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# NEW NUCLEAR SPIN RELAXATION PROCESS IN TWO DIMENSIONAL METALS IN HIGH MAGNETIC FIELDS*

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An unusual linear dependence of the nuclear relaxation rate upon the magnetic field is observed in the incommensurate linear chain compound $Hg_{3-\delta}AsF_6$. It is shown that the unique two-dimensional character of the conduction electrons motion in this metal enhances the relaxation rate in strong magnetic fields. The incommensurate lattice structure gives rise to very high cyclotron frequency in these fields and makes it possible to observe the enhancement.

## INTRODUCTION

Nuclear spin lattice relaxation studies have proved to be a very useful tool for investigating the low frequency electron spin dynamics in low dimensional (1D or 2D) conductor.[1,2] The frequency dependence of the nuclear spin relaxation rate ($T_1^{-1}$) is characteristic of the dimensionality; namely, in 1D $T_1^{-1} \alpha \omega_e^{-\frac{1}{2}}$ and in 2D $T_1^{-1} \alpha \ell n \omega_e^{-1}$ where $\omega_e$ is the electron spin Larmor frequency. The typical frequency dependencies arise from the long time behavior of diffusion processes in 1D and 2D. Thus, the above characteristics are realized in conductors where the electron

motion is governed by diffusion processes. The conditions underwhich the electronic susceptibility $\chi(q,\omega)$ behaves diffusively are $\omega\tau << q\ell << 1$ where $\ell$ and $\tau$ are the mean free path and the momentum relaxation time, respectively.[3] Indeed, for almost all of the low dimensional conductors investigated so far, $\omega_e\tau << 1$ and thus there is a finite range of small q values for which $\chi(q,\omega_e)$ is diffusive. Since $T_1^{-1}$ is directly related to $Im\chi(q,\omega_e)$ and the low q values are important especially in 1D and also in 2D, the dimensionality of the system is reflected through the nuclear relaxation measurements. On the other hand, for good metals for which $\omega_e\tau$ is not much smaller than 1, there is no range of small q-values for which $\chi(q,\omega_e)$ behaves diffusively. As a result, the diffusion process is not effective and the nuclear relaxation probes the "coherent" non-diffusive susceptibility. Here we deal with the case of 2D metals for which $\omega_e\tau\sim1$ and show that the planar cyclotron motion of the conduction electrons in high magnetic fields gives rise to an enhancement of the susceptibility (and $T_1^{-1}$) which varies linearly with the magnetic field.

The linear chain incommensurate mercury compound $Hg_{3-\delta}AsF_6$ is a unique low dimensional metal. Its structure of interpenetrating perpendicular sets of relatively weakly coupled linear mercury chains results in an unusual two-dimensional Fermi surface.[4,5] de Haas-vanAlphen[5,6] and magnetoresistance[7,8] studies showed that in this metal $\omega_c\tau > 1$ in relatively small external field ($\omega_c$ is the cyclotron frequency). We show that the enhancement of $T_1^{-1}$ observed in $Hg_{3-\delta}AsF_6$ can be accounted for by the mechanism mentioned above.[6] We further show that the incommensurate structure of this material plays a crucial role in the observation of this effect.

## SPIN CORRELATION FUNCTION IN A MAGNETIC FIELD

The nuclear spin relaxation rate is given by the Fourier transform of the local spin-spin correlation function $G(t) = < S(\vec{R},t)S(\vec{R},0) >$ as

$$T_1^{-1} = a^2\int_0^\infty \cos\omega_o\tau\, G(t)dt \equiv a^2\, J(\omega_o) \qquad (1)$$

where a is the electron-nucleus interaction constant (in frequency units) and $\omega_o$ is the resonant frequency. In metals, the correlation time for which G(t) has an appreciable value is usually very short and is determined by

$\hbar/E_F$ where $E_F$ is the Fermi energy (Fig. 1).

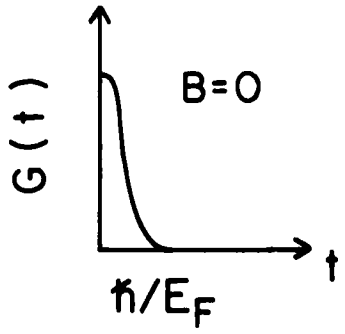


FIGURE 1 Temporal behavior of G(t) in zero magnetic field (schematic).

