Evidence of a spin-density wave in hcp zirconium

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Phonon spectra of both hcp Y and Zr have been calculated using shell-model pseudopotentials. Whereas Y displays no significant anomaly in its spectrum, the phonon frequency of the [001] LO branch of Zr exhibits a pronounced softening near the zone center. We have found that the anomalous behavior of the LO phonons in Zr can be well understood with the conjecture that Zr has a longitudinally polarized spin-density wave with wave vector $\mathbf{Q}=(2\pi/c)(0,0,1)$. This conjecture is consistent with several other experimental results for Zr, including its large magnetic anisotropy, its lack of anisotropic magnetic scattering, and the topology of its Fermi surface.

I. INTRODUCTION

The group-IV superconducting metals, i.e., Ti, Zr, and Hf, have attracted attention in recent years on account of anomalous features in several physical properties. These metals all have a hcp structure at low temperature, and undergo a phase transition to a bcc phase well above room temperature. Detailed experimental studies[1-3] with neutron scattering have shown that in the hcp phase the long-wavelength phonon frequencies of the $[00\xi]$ LO branch decrease with decreasing temperature and, eventually, at or below room temperature this branch exhibits a dip at the zone center. The frequencies of all other branches, on the other hand, were found to have a normal temperature dependence (increasing frequencies with decreasing temperature). The unexpected dips, while absent in the group-III hcp metals Sc and Y, resemble what was observed in Pb, where phonon dispersion curves display a downward cusp at the $\{100\}$ zone-boundary points for both transverse and longitudinal modes, and the sizes of the dips increase with decreasing temperature.[4] The anomalous behavior in Pb has been explained[5] successfully by assuming the existence of a cubic family of linear spin-density waves (SDW's). This hypothesis, however, has yet to be tested by neutron scattering.

The magnetic properties of Ti, Zr, and Hf also show puzzling features. Bulk magnetic susceptibility measurements[6] on these metals display considerable anisotropy, i.e., the susceptibility depends on whether the magnetic field is parallel or perpendicular to the (hexagonal) c axis. In Zr, for example, $\chi_{\parallel}$ is 1.65 times larger than $\chi_{\perp}$ at $T=70$ K. Moreover, the anisotropy decreases with increasing temperatures. The temperature dependence of the anisotropy of the susceptibility of seven hcp metals (Ti, Zr, Hf, Y, Re, Ru, and Os) has been studied by several groups,[6,7] and is illustrated in Fig. 1. Extrapolation of $\Delta\chi(T)\equiv\chi_{\parallel}-\chi_{\perp}$ to higher temperatures indicates that for these metals (excepting Re) the temperature at which the anisotropy disappears is rather close to the melting temperature. The sharp drop of $\Delta\chi$ in Ti and Zr near 1187 and 1167 K is believed to be associated with the hcp to bcc phase transition. The anisotropy at 70 K for seven metals is summarized in Table I.

