Nuclear Spin-Lattice Relaxation Induced by Thermally Fluctuating Flux Lines in Type-II Superconductors

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Thermal motion of the flux lines (FL) gives rise to fluctuating magnetic fields. These dynamic fields couple to the nuclei in the sample and relax the nuclear spins. Based on a model of harmonic fluctuations, we provide a theoretical description of the nuclear spin-lattice relaxation (NSLR) process due to the fluctuating FLs in clean type-II superconductors. At low fields, the calculated longitudinal relaxation rate $T_1^{-1}$ is enormously enhanced at temperatures just below $T_c$. At intermediate fields, the resulting $T_1^{-1}$ exhibits a peak structure as a function of temperature, which is eventually suppressed as the field is increased. The vibrational modes which have components propagating along the FLs play an essential role in the $T_1^{-1}$ process.

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Several recent experiments and theories have pointed to the importance of thermal fluctuations in the thermodynamics of type-II superconductors.\textsuperscript{[1, 2, 3]} These fluctuations also have pronounced effects on the dynamic properties of the system. Recent measurements of the NSLR rate $T_1^{-1}$ in the organic superconductor $\kappa$-(ET)$_2$Cu[N(CN)$_2$]Br (ET) by De Soto and Slichter et al. provide an excellent example of this.\textsuperscript{[4]} It was shown that there is an enormous enhancement of the relaxation rate of $^1$H nuclei in the superconducting state relative to the normal state Korringa value. It is believed that the experimentally observed peak structure does not belong to the category of the Hebel-Slichter coherence peak, rather it is caused by the thermal motion of the fluxoid system.\textsuperscript{[4]}

In this letter, we attempt to provide a theoretical description of NSLR in clean type-II superconductors induced by the fluctuating magnetic fields of the flux line lattice (FL). In the presence of fluctuating FLs, the local magnetic field at nuclear sites varies in time, which may contribute to the NSLR. This mechanism becomes dominant in a system such as ET when relaxation through other channels (e.g. through the interaction with conduction electrons) is suppressed.\textsuperscript{[4]} We will confine our study to the region $H \ll H_{c2}$, where the mean distance $a_0$ between FLs is much larger than the radius of the vortex core $\xi$ (coherence length). Since we aim to give a general treatment of the phenomenon, we will, for simplicity, consider a model of continuous FLs based on the 3D London description. The case of layered superconductors with the external magnetic field perpendicular to the layers (chosen along the z-direction) could be handled similarly.

We specify the axis of the $j$-th FL at time $\tau$ by a 3D vector $\vec{r}_j(\tau)$. The local field $\vec{B}(\vec{r}, \tau)$ varies with time $\tau$ only through the motion of the FL and is given by\textsuperscript{[3]}

$$
\vec{B}(\vec{r}, \tau) = \frac{\phi_0}{4\pi \lambda^2} \sum_j \int d\vec{s}_j \frac{\exp(-|\vec{r} - \vec{r}_j(\tau)|/\lambda)}{|\vec{r} - \vec{r}_j(\tau)|} = \frac{V}{(2\pi)^3} \int d^3k \vec{b}(k) \frac{1}{NL} \sum_j \int d\vec{z}_j e^{i\vec{k} \cdot (\vec{r} - \vec{r}_j(\tau))}, \quad (1)
$$

where $\vec{b}(k) = B(\hat{\vec{k}}k_\perp^2 - k_\perp k_z)/k_\perp^2(1 + k^2 \lambda^2)$, $\vec{k} = (k_\perp, k_z)$ and $k_\perp = (k_x, k_y)$, $\lambda$ is the magnetic penetration depth, the line integral $\int d\vec{s}_j$ is along the $j$-th FL, $V$ is the volume of the sample, and $N$ and $L$ are the number and the length of FL respectively. To obtain the Fourier transform of $\vec{B}(\vec{r}, \tau)$ we used $\nabla \cdot \vec{B} = 0$. A similar expression for the layered system can be found in Refs.\textsuperscript{3, 4, 5}.

