Ab initio study on magnetism suppression, anharmonicity, rattling mode, and superconductivity in Sc_6MTe_2 (M = Fe, Co, Ni)

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(Received 8 May 2024; revised 7 August 2024; accepted 19 August 2024; published 4 September 2024)

We perform a systematic *ab initio* study on phonon-mediated superconductivity in the transition-metal-based superconductors Sc_6MTe_2 (M = Fe, Co, Ni). Firstly, our charge analysis reveals significant electron transfer from Sc to M because of the substantial difference in the electronegativity, filling the 3d orbitals of M and suppressing magnetic instability. Secondly, we show that Sc_6FeTe_2 exhibits strong lattice anharmonicity. Moreover, for M = Fe and Co, we find low-frequency soft phonon bands of M, which can be interpreted as "rattling phonons" in the framework formed by Sc. While not observed in the case of M = Ni, the rattling phonons give rise to a prominent peak or plateau in the Eliashberg spectral function and enhance the pairing instability. By reproducing the experimental trend of superconductors by strategically combining nonsuperconducting and magnetic transition-metal elements.

DOI: 10.1103/PhysRevB.110.104505

I. INTRODUCTION

Identifying common characteristics among known superconductors bears significance for designing novel superconductors [1]. Despite the abundance of identified superconductors, platforms remain scarce, where superconductivity manifests across a series of materials. This is particularly true in compounds involving d electrons [2–12]. Recently, seven materials from a family of *d*-element-rich compounds Sc₆MTe₂ have been identified as possessing bulk superconductivity [12]. These materials have the Zr_6CoAl_2 -type structure, with M encompassing 3d, 4d, and 5d elements. Notably, the compounds with M = Fe, Co, and Ni show a significant material dependence in their superconducting transition temperatures (T_c 's) of 4.7 K, 3.6 K, and 2.7 K, respectively. Interestingly, the pairing mechanism is yet to be fully understood: In particular, it is unclear why superconductivity occurs in compounds containing magnetic elements and why T_c is highest with M = Fe and lowest with M = Ni.

To address these questions, we perform a systematic *ab initio* study on the electronic structures, phonon properties, and the electron-phonon coupling (EPC) for Sc₆*M*Te₂ (M = Fe, Co, Ni). We first perform the charge analysis based on the electronic structure, which shows an electron transfer from Sc to *M*. As a result, the 3*d* orbitals of *M* are almost fully occupied, which suppresses the local magnetic moments on *M*. By reproducing the chemical trend of T_c , we conclude that the target materials are phonon-mediated superconductors.

In addition, we show that the higher T_c in Sc₆FeTe₂ and Sc₆CoTe₂ can be ascribed to the strong EPC from the low-frequency phonon modes dominated by the displacements of M.

The crystal structure of Sc_6MTe_2 is characterized by the Sc cages encapsulating the isolated *M* atoms, indicating the system to be a host(Sc)-guest(*M*) compound. Due to such structural arrangement and the emergence of low-frequency phonons with single-atom vibrations, we would interpret such phonons as "rattling phonons". This phenomenon is often observed in materials where large freedom of motion is given to guest atoms within periodic cages with examples of skutterudites [13,14], pyrochlores [15–17], clathrates [13], brownmillerites [18], or quadruple perovskites [19].

Nonparabolic behavior of the potential energy surface due to atomic displacements, namely the lattice anharmonicity, can occur for a rattling vibrational behavior [20]. The effect of anharmonicity on superconductivity has been extensively studied [21–27], and the combining effect of rattling modes and anharmonicity on superconductivity was discussed for the β -pyrochlore oxides AOs_2O_6 [28] and intermetallic cage compounds MV_2AI_{20} [29]. Since rattling modes provide significant atomic displacement at the flat potential energy surface, the attractive interaction for the Cooper pair can be large and thus enhance T_c [28,29]. Although there are some cases where the rattling modes have only a moderate impact on superconductivity [30], we find that taking advantage of the strong EPC of the soft rattling phonons is an effective strategy to enhance superconductivity in Sc₆MTe₂.

The rest of this paper is organized as follows. Section II describes the theoretical methods, the crystalline structure of

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 Sc_6MTe_2 , and the computational details. Section III presents the calculated electronic structures, including the orbital projected density of states (DOS), band structures, and the Fermi surface. Then, we show the phonon band structures, atomprojected phonon DOS, and the corresponding Eliashberg spectral function to quantify the EPC strength. Further discussions on the suppression of magnetism and the emerging superconductivity in Sc_6MTe_2 are provided in Sec. IV. Finally, the conclusions drawn from this study are summarized in Sec. V.

II. THEORY AND COMPUTATIONAL METHODS

A. Electron-phonon coupling

Based on density functional theory (DFT), we can use density functional perturbation theory (DFPT) to calculate the EPC matrix element [31-33],

$$g_{mn,\nu}(\mathbf{k},\mathbf{q}) = \sum_{\kappa\alpha} \sqrt{\frac{\hbar}{2\omega_{\nu\mathbf{q}}}} \frac{e_{\kappa\alpha\nu\mathbf{q}}}{\sqrt{M_{\kappa}}} \times \left\langle \psi_{m\mathbf{k}+\mathbf{q}} \middle| \sum_{p} \frac{\partial V}{\partial u_{\kappa\alpha\mathbf{R}_{p}}} \frac{e^{i\mathbf{q}\cdot\mathbf{R}_{p}}}{\sqrt{N_{p}}} \middle| \psi_{n\mathbf{k}} \right\rangle, \quad (1)$$

where $\omega_{\nu \mathbf{q}}$ is the phonon frequency of mode ν and crystal momentum \mathbf{q} , $e_{\kappa\alpha\nu\mathbf{q}}$ is the polarization vector of site κ in the Cartesian direction α , M_{κ} is the mass of the atom at site κ . $\psi_{n\mathbf{k}}$ is the Kohn-Sham orbital with the band index *n* and wave vector \mathbf{k} . *V* is the self-consistent potential, and its first-order derivative with respect to $u_{\kappa\alpha\mathbf{R}_p}$ is calculated using DFPT [31–33]. Note that $u_{\kappa\alpha\mathbf{R}_p}$ is the atomic displacement in the lattice position \mathbf{R}_p of the *p*th unit cell. N_p is the number of primitive cells in the Born-von Kármán supercell.

