Measurement of the Zeeman effect in an atomic anion: Prospects for laser cooling of Os^-

A. Kellerbauer,* A. Fischer,† and U. Warring‡
Max-Planck-Institut für Kernphysik, Saupfercheckweg 1, 69117 Heidelberg, Germany
(Received 29 November 2013; published 29 April 2014)

The negative osmium ion Os^- is one of very few atomic anions potentially suitable for laser cooling. We have made a measurement of the Zeeman splitting of a bound-bound transition in $^{192}Os^-$ by studying the laser excitation from the $5d^76s^2\,{}^4F^e_{9/2}$ ground to the $5d^66s^26p\,{}^6D^o_{9/2}$ excited state in a homogeneous external magnetic field. The experimental Landé factors $g_J = 1.31(7)$ and $g_J = 1.50(8)$, respectively, agree well with calculated values. Both levels are found to split into 10 Zeeman sublevels, resulting in 28 allowed transitions of different relative intensities, in agreement with calculations based on pure and composite LS states. In view of the experimental results, the prospects for laser cooling of Os^- are discussed.

DOI: 10.1103/PhysRevA.89.043430 PACS number(s): 37.10.Rs, 32.60.+i, 32.80.Gc, 32.30.Bv

I. INTRODUCTION

Atomic and molecular anions play an important role in many fields of physics and chemistry, ranging from plasma physics [1] to microelectronics [2] and atmospheric science [3]. Negative ion sources are indispensable components of many types of particle accelerators [4]. Owing to their unique structure, anions are also studied as model systems to better understand the importance of correlation effects and to test atomic theory [5,6]. Finally, interest in negative ions has surged in the past few years following the observation of molecular anions in the interstellar medium [7]. Despite their importance, it is currently impossible to study negative ions at ultracold temperatures. Until now anions confined in electromagnetic traps may only be cooled to the temperature of the surrounding environment, typically a few kelvins, using electron cooling, buffer gas cooling, or resistive cooling [8–10].

To resolve this shortcoming, a technique for the production of ultracold negative ions has been proposed [11]. The method is based on the laser cooling of an atomic anion, which can in turn be used to sympathetically cool any other negative ion species which is simultaneously confined in the same ion trap. In this way, negative ions can in principle be cooled to microkelvin temperatures or below, depending on the linewidth of the cooling transition. However, the technique of laser cooling, well established for neutral atoms and atomic cations, has so far never been applied to anions. This is because the excess electron in an atomic anion is not bound by the Coulomb potential of a positive core but rather by quantum-mechanical correlation effects. The corresponding potential is comparatively weak and short ranged, and only allows for few (if any) bound excited states. Up to now, only very few anions are known to exhibit electric-dipole transitions between bound states, a requirement for the use of laser cooling. The very first such system, Os^-, was discovered in 2000 [12].

Our group has been studying the osmium anion by high-resolution laser spectroscopy. We have measured the transition frequency and cross section of the potential laser cooling transition [13], the hyperfine structure in isotopes with a nonvanishing nuclear spin [14], as well as the isotope shift in the anions of all seven stable Os isotopes [15]. Having established the existence of a transition potentially suitable for laser cooling, the Zeeman splitting of the relevant states in an external magnetic field of a few teslas (such as in an electromagnetic trap) must be examined. The Zeeman effect in atomic anions has been observed previously by two different methods: (1) by resolving the Zeeman substructure in the photodetachment threshold [16,17], and (2) by inducing microwave transitions between Zeeman sublevels followed by state-selective photodetachment [18]. In this article, we present a measurement of the Zeeman splitting of a bound-bound transition in an atomic anion, obtained by high-resolution laser spectroscopy on an Os^- beam. The experimental spectra allow the identification of the transition between Zeeman sublevels best suited for laser cooling, in terms of the intensities of the transition lines and the population of dark states.

II. THEORY

The line splitting in an external magnetic field results from the coupling of the atomic system with an external magnetic field $\vec{B}$ (see, e.g., Ref. [19]). The magnetic moment $\vec{\mu}$ of the ion is composed of orbital and spin contributions proportional to the total orbital angular momentum $\vec{L}$ and the total spin angular momentum $\vec{S}$, respectively. This description is valid in the Russell-Saunders (LS coupling) regime, where $\vec{L}$ and $\vec{S}$ are a good basis for describing the electronic wave function. While it is generally accepted that LS coupling only applies to light and moderately heavy elements, we find that this formalism describes the Os^- energy levels very well.