It is seen clearly from Eq. (1) that the thermal motions of FLs lead to fluctuating magnetic fields. These fields are coupled to the nuclear moments in the sample. This coupling is described by the interaction $\mathcal{H}(\tau) = -\gamma_n \hbar \sum_n \vec{I} \cdot \vec{B}(\vec{r}_n, \tau)$, where $\gamma_n$ is the nuclear gyromagnetic ratio, $\vec{I}$ is nuclear spin operator, and $\vec{r}_n$ is the position of the $n$-th nucleus. We then have\textsuperscript{[4]}

$$
\frac{1}{T_1} = 2\gamma_n^2 \sum_{\alpha=x,y} |\langle \uparrow | I_\alpha | \downarrow \rangle|^2 \int \frac{d\tau}{2\pi} \cos \omega_0 \tau (B_\alpha(\vec{r}, \tau)B_\alpha(\vec{r}, \tau)) , \quad (2)
$$

where $\omega_0 = \gamma_n B$, $\langle \uparrow | I_\alpha | \downarrow \rangle$ represents the thermal average of the FL configurations. Here we have taken an average over the relaxation rates of all the nuclei (denoted by the horizontal bar in Eq. (2)), which will be evaluated by spatial integration. Using Eq. (1) we recast Eq. (2) into

$$
\frac{1}{T_1} = \gamma_n^2 K(\omega_0, B) = \gamma_n^2 \frac{V}{(2\pi)^3} \int d^3k |\vec{b}_\perp(k)|^2 \frac{1}{NL} \tilde{S}(\vec{k}, \omega_0), \quad (3)
$$

where $K(\omega_0, B)$ is the fluctuation spectral density function, $\tilde{S}(\vec{k}, \omega_0) = [S(\vec{k}, \omega_0) + S(\vec{k}, -\omega_0)]$, and

$$
S(\vec{k}, \pm \omega_0) = \int_{-\infty}^{+\infty} \frac{d\tau}{2\pi} e^{\pm i\omega_0 \tau} \frac{1}{NL} \sum_{jj'} \int d\vec{z}_j \int d\vec{z}_{j'} \langle e^{i\vec{k} \cdot \vec{r}_j(\tau)} e^{-i\vec{k} \cdot \vec{r}_{j'}(\tau)} \rangle. \quad (4)
$$
is the dynamic structure factor of the FLs, which is entirely determined by the FL structure and corresponding time evolution spectrum, without reference to any properties of the nuclei. We emphasize that our result (3) is quite general, applying even to the flux liquid and glass states. In the following we will focus our study on the FLL state.

The problem of finding $T^{-1}_1$ is now reduced to obtaining the dynamic structure factor $S(k,\omega_0)$ of the FLs, which is a fundamental dynamic quantity and of importance in many other applications (e.g., the inelastic scattering). Evaluation of $S(k,\omega_0)$ requires information about the dynamics of the FL fluctuations. Bulaevskii et al. [8] have used the Langevin equations for the overdamped motion of pancakes in interpreting their $T^{-1}_1$ data in $Tl_2Ba_2CuO_6$, and found a monotonic decrease of $T^{-1}_1$ with temperature. At this time the dynamics of the FLs are much less well understood than their static properties. A clean translational invariant superconductor is thus a valuable model system which can provide significant insight into the physics involved. In this case it is clear that the vortex motion is reversible. Once this fundamental case is understood, various complications (e.g., including damping of the vortex motion) can be introduced.

For a clean superconductor, Fetter has derived a set of dynamic equations for the FLs by using the London model, which provides a basis for our study.[8] We assume that the deformation of a rectilinear FL is small so that the vortex axis can be specified by $\vec{r}_i(\tau) = (\overline{R}_i^{(e)} + \vec{u}_i(z,\tau), z)$, where $\vec{u}_i(z,\tau)$ is the displacement of the $i$-th FL from its equilibrium position $(\overline{R}_i^{(e)} , z)$. In the harmonic approximation to the total energy of the FLL, the motion of $\vec{u}_i(z,\tau)$ can be quantized by interpreting $u_{xi}(z)$ and $u_{yi}(z)$ as quantum mechanical conjugate variables.[8] This leads to