The EPC strength of a specific phonon mode ν and phonon momentum **q** can then be obtained via the Brillouin zone (BZ) integration [31,32,34],

$$\lambda_{\nu \mathbf{q}} = \frac{1}{N(\epsilon_{\mathrm{F}})\omega_{\nu \mathbf{q}}} \sum_{nm} \int_{\mathrm{BZ}} \frac{d\mathbf{k}}{\Omega_{\mathrm{BZ}}} |g_{mn,\nu}(\mathbf{k},\mathbf{q})|^{2} \\ \times \ \delta(\epsilon_{n\mathbf{k}} - \epsilon_{\mathrm{F}})\delta(\epsilon_{m\mathbf{k}+\mathbf{q}} - \epsilon_{\mathrm{F}}), \tag{2}$$

where $N(\epsilon_{\rm F})$ is the electronic DOS at the Fermi level $\epsilon_{\rm F}$ and $\epsilon_{n\mathbf{k}}$ is the state energy at momentum \mathbf{k} . $\Omega_{\rm BZ}$ is the volume of the BZ. We then introduce the isotropic Eliashberg spectral function [31,32,34]

$$\alpha^{2}F(\omega) = \frac{1}{2} \sum_{\nu} \int_{BZ} \frac{d\mathbf{q}}{\Omega_{BZ}} \omega_{\nu \mathbf{q}} \lambda_{\nu \mathbf{q}} \delta(\omega - \omega_{\nu \mathbf{q}}), \qquad (3)$$

from which the EPC constant λ is obtained as

$$\lambda = 2 \int \frac{\alpha^2 F(\omega)}{\omega} d\omega. \tag{4}$$

Through the celebrated McMillan-Allen-Dynes (MAD) formula [35–37], we can evaluate the superconducting temperature T_c by

$$T_{c} = \frac{\omega_{\log}}{1.2} \exp\left[-\frac{1.04(1+\lambda)}{\lambda - \mu^{*}(1+0.62\lambda)}\right],$$
 (5)

in which the prefactor includes the logarithmic average phonon frequency of the spectrum ω_{\log} where

$$\omega_{\log} = \exp\left[\frac{2}{\lambda} \int \frac{d\omega}{\omega} \alpha^2 F(\omega) \log\left(\omega\right)\right]$$
(6)

and μ^* is the pseudo-Coulomb parameter introduced to account for the screened electron-electron interaction.

B. Self-consistent phonon theory

The self-consistent phonon (SCPH) theory [38–40] is a mean-field theory of the lattice anharmonicity, in which we solve a self-consistent equation in terms of the renormalized effective frequencies $\Omega_{\nu q}$,

$$\Omega_{\nu \mathbf{q}}^{2} = \omega_{\nu \mathbf{q}}^{2} + \frac{1}{2} \sum_{\nu_{1} \mathbf{q}_{1}} F_{\mathbf{q} \mathbf{q}_{1}, \nu \nu_{1}} \frac{\hbar [1 + 2n_{B}(\Omega_{\nu_{1} \mathbf{q}_{1}})]}{2\Omega_{\nu_{1} \mathbf{q}_{1}}}, \quad (7)$$

where $n_B(\omega)$ is the Bose-Einstein distribution function. $F_{\mathbf{qq}_1,\nu\nu_1} = \Phi(\mathbf{q}\nu; -\mathbf{q}\nu; \mathbf{q}_1\nu_1; -\mathbf{q}_1\nu_1)$ is the fourth-order interatomic force constant (IFC), which is defined in reciprocal representation as [39]

$$\Phi(\mathbf{q}_{1}\nu_{1};\cdots;\mathbf{q}_{4}\nu_{4}) = N_{p}^{-1} \sum_{\kappa\alpha} (M_{\kappa_{1}}\cdots M_{\kappa_{4}})^{-\frac{1}{2}} e_{\kappa_{1}\alpha_{1}\nu\mathbf{q}_{1}}\cdots e_{\kappa_{4}\alpha_{4}\nu\mathbf{q}_{4}}$$
$$\times \sum_{p_{2},\cdots,p_{4}} \Phi_{\alpha_{1}\cdots\alpha_{4}}(0\kappa_{1};\cdots;p_{4}\kappa_{4}) e^{i(\mathbf{q}_{2}\cdot\mathbf{R}_{p_{2}}+\cdots+\mathbf{q}_{4}\cdot\mathbf{R}_{p_{4}})},$$
(8)

where $\Phi_{\alpha_1 \cdots \alpha_4}(p_1 \kappa_1; \cdots; p_4 \kappa_4)$ is the fourth-order derivative, or the quartic term of the Taylor expansion coefficient, of the Born-Oppenheimer potential energy surface U with respect to displacement $u_{\kappa\alpha\mathbf{R}_n}$,

$$\Phi_{\alpha_1\cdots\alpha_4}(p_1\kappa_1;\cdots;p_4\kappa_4) = \left.\frac{\partial^4 U}{\partial u_{\kappa_1\alpha_1\mathbf{R}_1}\cdots\partial u_{\kappa_4\alpha_4\mathbf{R}_4}}\right|_{u=0}.$$
 (9)

Equation (7) can be derived using the Dyson equation with a frequency-independent self-energy [39] or using the variational principle of the free energy [40]. Note that the polarization vectors are assumed not to change by the anharmonic effect in this paper.

As we discuss later in Sec. III B, we obtain imaginary frequency ($\omega_{\nu q}^2 < 0$) phonons in the harmonic phonon calculation for Sc_6FeTe_2 . This is inconsistent with the experiment because the imaginary modes indicate the instability of the identified crystal structure. Thus, we use the SCPH theory to obtain a stable phonon dispersion with real frequencies. Also, Eqs. (1) and (3) indicate that imaginary frequency phonons would give an ill-defined EPC matrix element and Eliashberg spectral function, which should be handled carefully for phonon-mediated superconductors [22-25,41,42]. While one may artificially set the imaginary phonon frequencies to zero or their absolute values or introduce a sizable electronic smearing parameter [43,44], in this paper, we employ a treatment that uses the SCPH theory. Namely, we obtain the SCPH frequencies $\Omega_{\nu q}$ and replace the harmonic frequencies $\omega_{\nu q}$ [45,46]. Note that using the SCPH theory is not entirely valid because firstly, it considers the finite-temperature renormalization of the phonon frequencies that originate from the



FIG. 1. Hexagonal crystal structure with space group $P\bar{6}2m$ (No. 189) and Brillouin zone of Sc_6MTe_2 (M = Fe, Co, Ni). (a) Side view and (b) top view of the crystal structure where one unit cell contains one formula unit. (c) The hexagonal Brillouin zone.

anharmonic effect. Secondly, the potential energy surface V in Eq. (1) is not renormalized. Nevertheless, we expect that this treatment is helpful because the SCPH frequencies reflect the overall curvature of the low-energy region of the potential energy surface, while the harmonic calculation is determined only by the curvature at the reference structure. The validity is also supported by the fact that the calculated T_c s agree fairly well with the experimental ones. Note that we carefully choose the temperature to estimate $n_B(\Omega_{\mu q})$ in the SCPH theory (T_{SCPH}) and check that the T_{SCPH} dependence of the calculated T_c is relatively small, which is further discussed in Appendix B.