The interaction Hamiltonian of an atom in an external magnetic field is given by $\hat{H}_{\mathrm{Zeeman}} = \vec{\mu}\cdot\vec{B}$. The energy shift due to a static external magnetic field is

$$\Delta E_{\mathrm{Zeeman}} = g_J \mu_B m_J B, \tag{1}$$

*To whom correspondence should be addressed: a.kellerbauer@cern.ch
†Present address: Institut für Nanotechnologie, Karlsruher Institut für Technologie, Hermann-von-Helmholtz-Platz 1, 76344 Eggenstein-Leopoldshafen, Germany.
‡Present address: Physikalisches Institut, Albert-Ludwigs-Universität Freiburg, Hermann-Herder-Strasse 3, 79104 Freiburg, Germany.

where μ_B denotes the Bohr magneton, m_J the projection of the total electronic angular momentum along the direction of the external field, and g_J the Landé factor. The latter is given by

$$g_J = 3/2 + \frac{S(S+1) - L(L+1)}{2J(J+1)} \quad (2)$$

(when setting the free-electron g factor to $g_s = 2$). With total angular momentum $J_{gnd} = J_{exc} = 9/2$, as obtained in Ref. [15], the ground and excited states are expected to split up into 10 Zeeman levels each. Applying the appropriate selection rule $\Delta m_J = 0, \pm 1$, one predicts a total of 28 allowed transitions. These comprise ten π transitions, nine σ^+, and nine σ^- transitions. The calculated Landé factors for the ground and excited states are $g_J = 1.33$ and $g_J = 1.56$, respectively. Quantitatively, the line splitting due to the Zeeman effect is expected to be about 10 GHz T^{-1}, many orders of magnitude larger than the natural linewidth of the transition for external magnetic fields of the order of T.

III. EXPERIMENT

In order to verify these theoretical predictions, we carried out laser spectroscopy on $^{192}Os^-$ in the static magnetic field of a superconducting solenoid magnet. In our apparatus (see Fig. 1), the laser is brought into transverse overlap with the ion beam via a set of mirrors and diaphragms located around the central ring of a stack of cylindrical electrodes at the center of the magnet. Due to the positioning of the central electrode and the laser entrance and exit apertures, the interaction region is limited to a volume where the magnetic-field magnitude and its homogeneity are maximal (homogeneity better than 0.1% within a radius $r < 10$ mm). A beam of mass-separated ions at a kinetic energy $E_{kin} = 2.5$ keV, consisting of more than 90% $^{192}Os^-$, is directed through the magnet along its axis such that the magnetic field is parallel to the ion beam. After interaction of the ions with the laser field, the osmium ions, now either in the ground or the excited state, are guided towards the detection setup. Here the excited ions are field neutralized by a strong longitudinal electric field and subsequently detected as neutrals on a microchannel plate detector (MCP). Since the

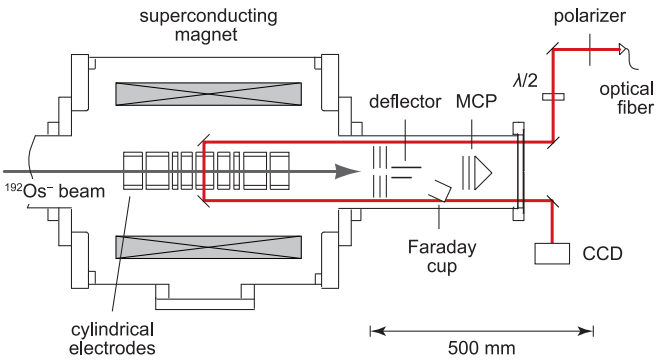

FIG. 1. (Color online) Experimental setup for transverse spectroscopy in the homogeneous-magnetic-field region of a superconducting magnet. The ion beam (gray) and laser beam (red) are superimposed perpendicularly at the center of the magnet.

electric field is not sufficient to detach electrons from ions in the ground state, these ions can be separated from the neutrals by deflecting them into a Faraday cup. The ion source and spectroscopy setup are described in more detail in Ref. [13].

