Using first-principles calculations, we examine the transition temperature $T_c$ of superconductivity in sodium tungsten bronze (Na$_x$WO$_3$, where $x$ is equal to or less than unity). Although $T_c$ is relatively low ($< 3K$), it is interesting that its characteristic exponential dependence on $x$ has been experimentally observed at $0.2 \leq x \leq 0.4$. On the basis of the McMillan equation for $T_c$ including the effect of plasmons, we succeed in reproducing the absolute values of $T_c$ and its $x$ dependence. We also find that the plasmon effect is crucial for the estimation of $T_c$ as well as phonons. Since the calculated $T_c$ may not exceed $\sim 20 K$ even for $x \leq 0.1$, the superconductivity at a low $T_c$ can be interpreted by the usual phonon mechanism, including the plasmon effect. On the other hand, a high $T_c$ up to about 90 K is found on the surface of a Na$_x$WO$_3$ system at $x \sim 0.05$ by recent experiments, cannot be explained by our results. This discrepancy suggests that another mechanism is required to clarify the nature of the high-$T_c$ superconductivity of Na$_x$WO$_3$.

Sodium tungsten bronze (Na$_x$WO$_3$) and related materials [A$_x$WO$_3$(A=K, Rb, Cs,..)] have been studied for a long time as typical oxide superconductors, where the concentration $x$ is equal to or less than unity.\(^1\)\(^-\)\(^9\) The transition temperature $T_c$ of superconductivity has been known to increase with decreasing $x$ and to be up to about 3 K for Na$_x$WO$_3$ (\(\sim 7 K\)) and Cs$_x$WO$_3$ at $x \sim 0.2$. It is interesting that the $x$ dependence of $T_c$ is experimentally given by $T_c \approx A \exp(-Bx)$ for $0.2 \leq x \leq 0.4$ for Na$_x$WO$_3$,\(^1\)\(^1\) where $A$ and $B$ are constants. Since $T_c$ is relatively low, the origin of this superconductivity has been explained by the usual phonon mechanism.\(^1\)\(^0\)

However, recent experiments on the surface of a Na$_x$WO$_3$ system showed that high-temperature superconductivity (HTS) at about 90 K or higher is achieved at $x \sim 0.05$ for Na$_x$WO$_3$.\(^1\)\(^1\),\(^1\)\(^2\) Furthermore, another experiment demonstrated that H$_x$WO$_3$ also shows HTS at about 120 K.\(^1\)\(^3\)

The result of scanning tunneling microscopy\(^1\)\(^1\)\(^1\) indicates that the $I-V$ characteristic behavior is well fitted to the BCS theoretical curve. It seems to suggest that the symmetry of a Cooper pair may be described by the $x$-wave and it does not contradict the phonon-mediated superconductivity. These results are extremely surprising because the values of $T_c$ are comparable to those of cuprate superconductors. If the HTS of Na$_x$WO$_3$ and/or H$_x$WO$_3$ are concrete, it is very interesting to clarify whether the mechanism underlying this superconductivity is the conventional one or not.

In this study, we examine the electronic state of Na$_x$WO$_3$ by first-principles calculations using ‘Quantum ESPRESSO’, which is an integrated software of Open-Source computer codes for electronic-structure calculations.\(^1\)\(^4\) Assumming the phonon-mediated superconductivity, we estimate $T_c$ as a function of $x$ on the basis of the McMillan formulaton\(^1\)\(^5\)-\(^1\)\(^7\) including the effect of plasmons.\(^1\)\(^8\)-\(^2\)\(^2\) Since $T_c$ can be obtained with high accuracy by first-principles calculations, we may obtain a clue distinguishing whether the observed HTS can be explained by the conventional mechanism or not.

In Fig. 1, we show a schematic structure of the unit cell of Na$_x$WO$_3$ for $x = 1$. It has a cubic perovskite structure, where the length of one side of the cube is $a$. Inside the cubic unit cell, one tungsten atom and six oxygen atoms form an octahedron, which we will call octahedral WO$_6$. First, we calculate the $x$ dependence of $a$ using a supercell consisting of four unit cells. It contains zero, one, two, three, and four Na atoms. Here, we use an $8 \times 8 \times 8$ Monkhorst–Pack (MP) grid for electronic Brillouin zone integration. Using a structural relaxation method, we determine the volume of the supercell and estimate the value corresponding to $a$ as the cubic root of one quarter of its volume. It is experimentally known that in a system of Na$_x$WO$_3$, the unit cell size $a$ decreases with decreasing $x$.\(^2\)\(^3\) In Fig. 2(a), we show $a$ as a function of $x$. It indicates that $x$ dependence of $a$ is given by $a \approx 3.826 + 0.082x$ [Å]. The result well reproduces that obtained by experiments as $a = 3.7845 + 0.0820x$ [Å].\(^2\)\(^3\)