C. Crystal structure and computational details

The crystal structure of Sc_6MTe_2 (M = Fe, Co, Ni) is hexagonal with the $P\bar{6}2m$ space group (No. 189), which contains one formula unit (f.u.) per unit cell. The Sc_6MTe_2 compounds are an ordered variant of the Fe_2P structure, belonging to a large family characterized by the Zr_6CoAl_2 -type structure [47,48]. The transition-metal elements M are surrounded by the Sc atoms forming tricapped trigonal prisms [47]. Such a local Sc cage with M bounded inside stacks up along the c direction, which forms a linear chain of M. The structures are shown in Fig. 1. In the current study, we perform full structural relaxation with the results summarized in Table I.

The structural relaxation and the calculations of electronic and phonon properties are conducted using DFT, and the corresponding electron-phonon properties are calculated using DFPT [33], as implemented in the QUANTUM ESPRESSO

TABLE I. Theoretical lattice constants (a, c, c/a) of Sc₆*M*Te₂ compared with the experimental data [47]. The number in the parenthesis denotes the percentage deviation of the theoretical lattice constants from the experiments.

Sc ₆ MTe ₂	<i>a</i> (Å)	<i>c</i> (Å)	c/a
Fe	7.703 (+0.306%)	3.788 (-1.272%)	0.492
	7.6795 ^a	3.8368 ^a	0.4996
Co	7.732 (+0.446%)	3.743 (-1.123%)	0.484
	7.6977 ^a	3.7855 ^a	0.4918
Ni	7.768 (+0.576%)	3.729 (-0.972%)	0.480
	7.7235 ^a	3.7656 ^a	0.4876

^aExperimental data taken from Ref. [47].

(QE) code [49]. All the calculations are performed using the scalar-relativistic projector-augmented wave (PAW) [50] pseudopotential with the generalized gradient approximation (GGA) in the form of Perdew-Burke-Ernzerhof (PBE) [51] from PSLibrary [52]. A plane-wave cutoff value of 952 eV and a Γ -centered 12 × 12 × 24 Monkhorst-Pack [53] *k* mesh was used to describe the electronic structure. For the DOS calculation, we apply the tetrahedron method [54]. The valence orbital set is $3s^23p^64s^23d^{1,6,7,8}$ for Sc, Fe, Co, Ni, and $5s^25p^4$ for Te. The phonon calculations are performed with a *q* grid of $2 \times 2 \times 2$. Also, the crystal structure is visualized by VESTA [55], and the Fermi surface is plotted by FermiSurfer [56].

We perform the SCPH calculation for Sc₆FeTe₂. The IFCs are generated using Vienna Ab Initio Simulation Package (VASP) [57,58] and ALAMODE package [39,40,59], where we use a $2 \times 2 \times 2$ supercell containing 72 atoms. The VASP calculations for the force data are performed using DFT with the PAW method [50] and the GGA in the form of PBE [51,60]. A plane wave cut-off energy of 520 eV is used. For the BZ integration, a Γ -centered k mesh of $4 \times 4 \times 4$ is used for the supercell. The harmonic IFCs are calculated by the frozen phonon method with atomic displacements of 0.01 Å. The anharmonic IFCs are computed using the compressive sensing method, which enables efficient calculation of IFCs from a small number of supercell calculations [61]. To generate the displacement-force data from which we extract the IFCs, we generate configurations with random atomic displacements using *ab initio* molecular dynamics (AIMD) simulation. To sample the low-energy region of the potential surface, we first perform the AIMD in VASP at 300 K for 5000 steps with a time step of 1 fs. We discard the first 1000 steps due to their dependence on the initial setting of the AIMD simulation and extract 80 snapshots from the rest of the trajectory. We further displace the atoms randomly by 0.04 Å to eliminate the correlation between the snapshots. DFT calculations of VASP compute the atomic force for each supercell, and up to the fourth-order IFCs are calculated to fit the displacement-force relation [40]. Note that the pseudopotentials used in both QE and VASP calculations are selected based on the best agreement between the results of the phonon dispersion under the harmonic approximation, and the results are given in Fig. 9 in Appendix A. Based on such a foundation, we can replace the dynamical matrix data in DFPT, which is used in QE, with the one from SCPH, which is generated



FIG. 2. Total and orbital-decomposed density of states (DOS) of (a) Sc_6FeTe_2 , (b) Sc_6CoTe_2 , and (c) Sc_6NiTe_2 . The Fermi level is set to zero, as denoted by the vertical dashed line.

by VASP plus ALAMODE. Note that there is no source code modification needed for QE in this procedure, and the technical details for this part will be presented in Notes 1 within the Supplemental Material (SM) [62].

Lastly, T_c is evaluated using the MAD formula [Eq. (5)], as explained in the previous section. The pseudo-Coulomb parameter μ^* is set to 0.1 to account for the screened Coulomb interaction.

III. RESULTS

A. Electronic structures

We examine the electronic structures of Sc_6MTe_2 . Figure 2 shows the total and orbital-decomposed DOS for Sc_6MTe_2 . We notice that there are sharp peaks in the DOS around 1.5 eV to 2.2 eV below the Fermi level, which is dominated by the *M*-3*d* orbitals. This suggests that the *M*-3*d* orbitals are localized in space because the narrow width of the peaks and the weak hybridization with other orbitals indicates weak hopping to the Sc atoms around them. We see that these peaks gradually get narrower and deeper below the Fermi level as we go from Fe toward Ni. This can be explained by the difference in the electronegativity of Fe (1.83), Co (1.88), and Ni (1.91) [63] as the electrons will be more localized and more strongly bound in atoms with larger electronegativity. Next, we discuss the electronic structures of Sc_6MTe_2 near the Fermi level. In Fig. 2, we observe hybridized DOS between Sc1, Sc2, and *M* within the energy range from –1 to 1 eV. Notably, there is a peak structure in the DOS at the Fermi level across all the materials, indicating a large DOS at the Fermi level. Large DOS at the Fermi level is beneficial for phonon-mediated superconductivity [64].

