β-Radiation Detected NMR in Solid and Liquid Neon *

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Z. Naturforsch. **49a**, 27–30 (1994); received July 23, 1993

Spin-polarized β-active ^{23}Ne ($T_{1/2}=37.2$ s) nuclei were created by capture of polarized cold neutrons in solid and liquid neon. The angular asymmetry of the β-radiation was used to monitor the nuclear polarization. From nuclear magnetic resonance curves the value for the dipole moment $|\mu(^{23}\text{Ne})|=1.079(1)$ n.m. (uncorrected) was obtained which is more precise than the literature value. Comparing the measured spin-lattice relaxation rates with those of stable ^{21}Ne the nuclear quadrupole moment of ^{23}Ne was determined. It agrees with a shell model calculation. The feasibility of ^{23}Ne-β-NMR for investigations in condensed rare gases in demonstrated.

Key words: β-radiation detected NMR, Solid and liquid Neon, $\mu(^{23}\text{Ne})$, $Q(^{23}\text{Ne})$, Nuclear spin relaxation.

1. Introduction

Nuclear magnetic resonance (NMR) is an important tool for the investigation of diffusive motion in rare gas solids and liquids [1, 2]. In neon, pulsed NMR experiments with the host species $^{21}\text{Ne}(I=\frac{3}{2})$ have been performed for both the solid and the liquid phases [3, 4]. It was found that quadrupolar processes dominate the spin-lattice relaxation (SLR) in the solid as well as in the liquid. From the temperature dependence of the SLR rate T_1^{-1} the self diffusion coefficients were determined.

Here first results of measurements with the short-lived β-emitter $^{23}\text{Ne}(I=\frac{5}{2}$, $T_{1/2}=37.2$ s) as NMR probe in condensed neon are reported. In the next section the experimental method will briefly be outlined and the sample cell for neon condensation will be described. Then the results of measurements of the ^{23}Ne resonance line and of the SLR rate T_1^{-1} in the solid and the liquid will be shown.

2. Experimental

In β-radiation detected NMR the angular asymmetry of the β-radiation of an ensemble of spin-polarized short-lived β-emitters in a sample is used for the measurement of the nuclear polarization. The β-emitters are produced in an on-line nuclear reaction. A detailed review on the method and its applications has been given in [5]; therefore only a brief description will follow.

The experiments were performed with the β-NMR spectrometer in the external neutron guide laboratory ELLA at the research reactor FRJ-2 of KFA Jülich [6]. β-active ^{23}Ne nuclei were produced in an (n, γ)-capture reaction with cold polarized neutrons from a neutron guide. The condensed neon sample containing the ^{22}Ne target nuclei was placed in the gap of an electromagnet. The β-radiation was detected by two double-coincidence counter telescopes mounted between the sample and the pole faces. The β-radiation asymmetry a_β is given by

$$a_\beta=\frac{Z_N-Z_S}{Z_N+Z_S}, \tag{1}$$

where Z_N and Z_S denote the counting rates in the northern and southern detectors, respectively. a_β is proportional to the dipolar nuclear polarization P thus allowing the detection of any changes of P due to resonance and/or relaxation via a_β.

The spectrometer can be run in two modes. In the time-resolved mode the time dependence of a_β and thus P is monitored after neutron activation pulses

* Presented at the XIIth International Symposium on Nuclear Quadrupole Resonance, Zürich, July 19–23, 1993.

** Also at: Institut für Festkörperforschung, Forschungszentrum Jülich.

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and the SLR rate T_1^{-1} can directly be obtained from the decay behaviour of $P(t)$.

In the time-integral mode the neutron beam is chopped with intervals short compared to the β-decay lifetime, thus achieving quasi-stationary activation conditions. For reduction of background counting rates the detection of β's is restricted to the 'beam-off' phases. In this mode, resonance curves are measured by irradiating the sample with radio frequency (rf) fields produced by a coil wound around the sample.

In both modes the neutron spin direction is reversed periodically to eliminate instrumental asymmetries. Important features of the β-NMR method for the present investigation of neon are the following: The probe nuclei are extremely diluted (typical abundance 10^{-18}), thus homonuclear dipole-dipole interactions of the probe nuclei are negligible and a spin temperature is not established. Furthermore, as a result of inhibited spin diffusion, β-NMR is generally not sensitive to paramagnetic impurities. Since ^{21}Ne is the only stable neon isotope with $I \neq 0$, β-NMR makes another probe nucleus available for the study of different SLR contributions. A disadvantage of the variant of β-NMR applied here is the need for an intense polarized neutron beam. Furthermore the observable SLR times T_1 are restricted to roughly $0.01\, T_{1/2} \leq T_1 \leq 100\, T_{1/2}$.

