Laser Cooling of Transition Metal Atoms

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We propose the application of laser cooling to a number of transition-metal atoms, allowing numerous bosonic and fermionic atomic gases to be cooled to ultra-low temperatures. The non-zero electron orbital angular momentum of these atoms implies that strongly atom-state-dependent light-atom interactions occur even for light that is far-detuned from atomic transitions. At the same time, many transition-metal atoms have small magnetic dipole moments in their low-energy states, reducing the rate of dipolar-relaxation collisions. Altogether, these features provide compelling opportunities for future ultracold-atom research. Focusing on the case of atomic titanium, we identify optical transitions that are suited for laser cooling. The high total angular momentum and electron spin of this state suppresses leakage out of the nearly closed optical transition to a branching ratio estimated below $\sim 10^{-5}$. Following the pattern exemplified by titanium, we identify optical transitions that are suited for laser cooling of elements in the scandium group (Sc, Y, La), the titanium group (Ti, Zr), the vanadium group (V, Nb), the manganese group (Mn, Tc), and the iron group (Fe, Ru).

Laser cooling and the achievement of quantum degeneracy of atomic gases has led to an ever broadening range of scientific investigations and applications. This growing impact on science and technology has been fueled by the availability of quantum gases produced from an increasing number of elements, each of which has a new set of properties that can enable a new family of experiments. For example, the fortuitous collisional properties of rubidium and sodium enabled the first realizations of scalar [1, 2] and spinor [3, 5] atomic Bose-Einstein condensation. The accessible Feshbach resonances of lithium allowed studies of Efimov states [6]. Isotopes of potassium and lithium allowed the study of resonantly interacting Fermi gases [7–10]. The detectability of single metastable helium atoms on micro-channel plate detectors allowed for studies of quantum atom optics [11]. The narrow lines of alkali-earth atoms and ytterbium enabled the realization of optical lattice clocks [12, 13]. The magnetism of chromium allowed for studies of quantum ferrofluids [14], accentuated by the even stronger magnetic dipole interactions of dysprosium [15] and erbium [16]. Gaining access to a greater variety of ultracold atomic gases can, therefore, be expected to broaden the impact of ultracold atomic physics even further.

Conversely, the limitations of present-day ultracold atom systems pose limitations on the range of scientific topics that they can be used to study. As an example, consider the use of anisotropic light-atom interactions to study gases in a stable mixture of internal states. Such anisotropic interactions lead to light-induced (ac Stark) energy shifts that depend on the polarization of light and the internal angular-momentum state of the atom, and also to Raman transitions between atomic angular-momentum states; these two phenomena represent the diagonal and the off-diagonal part of the second-order light-atom interaction, respectively. State-dependent optical manipulation of ultracold atoms enables a wide variety of studies, such as the realizations of spin-orbit coupling in a quantum gas [17] and of flux lattices with topological bands [18]; the use of spin-dependent optical lattices to simulate quantum magnetism [19], generate entangled cluster states [20], and rearrange atoms within optical lattices [21]; the optical detection [22] and manipulation [23] of spin textures; and more.

The majority of available ultracold atomic species – namely the alkali metals, alkali-earth atoms, metastable noble gases, and closed-shell atoms such as chromium and ytterbium – have electronic ground states with no electronic angular momentum, $L = 0$. This property greatly simplifies their optical spectrum, so that the line strength for atomic optical transitions is spectrally concentrated, split only by fine- and hyperfine-structure effects. As a result, the interaction between these atoms and light that is far-detuned from atomic resonances is isotropic, i.e. independent of the atomic spin state and the optical polarization. A residual anisotropy is only obtained by driving the atom near its atomic resonance, whereupon the fine and hyperfine level splittings are resolved. However, tuning close to resonance increases the incoherent light scattering rate, placing an inherent limit on the coherence of spin-dependent optical interactions. This limitation is particularly acute for lighter elements, where the fine- and hyperfine-splitting of excited states is small, including, for example, $^6\text{Li}$ and $^{40}\text{K}$, the most co-
monly used isotopes for realizing resonantly interacting Fermi gases.

Atoms with a non-zero electronic angular momentum in the ground state experience strongly anisotropic light-atom interactions even for far-off-resonant light. This occurs both because the excited-state electron configurations have spectral weight over a broad energy scale because of electron-correlation energy shifts, and also because of the presence of relatively strong transitions that drive electrons out of non-s orbitals. Such conditions are realized in several lanthanide elements that have been laser cooled [24–26] and brought to quantum degeneracy [15 16]. Optical spin-orbit coupling was achieved for Dy atoms with far-off-resonant light, for which the incoherent effects of spontaneous light scattering are negligible [27]. However, the ground-level electronic angular momentum of these atoms is large: \( L = 6 \) for Dy and \( L = 5 \) for Er. Consequently, these gases also have large ground-state magnetic moments (10 and 7 Bohr magnetons for Dy and Er, respectively). The strong magnetic dipole-dipole interactions in these gases leads to strong dipolar relaxation and atom loss when the gas is placed in a mixture of spin states [28].

