Magnetic quenching of the three-photon annihilation rate in some molecular solids

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The triple-coincidence technique, coupled with magnetic fields, can be used to feature the behavior of positron bound systems in condensed matter. In particular, data concerning the anomalous magnetic quenching of positronium in some nonmetallic solids are presented; comparisons with results from lifetime spectroscopy are made. Possible applications of the technique are discussed.

INTRODUCTION

The magnetic quenching of positronium[1] (Ps) has often been used to clarify several aspects of this exotic atom. For instance, shortly after the discovery of Ps, this technique was used for a first measurement of the Ps hyperfine splitting,[2] as well as to study the polarization of the positrons emitted by a ^{22}Na source.[3] The existence of a positron bound system in alkali halides was demonstrated by quenching the corresponding lifetime component by using magnetic fields.[4] Furthermore, the different structure of electron-positron correlated pairs in condensed matter with respect to Ps *in vacuo* has been shown through several magnetic quenching experiments:[5] it has been found that the electron density at the positron is generally lower than the same quantity for vacuum Ps.

Magnetic quenching has also been used as an heuristic method, for instance to discriminate between ortho-para conversion and oxidation reactions of Ps in the presence of paramagnetic compounds,[6,7] or to shed light on the origin of components in the positron lifetime spectra in some nonmetallic solids.[8]

Although the magnetic quenching method can be coupled, in principle, to every positron annihilation technique, most of the results were obtained through the lifetime spectroscopy because of its simplicity and high efficiency. However, the use of alternative techniques is always desirable, in the first place because the results of measurements acquire more reliability if they are obtained through different methods; and, secondly, because the explanation of a physical effect can be submitted to experimental cross tests if the data are achieved through several techniques. For these reasons, we deemed it worthwhile to carry out magnetic quenching measurements through an experimental technique, which is different from the lifetime spectroscopy. The data shown in this paper are achieved by using the three-γ coincidence technique; even though interesting results were obtained many years ago with this method in the presence of magnetic fields,[9,10] it was no longer exploited, perhaps because of the low efficiency shown in the first investigations. However, this drawback is only apparent, since in recent years we have seen an increase in the performance of the apparatus based on this method.[11] Actually, while taking our measurements we reached the conclusion that the times required to carry out a magnetic quenching experiment using the lifetime spectroscopy or the three-γ technique are nowadays comparable.

In order to check the usefulness of the aforementioned technique in the presence of magnetic fields, we investigated the magnetic quenching of positron bound systems in some nonmetallic solids, where the behavior of the system is already known from the lifetime spectroscopy. This choice is due to some unexpected results, discovered in the last decade: the magnetic quenching at low fields (0.1–0.7 T) in some polymers,[12,13] in a few molecular solids,[14,15] in liquid solutions of nitrobenzene in *n*-hexane[16] as well as in *n*- and iso-octane,[17] has been found to be much stronger than could have been expected on the basis of the Halpern theory. This phenomenon will be named here after the two people who independently observed it for the first time the Bisi-Rochanakij (BR) effect. These results are made all the more interesting by the many different explanations proposed. Actually, the BR effect was supposed to be due to different scaling factors for intrinsic decay rates and hyperfine splitting of the positron bound system under study in respect to vacuum Ps;[14] or to ortho-Ps quenching by diradical triplet excited states, whose formation (through intersystem crossing) should be dependent on the applied field.[18] The Strasbourg group explained the BR effect in liquid by invoking the formation of a complex between Ps and the solute ground state.[19] Another possibility taken into consideration was that the anomalous effect would originate from the motional magnetic field probed by the electron-positron pair moving in a sufficiently strong molecular field.[20] Mogensen proposed that a particular "swollen" Ps-molecule state, resulting from attachment and/or detachment of Ps to the molecule, gives the BR effect; he also predicted a qualitative dependence on the temperature of the process.[21] Eventually, a recent interpretation is that the effect is promoted by a positron interacting with two different electrons with unpaired spins.[22] The situation is complicated by the fact that the corresponding models are generally able to fit the experimental data quite well; this can be ascribed to the presence of several adjustable parameters. From this example we can realize the importance of having available data coming from different experimental methods: in such a case, comparing the various explanations would be made easier, since a multiparametrical model should fit all the results simultaneously.

To summarize, the aim of this work was twofold: to show that the magnetic quenching of the three-γ coincidence rate can supply comparable information to that obtained from lifetime spectroscopy and, at the same time, to give an experimental contribution to the problem of the understanding of the BR effect.