The resulting relaxation rate is given by the well known Korringa relation and it is field independent.

In a planar pure metal in a perpendicular magnetic field the electrons are localized on closed cyclotron orbits. (There is no component of the velocity along the field direction). Thus the local correlation function G(t) defined above is periodic in time with period determined by the cyclotron motion. Allowing for finite electronic relaxation time $\tau$, the correlation function G(t) is

$$G(t) = \sum_{n=0}^{\infty} G_o(t - 2\pi\, n/\omega_c) \exp(-2\pi n/\omega_c\tau) \qquad (2)$$

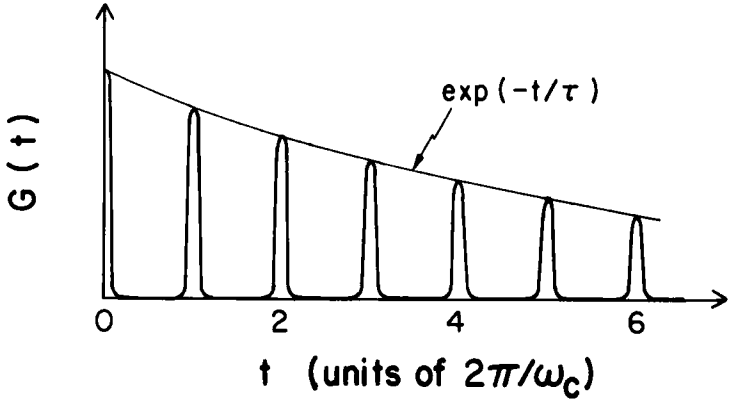


Figure 2 Temporal behavior of G(t) in strong magnetic fields, $\omega_c\tau >> 1$.

where $G_o(t)$ is the correlation function in zero field. G(t) is shown schematically in Fig. 2.

The Fourier transform of eq. (2) gives the spectral function $J(\omega_o)$ for a 2D metal,

$$J(\omega_o) = J_o(\omega_o)\ \mathrm{Re}\ \{(1-\exp(2\pi/\omega_c\tau + i2\pi\omega_o/\omega_c))^{-1}\} \quad (3)$$

where $J_o(\omega)$ is the spectral function in the absence of the field. Eq. (3) shows that (a) when $\omega_c\tau < 1$ $J(\omega)$ reduces to the usual zero field $J_o(\omega)$, and (b) when $\omega_c\tau >> 1$ the spin spectral function $J(\omega_o)$ has a special field dependence resulting from the orbital motion of the electrons. Also, the phase factor $\exp(i2\pi\omega_o/\omega_c)$ plays a crucial role in the behavior of $J(\omega_o)$. In fact, only when the phase factor is very close to unity the orbital effects could be observed.

Nuclear Spin Relaxation Behavior

Using eqs. (1) and (3) the expression for the nuclear relaxation rate becomes

$$\frac{T_1^{-1}}{(T_1^{-1})_o} = \frac{1-\exp(-X)\cos\gamma}{1+\exp(-2X)-2\exp(-X)\cos\gamma} \quad (4)$$

where $\gamma = 2\pi\omega_o/\omega_c$ $X = 2\pi/\omega_c\tau$ and $(T_1^{-1})_o$ is the usual metallic nuclear relaxation rate. For small phase parameters $\gamma << 1$, the ratio $T_1^{-1}/(T_1^{-1})_o$ increases first from 1 at low fields to a maximum value of approximately $(2\gamma)^{-1}$ at $X \approx \gamma$, and then decreases at very strong fields. This behavior is shown for $\gamma = 1/16$ in Fig. 3.

It is seen that $T_1^{-1}$ exhibits a nearly linear increase with the field for $1 < \omega_c\tau/2\pi < \gamma^{-1}$.