The laser light is delivered by a custom-built continuous-wave optical parametric oscillator (OPO) system (Xiton Photonics), which is pumped by a frequency-doubled Nd:YAG laser [20]. To ensure the stability of the laser beam alignment as well as a well-defined Gaussian beam profile, the linearly polarized laser light is transported to the entrance window into the vacuum with a polarization-maintaining optical fiber. An additional polarizer is placed in between to reduce the amount of depolarized light due to imperfections in the optical fiber. For a controlled rotation of the angle of the polarization ($\vec{P}$) with respect to the magnetic field, a $\lambda/2$ plate is also introduced into the laser path. This is used to selectively drive π transitions ($\vec{P} \parallel \vec{B}$) and σ transitions ($\vec{P} \perp \vec{B}$).

Spectroscopic measurements were performed for 12 different magnetic-field magnitudes in the range $B = 10$–100 mT. The wavelength of the (single) transition at $B \approx 0$ (magnet current $I = 0$ A, earth magnetic field not shielded) was found to agree with the result from the collinear setup in Ref. [13]. At each magnetic-field value, the spectra for parallel and orthogonal laser polarizations with respect to the field were recorded. Altogether 24 spectra, each containing 10 or 18 peaks, depending on the laser polarization, were taken. Typical spectra for both laser polarizations at $B = 35$ mT are shown in Fig. 2. The upper graph shows the resonances for the σ transitions and the lower one for the π transitions. The solid lines are fits of sums of Gaussian functions whose energy splitting is given by Eqs. (1) and (2). The fits use the following five parameters: the line intensities, peak width, the two Landé factors for the ground and excited states, and the magnitude of the magnetic field. The relative intensities were calculated from the relevant Clebsch-Gordan coefficients. The peak widths were held fixed at the value $\Gamma_{tot} = 110$ MHz found for the single peak at zero field. The resonance width is due to the Doppler broadening from the transverse velocity of the hot ions, clipped by the horizontal restrictions of the beam line ("geometrical cooling" [21]), as well as transit time broadening from the passage of the ion beam through the laser. The magnetic-field magnitude of the superconducting magnet was calculated from the power supply current used during a given measurement. Finally, the two g_J factors and the absolute intensity were extracted from the fits as the only free parameters. The fit routine was applied separately to each recorded spectrum.

The experimental results are displayed in Fig. 3. It shows that the g_J factors extracted across the entire range of magnetic-field magnitudes agree very well. We were therefore able to compute the weighted means of the Landé factors, indicated by the solid horizontal lines in the figure. The dashed horizontal lines correspond to the 1σ uncertainties of the weighted means. The final results are summarized in Table I, together with values calculated according to Eq. (2). The Landé g factors calculated for pure LS states are given in the second column. The third column shows Landé factors calculated for LS state compositions of the ground and excited states, as obtained in Ref. [22]. The agreement between the

