

Low-Magnetic Field Microwave Absorption in Superconductors and Conducting Polymers

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Abstract

Low-magnetic field microwave absorption (MA) in superconductors and conducting polymers is analysed in a low-field signal (LFS) version of the MA detecting method. The temperature dependences, hysteretic behavior and other properties of a LFS are compared in superconducting versus non-superconducting systems. Spin selective hopping processes between polarons and bipolarons is proposed to be one of the possible mechanisms of a LFS in non-degenerate conducting polymers.

1. Introduction

It is well established that all superconductors exhibit characteristic microwave absorption (MA) below the superconducting transition temperature (T_c) [1,2]. This MA changes with the magnetic field applied to the superconductor and can be detected as a change of a reflectivity or cavity quality factor by any microwave detecting setup. In most case these measurements are done by means of conventional ESR spectrometer. Although MA is detected in a range of magnetic field from the zero field up to several kOe, usually only a near zero-field region (about 50-200 Oe around the zero-field) is used for the search of superconductivity and study of superconducting properties. Accordingly, this absorption is often said to be low-field signal (LFS). At the same time, there is another scheme of MA detection called the magnetically modulated microwave absorption method (MAMMA method), the features of which will be analyzed shortly below. The method based on the MA detection (or its LFS version, LFS-method) has been widely applied for the study of conventional [1,3-5], high- T_c [1,2,6-9], organic [10-12], and novel alkali metal doped C_{60} superconductors [13-18]. Measurements of MA in superconducting (SC) state provides unique information about the T_c , granular nature, glassy behaviour and critical

state, critical fields in a Josephson medium of weakly coupled grains, fluxon behaviour, etc. Non-resonant MA proved to be a very sensitive contactless probe for the external effects, such as ambient atmosphere influence [19,20] or magnetic fields [21].

However, the appearance of an LFS-type structure in the microwave absorption spectrum does not directly imply the existence of superconductivity. For example, the weakly ferromagnetic phase of insulating gadolinium cuprate shows the LFS-type response below the crystalline temperature of magnetic ordering [22,23]. Also, a resonant near-zero field region has been observed in insulating crystals of $LiNO_3$ doped with Cu^{2+} [24], which is believed to be due to microwave absorption in $Cu^{2+} - O - Cu^{2+}$ triplets that is shifted to $H \sim 0$ [25]. To identify the LFS source as due to superconductivity or any other non-superconducting mechanism, the LFS should be tested at least on a hysteretic behavior. It has been shown that the superconducting LFS (SC-LFS) displays a hysteresis loop (resulting from critical state flux pinning forces on fluxon motion) at low modulation amplitudes [26-29], which can provide unequivocal evidence for the superconducting state.

Another type of LFS has been recently found in nondegenerate ground state conducting polymers (CP) [30]. Though the form of the LFS in CP is quite similar to that of the SC-LFS, it does not have a hysteresis and its temperature dependence is different. It has been proposed [30] that the LFS in conducting polymers (CP-LFS) might be a result of the increase of microwave absorption in low magnetic fields caused by negative a.c. magnetoresistance due to a spin selective hopping process in a pair of two paramagnetic polarons, whose rate is reduced by field-dependent singlet-to triplet transformation via hyperfine interaction [31]. In nature LFS is completely different from superconducting LFS. Recently, LFS was found below 110 K in shungite carbon [32,33] and it has simply been interpreted as a superconducting feature. This interpretation seems to be unbelievable since the signal in shungite carbon does not exhibit a hysteresis loop and the temperature dependence of intensity looks quite different. It should be noted that we have observed low temperature non-hysteretic LFS type structure in several dispersive solids, e.g. in a pyrex glass as well. Thus, the LFS of MA is itself a complex sophisticated phenomenon involving different mechanisms, and only characteristic features of LFS may allow to separate one origin from another and give the guide-points needed to choose the origin of LFS.

In this paper we present a comparative analysis of low-magnetic field microwave absorption in superconductors and conducting polymers and give the guidepoints needed to choose the origin of LFS.

2. Low-Magnetic Field Microwave Absorption in Superconductors

2.1. Modulated and Non-Modulated MA: Experimental Aspects

Since superconducting state microwave response of granular superconductors seems to be a common feature due to Josephson coupling between the grains [1,34] it can be de-

tected at the various frequencies and by the various types of microwave detecting setups. A few results published have been obtained by using of Q-band ESR spectrometers operating on about 20 GHz [5], 23 GHz [35], 35 GHz [36], and by special spectrometer with frequency variable within the range 1-38 GHz [36]. Several groups have reported the LFS study using NMR spectrometers operating in MHz frequency range [37]. However, most studies published have been done by means of conventional X-band ESR spectrometers operating on about 9-10 GHz using magnetic field modulation. The absorption can be detected both in very low magnetic field (even in the field of the earth [38,39]) and high magnetic field. Since a magnetic field H_{ext} at the sample loaded into the spectrometer cavity comprises three components,

$$H_{ext}(t) = H(t) + H_{mod} \sin(\omega_{mod}t) + H_{mw} \cos(\omega_{mw}t), \quad (1)$$

where $H(t)$ is a scanning magnetic field of a spectrometer d.c. magnet, H_{mod} is a modulation field amplitude changing with frequency $\omega_{mod} \sim 10 - 100$ kHz, and H_{mw} is a magnetic component of microwave field changing with frequency $\omega_{mw} \sim 10$ GHz, the final picture depends rather strongly on what kind of detecting scheme is used in the MA experiment.

