnetic, consistent with experiments. We further trace the difference in the spin excitations among ACo$_2$As$_2$ ($A=$Ba, Sr, Ca) to differences in the electronic correlation strength of the Co 3d $x^2$-$y^2$ orbitals and the electronic structures around the Fermi level which originate primarily from the different As heights in these cobalt pnictides compounds.
Fig. 1 The electronic band structure (a-f) and Co 3d partial density of states (g-i) of BaCo$_2$As$_2$ (a, d, g), SrCo$_2$As$_2$ (b, e, h), and CaCo$_2$As$_2$ (c, f, i) obtained by DFT+DMFT calculations (a-c), (g-i) and standard DFT calculations (d-f) in the paramagnetic state.

**Band structure and density of states**

Figure 1 shows the electronic band structure and Co 3d partial density of states of ACo$_2$As$_2$ (A=Ba, Sr, Ca) based on DFT+DMFT (Fig.1.a-c, g-i) and DFT calculations (Fig.1.d-f), respectively. The conduction bands are mostly of Co 3d $e_g$ orbital character as evident in the Co 3d partial DOS shown in Fig. 1g-i, in contrast to the iron arsenide compounds. The familiar 3d $t_{2g}$ bands giving rise to the hole (electron) Fermi-surface pockets around the Brillouin-zone center (corner) Γ (M) point in the iron arsenides locate 0.6-0.8 eV below the Fermi level, indicating a somewhat rigid-band shift of the Fermi level due to substitution of Fe by Co.

The overall electronic structures of ACo$_2$As$_2$ (A=Ba, Sr, Ca) are similar. The main difference lies in a flat conduction band of interest near the Fermi level around the M point. This band gives rise to a sharp peak in the partial DOS of the Co 3d $x^2-y^2$ orbital [Fig.1g-i] signaling this band of Co 3d $x^2-y^2$ orbital character is close to a Van Hove
singularity. The proximity to a Van Hove singularity near the Fermi level leads to high
density of states at the Fermi level and promotes ferromagnetic low-energy spin
excitations according to the Stoner criterion. The peak position in the partial DOS of
the Co 3d $x^2-y^2$ orbital is very close to the Fermi level in CaCo$_2$As$_2$ whereas it is about
35 meV above the Fermi level in BaCo$_2$As$_2$ and SrCo$_2$As$_2$, suggesting CaCo$_2$As$_2$ is in
close proximity to the Van Hove singularity while BaCo$_2$As$_2$ and SrCo$_2$As$_2$ are further
away from the Van Hove singularity, which is a main reason that CaCo$_2$As$_2$ has much
stronger ferromagnetic low-energy spin excitations than BaCo$_2$As$_2$ and SrCo$_2$As$_2$ as
shown later in the paper. Moreover, in CaCo$_2$As$_2$, two conduction bands around the M
point cross the Fermi level and each other to form two separated dumbbell-shape hole-
electron pockets, further promoting ferromagnetic (zero-momentum-transfer) low-
energy spin excitations.

The absence of ferromagnetism in BaCo$_2$As$_2$ has been discussed previously in the
context of SE + DDMFT calculations [17], where it is argued that the long-range
exchange interaction reduces the density of states at the Fermi level and avoids the
ferromagnetic order according to the Stoner criterion. Here we show that without taking
into account the long-range exchange interaction, correlation effects due to local
Coulomb interaction already avoid the ferromagnetic ordering in BaCo$_2$As$_2$.

The DFT band structures are also similar to the corresponding DFT+DMFT band
structures, subject to renormalization of the bandwidth and some shift of the band
positions. The bandwidths of the Co 3d bands around the Fermi level do not vary much
among the cobalt pnictide compounds. For example, the DFT+DMFT (DFT) parabolic
band around $\Gamma$ point (along the Y-$\Gamma$-X path) giving rise to an electron Fermi surface
pocket is mainly of Co 3d $x^2-y^2$ orbital character and has a bandwidth of 1.25 (1.55),
1.33 (1.66), and 1.32 (1.71) eV in BaCo$_2$As$_2$, SrCo$_2$As$_2$, and CaCo$_2$As$_2$, respectively.
This results in overall band renormalization factors of 1.23, 1.25 and 1.29 for the Co 3d
$x^2-y^2$ orbital in BaCo$_2$As$_2$, SrCo$_2$As$_2$, and CaCo$_2$As$_2$, respectively, which are
significantly smaller than the corresponding mass enhancement of above 2 at the Fermi
level calculated by $m^*/m_{band} = 1 - \partial \Sigma'(\omega = 0)/\partial \omega$ from the quasiparticle self-
energy $\Sigma'(\omega)$ as shown in Table I, suggesting significant nonuniform renormalization
of the band at different energies.