In all experiments reported here neon of natural isotopic composition (purity above 99.995%, supplied by Messer-Griesheim) was used. The neon was condensed into a cell mounted in the bath cryostat of the spectrometer. In order to reduce absorption of the β-particles from the ^{23}Ne decay, with an endpoint β-energy of about 4.4 MeV, the cell essentially consists of a metal frame with Mylar windows. The windows were sealed with indium wire. The cell was tested with a pressure of 10 bar thus allowing an extension of the liquid state range to higher temperatures. For resonance experiments the cell was equipped with an rf coil. The neon was filled through a stainless steel capillary connection with the room temperature port of the cryostat. The gas system outside the cryostat consisted essentially of a neon reservoir connected via a valve and a manometer. In all experiments the condensed neon was in thermodynamic equilibrium with the gas phase. For measurements in the liquid state the vapor pressure was used as input for a PID-temperature regulation using a heater consisting of a resistance wire wound around the sample cell. With this method of temperature control, the temperature could be kept constant within ± 0.06 K for several hours. For measurements in solid neon the standard temperature controller of the cryostat had to be used. This was less stable (deviation about ± 0.5 K over several hours), the temperature sensor being less sensitive.

3. Results and Discussion

3.1 Larmor resonance line of ^{23}Ne

In the time-integral mode of the spectrometer the β-asymmetry of ^{23}Ne was measured during irradiation of an rf field with increasing frequency ν swept over a region around the Larmor frequency. Examples for β-NMR signals in liquid and solid neon are shown in Figure 1. In order to increase the resonance signal the rf was frequency modulated with a width corresponding to the frequency increments of the scans. The lines were described by Lorentzian line-shape functions with linewidths of roughly 5 kHz.

The position of the resonance line can be used for a determination of the nuclear dipole moment μ of ^{23}Ne. For this purpose the value of the B field has to be determined precisely at the position of the sample cell. This was achieved by filling the cell with ^{7}LiF powder and making a measurement of the resonance frequency ν_L of the β-probe nucleus ^{8}Li ($I=2$, $T_{1/2}$, = 0.84 s). From the resonance experiment in liquid neon and a follow-up experiment on ^{8}Li with the same value of B, the dipole moment of ^{23}Ne is given by

$$|\mu(^{23}\mathrm{Ne})| = \frac{\nu_L(^{23}\mathrm{Ne})\, I(^{23}\mathrm{Ne})}{\nu_L(^{8}\mathrm{Li})\, I(^{8}\mathrm{Li})}\, \mu(^{8}\mathrm{Li})\,, \qquad (2)$$

where I denote the nuclear spins. The dipole moment of ^{8}Li is known with a high precision [7]. Systematic errors of this method for the determination of the nuclear dipole moment arise from a possible shift of the B field during the measurements of $\nu_L(^{23}\mathrm{Ne})$ and $\nu_L(^{8}\mathrm{Li})$ and a temperature dependent susceptibility of the stainless steel cryostat vessel since the β-NMR signal of ^{8}Li was recorded at higher temperatures. Possible drifts of the regulation of the magnet power supply are estimated to be below 10^{-4} within the run time of the measurements. A larger systematic error arises from shifts of B due to the variation of the susceptibility of stainless steel of the cryostat with temperature. From the decrease of B with decreasing temperature observed in a former measurement [8], a correction of 5×10^{-4} is estimated yielding