EXPERIMENTAL

The positron source consisted of a droplet of ^{22}Na from a carrier-free neutral solution, deposited between two identical circular kapton foils (1.08 mg/cm^2 each, diameter 20 mm), which were afterwards glued together. The activity was measured to be about 30 μCi. The investigated materials were three molecular solids: naphthalene, 2,5 diphenyloxazole (PPO) and byphenil, and the polymer polyethylene terephthalate (Mylar). Naphthalene, scintillation grade (purity $>99.5\%$) and PPO, scintillation grade (purity $>99.8\%$) were purchased from Fluka Chemie AG; byphenil (purity $>99.8\%$) and Mylar (density 1.375 g/cm^3, inherent viscosity 0.59) were purchased from Aldrich. They had already been studied with the lifetime spectroscopy coupled with magnetic fields (Refs. 14, 24, 23, and 13, respectively). The samples were formed by pressing the polycrystalline molecular solids; Mylar was supplied in the form of pellets that were afterwards compression molded. All the samples were in the form of 20-mm-diam. discs; the thickness was always about 200 mg/cm^2, that is, sufficient to stop all the positrons emitted by the radioactive source. This was inserted between two identical samples in the usual "sandwich" configuration.

The spectrometer consisted of three identical "channels," composed of an integral line—a 2″×2″ NaI(Tl) scintillator coupled to a photomultiplier tube—and a preamplifier stage, followed by an amplifier and a timing single channel analyzer. The outgoing signal from this last stage was sent to a coincidence unit, whose output fed a counter and timer. A block diagram of the apparatus is shown in Fig. 1. The scintillators were arranged in a symmetrical way (120° from each other); their symmetry axes were coplanar and in the same plane of the source. Each scintillator was surrounded by a truncated conical lead shield, to make certain that the crystals could not "see" each other. The opening of the shield allowed the corresponding scintillator to be seen by the positron source under a solid angle of 0.22 sterad. The γ rays collected by the scintillators were processed by the timing single channel analyzer, in order to accept only those quanta in the range 280–420 keV, that is, a region centered around 340 keV (indeed, each scintillator receives one third of the total annihilation energy, 1022 keV—without taking the spurious events into consideration).

A preliminary setting of the delays of each timing single-channel analyzer allowed three simultaneously generated events to be present at the input of the coincidence unit; the time resolution of the system was measured to be about 25 ns.

The assembly source sample was placed in the center of the gap of an electromagnet suitably designed for this kind of experiment. It consisted of two truncated-conical iron poles wrapped in coils made by a copper tube. The twofold function of the coil was to magnetize the iron, through a dc current, variable in the range 0–100 A, as well as to cool the magnet by means of water flowing inside the tube. The magnet was surrounded by an external iron covering, in order to supply a return path for the magnetic flux; in such a way, the effect of stray fields outside the magnet was minimized. Three symmetrical circular holes in the covering allowed the γ rays to be detected and, at the same time, made it possible to change the sample whenever required. The terminal part of the poles—on the opposite side of the gap—was threaded and laid through a flange to the covering; therefore, it was possible to change the gap by simply rotating the flange in respect to the poles.

Each photomultiplier tube was wrapped in a μ-metal shield, to protect it against stray fields; in any case, we performed preliminary tests in order to study the possible presence of parasitic effects induced by the magnetic field. So, an accurate comparison among the energy spectra for positrons annihilated in beryllium, recorded in the presence as well as in the absence of magnetic fields, allowed us to conclude that the centroid of the annihilation peak was stable—within the statistical fluctuations. Furthermore, by measuring the three quantum yield in beryllium in the presence of different fields we were able to con-

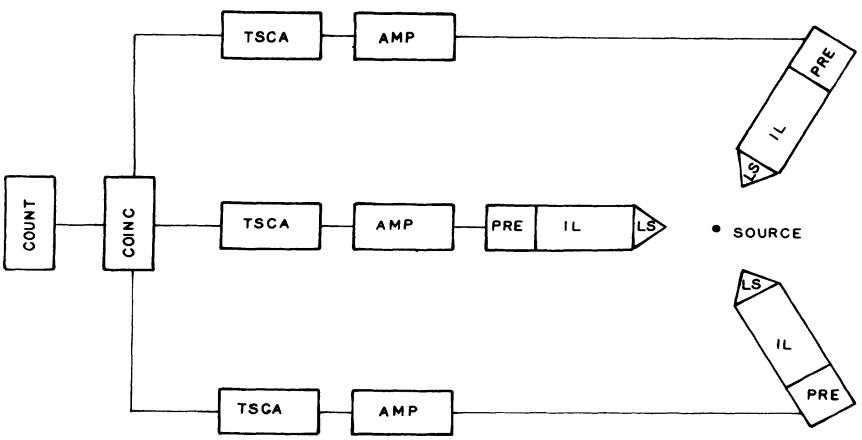

FIG. 1. Schematic block diagram of the experimental apparatus. The sample containing the source is inserted into the gap of a magnet (not shown for simplicity). LS: lead shield; IL: integral line; PRE: preamplifier; AMP: amplifier; TSCA: timing single channel analyzer; COINC: coincidence unit; and COUNT: counter and timer.

clude that it was identical, within the errors, to the same quantity, at zero field. Therefore, any influence of the field on the electronic apparatus could be excluded.