The overall mass enhancement of the Co 3d orbitals in ACo$_2$As$_2$ (A=Ba, Sr, Ca) are below 2 as shown in Table I, indicating much weaker electronic correlation effects than iron arsenide compounds, for which mass enhancement of about 3 was reported [24,25]. This is mainly due to an average increase of 0.2 of the Co 3d orbital occupation because of the substitution of Fe with Co. The Co 3d $e_g$ orbitals have stronger electronic correlation strength than the Co 3d $t_{2g}$ orbitals, in strong contrast to the iron arsenide compounds where the Fe 3d $t_{2g}$ orbitals have stronger electronic correlation strength than the Fe 3d $e_g$ orbitals. The Co 3d $x^2$-$y^2$ orbital has the strongest electronic correlation strength among all the Co 3d orbitals, and in CaCo$_2$As$_2$ its electronic correlation strength is comparable to that of the Fe 3d $t_{2g}$ orbitals in iron arsenides [25]. The stronger electronic correlation strength and higher density of states due to closer proximity to the Van Hove singularity of the Co 3d $x^2$-$y^2$ orbital in CaCo$_2$As$_2$ than in BaCo$_2$As$_2$ and SrCo$_2$As$_2$ result in much stronger ferromagnetic low-energy spin excitations in CaCo$_2$As$_2$ than in BaCo$_2$As$_2$ and SrCo$_2$As$_2$ as shown below.

Table I. Mass enhancement obtained from the quasiparticle self-energy and the CTQMC averaged orbital occupation of Co 3d orbitals from the charge self-consistent DFT+DMFT calculations.

|        | Mass enhancement | Orbital occupation |
|--------|------------------|--------------------|
|        | $z^2$  | $x^2$-$y^2$ | $xz/yz$ | $xy$ | $z^2$ | $x^2$-$y^2$ | $xz/yz$ | $xy$ |
| BaCo$_2$As$_2$ | 1.73  | 2.05       | 1.61   | 1.54 | 1.45 | 1.31       | 1.49   | 1.55 |
| SrCo$_2$As$_2$ | 1.76  | 2.16       | 1.59   | 1.52 | 1.43 | 1.29       | 1.50   | 1.56 |
| CaCo$_2$As$_2$ | 1.64  | 2.53       | 1.46   | 1.33 | 1.45 | 1.30       | 1.50   | 1.57 |

**Fermi surfaces**

The DFT+DMFT calculated three-dimensional FSs for ACo$_2$As$_2$ (A=Ba, Sr, Ca) are plotted in Fig. 2 in the PM tetragonal Brillouin zone. The corresponding two dimensional FS cuts in the $k_z = 0$ and $2\pi/c$ plane are displayed in the unfolded Brillouin zone of the one-Co unit cell. As shown in Fig.2(a-c), the FS has multi-sheets which are
large and quite three dimensional, in contrast to their Fe counterparts and consistent with the anisotropy of the measured resistivity [13] and the calculated optical conductivity shown below. As expected from the above analysis of the band structure in the vicinity of $E_F$, the FS topology of $\text{ACo}_2\text{As}_2 (A=\text{Ba, Sr, Ca})$ is quite different from that of the Fe pnictides. For example, the more or less circular hole FS pockets around the $\Gamma$ point in the $k_z=0$ plane for $\text{BaFe}_2\text{As}_2$ [25] are replaced by star-shape electron FS pockets (rather than hole pockets) in $\text{ACo}_2\text{As}_2 (A=\text{Ba, Sr, Ca})$, thus removing the electron-hole quasinesting scattering from $\mathbf{k}=(0,0)$ to $(1,0)$ in cobalt pnictide materials, as shown in Fig.2d-i. In addition, we find that $\text{CaCo}_2\text{As}_2$ shows two separated dumbbell-shape FS pockets at $\mathbf{k}=(1,0)$ in the $k_z=0$ plane, in contrast to an integrated dumbbell-shape FS pocket in $\text{BaCo}_2\text{As}_2$ and $\text{SrCo}_2\text{As}_2$. Similarly, we find that the FS pockets around $\mathbf{k}=(0,0)$ in the $k_z=2\pi/c$ plane are surrounded by two sets of four smaller FS pockets and exhibit changes of size and shape among $\text{ACo}_2\text{As}_2 (A=\text{Ba, Sr, Ca})$. At $\mathbf{k}=(1,1)$, the shape of the FS pocket in the $k_z=2\pi/c$ plane alters from star shape to square shape to circle shape in these cobalt pnictide compounds. In particular, comparing with $\text{BaCo}_2\text{As}_2$ and $\text{SrCo}_2\text{As}_2$, $\text{CaCo}_2\text{As}_2$ manifests a distinctive feature, where it has extra FS pockets at $\mathbf{k}=(1,1)$ in the $k_z=0$ plane and at $\mathbf{k}=(1,0)$ in the $k_z=2\pi/c$ plane, respectively.
Fig. 2 Three-dimensional Fermi surfaces for (a) BaCo$_2$As$_2$, (b) SrCo$_2$As$_2$ and (c) CaCo$_2$As$_2$ plotted in the PM tetragonal Brillouin zone obtained by DFT+DMFT calculations. The corresponding two-dimensional FS cuts in $k_z = 0$ and $2\pi/c$ plane of BaCo$_2$As$_2$ (d, e), SrCo$_2$As$_2$ (f, g), and CaCo$_2$As$_2$ (i, j) in the first Brillouin zone of the one-Co unit cell. The black, red, green, and blue colors denote the Co 3$d^{x^2}$, $x^2-y^2$, $xz/yz$, and $xy$ orbital characters, respectively. In panels (d-i), the units of the reciprocal-lattice vector $k_x$ and $k_y$ are $\sqrt{2}\pi/a$. 
Fig. 3 Dynamic spin structure factor $S(q, \omega)$ for BaCo$_2$As$_2$ (left column) and CaCo$_2$As$_2$ (right column). From top to bottom are the total $S(q, \omega)$ (a, e) and the diagonal component $S_{\alpha,\alpha}(q, \omega)$ ($\alpha$ is the orbital index) of Co 3$d$ $x^2$-$y^2$ orbital (b, f) and $z^2$ orbital (c, g) along the high-symmetry path (0, 0)→(1,0)→(1,1)→(0,0), and (d, h) the 2D plot in the L=0 plane with constant energy $\omega = 5 \text{ meV}$. The $S(q, \omega)$ of SrCo$_2$As$_2$ is very similar to that of BaCo$_2$As$_2$ thus is not shown. The wave vector is defined in terms of the reciprocal lattice of the one-Co unit cell.
Spin dynamics