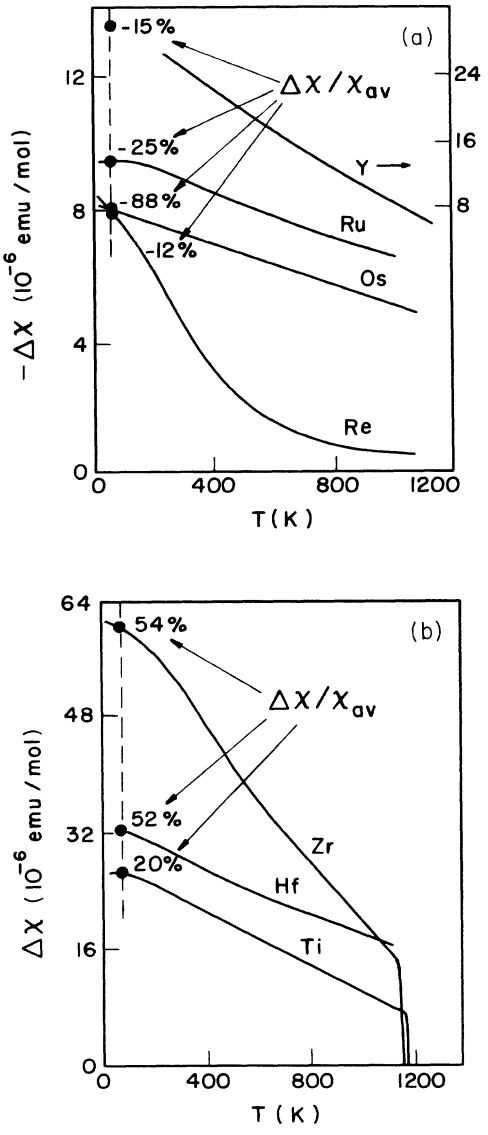

FIG. 1. Temperature dependence of the anisotropy of the magnetic susceptibility of (a) yttrium, ruthenium, osmium, and rhenium; (b) titanium, zirconium, and hafnium. Data are from Ref. 7. $\Delta\chi=(\chi_{\parallel}-\chi_{\perp})$ and $\chi_{av}=(\chi_{\parallel}+2\chi_{\perp})/3$.

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TABLE I. Components of the magnetic susceptibility of seven metals at $T=70$ K. $\chi_{av}=(\chi_{\parallel}+2\chi_{\perp})/3$ and $\Delta\chi=\chi_{\parallel}-\chi_{\perp}$. All susceptibilities are in μemu/mole.

Metal	$\chi_{\parallel}$	$\chi_{\perp}$	χ_{av}	$\Delta\chi/\chi_{av}$	Ref.
Y	171	199	190	−0.15	8,9
Ti	159	131	140	0.20	6,10
Zr	149	90	109	0.54	6
Hf	96	59	71	0.52	6
Re	61	69	66	−0.12	8
Ru	32.2	41.7	38.5	−0.247	10
Os	3.95	12.3	9.52	−0.877	11

The measured susceptibility, of course, is the sum of a series of diamagnetic and paramagnetic components. It has been suggested that the contribution of the itinerant-electronic orbital paramagnetism is responsible for the anisotropy.[6,7] However, a recent calculation based on the theory[12] of magnetic response of conduction electrons, which includes both orbital paramagnetism and Paul spin paramagnetism, shows the magnetic susceptibility of Zr to be insensitive to field direction, and so is essentially isotropic.[13] The field-induced neutron magnetic scattering in Zr at the reciprocal-lattice vectors $\{\mathbf{G}\}$ has been studied by Stassis *et al.*[14] Measurements for two field directions revealed no evidence of the large anisotropy observed in bulk susceptibility experiments.

It has been suggested that the softening of the [001] LO-phonon frequencies near the zone center is related to an incipient instability toward the formation of a charge-density wave,[2] and originates from the splitting of a doubly degenerate electronic band near the Fermi level caused by the lattice displacements associated with the zone-center [001] LO mode.[15] This explanation required, however, the Fermi-Dirac factors of electronic states near the Fermi level to readjust instantaneously during vibration, even though the optical phonon involved is the highest-frequency mode of the spectrum.

In view of the magnetic properties of the group-IV metals mentioned above, we believe that an interpretation based on the unique charge response[5] of a SDW state should be entertained. It has already been shown[16] that SDW states can cause a large anisotropy in spin susceptibility. For a linear SDW the axis of anisotropy is the SDW polarization vector; and for a spiral SDW the anisotropy axis is perpendicular to the plane of polarization. The sign of the anisotropy can be either positive or negative, depending on whether the SDW wave vector is greater than or less than $2k_F$ (the diameter of the Fermi surface).[16] Three of the metals listed in Table I have a positive anisotropy and four have negative values.

The purpose of this paper is to point out that the anomalous behavior of the [00ξ] LO phonons in hcp Zr can be understood if a linearly polarized SDW having wave vector $\mathbf{Q}=(2\pi/c)(0,0,1)$ is present. In this event the magnetic anisotropy and the phonon anomaly would have a common SDW origin which, in any case, needs to be present to account for the magnetic anisotropy. It is of interest to note that the Zr Fermi surface derived from an analysis of de Haas–van Alphen experiments,[17] shows that the proposed SDW $\mathbf{Q}$ vector almost spans the Fermi surface, as plotted in Fig. 2, a condition which can lead to the formation of a SDW ground state.[18]

We shall also calculate the phonon spectrum of hcp Y (having atomic number $Z=39$), which is adjacent to Zr ($Z=40$) in the Periodic Table. The result provides a reasonably good fit to the available experimental data. Other calculations[19,20] using pseudopotential models have been rather unsatisfactory. Our success for the case of Y indicates that the shell-model theory[21,22] of lattice dynamics is applicable even to metals at the beginning of a transition series. For this reason we would not expect a significant discrepancy to arise in Zr unless new physics, e.g., a linear SDW, intervenes.

