Early $^8Li^+$ β-NMR investigations in GaAs and Ge

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Abstract

In this paper, we describe initial studies of the structure and dynamics associated with $^8Li^+$ in bulk crystalline GaAs and Ge. At low temperatures in GaAs, the amplitude of the $^8Li^+$ resonance signal at ≈ 3 T indicates that a large fraction (at least 70%) of the Li end up in locations with cubic symmetry (i.e. the tetrahedral interstitial and substitutional sites). The linewidth of the β-NMR $^8Li^+$ resonance increases dramatically above 150 K, reaches a maximum at about 290 K, and decreases again. This suggests that the Li starts to change its location, probably from an interstitial to a substitutional site, at ≈ 150 K. Experiments in Ge are also described. In this sample, a narrow resonance is seen at low temperatures that is likely due to Li located at an interstitial site. Near room temperature, it appears that Li is converting to another site.

Key words: β-NMR, lithium, GaAs, Ge

1. Introduction

Recently, new facilities for carrying out $^8Li^+$ beta-detected nuclear magnetic resonance[1] (β-NMR) and beta-detected nuclear quadrupole resonance[2] (β-NQR) investigations of condensed matter systems were constructed at TRIUMF. One of the research programs that is being carried out using this technique is the investigation of the kinetics and structure of $^8Li^+$ in *bulk* crystalline semiconductors. There are a number of reasons for such experiments: (1) Li is an important impurity in many semiconductors, including GaAs and Ge. (2) Semiconductors are often used as substrates upon which thin films are grown, and hence must be understood in order to interpret results in such systems correctly. (3) Experiments in bulk semiconductors serve as a point of comparison for planned future studies in low-dimensional structures.

Here, we report of our early β-NMR studies on bulk GaAs and Ge. Future prospects are also de-

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scribed.

2. Experimental

More details on the $\beta-$NMR and $\beta-$NQR facilities can be found in other recent publications[1,2]. They are only briefly described in this section.

The Isotope Separator and Accelerator (ISAC) facility at TRIUMF has the ability to deliver a continuous beam of $\approx$ 30 keV ^{8}Li$^+$ with a typical flux of $\approx 10^7$/s. The ^{8}Li nucleus has a spin of 2, a lifetime of $\tau = 1.21$s, a gyromagnetic ratio $^8\gamma = 630.15$Hz/G, and an electric quadrupole moment $Q = +33$mB. The ^{8}Li$^+$ is then spin polarized using an optical pumping scheme that consistently produces ^{8}Li$^+$ with nuclear polarizations as high as $\approx$ 70%. Two stations, a low field spectrometer ("$\beta-$NQR" station, up to 150 G) and a high field spectrometer ("$\beta-$NMR" station, up to 9 T), are available for condensed matter research. The Li enters the $\beta-$NQR station with polarization transverse to the beam momentum while it enters the $\beta-$NMR station with polarization longitudinal to the beam momentum. In both situations, any static magnetic field is applied longitudinal to the initial Li polarization. The majority of the measurements reported here were undertaken on the $\beta-$NMR station.

In the resonance measurements, such as those reported in this paper, a linearly polarized RF magnetic field (perpendicular to the nuclear polarization) is stepped through a range of frequencies. A resonant loss of polarization, as reflected in the asymmetry of the beta decay particles in the directions forward and backward to the initial Li spin, is observed when the RF frequency matches the resonance frequency for the ^{8}Li nucleus. Spin-lattice relaxation, i.e. $1/T_1$, of the implanted ^{8}Li$^+$ can also be measured. Here, the ^{8}Li$^+$ beam is pulsed and the time dependence of the asymmetry is measured (in the absence of the RF field) after the beam pulse has ended.

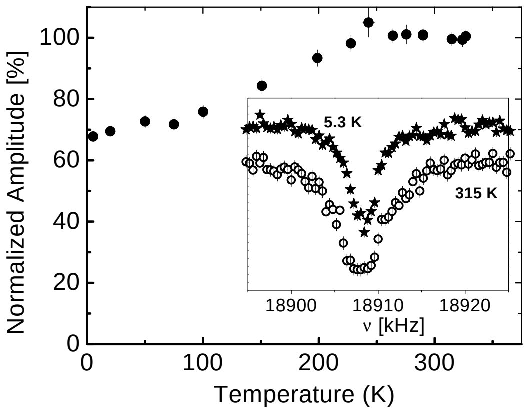

Fig. 1. Temperature dependence of the amplitude of the $\beta-$NMR resonance at the ^{8}Li$^+$ Larmor frequency in GaAs. The inset shows example resonance data for two temperatures. They are offset for clarity.