Here, we describe pathways for producing ultracold gases using a number of transition-metal elements, including those in the scandium group (Sc, Y, La), the titanium group (Ti, Zr, and possibly Hf), the vanadium group (V, Nb), the manganese group (Mn, Tc), and the iron group (Fe, Ru). Specifically, we find for all these elements that there is a strong, electric-dipole allowed, optical transition (see Tab. 1), with linewidth on the order of 10 MHz, which resembles the D2 line of alkali atoms in that an electron is driven from \( ns_{1/2} \) to the \( np_{3/2} \) state. The lower level on this transition, which we call the laser-cooling state, is either the atomic ground state (in Ru and Mn) or a metastable excited state (in the other cases). In all cases, these transitions are cycling, or at least very nearly so, and connect states with total angular momentum \( J \rightarrow J+1 \). As such, these transitions are suitable for standard laser cooling techniques such as Zeeman slowing [30], magneto-optical trapping [31], and polarization-gradient cooling [32, 33]. For all these elements, both the laser-cooling state and, with the exception of Mn and Tc, also the ground state have \( L \neq 0 \), offering the possibility of anisotropic light-atom interactions with far-off-resonant light. Following previous examples, we expect the addition of this new family of transition-metal elements to the rank of ultracold atomic gases will enable novel developments in ultracold atomic (and also molecular) physics.

This paper is organized as follows: in Section I we illustrate our proposed laser cooling scheme by focusing deeply on the example of atomic titanium. Through this example, we explain how the laser cooling level is identified as a highly spin-polarized configuration with a single electron in the valence \( s \) orbital. The case of titanium also exemplifies experimental techniques required to work with ultracold transition-metal atoms, and points to several potential scientific uses for quantum-degenerate transition-metal atomic gases. In Section II we describe laser cooling pathways for other atoms in the titanium group and in the iron group, which are all similar to the case of titanium. These include atomic iron, for which laser cooling has already been demonstrated [34] by the approach described in this paper. We conclude in Section III by describing briefly the opportunities for laser cooling atoms in the scandium, vanadium and manganese groups.

I. TITANIUM

A. Atomic properties

Atomic titanium (Ti) has four valence electrons surrounding a closed-shell core. In its lowest-energy term \( 1s^22s^22p^63d^24s^2 \). This level is split by fine structure into states with total angular momentum \( J = 2, 3 \) and 4, with the \( 3^3F_2 \) state being the overall ground state.

We outline several reasons why atomic titanium is an attractive choice for ultracold-atom experiments. For one, the non-zero \( (L = 3) \) orbital angular momentum of the ground state indicates that light-atom interactions remain anisotropic even for light that is far-detuned from atomic resonances. We have verified this property by computing the relative strengths \( \alpha_S(\omega), \alpha_V(\omega), \) and \( \alpha_T(\omega) \) of the scalar, vector, and tensor portions of the ground-state ac Stark shift, respectively, as a function of the optical frequency \( \omega \). Here, we express the total ac Stark shift \( \alpha_{mJ}(\omega) \), for an atom in the \( m_J \) magnetic sublevel defined with respect to a quantization axis \( \mathbf{b} \), as

\[
\alpha_{mJ}(\omega) = \alpha_S(\omega) + (\mathbf{k} \cdot \mathbf{b}) A \frac{m_J}{2J} \alpha_V(\omega) + \frac{1}{2} (3\epsilon \cdot \mathbf{b})^2 - 1 \frac{3m_J^2 - J(J+1)}{J(2J-1)} \alpha_T(\omega). \tag{1}
\]

In this expression, \( \mathbf{k} \) is the unit wavevector and \( A \) the helicity of the light field, and \( \epsilon \) is the optical polarization vector.

We calculate the ac Stark shift by taking into account all electric dipole optical transitions identified in the NIST Spectral Database [36] for neutral titanium, using the reported oscillator strengths for all lines for which such strengths are provided. In addition, there are several excited fine structure levels for which an oscillator strength is not directly reported, but for which we can estimate such strength based on that reported for a different level of the same configuration and term [37]. As an example, the \( a^3F_2 \rightarrow w^3D_1 \) and \( a^3F_2 \rightarrow w^3D_2 \) transitions have reported line strengths of 0.71 a.u. and 0.051 a.u. respectively, while the line strength of the \( a^3F_2 \rightarrow w^3D_3 \) transition is not reported. Using the results for LS coupling, we estimate that the unobserved
FIG. 1. (a) Vector(Re $\alpha_V$) and (b) tensor polarizability (Re $\alpha_T$) for Ti (solid black), Er (dashed red), and $^{40}$K (dotted blue) in their absolute ground states, scaled by the scalar polarizability Re $\alpha_S$ and plotted as a function of the wavenumber and wavelength of applied light. Arrows indicate all transitions to an excited 3$d^2$4$s$ state in Ti with an oscillator strength greater than 0.01. The data for Er, reproduced from Ref. [29], showing the dense spectrum above 20000 cm$^{-1}$ is omitted for clarity. (c) The figure of merit $\mathcal{M} = \Omega_R/\Gamma_{sp}$ is plotted over the narrow wavelength range indicated by a gray band in (a, b), with detuning measured relative to the $a^2F_2 \rightarrow z^2F_2$ transition. Solid horizontal lines indicate the maximal $\mathcal{M}$ achievable in the Raman coupling scheme for each element, while the vertical line indicates the corresponding wavelength ($\sim$ 522 nm) in Ti.

The anisotropy of light-atom interactions is characterized by the ratios Re($\alpha_V$)/Re($\alpha_S$) and Re($\alpha_T$)/Re($\alpha_S$). Figure 1 shows these calculated ratios for Ti, and compares them to similar ratios calculated for potassium (K) and erbium (Er). As expected, in the long-wavelength limit, the vector and tensor Stark shifts are strongly suppressed for the alkali-metal potassium. In contrast, for both titanium and for erbium, significantly anisotropic interactions are seen in wide parts of the optical spectrum.