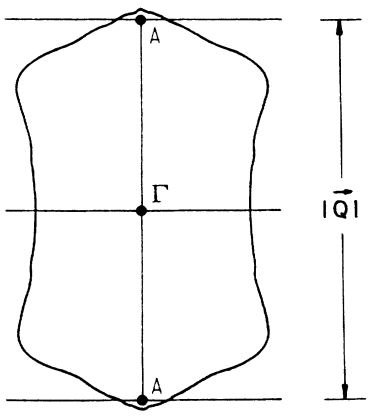

FIG. 2. (1,0,0) cross section of the fourth zone Fermi surface of Zr, determined from the de Hass–van Alphen experiment data of Ref. 13. The coordinates of the A points are $(0,0,\pm 0.5)$.

II. LATTICE DYNAMICS AND CHARGE RESPONSE OF A LINEAR SDW

The theory we shall use for studying the lattice dynamics of Y and Zr is the pseudopotential shell model developed by Wang and Overhauser.[21] This model has been successful in reproducing phonon spectra of more than a dozen simple metals,[22] including fct In and hcp Mg,[23] with only two (in most cases) or three adjustable parameters. The applicability of the theory to metals at the *beginning* of a transition series is the issue we shall investigate here.

The theory of Wang and Overhauser allows the last-filled shell of an ion to vibrate relative to its core. This feature turned out to be important for heavy metals such as Au and Pb. For lighter metals shell vibrations did not have a significant effect on the phonon spectrum, and we expect this to be the case for Y and Zr. Accordingly we shall ignore the deformation of each ion and let the shells move in unison with their ion cores. In a metal having l atoms per primitive cell the phonon frequencies corresponding to a wave vector $\mathbf{q}$ can be found by solving the following eigenvalue equation:

$$\omega_{\mathbf{q}}^2 \mathbf{u}_\alpha(i) = \sum_{j,\beta} D^*_{\alpha\beta}(i,j)\mathbf{u}_\beta(j) , \tag{1}$$

where $\mathbf{u}(i)$ is the displacement of the ith ion in a primitive cell and α and β $(=x,y,z)$ refer to the spatial components in a Cartesian-coordinate system. The dynamical matrix $D_{\alpha\beta}(i,j)$ is given by

$$D_{\alpha\beta}(i,i) = \frac{4\pi n}{lM}\left[\sum_{\mathbf{G}} \frac{|\bar{\rho}(\mathbf{G}+\mathbf{q})|^2(\mathbf{G}+\mathbf{q})_\alpha(\mathbf{G}+\mathbf{q})_\beta}{|\mathbf{G}+\mathbf{q}|^2} - \sum_{\mathbf{G}\,(\neq 0)}^{l} \frac{|\bar{\rho}(\mathbf{G})|^2 \mathbf{G}_\alpha \mathbf{G}_\beta}{|\mathbf{G}|^2} \sum_{k=1}^{l} \cos(\mathbf{G}\cdot r_{ik})\right] \tag{2}$$

for $i=j$, and

$$D_{\alpha\beta}(i,j) = \frac{4\pi n}{lM}\left[\sum_{\mathbf{G}} \frac{|\bar{\rho}(\mathbf{G}+\mathbf{q})|^2(\mathbf{G}+\mathbf{q})_\alpha(\mathbf{G}+\mathbf{q})_\beta}{|\mathbf{G}+\mathbf{q}|^2} e^{-i\mathbf{G}\cdot\mathbf{r}_{ij}}\right] \tag{3}$$

for $i\neq j$, where $\{\mathbf{G}\}$ are the reciprocal-lattice vectors, M is the ionic mass, and n is the density of primitive cells. $\bar{\rho}(\mathbf{Q})$ in Eqs. (2) and (3) represents the Fourier transform of the total pseudocharge density of an ion $\bar{\rho}(\mathbf{r})$, which includes two parts: the charge distribution of an ion core $\bar{\rho}_c(\mathbf{r})$, and the charge distribution of a last-filled electronic shell $\bar{\rho}_s(\mathbf{r})$. For both Y and Zr the last-filled shell is $4s^2 4p^6$. According to Ref. 21 the shell charge density can be represented by the one-parameter function,

$$\bar{\rho}_s(r) = -2e(2\pi)^{-3/2}R_s^{-3}\exp(-r^2/2R_s^2) - 6e[(2\pi)^{-3/2}/3]R_s^{-5}r^2\exp(-r^2/2R_s^2) . \tag{4}$$