As mentioned above, today there are a least two distinct schemes of the MA measurement: the first one is the recording of LFS and the second one is the MAMMA technique [11,40-44]. For the LFS version of MA, a spectrometer needs a minor modification. Additional d.c. Helmholtz coils must be attached to the spectrometer d.c. magnet poles to compensate for a residual magnet field. They allow magnetic field scanning around zero field within the range of $\pm 50 \sim 200$ Oe. In this method, a superconducting sample is cooled down to some chosen $T' < T_c$ in the zero d.c. magnetic field (zero field cooling, ZFC), followed by reversible H scanning within the range allowed by Helmholtz coils. ZFC is necessary to eliminate magnetic flux trapping effects [6-9]. A LFS at the next $T'' < T_c$ should be written after demagnetization of the sample at $T > T_c$, followed by ZFC. However, in many cases gradual decreasing of the temperature without demagnetization at $T > T_c$ is enough to obtain the main information about SC state.

In principle, microwave power absorbed by a sample is a function of an applied d.c. magnetic field and temperature,

$$P = P(H, T), \quad (2)$$

and the signal $I(H)$ recorded by an ESR spectrometer is proportional to a derivative of the absorbed power with respect to magnetic field. But in the case of a superconducting sample, modulated MA depends very much on the amplitude of the modulation field, [45,46]

$$I(H) \sim \left(\frac{\partial P}{\partial H} \right) H_{mod}. \quad (3)$$

On the other hand with the MAMMA method a temperature is scanned at a certain-constant magnetic field (from 100 Oe up to 10 kOe [40,41,44]) and the peak in microwave

response appears in the region of the superconducting phase transition, $T \approx T_c$. The idea behind the MAMMA technique is very clear. As was proposed by Kim et al. [40], the field modulation can be replaced by the temperature modulation in a hypothetical microwave spectrometer operating in the pure temperature mode with a temperature modulation amplitude T_{mod} [40],

$$\left(\frac{\partial P}{\partial H} \right)_T H_{mod} = \left(\frac{\partial P}{\partial T} \right)_H T_{mod}. \quad (4)$$

According to [40], the field modulation is equivalent to a temperature modulation in the region of the phase transition, and the spectrum recorded by a conventional ESR spectrometer in MAMMA scheme is the same as that of mentioned hypothetical temperature spectrometer. The MAMMA technique is good for a quick test of superconducting T_c , which the LFS method has the same advantages and provides complimentary information about the superconducting state.

In both case, LFS and MAMMA, the modulated magnetic field of about 0.05-10 Oe is always applied to the sample and affects the LFS. The third different method is a direct recording of MA absorption as a function of the d.c. magnetic field without magnetic field modulation. That was first reported by Pakulis and Osada [47] and repeated by other groups [48,49]. The absorption has been measured by using a diode detection of the microwave signal reflected at the cavity containing a superconducting sample. As the others, this method has some advantages since the part of Eq. (1) related to the modulation is eliminated. Consequently, this leads to a simplification of the situation. The method was productive in the analysis of the mechanism of MA in the range of magnetic field above the first critical field, H_{c1} [48,49].

In the following section we will concentrate mainly on the properties of LFS of MA.

2.2. Features of Low Magnetic Field Microwave Absorption in Superconductors

2.2.1. Critical temperatures and temperatures and temperature dependence of the LFS intensity

Since SC LFS of MA is a superconducting state phenomenon, one of the main LFS applications is the search of superconductivity in new materials and its T_c determination. As has been commonly accepted, the critical temperature corresponds to a temperature at which LFS appears (or disappears). The typical temperature dependence of the LFS intensity measured in the superconducting sample of $Y_1Ba_2Cu_3O_{7-\delta}$ containing only one superconducting phase is presented in Figure 1a. The LFS intensity increases quickly by a factor of several orders near the region of superconducting phase transition and saturates at low temperatures. Usually in a single-SC phase compound such dependence is smooth and does not exhibit any peaks or drops. For multiphase superconductors this dependence was shown to exhibit the drops [8]. The LFS intensity temperature depen-

dence for the sample $Bi_2Sr_2Ca_{n-1}Cu_nO_{2n+4}$ containing several superconducting phases is presented in Figure 1b (adopted from [8]). The features at 116K, 112 K, 87 K and 60 K have been interpreted as the critical temperatures of different superconducting phases [8]. All these SC phases have been proved by other methods later [50, 51]. This example shows that compared to the resistivity measurements or SQUID magnetometry the LFS method is more sensitive because it does not need a percolation through the whole bulk of material and can detect the local areas of superconducting phase surrounded by non-superconducting or even by other superconducting phase. The volume content of SC phase with $T_c = 116$ K was so small (0.01%) that it could not be detected by resistivity measurements or SQUID magnetometry [47]. At the same time, from the LFS measurements a bulk superconductor with $T_c=116$ K has been reported to exist in the Pb-doped Bi-Sr-Ca-Cu-O system [48]. and in the later case the difference between T_c measured in SQUID magnetometry and obtained from LFS was due to new superconducting phase.

The LFS method appeared to be very effective in the study of novel alkali-metal doped fullerene superconductors [13-18,52]. By means of LFS the evolution of superconductivity in bulk K_xC_{60} dependent on the variation of K-doping temperature and doping time was studied in details [15]. By using LFS a new Na-doped C_{60} superconductor having composition $Na_3N_xC_{60}$ was found and systematically studied in the compound prepared from sodium azide (Na_3) [53-58]. The critical temperature in $Na_3N_xC_{60}$ was found in the LFS measurement to vary within the range of 10-17 K. LFS is a good tool for the study of the thermal stability [56,59-61] or air-stability [20] of fullerene-based superconductors.

