Conduction electron spin-lattice relaxation time in the MgB$_2$ superconductor

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The spin-lattice relaxation time, $T_1$, of conduction electrons is measured as a function of temperature and magnetic field in MgB$_2$. The method is based on the detection of the $z$ component of the conduction electron magnetization under electron spin resonance conditions with amplitude modulated microwave excitation. Measurement of $T_1$ below $T_c$ at 0.32 T allows to disentangle contributions from the two Fermi surfaces of MgB$_2$ as this field restores normal state on the Fermi surface part with $\pi$ symmetry only.

INTRODUCTION

The conduction electron spin-lattice relaxation time in metals, $T_1$, is the characteristic time for the return to thermal equilibrium of a spin system driven out of equilibrium by e.g. a microwave field at electron-spin resonance (ESR) or a spin-polarized current. The applicability of metals in “spintronics” devices in which information is processed using electron spins [1] depends on a sufficiently long spin life-time. In pure metals $T_1$ is limited by the Elliott mechanism [2, 3], i.e. the scattering of conduction electrons by the random spin-orbit potential of non-magnetic impurities or phonons. In superconductors, the Elliott mechanism becomes ineffective and a long $T_1$ is predicted well below $T_c$. Here we report the direct measurement of the spin-lattice relaxation time of conduction electrons in MgB$_2$ in the superconducting state. The motivation to study the magnetic field and temperature dependence of $T_1$ is two-fold: i) to test the predicted lengthening of $T_1$ to temperatures well below $T_c$, ii) to measure the contributions to $T_1$ from different Fermi surface sheets and to compare with the corresponding momentum life-times, $\tau$.

The lengthening of $T_1$ has been observed in a restricted temperature range below $T_c$ in the fulleride superconductor, K$_3$C$_{60}$ by measuring the conduction electron-spin resonance (CESR) line-width, $\Delta H$ [4]. This method assumes $1/T_1 = 1/T_2 = \gamma_e \Delta H$, where $\gamma_e/2\pi = 28.0$ GHz/T is the electron gyromagnetic ratio, and $1/T_2$ is the spin-spin or transversal relaxation rate. It is limited to cases where the homogeneous broadening of the CESR line due to a finite spin lifetime outweighs $\Delta H_{\text{inhom}}$, the line broadening from inhomogeneities of the magnetic field. In a superconducting powder sample, the CESR line is inhomogeneously broadened below the irreversibility line due to the distribution of vortexes, which is temperature and magnetic field dependent. This prevents the measurement of $T_1$ from the line-width and calls for a method to directly measure $T_1$. Electron spin echo techniques, which usually enable the measurement of $T_1$, are not available for the required nanosecond time resolution range. The magnetic resonance technique, termed longitudinally detected (LOD) ESR [5, 6] used in this work allows to measure $T_1$’s as short as a few ns. The method is based on the measurement of the electron spin magnetization along the magnetic field, $M_z$, using modulated microwave excitation. $M_z$ recovers to its equilibrium value with the $T_1$ time-constant, thus the method allows the direct measurement of $T_1$ independent of magnetic field inhomogeneities.

MgB$_2$ has a high superconducting transition temperature of $T_c = 39$ K [7] and its unusual physical properties [8, 9, 10, 11] are attributed [12, 13] to its disconnected, weakly interacting Fermi surface (FS) parts. The Fermi surface sheets derived from B-B bonds with $\pi$ and $\sigma$ characters ($\pi$ and $\sigma$ FS) have smaller and higher electron-phonon couplings and superconductor gaps, respectively, and contribute roughly equally to the density of states (DOS). The strange band structure leads to unique thermodynamic properties: magnetic fields of about 0.3-0.4 T restore the $\pi$ FS well below $T_c$ for all field orientations in polycrystalline samples but the material remains superconducting to much higher fields. This is characterized by a small and nearly isotropic upper critical field, $H_{c2}^\pi \sim 0.3 - 0.4$ T [10, 14] and a strongly anisotropic one, $H_{c2}^\sigma = 2 - 16$ T [11, 12] related to the $\pi$ and $\sigma$ Fermi surface sheets, respectively. Our measurements at low fields and low temperatures determine $T_1$ from the $\pi$ FS alone, while high field and high temperature experiments measure $T_1$ averaged over the whole FS. We find
that spin relaxation in high purity MgB$_2$ is temperature independent in the high field normal state between 3 K and 50 K, indicating that it arises from non magnetic impurities. Spin relaxation times for electrons on the $\pi$ and $\sigma$ Fermi surface sheets are widely different but are not proportional to the corresponding momentum relaxation times.

