

Supplementary Material to “Separate tuning of nematicity and spin fluctuations to unravel the origin of superconductivity in FeSe”

Supplementary Note 1. Superconducting critical temperature in the strong coupling approximation.

The superconducting critical temperature T_c in the framework of the Eliashberg theory is given by the solution of the system of the linearized Eliashberg equations:^{1,2}

$$Z_{\alpha n} = 1 + T_c \sum_{\omega'_{n'}, \beta} \lambda_{\alpha\beta}^z(n - n') \frac{\text{sign}(\omega_{n'})}{\omega_n}, \quad (1)$$

$$Z_{\alpha n} \Delta_{\alpha n} = T_c \sum_{\omega'_{n'}, \beta} \lambda_{\alpha\beta}^\phi(n - n') \frac{\Delta_{\beta n'}}{\omega_{n'}}. \quad (2)$$

where $\omega_n = \pi T_c(2n + 1)$ are the Matsubara frequencies, $\Delta_{\alpha n}$ are the gap functions, $Z_{\alpha n}$ are the Z functions, and α, β are the band indices. The coupling functions

$$\lambda_{\alpha\beta}^{\tilde{\phi},z}(n - n') = 2\lambda_{\alpha\beta}^{\tilde{\phi},z} \int_0^\infty d\Omega \frac{\Omega B(\Omega)}{[(\omega_n - \omega_{n'})^2 + \Omega^2]} \quad (3)$$

are expressed through the normalized bosonic spectral function $B(\Omega)$. For the sake of simplicity we neglect the intraband interaction ($\lambda_{11}^{\phi,z} = \lambda_{22}^{\phi,z} = 0$). The interband coupling constants $\lambda_{\alpha\beta}^{\tilde{\phi}}$ are chosen to be negative (repulsive) due to the prevailing spin-fluctuation mechanism of the electron-electron interaction.^{3,4} The matrix elements $\lambda_{\alpha\beta}^z$ are positive. For simplicity we use the approximation $\lambda_{\alpha\beta}^z = |\lambda_{\alpha\beta}^{\tilde{\phi}}| \equiv |\lambda_{\alpha\beta}|$ and neglects the $\mathbf{k}$ -space anisotropy in the gap functions $\Delta_{\alpha n}$.

a. Spin-fluctuation model. A phenomenological model of spin fluctuation spectra based on NMR data was proposed for nearly antiferromagnetic metals by Millis et al.⁵ [Millis-Monien-Pines (MMP) model]. They argued that the low energy dynamical spin susceptibility may be represented in the following form:

$$\text{Im}\chi(\mathbf{q}, \omega) = \frac{\chi_0 \Gamma_{\text{sf}}}{\pi \omega_{\text{sf}}} \frac{(\omega/\omega_{\text{sf}})}{(1 + \xi^2 |\mathbf{q} - \mathbf{Q}|^2)^2 + (\omega/\omega_{\text{sf}})^2} \times \Theta(\Omega_c - |\omega|), \quad (4)$$

where $\mathbf{Q}$ is the nesting vector, Ω_c is the energy cut-off and

$$\omega_{\text{sf}} = \Gamma_{\text{sf}} \frac{a^2}{\pi^2 \xi^2},$$

where ξ is the correlation length, characterizing the the proximity to the antiferromagnetic instability. At the transition point it diverges $\xi^{-1} = 0$. The static spin susceptibility is

denoted as χ_0 , a is the lattice constant, and Γ_{sf} is the frequency scale characterizing the spin fluctuations. Within this model one gets the spin lattice relaxation rate in the following form at low temperatures (see also ref. 6.):

$$\Gamma = \frac{1}{T_1 T} = \gamma_g^2 \lim_{\omega \rightarrow 0} \sum_{\mathbf{q}} \frac{M^2 \chi''(\mathbf{q}, \omega)}{\omega} \sim \frac{\chi_0}{\xi^{-2}}. \quad (5)$$

b. Boson spectral function. The main input into the Eliashberg equations Eqs.(1) and (2) is the spectral function of the intermediate bosons $B(\mathbf{q}, \omega)$:

$$\alpha^2 F(\omega) = \frac{\sum_{\mathbf{k}, \mathbf{k}'} \delta(\epsilon_{\mathbf{k}}) \delta(\epsilon_{\mathbf{k}'}) |A(\mathbf{k}, \mathbf{k}')|^2 B(\mathbf{k} - \mathbf{k}', \omega)}{\sum_{\mathbf{k}} \delta(\epsilon_{\mathbf{k}})} \quad (6)$$

where $A(\mathbf{k}, \mathbf{k}')$ is the matrix element for the scattering an electron in Bloch state $\mathbf{k}$ to $\mathbf{k}'$ and