3. Results in GaAs and Discussion

Some initial $\beta-$NMR results in GaAs are discussed in this section. The experiments were conducted in the high-field spectrometer mentioned in Sec. 2. The ^{8}Li$^+$ is implanted into a semi-insulating GaAs sample ≈ 350 μm thick and the static magnetic field H_0 of approximately 3 T is applied parallel to a $\langle 100 \rangle$ direction.

The inset in Fig. 1 shows the $\beta-$NMR resonances near room temperature and at low temperature. At all temperatures studied, there is a signal of large amplitude at the ^{8}Li Larmor frequency, indicating that Li is located at highly symmetric sites and hence experiences no resolved quadrupole splitting. The possible cubic sites are the two interstitial tetrahedral or two substitutional sites in GaAs. As shown in Fig. 1, the amplitude of the resonance signal indicates that at low temperatures, about 70% of the Li end up in a high symmetry site, while at high temperatures, all of the Li are in such locations.

There is also a strong temperature dependence of the linewidth (assuming that the resonance lineshape is a Gaussian), as is shown in Fig. 2. At low temperatures, the linewidth is attributed to nuclear dipolar interactions between the static Li and other magnetic moments in the sample. The host ^{69}Ga, ^{71}Ga and ^{75}As nuclei (all of which

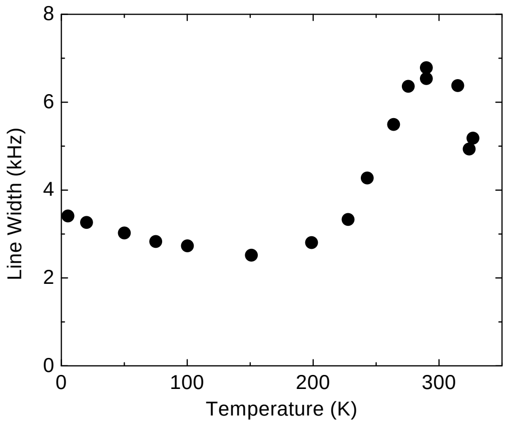

Fig. 2. Temperature dependence of the linewidth of the $\beta-$NMR resonance at the $^8Li^+$ Larmor frequency in GaAs.

have spin 3/2) are certain to contribute to the linewidth, as will intrinsic defects such as vacancies that are created during the implantation process. (There is also some additional broadening due to the RF power, but it is kept constant at all temperatures.) There appears to be a slight decrease in the linewidth from 5 K to 150 K, followed by a significant broadening above 150 K. The linewidth peaks at about 290 K before decreasing once again.

There is not enough information in our data to distinguish between the various high symmetry sites that are present in the GaAs zincblende lattice, or whether multiple sites are occupied at any particular temperature. However, comparison with other work on Li in GaAs provides clues. Particularly relevant publications are those of Lindner *et al.*[3] and Wahl[4] who reported investigations of the site changes associated with implanted Li in GaAs using α emission channeling. Lindner[3] concludes that below 220 K, Li preferentially occupies the tetrahedral interstitial site, whereas, above this temperature, substitutional sites are occupied: $Li_T^+ + V_{Ga}^{n-} \rightarrow Li_{Ga}^{(n-1)-}$ where $n = 2, 3$. The notation is as follows: Li_T^+ denotes Li^+ at the tetrahedral interstitial T position, V_{Ga} denotes the Ga vacancy, and Li_{Ga} symbolizes Li in a Ga substitutional site. Wahl[4] finds that in SI-GaAs, at 150 to 175 K, 59% of the Li^+ are in T sites, 22% are in S sites, and 19% are in random (R) sites. At 293 K, the fractions are f_T=15%, f_S=59% and f_R =26% respectively. This implies that at room temperature, significant conversion from T to S (substitutional) is occurring during the lifetime of the lithium. It is also useful to keep in mind that if isolated Li^+ (i.e. ionized donor) is in an interstitial position, the theoretically predicted stable equilibrium state is Li^+ in the T_{As} site, i.e. tetrahedral interstial surrounded by four nearest neighbor As atoms [5].

