β-detected NMR Study on Semi-Insulating GaAs

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β-NMR at ISAC (TRIUMF) is an effective and sensitive probe to study isolated impurities in semiconductors[1,2]. β-NMR uses a beam of highly polarized ^{8}Li (τ =1.2 s, γ =6.301 Mhz/T, Q=+31.4(2)mB) ions implanted into the sample. The Li beam of 28 keV enters the spectrometer (shown right) from the left, passing through a hole in the back detector and is implanted into the sample sitting at the end of the spectrometer. The Li beam can be decelerated from this maximum implantation energy by biasing the spectrometer to a high positive voltage. The nuclear polarization is monitored by detecting the e^- that is emitted preferentially along the Li spin direction during the anisotropic β-decay: ^{8}Li $\longrightarrow$ ^{8}Be + e^- + ν_e

High-field spectrometer

HV Isolator & Einzel Lens; Deceleration Space; Load-lock & gate valve; Cryostat (2-500 K); Beta Scintillation Detectors; S.C. Solenoid; UHV chamber; Bellows; Cryopump; 500mm

Layout of beam lines

Optics bench; Optical pumping region; Low field spectrometer; High field spectrometer; ^{8}Li$^+$ 30 keV beam; Na neutralizer cell; He re-ionizer cell; Laser optics bench
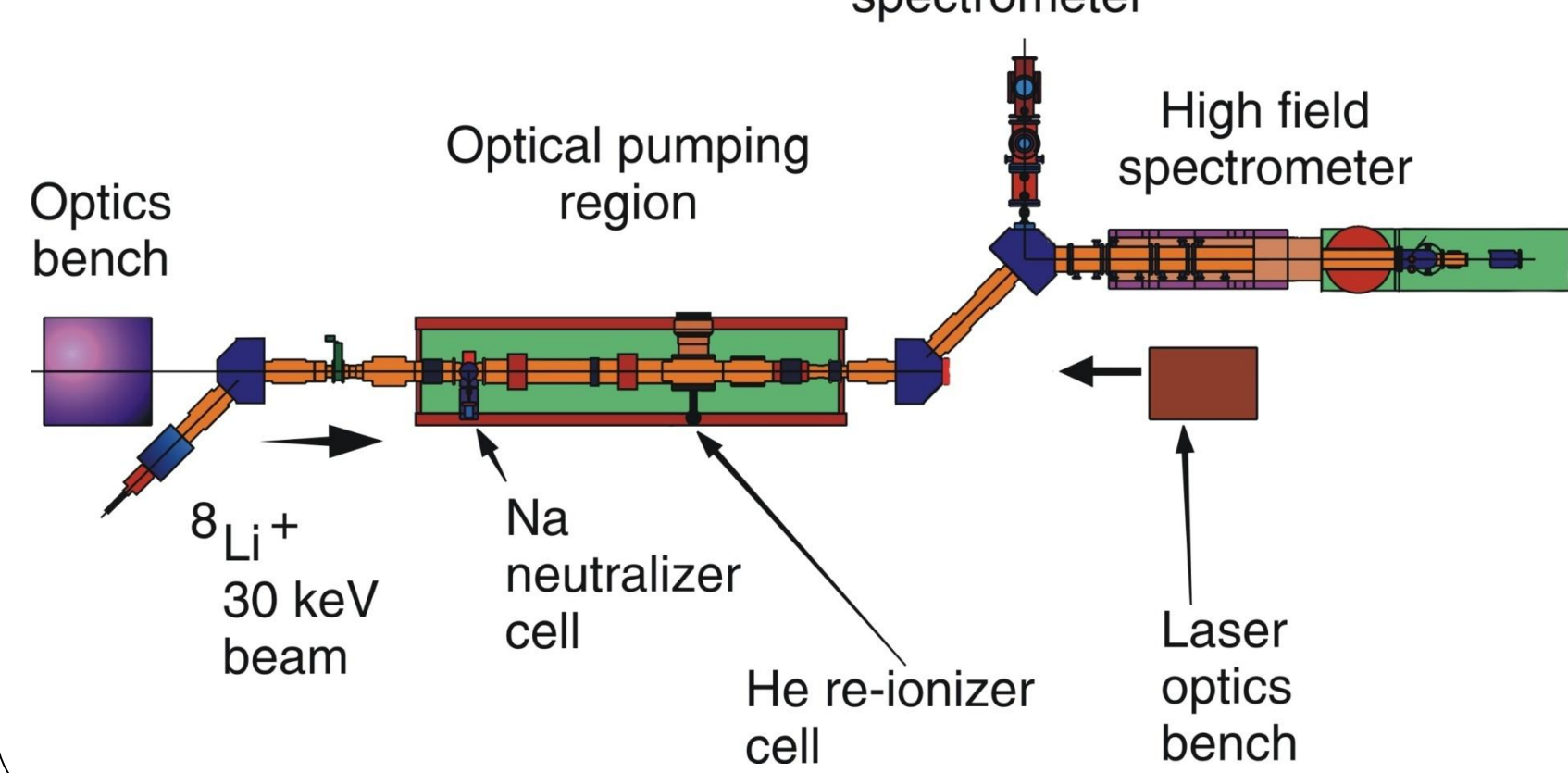
The β-NMR facility @ ISAC (TRIUMF)

