# Collinear laser spectroscopy of manganese isotopes using optical pumping in ISCOOL.

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Recently, optical pumping of ions has been achieved inside an ion beam cooler–buncher [1]. By illuminating the central axis of the cooler with laser light, subsequent decay populates selected ionic metastable states. This population enhancement is retained as the ion beam is delivered to an experimental station. In the case of collinear laser spectroscopy, transitions can then be excited from a preferred metastable level, rather than the ground– state. This proposal seeks to establish and develop the technique for ISCOOL. As a test of efficiency, this will be applied to the study of <sup>55</sup>−<sup>66</sup>Mn isotopes using collinear laser spectroscopy—expanding an earlier study where the benefit of the technique was demonstrated [2]. This will provide nuclear spins, magnetic–dipole and electric–quadrupole moments and changes in mean–square charge radii across the  $N = 40$  shell closure candidate and into a region where an onset of deformation, and a new "island of inversion", is predicted [3].

#### Physics case

A recent experiment performed collinear laser spectroscopy on the ground and isomeric– states of <sup>50−56</sup>Mn [2]. In this study, the behaviour of the mean–square charge radii was compared to the trend in neutron–separation energies. Such comparisons are now gaining theoretical attention [4]. While a close correspondence is found in isotope chains with higher– $Z$  [5], no such correspondence was seen in manganese. In particular, the course of the two–neutron separation energies appears smooth through the  $N = 28$  shell closure, and yet, the charge radii (and quadrupole moments) show a distinct minimum and a robust shell closure. Such shell effects are also investigated through the measurement of the nuclear moments, which are a very sensitive probe of the wave functions used in shell–model calculations [6].



Figure 1: Two–neutron separation energies, including the 2009 ISOLTRAP measurements of manganese [11].

Further from stability, the migration of single–particle levels result in a weakening or eradication of shell or sub-shell closures, while new candidates, such as those at  $N = 32$ and  $N = 34$ , emerge [7, 8]. Beyond these, the shell closure candidate at  $N = 40$  has received much attention. While  $B(E2)$  values in nickel decrease (and first excited  $2^+$ energies rise) towards  $N = 40$  [9], only a weak effect is seen in the two–neutron separation energies [10]. Subsequent ISOLTRAP mass measurements showed no enhanced binding at  $N = 40$  for manganese [11]. However, the course of the two–neutron separation energies appears to deviate beyond  $N = 34$  [11] (see figure 1) and appear different to those of iron. Measurements of excitation energies in chromium [12]  $(Z = 24)$  and in iron [3]  $(Z = 26)$ , show an increase in collectivity towards  $N = 40$  where it is suggested there may be a new "island of inversion" [3] similar to that around  $^{32}$ Mg. In manganese (Z = 25) rotational like structures have been observed in a study of  $57-60$ Mn isotopes [13]. Any increase in deformation can be investigated directly using optical techniques, which probe the spectroscopic quadrupole moments and mean–square charge radii. This will be the first investigation of these ground state properties in this region of the Segré chart.

Most manganese ground–state and isomeric–state spins are unassigned, or assignments are based on regional systematics. The study of gallium isotopes at ISOLDE [6], where <sup>73</sup>Ga was unexpectedly found to have a spin  $I = 1/2$  ground–state, is only a recent example of why such an approach should be taken with caution. Laser spectroscopy however, provides measurements of the spin.

For the odd–A manganese isotopes, the spins are all tentatively assigned  $I = 5/2$ from  ${}^{61}$ Mn onwards. For the even–A (odd–odd) manganese isotopes, all spin assignments made beyond <sup>56</sup>Mn are based on regional systematics or shell–model calculations. For <sup>58</sup>Mn, a series of spin assignments ( $I = 0, 1, 2, 3, 4$ ) were made for the ground and isomeric–states, with the current tentative assignments being a spin and parity of  $1^+$  for the ground–state and  $4^+$  for the isomeric–state. All that is known for certain is that the multipolarity of the transition between these two states is M3. However, GXPF1 shell– model calculations predict an inversion of these states, making the ground–state 4<sup>+</sup> and the isomeric–state  $1^+$  [14]. This is unlike  $60$ Mn where both the tentative experimental assignments and GXPF1 calculations suggest a  $1^+$  ground–state and  $4^+$  isomeric–state. However, for <sup>62</sup>Mn, it is suggested that the states are inverted, with a ground–state spin of  $3<sup>+</sup>$  or  $4<sup>+</sup>$  and a  $1<sup>+</sup>$  isomeric–state [7], contrary to various shell–model calculations, which all predict a  $2^+$  ground–state [15]. For <sup>64</sup>Mn, the ground–state is tentatively assigned  $1^+$ and for  $^{66}$ Mn it is tentatively assigned as  $2^+$  [16].