$$|\mu(^{23}\mathrm{Ne})| = 1.079(1)\ \text{n.m.} \qquad (3)$$

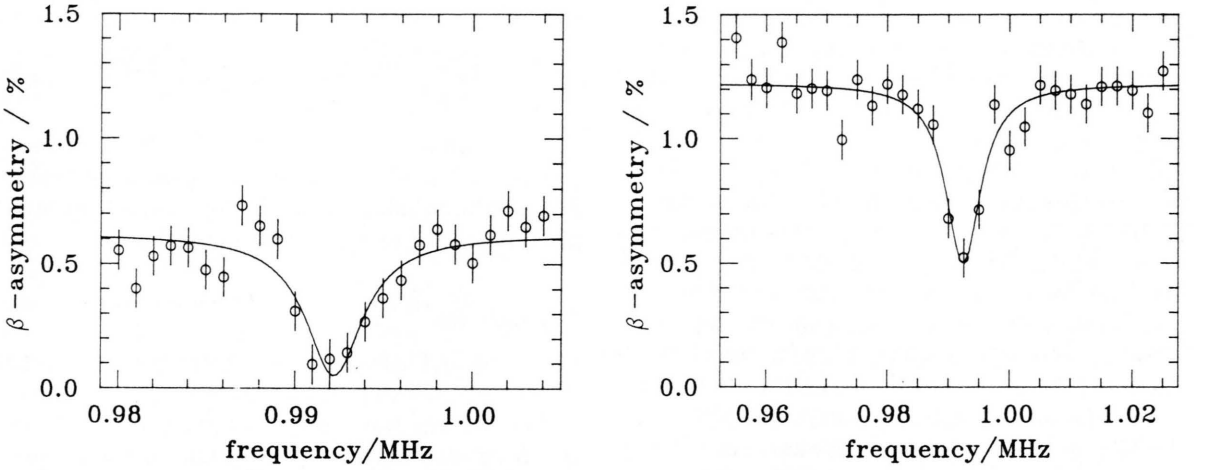

Fig. 1. β-NMR signal of ^{23}Ne at $B = 300$ mT in liquid neon at $T = 27.4$ K (left) and solid neon at $T = 19.7$ K (right).

which is much more precise than the literature value $-1.08(1)$ n.m. obtained by an atomic beam resonance method [9]. Correction for the chemical shift tabulated in [10] yields $|\mu(^{23}\text{Ne})| = 1.0795(10)$ n.m. Furthermore, in rare gases Van der Waals and exchange interactions lead to a small positive shift of the magnetic field at the nucleus. This shift increases with the density. It was experimentally observed for ^{83}Kr and ^{129}Xe [11] and amounted to 6.5×10^{-4} and 2×10^{-4}, respectively. For Ne no data are available.

3.2 SLR of ^{23}Ne in solid and liquid neon

In the time-resolved measuring mode of the spectrometer transients of the β-asymmetry were recorded in solid and liquid neon. Although the measurements suffered from background counting rates of approximately twice the counting rates of β's from ^{23}Ne, the transients could be analysed in terms of a $a_\beta(t) \propto \exp(-t/T_1)$ decay behaviour. The errors for the SLR rates T_1^{-1} were about 50% in the solid and about 10% in the liquid. Figure 2 is a semilogarithmic plot of T_1^{-1} of ^{23}Ne obtained at 300 mT as a function of inverse temperature, as usually used for the analysis of diffusion induced SLR rates. For comparison, quadrupolar SLR rates of ^{21}Ne taken from [3] are included. For both nuclei the same pronounced reduction of the SLR rate for the solid with respect to the liquid is observed. This observation was also made in T_1^{-1} measurements with ^{83}Kr and ^{131}Xe [12]. The shift of the T_1^{-1} scales in Fig. 2 should be noted. In the following it will be used for the estimate of the ratio of the nuclear quadrupole moments of ^{23}Ne and ^{21}Ne.

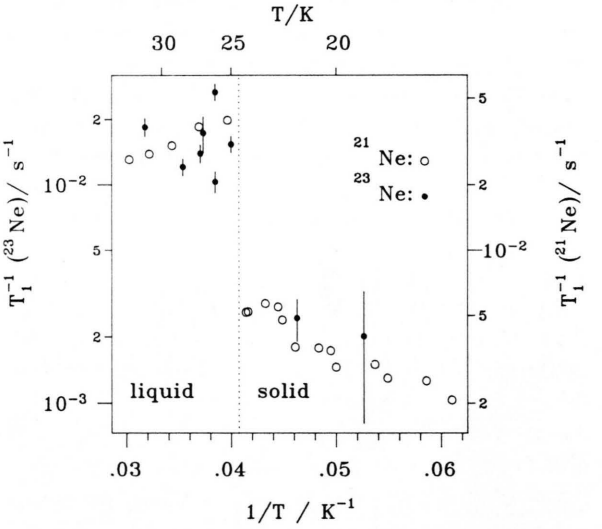

Fig. 2. Spin-lattice relaxation rate of ^{23}Ne (left hand scale, filled circles, $B = 300$ mT) and quadrupolar SLR rate of ^{21}Ne (right hand scale, open circles, $B = 892$ mT, data taken from [3]) as a function of inverse temperature in solid and liquid neon. From the shift of the T_1^{-1} scales the ratio of nuclear quadrupole moments is estimated.