In resonance measurement, a pulsed radio frequency (rf) field is applied perpendicular to the static magnetic filed (H_o=2.2T) with a frequency range ω ± Δω/2. And rf is on periodically for short time t_p~2π/Δω. The rf pulse is shaped to excite all spins in a frequency interval ω ± Δω/2. The difference in polarization after and before the pulse is proportional to the number of spins in that interval. With high repetition rate, frequency in rf mode is generated randomly to minimize memory effect and power broadening. Therefore, rf mode resonance is of higher resolution and provides details of magnetic structure in a sample.
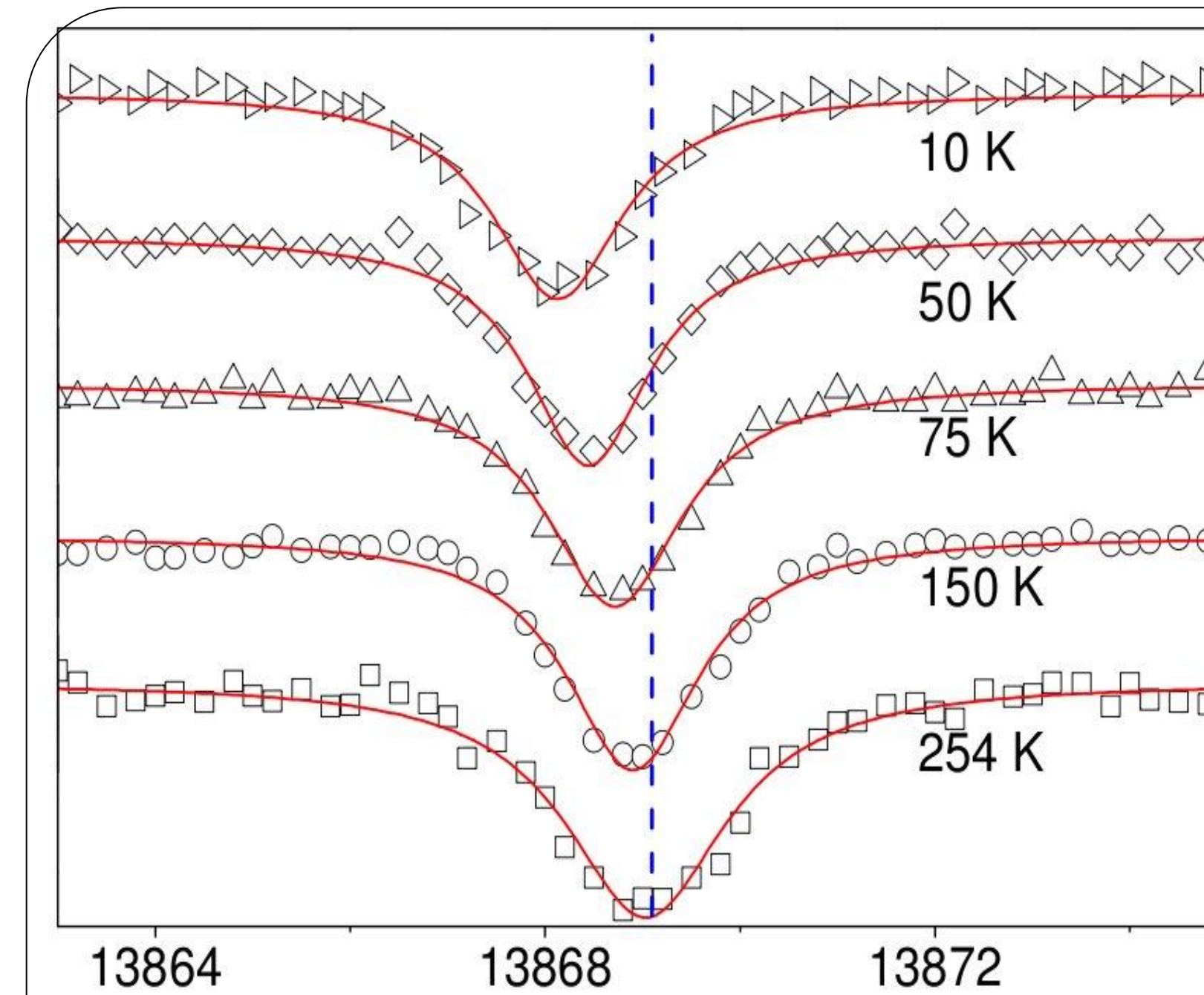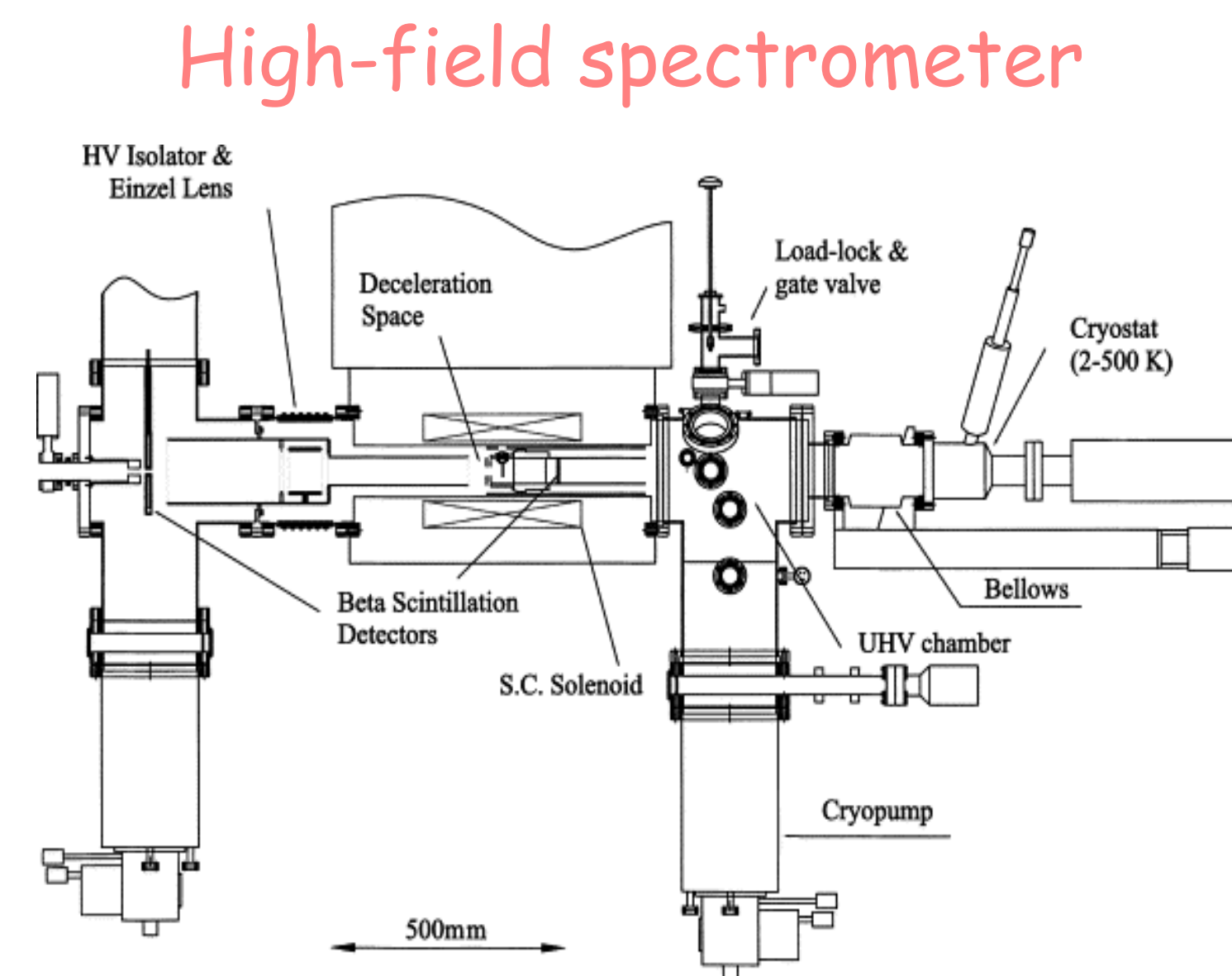
βNMR spectra as a function of temperature at full beam energy (28 keV). The resonance frequency shifts negatively when cooled, without any broadening. Spectra are offset for clarity. The resonance is fit to a single Lorentzian:

$$A(\nu) = \frac{A_o \sigma^2}{4(\nu - \nu_o)^2 + \sigma^2}$$

ν_o-frequency, A_o- amplitude, σ -width

The resonance shifts is calculated as
$$(\nu_o - \nu_{ref})/\nu_o$$
ν_{ref} -the resonance frequency of ^{8}Li in MgO at 300 K and converted to ppm

Temperature Dependence

➤ The resonance broadens above 150 K, consistent with CW mode data[5].

➤ The resonance starts to shift only below 50 K. The negative shift increases as temperature decreases and saturates at ~10 K.

➤ Above 150 K, the negative shift is small and temperature independent, consistent with Ga NMR shift measurements[6,7]. This shift may represent the chemical shift, which is generally small for Li.

MgO; saturated at ~10K
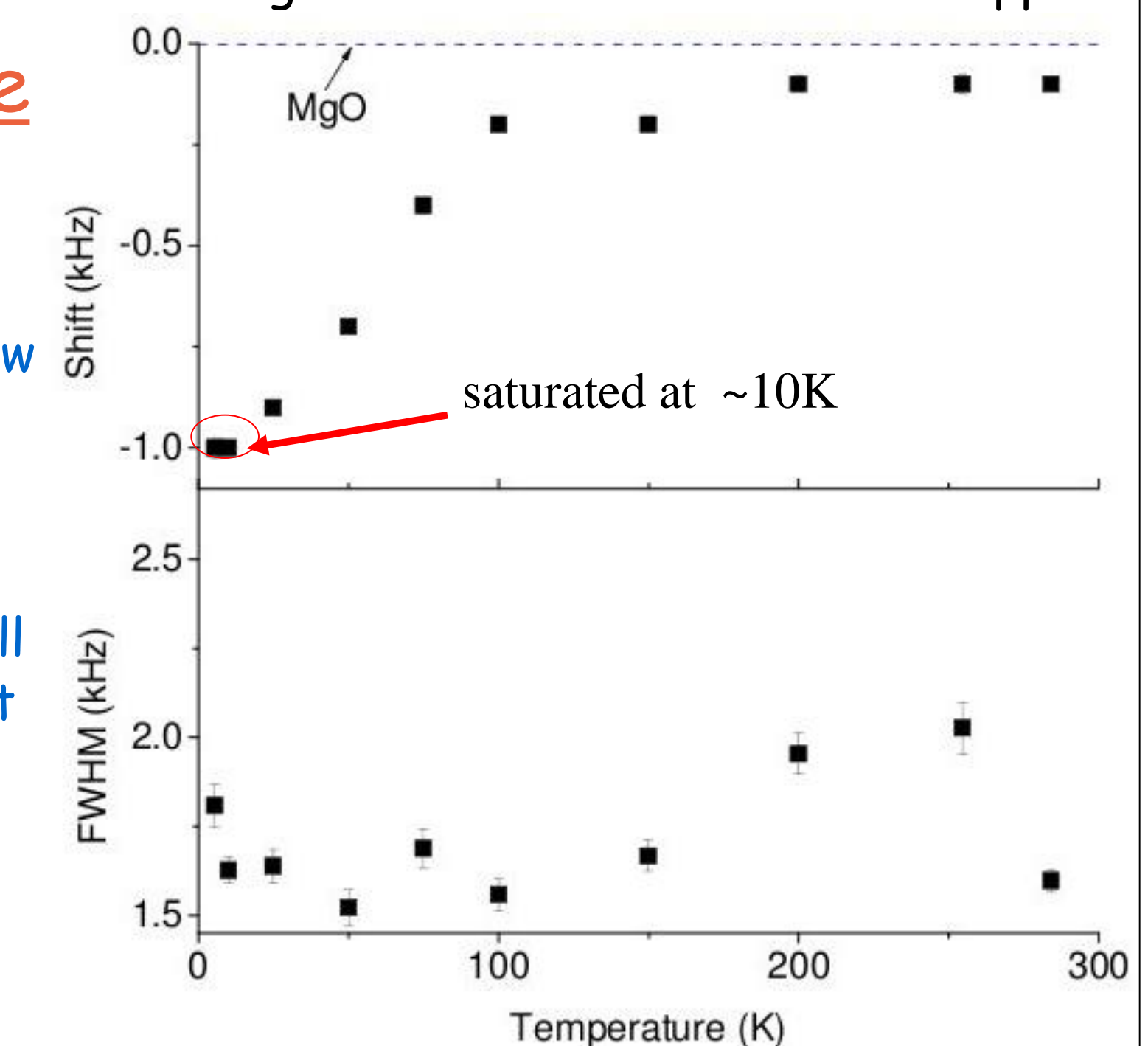