Spin assignments in the manganese chain also have an impact, via the decay chain, to those of other elements. In  $^{66}$ Mn for example, a change in the ground–state spin assignment would affect our understanding of the excited states in  ${}^{66}Fe$ —their feeding and spin assignments. Preliminary results from manganese nuclear–decay–spectroscopy experiments at ISOLDE suggest a necessary change to the spins and parities of the low– lying states in cobalt (see figure 2) [16]. In the  ${}^{66}\text{Mn} \rightarrow {}^{66}\text{Fe} \rightarrow {}^{66}\text{Co}$   $\beta$ -decay chain, it could even be that the ground–state spin of  $^{66}$ Mn affects the feeding of the levels in cobalt if, for example, a  $\beta$ -decaying isomeric state exists in <sup>66</sup>Fe.

Laser spectroscopy can directly measure the spins for all of the isotopes (and isomers of <sup>58</sup>,60,<sup>62</sup>Mn). The nuclear magnetic–dipole and electric–quadrupole moments are unknown for all manganese isotopes beyond <sup>56</sup>Mn. These will also be measured, and provide a stringent test for wave functions generated by new shell–model calculations.

## Production

The manganese yields from the on-line database are shown in figure 3 and date from the year 2000 [17]. Since then, improvements have been made to the RILIS scheme prompted by the transition to solid–state pump lasers [18]. Collinear laser spectroscopy has been performed on yields as low as 100 ions/s [19], and the recent manganese experiment was successful using yields of a similar order. With the  $6 \mu s$  bunches available from IS-COOL  $[6]$ , measurements up to and including  $66$ Mn will be possible. This will provide the nuclear spins, magnetic–dipole moments, electric–quadrupole moments and mean–square charge radii for all of these isotopes.

#### Use of the optical pumping technique

In the previous study  $[2]$  in Jyväskylä, collinear laser spectroscopy of a bunched ionic beam [20] was used. Charge exchange of the ionic beam to the neutral state was avoided, since this would have spread the population over a high density of atomic states, reducing the efficiency of spectroscopy from the ground–state. Unfortunately, the ionic transition from the (well populated) ground–state requires a wavelength of 230 − 260 nm—too short to be efficiently produced by high–resolution continuous–wave lasers. Instead, a 294.92 nm transition was chosen from an excited state at 9472.97 cm<sup>-1</sup>. In order to enhance the population of this metastable state, the axis and trapping region of the ion beam cooler–buncher was illuminated with a few mW of 230.5 nm light (figure 4). Unlike the high–resolution requirements for laser spectroscopy, broadband laser light is sufficient for the task and matches the Doppler broadened velocity profile well. The laser light can therefore be produced using pulsed lasers, which can readily be used to achieve higher harmonics and the required 230.5 nm wavelength. Although a weak transition from the



Figure 2: Levels in  $^{66}Fe$  and  $^{66}Co$  populated from the decay of  $^{66}Mn$  (preliminary) [16].



Figure 3: Manganese yields from the ISOLDE yields database (year 2000) using a uranium carbide target. These have since been improved as a result of the RILIS upgrade programme.

ground–state, the time spent exposed to the laser light in the cooler ensures efficient optical excitation. Indeed, a similar test [1] proved that 100% of the sample interacted with the laser, even with the cooler operating in continuous mode, with the ground–state of the emerging ion beam completely depopulated. In the scheme used for manganese the subsequent decay enhanced the population of the required metastable level, which persisted during delivery to the laser–spectroscopy station.

This proposal seeks to address the physics case above and in doing so establish such in–cooler optical pumping experiments using ISCOOL. To do so requires illumination of the central axis and/or trapping region of ISCOOL. Optical access can be achieved using a port on the outer side of the second magnet of the HRS. This will provide a direct "line of sight" to ISCOOL as shown in figure 5.