The SLR of ^{23}Ne in the solid as well as in the liquid is predominantly due to quadrupolar coupling. From the calculation of the dipolar SLR contribution of 5.9×10^{-3} s^{-1} for ^{21}Ne in the solid at 24.5 K given in [3], the dipolar contribution to T_1^{-1} of ^{23}Ne in the solid due to coupling to ^{21}Ne can be estimated by applying the scaling factor $[\gamma^2(^{23}\text{Ne})\, I(I+1) n_1]/[\gamma^2(^{21}\text{Ne})\, S(S+1) n_2]$ and accounting for the difference in Larmor frequencies. γ denotes the gyromag-

netic ratio, I, S the spins of ^{23}Ne and ^{21}Ne and n_1, n_2 the isotopic abundance of ^{21}Ne in the respective measurements. A dipolar SLR contribution of 6.1×10^{-4} s^{-1} to T_1^{-1} of ^{23}Ne is estimated to be negligible by comparison with the observed rates (see Figure 2). The relatively small ^{23}Ne dipolar SLR rate is due to the low isotopic abundance of ^{21}Ne in natural Ne(0.26 at.% ^{21}Ne) used in the β-NMR measurement with respect to that of the ^{21}Ne NMR study [3] where the sample was isotopically enriched to 51 at.% ^{21}Ne. Since homonuclear dipole-dipole interactions of the ^{23}Ne probe nuclei are negligible due to their extreme dilution, ^{21}Ne are the only magnetic nuclear species effective for dipolar SLR in the case of β-NMR. Hence β-NMR on ^{23}Ne with isotopically enriched ^{22}Ne offers the possibility of studying purely quadrupolar induced SLR.

In the liquid, the experimental conditions lie well within the extreme motional narrowing regime and the calculated dipolar T_1^{-1}-contributions are below 10^{-7} s^{-1} [3]. Thus the measured SLR rates of ^{23}Ne may be regarded as quadrupolar rates T_{1Q}^{-1}. Assuming the same relaxation mechanisms as for the quadrupolar SLR rate of ^{21}Ne in both the solid and the liquid, the following relation holds [13]

$$\frac{T_{1Q}^{-1}(^{23}\mathrm{Ne})}{T_{1Q}^{-1}(^{21}\mathrm{Ne})} = \frac{Q^2(^{23}\mathrm{Ne})}{Q^2(^{21}\mathrm{Ne})} \frac{s(5/2)}{s(3/2)} \tag{4}$$

with

$$s(I) = \frac{2I+3}{I^2(2I-1)} \tag{5}$$

and Q the nuclear quadrupole moment. From the comparison of the SLR rates of the two isotopes the ratio of the quadrupole moments can be obtained as

$$\left|\frac{Q(^{23}\mathrm{Ne})}{Q(^{21}\mathrm{Ne})}\right| = 1.40(36)\,. \tag{6}$$

With the known value $Q(^{21}\mathrm{Ne}) = +0.1029(75)$ barn [14] the value $|Q(^{23}\mathrm{Ne})| = 0.14(4)$ barn is obtained. This is in accordance with $Q(^{23}\mathrm{Ne}) = +0.144$ barn as given by a shell model calculation [15].

4. Conclusion

In these first experiments the feasibility of using the β-emitter ^{23}Ne as a probe for the investigation of solid and liquid neon was shown. The potential of β-NMR covers the measurement of NMR lines and SLR rates. Here the NMR line position was used for a determination of the nuclear magnetic moment of ^{23}Ne, and measurements of the SLR rate of ^{23}Ne were used to estimate the nuclear quadrupole moment. The use of isotopically ^{22}Ne enriched gas for the experiment should increase the signal/noise ratio such that a detailed investigation of the temperature dependence of T_1^{-1} becomes feasible. This sample is available and awaits the restart of the reactor. Then a system will be accessible for NMR studies, where no dipole-dipole interaction is effective. A further topic could be the investigation of the SLR in neon doped with other noble gases which would offer a system for the study of electric field gradients of defects in Van der Waals crystals.

Acknowledgement

The hospitality of the Institut für Festkörperforschung, KFA Jülich, is gratefully acknowledged. This work was sponsored by the BMFT.

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