The calculated dynamical spin structure factor $S(q, \omega) = \chi''(q, \omega) / (1 - \exp(-\hbar\omega/k_BT))$, which can be measured by inelastic neutron-scattering experiments [29], is shown in Fig.3 for BaCo$_2$As$_2$ (left column) and CaCo$_2$As$_2$ (right column). The dynamical spin structure factor of SrCo$_2$As$_2$ is similar to that of BaCo$_2$As$_2$ and hence is not shown. As is clear from Fig.3a & e, the low-energy spin excitations are much weaker in BaCo$_2$As$_2$ (and SrCo$_2$As$_2$) than in CaCo$_2$As$_2$. Two broad maxima appear at $q = (0, 0)$ and the spin-density wave ordering vector $q = (1, 0)$ in BaCo$_2$As$_2$ (see Fig. 3a-d), corresponding to FM spin excitations and stripe AFM spin excitations, respectively. In contrast, the one-order-of-magnitude stronger low-energy spin excitation in CaCo$_2$As$_2$ is mainly concentrated on the FM ordering vector $q = (0, 0)$ (see Fig. 3e-h), which is expected to diverge at zero energy and low temperature, leading to ferromagnetic ordering of the Co magnetic moments in the Co plane.

We further decompose the dynamical spin structure factor $S(q, \omega) = \sum_{\alpha,\beta} S_{\alpha,\beta}(q, \omega)$ into different orbital contributions, where $\alpha$ and $\beta$ are orbital indices. We find that the low-energy spin excitations are mainly contributed by the Co 3$d$ $e_g$ orbitals, in strong contrast to iron arsenides where the low-energy spin excitations are mainly contributed by Fe 3$d$ $t_{2g}$ orbitals [30,31]. As shown in Fig. 3b-c and Fig. 3f-g, the diagonal component $S_{\alpha,\alpha}(q, \omega)$ of the Co 3$d$ $x^2$-$y^2$ orbital is five times that of the 3$d$ $z^2$ orbital, whereas the intensities of the Co 3$d$ $t_{2g}$ orbitals are negligibly small. Therefore, the low-energy spin excitations are primarily contributed by the Co 3$d$ $x^2$-$y^2$ orbital due to the aforementioned reasons. Furthermore, the intensity of the Co 3$d$ $x^2$-$y^2$ orbital in CaCo$_2$As$_2$ is one order of magnitude larger than that in BaCo$_2$As$_2$ (Fig.3b, f) due to the stronger electronic correlation strength and closer proximity to the Van Hove singularity of the Co 3$d$ $x^2$-$y^2$ orbital in CaCo$_2$As$_2$ than in BaCo$_2$As$_2$ as discussed above. This explains why CaCo$_2$As$_2$ exhibits unique A-type AFM order at low temperature while BaCo$_2$As$_2$ and SrCo$_2$As$_2$ remain paramagnetic down to very low temperature (~2 K).
Optical conductivity

