β-Detected Nuclear Magnetic Resonance at ISAC

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We are developing a beta-decay nuclear magnetic resonance spectrometer which will accept an intense beam of low energy highly polarized radioactive ions produced by the ISAC facility. Although the main application is condensed matter physics the polarizer itself may also have applications in nuclear physics. What follows is an introduction into the scientific motivation, a description of the polarizer and spectrometer, and first test results from the spectrometer.

Introduction

Nuclear magnetic resonance (NMR) and related nuclear methods such as muon spin rotation are important probes of local electronic and magnetic properties of solids. A common feature in all forms of magnetic resonance is that one first creates nuclear polarization and then measures how that polarization evolves in time. The spin precession rate or Larmor frequency is a measure of the local magnetic field at the nucleus, whereas the spin relaxation rate senses the low energy dynamics. However, there are significant differences between methods which may influence particular applications. For example, in conventional magnetic resonance a small nuclear polarization is generated in thermal equilibrium by applying a large magnetic field and a pickup coil is used to detect the precession of the nuclear magnetization. Typically one needs about $10^{17}$ nuclear spins to generate a good NMR signal. On the other hand, in related nuclear methods such as muon spin rotation ($µ$SR) or $β$-detected NMR ($β$-NMR) a beam of highly polarized radioactive nuclei (or muons) is implanted into the material. The time evolution of the spin polarization is picked up through anisotropy in the distribution of particles from the decay of a radioactive nucleus or muon. The much larger initial polarization results in a factor of about $10^{10}$ fewer spins being required to obtain a signal from nuclear methods such as $µ$SR or $β$-NMR. This enhanced sensitivity means that nuclear methods are well suited to studies of dilute impurities or small structures with few host spins. Areas of application include many of the most interesting problems in condensed matter physics e.g. high Tc superconductors, magnetic multilayers, and semiconductor quantum wells.

The principles for $β$-NMR and $µ$SR are almost identical. In fact the two methods became possible at almost the same time when it was demonstrated in 1957 that parity is violated in weak interactions. In a decay mediated by the weak interaction the direction of the outgoing charged particle is correlated with the nuclear (or muon) spin at the instant of decay.

Muon spin rotation is now widely used as a sensitive probe of internal magnetic fields, to simulate the behaviour of hydrogen in semiconductors, and in studies of hydrogen chemistry and organic free radicals. The most important contributions from $µ$SR in chemistry and physics came after the building of the meson factories in the late 1970's and a period of development in the technique, $β$-NMR has been used by nuclear physicists studying properties of unstable nuclei. Although elegant experiments have been performed on a variety of materials such as simple metals [1], surfaces [2], disordered solids [3], and semiconductors [4] the method has not been developed as a probe of condensed matter to the same extent as $µ$SR. One reason for this is condensed matter studies typically require a high signal to noise ratio which is not as easy to achieve as in the case of muons. This situation is now changing since at facilities such as ISAC it should be possible to generate intense ($>10^8/s$) and highly polarized (80%) beams of various radioactive nuclei. [5] Under these circumstances one can expect large enhancements in the signal to noise with $β$-NMR. Furthermore such beams can be implanted at very shallow depths (1–1000 nm) whereas conventional muon beams are used primarily as a bulk probe of matter.

Most elements have at least one isotope that can be studied with $β$-NMR. Naturally the number of isotopes suitable for use as a probe in condensed matter is considerably smaller. The most essential requirements are: (1) a high production efficiency (2) a method to efficiently polarize the nuclear spins and (3) a high beta decay asymmetry. Other desirable features are: (4) small Z to reduce radiation damage on implantation, (5) a small value of spin so that the $β$-NMR spectra are relatively simple and (6) a radioactive lifetime that is not much longer than a few seconds. Table 1 gives a short list of the isotopes we have identified as suitable for development at ISAC. Production rates of $10^8/s$ are easily obtainable and in some cases such as $^8$Li we anticipate rates as high as $10^9/s$.

One of our objectives will be to develop new probes tailored for specific applications in condensed matter. For example the spin-$\frac{1}{2}$ $^{17}$Ne isotope should act as a pure magnetic sensor since it has no quadrupole interaction and is chemically inert. Oxygen is also interesting since oxides are playing a major role in research on superconductivity and magnetism. Furthermore, in the case of oxygen it will be possible to switch from an isotope with a nuclear quadrupole moment to one without a quadrupole moment (see Table 1). Although the quadrupole interaction can be very useful in distinguishing sites it is also desirable in some circumstances to eliminate the quadrupole interaction.
Polarizer

Ions emerging from an ISOL target are unpolarized. A fast collinear polarization technique can be used to generate high nuclear polarization in all the alkali metal atoms such as Li. This is possible since the ground state of a neutral alkali atom can be excited with visible or near visible lasers. The method has been used extensively by Neugart at al at ISOLDE to measure magnetic and electric quadrupole moments of unstable nuclei.[5]

Our first set of experiments will be on $^8$Li (nuclear spin I=2) which is the lightest isotope suitable for $\beta$-NMR. (see Table 1) The Li$^+$ ion beam is first neutralized by passing it through a Na vapour cell. Li$^+$ has a large cross section for charge exchange with Na ($> 10^{-15}$cm$^2$) and is neutralized efficiently with little or no change in the beam emittance. The neutral Li beam drifts 1.7m in a small longitudinal magnetic holding field of 1mT while being pumped with circularly polarized laser light brought in along the beam axis. The D1 atomic transition of neutral Li $2s^2S_{1/2} \rightarrow 2p^2P_{1/2}$ occurs at about 671 nm. Doppler broadening of the resonance is dramatically suppressed by accelerating the beam up to 30 keV and this reduces the required laser power. Tuning of the resonance is done by making fine adjustments to the beam energy (doppler shift) while stabilizing the laser frequency externally. Both the ground and first excited atomic levels are split by the hyperfine coupling between total spin states $F = 5/2$ and $F = 3/2$ and both of these hyperfine states must be pumped in order to maximize the polarization. The object is to pump all the atoms into the $F = 5/2, M_F = 5/2$ spin state which is fully nuclear polarized.

