# High-pressure study of layered nitride superconductors

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Pressure dependence of critical temperature, lattice constant, and phonon frequency has been investigated for layered nitride superconductors, $ZrNCl_{0.7}$ and $Li_{0.5}(THF)_yHfNCl$. The analysis of the data in terms of MacMillan's theory indicated that the relevant phonon frequencies are low ($\approx$50 and 100 $cm^{-1}$, respectively), and that the electron-phonon coupling constant $\lambda$ is larger than 3 in both compounds in sharp contrast with previous experimental and theoretical results. This result may suggest a possibility that other bosonic excitation than phonon additionally contributes to the pairing interaction in these materials.

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## I. INTRODUCTION

Identifying pairing-interaction in superconductors is of considerable importance and crucial for the complete understanding of the fascinating phenomena. In simple metals and alloys,[1] electron-phonon interaction plays the role, typically manifesting itself as an isotope effect on critical temperature ($T_c$). It is widely accepted that high-$T_c$ superconductivity in doped $C_{60}$ (Ref. 2) is also mediated by high-frequency intramolecular vibration and that in recently discovered $MgB_2$ (Ref. 3) by optical phonon mainly due to B ion, respectively. On the other hand, other interaction is believed to be responsible for pairing mechanism in some materials, such as high-$T_c$ cuprates,[4] ruthenate,[5] heavy-fermion compounds,[6] and organics.[7] Among superconductors with relatively high $T_c$, electron-doped $\beta$-ZrNCl and $\beta$-HfNCl still remain mysterious[8] as to the pairing mechanism.

It has been found[9–11] that $\beta$-ZrNCl and $\beta$-HfNCl become superconducting upon electron doping by means of alkali intercalation or chlorine deintercalation. Undoped compounds are band insulators with layered structure; double honeycomb Zr-N (or Hf-N) layers [depictured in the inset to Fig. 1(b)] are sandwiched by Cl layers. Band calculations[12–14] have indicated that the conduction band is derived mainly from Zr 4*d* and Hf 5*d* orbitals, respectively, while the valence band has mostly N 2*p* character. These calculations have also suggested that the electronic structure is two-dimensional, reflecting the layered nature of the crystal structure. The prediction was confirmed by magnetic measurements[15] and x-ray absorption measurement.[16] Recent NMR study[17] has shown that the superconducting phase has an even-parity spin-singlet order parameter. However, several experiments[17–20] have suggested possible unconventional superconductivity: A $\mu$SR study[18] has shown that the superconducting transition in this system can better be viewed as Bose-Einstein condensation type rather than conventional Bardeen-Cooper-Shrieffer type. Tou *et al.* have found large discrepancy[17,19] between the values of electron-phonon coupling constant $\lambda$ that were estimated from thermodynamic quantities and from an isotope effect on $T_c$, and have discussed a possibility that other boson may mediate the superconductivity. A recent tunneling spectroscopy measurement[20] has shown an anomalously large superconducting gap ratio $2\Delta/k_BT_c=5.0$–5.6 which well exceeds that of conventional strong-coupling superconductors[21] such as Pb (=4.50) or Hg (=4.59), where $\Delta$ denotes the superconducting gap. To gain more information on the microscopic mechanism of superconductivity, we have investigated the pressure dependence of three different physical quantities, namely, $T_c$, lattice constant, and optical phonon frequency. A high pressure study on the $T_c$ of a related material has already been reported by Shamoto *et al.*,[22] but the simultaneous determination of the pressure dependence of these quantities in this work allows us to make quantitative discussions on the parameters included in the MacMillan formula.[23] The analysis revealed that the relevant phonons have lower frequencies than 100 $cm^{-1}$, and that the electron-phonon coupling constant $\lambda$ is surprisingly large, provided that the present superconductor is within the MacMillan scheme. The unprecedentedly large value of $\lambda$ is in striking contrast with a theoretical prediction[14] and an estimate from thermodynamic quantities.[19] This might suggest that dynamical aspects of Coulomb interaction[24,25] must be taken into account in these superconductors.