$$u_{xi}(z,\tau) = \frac{1}{\sqrt{\rho \overline{\Omega} LN}} \sum_{\vec{q}_\perp} \frac{\hbar \Omega}{2\omega} (\alpha_{-\vec{q}_\perp} - q_z e^{-i\omega\tau} + a_{\vec{q}_\perp}^\dagger e^{i\omega\tau}) e^{i\vec{q}_\perp \cdot \overline{R}_i^{(e)} + iq_z z},$$

$$u_{yi}(z,\tau) = -\frac{1}{\sqrt{\rho \overline{\Omega} LN}} \sum_{\vec{q}_\perp} \frac{\hbar \Omega}{2\omega} [(\alpha+i\omega)\alpha_{-\vec{q}_\perp} - q_z e^{-i\omega\tau} + (\alpha - i\omega)a_{\vec{q}_\perp}^\dagger e^{i\omega\tau}] e^{i\vec{q}_\perp \cdot \overline{R}_i^{(e)} + iq_z z},$$

where $\overline{\Omega} = h/2m$, $\overline{\Omega} = \overline{\Omega} + \frac{1}{1+q^2_\perp \eta} + \xi$, $\rho$ is the mass density of the superelectrons, and the summation over $\vec{q}_\perp$ is restricted to the first Brillouin zone. In the above equations, $\overline{\Omega}$, $\alpha$, $\eta$, and $\xi$ are functions of $(\vec{q}_\perp, q_z)$ and are defined in Eqs. (43) and (44) of Ref. [8]. The corresponding Hamiltonian of the FLL reads $\mathcal{H}_{FLL} = \frac{1}{2} \Sigma_{\vec{q}_\perp, q_z} \hbar \omega (a_{\vec{q}_\perp q_z}^\dagger a_{\vec{q}_\perp q_z} + a_{\vec{q}_\perp q_z}^\dagger a_{\vec{q}_\perp q_z})$, with the dispersion relation given by

$$\omega^2(\vec{q}_\perp, q_z) = (\overline{\Omega} + \frac{1}{1+q^2_\perp \eta})^2 - (\overline{\alpha}^2 + \overline{\xi}^2).$$

Here $a_{\vec{q}_\perp q_z}^\dagger$ and $a_{\vec{q}_\perp q_z}$ obey boson commutation relations, and represent the creation and annihilation operators for a single vibrational quantum (which will be referred to as a “fluxon”) in the normal mode with wave vector $(\vec{q}_\perp, q_z)$ and frequency $\omega(\vec{q}_\perp, q_z)$.

We will now derive $S(k,\omega_0)$ resulting from Fetter’s dynamics. [8] As usual, we use the Bloch identity [9] to write $\langle e^{i\vec{k}_\perp \cdot \vec{u}_j(z,0)} e^{-i\vec{k}_\perp \cdot \vec{u}_j(z',\tau)} \rangle = e^{-2\overline{\Omega} W(\vec{k}_\perp) e^{i\vec{k}_\perp \cdot \vec{u}_j(z,0) \vec{k}_\perp \cdot \vec{u}_j(z',\tau)}}$, where $W(\vec{k}_\perp) = \frac{1}{2} \langle [\vec{k}_\perp, \vec{u}_j(z,0)]^2 \rangle$ is the Debye-Waller factor. The lowest order contribution arises from the one-fluxon process, in which a nucleus flips its spin and the FLL creates or annihilates a fluxon to conserve the total energy. Combining the Bloch identity and Eqs. (4)-(6), and keeping only the one-fluxon
transverse field is required for longitudinal relaxation). This indicates the essential role played by these modes. First, we notice that the modes propagating perpendicular to the FL direction (⊥) contribute to those of a single FL. At high fields ($T_1^{-1} \propto h/\rho c_{\lambda e}$, and $T_1^{-1}$ is no transverse field associated with these modes (as can be seen from Eq. (2), a fluctuating $\lambda/\xi$ term, we obtain