**EXPERIMENTAL**

The same MgB$_2$ samples were used as in a previous study [17]. Thorough grinding, particle size selection and mixing with SnO$_2$, an ESR silent oxide, produced a fine powder with well separated small metallic particles. The nearly symmetric appearance of the CESR signal [18] proves that penetration of microwaves is homogeneous and that the particles are smaller than the microwave penetration depth of $\sim 1 \mu$m. SQUID magnetometry showed that grinding and particle selection do not affect the superconducting properties. The particles are not single crystals but rather aggregates of small sized single crystals. Continuous wave (cw) and longitudinally detected ESR experiments were performed in a home-built apparatus [6] at 9.1 and 35.4 GHz microwave frequencies, corresponding to 0.32 and 1.27 T resonance magnetic fields. The 9.1 GHz apparatus is based on a loop-gap resonator with a low quality factor ($Q \sim 200$) and the 35.4 GHz instrument does not employ a microwave cavity at all. The cw-ESR was detected using an audio frequency magnetic field modulation. Line-widths are determined by Lorentzian fits to the cw-ESR data. For the LOD-ESR, the microwaves are amplitude modulated with $f = \Omega/2\pi$ of typically 10 MHz and the resulting varying $M_z$ component of the sample magnetization is detected with a coil which is parallel to the external magnetic field and is part of a resonant circuit that is tuned to $f$ and is matched to 50 Ohms. cw-ESR at 420 GHz (centered at 15.0 T) was performed at EPFL using a quasi-optical microwave bridge with no resonant cavities.

**RESULTS**

**Relaxation in the normal state**

The low temperature behavior of the spin-lattice relaxation time in MgB$_2$ in the normal state can be measured using cw-ESR from the homogeneous line-width, $\Delta H_{\text{hom}}$, using $1/T_1 = \gamma_e \Delta H_{\text{hom}}$ at high fields, $H > H_{c2}$ that suppresses superconductivity. The maximum upper critical field is $H_{c2,\text{max}} \sim 16$ T for particles with field in the $(a,b)$ crystallographic plane in the polycrystalline sample at zero temperature [19]. We did not observe any effects of superconductivity on the CESR, at 15 T it is suppressed in the full sample above a temperature of a few K. Fig. 1 shows that the temperature dependence of the CESR line-width at 15 T is small below 40 K.

The CESR line-width is magnetic field dependent as shown in Fig. 2 at 40 K: it is linear as function of magnetic field with $\Delta H = \Delta H_0 + b \cdot H$, where $\Delta H_0 = 0.90(1)$ mT is the residual line-width and $b = 0.057(1)$ mT/T. The residual homogeneous line-width corresponds to $T_1 = 6.3$ ns at 40 K. The linear relation can be used to correct the 15 T CESR line-width data to obtain the homogeneous contribution, $\Delta H_{\text{hom}}(T) = \Delta H(15 \text{ T}, T) - 15 \text{T} \cdot b$ as the magnetic field dependence is expected to be temperature independent. We show the homogeneous line-width in Fig. 1. We find that it is
FIG. 3: Inhomogeneous CESR line broadening in MgB$_2$ below $T_c$ at 0.32 T. Full and open symbols show the CESR line-width for up and down magnetic field sweeps, respectively. Inset shows the data near $T_c$. Note the line narrowing between $T_c$ and $T_{irr}$ and the field sweep direction dependent line-widths below the irreversibility temperature.

temperature independent within experimental precision between 3 and 50 K. This means that the spin-lattice relaxation time flattens to a residual value that is given by non-magnetic impurities.