In experiments where the external magnetic field is not perpendicular to the metallic plane the trajectory of the electrons is still planar and it is determined by the component of the field perpendicular to the the plane. Therefore, to obtain the expression for the angular dependent relaxation rate, $T_1^{-1}(\theta)$, $\omega_c$ in eq. (4) is to be replaced by $\omega_c\cos\theta$ where $\theta$ is the angle between $\vec{B}$ and the normal to the plane. In a sample composed of many randomly oriented grains, $T_1^{-1}(\theta)$ is to be averaged over all possible directions, thus

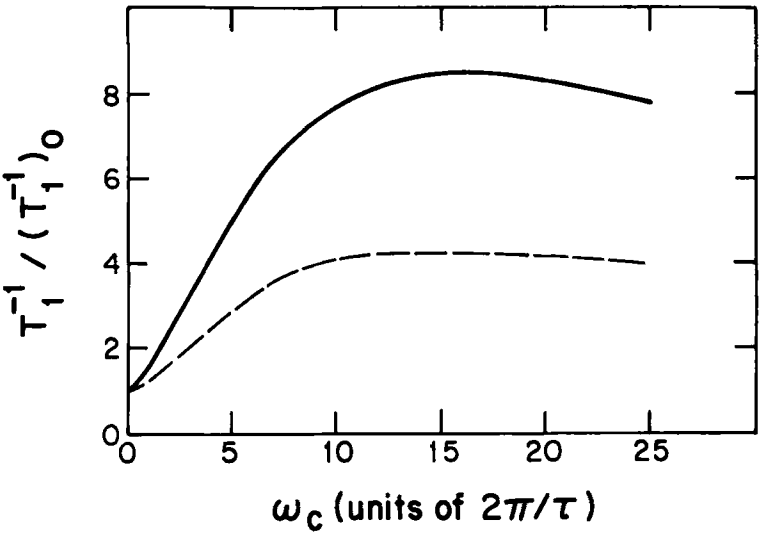


FIGURE 3 Normalized relaxation rate as a function of $\omega_c$, for $\gamma = 1/16$ (see eq. (4)). Solid line - magnetic field perpendicular to the plane. Broken line - powder.

$$T_1^{-1}(\text{powder}) = \int_0^{\pi/2} T_1^{-1}(\theta) \sin\theta \, d\theta \qquad (5)$$

The broken line in Fig. 3 shows the variation of $T_1^{-1}$ (powder) vs $\omega_c$. The general behavior is similar to that of eq. (4), although the enhancement of $T_1^{-1}$ is less pronounced.

## EFFECT OF OUT-OF-PLANE MOTION

We consider now a material composed of a large number of parallel planes with small, but finite, interplane interaction. There are two main cases to discuss: incoherent (or diffusive) vs coherent interplane motion. The effect of diffusive interplane hops is to shorten the effective electronic time $\tau$. Thus in eq. (2) $1/\tau$ is to be replaced by $1/\tau + 1/\tau_\rho$, where $1/\tau_\rho$ is the characteristic hopping rate between adjacent planes. The effect of a coherent interplane motion, on the other hand, is to change the phase factor $\gamma$ in eq. (3). Detailed calculation[9] of this effect shows that in order to account for the change in kinetic energy of the electrons, $\gamma$ is to be changed to

$$\gamma = 2\pi/\omega_c(\omega_o + v_{//}/c)$$

where $v_{//}$ is an average velocity perpendicular to the planes and c is the interplane separation.

Thus, in either the coherent or incoherent out-of-plane motion its effect is to destroy the "orbital" enhancement predicted for the nuclear relaxation rate, since the periodic nature of the local correlation function G(t) is lost.

## COMPARISON WITH EXPERIMENT

Mercury spin lattice relaxation rates were measured in a powder sample of the 2D metallic $Hg_{3-\delta}AsF_6$, as a function of the magnetic field at 4.2K(Fig. 4).