Recently groups of Metzger and Maruyama have succeeded in detection of LFS, that is superconductivity in 50 layered Langmuir-Blodgett film of C_{60} doped with potassium [62]. The superconducting transition occurred at about 8.1 K, a temperature lower than the critical temperature $T_c=19$ K observed in bulk K_3C_{60} [13,15]. This decreased T_c was explained by an existence of defects in the C_{60} Langmuir-Blodgett film.

2.2.2. Hysteresis of LFS

A LFS of MA in the granular superconductors exhibits exceptional hysteresis which depends on the modulation field amplitude. With small modulation field LFS changes sign whenever there is a change in direction of scanning d.c. magnetic field. The altitude of the hysteresis loop decreases with the increase of H_{mod} .

In fact, any explanation of the origin of LFS hysteresis as well as of the other properties of MA are related to the possible mechanisms of this phenomenon. Though a number of mechanisms are proposed for MA in superconducting state, none of them can completely explain all the features, since MA being a complicated phenomenon includes many different mechanisms. One approach to a mechanism of LFS and its hysteresis is presented in paper of Blazey and Portis with co-workers [1,26-29]. According to their approach MA is, in fact, dissipation of microwave power in bulk superconductor due to viscous fluxon motion [63]. Josephson junctions connecting the grains allow the flux to enter at a very low effective H_{c1}^* [1]. The change of the LFS sign at the reverse sweep is related to the change of the critical state at the surface [1,28], i.e. to the pinning and depinning

of fluxons during each modulation cycle. Since H_{mw} and H_{mod} are always applied to the sample in a cavity, the critical current J_{cs} already flows at the surface of grain at these low fields and contributes to the microwave absorption. The surface critical current reverses with reversing the field sweep only after a certain field interval equal to twice the Josephson critical field [1]

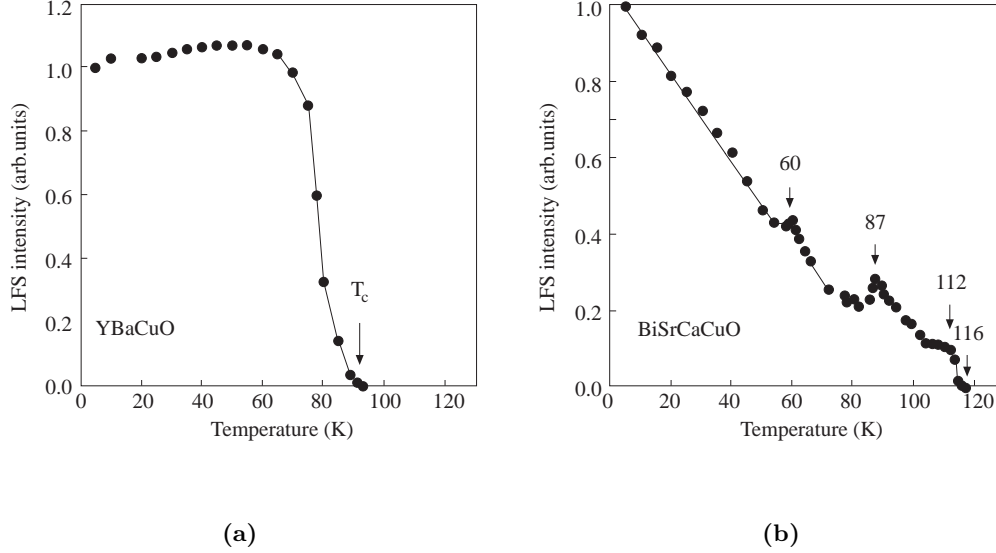


Figure 1. Typical temperature dependencies of the LFS peak-to-peak amplitude in the single superconducting phase $Y_1Ba_2Cu_3O_{7-\delta}$ (a) and in $Bi_2Sr_2Ca_{n-1}Cu_nO_{2n+4}$ (b) containing several superconducting phases

$$2H_{c1}^* = \frac{4\pi}{c\lambda_J J_{cs}}, \quad (5)$$

where λ_J Josephson penetration depth. The same change of the critical current occurs over a modulation field cycle. Typical dependence of the MA level on H_{mod} at a fixed magnetic field of 20 G obtained for Na-doped C_{60} superconductor is presented in Figure 2. (adopted from [64]). Such dependence usually exhibits two linear parts [1,56, 64]. These two regions involve two different contributions [1,64]: the signal induced by the surface critical current for $H_{mod} < H_{c1}^*$, and the signal caused by changing the fluxon density for $H_{mod} > 2H_{c1}^*$, where MA absorption is proportional to the concentration of fluxons [45]. The minimum point between these two regions corresponds to $H_{mod} = 2H_{c1}^*$. On increasing the temperature, the minimum point shifts to lower field.