In Figs. 3(a)-3(c), we show the detailed orbital-projected band structures and the corresponding Fermi surface in Figs. 3(d)-3(f). In Figs. 3(a)-3(c), all the band structures possess a hole pocket around the Γ point. This hole pocket at Γ primarily comprises hybridized M-3 d_{xy,x^2-y^2} and Sc2-3 d_{z^2} orbitals. From Figs. 3(d)-3(f), we see that the hole pocket forms a cylindrical Fermi surface around Γ , and the size of this cylindrical patch decreases from M = Fe to Ni, as clearly depicted in the Γ -M-K section in Figs. 3(d)-3(f). We do not observe any Fermi surface pockets around the M and K points. On the other hand, the projected band structures along A-L-H change through electron filling. For example, from Figs. 3(a) and 3(b), along A-L-H, we can see that for M = Fe, the M-3 $d_{xz,yz}$ and Sc2-3 $d_{xz,yz}$ bands are not occupied while they are occupied in the case of M = Co. Moreover, the Fermi surface on the A-L-H plane is more complicated than the hole pocket surrounding the Γ point. This originates from the less dispersive bands with more crossings across the Fermi level compared to that of the Γ -M-K plane, as we can see from Figs. 3(a)-3(c). The same observation can also be made by looking at the A-L-H section of the Fermi surface at Figs. 3(d)-3(f).

Lastly, we discuss the magnetism suppression in Sc₆MTe₂ (M = Fe, Co, and Ni). In the experiments [12,47], no sign of stable ferromagnetism is reported, which is quite non-trivial because they include well-known magnetic elements M = Fe, Co, and Ni. Notably, possibilities of paramagnetism, especially in the case of Sc₆FeTe₂ has been suggested [12]. In fact, no magnetic states can be stabilized for M = Co, Ni in our spin-polarized self-consistent DFT calculations. Also for M = Fe, it is not certain that there is a metastable ferromagnetic state [65].

To further investigate the underlying reason, we perform the Bader charge analysis, which is a useful charge counting analysis done by segregating atoms along the minimum in the electron density [66]. The Bader charges, which are the counted valence electrons, and the corresponding nearest oxidation state are given in Table II. Interestingly, every Sc atom donates roughly one electron to the neighboring M and Te, transferring six electrons. The pair of Te atoms would share three electrons, and M would take the remaining three electrons. Note that the oxidation state for Ni in Sc₆NiTe₂ is in between a 2- and 3- state while Co and Fe are 3-, indicating a stronger hybridization of the outer electrons of Ni. Nevertheless, the 3d electron shell of Co and Ni is fully occupied with Fe having one vacancy left. This explains why the potential magnetism brought by Co and Ni does not show up in the Sc_6MTe_2 systems while Sc_6FeTe_2 shows stronger magnetic interaction in the family but no stable long-range ferromagnetism. Note that this electron transfer occurs due to the difference in electronegativity where Sc, Fe, Co, Ni, and Te are of value 1.36, 1.83, 1.88, 1.91, and 2.10, respectively [63]. Crucially, this electron reallocation prevents the potentially



FIG. 3. Electronic band structures around the Fermi level (0 eV) of (a) Sc_6FeTe_2 , (b) Sc_6CoTe_2 , and (c) Sc_6NiTe_2 color coded with the *d*-orbital projections. Note that the projected density of states for the *d* orbitals not plotted contribute at least three times less to the Fermi level than those that are plotted. The Fermi surface, color coded by band indices, and two sections along the Γ -M-K and A-L-H plane for Sc_6MTe_2 where M = Fe (d), Co (e), and Ni (f).

detrimental effect on superconductivity caused by the partially filled 3*d* shell. Moreover, since the Fe 3*d* shell is not fully filled, we expect a more unstable nonmagnetic state than the rest, resonating with the absence of stable ferromagnetic results in DFT self-consistent calculation for M = Co and Ni. The magnetic instability of the Sc₆MTe₂ system, especially for M = Fe, is an interesting topic but beyond the scope of this paper.

TABLE II. Results of Bader charge analysis for Sc_6MTe_2 (M = Fe, Co, Ni). The closest ideal ionic state, as well as its oxidation state, is also given. The valence orbitals used in the present calculations are $3s^23p^64s^23d^{1,6,7,8}$ for Sc, Fe, Co, Ni, and $5s^25p^4$ for Te. The Bader charge of each neutral atom is thus 11, 16, 17, 18, 6 for Sc, Fe, Co, Ni, and Te, respectively.

	Bader charge	Oxidation state
Compound	(e ⁻)	(e ⁻)
Sc ₆ FeTe ₂		
Sc1	9.95	$10 (Sc^{1+})$
Sc2	9.88	$10 (Sc^{1+})$
Fe	18.97	$19 (Fe^{3-})$
Те	7.74	$8 (Te^{2-})$
Sc ₆ CoTe ₂		
Sc1	9.94	$10 (Sc^{1+})$
Sc2	9.91	$10 (Sc^{1+})$
Co	19.92	$20 (Co^{3-})$
Те	7.76	$8 (Te^{2-})$
Sc ₆ NiTe ₂		
Sc1	9.96	$10 (Sc^{1+})$
Sc2	9.94	$10 (Sc^{1+})$
Ni	20.69	20 (Ni ²⁻) or 21 (Ni ³⁻)
Те	7.78	8 (Te ^{2–})

B. Phonon properties

Now, we discuss the phonon properties of the Sc_6MTe_2 systems. The calculated phonon bands are given in Figs. 4(a), 4(c), and 4(e). As shown in Fig. 4(a), under the harmonic approximation, imaginary frequency phonon bands exist along the Γ -M-K plane of Sc₆FeTe₂. We mark the imaginary frequency phonon bands at the M point as "a" in Fig. 4(a). The displacement amplitude along the polarization vector of such a band is displayed in Fig. 5(a). We see that the origin of such an imaginary frequency mode is the in-plane vibration of the Fe atoms. On the other hand, Sc₆CoTe₂ and Sc₆NiTe₂ do not have imaginary frequencies under the harmonic approximation, as shown in Figs. 4(c) and 4(e). This suggests that Fe atoms in Sc₆FeTe₂ will show larger atomic displacements in the in-plane direction and manifest stronger anharmonicity compared to M =Co and Ni cases. Note that anharmonicity refers to the deviation of the potential energy surface from the parabolic behavior near the reference structure.

