Relation between the dielectric function and nuclear spin-lattice relaxation by thermal phase fluctuations of a pinned spin-density wave

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We present a model for the nuclear spin-lattice relaxation rate (1/T1) by quasi-one-dimensional phase fluctuations of a pinned, incommensurate spin-density wave (SDW) phase of (TMTSF)2PF6 and similar materials is thermal fluctuations of the SDW phase. In this paper we point out the relation of this relaxation mechanism to the frequency (ω) and wave vector (k) dependence of the imaginary part of the dielectric function [εi(ω)] of the SDW condensate. Our goal is to provide a framework for the interpretation of nuclear spin-lattice relaxation measurements caused by SDW phase fluctuations for use as a probe of SDW dynamics.

The relaxation process described here is based upon the following physical picture. Thermal phase motion of the SDW generates the fluctuating magnetic field responsible for nuclear spin-lattice relaxation. Such fluctuations also correspond to local polarization charge transport of the SDW condensate and are, therefore, related to εi(ω) through a Kubo relation or, equivalently, the fluctuation-dissipation theorem.

One of the important aspects of this spin-lattice relaxation by phase fluctuations in Bechgaard salts is that, in contrast to conventional electrical transport measurements, the NMR process responds mainly to the motion of the condensed phase (the SDW) and not to transport by thermally excited normal carriers. Thus, it complements electrical transport measurements, which include both contributions to the current.

There are several problems that have elements in common with the present work. One example is the description by Blicke of spin-lattice relaxation by phase fluctuations associated with ferroelectric displacement transitions. In that case, the coupling to the nuclei is quadrupolar and the relation to the εi(ω) is not spelled out. Later, this approach was applied to spin-lattice relaxation in charge-density-wave systems, where the coupling is also to the quadrupole moment of the nucleus. Another important precedent is the description of magnetic spin-lattice relaxation pioneered by Moriya, which has several features that are closely analogous to the present paper, such as use of the fluctuation-dissipation theorem and a result for 1/T1 in terms of the frequency- and wave-vector dependence of the electron-spin susceptibility.

In this paper we use SI units for electrical and magnetic quantities. The next section presents the model for the spin density and applies it to obtain the corresponding magnetic field of the SDW. Its thermal fluctuations are used to obtain the formula for 1/T1. We then conclude with a brief discussion of the results.

II. MODEL

A. Description of the spin-density wave

One of the important considerations is that the magnetic field associated with the SDW must be specified at the spatial points where the nuclei are located. The intended application is for molecular conductors, where the spin density is associated with the electronic states of a single molecular entity, such as one TMTSF molecule in the Bechgaard salts. In this case, one views the spin density as being rigid within a given molecule but with a phase that varies from one molecule to the next. This situation is taken into account by modeling the spin density ρ with

\[ \rho(w_j + r_j, t) = f_j \hat{\sigma} \rho_5(r_j, t), \] (1)

where \( w_j \) is the coordinate of the \( j \)th site is expressed in molecular coordinates, \( t \) is time, \( r_j \) is the position of the \( j \)th molecule, \( f_j \) is the fraction of the total spin density of the molecule at the \( j \)th site, and \( \hat{\sigma} \) is a unit vector that designates the polarization of the SDW. The function \( \rho_5 \) is written as

\[ \rho_5 = \rho_{05} \Re \exp[i(\mathbf{Q} \cdot \mathbf{r}_j + \phi(r_j, t))], \] (2)

where \( \rho_{05} \) is the SDW amplitude, \( \Re \) denotes the real part, and \( Q \) is the SDW wave vector. In the following we will not write \( \Re \) except where it is needed explicitly. The amplitude \( \rho_{05} \) can alternatively be expressed as \( \rho_{05} = \eta \mu_B \), where \( \eta \) is the dimensionless amplitude order parameter of the SDW and \( \mu_B \) is the Bohr magneton.
static part $\phi_0(\mathbf{r}_j)$ that includes the internal distortion at $\mathbf{r}_j$ and a part $\delta \phi(\mathbf{r}_j, t)$ that represents the time dependence of $\phi$ at $\mathbf{r}_j$, so that

$$
\rho_s = \rho_{0s}(\mathbf{r}_j) + \delta \rho_s(\mathbf{r}_j, t),
$$

(7)

It will also be assumed that all driven and thermal fluctuation motion of the SDW is phase variation along the chains; i.e., $Q \approx Q = 2 \pi / \lambda$ and $\mathbf{r} \approx x$, which is expressed in terms of a local condensate displacement

$$
u(x, t) = (\lambda / 2 \pi) \delta \phi(\mathbf{r}_j, t) = \sum_k u_k(t) \exp[i(kx - x_0)],
$$

(4)

where $x_0$ is the coordinate of the origin of the fluctuation in the chain. It is assumed to be uniformly distributed over an ensemble of systems. The range of $k$ is

$$
k = \frac{2 \pi n}{N \lambda} \quad - \frac{N}{2} \leq n \leq \frac{N}{2} - 1,
$$

(5)

where $n$ is an integer, $N$ is the number of SDW periods, and the corresponding condensate velocity is

$$
u_s(x, t) = \frac{\partial u(x, t)}{\partial t} = (\lambda / 2 \pi) \frac{\partial \phi(\mathbf{r}_j, t)}{\partial t}.
$$

(6)

The condition that $u(x, t)$ is real requires $u_{-k}(t) = u_k^*(t)$.