In Ref.[6], Fistul et.al. excluded the Knight shift because the carrier concentration is so low. And the shift saturation below 10 K indicates that it is NOT a paramagnetic shift resulting from the hyperfine field of paramagnetic impurities with low concentration.
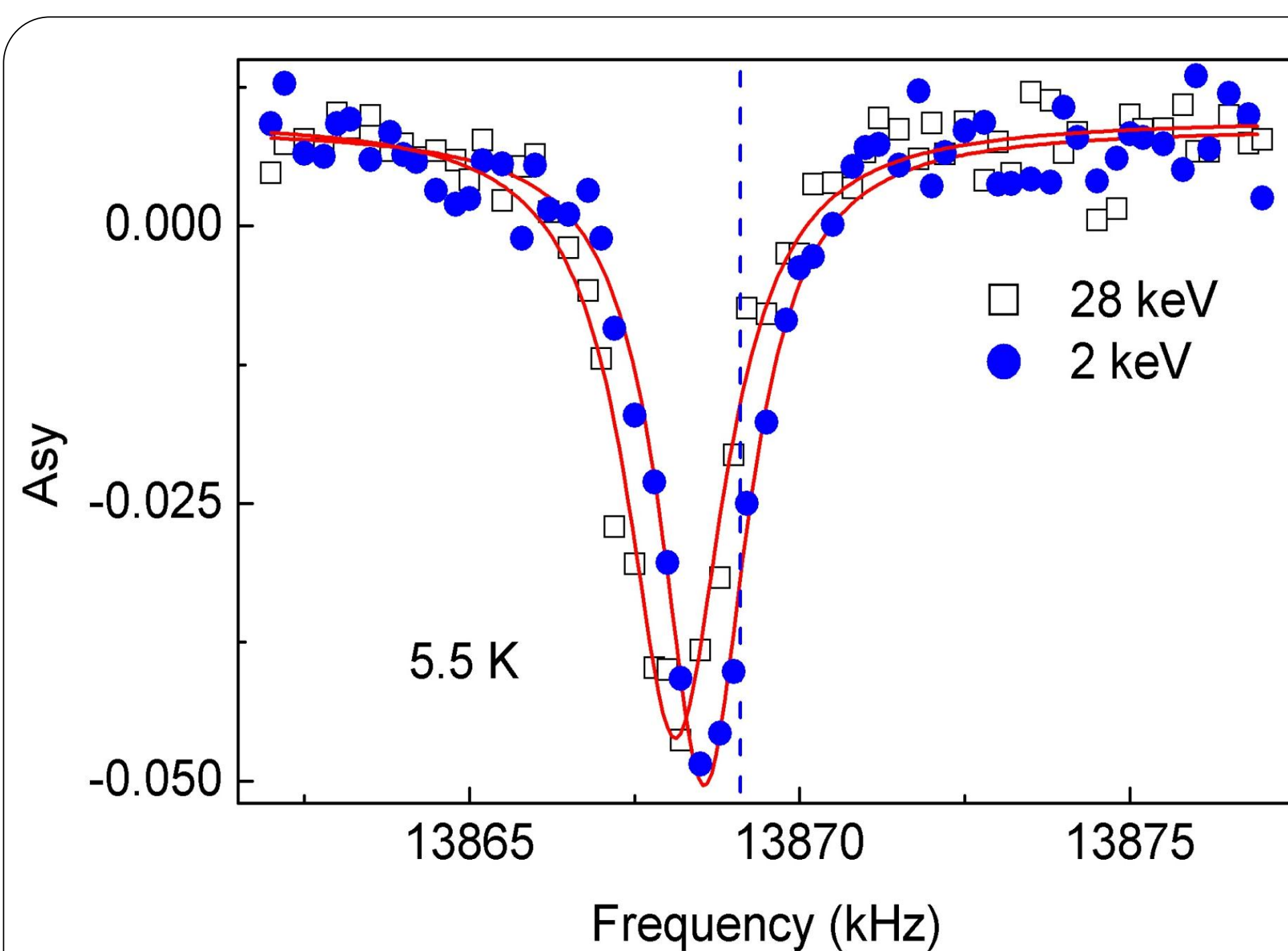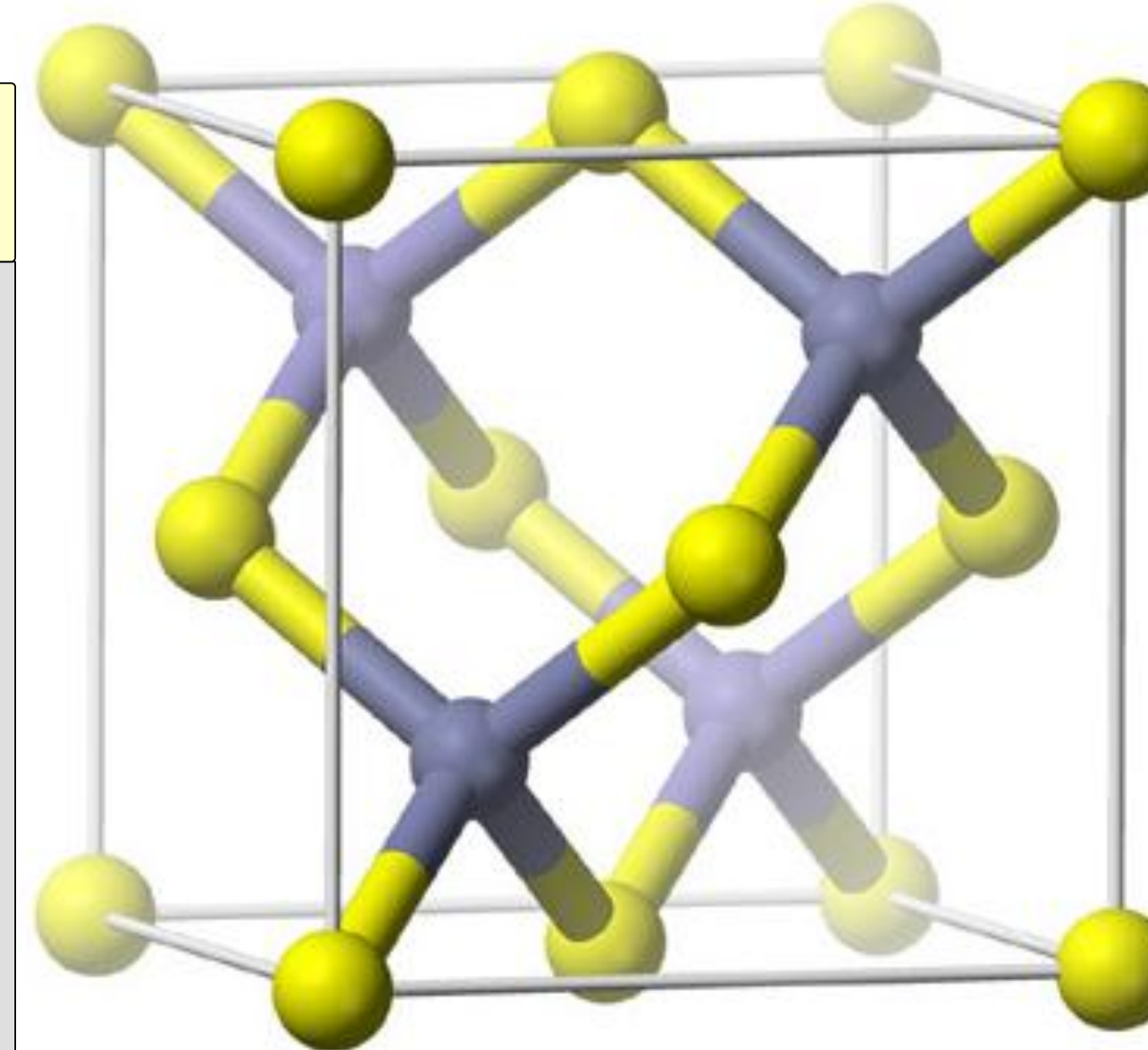
Other thin films (e.g. Fe)
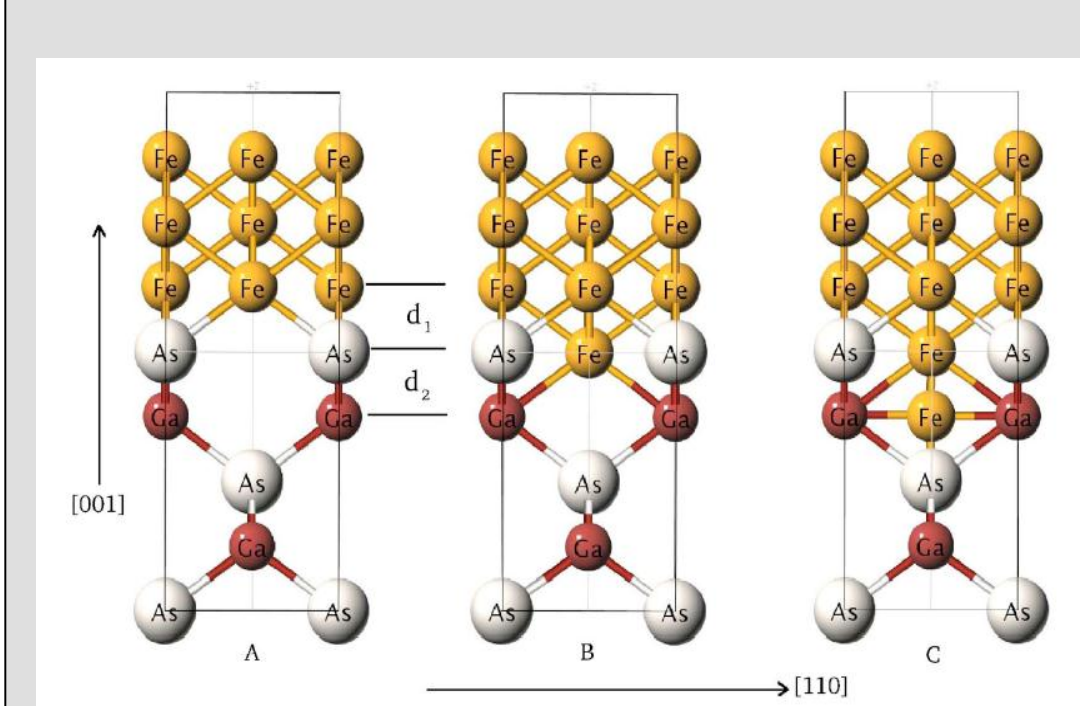
GaAs