However, the dynamical instability (i.e., the imaginary phonon frequency) is incorrectly given by the harmonic approximation because no structural phase transitions have been observed in the experiment [12]. This is because the potential energy surface of Sc₆FeTe₂ is very flat, as shown in Fig. S1 within the SM [62]. Therefore, we use the SCPH theory to obtain a positive dispersion in a physically reasonable way, as we discussed in Sec. II B. The obtained SCPH dispersion with $T_{\text{SCPH}} = 20 \text{ K}$ is displayed with magenta lines in Fig. 4(a). Comparing the harmonic and SCPH bands, the higher-frequency portion of the phonon bands remains unchanged, while the imaginary phonon modes are stabilized, resulting in dispersive phonon bands in the low-frequency region. Importantly, we notice that the flat degenerate phonon modes around 75 cm⁻¹ on the Γ point are also restored for Sc₆FeTe₂



FIG. 4. Phonon band structures, total and *M*-projected phonon density of states (PhDOS) of (a)(b) Sc_6FeTe_2 , (c)(d) Sc_6CoTe_2 , and (e)(f) Sc_6NiTe_2 . The magenta curves in (a) and (b) are the anharmonicity-renormalized phonon bands. The displacement pattern of the phonon bands indicated by markers "a", "b", and "c" are given in Fig. 5. Also, the red arrows point to the localized phonon bands arising from the Fe and Co atoms.

as we see such a common feature for M = Co and Ni as well, implying that anharmonicity is crucial for this mode. In Notes 2 within the SM [62], we demonstrate the increasing degree of anharmonicity by the increasing flatness of the potential



FIG. 5. Displacement pattern of (a) the imaginary frequency phonon mode at the M point of Sc_6FeTe_2 [marker "a" in Fig. 4(a)], (b) the doubly degenerated phonon bands number 4 and 5 around 75 cm⁻¹ at the Γ point of Sc_6FeTe_2 [marker "b" in Fig. 4(a)], and (c) Sc_6CoTe_2 [marker "c" in Fig. 4(c)]. All displacement modes shown here are in-plane.

energy surface from M = Ni to Fe by calculating the total energy with respect to the displacement of these modes.

In Figs. 4(a), 4(c), and 4(e), we can see common features across M = Fe, Co, and Ni. All spectral widths are roughly $250 \,\mathrm{cm}^{-1}$. Also, the phonon band structures of $\mathrm{Sc}_6 M \mathrm{Te}_2$ are generally dispersed in the low-frequency region and flatter in higher-frequency regions. For Sc₆NiTe₂, the phonon bands are more dispersive than the others. The similarity of the phonon band structures can also be well observed by the total phonon density of states (PhDOS), as given in Figs. 4(b), 4(d), and 4(f). We can see that the peak positions and magnitudes of the total PhDOS are qualitatively similar for M = Fe, Co,and Ni. Interestingly, the difference lies in the M atomprojected PhDOS. In comparing Figs. 4(b), 4(d), and 4(f), as indicated by the red arrows, around 75 cm^{-1} , only the renormalized phonon bands of Sc₆FeTe₂ and the harmonic phonon bands of Sc₆CoTe₂ have a PhDOS peak. This indicates the emergence of localized M phonon modes for only Sc₆FeTe₂ and Sc₆CoTe₂. Thus, we take the phonon bands around 75 cm⁻¹ at the Γ point as representatives to investigate their vibration patterns. They are doubly degenerated phonon bands with index numbers 4 and 5 for both materials, and we mark them as "b" in Fig. 4(a) and "c" in Fig. 4(c). In Figs. 5(b) and 5(c), we see that these modes originate from in-plane vibrations of M. On the contrary, the Ni PhDOS of Sc₆NiTe₂ is fairly uniform throughout the spectrum, as shown in Fig. 4(f).

Significantly, we realize that such a difference in the distribution of the *M*-projected PhDOS is related to the EPC strength. Figure 6 displays the phonon bands of Sc₆*M*Te₂ with indication of the relative magnitude of the EPC strength $\lambda_{\nu q}$ [Eq. (2)]. Note that a hot spot of $\lambda_{\nu q}$ indicates a strong pairing interaction between electrons at **k** and **k**+**q** mediated by the ν phonon mode. For M = Fe and Co, Sc₆*M*Te₂ has large $\lambda_{\nu q}$ in phonon modes with frequencies of ~25–40 cm⁻¹ around M and K. For M = Fe, as is obvious from Fig. 4(a), the anharmonic effect is significant in this segment of the phonon bands. The second hot spot lies around 75 cm⁻¹. This segment highlights the difference in the *M* atom-projected



FIG. 6. Phonon band structure color coded with the relative size of electron-phonon coupling strength $\lambda_{\nu q}$ for Sc₆*M*Te₂. Orange lines represent the phonon bands. Note that the thickness of the line is normalized to $\lambda_{\nu q}$ of each material.

PhDOS as observed through Figs. 4(b), 4(d), and 4(f). Also, these hot spots indicate the importance of applying the SCPH to estimate the phonon frequency for this energy range. Moreover, based on the results of harmonic phonons in Sc₆CoTe₂, the similarity in the EPC distribution between M = Fe and Co supports the sufficiency of our method for M = Fe as we capture the main physics in the system. That is, if we look at Fig. 6(c), we would see that $\lambda_{\nu q}$ of Sc₆NiTe₂ is uniformly distributed with respect to phonon momentum and frequency. From Fig. 6, we reveal the significance of these low-frequency phonons in understanding the chemical trend of the EPC and consequently T_c for Sc₆MTe₂ (M = Fe, Co, Ni). In Sec. IV A, we will further discuss that we could interpret these low-frequency phonons that generate strong EPC as "rattling" phonons.

C. Eliashberg function and superconductivity

In Figs. 7(b), 7(d), and 7(f), the calculated Eliashberg spectral function $[\alpha^2 F(\omega)]$ is depicted alongside the PhDOS. $\alpha^2 F(\omega)$ provides insight into the EPC strength (λ). Typically, peaks in $\alpha^2 F(\omega)$ align with those in PhDOS. However, a significant $\alpha^2 F(\omega)$ contribution to λ emerges in the 60 to 80 cm^{-1} range for Sc₆FeTe₂ and Sc₆CoTe₂, which is absent in Sc₆NiTe₂. This distinction can be attributed to the presence or absence of the M-dominated low-frequency phonons discussed in the previous subsection. For Sc₆FeTe₂, $\alpha^2 F(\omega)$ manifests a wide plateau in 20 to $50 \,\mathrm{cm}^{-1}$ frequency range, yielding significant contributions to λ . At higher-frequency regions above $80 \,\mathrm{cm}^{-1}$, the EPC contribution coincides with the peaks of PhDOS, initially from the Te phonon bands followed by the Sc phonon bands. Figure 7 illustrates, firstly, the critical role of the M atom in the EPC of Sc_6MTe_2 . Second, it shows a correlation between low-frequency vibrations of Mand enhanced EPC. Finally, it demonstrates the dominance of the anharmonicity-driven EPC for M = Fe.