Fig. 4 shows the in-plane and out-of-plane optical conductivity of ACO$_2$As$_2$ (A=Ba, Sr, Ca) in the PM state calculated by DFT+DMFT. In Fig. 4a, a broad peak, due to interband transitions, is centered around 1.0 eV for in-plane optical conductivity in ACO$_2$As$_2$ (A=Ba, Sr, Ca) compounds. In contrast, the out-of-plane optical conductivity does not show a clear characteristic peak, whereas it does exhibit obvious differences among ACO$_2$As$_2$ (A=Ba, Sr, Ca) compounds. Near zero frequency, the optical conductivity spectrum in the PM phase shows Drude-like behavior as shown in Fig. 4, where the out-of-plane Drude weight is substantially larger for the cobalt pnictide compounds. The plasma frequencies $\omega_{p,xx}$ and $\omega_{p,xz}$ along the x and z directions for ACO$_2$As$_2$ (A=Ba, Sr, Ca) and BaFe$_2$As$_2$, obtained from both DFT+DMFT and standard DFT calculations, are shown in Table II. The plasma frequencies from DFT+DMFT calculations are obtained by fitting the calculated low-energy optical conductivity to the Drude function $\sigma_1(\omega) = \frac{\omega_p^2 \Gamma}{\omega^2 + \Gamma^2}$ where $\omega_p$ is the plasma frequency and $\Gamma$ is the scattering rate. The in-plane plasma frequency $\omega_{p,xx}$ is obviously larger in ACO$_2$As$_2$ (A=Ba, Sr, Ca) than in BaFe$_2$As$_2$, especially for DFT+DMFT calculations. The average mass enhancement $\frac{m}{m_{\text{band}}} = \frac{\omega_p(DFT)^2}{\omega_p(\text{DMFT})^2}$ is much smaller for the cobalt pnictide compounds, indicating weaker electronic correlations, consistent with the estimation from the electronic bandwidth, the quasiparticle self-energy, and
experimental results.

The in-plane and out-of-plane conductivity anisotropy, defined as \( \sigma_{1,x}/\sigma_{1,z} = \omega_{p,xx}^2/\omega_{p,zz}^2 \) (assuming the same scattering rate at low temperature), is above 10 in BaFe\(_2\)As\(_2\) as shown in Table II, suggesting strong in-plane and out-of-plane anisotropies and semi-two-dimensional electronic structures in BaFe\(_2\)As\(_2\). In contrast, the in-plane and out-of-plane conductivity anisotropies are around unity in AC\(_2\)As\(_2\) (A=Ba, Sr, Ca). Therefore, AC\(_2\)As\(_2\) (A=Ba, Sr, Ca) has three-dimensional electronic structures as shown above. In particular, CaCo\(_2\)As\(_2\) has better conductivity along the \( c \) axis than in the \( ab \) plane.

Table II. The in-plane and out-of-plane plasma frequency (\( \omega_{p,xx} \), \( \omega_{p,zz} \), respectively), the in-plane and out-of-plane conductivity anisotropy \( \sigma_{1,x}/\sigma_{1,z} = \omega_{p,xx}^2/\omega_{p,zz}^2 \) and the average mass enhancement along \( xx \) and \( zz \) directions in AC\(_2\)As\(_2\) (A=Ba, Sr, Ca) and BaFe\(_2\)As\(_2\) from Ref. [24, 25].

|          | \( \omega_{p,xx} \) (eV) | \( \omega_{p,zz} \) (eV) | \( \omega_{p,xx} \) (eV) | \( \omega_{p,zz} \) (eV) | \( \sigma_{1,x}/\sigma_{1,z} \) | \( \sigma_{1,x}/\sigma_{1,z} \) | Mass enhancement \( xx \) | Mass enhancement \( zz \) |
|----------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| BaCo\(_2\)As\(_2\) | 3.66            | 3.37            | 2.77            | 3.06            | 1.18            | 0.82            | 1.75            | 1.21            |
| SrCo\(_2\)As\(_2\) | 3.54            | 3.45            | 2.95            | 2.96            | 1.05            | 0.99            | 1.44            | 1.36            |
| CaCo\(_2\)As\(_2\) | 2.87            | 3.57            | 2.37            | 3.20            | 0.65            | 0.55            | 1.47            | 1.24            |
| BaFe\(_2\)As\(_2\) | 2.63            | 0.77            | 1.60            | 0.5             | 11.7            | 10.2            | 2.7             | 2.4             |

