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We present first-principles calculations of the coupling of quasiparticles to spin fluctuations in iron selenide and discuss which types of superconducting instabilities this coupling gives rise to. We find that strong antiferromagnetic stripe-phase spin fluctuations lead to large coupling constants for superconducting gaps with $s_\pm$-symmetry, but these coupling constants are significantly reduced by other spin fluctuations with small wave vectors. An accurate description of this competition and an inclusion of band structure and Stoner parameter renormalization effects lead to a value of the coupling constant for an $s_\pm$-symmetric gap which can produce a superconducting transition temperature consistent with experimental measurements.

Introduction.—The discovery of superconductivity in iron-based compounds with transition temperatures higher than 50 Kelvin in 2008 [1] has generated considerable interest in recent years and led to intense research activity. These materials consist of iron atoms with additional pnictogen or chalcogen atoms located above and below the plane of the iron atoms. Additional intercalating layers give rise to various families of iron-based compounds, such as the 1111-, 122-, 111-families [2]. Iron selenide (FeSe) belongs to the structurally simplest family, the 11-family, which does not contain a spacer layer between the iron-selenium layer and exhibits superconductivity with transitions temperatures up to 37 Kelvin under pressure [3, 4]. Recently, there has also been considerable interest in the properties of FeSe monolayers, where superconductivity with transition temperatures exceeding 60 Kelvin has been reported [5, 6].

An important question in these materials is the nature of the microscopic pairing mechanism and the symmetry of the superconducting gap. As in the cuprates, superconductivity typically emerges in the iron-based compounds when an antiferromagnetic parent state is doped. This observation led to the proposal [7] that spin fluctuations (paramagnons) could act as the superconducting glue in these materials [7]. However, in contrast to the cuprates, the iron-based materials generally have multiple Fermi surfaces with several electron pockets at the M-point of the Brillouin zone (corresponding to the unit cell containing two iron atoms) and multiple hole pockets at the Γ-point. Mazin *et al.* suggested that spin-fluctuation mediated scattering of Cooper pairs between electron pockets and hole pockets gives rise to an $s_\pm$-symmetric superconducting gap [7], which has a constant absolute magnitude, but switches sign between electron and hole pockets. The sign-changing gap was predicted to give rise to a resonance in the neutron scattering spectrum [8], which was subsequently observed in several experiments [9, 10].

Many theoretical approaches have been developed to study spin-fluctuation mediated superconductivity in the iron-based compounds. In purely empirical approaches, both the electronic band structure and the interacting spin susceptibility are parametrized using experimental data (such as angle-resolved photoemission, nuclear magnetic resonance and neutron scattering results)[11, 12]. In another approach [13, 14], a theoretical band structure from a density-functional theory calculation is used to parametrize a tight-binding Hamiltonian with added interaction parameters (such as the Hubbard U or Hund's J) adjusted to reproduce experimental findings. Then, superconducting properties of the resulting Hamiltonian are studied.

While the aforementioned theories have been very instructive, their applications have been limited by the availability of concrete experimental data needed to determine their input parameters. Hence, there is a need for a fully first-principles theory without empirical parameters. We have recently developed such an *ab initio* theory [15] for the spin fluctuation-electron coupling based on the work of Overhauser and coworkers on the homogeneous electron gas [16, 17].

In this paper, we apply our first-principles theory of the electron-spin fluctuation coupling to iron selenide, the structurally simplest iron-based superconductor. We reveal a complex interplay between different spin fluctuations. In agreement with experiment, superconductivity depends sensitively on the height of the selenium atoms relative to the iron atom plane. In the vicinity of a transition to an antiferromagnetic stripe phase, superconductivity with an $s_\pm$-symmetric gap function becomes favorable.

Methods.—The superconducting order parameter, the gap function, is typically expressed as $\Delta(\boldsymbol{k}) = |\Delta|g(\boldsymbol{k})$ with $g(\boldsymbol{k})$ describing the symmetry of the gap function