The pseudocharge density of an ion core can be taken to be a Gaussian, i.e.,

$$\rho_c(\mathbf{r}) = e(z+v)(2\pi)^{-3/2}R_c^{-3}\exp(-r^2/2R_c^2) , \tag{5}$$

where $z=8$ is the number of electrons in the shell and v is the valence, the number of conduction electrons per atom. R_s and R_c in Eqs. (4) and (5) are the crucial parameters used to fit the phonon spectrum.

The test-charge–test-charge dielectric function appearing in Eqs. (2) and (3) can be written as

$$\epsilon(\mathbf{q}) = 1 + \frac{Q(\mathbf{q})}{1 - G_+(\mathbf{q})Q(\mathbf{q})} . \tag{6}$$

For a free-electron-like metal without a SDW, $Q(\mathbf{q})$ in the above equation is just the Lindhard response function

$$Q(\mathbf{q}) = \frac{m_b e^2}{\hbar^2 k_F x^2}\left[\frac{1}{2} + \frac{1-x^2}{4x}\ln\left|\frac{1+x}{1-x}\right|\right] \tag{7}$$

with $x=|\mathbf{q}|/2k_F$. The effective band mass m_b can be deduced from the experimental values for the thermal mass m_{th} and the electron-phonon coupling constant λ since

$$m_{\text{th}} = m_b(1+\lambda) . \tag{8}$$

However, in view of the band-structure complexity encountered on entering a transition series, it may be propitious to treat m_b as an extra adjustable parameter. G_+ in Eq. (6) is the spin-symmetric exchange and correlation local field factor for which we use the traditional form (proposed by Hubbard[24]), but with the correct limiting behavior at $x=0$ and $x=\infty$,

$$G_+(x) = \frac{1.1x^2}{1+1.7x^2} . \tag{9}$$

The charge response of a linear SDW state has been studied[5] previously by incorporating the spin-split-phason mechanism.[25] The existence of a SDW in an electron gas introduces an additional charge response $Q^{\text{SSP}}(\mathbf{q})$ over and above the usual Lindhard response. A detailed discussion of the spin-split-phason mechanism and the function $Q^{\text{SSP}}(\mathbf{q})$ can be found in Ref. 5. The most important feature of this new response is that it has small three-dimensional, Gaussian-like peaks for $\mathbf{q}$ near $\mathbf{Q}$ and $-\mathbf{Q}$, where $\mathbf{Q}$ is the SDW wave vector. Accordingly, if we assume that there is a SDW in Zr with $\mathbf{Q}=(2\pi/c)(0,0,1)$, $Q^{\text{SSP}}(\mathbf{q})$ can be represented by

$$Q^{\text{SSP}}(\mathbf{q}) = \frac{A}{x^2}[\exp(-\Pi_+)+\exp(-\Pi_-)] , \tag{10}$$

with

$$\Pi_\pm = B_{ab}^2 W_\pm^{ab\,2} + B_c W_\pm^{c2} , \tag{11}$$

where $\mathbf{W}_\pm = \mathbf{q} \mp \mathbf{Q}$; $x=|\mathbf{q}|/2k_F$. B_{ab} and B_c are, respectively, the inverse widths of the peaks in the basal plane and along the c axis; and $W_\pm^{ab}$ and $W_\pm^c$ are the basal plane and c-axis components of $\mathbf{W}_\pm$. We find that the anomalous behavior of the [001] LO phonons in Zr can be satisfactorily explained by adding $Q^{\text{SSP}}(\mathbf{q})$ to Eq. (7). The coefficient A in Eq. (10) and the B's in Eq. (11) are determined by fitting the calculated dip in the LO frequencies near the zone center to the observed spectrum.