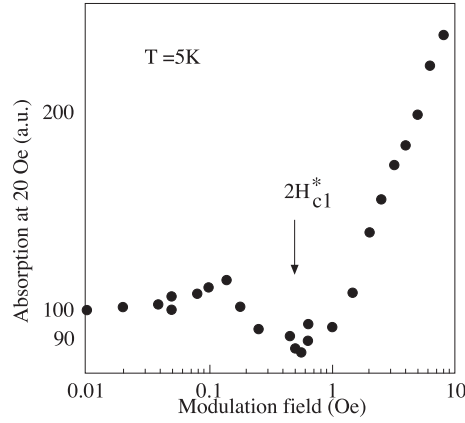


Figure 2. Typical dependence of the microwave absorption upon modulation field amplitude H_{mod} at a certain fixed d.c. magnetic field 20 Oe measured in Na-doped C_{60} superconductor (adopted from Ref [56])

An alternative approach to the explanation of hysteretic behavior and, correspondingly, a mechanism of magnetically depended MA has been proposed by Pakulis and Osada [47], who related an absorption of microwaves to the resistivity losses on normal charge carriers appearing above H_{c1} and associated the hysteresis with a magnetization changing under flux trapping. The same relationship between MA and magnetization has been pointed out by Rubins et al. [65]. The abrupt change of slope occurring at low fields in the magnetization curve and which could be seen as a peak in derivative MA is consistent with the sudden drop in d.c. susceptibility predicted by Ebner and Stround [66,67].

One more mechanism of MA is the high-frequency a.c. susceptibility [68].

2.2.3. LFS peak position and oscillations

The low field signal is a microwave response written by scanning the magnetic field from the negative to positive field through zero. The sign of the LFS shape is opposite to a conventional ESR signal. The integrated signal exhibits a minimum of absorption exactly at $H = 0$ Oe, while the integrated ESR signal has a maximum at the same point. Then if one were to look at only the right part of LFS starting from $H=0$ Oe one can see that the derivative of absorption, $dP/dH = f(H)$, of the sample cooled in zero magnetic field has a peak at $H_p = 0.1 - 100$ Oe. If there is a current loop with the radius r , the flux starts to penetrate into the loop at the field of $H_0 \sim \Phi_0/2\pi r^2$, where $\Phi_0 = ch/2e = 2 \times 10^{-7}$ Oe * cm² is the flux quantum. The size of the loops in granular superconductor varies over a wide range, and there is a maximum in the size distribution of the loops. Most groups [26] associate this field H_p with the maximum in the size distribution of the loops, $H_p \simeq H_0$, and use it to deduce an average projected area $\langle S \rangle = \pi r^2$ of the superconducting loop.

$$\langle S \rangle = \Phi_0/2H_p. \quad (6)$$

It should be pointed out that the loop may encompass either only one grain or several grains connecting to each other and making the loop. Accordingly with Ebner and Stroud, when the applied magnetic field induces a current greater than the critical current the junction breaks down, emits a flux quantum accompanied by MA. H_p occurs at the values of the magnetic field where one can initiate the maximum number of flux slips.

Summarising this paragraph, though a number of mechanisms are proposed for MA in a SC state, none of them can completely explain all the features, since MA includes many different mechanisms. Though one of the explanations of MA has been proposed to be the high-frequency a.c. susceptibility [68], most properties, however, are explained, as mentioned above, in terms of the SC grains weakly coupled through Josephson contact [1]. In this case MA occurs due to either a viscous motion of the Josephson vortices [45] and hypervortices [38,39] or the current slippage processes in the loops of weakly coupled SC grains [69,70]. The other mechanisms can contribute to MA near T_c . Such as the dissipative relaxation of Abrikosov vortices in a microwave field which appear in the bulk at $H > H_{c1}$ [71], and the second one is caused by electrons in the normal state in the cores of vortices at $H_{c1} \ll H \ll H_{c2}$ [47,72]. As the mechanisms may be, the MA in SC state is characterized by several features: characteristic temperature dependence, hysteretic behaviour, oscillations.

Now we will change our subject to analysis the microwave absorption in nondegenerate conducting polymers which has completely different properties and origin.

3. Low-Field Microwave absorption in Conducting Polymers

3.1. Experimental

Microwave absorption measurements in conducting polymers have been carried out using the same experimental setup and conditions which have been used for the study of high- T_c oxides. A LFS was detected in nondegenerate CP such as poly(p-phenylene) (PPP), poly(p-phenylene vinylene) (PPV), but not was detected in degenerate CP polyacetylene. The LFS in CP was found to depend on the doping condition. Most of the experiments to be described in this paper are on nominally undoped PPP prepared by the Kovacic method [27]. As a consequence of impurities, the as-synthesized samples are very lightly p-doped. Nominally undoped PPP films were prepared by vacuum deposition of ppp [28]. Other conducting polymers consisted of polyperinaphthalene (PPN), undoped or lightly p-doped [29], and undoped PPV [30]. By undoped we mean that the samples are not intentionally doped. The “undoped” samples contain small concentrations of predominantly p-dopants as impurities.

3.2. Properties of LFS in Conducting Polymers in Comparison with those in High T_c Cuprates

3.2.1. The signal shape and hysteresis

All of the conducting polymers under study showed LFS (Figure 3).