In Fig. 2(b), we also show the density of states (DOS) for the same supercells. For $x = 0$, the energy gap appears at the Fermi energy and the system becomes an insulator. On the other hand, the value of DOS at the Fermi energy is finite and the system is metallic for $x > 0$. We confirmed that the DOS of one Na atom is less than $10^{-3}$ [st/eV/spin/cell] near the Fermi energy, and it is completely negligible for $x \geq 0.24$. Furthermore, the energy dependences of DOS are very similar except for the position of the Fermi energy, as shown in Fig. 2(b). Therefore, the role of the Na atom is considered to only provide an electron into the conduction band of the sys-
term of WO$_3$. These results suggest that the rigid-band picture well stands, as shown by Raj et al.\textsuperscript{7} Therefore, to calculate in the case of an arbitrary $x$, we adopt the rigid-band approximation (RBA).\textsuperscript{25} This approximate method introduces a fictitious carrier in the target system by assuming a rigid-band. It changes only the Fermi energy of the system according to the carrier density. Although it is a simple approximation, it allows us to systematically analyze the electronic state of Na$_x$WO$_3$ for $x \leq 1$.\textsuperscript{22,25} Hereafter, we denote the result obtained by adopting RBA for the WO$_3$ system as WO$_3$\textsuperscript{RBA}$^-$ at an arbitrary $x$, whereas that for NaWO$_3$ as NaWO$_3$\textsuperscript{RBA}$^+$. Here, the notation of RBA$^-$ means the case of electron doping in WO$_3$, and RBA$^+$ means that of hole doping in NaWO$_3$.

![Fig. 2](image-url)

Fig. 2. (Color online) (a) Calculated equilibrium values of $a$ as a function of $x$, where $x$ is composition ratio of Na. (b) Density of states of states of Na$_x$WO$_3$ calculated for the supercell, where $x = 0$ (dotted-dashed line), 0.25 (dotted line), 0.5 (solid line), 0.75 (broken line), and 1.0 (solid line), where the values of DOS ($N(\varepsilon)$) are normalized by a single unit cell.

If the superconductivity of Na$_x$WO$_3$ is caused by phonon-mediated attraction, $T_c$ can be estimated by the following McMillan equation:

$$ T_c = \frac{\omega_{\text{log}}}{1.2} \exp\left(-\frac{1.04(1+\lambda)}{\lambda - \mu^*(1+0.624)}\right), \quad (1) $$

where $\omega_{\text{log}}$ is the logarithmic average frequency, which means a characteristic phonon frequency of the system, $\lambda$ is the electron-phonon coupling constant, and $\mu^*$ is the Coulomb pseudopotential, which is usually treated as a constant of about 0.1 for metals.\textsuperscript{17} However, $\mu^*$ is dependent on the electron density of the system, $n$ for $n \lesssim 10^{22}$ cm$^{-3}$ owing to the effect of plasmons.\textsuperscript{18,22} When $n$ decreases, $\mu^*$ also decreases and it even becomes negative at $n \sim 3 \times 10^{21}$ cm$^{-3}$.\textsuperscript{22,26} In the system of Na$_x$WO$_3$, $n$ is given by $\approx 1.7 \times 10^{22}$ cm$^{-3}$ for $x = 1$, and $\mu^*$ is expected to be considerably smaller than 0.1 in the region of $x \lesssim 0.5$.\textsuperscript{10,22}

In this work, we adopt a simple phenomenological relation,

$$ \mu^* = 0.05[\log_{10}(x) + \log_{10}(1 + 40x^2)], \quad (2) $$

which well reproduces the $x$ dependence of $\mu^*$ obtained in the previous work for $0.01 \leq x \leq 0.5$.	extsuperscript{22} This relation allows us to estimate $T_c$ including the plasmon effect through the McMillan equation (1). Using the first-principles calculations, we can estimate two parameters $\omega_{\text{log}}$ and $\lambda$ precisely. Combined with the above $\mu^*$, we can obtain the concrete value of $T_c$ by this formulation.