A comparison between the optical response of Ti and Ca is illuminating since both atoms have a filled 4$s^2$ electron configuration in the ground state. The optical response of calcium in its ground state is dominated by the 4$s^2 \rightarrow 4s4p(1P^1)$ transition at 432 nm wavelength, with linewidth of $2\pi \times 34.7$ MHz. For titanium, transitions that similarly drive from the 3$d^2$4$s$ ground-state configuration to the 3$d^2$4$s$4$p(1P^1)$ excited-state configuration are also strong. However, in titanium, this excited-state configuration is split into numerous terms, several of which are labeled in Fig. 1 with the optical coupling strength to each term depending on the optical polarization and the angular momentum state of the $a^2F_2$ ground-state atom. The excited energy terms are split by large electron-correlation energies, leading to large differential detunings for long-wavelength light from each of the relevant excited states. Altogether, the state-dependent coupling and the large energy differences of these many states results in a strongly anisotropic light-atom interaction.

An additional comparison between the optical response of Ti and K is valuable as well, since both elements possess fermionic isotopes and are thus suited to studies of fermionic quantum matter. In K, two levels contribute to the response, the D1 line ($4s^2S_{1/2} \rightarrow 4p^2P_{1/2}$) at 770.1 nm wavelength and linewidth $2\pi \times 6.01$ MHz, and the D2 line ($4s^2S_{1/2} \rightarrow 4p^2P_{3/2}$) at 766.7 nm wavelength and linewidth $2\pi \times 5.94$ MHz. The small differential detuning between these fine structure lines ($\sim$ 1 THz) limits the anisotropic vector light shift. In Ti, it is not fine structure splitting that leads to differential detuning, but rather the energy splitting associated with electron-
electron correlation (~100 THz). This large differential detuning leads to much larger anisotropic light shifts in Ti relative to K.

An important figure of merit for cold-atom experiments that make use of anisotropic light-atom interactions is the ratio $M = \Omega_R/\Gamma_{sp}$ between the coherent and the incoherent parts of the interaction. Here, $\Omega_R$ is the Rabi frequency for Raman transitions between different angular-momentum states within the electronic ground state, or, equivalently, the vector or tensor ac Stark shift divided by $\hbar$, while $\Gamma_{sp}$ is the total spontaneous emission rate. Highly coherent (large $M$) interactions can be achieved for light that is far red-detuned from all optical transitions. Denoting $M_{VT}$ for the vector and tensor coupling figures of merit respectively, in the far red-detuned regime, we estimate the average figure of merit as $M_{VT} \approx \Re \alpha_{VT}/|\Im \alpha_{S}|$. For example, considering light at a wavelength of 1 $\mu$m, as commonly used in optical-lattice experiments, we expect $M_{VT} \sim 4 \times 10^5$, $M_T \sim 5 \times 10^6$ for Ti, compared to $M_{VT} \sim 4 \times 10^3$, $M_T \sim 1 \times 10^6$ for Er, and $M_{VT} \sim 1.5 \times 10^5$, $M_T \sim 1.5 \times 10^6$ for $^{40}$K.

Alternately, highly coherent light-atom interactions can be found for light at frequencies that are closer to specific atomic resonances. For example, Refs. [39] consider the use of narrow-line excitations to achieve anisotropic light-atom interactions in Er. Considering scattering just from the narrow-line resonance itself, the ratio $M$ generally increases with detuning $\Delta$ between the optical and resonance frequencies, with $\Omega_R \propto \Delta^{-1}$ and $\Gamma_{sp} \propto \Delta^{-2}$. However, at large enough $\Delta$, spontaneous emission from all other atomic resonances becomes dominant, establishing an upper limit of $M$. Near a particular atomic resonance, the figure of merit can be approximated as

$$M \approx \frac{2\Delta_j}{\Gamma_j + \sum_{j' \neq j} \Gamma_{j'} \frac{\Delta_j \Delta_j}{\Delta_{j'}}} \quad (2)$$

where $\Gamma_j$ is the optical detuning from the particular transition chosen, and $\Gamma_j$ is summed over all other transitions. For Er, this expression gives a maximum of $M \sim 8 \times 10^6$. A similar approach appears possible in atomic Ti. For example, examining the spectrum of Ti, we find a similar figure of merit, $M \sim 6 \times 10^6$ for generating state-dependent optical interactions with light that is detuned from the transition between the atomic ground state and the $3d^2(3F)4s4p(3P^2)z^2F_2$ excited state, with wavelength 518 nm. It is notable that this highly coherent, anisotropic light-atom interaction is achieved with light near the 530 nm wavelength range that is easily produced at high power by frequency-doubled laser systems. Second, unlike for the ground states of lanthanide atoms, the $a^3F_2$ ground state of Ti is relatively stable against inelastic magnetic-dipolar relaxation. The rate of dipolar relaxation induced by the long-range dipole-dipole interaction in ultracold gases, as considered in Refs. [41][43], scales (for the magnetic dipole - magnetic dipole case) as the square of the magnetic moment $\mu = g_J J \mu_B$, with $g_J$ being the Landé $g$-factor, $J$ being the atomic total angular momentum, and $\mu_B$ being the Bohr magneton. Magnetic dipolar relaxation in highly magnetic atoms such as Dy [28] and Cr [43], with magnetic moments of $\mu = 10$ and 6 $\mu_B$, respectively, is rapid, leading to spin-polarization lifetimes on the order of 1 second or below under common experimental gas densities ($\sim 10^{13}$ cm$^{-3}$) and applied magnetic fields ($\sim 1$ G). In contrast, for alkali atoms, with magnetic moments of 1 $\mu_B$ and below, the minutes-long stability of magnetically trapped atoms indicates the dipolar relaxation rate to be very small.