By integrating $\alpha^2 F(\omega)/\omega$ over frequency, we obtain the EPC constant λ [Eq. (4)]. As shown in Table III, the resultant λ values are 1.78, 0.75, and 0.46 for M = Fe, Co, and Ni, respectively. Utilizing the calculated λ , we predict T_c

values using the MAD formula [Eq. (5)] [35–37]. The pseudo-Coulomb interaction (μ^*) is treated as an empirical parameter and is set to 0.1. The predicted T_c values for Sc₆MTe₂ are 9.4 K, 6.2 K, and 1.6 K for M = Fe, Co, and Ni, respectively. The calculated values of T_c align with the observed chemical trends [12], with M = Fe ranking the highest and M = Ni the lowest. Also, the predicted values of T_c are comparable to the measured values. Thus, we have revealed that the Sc₆MTe₂ (M = Fe, Co, Ni) are phonon-mediated superconductors. Note that the calculated λ and T_c of Sc₆FeTe₂ is little affected by T_{SCPH} as shown in Table III. While we use the calculation of $T_{\text{SCPH}} = 20$ K as the representative, the discussion remains robustly for a wide range of T_{SCPH} . See Appendix B for further discussions on T_{SCPH} of Sc₆FeTe₂ and comments on the effect of SCPH toward Sc₆CoTe₂.

Finally, we discuss the possible origins of the deviation between the calculated and experimental T_c . First of all, we notice that the T_c of M = Fe and Co is overestimated but not for M = Ni. We have shown in previous sections that the main difference between these two cases is the existence of soft phonon modes in the former (Figs. 4 and 6). In the present case, although λ of the soft phonon modes could be

TABLE III. Calculated electron-phonon coupling constant λ , logarithmic average phonon frequency ω_{\log} , total $[N(\epsilon_{\rm F})]$ and *M*projected $[N_M(\epsilon_{\rm F})]$ density of states at the Fermi level, and the superconducting transition temperature (T_c) of Sc₆*M*Te₂ (*M* = Fe, Co, Ni). The last column includes the temperature $(T_{\rm SCPH})$ used in the SCPH theory. The smearing parameter is 0.02 eV, and the pseudo-Coulomb parameter μ^* is 0.1.

System M	λ	ω_{\log} (K)	$N(\epsilon_{\rm F})$ (1/eV/f.u.)	$N_M(\epsilon_{\rm F})$ (1/eV/M)	<i>T_c</i> (Exp. ^a) (K)	T _{SCPH} (K)
Fe	1.78	72	9.04	1.16	9.4 (4.7)	20
Fe	1.67	79	9.04	1.16	9.9 (4.7)	50
Fe	1.46	95	9.04	1.16	10.5 (4.7)	100
Co	0.75	152	16.30	2.03	6.2 (3.5)	
Ni	0.46	187	12.46	0.79	1.6 (2.7)	

^aExperimental data taken from Ref. [12].



FIG. 7. Atom-decomposed phonon density of states (phonon DOS), as well as the Eliashberg spectral functions $\alpha^2 F(\omega)$ and the frequencyresolved electron-phonon coupling strength $\lambda(\omega)$ of (a)(b) Sc₆FeTe₂, (c)(d)Sc₆CoTe₂, and (e)(f) Sc₆NiTe₂. Note that $\lambda(\omega) = 2 \int_0^{\omega} \frac{\alpha^2 F(\omega')}{\omega'} d\omega'$ and thus the electron-phonon coupling constant λ is the value obtained after integrating over the whole phonon spectrum.

overestimated, the main argument of soft modes to enhance T_c still holds.

Next, one other possibility lies in the influence of spin fluctuation coming from the *d*-electrons in the system. Kawamura et al. [67] carried out systematic ab initio calculations, including both EPC and spin fluctuation based on density functional theory for superconductors (SCDFT) for all the elemental metals. Their SCDFT calculations showed that for some elemental metals, including spin fluctuation could significantly reduce the phonon-mediated T_c . In particular, they reported that elemental Sc metal has an EPC constant λ of 0.398 and a T_c of 2.7 K when the spin fluctuation is not considered. However, when the spin fluctuation is taken into account, the superconductivity disappears, agreeing with the experiments. We notice that the DOS at the Fermi level in hexagonal Sc is rather large, being 2.01 states/eV/Sc atom [67], and thus, spin fluctuation would be quite significant. Figure 2 shows that the projected DOS for Sc at the Fermi level in Sc₆MTe₂ is also rather large, being 1.65, 2.06, and 1.38 states/eV/Sc atom, respectively, for M = Fe, Co, and Ni. Since two-thirds of atoms in Sc_6MTe_2 (M = Fe, Co, Ni) are Sc, the spin fluctuation in Sc₆MTe₂ may affect the predicted phonon-mediated T_c in Sc₆MTe₂. In particular, this perhaps explains why the calculated T_c values in M = Fe and Co are higher than the experimental values (Table III).

Also, such deviation of T_c could arise from the isotropic gap function that the MAD formula assumes. Given the complicated Fermi surface of Sc₆*M*Te₂, it is an interesting future problem of how much the predicted T_c is affected if we go beyond the MAD formula. In conclusion, our findings suggest that superconductivity in Sc_6MTe_2 (M = Fe, Co, Ni) is phonon mediated, where low-frequency M vibrations and anharmonic phonon modes play a crucial role. Also, our study suggests that by utilizing the electronegativity difference, we can manipulate the electron fillings of the d orbitals to make the combination of a nonsuperconducting elemental metal, Sc in the present case, and magnetic element become superconducting. In general, this proposition could serve as a potential explanation or direction of investigation for Sc-included systems, including one recent report on the discovery of superconductivity of ScPdGe and ScPdSi [68].

IV. DISCUSSIONS

In this section, we further extend the discussions toward the rattling phonons and the magnetic suppression of Sc_6MnTe_2 .

A. Rattling phonons

From our results (Figs. 4, 6, and 7), we show that low-frequency phonons from M are the key factor in understanding the trend in T_c for Sc₆MTe₂. Also, note that in Fig. 5, we learn that these phonons are mainly single-atom vibrations. These results prompt us to interpret this key factor to be "rattling phonons", where atoms "rattle" alone in the system, producing low-frequency vibrational modes and large EPC [28,29,69].