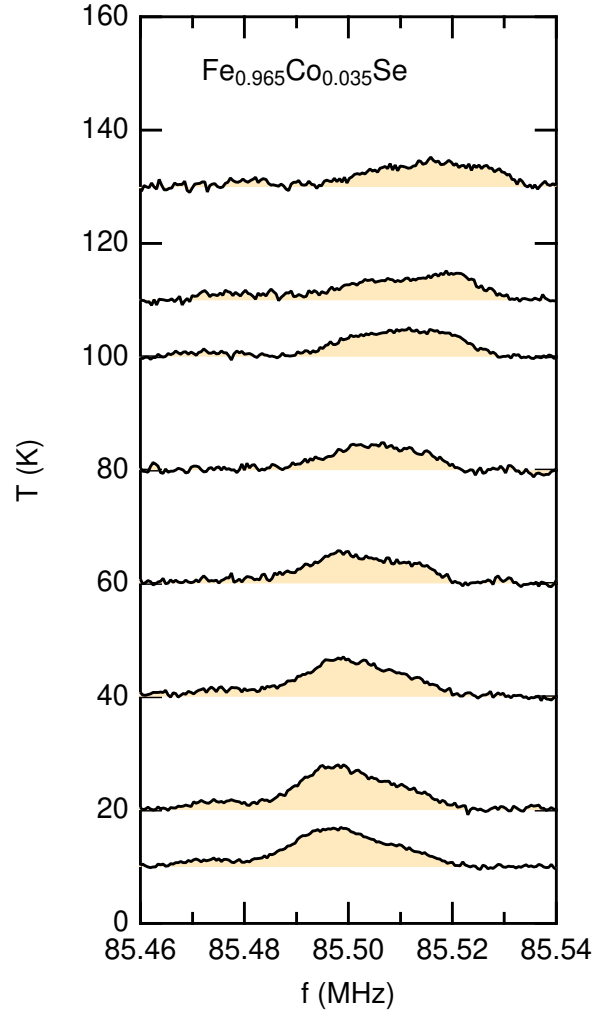
$$B(\mathbf{q}, \Omega) = -\frac{1}{\pi} N(0) \text{Im} \chi(\mathbf{q}, \Omega) \quad (7)$$

is the spectral function of the spin fluctuations normalized by the density of states at the Fermi level $N(0)$. As it was pointed out in refs. 7–9, that use of the MMP spectrum in the Eliashberg equation leads to overestimation of T_c and superconducting gaps due to a long tail at high frequencies $\propto 1/\omega$. Here, we adopt the phenomenological approach proposed by Popovich et al.⁹ The low energy part of the bosonic function is given by Eq. (4) and the function decays fast after a characteristic cut-off energy. The cut-off energy is determined by the band structure and in the leading approximation can be taken independent on the distance to the quantum critical point.

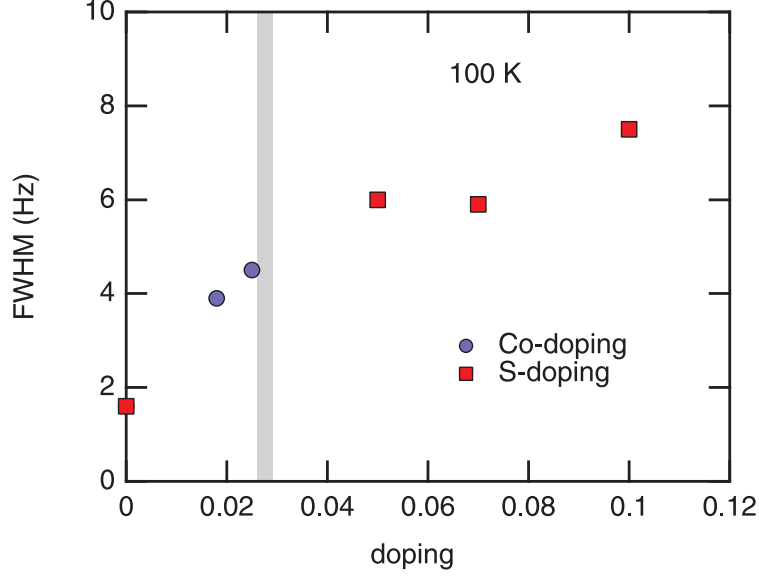
Since we consider the leading term, we neglect the momentum dependence of $A = g = \text{const.}$ Performing the momentum integration in Eq. (6) we get for small ω :