[18]. To investigate which types of superconducting instabilities an effective spin-fluctuation-mediated electron-electron interaction V_{SF} gives rise to, we compute the electron-spin fluctuation coupling strength for different symmetry functions $g(\boldsymbol{k})$ according to [18]

$$\lambda[g] = -D_F \times \tag{1}$$

$$\frac{\sum_{n\boldsymbol{k},n'\boldsymbol{k'}} g(\boldsymbol{k})V_{SF}(n\boldsymbol{k},n'\boldsymbol{k'})g(\boldsymbol{k'})\delta(\epsilon_{n\boldsymbol{k}}-\epsilon_F)\delta(\epsilon_{n'\boldsymbol{k'}}-\epsilon_F)}{[\sum_{n\boldsymbol{k}} g^2(\boldsymbol{k})\delta(\epsilon_{n\boldsymbol{k}}-\epsilon_F)]^2}, \tag{2}$$

where ϵ_F denotes the Fermi energy and D_F is the density of states per spin at the Fermi energy. We define $V_{SF}(n\boldsymbol{k},n'\boldsymbol{k'})$ as the matrix element of the spin-fluctuation mediated interaction for the scattering of the spin-singlet Cooper pair $(n\boldsymbol{k}\uparrow, n-\boldsymbol{k}\downarrow)$ to $(n'\boldsymbol{k'}\uparrow, n'-\boldsymbol{k'}\downarrow)$ (with n denoting a band and $\boldsymbol{k}$ a $\boldsymbol{k}$-point in the Brillouin zone). We follow the standard convention [18] and normalize $g(\boldsymbol{k})$, such that $D_F = \sum_{n\boldsymbol{k}} g^2(\boldsymbol{k})\delta(\epsilon_{n\boldsymbol{k}}-\epsilon_F)$.

The effective spin-fluctuation mediated interaction is obtained using a recently developed first-principles formalism [15, 19] based on the work of Overhauser *et al.* for the homogeneous electron gas [16, 17]. In this approach, the effective interaction is expressed as the sum of a bare Coulomb interaction, a contribution arising from charge fluctuations and a contribution arising from spin fluctuations, which is given by

$$V_{SF}(\boldsymbol{r},\boldsymbol{r'},\omega) = 3 \times$$

$$\int d\boldsymbol{r}_1 \int d\boldsymbol{r}_2 I_{xc}(\boldsymbol{r},\boldsymbol{r}_1)\chi_S(\boldsymbol{r}_1,\boldsymbol{r}_2,\omega)I_{xc}(\boldsymbol{r}_2,\boldsymbol{r'}), \tag{3}$$

with $I_{xc}(\boldsymbol{r},\boldsymbol{r'}) = \delta^2 E_{xc}/[\delta m(\boldsymbol{r})\delta m(\boldsymbol{r'})]$ (E_{xc} is the exchange-correlation energy and $m(\boldsymbol{r})$ the spin density). Also, $\chi_S(\boldsymbol{r},\boldsymbol{r'},\omega)$ denotes the interacting spin susceptibility of a nonmagnetic system, which is obtained by solving the Dyson-like equation of time-dependent density-functional theory [20]

$$\chi_S(\boldsymbol{r},\boldsymbol{r'},\omega) = \chi_0(\boldsymbol{r},\boldsymbol{r'},\omega)$$

$$+ \int d\boldsymbol{r}_1 \int d\boldsymbol{r}_2 \chi_0(\boldsymbol{r},\boldsymbol{r}_1,\omega)I_{xc}(\boldsymbol{r}_1,\boldsymbol{r}_2)\chi_S(\boldsymbol{r}_2,\boldsymbol{r'},\omega), \tag{4}$$

with χ_0 denoting the non-interacting susceptibility.

The physically appealing effective spin fluctuation-mediated interaction of Overhauser *et al.* can be put on a firm theoretical footing by an analysis of Feynman diagrams [21, 22]. While this theory neglects certain diagrams corresponding to nonlinear polarization processes, it should be valid in the vicinity of the Fermi surface and thus describe superconducting properties accurately.

Computational details.—To obtain a mean-field theory starting point for the calculation of the spin-fluctuation mediated interaction, we carry out density-functional theory calculations in a plane-wave basis using the Quantum ESPRESSO program package [23]. We employ the local-density approximation (Perdew-Zunger parametrization) and norm-conserving pseudopotentials with a 55 Ry energy cutoff. For iron, the non-linear core correction is used. We use experimental lattice constants ($a = 3.77$ Å, $c = 5.52$ Å) [24] of the tetragonal phase. We have carefully verified that our band structures and magnetic phase diagrams agree with all-electron results [25, 26].