Here, we discuss the phonon dispersion of a system with rattling phonons, which in our case are the M = Fe and Co



FIG. 8. Rattling phonon modes of Sc₆FeTe₂ and Sc₆CoTe₂. Phonon band structures in low-frequency regions are color coded based on their relative magnitude of atomic displacement for the rattler (*M* atom) and the cage (Sc atoms) for M = Fe (a), Co (b), and Ni (c). Insets of each are the phonon band structures in low-frequency regions color coded with the relative size of the electron-phonon coupling strength $\lambda_{\nu q}$. (d) Schematic of the avoid crossing between the rattling mode and longitudinal acoustic mode as well as the displacement pattern around the crossing for M = Fe and Co.

phonon modes. Christensen et al. [70] and Baggioli et al. [71] showed by inelastic neutron scattering experiments and analytical model calculations that rattling phonons appear as flat phonon modes dominated by guest atom vibrations along the energy region of the acoustic phonon modes. Also, with increasing interaction between the guest rattler and the host cage, the avoid crossings between the two modes would appear [71]. In Figs. 8(a)-8(c), we present the lowfrequency segment of the phonon band structures. Notably, from Figs. 8(a) and 8(b), in both Sc₆FeTe₂ and Sc₆CoTe₂, the flat phonon band around 75 cm⁻¹ predominantly reflects the M vibration mode. It persists over a wide range along the high symmetry line, particularly the A-L-H plane in the energy region of the acoustic modes, displaying the existence of rattling modes. On the contrary, from Fig. 8(c), we notice the absence of such a feature in Sc₆NiTe₂. This is consistent with the PhDOS shown in Figs. 4. Although structuralwise, M atoms are all located inside the Sc cage, but from the absence of rattling modes for Sc₆NiTe₂, we can infer that the Ni atoms are likely to bond more strongly to the surroundings than Fe or Co. In Appendix C, we show the results of the on-site force constant and the responsive force due to *M* displacement to support this argument.

As circled in green in Figs. 8(a) and 8(b), we can discern a gap between the rattling phonons and the longitudinal acoustic mode along the Γ -M and Γ -A line, displaying the dynamical coupling between rattlers and the cage [70,71]. Such a feature

is also seen in Sc₆NiTe₂ but is expected given that the Sc and Ni vibrations in this energy region are highly entangled. Figure 8(d) shows the schematic of the avoided crossing and the displacement pattern of M = Fe and Co around the gap. We notice the displacement of the Sc cage to dominate along the acoustic mode before and after the gap. Moreover, we notice the dominance of the rattler displacements from Fe and Co to persist on the rattling phonon band.

Finally, we comment on the relation between EPC and rattling phonons. The insets of Fig. 8(a)-8(c) are the low-frequency segment of Fig. 6, we clearly see that the EPC of Sc₆FeTe₂ and Sc₆CoTe₂ is highly concentrated in the region of the rattling phonons. This suggests that Sc₆MTe₂ is a case to demonstrate the EPC driven by these localized rattling modes that can be further tuned by the degree of lattice anharmonicity.

B. Magnetic suppression of Sc₆MnTe₂

Now let us inquire further into the absence of the magnetic moment on M in Sc₆MTe₂ [12]. As elaborated in the last part of Sec. III A, our Bader charge analysis reveals an electron transfer from Sc to M because Sc displays the lowest electronegativity among the constituent atoms. This electron transfer provides enough electrons to saturate the 3d orbitals of Fe, Co, and Ni, thus suppressing the formation of the magnetic moment on M. In the experiments by Shinoda *et al.*



FIG. 9. Phonon band structures calculated under the harmonic approximation using (a) DFPT in the QE package and (b) frozen phonon method in the VASP plus ALAMODE package.

[12], superconductivity is not observed for M = Mn. Since Mn has a much lower electronegativity of 1.55 compared with that of Fe, Co, and Ni [63], the large electron transfer from Sc to M mentioned above would be weaker; thus, Sc₆MnTe₂ may have a magnetic ground state. This suggests that superconductivity in Sc₆MnTe₂ is perhaps suppressed by magnetic pair breaking [72] due to the magnetic moments on the Mn atoms. To verify this hypothesis, we further perform self-consistent spin-polarized electronic structure calculations for Sc₆MnTe₂. Indeed, we find that Sc_6MnTe_2 has a stable ferromagnetic state with a local magnetic moment of $1.4\mu_B$ on Mn. The total energy of the ferromagnetic state is 25 meV/f.u. lower than the nonmagnetic state. Moreover, we perform the Bader charge analysis toward Sc₆MnTe₂ with the final results giving us 1.98, 10.01, and 7.71 e^- for the Sc, Mn, and Te atoms, respectively. The Bader charges correspond to the nearest oxidation state of 2 e⁻ (Sc¹⁺), 10 e⁻ (Mn³⁻ = Ni valence state $4s^23d^8$), and $8e^-$ (Te²⁻).

V. CONCLUSIONS

In conclusion, we have carried out a systematic ab initio computational study on the magnetic, electronic, phonon, and superconducting properties of newly reported transitionmetal-based superconductors Sc_6MTe_2 (M = Fe, Co, Ni) [12] by using theoretical methods of DFT, DFPT, and SCPH. First, our DFT calculations reveal that Sc_6MTe_2 (M =Fe, Co, Ni) have nonmagnetic metallic ground states with multiple Fermi surface pockets. The Bader charge analysis indicates a significant electron transfer from Sc to M(where M = Fe, Co, Ni) due to the substantial electronegativity difference. This charge transfer fills the M 3d orbitals, thus suppressing the magnetism from the M atoms. Second, combining DFPT and SCPH, we obtain the renormalized phonon bands for the anharmonic Sc₆FeTe₂ as we observe flat phonon bands near 75 cm^{-1} and dispersive low-frequency phonon bands emerging after considering the anharmonic effect. Interestingly, these two emerging features are rattling



FIG. 10. (a) Phonon band structures and (b) Eliashberg spectral functions $\alpha^2 F$ of Sc₆FeTe₂ calculated using different T_{SCPH} values in the SCPH treatment of anharmonicity.

phonon bands holding large EPC at low-frequency regions in Sc_6MTe_2 when M = Fe and Co. Quantitatively, they also give rise to prominent peaks in the calculated Eliashberg spectral function. These rattling mode-enhanced EPCs lead to a rather high superconducting transition temperature (T_c) of $\sim 10 \,\mathrm{K}$ for M = Fe. Our DFPT calculations also predict $\text{Sc}_6 M \text{Te}_2$ (M = Co and Ni) to be phonon-mediated superconductors with lower T_c values of 6.2 K and 1.6 K, respectively. The chemical trend of the predicted T_c for Sc₆MTe₂ from M = Fe to Co and Ni agrees well with the experimental observation [12]. Importantly, we attribute such a chemical trend to the phonon bands where we observe the progression of increasing rattling behavior from M = Ni to Co and then the increasing anharmonicity from M = Co to Fe. In short, our *ab initio* study not only shows that the superconductivity observed in 3d transition-metal compounds Sc_6MTe_2 (M = Fe, Co, and Ni) is phonon mediated but also suggests a way to design transition-metal-based superconductors by combining nonsuperconducting and magnetic metal elements with different electronegativities.