To calculate $x$ dependences of $\omega_{\text{log}}$ and $\lambda$ by RBA, we use two independent results, WO$_3$\textsuperscript{RBA}$^-$ and NaWO$_3$\textsuperscript{RBA}$^+$ in the case of $x \geq 0.4$. We assume that the electronic properties of Na$_x$WO$_3$ corresponding to an arbitrary $x$ are approximately obtained by prorating both results, such as $(1-x)$WO$_3$\textsuperscript{RBA}$^-$ $+ x$NaWO$_3$\textsuperscript{RBA}$^+$. Here, we use the $24 \times 24 \times 24$ MP grid for the electronic state and the $4 \times 4 \times 4$ grid for the phonon calculation.\textsuperscript{27} The unit cell size is chosen to be the same as that given in Fig.1 for both WO$_3$ and NaWO$_3$ unit cells. This approximation method may correspond to mean field approximation, which neglects the effect of randomness produced by Na atoms.

In Fig. 3(a), we show $\omega_{\text{log}}$ as a function of $x$. $\omega_{\text{log}}$ seems to be almost constant and is roughly given by 500 K. The difference in $\omega_{\text{log}}$ between WO$_3$\textsuperscript{RBA}$^-$ and NaWO$_3$\textsuperscript{RBA}$^+$ is small. Therefore, the prorating method may be a good approximation to consider the system of Na$_x$WO$_3$. In Fig. 3(a), the values of $\lambda$ and $-\mu^*$ as functions of $x$ are given. The result indicates that the difference in $\lambda$ between WO$_3$\textsuperscript{RBA}$^-$ and NaWO$_3$\textsuperscript{RBA}$^+$ is very small. $\lambda$ increases with decreasing $x$, which is roughly similar to the result obtained by Mascello et al.\textsuperscript{28} The figure also shows that $-\mu^*$ increases with decreasing $x$, and its rate of change is almost the same as that of $\lambda$. This
means that the plasmon effect described by $\mu'$ plays as important role in the superconductivity of Na$_x$WO$_3$ as the phonon-mediated attraction described by $\lambda$.

![Fig. 4](image)

**Fig. 4.** (Color online) (a) Phonon dispersions of WO$_3^{RBA}$ (solid lines) and Na$_x$WO$_3^{RBA}$ (dotted lines) for $x = 0.4$. (b) Phonon DOS, $F(\omega)$ of WO$_3^{RBA}$ (solid line) and Na$_x$WO$_3^{RBA}$ (dotted line) for $x = 0.4$. (c) Electron-phonon spectral function $\alpha^2 F(\omega)$ for $x = 0.4$, where the dashed (WO$_3^{RBA}$) and dotted-dashed (Na$_x$WO$_3^{RBA}$) lines stand for $\lambda(\omega)$ defined as $\lambda(\omega) = 2 \int_0^{\omega} \alpha^2 F(\omega')/\omega' d\omega'$.

For $x \leq 0.4$, the symmetry of a unit cell is not cubic, but tetragonal, orthorhombic, or triclinic depending on the value of $x$. In this case, the shape of a unit cell changes and the arrangement of octahedral WO$_6$ becomes complicated. However, the distortion of octahedra WO$_6$ is small and its structure is close to that when it is inside a cube. Therefore, we consider only the contribution of part of WO$_3$, that is, octahedral WO$_6$, and neglect the effects of Na atoms except for the effect of supplying carriers to the system. From the result for $x \geq 0.4$ shown in Fig. 3, the effect of Na atoms is expected to be small in the region of $x \leq 0.4$. Therefore, this approximated treatment may be allowed for the estimation of $T_c$. In fact, DOS of Cs$_8$WO$_3$, which has a hexagonal structure, is similar to that of Na$_x$WO$_3$ near the Fermi energy despite the difference in crystal structure. Furthermore, the absolute value and the $x$-dependence of $T_c$ obtained in experiments seem to be also similar for both systems.