## II. EXPERIMENT

Cl-deintercalated $\beta$-ZrNCl ($ZrNCl_{0.7}$) and Li and tetrahydrofuran (THF) cointercalated $\beta$-HfNCl [$Li_{0.5}(THF)_yHfNCl$] were prepared for the high-pressure experiments. Details of sample preparation have already been reported elsewhere.[9,11] After the intercalation or deintercalation procedure was made, samples have been kept in evacuated sealed glass tubes, or carefully treated in a glove box with high-purity Ar gas to avoid their degradation due to moisture and/or oxygen. Determination of $T_c$ under pressure was made by means of ac-susceptibility measurement in a piston-cylinder-type clamp cell. We estimated the pressure difference in the clamp cell between at room temperature and at low temperatures by measuring the pressure dependence of $T_c$ of Pb prior to the

 

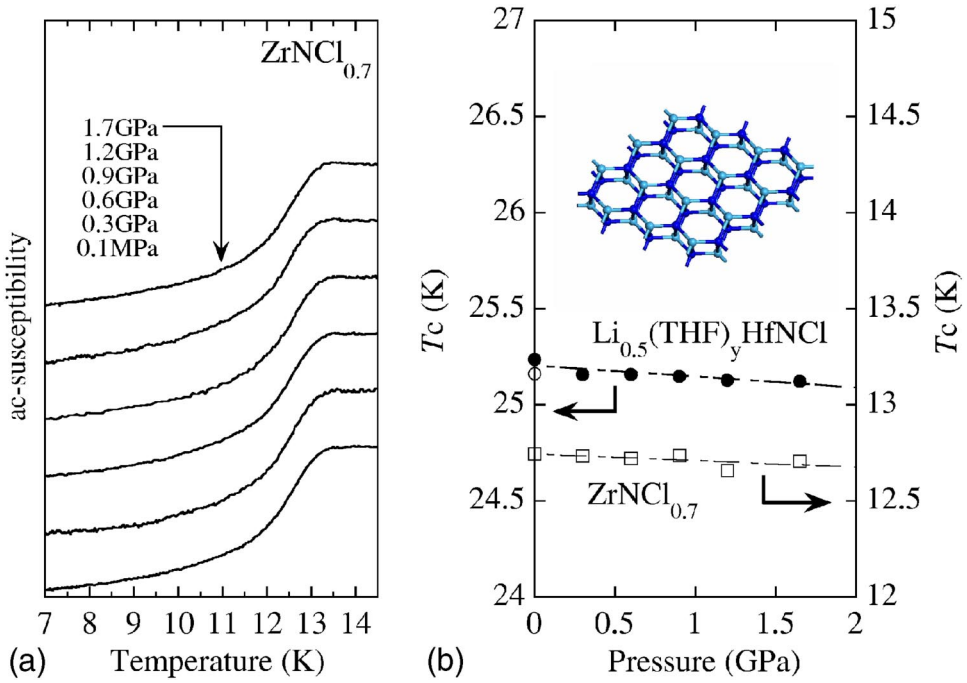


FIG. 1. (Color online) (a) ac-susceptibilities of $ZrNCl_{0.7}$ are plotted against temperature for various values of applied pressure. (b) Pressure dependence of $T_c$ for $ZrNCl_{0.7}$ and $Li_{0.5}(THF)_yHfNCl$ as determined from ac-susceptibility. An open circle for $Li_{0.5}(THF)_yHfNCl$ at zero pressure represents the $T_c$ value after the pressure was completely released. Inset depicts double honeycomb Zr-N (or Hf-N) layer.

sample measurement, and calibrated the pressure value for the separate sample measurements, using this information. High-pressure synchrotron x-ray diffraction measurements were done with a use of diamond anvil cell at BL10XU, SPring-8. The wavelength of the monochromated x-ray was 0.49583(5) Å and the diffraction patterns were detected using an image-plate. We also used a diamond anvil cell for the high-pressure Raman scattering experiments. A 532 nm light was used for excitation, and backward scattered light was collected and dispersed by a single monochrometer equipped with a liquid-nitrogen-cooled charge-coupled device detector. In all the high-pressure experiments, fluorinert was used as the pressure transmitting medium, and the data was taken in pressure increasing run. In the diamond anvil cell experiments, fluorescence of ruby was used to monitor the pressure in the diamond anvil cell, and the measurements were done at room temperature.