For Ti, the $J = 2, 3$, and 4 ground states have magnetic moments of $g_J J \mu_B = 4/3, 13/4$, and 5 $\mu_B$, respectively. Since the magnetic moment of the overall ($J = 2$) ground state is similar in magnitude to that of an alkali atom, we expect dipolar relaxation in a $a^3F_2$ Ti gas to be very slow. Additional inelastic decay paths could be opened by interactions between the open d shells of the colliding atoms. However, several studies, e.g. theoretical studies on Sc [44] and experiments on buffer-gas-cooled Ti gases [45], indicate that such inelastic processes are suppressed by the fact that the 3d shell is submerged below the closed 4s shell. Altogether, collisions among ground-state Ti atoms should be predominantly spin-preserving, allowing, for example, for magnetic trapping in weak-field seeking states and for studies of spinor gases with conserved magnetization.

Finally, Ti is endowed with several stable isotopes. These include three bosonic isotopes, $^{46}$, $^{48}$, $^{50}$Ti, with relatively large natural abundance (8%, 74%, and 5%, respectively). All three bosonic isotopes have nuclear spin $I = 0$, leading to a simple atomic level structure that is free of hyperfine structure. As quantum degenerate gases, these three isotopes, in their $a^3F_2$ ground state, may present examples of $J = 2$ spinor Bose-Einstein condensates, which are predicted to manifest three different types of magnetic ordering: ferromagnetic, nematic and tetragonal [47][49]. The choice of ground state depends on the relative strengths of three s-wave scattering lengths that characterize the low-energy collisions between $J = 2$ bosons. Both the nematic and tetragonal phases, owing to their discrete rotational symmetries, support structures such as fractional superfluid vortices [50][52], whose non-abelian character can lead to complex vortex collision and rung dynamics [53].

Ti also has two stable fermionic isotopes, $^{47}$, $^{49}$Ti, with nuclear spins $I = 5/2$ and 7/2, and abundances of 7% and 5%, respectively. Given the strong anisotropy of light-atom interactions, and the stability of spin mixtures against dipolar relaxation, these isotopes could be used to study Cooper pairing in spin-orbit coupled Fermi gases. Such systems, which are under investigation also within solid-state devices, are predicted to support topological superfluidity and Majorana fermions [53][56], with relevance to topological quantum computing [57][59].
B. Laser cooling scheme

Many successful realizations of laser cooling make use of atomic transitions that are approximately closed, driving an atom from a low-energy state of angular momentum \( J \) to an excited state of angular momentum \( J + 1 \). The closed nature of the transition ensures that an atom will scatter the large number of photons (10^4 or greater) required to reduce its velocity through the optical scattering force, returning always to the laser-cooling state. The \( J \rightarrow J + 1 \) nature of the transition provides for optical pumping of the atom into a brightly scattering state and enables techniques such as magneto-optical trapping and polarization-gradient cooling.

There is no \( J \rightarrow J + 1 \) transition from the \( a^3F \) ground term of titanium that satisfies these criteria. The optical spectrum of this ground term, dominated by excitation of one of the \( 4s^2 \) electrons of the ground configuration, resembles that of calcium. As in the case of calcium, laser cooling via one of these transitions is unfavorable since there is significant leakage from the excited states to other even-parity metastable states.

Instead, a suitable laser-cooling transition is found among the family of lines that arise after exciting an electron to a metastable alkali-like configuration \( 3d^54s^1 \). The lowest energy level of this metastable configuration, the \( a^5F \) level, is split via fine structure into \( J = 1 \) to \( J = 5 \) states. While the lifetime of these states has not been measured, the two dominant decay mechanisms, magnetic dipole radiation between fine structure states and spin-forbidden electric quadrupole decay to the ground state, are weak transitions. Comparing to similar atoms, such as Fe, we expect the radiative lifetime of these states to be on the order of 10-100 s. The highest-\( J \) state, labeled \( a^5F_5 \) and with an energy of 6843 cm\(^{-1}\) above the ground state, has a strong \( J \rightarrow J + 1 \) transition to the odd-parity \( y^5G_6^0 \) state (at energy 29611 cm\(^{-1}\)). On this transition, at a wavelength of 498.3 nm, the valence \( 4s_{1/2} \) electron is excited to the \( 4p_{3/2} \) level. The remaining valence electrons remain in the spin polarized \( 3d^5(4F_{9/2}) \) configuration. As such, the transition resembles the D2 \( 4s \rightarrow 4p \) transition in potassium if we regard Ti’s \( 3d^5 \) valence electrons as part of the inert atomic core.

The linewidth of this transition is comparable to alkali atom cooling transitions with \( \Gamma = 2\pi \times 10.5 \text{ MHz} \), with a corresponding Doppler cooling limit of \( T_D = 250 \text{ µK} \). This line is amenable to all the standard tricks of laser-cooling: Zeeman slowing, magneto-optical trapping, and polarization-gradient cooling. Furthermore, the line is sufficiently broad to provide a strong cooling force.

This transition appears to be a cycling transition because of the spin-polarized nature of the \( 3d^5 \) electrons and the maximal \( J = 5 \) value of the total electronic angular momentum. Electric dipole transitions could, in principle, lead from the odd-parity excited state to four other even-parity states: the \( a^5G_5 \) state (at energy of 15220 cm\(^{-1}\)), the \( a^3H_{5,6} \) states (18141 cm\(^{-1}\) and 18192 cm\(^{-1}\), respectively), and the \( a^1H_5 \) state (20796 cm\(^{-1}\)). However, these transitions are all spin forbidden, requiring a reconfiguration of the \( 3d^5 \) valence electrons. Focusing on the leakage into states with \( J = 5 \), we estimate the residual branching ratio for spontaneous emission into these states by considering the degree to which a spin-orbit interaction term would mix these states with the \( a^5F_5 \) state. We quantify the strength of the spin-orbit mixing energy from the fine-structure splitting of the \( a^3F_5 \) level, and then apply second-order perturbation theory to estimate the branching ratio as \( \delta = E_{a^5F_5} - E_{a^5F_1} \) is the fine structure splitting and \( \Delta = E_5 - E_{a^5F_5} \) is the difference in energy between a given leakage state \( i \) and the \( a^5F_5 \) state. Under these assumptions, we expect that the leakage from the cooling transition to be around \( 3 \times 10^{-5} \), low enough to perform laser cooling and trapping without the need for constant repumping lasers.