We then compute the non-interacting susceptibilities on a $32 \times 32 \times 1$ $\boldsymbol{k}$-point grid in the Brillouin zone using 50 empty states and a plane-wave cutoff of 45 Ry. For this, we use the BerkeleyGW program package [27]. Next, we compute the interacting spin susceptibilities. Within the local-density approximation, we need to evaluate $I_{xc}^{LSDA}(\boldsymbol{r},\boldsymbol{r'}) = f(n(\boldsymbol{r}))\delta(\boldsymbol{r}-\boldsymbol{r'})$ with $n(\boldsymbol{r})$ being the ground-state density (including the core contribution) and $f(n)$ is obtained analytically by computing the second derivative of the exchange-correlation energy with respect to the spin density. Finally, the Fermi surface averages in Eq. (2) are evaluated using a Lorentzian representation of the δ-function $[\delta_\eta(\omega) = 1/\pi \times \eta/(\omega^2+\eta^2)]$ with $\eta = 0.1$ eV.

Results—. Figure 1(a) shows the magnetic moments of various magnetic phases of FeSe as a function of the selenium height, which has been identified as a crucial parameter for the occurence of superconductivity in this system and other iron-based compounds [28]. At low selenium heights, the system is nonmagnetic. As the height is increased, a transition to an antiferromagnetic stripe phase occurs. At even higher Se heights, an antiferromagnetic checkerboard phase also has a lower energy than the nonmagnetic state and a moment develops. Finally, at Se heights larger than 1.4 Å a ferromagnetic moment is observed. Note that bulk FeSe is experimentally known to be nonmagnetic [3], while a recent ARPES experiment reported indications that multilayer FeSe is in a stripe phase state [29].

Figure 1(b) shows the DFT-LDA band structure of FeSe at the experimental selenium height of 1.47 Å[24]. We observe three hole pockets in the vicinity of the Γ-point and two electron pockets near the M-point (note that a Brillouin zone corresponding to a unit cell containing *two* iron atoms is used). While DFT-LDA band structures agree qualitatively with experimental ARPES measurements for many iron-based compounds, the effective masses of the electron and hole pockets near the Fermi level are typically underestimated by a factor of 2 or 3 [30, 31]. A similar finding was reported in a recent ARPES experiment on FeSe multilayers [29]. In Fig. 1(b) we also show the renormalized band structure obtained by dividing all DFT-LDA energies by a factor of 3. We note that a recent Shubnikov-de Haas oscillation measurement reported significantly reduced Fermi surfaces [32]. Further experimental work is necessary to fully resolve this issue.

Figure 2 shows the real part of the DFT-LDA static non-interacting and interacting spin susceptibilities at
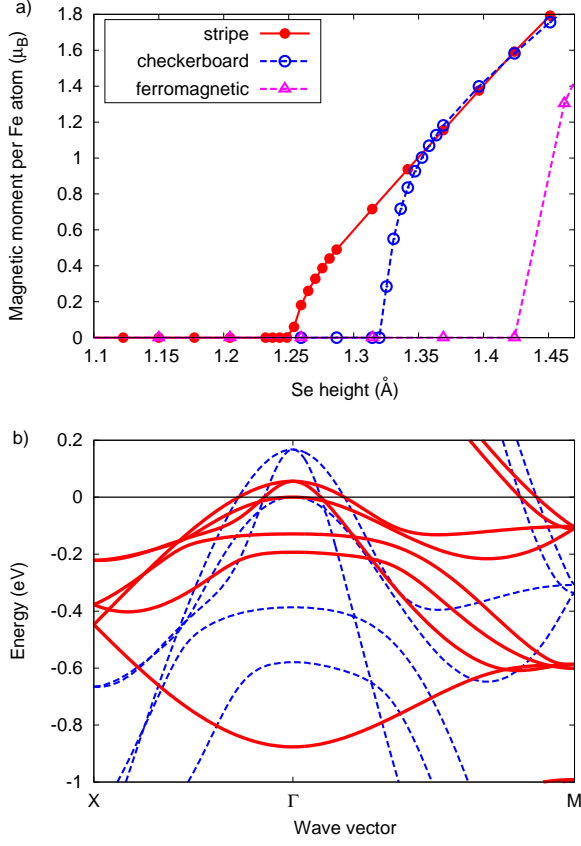
FIG. 1. (a): Magnetic moment per iron atom in the stripe an-
tiferromagnetic phase (red filled dots), checkerboard antifer-
romagnetic phase (blue empty dots) and ferromagnetic phase
(magenta diamonds) as function of the height of the selenium
atoms above (and below) the plane of the iron atoms. (b)
DFT-LDA band structure of iron selenide at the experimen-
tal selenium height (dashed blue lines) and the renormalized
band structure (solid red lines), where all energies have been
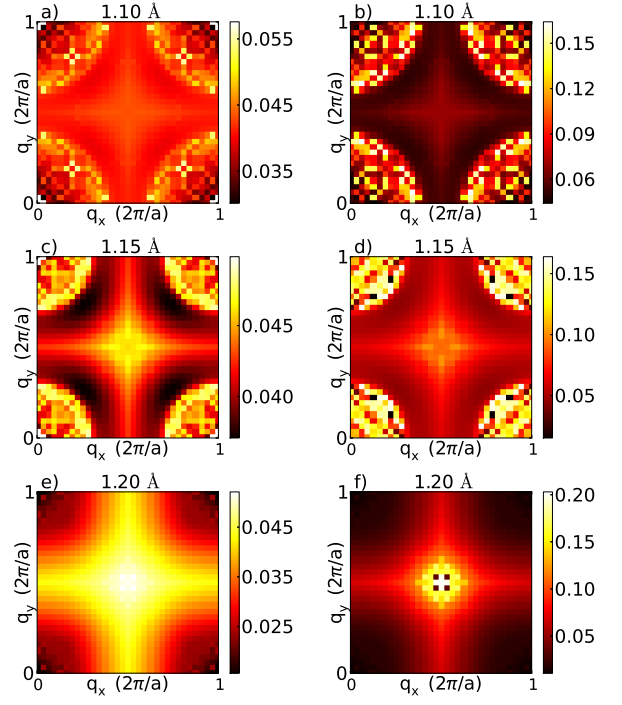divided by a factor of 3 [29].