To examine the effect of Na atoms in more detail, we calculate phonon dispersion, phonon DOS, and the electron-phonon coupling function $\alpha^2 F(\omega)$ for WO$_3^{RBA}$ and Na$_x$WO$_3^{RBA}$ at $x = 0.4$. Here, an integral of $\alpha^2 F(\omega)/\omega$ is related to $\lambda$ as $\lambda(\omega) = 2 \int_0^{\omega} \alpha^2 F(\omega')/\omega' d\omega'$. Figure 4(a) indicates that the phonon dispersions of both WO$_3^{RBA}$ and Na$_x$WO$_3^{RBA}$ are very similar, except for the almost flat mode of NaWO$_3^{RBA}$ at $\omega \approx 18$ meV. As shown in Fig. 4(b), this mode appears as a sharp peak at $\approx 18$ meV in the phonon DOS of NaWO$_3^{RBA}$, whereas there is no corresponding peak in that of WO$_3^{RBA}$. It can be interpreted as an oscillation mode of Na atoms in the cell of NaWO$_3$. Since there seems to be no dispersion, the vibration of Na atoms behaves as an Einstein phonon with a frequency $\approx 18$ meV. It also indicates that Na atoms are isolated in the crystal, and phonons of Na have little effect on those of other atoms. A similar situation has been noted on the Rb$_x$WO$_3$ and Cs$_x$WO$_3$ systems.

Figure 4(c) shows that the main contribution to $\lambda$ comes from part of WO$_3$ and the contribution from Na may be negligible. This result is consistent with the electron DOS of Na atoms being almost zero at the Fermi energy. This figure also shows that the contribution of acoustic phonon modes near the $\Gamma$-point ($\omega \leq 30$ meV) is not large, and the main part of $\lambda$ comes from the optical modes of WO$_3$ for $\omega \geq 30$ meV, by considering it against the result showing in Fig. 4(a). If the long-wavelength modes such as acoustic phonons are not essential for the evaluation of $\omega_{\log}$ and $\lambda$, the arrangement of octahedral WO$_6$ is expected to be not important, and the above-mentioned simplifications omitting the effects of Na atoms may be justified for the estimation of $T_c$.

Since the tungsten atoms are staggered in adjacent unit cells in the case of $x \leq 0.3$, we use a supercell made of two unit cells of WO$_3$ as a new unit cell, 2WO$_3$, to calculate $\omega_{\log}$ and $\lambda$. The structure of 2WO$_3$ and atomic positions are determined by the structural relaxation within RBA. For $x = 0.2$ and 0.3, we find that the suitable structure of WO$_3$ is tetragonal, whereas it is orthorhombic for $x = 0.1$ owing to the subsequent structural transition.

![Fig. 5](image)

**Fig. 5.** (Color online) Calculated $T_c$ as a function of $x$ with the results of experiments. Here, solid circles represent our calculated $T_c$ and double circles with the dotted line represent that excluding the plasmon effect. Open circles indicate experimental $T_c$ of Na$_x$WO$_3$, open triangles represent that of Cs$_x$WO$_3$, crosses indicate K$_x$WO$_3$, and open squares indicate Rb$_x$WO$_3$. Double triangles with the dotted line indicate the theoretical estimation of $T_c$ for Cs$_x$WO$_3$ obtained by Pellegrini et al. and solid triangles indicate that added to the plasmon effect. The inset shows a semi-log graph for $T_c$ as a function of $x$. In Fig. 5, we show the calculated $T_c$ as a function of $x$ with several results of experiments and that of theoretical result for
Cs$_3$WO$_3$.\(^{33}\) Here, to clarify the effect of plasmons, we show the theoretical results with and without the plasmon effect. The former (solid circles) is in good agreement with the experimental results of Na$_8$WO$_3$ (open circles), whereas values of the latter\(^{24}\) (double circles) are close to 0K, indicating that they are too low. These results suggest that the plasmon effect is important to explain the superconductivity of Na$_8$WO$_3$ as well as the phonon mechanism.

The theoretical estimation obtained by Pellegrini et al.\(^33\) for Cs$_3$WO$_3$ (double circles) does not include the plasmon effect. Therefore, if we add the effect to their result, the obtained values of $T_c$ (solid triangles) increase and seem to be in good agreement with the result of the experiment (open triangles). Here, to estimate $T_c$, we combined the values of $\lambda$ obtained by Pellegrini et al.\(^33\) and the phenomenological relation (2) for $\mu^*$, and $\omega_{\text{log}}$ is assumed to be 200 K for simplicity. This result also indicates the importance of the plasmon effect for superconductivity.\(^{18-20,22}\) It is interesting that absolute values and the $x$ dependence of $T_c$ for Na$_8$WO$_3$ seem to be similar to those for Cs$_3$WO$_3$ and other materials despite differences in the crystal structures.\(^{31,33}\) This suggests that the octahedra common in these materials are mainly responsible for their superconductivity.