Relaxation in the superconducting state

In type II superconductors, CESR arises from thermal excitations and from normal state vortex cores. The inhomogeneity of the magnetic field in the vortex lattice or glass states does not broaden the CESR line. The local magnetic field inhomogeneity is averaged since within the spin life-time itinerant electrons travel long distances compared to the inter-vortex distance [4]. This is in contrast to the NMR case where the line-shape is affected: the nuclei are fixed to the crystal and nuclei inside and outside the vortex cores experience different local fields [20]. In other words, a superconducting single crystal sample would display a narrow conduction electron ESR line if there were no irreversible effects. However, the CESR line is inhomogeneously broadened below the irreversibility line for a superconducting powder sample: the vortex distribution depends on a number of factors such as the thermal and magnetic field history, grain size and, for an anisotropic superconductor such as MgB$_2$, on the crystal orientation with respect to the magnetic field also. The resulting inhomogeneous broadening of the CESR line gives $1/\gamma \Delta H_{\text{inhom}} = T_2^* \ll T_1$, and $T_1$ cannot be measured from the line-width. In Fig. 3 we show this effect: above $T_c$, MgB$_2$ has a relaxationally broadened line-width of $\Delta H = 0.9$ mT. Between $T_c$ and the irreversibility temperature at the given field, $T_{irr}$, the CESR remains homogeneous and narrows with the lengthening of $T_1$. However, below $T_{irr}$ it broadens abruptly and the line-width depends on the direction of the magnetic field sweep: for up sweep it is broader than for down sweeps due to the irreversibility of vortex insertion and removal.

To enable a direct measurement of the $T_1$ spin lattice relaxation time, one has to resort to time resolved experiments. Conventional spin-echo ESR methods are limited to $T_1$'s larger than a few 100 ns. To measure $T_1$'s of a few nanoseconds, the so-called longitudinally detected ESR was invented in the 1960's by Hervé and Pescia [21] and improved by several groups [22, 23]. The method is based on the deep amplitude modulation of the microwave excitation with an angular frequency, $\Omega \sim 1/T_1$. When the sample is irradiated with the amplitude modulated microwaves at ESR resonance, the component of the magnetization along the static magnetic field, $M_z$, decays from the equilibrium value, $M_0$, with a time constant $T_1$. $M_z$ relaxes back to $M_0$ with a $T_2$ relaxation time when the microwaves are turned off. The oscillating $M_z$ is detected using a coil which is part of a resonant rf circuit. The phase sensitive detection of the oscillating voltage using lock-in detection allows the measurement of $T_1$ using $\Omega T_1 = v/u$ [3, 21], where $u$ and $v$ are the amplitudes of the in- and out-of-phase components of the oscillating magnetization after corrections for instrument related phase shifts. The principal limitation of the LOD-ESR technique is its 3-4 orders of magnitude lower sensitivity compared to conventional cw-ESR. The LOD-ESR method and the experimental apparatus are detailed in Refs. [3, 6].

To prove that the LOD-ESR signal of the itinerant electrons is detected in the superconducting phase, we compare in Fig. 3 the LOD-ESR signal with that measured with conventional continuous-wave CESR (referred to as CESR in the following) of MgB$_2$ in the normal and superconducting states. The CESR signal is the derivative of the absorption due to magnetic field modulation used for lock-in detection. This signal was previously identified as the ESR of conduction electrons in MgB$_2$ in the superconducting and normal states [12, 17, 24] and its characteristics have been discussed in detail [13, 17]. Above $T_c$ at 40 K, the CESR line is relaxationally broadened. Below $T_c$, it is inhomogeneously broadened and is diamagnetically shifted, i.e. to higher resonance fields. The irreversible effects also contribute to a non-linear baseline known as the non-resonant microwave absorption [25]. The intensity of the CESR signal decreases below $T_c$ as we discussed previously [17], due to the vanishing of normal state electrons.