➤ Diamagnetic semiconductor. Its susceptibility is small and weakly temperature-dependent[3]

➤ Li is one of the common impurities in GaAs, but little is known about the NMR of isolated Li in GaAs.

➤ Widely used as substrate in the hetero-structures, e.g. perfect lattice match with Fe.

➤ The ^{8}Li behaviour in GaAs crystal is the "baseline" of studies on GaAs-based heterostructures.

➤ GaAs (001) is a polar surface, and it reconstructs forming dimers. For a clean surface, there is no band-bending.

➤ Native oxide surface layer modifies the electronic structure. It may become negatively charged, creating a depletion layer in the near surface region.

In undoped GaAs, the depletion region is much larger than the implantation depth. However, some infrared illumination from the beamline upstream may photo-excite some carriers in the near surface region, and therefore results in a larger carrier concentration, which reduces the barrier width.

$GaAs$; Φ_o; E_c; E_F; vacuum; ω; E_v

$$w = \sqrt{\frac{2\epsilon\epsilon_0 e\Phi_o}{ne}}$$

ε : static dielectric permittivity
Φ_o: barrier height
n : carrier concentration

Some possible sources related to the observed shift:

➤ Magnetic field effects on far-infrared photoexcitation of compensating impurities in semi-insulating GaAs[8].