Based on these investigations, we suggest the following explanation for our data: at low temperatures, a significant fraction of the Li ends up in a T_{As} site with possible occupancy of a T_{Ga} or substitutional site. However, as the temperature is raised above $\approx$ 200K, the environment around many of the Li ions is changing significantly. These Li approach nearby implantation-created vacancies, and the broadening of the resonance signal is due to small unresolved quadrupolar splittings. At higher temperatures, Li preferentially occupies the V_{Ga} (e.g. substitutional Li_{Ga}) where the quadrupolar splitting is identically zero, and hence the linewidth is observed to decrease again. Whether such a picture is consistent with measurements of the intrinsic diffusivity of Li in GaAs[6] requires further analysis, as well as additional site sensitive measurements. In addition, we cannot at this stage rule out that the vacancy is moving, as suggested by Lindner *et al.* [3].

4. Results in Ge and Discussion

We have also initiated studies in an $n-$type Ge sample. The temperature dependence of the resonance spectra are shown in Fig. 3 under an applied magnetic field of 4.1 T along a $\langle 100 \rangle$ direction. As is seen, the resonance is sharp at low temperatures, but becomes significantly broader above $\approx$ 270 K. This is analogous to the behavior in GaAs described in Sec. 3. (The shift in the positions of the resonances could be due to a drift in the magnetic field, and is currently being investigated.) The intrinsic linewidth is much narrower in Ge than in GaAs, and is likely due to the fact that only 7.7% of the host Ge nuclei have non-zero spin (while 100% of the host nuclei in GaAs have non-zero spin).

A second peak at higher frequency makes its ap-

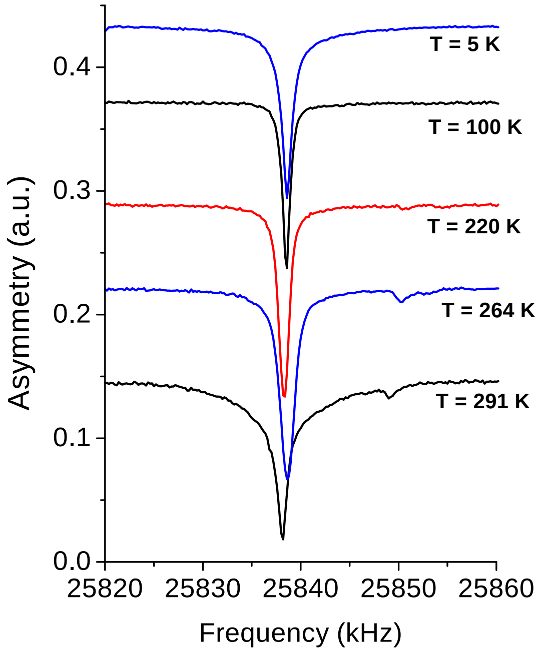

Fig. 3. $\beta-$NMR resonances at 4.1 T at various temperatures in Ge. The curves are offset vertically for clarity.

pearance above ≈ 270 K (at ≈ 25850 kHz in Fig. 3). The peak is currently unassigned but is probably due to conversion from the interstitial Li at low temperatures to another site at higher temperatures. The large frequency shift associated with Li in the latter is puzzling and is being subjected to further studies.

5. Summary and Future Prospects

In this paper, we have discussed some initial measurements of ^{8}Li in bulk GaAs and Ge using the new $\beta-$NMR facility at TRIUMF. Measurements that can provide site sensitive information such as level crossing cross-relaxation [7–11], double resonance, etc. are planned for the future in order to enhance our understanding of the behavior of Li in these semiconductors. The capability to carry out measurements at higher temperatures is being explored.

In contrast to μSR studies of semiconductors, interactions of the implanted probe with other defects and impurities in the sample appear to be quite prominent. This is partly due to the long lifetime of ^{8}Li (compared to μ^+). Furthermore, the ability to carry out $\beta-$NMR investigations on low dimensional materials was not utilized in the current studies, but is definitely a capability that we will make use of in the near future.

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