The LOD-ESR signal shows the same characteristics as the CESR below $T_c$: it is broadened, shifted to higher fields and its intensity decreases. The values for the temperature dependent diamagnetic shifts and broadening and the relative intensity change agree for the two kinds of measurements within experimental precision (not shown). This unambiguously proves that the LOD-
that of weak-coupled BCS theory by Yafet [3]. He concluded that a type I superconductor was calculated in the framework of superconductor in finite fields with state excitations. However, no theory exists for a type II.

On the other hand, the field independence between 0.32 and 1.27 T of $T_c$ below $T_c$ below $T_c$ is surprising. The lengthening of $T_1$ below $T_c$ in zero magnetic field for an isotropic, type I superconductor was calculated in the framework of weak-coupled BCS theory by Yafet [3]. He concluded that $T_1$ lengthens as a result of the freezing of normal state excitations. However, no theory exists for a type II superconductor in finite fields with $H_{c2}$ anisotropy such as MgB$_2$, thus here the $T_1$ data are analyzed phenomenologically in the framework of the two-band/gap model of MgB$_2$.

In the following, we deduce the residual (low temperature), impurity related spin scattering contributions of the $\sigma$ and $\pi$ Fermi surface sheets. The DOS is distributed almost equally on the FS sheets of MgB$_2$: $N_\pi/(N_\pi + N_\sigma) = 0.56$ [13], where $N_\pi$ and $N_\sigma$ are the DOS of the two types of FS sheets. A magnetic field of $\sim 0.3 - 0.4$ T closes the gap on the $\pi$ FS sheets but leaves the gap on the $\sigma$ sheet almost intact [10, 17]. This suggests that well below $T_c$, our experiment at 0.32 T measures exclusively the relaxation of electrons on the fully closed $\pi$ FS sheets. Since $T_1$ at 0.32 T increases slowly with temperature between 10 and 20 K, we extrapolate $T_{1\pi} \approx T_1(10$ K, 0.32 T) = 20(2) ns for the $\pi$ FS.

In order to separate the contribution of the $\sigma$ FS to the relaxation rate in the normal state, $1/T_{1\sigma}$, we assume that inter-band relaxation is negligible and $1/T_{1n}$ is equal to the average of the spin-lattice relaxation rates on the two FS’s weighted by the corresponding DOS:

$$\frac{1}{T_{1n}} = \frac{N_\pi/T_{1\pi} + N_\sigma/T_{1\sigma}}{N_\pi + N_\sigma}$$

(1)

Here $T_{1\sigma}$ is the spin-lattice relaxation time on the $\sigma$ FS. The 15 T measurement shows that $1/T_{1n}$ changes little with temperature between 3 K and 40 K. Thus we find $T_{1\sigma} = 3.4(5)$ ns for the contribution of the $\sigma$ FS sheets using $T_{1\pi} = T_1(T_c) = 6.3$ ns, $T_1(10$ K, 0.32 T) = 20(2) ns and Eq. (1).

