Extension of susceptibilities, screened exchange and spin-fluctuation integrals into ultrasoft pseudopotentials

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In the Eliashberg theory or density functional theory for superconductors (SCDFT) [1], where the electron-phonon, screened Coulomb, spin-fluctuation mediated interactions are included non-empilically, we need to compute the folowing product of two Kohn-Sham orbitals

$$\rho_{n\mathbf{k}n'\mathbf{k}'}(\mathbf{r}) = \varphi^*_{n\mathbf{k}}(\mathbf{r})\varphi_{n'\mathbf{k}'}(\mathbf{r}), \tag{1}$$

where n (n') and $\mathbf{k}$ ($\mathbf{k}'$) are the band index and Bloch wavenumber, respectively. To perform this product together with the ultrasoft pseudopotentials (USPP) or projector augumented waves (PAW) that are widely used because of the good accuracy and reasonable numerical costs [2], we need a correction term for the norm-conservation. Such a correction is originally proposed for the calculation of the susceptibility [3] as

$$\Delta\rho_{n\mathbf{k}n'\mathbf{k}'}(\mathbf{r}) = \sum_{\tau i i'} \langle\varphi_{n\mathbf{k}}|\beta_{\tau i'}\rangle\langle\beta_{\tau i}|\varphi_{n'\mathbf{k}'}\rangle Q_{\tau i i'}(\mathbf{r}), \tag{2}$$

where $\beta_{\tau i}$ is the projector dual to the atomic pseudo orbital $\psi^{PS}_{\tau i}$ at atom τ and orbital i, and the augmentation charge $Q_{\tau i i'}(\mathbf{r})$ is computed from the pseudo (PS) and all-electron (AE) atomic orbitals as follows:

$$Q_{\tau i i'}(\mathbf{r}) \equiv \psi^{AE*}_{\tau i}(\mathbf{r})\psi^{AE}_{\tau i'}(\mathbf{r}) - \psi^{PS*}_{\tau i}(\mathbf{r})\psi^{PS}_{\tau i'}(\mathbf{r}). \tag{3}$$

To utilise this correction to the calculation of the spin-fluctuation mediated interaction,

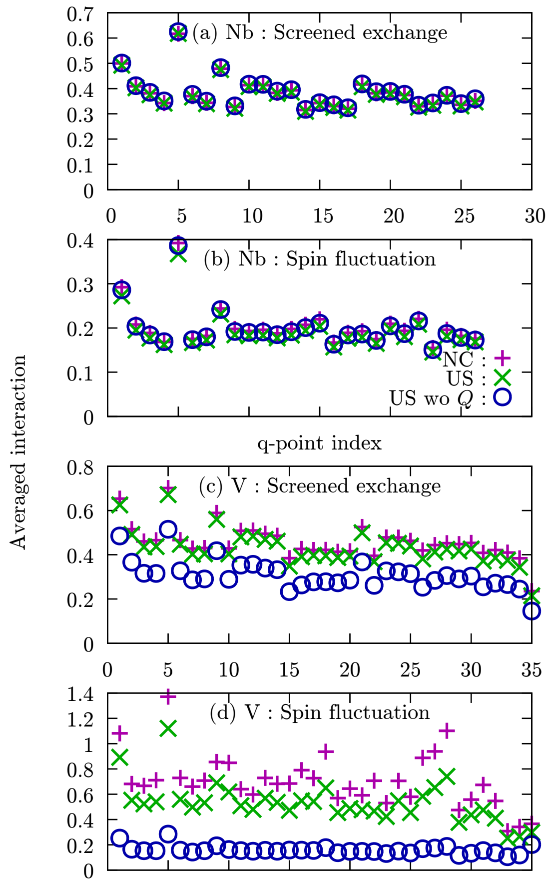

Figure 1: Spin-fluctuation mediated- (a, c) and screened exchange- (b, d) interactions averaged over the Fermi surface of Nb (a, b) and V (c, d). "+", "×", and "∘" idicate results with norm-conserving (NC) ultrasoft (US) with and without augumentation charge $Q(\mathbf{r})$, respectivelly.

we implemented this formalism into our first-principles program package Superconducting-Toolkit [4] which is based on SCDFT.

Figure 1 shows the screened exchange and spin-fluctuation interactions averaged over the Fermi surface of Nb and V. In Nb, the effect of the augmentation charges Q is small because the USPP for this atom is almost norm-conserving. At the same time, V has a significant contribution from Q due to the nodeless 3d orbitals. In Fig. 1(d), we can see a deviation between the result by NC and US pseudopotential even if we include the correction. This deviation may be because the exchange-correlation kernel included in the spin-fluctuation is sensitive to the charge density in the vicinity of atoms.

We also performed the benchmark of the calculation of T_c for 15 materials, namely Al, V, Ta, In, Zn, Cd, Sn, ZrN, TaC, MgB_2, H_3S (at a pressure of 200 GPa), CaC_6, YNi_2B_2C, and V_3Si. Figure 2 shows the experimental, and calculated T_c; we performed four kind of calculations by changing superconducting density functional, namely the conventional plasmon-assisted [5], Sanna's Eliashberg-mimic (Sanna) [6], Sanna+Coulomb renormalization ($Z_{\mathbf{C}}$) [7], and Sanna + $Z_{\mathbf{C}}$ + Spin-fluctuation [9, 8] functional.

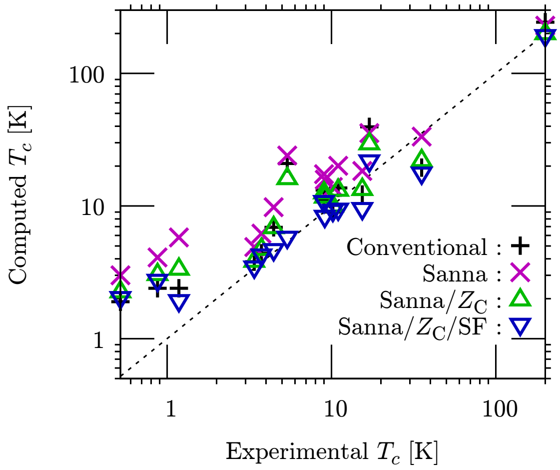

Figure 2: Computed- and experimental T_c. "+", "×", "△", and "▽" indicate result with conventional plasmon-assisted [5], Sanna's Eliashberg-mimic (Sanna) [6], Sanna+Coulomb renormalization ($Z_{\mathbf{C}}$) [7], and Sanna + $Z_{\mathbf{C}}$ + Spin-fluctuation [9, 8] functional, respectively.

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