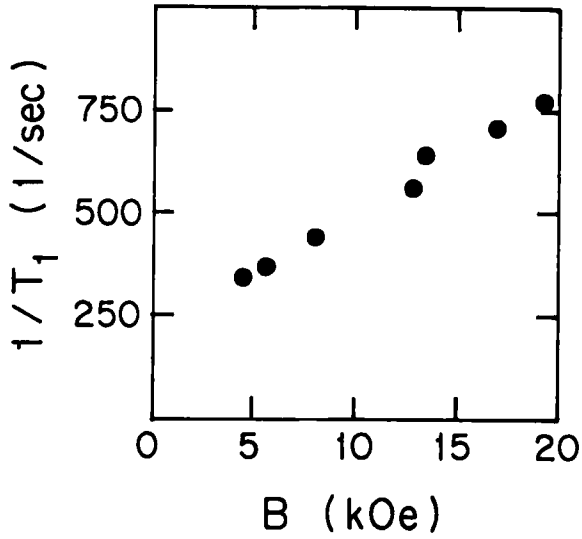


FIGURE 4 Mercury spin relaxation rate as a function of the field in $Hg_{3-\delta}AsF_6$ powder at 4.2K.

It is seen that the rate increases linearly with the field in the range 4 to 20 kOe.

The mechanism discussed above predicts such a behavior under the following conditions: (a) $\gamma << 1$ (b) $1 < \omega_c\tau/2\pi < \gamma^{-1}$ and[5] (c) $v_{//}/c < \omega_c$. In $Hg_{3-\delta}AsF_6$ the Fermi surface consists of straight cyclinders with square cross section in the $k_x$-$k_y$ plane. The incommensurability of the mercury chain lattice with the tetragonal lattice of this compound imposes superlattice structure

which results in an arbitrary small cross sectional areas in the $k_x$-$k_y$ plane.[5,6] Cyclotron orbits in an external field will, however, be finite due to magnetic breakdown effects. As a result, the corresponding cyclotron frequencies will be large but finite. de Haas-van Alphen effect studies in the field range 25 to 55 kOe showed indeed a large number - at least eight - of orbits.[6] The estimated cyclotron frequencies are in the range of 8 to 50 times the free electrons cyclotron frequency (which is equal to $\omega_e$). In lower fields $\omega_c/\omega_e$ is expected to be even larger. Thus, using for the hyperfine interaction $\omega_o \approx \omega_e$ in eq. (4), we have for the phase factor $\gamma < 0.1$ for the smallest orbit observed at 25 kOe. The observation of a straight cylinder Fermi surface in the de-Haas-van Alphen experiment[5], indicates relatively small values for $\nu_{//}$. Furthermore, the anisotropy of 100 observed in the conductivity suggests also a relatively small $\nu_{//}$. It seems therefore that the conditions (a), (b), and (c) above are fulfilled and the observed behavior of $T_1^{-1}$ in $Hg_{3-\delta}AsF_6$ is the result of the "orbital" enhancement effect.

REFERENCES

* Supported by the U. S. - Israel Binational Science Foundation (BSF), Jerusalem, Israel.

1. G. Soda, D. Jerome, M. Weger, J. Allizon, J. Gallice, H. Robert, J. M. Fabre and L. Giral, J. Phys. (Paris) 38, 931 (1977).
2. F. Devreux and M. Nechtschein, Lecture Notes in Physics Vol. 95, Proceedings of the International Conference on Quasi-One-Dimensional Conductors, Dubrovnik, Yugoslavia (1978), ed. S. Barisic, A. Bjelis, J. R. Cooper and B. Leontic, p. 145.
3. P. Fulde and A. Luther, Phys. Rev. 170, 570 (1968).
4. E. Ehrenfreund, P. R. Newman, A. J. Heeger, N. D. Miro and A. G. MacDiarmid, Phys. Rev. B16, 1781 (1977).
5. F. S. Razavi, W. R. Datars, D. Chartier and R. J. Gillespie, Phys. Rev. Lett. 42, 1182 (1979).
6. F. S. Razavi, Ph.D., thesis, McMaster University, 1980 (unpublished).
7. D. P. Chakraborty, R. Spal, A. M. Denenstein, K.-B. Lee, A. J. Heeger and M. Ya. Azbel, Phys. Rev. Lett. 43, 1832 (1979).
8. M. Weger, D. Moses, A. M. Denenstein, R. Spal, K.-B. Lee, P. J. Nigrey and A. J. Heeger, Phys. Rev. B23, 5993 (1980).
9. E. Ehrenfreund, A. Ron and M. Weger (to be published).