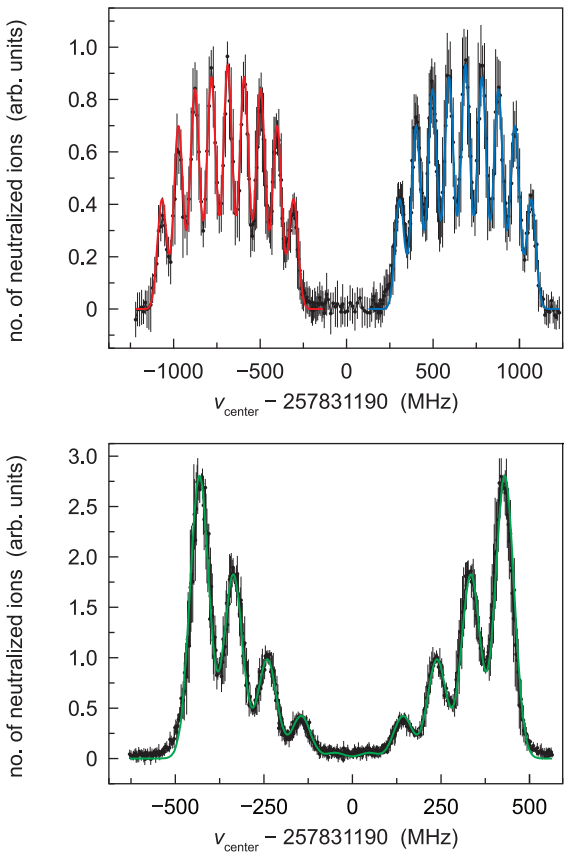

FIG. 2. (Color online) Measured absorption spectra in an external magnetic field of 35 mT. Top: 2× 9 peaks of the σ transitions; bottom: 10 peaks of the π transitions. Experimental data are represented by black dots with vertical error bars, and the corresponding fits (see text) by solid lines.

experimental and the theoretical g_J factors, particularly for the composite LS states, is found to be very good. The calculated magnetic-field magnitude is the dominant source of systematic uncertainty for the experimental values. Its estimated error is ±5%, which has to be added to the statistical uncertainties

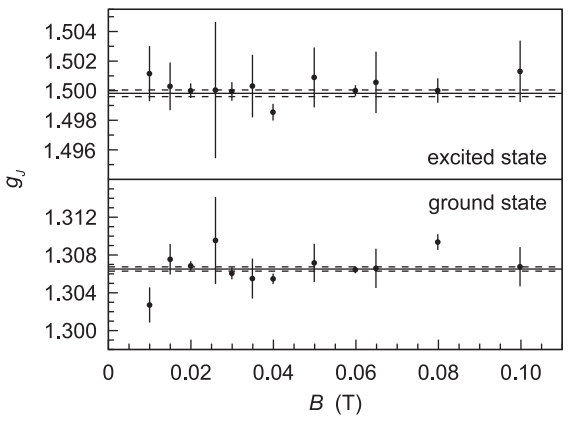

FIG. 3. Landé g factors extracted from the fits of the experimental Zeeman spectra (see text). The weighted means are indicated as the bands (1σ uncertainties) between the dashed horizontal lines.

TABLE I. Comparison of experimental values of the Landé g factors with the calculated values, for pure and composite LS states (see text). The experimental uncertainty mainly results from the calculated magnitude of the magnetic field (±5%).

	Landé factor g_J		
	Calculated		
State	Pure LS	Composite LS	Experimental
$^6D_{9/2}$ (exc)	1.56	1.52	1.50(8)
$^4F_{9/2}$ (gnd)	1.33	1.31	1.31(7)

from the fits. It results in a corresponding uncertainty in the experimental Landé factors. For a more precise experimental determination, an independent measurement of the magnetic field, at the site of the interaction of the laser field with the ions, would have to be established.

IV. RESULTS AND DISCUSSION

While we achieved a precision of $\approx 10^{-7}$ for the transition frequencies of all allowed transitions between Zeeman sublevels, the Landé g factors were determined with an uncertainty of $< 2 \times 10^{-4}$ (neglecting the uncertainty of the magnetic-field magnitude). This is because the Zeeman splitting is a small correction to a large quantity. Resolved photodetachment thresholds, as in Ref. [17] or, more recently, in Ref. [23], have not yet been successfully used to extract numerical values for g_J. On the other hand, the direct measurement of the microwave transitions between Zeeman sublevels of the $^2P_{3/2}$ ground state in $^{32}S^-$ by Jopson and Larson [18] yielded a Landé g factor with a remarkable precision of 2×10^{-5}. At this level, the result becomes sensitive to the anomalous g factor of the electron, as well as relativistic and diamagnetic effects.

Our results, along with those of Refs. [13,14], allow us to discuss the prospects of laser cooling Os^-. Figure 4 shows a partial energy-level diagram in which the Zeeman levels with $m_J < 0$ of the $^4F^e_{9/2}$ ground state and the $^6D^o_{9/2}$ excited state are indicated. The Zeeman levels of the intermediate $^4F_{7/2}$ state are also shown. The π transition between the $m_J = -9/2 \longleftrightarrow -9/2$ sublevels (green arrow) is the one with the strongest coupling. It would therefore be a natural choice for a laser-cooling transition. However, the energy-level diagram shows that this transition is not closed. The excited state may decay back to the ground state with the cooling rate $\Gamma_c \approx 50$ Hz [13] (see also below). In addition, it may decay to the $m_J = -7/2$ sublevel of the ground state *or* to the $m_J = -7/2$ sublevel of the intermediate $^4F^e_{7/2}$ state. Once either of these states is populated, the ion is no longer addressed by the laser.