The \( a^5F_5 \) state supports an additional cycling transition. This transition to the \( z^5G_6^0 \) state has an optical wavelength of 1040 nm and a linewidth of \( 2\pi \times 11.3 \text{ kHz} \), giving a Doppler temperature of \( T_D = 0.27 \text{ µK} \). As has been demonstrated with Sr, Ca, Yb, Dy, and Er, a second-stage magneto-optical trap using this narrower line could significantly increase the phase space density of the laser cooled gas. Furthermore, there are even fewer leakage channels for emission out of the \( z^5G_6^0 \) state than out of the \( y^5G_6^0 \) state, increasing the number of photons that could be scattered without the need for repumping. By the same method of estimation, we expect the leakage from this transition to be around \( 1 \times 10^{-7} \).

Producing an atomic beam of Ti through direct sublimation requires a Ti source to be heated to temperatures upwards of 1500 K. At this high temperature, almost 0.1% of the sublimated atoms will be thermally excited into the metastable laser-cooling state. A higher population in the laser-cooling state can be achieved by optical pumping of ground-level atoms. Specifically, we identify three promising schemes for transferring atoms from the \( a^3F_2 \) state into the \( a^5F_5 \) state: one-color optical pumping via the \( z^5D_4^1 \) state (wavelength \( \lambda = 546.20 \text{ nm} \), linewidth \( \Gamma = 2\pi \times 0.3 \text{ MHz} \), transfer efficiency \( \eta = 0.88 \)), one-color optical pumping via the \( y^5G_5^0 \) state (\( \lambda = 350.76 \text{ nm} \), \( \Gamma = 2\pi \times 16 \text{ MHz} \), \( \eta = 0.9 \)), or two-color pumping via the \( y^5G_5^0 \) state (\( \lambda = 378.99 \text{ nm} \), \( \Gamma = 2\pi \times 12.7 \text{ MHz} \)) and \( y^5F_5^0 \) state (\( \lambda = 451.40 \text{ nm} \), \( \Gamma = 2\pi \times 15.7 \text{ MHz} \)). In the two-color scheme, the first transition pumps the majority of the atoms into the \( a^3F_4 \) state, which is then pumped by the second transition to the laser coolable \( a^5F_5 \) state, for a total efficiency of \( \eta = 0.995 \). There are multiple routes to transfer atoms from the \( a^3F_4 \) state to the \( a^5F_2 \) ground state; one simple path would be to drive the \( a^5F_5 \rightarrow z^3F_3 \) transition at 758 nm, which transfers atoms to the \( a^1F_3 \) and \( a^3F_3 \) states. Simultaneously driving the \( a^3F_4 \rightarrow y^3F_3^0 \) and \( a^5F_3 \rightarrow y^5F_2^0 \) transitions at the wavelengths of 403 nm and 401 nm leads to a total transfer efficiency of \( \eta = 0.814 \) for transferring \( a^5F_5 \) atoms to the \( a^3F_2 \) ground state. More efficient schemes
FIG. 2. Energy levels of titanium group elements Ti and Zr. Here, and in subsequent energy-level figures, black (red) lines indicate even (odd) parity states, and thicker lines highlight states relevant for laser cooling. Levels are arranged by total electronic angular momentum $J$ and ordered by energy. (a) Shows a simplified energy level diagram of Ti, highlighting the important levels and transitions involved in the entire laser cooling scheme. The $a^3F_J$ fine structure ground levels and $a^5F_5$ metastable laser-cooling level are the relevant even parity states. The $z^5D_4$ level is one of several intermediate states that could be used to optically pump atoms into the $a^3F_5$ state, with the $546 \text{ nm}$ transition indicated as well as the dominant decay path. The two excited levels for laser cooling, the $z^5G_6^e$ and $y^5G_6^e$, are also identified and can support type I MOTs with transitions at 1040 nm (red) and 498 nm (blue) respectively. (b) A more complete diagram of Ti levels is given, including the full configuration and term symbol for the ground states and laser cooling excited states. For simplicity, optical pumping transitions are suppressed. All levels below the $y^5G_6^e$ state are shown. (c) Zr level structure. All levels below the $y^5G_6^e$ are shown. The laser-cooling transitions, analogous to those in Ti, are shown as well.

come at the cost of greater experimental complexity.

II. TITANIUM AND IRON GROUP ATOMS

A. Titanium group

Other elements in the titanium group, zirconium (Zr) and hafnium (Hf), have similar atomic structures and spectra to Ti. There are two low-energy, even-parity electronic configurations, in which the four valence electrons reside in either an alkali-earth-like configuration of $(n - 1)d^2 ns^2$ or an alkali-like configuration $(n - 1)d^3 ns^1$ where $n$ is the principal quantum number of the valence shell ($n = 4, 5, 6$). The ground level has the configuration $(n - 1)d^2 ns^2$ and term symbol $a^3F_2$, with fine-structure splitting to other $J$ levels on the order of 100 cm$^{-1}$. The lowest-energy state with the $(n - 1)d^3 ns^1$ electron configuration has a term symbol $a^5F$ and the spin-polarized laser-cooling state is identified as the highest fine-structure state with $J = 5$.