IV. Discussions

The dimensionality of the electronic structures and the electronic correlation strength are closely connected to the crystal structure parameters [25]. In Table III, we collect the \( c \)-lattice constant which is twice the interlayer distance of the neighboring Co (Fe) layers, the bond length of Co-Co (Fe-Fe) and Co-As (Fe-As), and the anion As height from the Co (Fe) layer in AC\(_2\)As\(_2\) (A=Ba, Sr, Ca) [10-12] and BaFe\(_2\)As\(_2\) [32]. The substantial reduction of the interlayer distance in the cobalt arsenides from BaFe\(_2\)As\(_2\),
~20% in CaCo₂As₂, is responsible for the significantly increased three-dimensional electronic structures and decreased in-plane and out-of-plane anisotropies in the cobalt arsenides shown above. While the nearest-neighbor Co-Co and Fe-Fe distances do not differ much, the As height in the cobalt arsenides is substantially lower than that in BaFe₂As₂, leading to a strongly distorted As₄ tetrahedron away from its perfect shape and different crystal field splitting, which reduces the electronic correlation strength of the 3d t²₋₂g orbitals and promotes the electronic correlation strength of the 3d eₓ²₋ᵧ² orbitals [25]. Among ACo₂As₂ (A=Ba, Sr, Ca), CaCo₂As₂ has the largest nearest-neighbor Co-Co bond length and lowest As height, therefore its Co 3d x²₋ᵧ² orbital has the strongest electronic correlation strength and plays a crucial role in its unique A-type AFM order at low temperature.

Table III. The c-lattice constant, bond length of Co-Co (Fe-Fe) and Co-As (Fe-As), and the anion As height from Co (Fe) layer in ACo₂As₂ (A=Ba, Sr, Ca) [10-12] and BaFe₂As₂ [32].

|          | c (Å)    | Co-Co (Å) | Co-As (Å) | hCo-As (Å) |
|----------|----------|-----------|-----------|------------|
| BaCo₂As₂ | 12.670   | 2.798     | 2.356     | 1.278      |
| SrCo₂As₂ | 11.773   | 2.791     | 2.352     | 1.280      |
| CaCo₂As₂ | 10.273   | 2.817     | 2.327     | 1.204      |
| BaFe₂As₂ | 13.017   | 2.802     | 2.403     | 1.360      |

V. Conclusions
While the cobalt arsenides ACo₂As₂ (A=Ba, Sr, Ca) are isostructural to their iron counterparts AFe₂As₂, they show quite different (three-dimensional) electronic structures and (strong ferromagnetic) spin excitations. Except the 3d x²₋ᵧ² orbital, the mass enhancement of the Co 3d orbitals is below 2, indicating much weaker electronic correlation strength than that in the corresponding iron compounds. The Co 3d eₓ²₋ᵧ² orbitals dominate the electronic states around the Fermi level and have stronger electronic correlation strength than the Co 3d t²₋₂g orbitals, in contrast to the iron arsenide compounds where the Fe 3d t²₋₂g orbitals have stronger electronic correlation strength than the Fe 3d eₓ²₋ᵧ² orbitals [25]. The Co 3d x²₋ᵧ² orbital has the strongest electronic
correlation strength among all the Co 3d orbitals, especially in CaCo$_2$As$_2$, which is comparable to the correlation strength of the Fe 3d $t_{2g}$ orbitals in BaFe$_2$As$_2$. The conduction band of primarily Co 3d $x^2$-$y^2$ orbital character around the M point is close to a Van Hove singularity, which promotes ferromagnetic low-energy spin fluctuations. The low-energy spin excitations are mainly contributed by the Co 3d $x^2$-$y^2$ orbital. The combined effects of strong electronic correlation strength and high density of states at the Fermi level (due to a Van Hove singularity just above the Fermi level) of the Co 3d $x^2$-$y^2$ orbital are responsible for the unique A-type AFM order observed in CaCo$_2$As$_2$. Despite substantial ferromagnetic low-energy spin excitations, BaCo$_2$As$_2$ and SrCo$_2$As$_2$ remain paramagnetic down to very low temperature because the Co 3d $x^2$-$y^2$ orbital has weaker electronic correlation strength and is further away from the Van Hove singularity. The unique magnetic properties of CaCo$_2$As$_2$ originate from the increased nearest neighbor Co-Co distance and the significantly reduced As height from the Co plane.

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