The decay rate to the neighboring sublevel of the ground state can be calculated from the Clebsch-Gordan coefficients as $\Gamma_c/4.5$. The decay rate to the $m_J = -7/2$ sublevel of the intermediate state has been found to be $\Gamma_c/7.4$ from relativistic configuration interaction calculations [22]. Despite the lower rates, all ions will eventually end up in either one of these two dark states. At this point, cooling will stop unless the ions are repumped to the $^6D_{9/2}$, $m_J = -9/2$ excited state. Assuming a

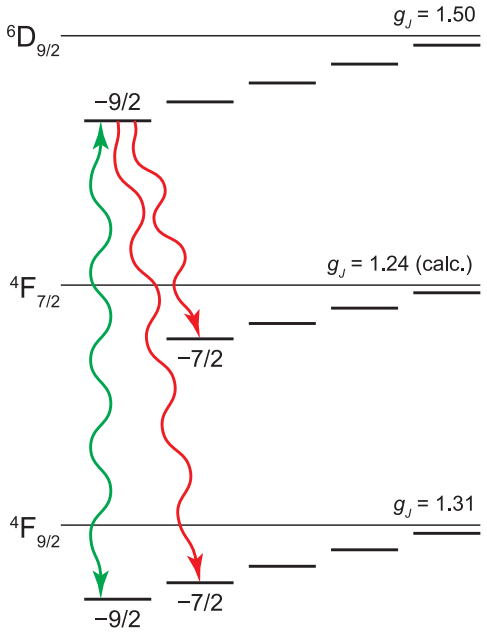

FIG. 4. (Color online) Partial energy-level diagram of $^{192}Os^-$ in an external magnetic field. Zeeman sublevels are not to scale. The potential laser cooling (π) transition is represented as a green arrow, and the undesired (σ^+) decays to dark states are indicated by red arrows.

magnetic field of 1 T, the required laser detuning for repumping the $^4F_{9/2}$, $m_J = -7/2$ state is $\Delta\nu = 18.6$ GHz. This near wavelength could be produced by electro-optical modulation of the cooling laser [24]. An additional laser system with a wavelength $\lambda = 2.29$ μm would be required to repump the intermediate $^4F_{7/2}$, $m_J = -7/2$ state.

Our prior measurement of the resonant cross section of the laser cooling transition [13] indicates that the transition rate is only $\Gamma_c \approx 50$ Hz, a factor of 30 smaller than previously found. The low rate is likely due to the fact that the transition is spin forbidden ($S = 3/2 \longleftrightarrow 5/2$). While a narrow natural linewidth corresponds to a low Doppler temperature ($< \mu$K), the low cooling rate also means that cooling takes longer. Furthermore, competing heating effects, for instance, due to the absorption of black-body radiation, must be overcome. In order to limit the required cooling time to a reasonable level, the Os^- ions must be precooled to liquid-helium temperature by other means prior to laser cooling. The laser cooling from 4 K to the Doppler temperature would then take ≈ 5 min, assuming the transition is saturated.

To conclude, the Zeeman splitting of a bound-bound transition in an atomic anion has been experimentally investigated. The full spectra, each comprising 28 σ and π resonances, have been visualized by optical spectroscopy on $^{192}Os^-$ in magnetic fields $B = 10$–100 mT. The Landé g factors $g_J = 1.31(7)$ ($^4F_{9/2}$ ground state) and $g_J = 1.50(8)$ ($^6D_{9/2}$ excited state) were found to be in good agreement with the calculated values, especially those taking into account composite LS states. The resulting energy-level diagram suggests the transition between the $m_J = -9/2 \longleftrightarrow -9/2$ Zeeman sublevels as a suitable laser cooling transition. The decay of the excited state to two "dark" states will require the use of two repumping wavelengths. Furthermore, the low cooling transition rate results in a long cooling time even if the anions are precooled. Thus, laser cooling of Os^- appears feasible but challenging. It may be worthwhile to investigate other predicted bound-bound electric-dipole transitions in lanthanide anions [22], particularly La^-, with a view to the laser cooling of atomic anions.