$$S(\vec{k}, \omega_0) = \frac{h}{2\rho c_{\lambda e}} e^{-2W(\vec{k}_{||})} \sum_{\vec{q}_{\bot}, \vec{a}_{\bot}} \sum_{\vec{r}} \frac{1}{\omega} \Omega_{\vec{q}_{\bot}}(\omega + \omega_0) \delta_{\vec{K}_{\bot} \vec{q}_{\bot}} \delta_{\vec{K}_{\bot} + \vec{R}} \delta(\omega - \omega_0) \delta_{\vec{K}_{\bot} \vec{q}_{\bot} + \vec{R}}$$

where $\vec{R}$ is the reciprocal lattice vector, $\Omega' = \Omega + \frac{1}{1 + q_\perp^2 \lambda^2} \bar{\Omega} - \bar{\xi}$, and $n = \langle a_{\vec{q}_{\bot} \vec{a}_{\bot}}^\dagger a_{\vec{q}_{\bot} \vec{a}_{\bot}} \rangle$ is the Bose-Einstein occupation factor of fluxons.

Upon substituting Eq. (8) into Eq. (3), we obtain

$$\frac{1}{T_1} = \frac{\gamma_n^2 \hbar B^2}{2\rho c_{\lambda e} L} \sum_{\vec{q}_{\bot}, \vec{a}_{\bot}} \sum_{\vec{r}} \frac{q_\perp^2 \lambda^2}{(q_\perp + R)^2} \frac{e^{-2W(q_\perp + R)}}{\omega} \coth \left( \frac{h \omega}{2k_B T} \right) \times \left[ \Omega(q_x + K_x)^2 + \Omega'(q_y + K_y)^2 - 2\pi^2(q_x + K_x)(q_y + K_y) \right] \delta(\omega - \omega_0)$$

For arbitrary fields and temperatures the quantity in the summation is a complicated function of $\vec{R}$ and $(q_\perp, q_\parallel)$, and the summation can only be evaluated numerically; however, in some special cases and with some restrictions, analytical results can be found. The simplest case is a single FL in a bulk sample (this also includes the case of well-separated FLs, i.e. $a_0 \gg \lambda$). In this case the motion is entirely self-induced and only waves propagating along the FL ($q_\parallel = 0$) exist. Thus we have $\tau = \eta = \xi = 0$. In the long wave length limit the dispersion relation can be written as

$$\bar{\Omega}(q_x) \approx \frac{\gamma}{4\pi} \left[ \ln \frac{2\lambda}{\xi} - \gamma + \frac{1}{4} q_\perp^2 \lambda \right]$$

where $\gamma = 0.5772$ is Euler’s constant. We then obtain

$$\frac{1}{T_1} = \frac{\hbar \phi_0 c_{\lambda e} q_{\perp_0}^2}{16\pi^2 \rho c_{\lambda e}^2} \frac{\hbar \omega_0}{2k_B T} \left[ \frac{1}{1 + q_{\perp_0}^2 \lambda} e^{-\frac{\omega_0}{h^2} \frac{q_{\perp_0}^2 \lambda}{\omega^2}} - \frac{\langle u^2 \rangle}{\lambda^2} E(1 + q_{\perp_0}^2 \lambda^2) \right]$$

where $E(x)$ is the exponential integral. $q_{\perp_0}$ is determined by $\omega(q_{\perp_0}) = \omega_0$. The dashed curve in Fig. 1 shows $T_1^{-1}$ as a function of $t$, where $\lambda = \lambda_0/\sqrt{T - T_c}$, $\langle u^2 \rangle / \lambda^2 \approx \beta t$,\footnote{\[13\]} $\beta = \frac{4\pi k_B T}{\pi \rho c_{\lambda e} \lambda^2 \ln(\lambda/\xi)}$ and $t = T/T_c$. Here we have chosen $\omega_0 = 5$ MHz, $\gamma_n = 2.675 \times 10^4 s^{-1} Gauss^{-1}$, $\lambda_0 = 2000 \AA$, and $\beta = 0.1$.