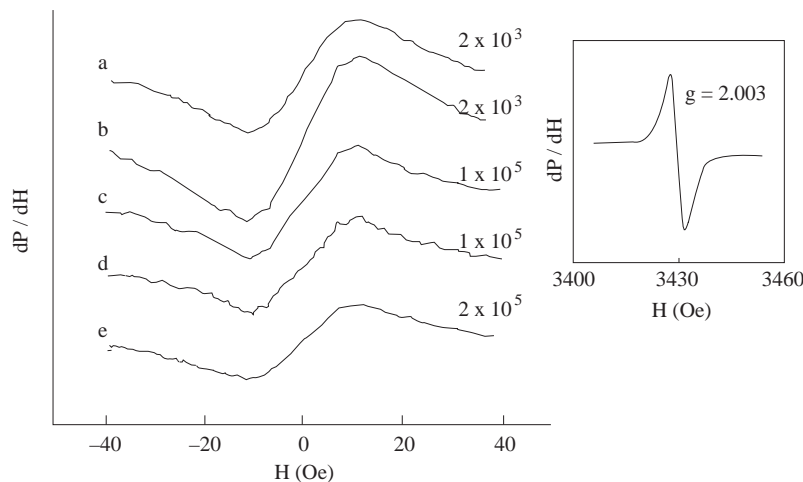


Figure 3. The low signal of microwave absorption in nondegenerate conducting polymers (CP-LFS): consequently, in undoped polyparaphenylene (PPP)-powder (a); undoped PPP thin film (b); polypiperinaphthalene (PPN) (c), polypyrrol (d); Poly(p-phenylene vinylene) (PPV) (e). Inset shows regular ESR signal of the polarons typical for those polymers

Among them the most intense signal was exhibited by the undoped PPP prepared using $CuCl_2 - AlCl_3$, which was a factor of 5 more intense than the LFS in undoped PPP prepared using $MoOCl_4$, and was larger much more than that in PPV, PPN and polypyrrol. As seen in Figure 3 the shape of the LFS is quite similar to LFS detected in high T_c cuprates and is opposite to the usual ESR derivative signal presented in an inset of Figure 3. The ESR signal with $g = 2.003$ in this inset is the paramagnetic resonance of polarons. As mentioned in our previous paper [25], along with the LFS and ESR of polarons a few paramagnetic resonance signals associated with Cu, Fe, and Mo cations were observed in the spectra recorded out to 7000 Oe as well. We shall skip consideration of those signals in present paper.

No critical state hysteresis loop with decreasing and increasing d.c. magnetic field was observed in the LFS of all the measured CP, suggesting a non-superconducting and non-magnetic origin for the LFS in CP.

3.2.2. The LFS intensity temperature dependence

Figure 5 shows the temperature dependence of the CP-LFS amplitude (designated on

a Figure 4 as I^{GFS}) normalized to the lowest temperature amplitude measured at 4.2 K for undoped PPP samples (synthesized using $CuCl_2 - AlCl_3$) in comparison with the analogous dependence of $Y_1Ba_2Cu_3O_{7-\delta}$ ceramic. The LFS becomes detectable below about 120 K in undoped PPP and 160 K in the lightly sodium-doped samples. The LFS amplitude increases rapidly with the temperature lowering, and becomes approximately three orders of magnitude larger than that at 120-160 K. A clear onset point followed by means field saturation at low temperatures, generally observed in the cuprate superconductors was found for conducting polymers.

3.2.3 A possible mechanism of microwave absorption in nondegenerated conducting polymers

In the case of electron spin resonance (ESR), a sample loaded into the microwave field $H_{mw} \cos(\omega_{mw})$ absorbs power $P = \omega_{mw} \chi'' H_{mw}^2 / 2$, where H_{mw} is a magnetic component of a microwave, ω_{mw} is a microwave frequency, and χ'' is an imaginary part of dynamic magnetic susceptibility of the sample. Does the LFS in CP have the resonance origin like ESR signal? The left part of the CP-LFS derivative spectra taken from $H = 0$ Oe up to approximately 500 Oe itself is similar to the ESR signal of Dysonian line shape (Figure 3). If one speculates that this is the ESR signal, the g-value could be estimated to be $g=127$ for a PPP polycrystalline powder and $g=132$ for a PPP polycrystalline film. But if this is the case, it is difficult to explain such large g-values by an existing in PPP of paramagnetic species resonating at $H \sim 30 - 50$ Oe. As well known, H_{mw} is maximal at the center of ESR spectrometer rectangular cavity and is minimal at the half of distance between the center and the cavity wall. Vice versa, the electrical component of a microwave E_{mw} is maximal at the later point and is minimal in a cavity center [73]. Assuming, that this minimal E_{mw} at the sample place at the cavity center effects the microwave absorption, one can consider another mechanism of LFS of MA in PPP which has been recently proposed [30] based on Kivelson's hopping model of conductivity [74,75].

In the case of nondegenerate CP, the charge carriers are charged polarons (P) and bipolarons (BP). The CP-LFS mechanism proposed involves the polaron and bipolaron hopping transport and spin flip processes between singlet (S) and triplet (T_{-1}, T_0, T_{+1}) state of polaron-bipolaron pairs. The undoped PPP samples in which LFS was observed have d.c. conductivities which are low, less than 10^{-11} S/cm. However, from ESR measurements, it is evident that about 10^{17} spins/cm³ are present in the sample volume, most probably as charged polarons (P^+) which are formed by unintentional doping with the oxidant used for polymerization (e.g. $CuCl_2 - AlCl_3$ or $MoOCl_4$). Light sodium doping of the PPP increases the $g=2$ ESR intensity by an amount which would correspond to a polaron concentration of about 10^{18} cm⁻³. A temperature dependent equilibrium exists between polaron, bipolaron, and polaron pair concentrations [74]. In the case of the heavily doped PPP, most of the charges are in the form of bipolarons. From the entropic viewpoint, separated polarons are favorable at high temperatures, while the amount of spinless species (bipolarons) or coupled polaron pairs increase with decreasing temperature.