For normal metals with a simple Fermi surface, the so-called Elliott relation [2, 3, 26, 27] holds, which states:

![FIG. 4: ESR (a-b) and LOD-ESR (c-d) spectra of MgB$_2$ at 9.1 GHz (0.32 T). a) and c) at 40 K in the normal state, and b) and d) in the superconducting state at 15 K. Solid and dashed curves are the in- and out-of-phase LOD signals, respectively and are offset for clarity. Vertical solid lines indicate the resonance field above $T_c$. Note the diamagnetic shift and broadening for both kinds of spectra below $T_c$. Also note the rotated phase of the in-phase and out-of phase channels upon cooling.](image)

![FIG. 5: Spin-lattice relaxation time as a function of the reduced temperature in MgB$_2$ at 0.32 (■) and 1.27 T (○) magnetic fields. Representative error bars are shown for some of the data. Dashed curve shows $T_1$ corresponding to $\Delta H_{1\text{hom}}$ in the 15 T measurement such as in Fig. 4 with the reduced temperature normalized to 39 K.](image)
that for a given type of disorder (e.g. phonons or dislocations) $T_1$ is proportional to the momentum relaxation time, $\tau$. The proportionality constant depends on the spin orbit splitting of the conduction electron bands and has been estimated in a number of metals from the shift of the CESR from the free electron value. Metals with complicated Fermi surfaces i.e. with great variations of the electron-phonon coupling on the different FS parts are known to deviate from the Elliott relation [28] and calculation of $T_1$ requires to take into account the details of the band-structure [29, 30, 31]. Examples include polycrystalline elemental metals such as Mg or Al. Clearly, a calculation of $T_1$ is required for MgB$_2$, which takes into account its band structure peculiarities. Comparison of spin scattering and momentum scattering times of the two types of Fermi surfaces is instructive. The relative values of $\tau$ for the two FS parts, $\tau_\pi$ and $\tau_\sigma$, and for interband scattering were estimated by Mazin et al. [32]. A very small impurity interband scattering and $\tau_\pi < \tau_\sigma$, i.e. a larger $\pi$ intraband scattering relative to $\sigma$ intraband scattering was required to explain the rather small depression of $T_c$ in materials with widely different conductivities. De Haas-van Alphen [33] and magnetoresistance [34] measurements of high purity samples yield $\tau_\pi < \tau_\sigma$ also. Such a behaviour could rise from Mg vacancies, which perturb more electrons of the $\pi$ band relative to the $\sigma$ band. However, our spin scattering data do not support this. In contrast to momentum scattering, spin scattering is stronger on the $\sigma$ FS: $T_{1\pi}/T_{1\sigma} = 6 : 1$ in high purity samples and low temperatures. The relative values of $T_1$ and $\tau$ for the two FS do not necessarily follow the same trend, spin relaxation times at low temperatures depend on spin orbit relaxation on impurities while momentum relaxation is due to potential scattering. However, a defect center such as a Mg vacancy with a strong modification of the electron-phonon coupling and an atomic number strongly differing from that of the regular atoms constitutes the compound would greatly affect $T_1$ compared to $\tau$. In the two gap model Mg defects are expected to shorten $T_{1\pi}$ more than $T_{1\sigma}$ and thus are unlikely to be the dominant scatterers.

A final note concerns the validity of the above analysis of $T_1$’s in the framework of the two-band/gap model. The field independence of the lowest temperature $T_1$ for 0.32 and 1.27 T is unexpected within this model. The spin susceptibility increases strongly between these fields and more normal states are restored at 1.27 T than expected from the closing of the gap on the $\pi$ FS sheets alone. Based on this, one would expect to observe additional spin scattering from the restored $\sigma$ FS parts, which is clearly not the case. This also indicates that a theoretical study, which takes into account the peculiarities of MgB$_2$ is required to explain the anomalous spin-lattice relaxation times.

In conclusion, we presented the measurement of the spin-lattice relaxation time, $T_1$, of conduction electrons as a function of temperature and magnetic field in the MgB$_2$ superconductor. We use a novel method based on the detection of the $z$ component of the conduction electron magnetization during electron spin resonance conditions with amplitude modulated microwave excitation. Lengthening of $T_1$ below $T_c$ is observed irrespective of the significant CESR line broadening due to irreversible diamagnetism in the polycrystalline sample. The field independence of $T_1$ for 0.32 T and 1.27 T allows to measure the separate contributions to $T_1$ from the two distinct types of the Fermi surface.

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