III. CALCULATED RESULTS

A. Yttrium

Figure 3 is a plot of the theoretical phonon spectrum of Y along several symmetry directions together with the data due to Sinha *et al.*[26] Only the two parameters R_s and R_c are used in fitting the eight available frequencies at the Γ, M, and A points. The band mass m_b in Eq. (7)

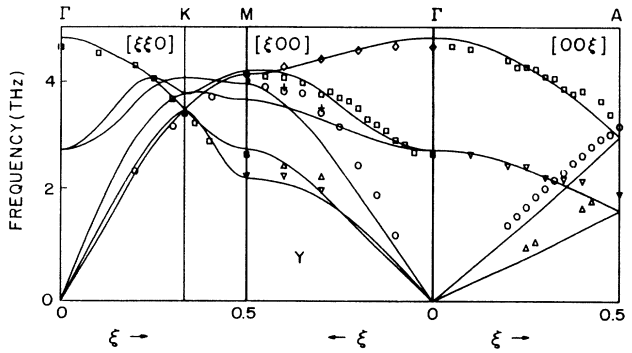

FIG. 3. Phonon spectrum of Y. Data are taken from Sinha *et al.* (Ref. 26) for $T=295$ K. Solid lines are our calculated results with $R_s=1.26/(2k_F)$ and $R_c=2.16/(2k_F)$. The reciprocal-lattice vector sums in Eqs. (2) and (3) included all terms for which $|\mathbf{G}| \leq 10(2\pi/c)$.

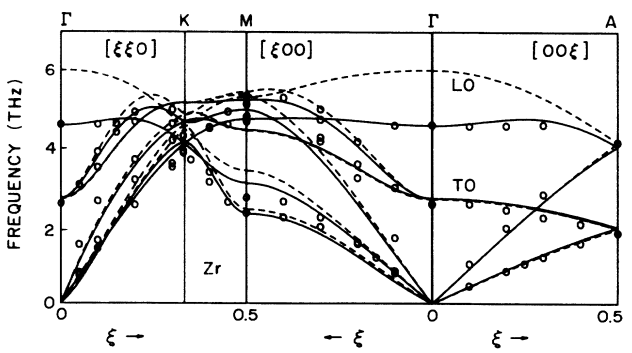

FIG. 4. Phonon spectrum of Zr. Data are taken from Stassis *et al.* (Ref. 1) for $T=295$ K. The dashed lines are our two-parameter fit with $R_s=3.48/(2k_F)$ and $R_c=3.37/(2k_F)$. The solid lines are the calculated results including the influence of the extra charge response caused by the SDW, Eq. (10), with $A=0.093$, $B_{ab}=2.5/(2k_F)$, and $B_c=8.5/(2k_F)$; R_s and R_c are unchanged.

is fixed at $7.6m_e$ by experimental values of $m_{\rm th}$ [$=10.64m_e$ (Ref. 27)] and λ [$=0.4$ (Ref. 28)] according to Eq. (8). The general agreement between theory and experiment is reasonably good. The dispersion curves of Y show no significant anomaly. We have also treated m_b as a third adjustable parameter, but the improvement in the fit was hardly noticeable.

We note that the magnetic susceptibility of Y in Fig. 1(a) also exhibits a considerable anisotropy although the relative magnitude is not as large as that in Zr. Recent studies[29,30] of neutron scattering, heat capacity, and susceptibility indicate that the dilute magnetic alloy $Y_{1-x}Gd_x$ (where $x>0.01$) orders into a long-range helical antiferromagnet at low temperatures via Overhauser's SDW mechanism.[31] The magnetic order has a periodic incommensurate spin structure with the Gd moments directed in the basal plane and a propagation wave vector $|\mathbf{Q}|=0.28(2\pi/c)$ parallel to the c axis. It is possible that pure Y metal may have a small amplitude SDW, which would cause the observed anisotropy in its magnetic susceptibility.[16]