We will also assume that the thermal fluctuations are small in the sense $u(x, t) \ll \lambda / 4$, in which case

$$
\rho_s = \rho_{0s}(\mathbf{r}_j) + \delta \rho_s(\mathbf{r}_j, t), \quad \rho_{0s}(\mathbf{r}_j) = \rho_{0s} \exp[i(\mathbf{Q}_j + \phi_0(\mathbf{r}_j))], \quad \delta \rho_s(\mathbf{r}_j, t) = \rho_{0s} \frac{2 \pi i u(x, t)}{\lambda},
$$

(8)

$$
u_s(x, t) = \frac{\partial u(x, t)}{\partial t} = \frac{2 \pi i u(x, t)}{\lambda} \sum_k u_k(t) \exp[i(kx - x_0)].
$$

(9)

The $x$ dependence of $\phi_0$ represents static distortions of the SDW by pinning centers, boundaries, solitons, discommensurations, etc., and $u_k(t)$ models its phason deformation modes. It is thermal fluctuations of these modes that produce the spin-lattice relaxation considered in this paper.

Associated with the thermal and driven motion of the SDW condensate there is a corresponding charge transport.\(^6\) It is responsible for the huge dielectric response of the pinned condensate\(^8\) and the additional current and narrow band noise\(^9\)–\(^11\) observed above the depinning threshold. Following recent work on this topic, we will assume that this motion carries the entire charge of the condensate.\(^11,12\)

Now consider the electrical response $u(x, t)$ of the pinned SDW to a small amplitude electric field $E(x, t)$. Although there are well-known nonlinear and memory effects associated with SDW transport, we will assume that $u$ is a linear response to the applied field. Electrical transport\(^8\) and NMR (Refs. 11 and 12) measurements have shown that linear response is a reasonable approximation for a small driving force well below the depinning threshold. Corresponding to this electric field

$$
E(x, t) = \sum_k E_k \exp[i(kx - \omega t)] = \sum_k E_k(\omega) \exp[ikx],
$$

(10)

there will be an electric polarization $P$ of the form

$$
P(x, t) = \sum_k P_k \exp[i(kx - \omega t)] = \sum_k P_k(\omega) \exp[ikx]
$$

(11)

with

$$
P_k(\omega) = \varepsilon_0 \alpha_k(\omega) E_k(\omega) = n u_k(\omega)
$$

(12)

and

$$
u_k(\omega) = \frac{1}{2 \pi} \int_{-\infty}^{\infty} u_k(t) \exp[-i \omega t] dt.
$$

(13)

where $\varepsilon_0$ is the dielectric constant of free space, $n$ is the condensate charge density (in C/m\(^3\)), and $\alpha_k(\omega)$ is the electric susceptibility component at $k$ and $\omega$. It is complex and related to $\epsilon_k(\omega)$ by\(^6\)

$$
\alpha_k(\omega) = \alpha_k'(\omega) + i \alpha_k''(\omega) = \frac{\epsilon_k(\omega)}{\varepsilon_0} - 1.
$$

(14)

These quantities are also related to the electrical conductivity $\sigma$ through the relations

$$
\sigma(\omega) = \sigma'(\omega) + i \sigma''(\omega)
$$

$$
= \varepsilon_0 \left[ 1 + i \frac{\sigma'(\omega)}{\sigma(\omega)} \right]
$$

$$
= \frac{\varepsilon_0 \left[ 1 - \frac{\sigma''(\omega)}{\sigma(\omega)} + i \frac{\sigma'(\omega)}{\sigma(\omega)} \right]}{\varepsilon_0 - 1}.
$$

(15)

In the next section, the charge response will be related to $1/T_1$ through the Kubo formula (or the fluctuation-dissipation theorem) in the classical limit.\(^2\)

$$
\alpha_k''(\omega) = \frac{\omega_n e}{2 \varepsilon_0 k_B T} \int_{-\infty}^{\infty} \langle u_k(t) u_k(0) \rangle \exp[i \omega t] dt = \frac{\epsilon_k''(\omega)}{\varepsilon_0},
$$

(16)

where $\langle u_k(t) u_k(0) \rangle$ is the ensemble autocorrelation function of $u_k(t)$.