a. Zirconium: The level structure of Zr is shown in Fig. 2. The laser-cooling state lies $5889 \text{ cm}^{-1}$ above the ground state. A strong laser-cooling transition to the $y^5G_6^e$ state lies at a wavelength of 468.9 nm. An additional closed cooling transition, to the $z^5G_6$ state, occurs at the wavelength 901.8 nm. Line widths for these two transitions have not been measured. However, one expects linewidths similar to those found in Ti, i.e. that the blue-light transition, which involves a $(n - 1)d^3 ns \rightarrow (n - 1)d^3 np$ electron configuration change, will have a linewidth in the range of $2\pi \times 10 \text{ MHz}$, while the red transition, which involves a $(n - 1)d^3 ns \rightarrow (n - 1)d^2 ns np$ electron configuration change, will be substantially narrower. Given the increased spin-orbit coupling of Zr, leakages from the excited states are expected be greater than in Ti. We expect the leakage for the broad (narrow) transition to be around $1 \times 10^{-4}$ ($1 \times 10^{-5}$). Zr possesses six stable or long-lived naturally abundant isotopes: four bosonic isotopes (90$^{90}, 92$,$^{92}, 94$,$^{94}, 96$,$^{96}$Zr) with nuclear spin $I = 0$, and two fermionic isotope (91$^{91}, 93$Zr) with $I = 5/2$.

b. Hafnium: The spectrum of Hf is too poorly known to determine whether the analogous laser-cooling transition exists below the first ionization limit (55047.9 cm$^{-1}$). However, there is a metastable state that should be long lived and might support laser cooling: the $5d^3(4F)6s a^3F_5$ state with an energy of 17901.28 cm$^{-1}$ above the ground state.

B. Iron group

The elements in the iron group, iron (Fe), ruthenium (Ru), and osmium (Os), also have two low energy even parity electronic configurations: an alkali-earth-like valence configuration $(n - 1)d^6 ns^2$ and the alkali-like valence configuration $(n - 1)d^7 ns^1$. As in the titanium group, a cycling transition exists on the maximum-$J$ level of the lowest-energy alkali-like level, also given by the term symbol $a^5F_5$. However, because the va-
The laser-cooling transition at 358 nm wavelength drives the atom to the $z^5G_6$ excited state. Laser cooling on this transition has been demonstrated. Indicated by the dashed gray arrow is the decay of the $a^2F_3$ state in Fe, predicted to occur at a rate of $2 \times 10^{-3}$ s$^{-1}$. In Ru, the $a^2F_3$ laser-cooling state is also the overall ground state, eliminating the need for optical pumping into a metastable state. The 350 nm wavelength laser-cooling transition, connected to the $z^5G_6$ state is indicated.

The laser cooling of iron (Fe) has been realized by the Bastin group. The ground level of Fe has an electron configuration of $3d^64s^2$ and term symbol $a^2D_J$, with total angular momenta $J$ ranging from $J = 0$ to $J = 4$. The overall ground state, $a^2D_1$, has a large magnetic moment and is expected to undergo significant magnetic dipole-dipole relaxation. The metastable cooling state $3d^7(4F)4s$ $a^5F_5$, with an energy of 6928 cm$^{-1}$, has an estimated lifetime of 300 - 1000 s, allowing ample time for cooling and trapping of Fe atoms. This state has a closed transition to the excited state $3d^7(4F)4p$ $z^5G_6^*$ with a wavelength of 358 nm and a linewidth of $2\pi \times 16.2$ MHz. Similar to Ti, it is possible to increase the population of atoms in the metastable cooling state via optical pumping; in Ref. 34, the 372-nm-wavelength transition to the $z^5F_5$ state is used. While not reported in the literature, we expect the leakage from the cooling transition to be around $4 \times 10^{-5}$.

Ruthenium (Ru) may be a surprisingly convenient choice for laser cooling. For this atom, the laser-cooling state $a^5F_5$ is indeed the overall ground state, obviating the need for optical pumping to drive atoms into the laser-cooling state. Laser cooling would proceed directly through illumination at a wavelength of 350 nm nearly resonant with the $z^5G_6^*$ excited state. The leakage from this transition is expected to be around $7 \times 10^{-4}$. In other words, laser cooling Ru may be as straightforward as laser cooling an alkali atom.

Ru has several additional features of interest. First, the spectrum of Ru should contain a narrow-line electric-quadrupole transition from the metastable even-parity $4d^7(a^4F)5s$ $a^5F_1$ state up to the low-lying $4d^95s^2$ $a^4D_0$ excited state. This transition frequency should be insensitive at first order to applied magnetic fields. Moreover, light at the transition wavelength of 1566 nm could be transmitted at low loss through optical fibers. Altogether, this transition may offer advantages as an optical clock standard. Second, Ru has a large number of stable isotopes, including five bosonic isotopes (96, 98, 100, 102, 104) Ru with a nuclear spin $I = 0$. The isotope shifts on the various narrow-line transitions in Ru could be leveraged to search for forces outside the Standard Model.

As with Hf, the spectrum of Os is not known well enough to determine whether there is a closed laser-cooling transition below the first ionization limit. However, the $5d^7(4F)6s$ $a^5F_5$, metastable state (energy of 5144 cm$^{-1}$) would likely have a long lifetime and could support laser cooling.

III. SCANDIUM, VANADIUM AND MANGANESE GROUP ATOMS

The general method of cooling gases of transition metals proposed herein – employing a long-lived alkali-like metastable state with configuration $(n-1)d^2ns^1$ consisting of a polarized submerged shell core and a single valence $s$ electron – is applicable also to several other transition-metal atoms. Table I gives the relevant properties for the metals of the scandium, vanadium and manganese group, elements that we have additionally identified as amenable to laser cooling. We discuss these cases briefly below.