ACKNOWLEDGMENTS

M.-C.J. and G.-Y.G. acknowledge the support from the Ministry of Science and Technology and the National Center for Theoretical Sciences (NCTS) of the R.O.C. M.-C.J. was supported by the IPA Program, RIKEN. R.M. was supported by Grant-in-Aid for JSPS Fellows (No. 22KJ1028). R.A. was supported by JSPS KAKENHI Grants No. JP22H00110 and No. JP24H00190.

APPENDIX A: COMPARISON OF THE HARMONIC PHONONS USING DFPT AND THE FROZEN PHONON METHOD

In Sec. II, we mentioned replacing the harmonic phonon bands with the renormalized phonon bands from the SCPH theory to evaluate the EPC matrix element. Equation (7) shows that the fourth-order IFCs are corrected on the TABLE IV. Results of the on-site force constant $\Phi_{xx,yy}(0, 0)$ and the Hellmann-Feynman (HF) force on *M* atom with a 0.02 Å displacement for Sc₆*M*Te₂ (*M* = Fe, Co, Ni).

System	$\Phi_{xx,yy}(0,0)$ (Ry/Bohr ²)	HF force (Ry/Bohr)
Sc ₆ FeTe ₂	0.056635	-0.001588
Sc ₆ CoTe ₂	0.063625	-0.002491
Sc ₆ NiTe ₂	0.103335	-0.002530

harmonic phonon frequency. Thus, to merge the information and calculate the EPC matrix element [Eq. (1)], we need to double-check the agreement among the harmonic phonons. Figure 9 displays the dispersion relations calculated using DFPT and the frozen phonon method. Clearly, Fig. 9 shows that the two sets of dispersion relations agree in the frequency range above 80 cm^{-1} . Also, the imaginary frequency modes on the Γ -M-K plane appear in both cases. This is thus an appropriate foundation for further calculating the EPC using the renormalized phonon frequencies and eigenvectors from the SCPH theory.

APPENDIX B: EFFECT OF T_{SCPH} ON PHONON AND ELECTRON-PHONON PROPERTIES

In this Appendix, we examine the effect of T_{SCPH} on the phonon properties and EPC. As mentioned in Sec. II B, a T_{SCPH} of 20 K was used in the Bose-Einstein distribution of the SCPH equation [Eq. (7)]. We choose the temperature at which the phonon bands are stable but softer than Sc₆CoTe₂, reflecting the stronger anharmonicity in Sc₆FeTe₂. We consider that the treatment is valid because no structural phase transition has been experimentally observed for Sc₆FeTe₂. Furthermore, as Table III shows, T_c remains almost the same when T_{SCPH} increases. This indicates that the choice of $T_{\text{SCPH}} = 20$ K is appropriate.

Nevertheless, we do notice that λ drops and ω_{\log} rises with increasing T_{SCPH} (Table III). In Fig. 10, we show the phonon bands and $\alpha^2 F(\omega)$ for three different T_{SCPH} values. From Fig. 10(a), we notice that the phonon bands in the frequency region above 100 cm⁻¹ are not affected by the T_{SCPH} value. However, for the flat phonon band near 75 cm⁻¹ and the dispersive phonon bands below, the frequencies of the phonon bands, especially near the M point, do increase as T_{SCPH} rises. This results in the visible upward shift of the bottom of the Eliashberg spectral function as T_{SCPH} increases [see Fig. 10(b)].

Importantly, we note from Eqs. (3), (2), and (6) that $\lambda \propto \omega^{-1}$ and $\omega_{\log} \propto \log(\omega)$. Since T_{SCPH} only affects the phonon frequency, we can see why λ drops as ω_{\log} rises due to the increase of T_{SCPH} . Furthermore, due to the competing scaling relations of λ and ω_{\log} with respect to ω , T_c of Sc₆FeTe₂ evaluated using Eq. (5) does not significantly depend on T_{SCPH} .

Last, we comment on the possible effect of SCPH on Sc_6CoTe_2 . From Fig. 6, we can spot the phonon softening around the M point for Sc_6CoTe_2 , which is one of the key indicators that there exists a chemical trend in the phonon structures of Sc_6MTe_2 . However, in the case of Sc_6CoTe_2 , since its phonon can be captured by the harmonic approximation (Fig. 4), we would expect less hardening effect from SCPH. Also, at a temperature as low as T_c , the renormalization of the phonon band structures would be small. Moreover, given the similarity between M = Fe and Co, we would expect the T_c to rise as well for Sc_6CoTe_2 if we introduce T_{SCPH} to it, although the changes are expected to be small.

APPENDIX C: ON-SITE FORCE CONSTANT AND HELLMANN-FEYNMANN FORCE OF Sc_6MTe_2 (M = Fe, Co, Ni)

In this Appendix, we discuss the level of bonding between the *M* atoms and the rest by calculating the on-site force constant $\Phi_{xx,yy}^M(0,0)$ in the real-space representation for the *M* atom and the Hellmann-Feynman force on *M* atom with a 0.02 Å displacement along the *x* direction of Sc₆*M*Te₂ (*M* = Fe, Co, Ni). The on-site force constant shows the restoring force when the *M* atom is moved while others are fixed. $\Phi_{xx,yy}^M(0,0) = \frac{1}{2}(\Phi_{xx}^M(0,0) + \Phi_{yy}^M(0,0))$ and $\Phi_{\alpha\beta}^M(a,b) = \sum_{\kappa} \Phi_{\alpha\beta}(a\kappa,bM)$ where α,β are the Cartesian directions, κ is the atomic site index, *M* is the atomic site in interest, and *a*, *b* are the supercell indices. Note that a positive value indicates the stability of the *M* atom position and a negative value of $\Phi_{xx,yy}^M(0,0)$ indicates *M* atom to be off-center [73].

From Table IV, we notice that the Ni atom has the largest on-site force constant, which can also be observed by the largest force acting on Ni when it is displaced for 0.02 Å away from equilibrium. The same trend can be observed throughout M =Co and Fe. Therefore, the Ni atoms are more strongly bonded to the Sc cage, as also shown from the dispersive nature of phonon bands, as shown in Fig. 4.

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