ELSEVIER

Physica B 284–288 (2000) 1700–1701

PHYSICA B

www.elsevier.com/locate/physb

Double-spin-flip mode of rhodium nuclei

Tauno Knuuttila[a,*], Juha Martikainen[a], Finn Rasmussen[b], Juha Tuoriniemi[a]

[a] *Low Temperature Laboratory, Helsinki University of Technology, P.O. Box 2200, FIN-02015 HUT, Finland*
[b] *Niels Bohr Institute, University of Copenhagen, Universitetsparken 5, 2100 København Ø, Denmark*

Abstract

We have observed the double-spin-flip mode, corresponding to one photon flipping two nuclear spins, in rhodium with low-frequency SQUID NMR at ultralow temperatures. The shift and magnitude of the absorption line at around twice the Larmor frequency has been studied. The shift of the double-spin-flip mode can be used to calculate the relative strength of the Ruderman–Kittel exchange interaction, for which we obtain $R = -0.9 \pm 0.1$. © 2000 Elsevier Science B.V. All rights reserved.

Keywords: Double spin flip; Rhodium; SQUID NMR

In solids, the dipole–dipole interaction between the nuclei allows one photon to flip two or more spins, introducing resonance lines at integer multiples of the Larmor frequency, $f_0 = \gamma B/2\pi$. These lines can be observed when the external magnetic field becomes comparable in size with the internal fields arising from the nuclear magnetic moments. Experimentally, the double-spin-flip resonance line was first observed indirectly by Anderson [1]. In copper, the frequency shift of the double-spin-flip line was used to determine the strength of the indirect exchange interaction [2].

We have observed the double-spin-flip resonance in rhodium metal, in both positive and negative temperatures, by utilizing low-frequency continuous wave NMR.

The sample was a high-purity single crystal with dimensions $25 \times 4 \times 0.4$ mm^3 [3]. It was polarized in a powerful cascade nuclear demagnetization cryostat [4]. The first nuclear stage, a massive copper stage with 97 moles in 9 T field, was able to cool the Rh-sample to below 100 μK for up to several weeks. The sample, acting as a second nuclear cooling stage, was polarized by a 7.5 T magnet. After a typical polarization time of 50–70 h, the sample was adiabatically demagnetized to the measuring fields below 1 mT. The highest nuclear polarization obtained was $p = 0.86$.

The negative spin polarizations were produced by inverting an external magnetic field of 400 μT in about 1 ms, a time much shorter than the spin–spin relaxation time of rhodium, 10.2 ms. Due to the bulk nature of the sample, the losses were considerable and the maximum negative polarization achieved was $p = -0.49$.

The NMR signal was recorded with a superconducting planar second-order gradiometric pick-up coil connected to a DC-SQUID. The sample was oriented so that the polarizing field and the pick-up were along the longest dimension, and the NMR measuring field along the second longest side. The spectra were measured by sweeping the magnetic field, as frequency sweeps would have produced additional difficulties in later analysis, due to the varying skin depth. An AC-excitation field of 50 nT was used to measure the double-spin-flip signals.

Fig. 1 shows two resonance spectra measured at $p = 0.39$ and -0.39, respectively. The excitation frequency was 431 Hz and the magnetic field was swept from 600 μT down to zero in 8 min. The phase of the recorded signal has been adjusted to best fit a Lorentzian line to the Larmor peak and a constant background has been subtracted. From the figure, it can immediately be noted, that at positive temperatures the mutual interactions between the spins shift the resonances closer to each

---

[*] Corresponding author.
E-mail address: tauno.knuuttila@hut.fi (T. Knuuttila)

0921-4526/00/$ - see front matter © 2000 Elsevier Science B.V. All rights reserved.
PII: S0921-4526(99)02944-0

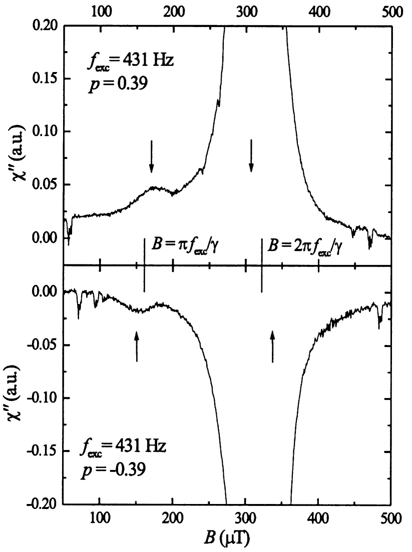

Fig. 1. Absorption parts of resonance spectra measured at $p = 0.39$ and -0.39. The two vertical lines, $B = \pi f_{\mathrm{exc}}/\gamma$ and $B = 2\pi f_{\mathrm{exc}}/\gamma$, show the Larmor and double-Larmor positions, whereas the arrows indicate the peak positions from Lorentzian fits to the resonances. The main Larmor line is slightly asymmetric, which makes the peak position appear off center. The peak of the signal and the center frequency of the fit do, however, coincide very well.

other, whereas at negative temperatures the peaks move further away from each other.

The double-spin-flip satellite was clearly observable at excitation frequencies from 131 Hz up to 831 Hz. At lower frequencies the satellite was drowned by the Larmor peak, whereas at higher frequencies the intensity, behaving as $1/f^2$, became too small.

The resonance frequencies of the Larmor and the double-spin-flip line are given by [2,5]

$$f_1 = f_0 + \tfrac{1}{2}(1 - 3D)f_s p, \qquad (1)$$

$$f_2 = 2f_0 + 2(R + \tfrac{1}{3} - D)f_s p, \qquad (2)$$

where D is the demagnetization factor along the static field and $R = \sum_j J_{ij} I/hf_s$ is the relative exchange parameter. $f_s = \mu_0 \mu^2 \rho / Ih + f_{ae}$ is the "dipolar frequency", where ρ is the atom number density and f_{ae} the anisotropic exchange part.

From the measured shifts of the resonance lines we can deduce the values of R and f_s. Taking into account the effects of eddy currents and the demagnetization factor $D = 0.08$, we obtain $R = -0.9 \pm 0.1$ and $f_s = 74 \pm 6$ Hz. Taking just the pure dipolar interaction for Rh gives $f_s = 54$ Hz, and the remaining shift is attributed to the anisotropic indirect exchange term $f_{ae} = 20 \pm 6$ Hz [6].

Acknowledgements

This work was supported by the EU program ULTI II.

References

[1] A.G. Anderson, Phys. Rev. 115 (1959) 863.
[2] J.P. Ekström et al., Physica B 98 (1979) 45.
[3] MaTeck Büro für Forschungsmaterialien Karl Heinz-Bechurts-Str. 13, D-52428 Jülich, Germany.
[4] W. Yao et al., submitted to J. Low-Temp. Phys.
[5] P.L. Moyland, P. Kumar, J. Xu, Y. Takano, Phys. Rev. B 48 (1993) 14020.
[6] A. Narath, A.T. Fromhold Jr., E.D. Jones, Phys. Rev. 144 (1965) 428.