A. Scandium group

Atoms in the scandium group – Sc, Y, and La – have a ground state characterized by two valence $s$-electrons ($(n-1)d^1ns^2$ $a^2D_{3/2}$, where $n$ is the principal quantum number of the valence shell ($n = 3,4,5,6$)) and a spin-polarized metastable alkali-like configuration ($(n-1)d^2ns^1$ $a^4F_I$). The laser-cooling state is given by the highest fine-structure level $J = 9/2$ of this metastable state from which a strong, closed $J + 1$ transition exists up to an odd parity excited state $z^5G_{11/2}^*$ (see Fig. 4 for atomic level structures).

b. Scandium: In scandium (Sc), the cooling scheme proceeds without any mitigating factors. The laser-cooling transition occurs at 567.3402 nm with a
### TABLE I. Laser cooling lines of transition metals. Configurations, terms, and essential cooling line information is summarized. Values with a (*) indicate that the wavelength has been inferred from energy level data, not measured directly. All values are drawn from the NIST database [36].

| Atom | Ground state config. | term | Metastable state config. | term | Excited state config. | term | Cooling trans. (nm) | Linewidth (MHz/2π) |
|------|----------------------|------|--------------------------|------|----------------------|------|--------------------|--------------------|
| Sc   | 3d4s2                | a2D3/2 | 3d2(3F)4s                | a4F9/2 | 3d2(3F)4p            | z4G11/2 | 547                | 10.0               |
| Y    | 4d5s2                | a2D3/2 | 4d2(3F)5s                | a4F9/2 | 4d2(3F)5p            | z4G11/2 | 547                | 10.0               |
| La   | 5d6s2                | a2D3/2 | 5d2(3F)6s                | a4F9/2 | 5d2(3F)6p            | G11/2 | 625                | 5.86               |
| Ti   | 3d4s2                | a2F4  | 3d2(3F)4s                | a5F5  | 3d2(3F)4p            | y5G6  | 498                | 10.5               |
| Zr   | 4d5s2                | a2F4  | 4d2(3F)5s                | a5F5  | 4d2(3F)5p            | y5G6  | 467*               | —                  |
| V    | 3d4s2                | a2F3/2 | 3d2(3H)4s                | a5H13/2 | 3d2(3H)4p            | z4I15/2 | 445                | 13.0               |
| Nb   | 4d4(4D)5s            | a6D1/2 | 4d4(3H)5s                | a5H13/2 | 4d4(3H)5p            | z4I15/2 | 463*               | —                  |
| Mn   | 3d4s2                | a6S5/2 | —                         | —                  | 3d4(6S)4s4p(3P) | z6P7/2 | 403                | 2.71               |
| Tc   | 4d5s2                | a6S5/2 | 4d4(3H)5s                | a5H13/2 | 4d4(3H)5p            | z4I15/2 | 405*               | —                  |
| Fe   | 3d4s2                | a5D4  | 3d2(4F)4s                | a5F5  | 3d2(4F)4p            | z5G6  | 358                | 16.2               |
| Ru   | 4d4(4F)5s            | a5F5  | —                         | —                  | 4d4(4F)5p            | z5G6  | 350                | 13.7               |

FIG. 4. Energy levels of scandium group elements (a) Sc, (b) Y, and (c) La. The a2D3/2 ground term and a4F9/2 metastable laser-cooling state are highlighted in all diagrams. For Sc, the laser-cooling transition at 549 nm wavelength connects to the z4G11/2 state. In Y, the analogous transition occurs at wavelength 547 nm; also indicated is the narrow laser-cooling transition for the ground state at wavelength 950 nm. For La, two laser-cooling states at wavelengths 625 nm and 406 nm connect to two z4G11/2 states. The grey dashed arrows indicated two potential spin-allowed leakage paths that may require repumping.

The linewidth of 2π × 8.6 MHz [71]. Sc possesses one stable isotope, bosonic 45Sc, with nuclear spin I = 7/2.

b. Yttrium: In Y, there is the laser cooling line from a metastable state as described above, with a wavelength of 546.7986 nm [72] and a linewidth of 2π × 10.0 MHz [73]. Yttrium possesses one stable isotope, bosonic 89Y, with nuclear spin I = 1/2.

A second transition may also be considered, between the overall ground state 4d5s2 a2D3/2 and the lowest odd-parity excited state, 5s25p z2P1/2. As a J → J – 1 transition, this excitation would be suited to a type II magneto-optical trap [74]. While the transition itself has not yet been observed, its wavelength of 949.7429 nm is known [75]. There are no possible leakage channels for
this transition, allowing a clear cycling transition to be used. It would likely be a relatively narrow E1 transition, since it involves the transition of the inner shell 4d electron to the 5p state.

c. Lanthanum: Lanthanum (La) returns to the pattern of only being coolable from a metastable state. However, it has two possible cycling transitions available in this state. One laser-cooling transition connects the metastable 5d 2 3F6s 4F9/2 and the 5d 2 3F6p 4G11/2 state, occurring at 625.166 nm with a linewidth of 2π × 5.86 MHz. A second transition connects the metastable state to the excited state 4f5d 2 3G7/2 6s 4G11/2, with wavelength 406.148 nm and linewidth 2π × 8.9 MHz. Lanthanum possesses one stable isotope, bosonic 139La, with a nuclear spin I = 7/2. However, fermionic 138La is long lived, with a half-life of 1.02 × 10^{11} yr, which could allow an isotopically enriched sample of fermionic atoms to be studied. 138La has a nuclear spin I = 5.