Spectroscopy is performed using the singly–charged ionic beam. More elaborate components such as a charge–exchange cell are not required, thus reducing the complexity of the collinear spectroscopy. The "light collection region" shown in figure 6 contains a removable aperture for aligning the laser and ion beams, Doppler tuning electrode and the optical detection system. These components are contained within a standard beam cube, have been successfully tested and can be located on any "spare" line at ISOLDE (LA1 or LA2) without disrupting any existing laser programme. In the technique of collinear laser spectroscopy, the ions are accumulated in ISCOOL every  $10 - 200$  ms cycle and released in bunches of 6  $\mu$ s temporal length. A gate is applied to the photon signal which defines the laser/ion–bunch interaction time and all photons arriving outside of this window are vetoed. This enables the background caused by the continuous scattering of laser light to be suppressed by four orders–of–magnitude.

#### Application of optical pumping beyond manganese



Figure 4: Optical pumping and spectroscopy scheme to be used.



Figure 5: Laser access to ISCOOL can be obtained through a port in the outer side of the second HRS magnet. This will allow optical pumping of the ions in the trapping region of ISCOOL to populate selected ionic metastable states in the emerging ion beam.



Figure 6: The "cube" spectroscopy apparatus for spectroscopy of ionic beams.

With optical pumping established at ISOLDE, the applications will reach beyond the physics case and techniques presented here. Although we do not currently request beam time for these, the following are to give a flavour of possible future use.

#### (1) Optical pumping to perform laser spectroscopy from metastable states.

In the case of manganese, the wavelengths of transitions from the ground–state are too short to be efficiently produced using the continuous–wave lasers used for high–resolution laser spectroscopy. Population enhancement of a metastable state using optical pumping permits a more suitable transition to be chosen from this level. In other situations (eg. niobium), the ground–state transition may be too weak—a problem for collinear laser spectroscopy, but not for the extended ion–laser interaction time in the cooler [1]. In studies of yttrium [21, 22] and niobium [1], the ionic ground–state has an atomic spin  $J = 0$ . This restricts all transitions to  $J = 0 \rightarrow J = 1$ , which produces a hyperfine structure with a maximum of only three peaks—sufficient to provide the mean–square charge radii and nuclear moments only in cases where the nuclear spin is already known. Transitions from metastable states with  $J > 0$  produce more peaks, allowing all four of these quantities to be unambiguously measured. This is the case for the proposed study of lutetium (CERN-INTC-2010-005/I-085) for which optical pumping in ISCOOL will be required.

#### (2) Purification to provide a beam of a single element.

Before the introduction of the bunching technique, the photon background was suppressed by detecting photons in delayed coincidence with the corresponding ion [23] or atom [24]. With position correction of the photon detection [25, 26] using segmented photomultiplier tubes, and now cooled beams, the arrangement is theoretically capable of performing measurements on fluxes of only a few ions per second. This is superior to the bunching technique (which requires  $\sim 100/s$ ) but in practice the sensitivity is limited by isobaric components of the beam triggering "false" coincidences. In–cooler optical pumping may be developed to step–wise resonantly ionize the ions to the doubly–charged state. On release of the ions from the cooler in bunches, these doubly-charged ions will travel with twice the energy and be fully separated in TOF analysis from the remaining ions. The latter can be electrostatically deflected away from the beam path. The selected ion bunch will contain doubly-charged ions of the selected element and mass only. No other ion passed by the HRS will have been (doubly) ionized to then have half the selected mass to charge ratio on leaving the cooler. Ultra–pure beams of single  $(N, Z)$  would then be available for efficient laser spectroscopy or for any other experiment at ISOLDE using ISCOOL.

#### Beam request

This proposal requires the use of the HRS (with a uranium carbide target), RILIS for the enhancement of the manganese yield, ISCOOL and the LA1 or LA2 beamlines. We request 18 shifts of beam time and estimate a shift per isotope close to stability (allowing for optimisation) and 2 shifts per isotope further from stability. Stable beam time will also be required at least for a day prior to each radioactive beam period to enable setting up and initial optimisation.

#### Safety

Access will be required to the HRS/ISCOOL area for the initial alignment of the laser path into ISCOOL. Once established, no further intervention will be required and fine tuning can be done without further intervention (as is the case with RILIS). All laser beams will be concealed and present no risk. The production of the light for in–cooler optical pumping will be provided by the RILIS cabin and will be controlled by their safety guidelines. Similarly, the laser–spectroscopy station will be fully enclosed and laser light will be fibre coupled from the CRIS (IS457) laser table, with existing laser safety procedures respected.

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