### III. RESULTS

In Fig. 1(a), we display temperature dependence of the ac-susceptibility of $ZrNCl_{0.7}$ sample for various values of pressure. At ambient pressure, the onset of superconductivity is clearly signaled by a decrease of the ac-susceptibility at around 13 K. As the pressure is increased up to 1.7 GPa, the onset temperature gradually decreases, but the change is rather small. Figure 1(b) displays pressure dependence of $T_c$, together with that of $Li_{0.5}(THF)_yHfNCl$. Here, $T_c$ is defined as the temperature where the diamagnetic signal becomes $\frac{1}{5}$ of the low temperature saturation value. An open circle for Hf compound at zero pressure indicates $T_c$ value after the pressure was completely released. This value is within the range of experimental error, ensuring the stability of the material under pressure. The rates of the decrease in $T_c$ are $(1/T_c\, dT_c/dP) = -(2.7 \pm 1.9) \times 10^{-3}$/GPa and $-(2.3 \pm 1.1) \times 10^{-3}$/GPa for $ZrNCl_{0.7}$ and $Li_{0.5}(THF)_yHfNCl$, respectively.[26] Compared with the values reported for $K_3C_{60}$ $[(1/T_c d\, T_c/dP) = -0.41/\mathrm{GPa}]$ (Ref. 27) or $MgB_2$ $[(1/T_c)(dT_c/dP) = -(4.3-5.4) \times 10^{-2}/\mathrm{GPa}]$,[28,29] these values are quite small, but in the same order of magnitude as a previous result $[(1/T_c)(dT_c/dP) = -6.7 \times 10^{-3}/\mathrm{GPa}]$ (Ref. 22) on a similar material ($Na_{0.3}HfNCl$) with slightly lower $T_c$(=19.4 K).

To estimate pressure dependence of the electronic density of states, we have made x-ray diffraction measurements under high pressure up to 5.2 GPa and have determined the lattice constant. In Fig. 2(a), we plot the diffraction profiles of $ZrNCl_{0.7}$ at several values of pressure. As the pressure is increased, all the peaks show a slight shift to a higher angle, but there seems to be no structural phase transition below the highest pressure (5.2 GPa) in this study. Both (003) peak and (110) peak shift to higher angle by about 0.2° with an application of 5.2 GPa. When we take the difference in the $2\theta$ values of (003) and (110) peaks into account, this result indicates that $c$-axis direction is more compressible than in-plane directions.

In Fig. 2(b), normalized lattice parameters of $ZrNCl_{0.7}$ are plotted together with the least-squares fit to the semiempirical Murnaghan relation[30] of $P = (K_x/K_x')[(x_0/x)^{K_x'} - 1]$, where $x = a, c$. This clearly exemplifies the different compressibilities along the $c$-axis and within the conduction plane. The linear compressibilities are $(1/a)(da/dP) = -2.8(3) \times 10^{-3}$/GPa and $(1/c)(dc/dP) = -3.6(5) \times 10^{-2}$/GPa for the $a$- and $c$-axes, respectively. The compressibility along the $c$-axis is more than ten times as large as that within the plane. This anisotropy is rationalized by the two-dimensional character of the material, where inter- and intra-layer bonding nature are of van der Waals and covalent, respectively.

Next, we have investigated the lattice dynamics under pressure by measuring the phonon Raman scattering. In Fig.