Lanthanum also has a difficulty not present in the other elements that have been discussed: from both of the excited states that could be addressed for laser cooling, there is a spin-allowed electric dipole transition to the 5d 4 4F9/2 state. While decay to this state has not been observed, it could be a stronger source of leakage than in the other examples considered thus far. If these leakage channels are significant, they could be repumped to replenish the atoms in the metastable cooling state.

**B. Vanadium group**

In both vanadium (V) and niobium (Nb), the alkali-like, even-parity state with stretched J, the (n – 1)d 2 ns 1 a 4H13/2 state, is viable for laser cooling. In both atoms, it connects to a cycling transition with the (n – 1)d 2 ns 1 z 4I15/2 state.

a. Vanadium: For V, the laser-cooling transition occurs at wavelength 445 nm with a linewidth of Γ = 2π × 13 MHz. There is only one other J = 13/2 even parity state where leakage from this cycling transition could occur, we estimate the leakage to be 2 × 10^{-6}. The radiative lifetime of the a 4H13/2 state has been calculated to be ~ 1.2 s, occurring via electric quadrupole decay to the a 4F9/2 state.

b. Niobium: For Nb the laser-cooling transition occurs at wavelength 463 nm. The linewidth is not known. However, there are no other J = 13/2, even parity states in Nb available for leakage. Thus, we expect the cycling transition, once observed, will be fully closed with respect to electric dipole decays. However unobserved even parity states could in principle introduce leakage channels not listed in atomic structure databases. In this case, the electric quadrupole decay would be the dominant single particle loss mechanism for trapped atoms.

**C. Manganese group**

Elements of the manganese group, manganese (Mn) and technetium (Tc), have ground state electronic configurations (n – 1)d 2 ns 2 and term a 6S5/2 (Fig. 6). The stretched J, alkali-like metastable state potentially suitable for laser cooling is the (n – 1)d 6 ns a 4H13/2 state.

a. Manganese: The laser-cooling transition of the metastable state in Mn occurs at a wavelength of 406

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**Figure 5.** Energy levels of vanadium group elements (a) V and (b) Nb. The fine structure ground states (a 4F for V, a 4D for Nb), metastable laser-cooling state a 4H13/2, and z 4I15/2 state are indicated. The laser-cooling transitions (wavelengths 445 nm for V, 463 nm for Nb) are also indicated. In dashed gray is indicated the dominant decay path of the metastable state of Nb atoms, an E2 transition with A_{nl} ∼ 2π × 130 mHz.

**Figure 6.** Energy levels of manganese group elements (a) Mn and (b) Tc. Both Mn and Tc possess a viable cycling transition (wavelengths 406 nm and 378 nm) between the metastable a 4H13/2 state and the z 4I15/2 state. Mn is also likely laser-coolable directly in the ground state. The three possible laser-cooling transitions are at a wavelength of 403 nm and connect the a 6S5/2 ground state to the z 4F_{J} states.
nm. Its linewidth has not been measured. We expect this transition to be nearly closed, with an estimated leakage of $2 \times 10^{-5}$. Mn presents a unique challenge for optical pumping due the high energy of the metastable $a^4H_{13/2}$ state with respect to the $3d^5 4s^2 a^6S_{5/2}$ ground state ($34139$ cm$^{-1}$).

However, this ground state may be interesting in its own right, as it possesses several nearly closed electronic transitions with wavelengths near 403 nm that connect to the $3d^5 (6S) 4s 4p(3P') z^0P_z$ states, while these states likely have spin-allowed leakage channels, the low frequency of these leakage transitions, and the fact that these leakages involve inner electron transitions, may imply that the branching ratio for leakage is very low.

b. Technetium: Unlike in Mn, there is not a closed transition in the ground state of Tc. However the metastable cooling state $(4d^8(3H) 5s 5^1H_{13/2})$ is of much lower energy ($16553$ cm$^{-1}$) than in Mn, and is thus likely easier to access. The laser-cooling transition from this state connects to the $4d^8(3H) 5p 4^3I_{15/2}$ state and occurs at the wavelength 378 nm. Its linewidth has not been reported. Tc is a radioactive element that is not naturally occurring, however bosonic $^{97}$Tc and fermionic $^{98}$Tc have lifetimes over $10^6$ years, while the bosonic $^{99}$Tc - commonly used in medicine - has a lifetime over $10^6$ years.

IV. CONCLUSION

By examining the structure of ground and metastable states of transition-metal atoms, we have identified a number of optical transitions that resemble the principal atomic transitions of alkali atoms, in that a single s-orbital valence electron is excited to the p-orbital at the same principal quantum number. Thus, these transition-metal optical transitions appear suited for laser cooling, i.e. they have large oscillator strength, are nearly cycling with little leakage to additional states, and have the right $J \rightarrow J+1$ angular momentum structure to support many standard laser-cooling techniques. We focus most of our attention on the example of atomic titanium, given that the level structure for this atom is well characterized, that the atom has numerous abundant bosonic and fermionic isotopes, and that its ground state presents attractive features for ultracold atomic physics experiments. The example set by titanium allows us to sift through the level structures of ten other transition-metal elements, and to identify laser-cooling transitions for each.

There remain many potential pitfalls in applying laser cooling to these atomic elements. For example, atoms in metastable states may be subject to strong collisional decay, including light-induced decay in the presence of laser-cooling and optical-pumping light. Even if laser cooling is successful, further cooling toward quantum degeneracy may require multi-step optical pumping to the electronic ground state. For some elements, inelastic decay channels may be unexpectedly high. Nevertheless, with such a large selection of elements (11) and of stable (26 total) and long-lived (9 total with $> 10^6$ yr half-life) isotopes potentially made available for ultracold atomic physics experiments, the investment of efforts to realize laser-cooling of these elements is warranted.

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