➤ light-induced ^{69}Ga hyperfine shift in semi-insulating GaAs[9].

Electrons trapped at the optically relevant defects interact with the nuclear spins. The hyperfine interaction between the localized electrons and surrounding nuclei, which is responsible for the nuclear polarization, is expected to produce a resonance shift[10].

βNMR spectra of the undoped GaAs crystal at 5.5 K with beam energy of 28 keV and 2 keV in the magnetic filed of 2.2 T. The blue dashed line is the reference resonance frequency of 8Li+ in MgO.

➤ This resonance shift is likely intrinsic to GaAs crystal.

➤ The shift at 2 keV is about -40ppm relative to MgO, comparable with Knight shift of Li in some noble metals[4].
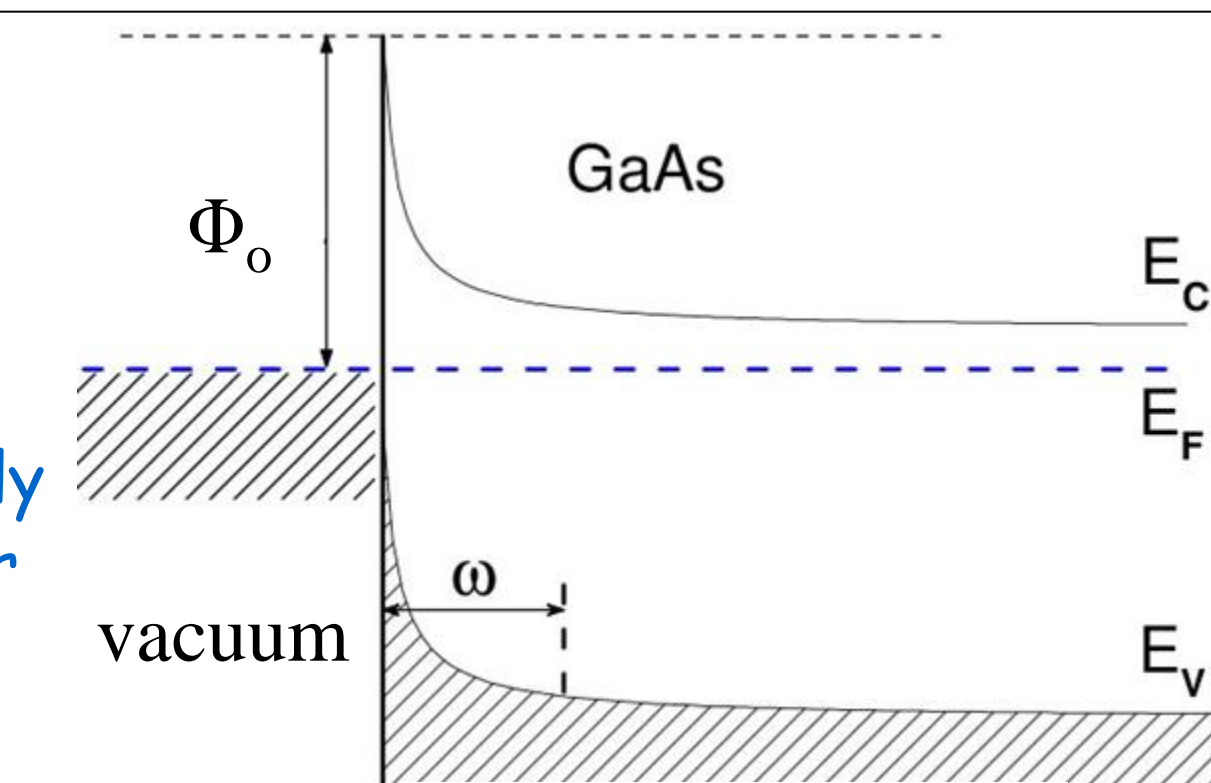
➤ The resonance shifts negatively without broadening as the beam stops deeper into GaAs.

Depth dependence

➤ The resonance shifts negatively at room temperature, and becomes more negative below 150 K.
➤ The low temperature shift is NOT likely a chemical or Knight shift.
➤ The resonance does NOT exhibit a Curie temperature dependence.
➤ The depth dependence suggests it may related to band bending. A non-equilibrium carrier distribution near the surface, caused by unintentional infrared illumination, may narrow the width down.
➤ The mechanism for the shift remains uncertain.

Conclusions

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