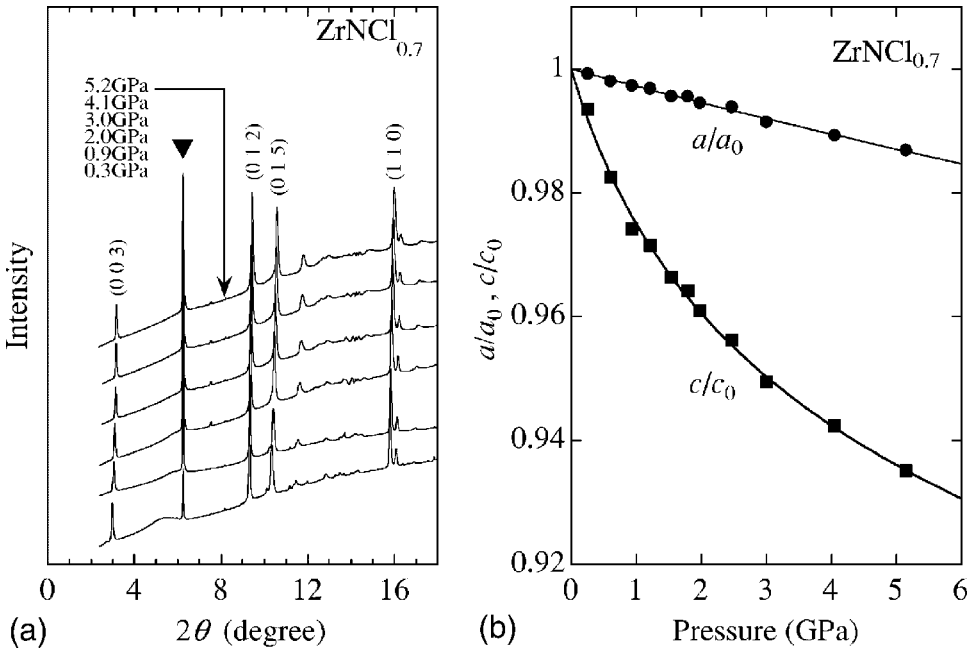


FIG. 2. (a) X-ray diffraction profiles of $ZrNCl_{0.7}$ at room temperature for various values of applied pressure. A peak indicated by a triangle represents a diffraction peak from the diamond. (b) Pressure dependence of normalized lattice constants for $ZrNCl_{0.7}$, together with fitting curves to a Murnaghan relation.

3(a), we show the Raman spectra of $ZrNCl_{0.7}$ under several pressure values. According to a recent Raman scattering study,[31] three peaks labeled as A, B, and C are assigned as out-of-plane Cl-, out-of-plane Zr-, and in-plane N-vibration, respectively. All the peaks display a slight upward shift against pressure. Figure 3(b) shows the pressure dependence of the normalized phonon frequencies, together with a linear fit to the data. Although the rate of the frequency shift depends on the particular phonon mode, it is within the range of $0.005/\mathrm{GPa} < (1/\omega)(d\omega/dP) < 0.02/\mathrm{GPa}$.

### IV. ANALYSIS AND DISCUSSION

We analyze the data of $ZrNCl_{0.7}$ in terms of MacMillan's formula.[23] This formula has been used as a versatile prescription for phonon-mediated superconductors, including the strong-coupling one. Recently, the pressure dependence of $T_c$ in $MgB_2$ has successfuly been explained[28,32] in terms of this formula. According to the theory,[23] $T_c$ is given by

$$k_B T_c = \frac{\hbar\langle\omega_{ph}\rangle}{1.2} \exp\frac{-1.04(1+\lambda)}{\lambda - \mu^*(1+0.62\lambda)}, \tag{1}$$

where $\langle\omega_{ph}\rangle$ represents relevant phonon frequency. $\lambda$ and $\mu^*$ are written[23,33] as $\lambda = N(0)\langle I^2\rangle / M\langle\omega_{ph}^2\rangle$ and $\mu^* = \mu/[1+\mu\ln(E_F/\langle\omega_{ph}\rangle)]$. $N(0)$, $I$, and $M$ are the density of states at the Fermi energy, electron-phonon matrix element, and atomic mass, respectively. $\mu$ is expressed as $\mu = N(0)V_c$, where $V_c$ represents the screened Coulomb interaction. $\lambda$ and $\mu^*$ vary with application of pressure *via* the change of $N(0)$, $\langle\omega_{ph}\rangle$, and $E_F$. In the present system, band calculations[12–14] show that only single band is relevant to the electronic states near the Fermi energy and that the electronic state is almost two-dimensional. Therefore, we can safely make a simple approximation of $N(0) \propto t^{-1}$, where $t$ is the intraplane transfer integral between the nearest neighbor Hf and N sites. Furthermore, $t$ depends on the lattice constant $a$ as $t \propto a^{-3.5}$ according to Harrison's parametrization.[34] $E_F$ also depends on $t$ as $E_F \propto t^{-1}$. Using these relations, we can derive[35]

$$\frac{1}{T_c}\frac{dT_c}{dP} = \frac{1}{\langle\omega_{ph}\rangle}\frac{d\langle\omega_{ph}\rangle}{dP} + \frac{1.04(1+0.38\mu^*)\lambda}{[\lambda-\mu^*(1+0.62\lambda)]^2}\left[\frac{3.5}{a}\frac{da}{dP} - \frac{2}{\langle\omega_{ph}\rangle}\frac{d\langle\omega_{ph}\rangle}{dP}\right] - \frac{1.04(1+\lambda)(1+0.62\lambda)(\mu^*)^2}{[\lambda-\mu^*(1+0.62\lambda)]^2}\left[\frac{3.5}{a}\frac{da}{dP} - \frac{1}{\langle\omega_{ph}\rangle}\frac{d\langle\omega_{ph}\rangle}{dP}\right]. \tag{2}$$