$$\alpha^2 F(\omega) \approx \frac{g^2 N(0)}{4\pi^2 (\xi/a)} \frac{\omega}{\omega_{\text{sf}}} \sim g^2 N(0) \frac{\xi}{a} \frac{\omega}{\Gamma_{\text{sf}}}. \quad (8)$$

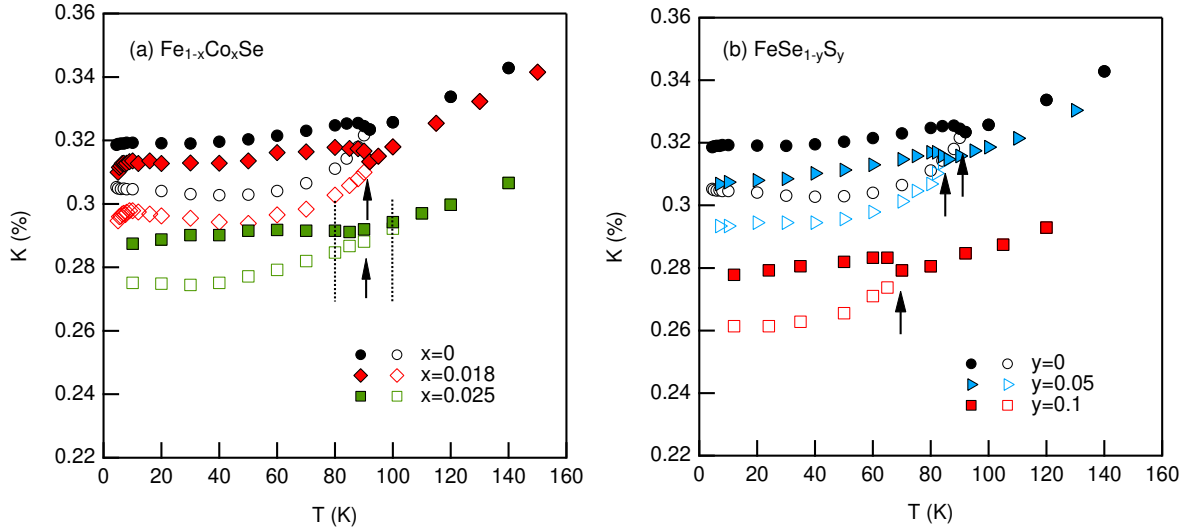
It determines that $\lambda \sim \xi \sim \sqrt{\Gamma}$. In Fig. 3 of the main text we show the experimental and calculated T_c vs. λ and $\sqrt{\Gamma}$ correspondingly. The fit gives the scale of the coupling constants.



Supplementary Figure 1. Temperature dependence of ^{77}Se spectrum at 3.6%Co doping. The full-width-at-half-maximum (FWHM) significantly increased at this doping. Since the splitting of the line due to nematicity is smaller than the FWHM, T_{nem} cannot be identified.



Supplementary Figure 2. Doping dependence of ^{77}Se full-width-at-half-maximum (FWHM) at 100 K. The gray line denotes the existence of a critical Co doping.



Supplementary Figure 3. ^{77}Se Knight shift data in (a) $\text{Fe}_{1-x}\text{Co}_x\text{Se}$ (b) $\text{FeSe}_{1-y}\text{S}_y$. The arrow indicates the nematic transition temperature T_{nem} . For $x = 0.025$ in (a), T_{nem} cannot be identified accurately, but is in the range 80-100 K. It should be noted that the Knight shift splitting $\Delta\mathcal{K}(T)$ is sensitive to an inevitable slight misalignment of the sample with respect to H . Therefore, the small variation of $\Delta\mathcal{K}(T)$ with doping should be treated as an experimental error.

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