Kivelson's equations for the d.c. and a.c. conductivity of polyacetylene were slightly modified and applied by Kuivalainen et al. to conduction processes in PPP [75]:

$$\sigma_{dc}(T) = \frac{Ae^2\gamma(T)}{k_B T} \left[\frac{\xi}{R_0^2} \right] \frac{Y_p Y_{bp}}{(Y_p + Y_{bp})^2} e^{-2BR_0/\xi}, \quad (7)$$

$$\sigma_{ac}(T) = \sigma_{dc}(T) + \frac{e^2}{384\hbar} \left[\frac{C_{imp}^2}{k_B T} \right] \frac{Y_p Y_{bp}}{(Y_p + Y_{bp})^2} \xi_{\parallel}^3 \xi_{\perp}^2 \hbar\omega \left[\ln \left[\frac{2\omega}{\Gamma_0} \right] \right]^4. \quad (8)$$

Here, $A = 0.45$; $B = 1.39$; Y_p and Y_{bp} are the concentrations of polarons and bipolarons, respectively; $R_0 = (3/4\pi C_{imp})^{1/3}$ is the typical separation between impurities whose concentration is C_{imp} ; ξ_{\parallel} and ξ_{\perp} are the average decay lengths of a polaron or bipolaron wave function; ξ_{\parallel} and ξ_{\perp} being the decay lengths parallel and perpendicular to the polymer chain, respectively, and

$$\Gamma_0 = \left[Y_p Y_{bp} / (Y_p + Y_{bp})^2 \right] \gamma(T) \quad (9)$$

where, here,

$$\gamma(T) = \gamma_0 [T/300K]^{n+1} \quad (10)$$

is the transition rate of an electron between polaron and bipolaron states depending on the temperature.

Assuming that magnetic field influences the conductivity, and that at low temperature $\sigma_{ac} \gg \sigma_{dc}$ [75], Eq.(8) may be transformed to:

$$\sigma_{ac}(T, H) = \frac{D}{T} \left[\ln \left[\frac{2\omega}{\Gamma(T, H)} \right] \right]^4, \quad (11)$$

here

$$D = \frac{e^2}{384\hbar} \left[\frac{C_{imp}^2}{k_B} \right] \frac{Y_p Y_{bp}}{(Y_p + Y_{bp})^2} \xi_{\parallel}^3 \xi_{\perp}^2 \hbar\omega \quad (12)$$

and

$$\Gamma(T, H) = \Gamma_0 \tilde{\gamma}(H), \quad (13)$$

where $\tilde{\gamma}(H)$ is the component of the transition rate depending on magnetic field. Since a conventional ESR spectrometer measures a derivative of modulated microwave absorption, dP/dH not by direct absorption, it is difficult to determine a value $P(T, H)$ (which is actually MA of the sample loaded into the cavity) without special modifications of the spectrometer. For the LFS in conducting polymers, in the absence of d.c. magnetic field ($H = 0$) this value $P(T, 0)$ is minimal and increases with magnetic field up to 20-50 Oe,

thus $(P(T, H) > P(T, 0))$ as it is schematically presented in Figure 4. The CP-LFS intensity (I^{LFS}) taken from the integrated signal as it is shown in Figure 4 is proportional to the change of microwave conductivity due to magnetic effect and is given by:

$$\Delta\sigma(\omega, T, H) = [\sigma(\omega, T, H) - \sigma(\omega, T, 0)] \sim I^{LFS}(T, H) = [P(T, H) - P(T, 0)]. \quad (14)$$

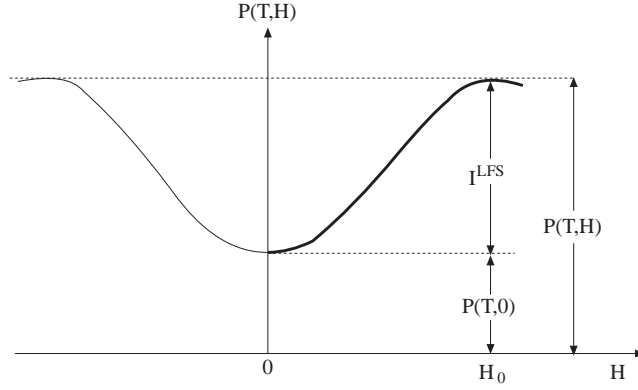


Figure 4. The temperature dependence of the peak-to-peak amplitude of the CP-LFS in the undoped PPP normalized to the amplitude at 4.2 K

3.2.4 Temperature dependence of CP-LFS

We have shown previously that the temperature dependence of the LFS intensity in CP could be approximated by CT^α (where α is about -1.8 and C is a temperature independent constant) [30]. The same law CT^α with C constant and $\alpha = -2$ was obtained for the LFS intensity temperature dependence in insulating Mn-V-Mo oxides [76], where the existence of polarons and bipolarons was suggested theoretically [77]. Below we will try to approximate the CP-LFS temperature dependence using Eq. (14). However, one should keep in mind that this procedure looks rather artificial and has an illustrative meaning. After normalization of the CP-LFS intensity to that at some fixed temperature T_0 and magnetic field H_0 , one can write the following equation which can be used for the fitting procedure:

$$\frac{\Delta\sigma(\omega, T, H_0)}{\Delta\sigma(\omega, T_0, H_0)} \frac{\sigma(\omega, T, H_0) - \sigma(\omega, T, 0)}{\sigma(\omega, T_0, H_0) - \sigma(\omega, T, 0)} = \frac{I^{LFS}(T)}{I_0^{LFS}(T_0)}. \quad (15)$$