FIG. 2. Real parts of the static non-interacting [(a), (c),
(e)] and interacting [(b), (d), (f)] spin susceptibilities of iron
selenide for selenium heights of 1.10 Å [(a) and (b)], 1.15 Å [(c)
and (d)] and 1.20 Å [(e) and (f)]. Shown is the $\boldsymbol{G} = \boldsymbol{G}' = 0$
component of the susceptiblity matrix in Fourier space.

three Se heights in the vicinity of the transition from the
nonmagnetic phase to the antiferromagnetic stripe phase.
At the smallest Se height (1.10 Å), we observe strong
features in the non-interacting susceptibility near $\boldsymbol{q} = 0$.
In the interacting spin susceptibility, these features are
strongly enhanced indicating that at these wave vectors
the Stoner condition, which states that magnetism occurs
if $I_{xc}\chi_0(\boldsymbol{q})$ (in matrix notation) has eigenvalues equal to
unity, is almost fulfilled.

At a selenium height of 1.15 Å [see Figs. 2b) and c)],
another feature at $\boldsymbol{q} = (1/2,\ 1/2,\ 0)\frac{2\pi}{a}$ emerges in ad-
dition to the structure near $\boldsymbol{q} = 0$ in the non-interacting
susceptibility. This feature results from spin fluctuations
with stripe phase character. When interactions are in-
cluded, this new feature, however, becomes weaker.

Finally, at a selenium height of 1.20 Å [see Figs. 2e)
and f)], the system is very close to the transition to the
antiferromagnetic stripe phase, see Fig. 1(a). Now, the

peak at $\boldsymbol{q} = (1/2,\ 1/2,\ 0)\frac{2\pi}{a}$ becomes very strong and is
further enhanced when interactions are included. Again,
the divergence of the interacting spin susceptibility indi-
cates that the Stoner criterion is almost fulfilled signaling
the onset of a transition to a stripe phase.

To gain further insight, we fit our results to a model
where off-diagonal elements of the non-interacting sus-
ceptibilities in a plane-wave basis are neglected (neglect-
ing the so-called local-field effects). We then adjust the
Stoner parameter I_{xc}, which now is a single number, un-
til the model reproduces the diagonal elements of the
previously computed interacting spin susceptibility. In
this way, we extract $I_{xc} = 0.43$ eV (per Fe atom), which
agrees very well with the Stoner parameter for bcc iron
$I_{xc} = 0.46$ eV [33]. In addition, we find that the same
value of I_{xc} can be used for all selenium heights that were
studied. This indicates that the Stoner parameter is only
weakly dependent on the crystalline environment.