**B. Spin-lattice relaxation**

In this section we calculate the contribution to $1/T_1$ for nuclei with spin $I = 1/2$ caused by phase fluctuations of the SDW. The underlying physics is that thermal phase fluctuations cause a time-dependent magnetic field at the site of each nucleus. In the semiclassical picture, it is the component of this field perpendicular to the applied field $B_0$ [$\delta B_{x} (x_j)$] that causes the nuclear spin-lattice relaxation. According to NMR theory, $1/T_{1,j}$ of the nucleus at the site $l$ in the $j$th cell can be written as\(^5,13\)

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fluctuations will be suppressed because of their large energy can be ignored. Since it is expected that large-wave-vector $k$ field. It can be shown, however, that for all but relatively small values of $k$ are weighted equally and the only $k$ dependence to the coupling is that of $u(x,t)$.

Since the dipolar contribution at a nucleus depends on the distribution of nearby spin density, it has a more complicated spatial dependence. Roughly speaking, the main contribution to $\delta B_{\perp}(x_j,t)$ is proportional to the hyperfine one. If there is an additional phase shift or polarization direction change for this contribution, the result can be easily extended to include them. By making these approximations, one obtains

$$B_{\perp}(x_j,t) = B_{\perp}\cos(Q x_j + \phi_0(x_j)) + \delta \phi(x_j,t)$$

(18)

with the fluctuating component [see Eq. (9)]

$$\delta B_{\perp}(x_j,t) = \frac{2 \pi B_{\perp}}{\lambda} \sin(Q x_j + \phi_0(x_j))$$

$$\times \sum_k u_k(t) \exp(ik(x_j-x_0)).$$

(19)

Combining Eq. (19) with Eq. (17) then gives

$$\frac{1}{T_{1,i,l}} = \frac{\gamma_n^2}{2} \int_{-\infty}^{\infty} \langle \delta B_{\perp}(x_j,t) \delta B_{\perp}(x_j,0) \rangle \exp(i \omega_n t) dt,$$

where $\gamma_n$ is the nuclear gyromagnetic ratio, $\omega_n = \gamma_n B_0$ is the nuclear Larmor frequency, and the brackets are an ensemble average.

There are two predominant origins of $\delta B_{\perp}(x_j,t)$ for nuclear spin-lattice relaxation: the contact hyperfine field and the dipolar field of the spin density (we ignore the spin-orbit contribution). The contact term is proportional to the spin density at the nuclear site, so that its contribution has the spatial dependence of $\delta \rho(x_j,t)$ shown in Eqs. (8) and (9). Because this interaction is completely local, all values of $k$ are weighted equally and the only $k$ dependence to the coupling is that of $u(x,t)$.

Using well-established methods.

Equations (20) and (21) show that nuclear spin-lattice relaxation by SDW phase oscillations can be used to probe the frequency dependence of the imaginary part of the condensate dielectric function in the ordered phase. This result differs from most electrical transport measurements by its inclusion of all values of $k$ rather than only $k = 0$. A detailed interpretation of this difference is expected to rely on models for the dynamics of SDW’s.

In principle, neutron diffraction can be used to obtain the individual components $\epsilon_i(\omega)$. Despite several attempts, this approach has not yet been successful because of several very unfavorable conditions of the materials (small samples, small magnetic order parameter, and a relatively large concentration of protons).

On the other hand, proton NMR investigations of the SDW’s in Bechgaard salts can be carried out over very wide temperature and frequency ranges. The viable frequency range for such studies with proton NMR extends from a few kHz (using magnetic-field cycling) to 2 GHz with the 45 T

III. DISCUSSION AND CONCLUSIONS

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hybrid magnet under construction at the National High Magnetic Field Laboratory in Tallahassee, Florida. By exploiting this approach, it should be possible to characterize the dissipative behavior of SDW’s over an extremely broad range of frequency and temperature.

The analysis presented here has been applied to the case of a pinned SDW. There is, however, evidence from a single measurement, that \(1/T_1\), and therefore \(\Sigma \epsilon_k^b(\omega)\), is changed very little when the SDW slides. It should be interesting to see if this is a general feature of incommensurate SDW’s in Bechgaard salts and to follow up its microscopic ramifications.

In summary, we have presented a model for \(1/T_1\) by thermal phase fluctuations of an incommensurate SDW. The primary result is that \(1/T_1\) is proportional to \(\Sigma \epsilon_k^b(\omega)\).

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6 For a recent review of this subject, see G. Grüner, Density Waves in Solids (Addison-Wesley, Menlo Park, CA, 1994).
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