Substituting Eqs.(9-11) in Eq.(15) gives:

$$Y = \frac{1}{M} \left\{ [\ln \alpha_1 + (n+1)X]^4 - [\ln \alpha_0 + (n+1)X_0]^4 \right\}, \quad (16)$$

where

$$\begin{aligned}
 Y &= \frac{T}{T_0} \times \frac{I^{LFS}(T)}{I^{LFS}(T_0)}. \\
 X &= \ln \left(\frac{300K}{T} \right); \quad X_0 = \ln \left(\frac{300K}{T_0} \right) \\
 M &= [\ln \alpha_1 + (n+1)X_0]^4 - [\ln \alpha_0 + (n+1)X_0]^4 \\
 \alpha_0 &= \frac{2\omega}{Y_p Y_{bp} / (Y_p + Y_{bp})^2} \times \frac{1}{\tilde{\gamma}(0)}; \quad \alpha_1 = \frac{2\omega}{Y_p Y_{bp} / (Y_p + Y_{bp})^2} \times \frac{1}{\tilde{\gamma}(H_0)}.
 \end{aligned}$$

After cumbersome transformation Eq.(16) can be presented as the third order polinomial regression:

$$Y = \beta_0 + \beta_1 X + \beta_2 X^2 + \beta_3 X^3, \quad (17)$$

here

$$\begin{aligned}
 \beta_0 &= \frac{1}{M} \left[(\ln \alpha_1)^4 - (\ln \alpha_0)^4 \right] \\
 \beta_1 &= \frac{4(n+1)^2}{M} \left[(\ln \alpha_1)^3 - (\ln \alpha_0)^3 \right] \\
 \beta_2 &= \frac{6(n+1)^2}{M} \left[(\ln \alpha_1)^2 - (\ln \alpha_0)^2 \right] \\
 \beta_3 &= \frac{4(n+1)^3}{M} [\ln \alpha_1 - \ln \alpha_0].
 \end{aligned}$$

As it is seen in Figure 6, experimental points are well fitted by this regression. I_0^{LFS} is the LFS intensity at $T_0 = 6.4K$ chosen as the normalization temperature.

3.2.5. Magnetic field dependence of microwave absorption

An analogous procedure applied above for the approximation of the temperature dependence of LFS intensity at fixed magnetic field was shown to be much more cumbersome when T is fixed and H is varied. So only the qualitative explanation of magnetically depending microwave absorption is presented below.

CP-LFS measurement shows an increase of I^{LFS} with magnetic field which corresponds to $\Delta\sigma(\omega, H) = \sigma(\omega, H) - \sigma(\omega, 0) > 0$ or to a negative a.c. magnetoresistivity. To explain this negative magnetoresistance we have considered the transitions between polaron and bipolaron triplet (T_{+1}, T_0, T_{-1}) and singlet (S) states.

In the general case of singlet-triplet pair, transition between T_0 and S states is induced by difference between Zeeman precession frequencies of electrons and hyperfine interaction between electron and magnetically active nuclei [31]. In the former case, the characteristic

spin evolution time is $\tau_{ev} \simeq (\Delta g \beta H)^{-1}$, where Δg -value is the g-value difference within the pair, β is Bohr magneton (Δg -value mixing mechanism), and in the latter case, $\tau_{ev} \simeq (g_e a_{eff})^{-1}$, where g_e -value is electron Gyromagnetic ratio, a_{eff} is the hyperfine interaction constant (hyperfine mechanism) [31]. Since Δg -value of a polaron-bipolaron pair is negligible and the major increase in microwave absorption occurs at low magnetic fields, $H \sim 10 - 15$ Oe, Hyperfine mechanism is most reasonable for the explanation of the CP-LFS phenomenon. This mechanism can provide a magnetoresistance which arises at small fields comparable with $a_{eff} = 10 - 70$ Oe and saturates at $H > a_{eff}$ [31]. The hyperfine interaction mechanism has been used by Francevich et al. [78-80] to explain the observed positive d.c. magnetoresistance of nominally undoped polyacetylene, weakly doped polacetylene, nominally undoped PPP, and some other polymers. Since the hopping processes considered by these authors results in a positive d.c. magnetoresistance, while the present CP-LFS corresponds to a negative a.c. magnetoresistance, at least at microwave frequencies, we have proposed different spindependent hopping process which result in a sign change for magnetoresistance in goin form d.c. to microwave frequencies [30].

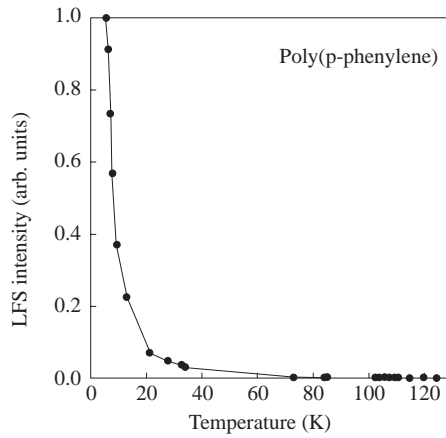


Figure 5. Schematik representation of the LFS of conducting polymer in the vicinity of the zero d.c. magnetic field as it is derived by integration of the experimental derivative signal. (See text for details.)