Next, we compute the electron-spin fluctuation cou-
pling strength and evaluate $\lambda[g]$ for the lowest order sym-
metry functions $g(\boldsymbol{k})$ of the tetragonal crystal. Specif-
ically, we compute the coupling strength for s-wave
$[g(\boldsymbol{k}) = 1]$, $s_{\pm}$ $[g(\boldsymbol{k}) \propto \cos(k_x) + \cos(k_y)]$, $d_{x^2-y^2}$ $[g(\boldsymbol{k}) \propto$
$\cos(k_x) - \cos(k_y)]$ and d_{xy} $[g(\boldsymbol{k}) \propto \sin(k_x)\sin(k_y)]$ sym-
metries. Fig. 3(a) shows our results for the various cou-
pling strengths as function of the selenium height. In the
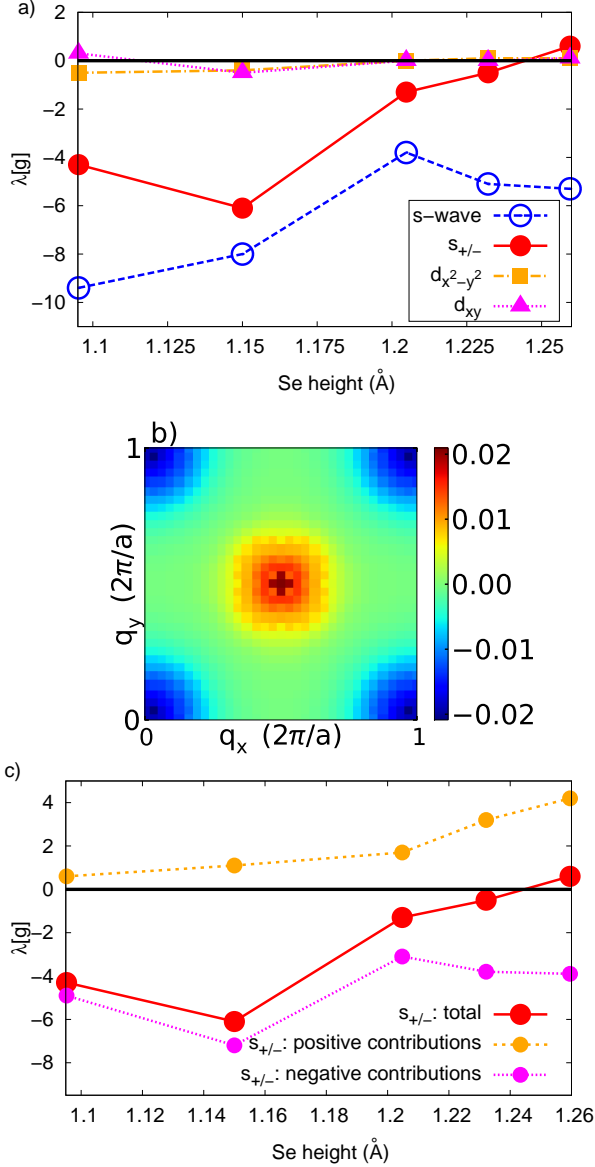singlet channel, the spin-fluctuation mediated interaction

FIG. 3. (a) Electron-spin fluctuation coupling constant for various gap symmetries as function of the selenium height. Positive coupling constants indicate that the system may undergo a transition to a superconducting phase at sufficiently low temperatures. (b) Contributions to the total electron-spin fluctuation coupling constant for an $s_\pm$-symmetric gap from the $\boldsymbol{q}$-points in the Brillouin zone at a selenium height of 1.20 Å. (c) Positive and negative contributions to $\lambda[s_\pm]$ as function of the selenium height.

between electrons is *repulsive* and therefore the coupling strength for s-wave symmetry is always negative. The vicinity to a magnetic phase transition gives rise to an *almost singular* spin-fluctuation mediated interaction and large, negative coupling constants $\lambda_s \ll -1$. This repulsion arising from magnetic interactions is much larger than the repulsion from the screened Coulomb interactions in standard metals, where $\lambda_s = -\mu \approx -0.2$ [34].

Note that this repulsion must also be overcome by other pairing mechanisms, such as phonons.