As can be seen from Eq. (9), for arbitrary vortex density, $T_1^{-1}$ is a sum of contributions from all normal modes. It is instructive to analyze the contributions arising from two special types of modes. First, we notice that the modes propagating perpendicular to the FL direction ($q_\parallel = 0$), which corresponds to the vibrations of the rigid FLs, do not contribute to $T_1^{-1}$, because there is no transverse field associated with these modes (as can be seen from Eq. (2), a fluctuating transverse field is required for longitudinal relaxation). This indicates the essential role played by the modes with $q_\parallel \neq 0$ in the $T_1^{-1}$ process. Second, consider the vibrations propagating along the FLs (also known as the “helicon modes”\footnote{\[12\]}). In the dilute limit these modes become identical to those of a single FL. At high fields ($a_0 \ll \lambda$), the dispersion relation of such modes reads

$$\bar{\omega}(q_\parallel) \approx \frac{\gamma}{4\pi} \left[ \ln \frac{2\lambda}{\xi} - \gamma + \frac{1}{4} q_\perp^2 \lambda \right]$$

in the long wave-length limit.\footnote{\[14\]} In this case we also have $K^2 \lambda^2 \gg 1$ for $\vec{K} \neq 0$. This allows us to keep the $\vec{K} = 0$ term only in Eq. (9) and to obtain the contribution $\frac{dT_1^{-1}}{dq_\perp |_{q_\parallel=0}}$ from these so-called “helicon” modes:\footnote{\[12\]}:

$$\frac{dT_1^{-1}}{dq_\perp |_{q_\parallel=0}} = \frac{\hbar \phi_0 c_{\lambda e} q_{\perp_0}}{16\pi^2 \rho c_{\lambda e}^2} \frac{\hbar \omega_0}{2k_B T} \frac{q_{\perp_0}^2 \lambda}{(1 + q_{\perp_0}^2 \lambda^2)^2}$$

(11)
The NSLR rate $T_1^{-1}$ arises, in general, from the contributions of both the “helicon” modes and those branches with $\vec{q}_\perp \neq 0$. But one should note that not all $\vec{q}_\perp \neq 0$ branches contribute to the $T_1^{-1}$ process. This is seen clearly from Fig. 2, in which we have plotted $\omega(q_\perp, q_z)/\omega_0$ as a function of $q_z$ for a few values of $q_\perp$. The branches with $\omega(q_\perp, q_z = 0)/\omega_0 > 1$ do not contribute to $T_1^{-1}$ due to the $\delta$-function in Eq. (9). For $^1\text{H}$ we have numerically evaluated Eq. (9) for $\omega_0 = 70$ and 1000 MHz (corresponding to $B = 0.37$ and 3.74 Tesla, respectively). The resulting $T_1^{-1}$'s are plotted as a function of temperature in Fig. 1 as the solid and dotted curves. Notably, a relaxation peak is found below $T_c$. Note that the results shown in Fig. 1 only contain the contributions from the fluctuating FLL only. At very high fields, the relaxation peak could be absent if $T_c(B)$ becomes less than the temperature at the peak, or if the relaxation due to the FLL is so greatly suppressed that it is slower than that due to other sources, e.g. the interaction with the current carriers.

We now address the behavior of $T_1^{-1}$ at different fields in more detail. First of all, we mention that at typical NMR frequencies (from a few to several hundred MHz) the condition $\frac{h\omega}{k_BT_c} \ll 1$ is satisfied for superconductors with $T_c \sim 10K$. The low frequency region corresponds to fields of a few hundred Gauss. The FLs are well-separated, and are very soft in this case. $T_1^{-1}$ can be described by Eq. (10) and from Fig. 1 we see that $T_1^{-1}$ is enormously enhanced right below $T_c(B)$. Note that so far there seems to be no experimental data published in this regime. It would be interesting to carry out such measurements. When the field increases to $a_0 \sim \lambda$, the interaction between the FLs begins to play a role. This interaction promotes the modes with $\vec{q}_\perp \neq 0$ and stiffens the FLs. In the high frequency range $(a_0 \ll \lambda)$, only the $\tilde{K} = 0$ term gives important contributions to $T_1^{-1}$. The magnitude of $T_1^{-1}$ is greatly suppressed in comparison with the single FL case, as can be seen in Fig. 1. A close examination of Eq. (9) suggests that this suppression comes from the decrease of the thermal factor coth $\frac{h\omega}{2k_BT_c}$, the density of states of fluxons at $\omega_0$, and the resonant wave number $q_\omega$. These effects overcome the increase brought on by the prefactor $\omega_0^2$ in Eq. (9). One should note that the Fetter’s vortex dynamics breaks down if the field is close to $H_{c2}$ so that vortex fields overlap strongly. In this case the nonlocal effects (wave vector dependence of the elastic moduli) act to enhance the fluctuations, and a Ginzburg-Landau type of approach becomes relevant.