B. Zirconium

The band mass m_b of Zr is found to be $1.6m_e$ from Eq. (8), using $m_{\rm th}=2.24$ (Ref. 27) and $\lambda=0.4$.[28] We again adjust the two parameters R_s and R_c to fit the seven frequencies at (0,0,0) TO, (0,0,0.5) TA, (0,0,0.5) LA, (0.1,0,0) $TA_{\parallel}$, (0.5,0,0) $TA_{\parallel}$, (0.5,0,0) $TO_{\parallel}$, and (0.5,0,0) $TO_{\perp}$. The calculated phonon spectrum resulting from this fit is displayed as dashed lines in Fig. 4. The data points in the figure are taken from the inelastic neutron-scattering measurements (at 295 K) due to Stassis *et al.*[1] This calculation (without a SDW) shows a qualitatively good agreement with experiment in general except for the [00ξ] LO phonons near the zone center Γ, where the observed frequencies are considerably smaller than the calculated ones. The discrepancy cannot be eliminated either by treating the band mass as an adjustable parameter or by including the exchange interaction between conduction electrons and the electrons of the last-filled shells.[21] It is, nevertheless, possible to reproduce the [00ξ] LO branch if we allow the last-filled shells to vibrate relative to their ion cores. However, the required readjustment of R_s and R_c leads to bizarre behavior of the other phonon branches. The fact that Y has a normal phonon spectrum whereas Ti (which is in the same group as Zr, but lighter) exhibits a similar, but more pronounced dip[2] at the zone center implies that the anomalous behavior is driven by an unusual conduction-electron response function, and not by the shell vibrations relative to each ion core.

As we have reviewed in Sec. II, an electron gas with a linear SDW has an additional charge response, Eq. (10), which must be added to the usual Lindhard response function Eq. (7). The solid lines in Fig. 4 are the calculated dispersion curves of Zr which include the influence of a SDW with $\mathbf{Q}=(2\pi/c)$ (0,0,1). All branches ([$\xi\xi$0] $TO_{\perp}$, [ξ00] $TO_{\perp}$, and [00ξ] LO) that have been affected by the softening of the optical phonons near the zone center are now well reproduced. The agreement between theory and experiment along the [ξ00] $TA_{\perp}$ branch is also improved. The additional charge response caused by the SDW has little effect on the long-wavelength acoustic phonons.

IV. CONCLUSIONS

We have proposed a successful theoretical model which reproduces the observed phonon spectra of Y and Zr. The two elements have the same ion-core electronic configuration but differ by unity in the number of conduction electrons. This small difference is nevertheless dramatically reflected in their phonon spectra. While Y has essentially a normal behavior, Zr displays a pronounced frequency softening for the zone center [00ξ] LO mode. We have shown that the anomalous dispersion in Zr can be well explained if Zr has a linear SDW with wave vector $\mathbf{Q}=(2\pi/c)$ (0,0,1). The temperature dependence of the [00ξ] LO phonon frequencies, mentioned in the Introduction, can be understood either as a decrease in SDW amplitude with increasing temperature or as a consequence of thermally induced phase modulation of the SDW, which causes a reduction (and broadening) of the additional charge response peaks given by Eq. (10).

The SDW interpretation of the phonon anomaly in Zr elaborated on in this paper is consistent with several other experimental facts, such as the magnetic anisotropy,[6,7] the lack of anisotropic magnetic scattering,[10] and the topology of the Fermi surface.[17] The study of the spin response[16] of a SDW indicates that the large anisotropy in magnetic susceptibility of Zr would require a SDW with a polarization vector parallel to the hexagonal, c axis. This means that the SDW we envision here must be longitudinally polarized. Since a longitudinal, free-electron SDW produces no magnetic induction **B**, *it is invisible to magnetic neutron scattering.* The only way to detect a nearly-free-electron, longitudinal SDW is to observe magnetic satellites at $\mathbf{Q}+\mathbf{G}$, where **G** is a reciprocal-lattice vector (not parallel to **Q**). Such satellites would be extremely weak because of the small admixture of $\mathbf{k}+\mathbf{G}$ wave-function components into the plane waves of wave vector **k** (by the crystalline potential).

The decrease in magnetic anisotropy with increasing temperature can be attributed to the temperature dependence of the SDW energy gap, which is similar[18] to that of any second-order phase transition, e.g., the energy gap in a superconductor. It should be pointed out that in addition to Ti, Zr, and Hf, Re and Tc exhibit similar anomalous behavior in their phonon spectra.[2,32] The available data for the magnetic susceptibility of Re, shown in Fig. 1(a), also reveals a strong temperature-dependent anisotropy. We are therefore tempted to suggested that Ti, Hf, Re, and Tc might also have a longitudinally polarized SDW with $\mathbf{Q}=(2\pi/c)$ (0,0,1). The fact that this **Q** is commensurate with the lattice implies that SDW effects on nuclear magnetic resonance can be avoided if the SDW phase is chosen so that nodes fall on all atomic sites.

ACKNOWLEDGMENTS

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