Figure 3(a) shows that d-wave coupling constants are quite small, but can be positive and thus give rise to superconductivity at sufficiently low temperatures. Most interestingly, we observe that the $s_\pm$ coupling constant is large and negative for small selenium heights, but quickly increases and eventually becomes positive near the transition to a stripe phase reaching values of approximately unity. We expect that this value of the $s_\pm$ coupling constant is reduced by the other contributions to the effective interaction (such as the bare Coulomb interaction and the charge fluctuation-mediated interaction) only by a *small amount*, as these contributions are much more isotropic and thus produce small values when integrated against the anisotropic $g(\boldsymbol{k})$ in Eq. (2).

Figure. 3(b) shows the contributions to the total $s_\pm$ coupling strength from all $\boldsymbol{q}$-points in the Brillouin zone at a selenium height of 1.20 Å. We observe significant cancellations between positive and negative contributions. Fig. 3(c) shows the total positive and total negative contributions to the $s_\pm$ coupling strength as function of the selenium height. While the negative contributions remain relatively constant, the positive contributions increase rapidly as stripe phase spin fluctuations become enhanced. These spin fluctuations scatter electrons from the hole pockets at the Γ-point to the electron pockets near the M-point. In the $s_\pm$ scenario, the superconducting order paramater switches sign between the Γ- and the M-point and therefore such scattering events are favorable for the emergence of superconductivity. On the other hand, spin fluctuations with wave vectors in the vicinity of the Γ-point scatter Cooper pairs only from electron pockets to other electron pockets or from hole pockets to other hole pockets. Their contribution to the $s_\pm$ coupling constant is *negative*. Suppression of such fluctuations (for example, by application of pressure) could provide a path towards higher transition temperatures [4, 12].

Finally, we discuss corrections to the presented first-principles framework that arise from the lack of self-consistency in our calculations. Specifically, the non-interacting spin susceptibility should be computed from a quasiparticle band structure including renormalization effects arising from many-electron interactions instead of the mean-field DFT-LDA band structure. While such renormalization effects are reproduced by "beyond-DFT" approaches, such as GW theory [35] or dynamical mean-field theory [25], good agreement with experimental ARPES measurements can be obtained by simply dividing all DFT-LDA band energies by a factor α, i.e. $\xi_{n\boldsymbol{k}} = \epsilon_{n\boldsymbol{k}} - \epsilon_F \rightarrow \xi'_{n\boldsymbol{k}} = \xi_{n\boldsymbol{k}}/\alpha$. Based on a recent ARPES experiment on FeSe multilayers [29], we use $\alpha = 3$, see Fig. 1(b). This rescaling of the band energies renormalizes the non-interacting susceptibility according to $\chi'_0 = \alpha\chi_0$ and also the density of states at the Fermi

level $D'_F = \alpha D_F$.

In addition to the electronic band structure, also the Stoner parameter is renormalized in the vicinity of a magnetic phase transition where spin fluctuations are significant [36, 37]. To include this effect, we also rescale the Stoner parameter according to $I'_{xc} = I_{xc}/\beta$. We use $\beta = 4.17$ which reproduces the experimental magnetic moment and critical doping strength in LaOFeAs [38] (note that $\beta = \alpha \times s$ with $s = 1.39$ being the rescaling parameter for I_{xc} neglecting band structure renormalization effects).

We now compute the coupling constant for the $s_\pm$-symmetric gap in FeSe at the experimental selenium height employing a renormalized band structure and Stoner parameter. This yields $\lambda[s_\pm] = 0.30$. Including the effect of charge fluctuations results in a slightly reduced value of $\lambda[s_\pm] = 0.28$. A crude estimate of the superconducting transition temperature $T_c = \omega_{SF}e^{-1/\lambda[s_\pm]}$ (where we use $\omega_{SF} = 15$ meV, a typical energy scale for spin fluctuations in iron-based compounds[9]) yields $T_c = 5$ Kelvin consistent with the experimental transition temperature of 8 Kelvin in FeSe [3, 4] (note that allowing $g(\boldsymbol{k})$ to be a *sum* of symmetry functions further increases λ [18] and our calculation therefore provides a *lower bound* for the transition temperature). In the current work, we have not considered the effect of phonons which was recently shown to be significant in the FeSe monolayer [39]. Future work is needed to investigate the interplay of spin fluctuations and phonons in iron-based superconductors.

Note added: After submission of the present manuscript, a similar calculation on spin fluctuation-mediated superconductivity in iron selenide was published [40].

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