The final step is to ionize the neutral beam by passing it through a He gas cell. Results from the test stand (see elsewhere in this report) show that this can be done with high efficiency and only a nominal increase in the emittance. The beamline after this point has been designed to accept a large emittance beam (200 mm-mrad). After reionization the polarized Li$^+$ ion beam passes through two 45° electrostatic bends which preserve the longitudinal polarization. The final section before the magnet has three Einzel lenses, two pairs of steering plates and three adjustable collimators which control the beam spot on the sample. This eliminates the need for collimation downstream of the back counter since all the ions passing through the back detector stop in the sample. Only betas originating from the sample reach the detectors. This final leg in the beamline is ultra-high vacuum (UHV) compatible in order to reduce the pressure from $10^{-7}$ torr upstream of the spectrometer to $10^{-10}$ torr in the main chamber. This is necessary to avoid accumulating residual gases which would otherwise be condensed onto the surface of the sample at low temperatures.

Spectrometer

Figure 1 shows a schematic of the high field $\beta$-NMR spectrometer which will be complete in a few months when a new 9T magnet arrives. The main function of the spectrometer is to record the beta-decay anisotropy from spin polarized ions implanted into the sample, either as a function of RF frequency or time after implantation. The nuclear polarization is longitudinal with respect to both the beam and the axis of high homogeneity 9T solenoid. This geometry is required for measurements in high magnetic fields where both the incoming ions and outgoing betas are strongly influenced by the magnetic field. All elements of the spectrometer are UHV compatible and a base pressure of $10^{-10}$ torr has been reached. Two large cryopumps are used to achieve the desired UHV vacuum condition. The cryostat is mounted on a large bellows so that it can be removed from the magnet bore in order to change the sample though a load lock on top of the main vacuum chamber. Fast plastic scintillators are used to detect the betas from $^8$Li $\rightarrow ^7$Be $+ \nu_e + e^-$ for which the end point energy is 13 MeV. This is enough to easily penetrate thin stainless steel windows. Since we anticipate that the counting rates may reach $10^8/s$ the detectors are segmented into 16 elements each to reduce distortions in the time histograms due to detector deadtime. The plastic scintillators and light guides are held in reentrant stainless steel housings with thin stainless steel windows which isolate the detectors from the UHV vacuum chamber but allow transmission of the low energy betas. In order to detect betas in the backward direction (opposite to the beam) it is necessary to place the detector outside the magnet since the betas are confined to the beam axis while inside the bore of the magnet. Simulations and now experiment show that as the betas emerge from the magnet bore in high field the angular cone of emission collapses. Consequently the two detectors shown in Fig. 1 intercept betas over very similar solid angles even though they appear very asymmetric.

In December 1999 we had our first test run to commission the beamline and spectrometer. Both parts were very successful. Figure 2 shows our first lifetime spectrum for $^8$Li measured with unpolarized beam. The $^8$Li rate at the spectrometer is estimated to be $10^7/s$ with a Ta target confirming that we should easily reach $10^8/s$ with 100$\mu$A on a carbon target. This lifetime spectrum, uncorrected for background or deadtime, is undistorted indicating that the detector segments can easily operate at 1MHz.

One of the most important features of the spectrometer is that the ions can be implanted over a wide range of...
of energies (1–90 keV) with the same high efficiency. This will be tested in July 2000 by floating the spectrometer and all ancillary equipment (magnet, power supplies, cryostat, thermometry etc.) on a high voltage platform. For example by applying a bias voltage to the platform between +29 keV and -60 keV one can adjust the energy of implantation in the range (1–90 keV). For $^8$Li this corresponds to an average implantation depth of between 6 nm and 300 nm respectively.

Table 1. Examples of isotopes suitable for $\beta$-NMR.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>$T_{1/2}$ (s)</th>
<th>$\gamma$ (MHz/T)</th>
<th>$\beta$-Decay Asy (A)</th>
<th>Flux ($s^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^8$Li</td>
<td>2</td>
<td>0.8</td>
<td>6.2</td>
<td>0.33</td>
</tr>
<tr>
<td>$^{11}$Be</td>
<td>1/2</td>
<td>13.8</td>
<td>22</td>
<td>0.33</td>
</tr>
<tr>
<td>$^{15}$O</td>
<td>1/2</td>
<td>122</td>
<td>10.8</td>
<td>0.7</td>
</tr>
<tr>
<td>$^{19}$O</td>
<td>5/2</td>
<td>26.7</td>
<td>4.7</td>
<td>0.7</td>
</tr>
<tr>
<td>$^{17}$Ne</td>
<td>1/2</td>
<td>0.1</td>
<td></td>
<td>0.33</td>
</tr>
</tbody>
</table>

References


Fig. 1. High Field $\beta$-NMR spectrometer at ISAC. The beam enters from the left, passes through a small hole in the backward scintillation detector and is focussed onto the sample with the Einzel lens. Betas are detected with two segmented scintillation counters centered on the axis of the superconducting solenoid.

Fig. 2. First $^8$Li decays at the spectrometer in December 1999. During this run the beam was turned on for the first 2 s of each 10 s cycle. Note the single exponential decay with the characteristic lifetime of $^8$Li. The data accumulation rate is about $3.2 \times 10^6$ per hour or about 100 times that in a typical $\mu$SR measurement.