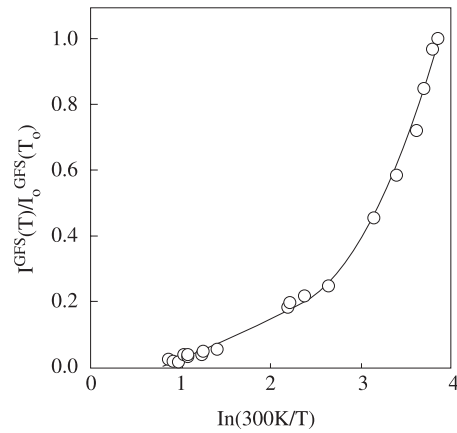


Figure 6. Experimental data are fitted by Eq.(17). I_0^{LFS} is the LFS intensity at $T_0 = 6.4$ K chosen as normalization temperature

Polarons and bipolarons are trapped at low temperatures in close proximity to dopant ions by coulomb energies of about 0.2 eV, since coulombic screening is not effective at low dopant concentrations. Consequently, the predominant hopping process for microwave conductivity should be highly localized and correspond to the transfer of polarons or bipolarons back and forth between chains which are nearest neighbor to the dopant ions.

The following hopping processes between chain segments 1 and 2 in the same proximity to a dopant ion can be considered.

$$P_1 \xrightarrow{k_p} P_2, \quad (18)$$

$$BP_1 \xrightarrow{k_{bp}} BP_2, \quad (19)$$

$$BP_1 + P_2 \xrightarrow{k_{pbp}} BP_2 + P_1, \quad (20)$$

where the subscripts denote the chain upon which the P and BP is located. However, these three processes (18, 19, 20) or (18-20) are not spin dependent, since the spin before and after the hop is unchanged. Only the process of the hopping of a charge from a polaron on a one chain to a polaron on a second chain with a bipolaron formation, $P_1 + P_2 \rightarrow BP_1$ or BP_2 , and the reverse process is spin dependent. The hopping is allowed from the singlet state of polaron pair, $(P_1 + P_2)^S$, and is forbidden from the triplet pair, $(P_1 + P_2)^T$, as presented below.

$$\left. \begin{array}{l} (P_1 + P_2)^S \xrightarrow{k} BP_1 \text{ or } BP_2 \xrightarrow{k'} (P_1 + P_2)^S \\ (P_1 + P_2)^T \xrightarrow{\text{forbidden}} BP_1 \text{ or } BP_2 \xrightarrow{\text{forbidden}} (P_1 + P_2)^T \\ (P_1 + P_2)^S \xleftarrow{k_{ST}} (P_1 + P_2)^T \end{array} \right\} \quad (21)$$

At $H = 0$, the singlet state is mixed equally with all three components of triplet state, T_{-1}, T_0 and T_{+1} when the life time relevant for hyperfine mixing is negligible. Consequently, the effective hopping rate at zero field for polaron recombination to form a bipolaron by an interchain hop is reduced from that for an allowed hop $(P_1 + P_2)^S \rightarrow BP$, since the weight of the singlet state is only 1/4. However, when $H > a_{eff}$ the singlet state S is mixed only with the T_0 component of a triplet, while T_{-1} and T_{+1} are Zeeman shifted in energy and cannot be populated by hyperfine interactions. Hence, the weight of singlet state (w_S) is 1/2, as opposed to the above discussed value of $w_S = 1/4$ for $H = 0$. BP hopping by the above mechanism depends sequentially upon (18) formation of an interchain polaron pair from a bipolaron (with rate constant k') and (19) the reverse process to reform the bipolaron on either the original chain or on neighboring chain (with rate constant from the pure singlet state of k). Consequently, the frequency of dopant-localized bipolaron hopping (ν) is obtained as follows in the limit where processes (18-20) can be neglected:

$$\nu = \left[(k')^{-1} + (w_S k)^{-1} \right]^{-1}, \quad (22)$$

where w_S is the above mentioned field-dependent singlet contribution to the polaron pair distribution. Since w_S increases from 1/4 to 1/2 as H increases from zero to about a_{eff} ,

and since a.c. conductivity is proportional to ν , the maximum low field $\Delta\rho_w/\rho_w$ can vary on going from $H = 0$ to $H \sim a_{eff}$ from near zero (when $k/k' \gg 1$) to 0.5 (when $k/k' \ll 1$). This maximum predicted effect, which corresponds to negative magnetoresistance, will decrease in magnitude as the spin-independent hopping processes (from (18) to (20)) become important.

In other words, if the lifetime of the $(P_1 + P_2)^S$ pair (t) is longer than $\tau_{ev} \simeq (g_e a_{eff})^{-1}$, but smaller than the spin-lattice relaxation time $T_1 (g_e a_{eff} < t < T_1)$, then the triplet states T_{-1} and T_{+1} are populated at high field neither through hyperfine interactions nor from thermal equilibrium with lattice. However, S, T_{+1}, T_0 , and T_{-1} are equally populated at $H = 0$ under these same conditions. Consequently, a decreased triplet state contribution and a correspondingly increased singlet state contribution with increased magnetic field increases the hopping frequency due to mechanism (21), which leads to increased microwave absorption for increased magnetic field.

4. Conclusion

In conclusion, we have considered comparatively the features of the low field microwave absorption in superconductors and conducting polymers. A LFS in nondegenerate conducting polymers was examined from the viewpoint of the negative magnetoresistance caused by polaron-bipolaron spin-dependent hopping processes. However, it should be noted that the different mechanisms might contribute to the low field magnetoresistance of CP. As an example, the spin-flip hop model of Movagher-Schweitzer-Osaka [81,82] provides an alternate or additional possible explanation for the negative magnetoresistance evidenced by the LFS.

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