Given the pressure dependence of $T_c$ and lattice constant, we have four unknown parameters in Eqs. (1) and (2), namely, $\lambda$, $\mu^*$, $\langle\omega_{ph}\rangle$, and $(1/\langle\omega_{ph}\rangle)(d\langle\omega_{ph}\rangle/dP)$. When we take a set of values of $\langle\omega_{ph}\rangle$ and $(1/\langle\omega_{ph}\rangle)(d\langle\omega_{ph}\rangle/dP)$, then $\lambda$ and $\mu^*$ are determined from Eqs. (1) and (2). Therefore, $\lambda$ and $\mu^*$ are functions of $\langle\omega_{ph}\rangle$ and $(1/\langle\omega_{ph}\rangle)(d\langle\omega_{ph}\rangle/dP)$. In the present case, a relation of $\mu \approx 0.5$ holds.[35] Then, $\mu^* = \mu/[1+\mu\ln(E_F/\langle\omega_{ph}\rangle)]$ must satisfy a relation of

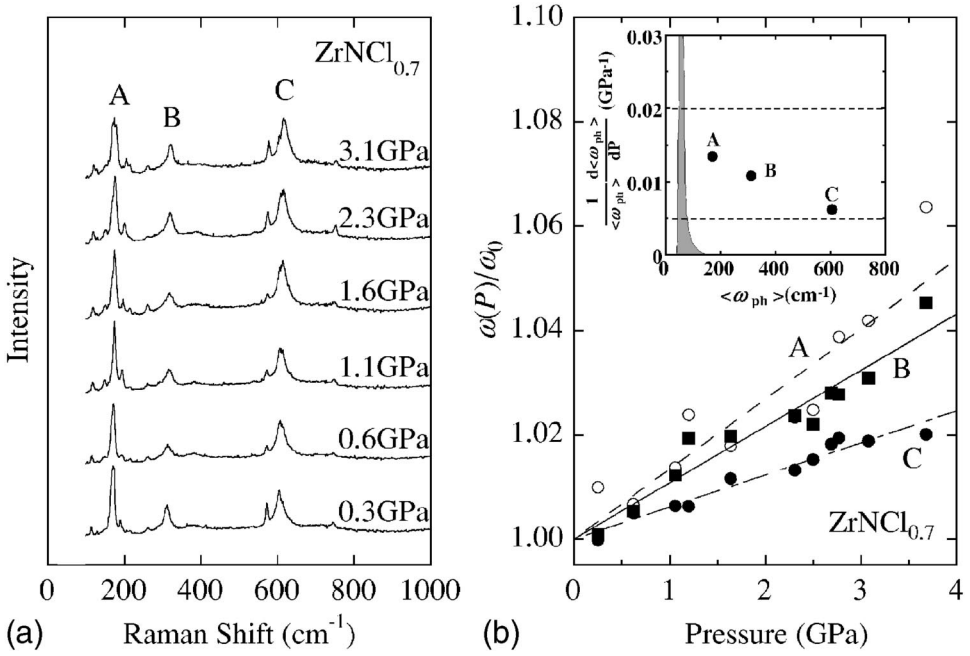


FIG. 3. (a) Raman scattering spectra of $ZrNCl_{0.7}$ for various values of applied pressure. (b) Pressure dependence of normalized phonon frequencies. Straight lines are least-squares fit to the data. Inset: Shaded area represents the region where the necessary condition of $0<\mu^*<0.5$ is fulfilled in the plane of $\langle\omega_{\rm ph}\rangle$ versus $(1/\langle\omega_{\rm ph}\rangle)(d\langle\omega_{\rm ph}\rangle/dP)$. Closed circles show $\omega$ versus $(1/\omega)(d\omega/dP)$ correlation that were observed for phonon lines A, B, and C. Horizontal broken lines are upper and lower limit for $(1/\langle\omega_{\rm ph}\rangle)(d\langle\omega_{\rm ph}\rangle/dP)$.

$0<\mu^*<0.5$ since $E_F/\langle\omega_{\rm ph}\rangle$ is obviously larger than 1. The region of $[\langle\omega_{\rm ph}\rangle,(1/\langle\omega_{\rm ph}\rangle)(d\langle\omega_{\rm ph}\rangle/dP)]$ that satisfies the above constraint is represented as shaded area in the inset to Fig. 3. Horizontal dashed lines correspond to lower and upper limits for $(1/\langle\omega_{\rm ph}\rangle)(d\langle\omega_{\rm ph}\rangle/dP)$ (=0.005 and 0.02 $\mathrm{GPa}^{-1}$, respectively), which were imposed by the Raman scattering measurement under pressure. When the errors of $(1/T_c)(dT_c/dP)$ and $(1/a)(da/dP)$ are taken into account, the lower and higher frequency boundaries slightly shift, but the change is at most $\pm 10$ $\mathrm{cm}^{-1}$ for any values of $(1/\langle\omega_{\rm ph}\rangle)(d\langle\omega_{\rm ph}\rangle/dP)$ between 0.005 and 0.02. Therefore, this result definitely rules out the relevance of the high frequency phonon associated with the light N atom, but strongly suggests the importance of the low frequency phonons around 50 $\mathrm{cm}^{-1}$.