In an actual Na$_8$WO$_3$ system, it becomes an insulator when $x$ is smaller than about 0.2 and its superconductivity is not observed. Because the structural transition occurs and the crystal symmetry of the system changes, the effect of the Anderson localization may be enhanced, which causes the metal-insulator transition observed at $x \approx 0.2$.\(^7\) Note that our result is obtained from a perfect crystal without randomness, rather than realistic crystals with randomness.

The inset in Fig. 5 shows a semi-log plot of $T_c$. It indicates that the $x$-dependence of $T_c$ seems to be given by $T_c \propto A \exp(-Bx)$ for $0.1 \leq x \leq 0.6$, where A and B are constants. We find these values to be $A=20.7$ [K] and $B=9.3$ by fitting to the data of Na$_8$WO$_3$. This result gives a theoretical base for understanding the exponential behavior of $T_c$ obtained by the experiment for $0.2 \leq x \leq 0.4$.\(^1\) The inset also indicates that $x$-dependences are almost the same regardless of the material except for the theoretical results which have no plasmon effect. By including the plasmon effect to the theory, we can obtain good agreement with the experiments.

Finally, we discuss the relationship between our result and the HTS of Na$_8$WO$_3$. If we use the above fitting equation for $T_c$, it becomes about 13 K at $x = 0.05$. This result is clearly inconsistent with the result of the experiment on HTS. Of course, HTS is observed on the surface and we should pay attention to the dimensionality of the system. The surface superconductivity has already been examined by the first-principles calculations, although the result is limited to hole doped diamond systems.\(^{35,36}\) Results indicate that the electronic state forms a characteristic bound state on the surface. However, the values of $\omega_{\text{log}}$ and $\lambda$ are comparable to those of the bulk, and no special mechanism was found to significantly increase these values.\(^{37}\)

On the other hand, the absolute value of $\mu^*$ increases with decreasing dimensionality of the system. Using the result of Takada,\(^8\) we find that the absolute value of $\mu^*$ increases by about 0.05 at $x = 0.05$ for a pure two-dimensional system.\(^{38}\) If we assume $\omega_{\text{log}} = 500$ K and $\lambda = 0.3$,\(^39\) which are typical values for a bulk system as shown in Fig. 3, $T_c$ including the enhancement of $\mu^*$ becomes about 20K. However, this value is far from ~90 K of HTS. Therefore, it is difficult to explain the HTS of Na$_8$WO$_3$ by the conventional phonon mechanism including the plasmon effect, and some new mechanisms such as orbital fluctuation\(^{40}\) may be required.

In summary, we have investigate the $x$ dependence of $T_c$ in sodium tungsten bronze (Na$_8$WO$_3$) first-principles calculations. We find that the superconductivity is dominated by a part of WO$_3$, and the role of Na atoms is almost limited to providing carriers to the conduction band of the system. Combined with the McMillan equation and the phenomenological relation of $\mu^*$ including the plasmon effect, we show that $T_c$ is given as $T_c \approx 20.7 \exp(-9.3x)$ for $0.1 \leq x \leq 0.6$, which is consistent with the result obtained by the experiment for $0.2 \leq x \leq 0.4$. We have found that plasmons are as important as phonons for the superconductivity of Na$_8$WO$_3$. Although $T_c$ increases with decreasing $x$, it may not exceed ~20K for small $x$. This result is inconsistent with HTS up to about 90 K at $x \sim 0.05$ observed in the recent experiment for the surface. This discrepancy may require some new mechanism to explain the HTS of Na$_8$WO$_3$ beyond the conventional phonon mechanism.

In this work, we used the Gaussian smearing method to obtain $\lambda$, $\omega_{\text{log}}$, and $\alpha^2 F(\omega)$. The possible errors of these values are not large.\(^{27}\) However, more efficient method introduced by Koretsune and Arita\(^{41}\) may give more accurate results for these values. In addition, we used the phenomenological relation of $\mu^*$ and the McMillan equation to calculate $T_c$, but it is insufficient from a theoretical viewpoint. More microscopic theories such as superconducting density functional theory including the plasmon effect will be required.\(^{20,21}\) We would address these problems in our future study.

Acknowledgment This work was supported by JSPS KAKENHI Grant Numbers 17K05539 and 19K03716. The authors thank Takuya Sekikawa, Rai Watabe, Jun Ishizuka, and Kouki Hara for valuable discussions.

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