ACKNOWLEDGMENTS

This work was supported by the German Research Foundation (DFG) under Contract No. KE1369/1-1, as well as by the European Research Council (ERC) under Grant No. 259209 (UNIC). We thank the MPIK accelerator group and workshop, in particular, M. König, V. Mallinger, and M. Beckmann. The assistance of the group of S. Jochim for the wave meter calibration is gratefully acknowledged. We are indebted to C. W. Walter (Denison University, Granville, OH, USA) for his valuable comments on the manuscript.

[1] B. Song, N. D'Angelo, and R. L. Merlino, Phys. Fluids B **3**, 284 (1991).
[2] S. K. Guharay, AIP Conf. Proc. **1390**, 661 (2011).
[3] N. L. Aleksandrov, Plasma Sources Sci. Technol. **3**, 226 (1994).
[4] J. Ishikawa, in *Handbook of Ion Sources*, edited by B. H. Wolf (CRC Press, Cleveland, OH, 1995).
[5] N. Berrah, J. D. Bozek A. A. Wills, G. Turri, H.-L. Zhou, S. T. Manson, G. Akerman, B. Rude, N. D. Givson, C. W. Walter *et al.*, Phys. Rev. Lett. **87**, 253002 (2001).
[6] L. G. Gerchikov and G. F. Gribakin, Phys. Rev. A **77**, 042724 (2008).
[7] T. P. Snow, N. D'Angelo, and V. M. Bierbaum, Annu. Rev. Anal. Chem. **1**, 229 (2008).
[8] G. Gabrielse, X. Fei, L. A. Orozco, R. L. Tjoelker, J. Haas, H. Kalinowsky, T. A. Trainor, and W. Kells, Phys. Rev. Lett. **63**, 1360 (1989).
[9] D. Gerlich, Phys. Scr., T **59**, 256 (1995).
[10] G. Gabrielse, A. Khabbaz, D.S. Hall, C. Heimann, H. Kalinowsky, and W. Jhe, Phys. Rev. Lett. **82**, 3198 (1999).
[11] A. Kellerbauer and J. Walz, New J. Phys. **8**, 45 (2006).
[12] R. C. Bilodeau and H. K. Haugen, Phys. Rev. Lett. **85**, 534 (2000).
[13] U. Warring, M. Amoretti, C. Canali, A. Fischer, R. Heyne, J. O. Meier, Ch. Morhard, and A. Kellerbauer, Phys. Rev. Lett. **102**, 043001 (2009).
[14] A. Fischer, C. Canali, U. Warring, A. Kellerbauer, and S. Fritzsche, Phys. Rev. Lett. **104**, 073004 (2010).

[15] A. Kellerbauer, C. Canali, A. Fischer, U. Warring, and S. Fritzsche, Phys. Rev. A **84**, 062510 (2011).
[16] W. A. M. Blumberg, R. M. Jopson, and D. J. Larson, Phys. Rev. Lett. **40**, 1320 (1978).
[17] R. E. Elmquist, C. J. Edge, G. D. Fletcher, and D. J. Larson, Phys. Rev. Lett. **58**, 333 (1987).
[18] R. M. Jopson and D. J. Larson, Phys. Rev. Lett. **47**, 789 (1981).
[19] C. J. Foot, *Atomic Physics* (Oxford University Press, New York, 2005).
[20] P. Groß, I. D. Lindsay, C. J. Lee, M. Nittmann, T. Bauer, J. Bartschke, U. Warring, A. Fischer, A. Kellerbauer, and K.-J. Boller, Opt. Lett. **35**, 820 (2010).
[21] W. Demtröder, *Laser Spectroscopy*, 4th ed. (Springer, Berlin, 2008), Vol. 2.
[22] S. M. O'Malley and D. R. Beck, Phys. Rev. A **81**, 032503 (2010).
[23] A. Joiner, R. H. Mohr, and J. N. Yukich, Phys. Rev. A **83**, 035401 (2011).
[24] D. Leibfried, F. Schmidt-Kaler, M. Weitz, and T. W. Hänsch, Appl. Phys. B **56**, 65 (1993).