To estimate the value of $\lambda$, we assume $\mu^*=0.14$. We adopted this value, using $\mu\approx 0.5$, $E_F\approx 1$ eV, and $\langle\omega_{\rm ph}\rangle \approx 50$ $\mathrm{cm}^{-1}$, and this value is very close to the commonly accepted values in many superconductors.[21] This particular value of $\mu^*$ also gives a trajectory within the shaded area, on which the $\lambda$ value varies from 3.2 to 3.7. This value is suprizingly large since the largest value reported thus far is 3.0 and most of the superconductors have $\lambda$ values less than 2 (Ref. 21).

As for the $Li_{0.5}(THF)_yHfNCl$, we made the same set of analysis based on the MacMillan scheme, assuming that the $(1/da)(da/dP)$ value of $Li_{0.5}(THF)_yHfNCl$ is the same as that of $ZrNCl_{0.7}$ since $(1/da)(da/dP)$ is not available for $Li_{0.5}(THF)_yHfNCl$. The obtained result is very similar to the case of $ZrNCl_{0.7}$: Relevant phonon frequency is about 100 $\mathrm{cm}^{-1}$, and the $\lambda$ is in the range of 3.3–3.9.

The above results clearly show that the relevant phonons are of low frequency (less than 100 $\mathrm{cm}^{-1}$), and that these superconductors have the largest coupling constant ever found, provided that the MacMillan scheme is applicable to these materials. In strong-coupling superconductors, the superconducting gap ratio $2\Delta/k_BT_c$ is known to obey[36] a relation of

$$\frac{2\Delta}{k_BT_c}=3.53\left[1+12.5\left(\frac{T_c}{\langle\omega_{\rm ph}\rangle}\right)^2\ln\left(\frac{\langle\omega_{\rm ph}\rangle}{2T_c}\right)\right]. \qquad (3)$$

In the case of $Li_{0.5}(THF)_yHfNCl$, this value is calculated to be 5.0 with the use of $\langle\omega_{\rm ph}\rangle\approx 100$ $\mathrm{cm}^{-1}$. This result coincides with the observed value (=5.0–5.6) by a recent tunneling spectroscopy.[20] The $\mu$SR result[18] is also in accord with the strong coupling since the Bose-Einstein condensation can be thought of as the $\lambda\rightarrow\infty$ limit of the BCS-type superconductivity.

On the other hand, the large $\lambda$ over 3 is inconsistent with some experiments and theory: Weak electron-phonon interaction and small density of states at the Fermi level are suggested from the analysis of thermodynamic quantities.[19] The band calculation[14] also predicts too weak electron-phonon interaction to reproduce $T_c=25$ K. One possibility to reconcile all the theory and experiments is that other bosonic excitation than phonon additionally contributes to the pairing interaction.[24,25] (Phonon must play at least some role in the pairing mechanism in view of the finite isotope effect.) According to Bill *et al.*,[25] for example, $T_c$ as high as 25 K can be expected even for $\lambda=0.3$ if they take into account an additional pairing-channel from acoustic plasmon that originates from the dynamical part of the Coulomb interaction, which is neglected in the MacMillan framework.

## V. SUMMARY

We have investigated the pressure dependence of $T_c$, lattice constant, and phonon frequency of superconducting layered nitrides, $ZrNCl_{0.7}$ and $Li_{0.5}(THF)_yHfNCl$. The analysis revealed that the superconducting pairing is mediated by low frequency phonons with $\approx$50 and 100 $\mathrm{cm}^{-1}$, respectively, and that the coupling constant $\lambda$ is larger than 3 in both

materials, if MacMillan's scheme is applicable to the present system. However, such a large coupling constant makes a striking contrast with a theory[14] and an estimate[19] from thermodynamic quantities. This might imply that the MacMillan scheme is not sufficient for these materials and that the dynamical aspect of Coulomb interaction that is neglected in the framework must be taken into account. Further experimental and theoretical studies are needed to clarify the details of pairing mechanism and the origin of the high critical temperature in this system.

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