The fluctuation spectral density function $K(\omega, B)$ contains detailed dynamic information of the system. It depends on both frequency $\omega$ and magnetic field $B$. In Fig. 3 we plot $K(\omega, B)$ vs $\omega$ for a few $B$'s of interest. At a fixed $B$, the function has a maximum at a frequency $\omega_m$ which shifts upward as the field increases. The field dependence of $K(\omega, B)$ at a given frequency $\omega$ can also be seen from Fig. 3. At low frequencies, $K(\omega, B)$ decreases with the field, which has been observed experimentally. At high frequencies it increases with $B$, and at intermediate frequencies it has a peak structure as a function of $B$. These are very interesting predictions which can be tested experimentally by measuring the NSLR rates of different type of nuclei, or different transitions of a nucleus with $I > 1/2$. The measurement of $K(\omega, B)$ provides a crucial test for a vortex dynamics.

In summary, we have studied the NSLR caused by thermally fluctuating FLL in a clean type-II superconductor. Its applications for the vortex dynamics go beyond the NMR. The NMR approach to the FLL state is quite diagnostic, and provides a sensitive probe to the dynamic fluctuations of the vortices. The simple harmonic fluctuations of the FLL seems to render a good description of the overall features of $T_1^{-1}$ obtained in ET. We found that the fluctuation effects on the $T_1^{-1}$ process is most pronounced at low fields, where the FL is easy to “bend” and the thermal excitations of the fluxons with the frequency $\omega_0$ is effective. In the above discussions, we have
ignored the effects of damping of the vortex motion. We expect that inclusion of weak damping would not change our results significantly, and that the theory should be directly applicable to systems with sufficiently large Hall angle\[16\]. The harmonic and the overdamped vortex motions (this is believed to be the case in YBCO) represent two different important regimes. The latter also deserves further study. While at present there is still no well-accepted dynamic theory of the vortices, we believe that our calculations shed important light on this subject. Finally, we mention that the effects discussed in this paper should be more pronounced in layered superconductors since in that case the FLs are more flexible.\[2\]

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FIGURE CAPTIONS

FIGURE 1. $T_1^{-1}$ as a function of $T/T_c$ at $\omega_0 = 5$ MHz (dashed curve, represents the case of a single FL), and at $\omega_0 = 70$ (solid curve) and 1000 MHz (dotted curve). The unit of $T_1^{-1}$ is chosen to be $\frac{\hbar \gamma_n \phi_0}{8\pi^2 \rho \lambda^2 \kappa \lambda_0^3}$, which is about $10^{-3}$ to 100 Hz for typical type-II superconductors. Note that we have plotted $T_1^{-1}$ all the way up to $T/T_c = 1$. In reality, the $T_1^{-1}$ is only valid up to $T_c(B)$. Above $T_c(B)$, the system goes to the normal state.

FIGURE 2. $\omega(q_z, q_\perp)/\omega_0$ at $\omega_0 = 1000$ MHz and $T/T_c = 0.4$ for $q_\perp \lambda = 0, 0.15, 0.31, 0.46, 0.61, 0.76$ and 0.92 (from bottom to top).

FIGURE 3. Fluctuation spectral density functions at $B = 1000$ (solid curve), 3000 (dotted curve), and 6000 Gauss (dashed curve).