THEORY OF THE EXPERIMENT

It should be remembered that a positron-electron bound pair in condensed matter differs from positronium in vacuum, because of (i) the possibility for the positron to be annihilated by an external electron—"pickoff" annihilation—instead of its own electron (intrinsic annihilation) and (ii) polarization effects, induced by local electric fields, which tend to reduce the electron density at the positron.[25] For these reasons, we prefer to call quasipositronium (qPs) such a positron-electron pair, in order to distinguish it from vacuum positronium (Ps), according to Dupasquier.[26] The polarization effect is usually taken into account in a phenomenological way by introducing a relative contact density:[27]

$$\eta=|\Phi(0)|^2_{\mathrm{qPs}}/|\Phi(0)|^2_{\mathrm{Ps}} . \tag{1}$$

Since both the intrinsic decay rates Γ of singlet and triplet qPs ground-state sublevels, as well as the energy separation between them—the hyperfine splitting δW—are proportional to $|\Phi(0)|^2$, it immediately results:[28]

$$(\Gamma_{s,t})_{\mathrm{qPs}}=\eta(\Gamma_{s,t})_{\mathrm{Ps}},\quad (\delta W)_{\mathrm{qPs}}=\eta(\delta W)_{\mathrm{Ps}} , \tag{2}$$

where $(\Gamma_{s,t})_{\mathrm{Ps}}=8$ and $0.0072\ \mathrm{ns}^{-1}$ for singlet and triplet substates, respectively, and $\delta W=8.41\times10^{-4}$ eV. In conclusion, we can write for the ortho- and para-qPs decay rates in matter:[29]

$$\Gamma_3=\eta(\Gamma_t)_{\mathrm{Ps}}+\Gamma_p \quad (\text{ortho}) , \tag{3a}$$

$$\Gamma_0=\eta(\Gamma_s)_{\mathrm{Ps}}+\Gamma_p \quad (\text{para}) , \tag{3b}$$

where Γ_p is the pickoff annihilation rate. Due to the presence of the pickoff process, a qPs atom, initially formed in triplet state, can disappear through two-γ annihilation; we recall that the selection rules only allow annihilation into three photons for triplet Ps in vacuum. Therefore, the three photons annihilation rate for qPs is

$$\Gamma_3^{3\mathrm{ph}}=\eta(\Gamma_t)_{\mathrm{Ps}}+\Gamma_p/372 \quad (\text{ortho}) , \tag{4a}$$

$$\Gamma_0^{3\mathrm{ph}}=\Gamma_p/372 \quad (\text{para}) . \tag{4b}$$

The factor 372, which appears in the preceding equation results from the ratio between the intrinsic decay rates into three and two photons, averaged on the relative spin populations for ortho and para sublevels.[30]

By turning on a static, magnetic field, the $m=0$ sublevels become coupled; on the contrary, the $m=\pm1$ sublevels are unaffected by the field;[1] a report of the magnetic quenching theory has been given in.[31] Here we recall that the mixed $m=0$ substates ("perturbed" ortho and para sublevels) exhibit a quadratic Zeeman effect as far as their energy levels are concerned; their decay rates Γ'_3 and Γ'_0 are linear combinations of the unperturbed decay rates Γ_3 and Γ_0, with field dependent coefficients:

$$(\Gamma')_3=\frac{\Gamma_3+y^2\Gamma_0}{1+y^2} \quad (\text{perturbed ortho}) , \tag{5a}$$

$$(\Gamma')_0=\frac{\Gamma_0+y^2\Gamma_3}{1+y^2} \quad (\text{perturbed para}) , \tag{5b}$$

where $y=(\sqrt{1+x^2}-1)/x$, $x=4\mu_B H/\delta W$; μ_B is the Bohr magneton and H the applied field. It follows that, in such a case, the decay rate into three photons for the $m=0$ sublevels is

$$(\Gamma')_3^{3\mathrm{ph}}=\frac{\eta(\Gamma_t)_{\mathrm{Ps}}}{1+y^2}+\Gamma_p/372 \quad (\text{perturbed ortho}) \tag{6a}$$

$$(\Gamma')_0^{3\mathrm{ph}}=\frac{\eta(\Gamma_t)_{\mathrm{Ps}}y^2}{1+y^2}+\Gamma_p/372 \quad (\text{perturbed para}) \tag{6b}$$

which obviously reduce to Eq. (4) for vanishing field.

The contribution to the three quantum yield from the various sublevels can be accounted for as follows. If I is the fraction of positrons forming qPs, we have, at zero field

$$C_t(0)=\frac{3}{4}I\frac{\Gamma_3^{3\mathrm{ph}}}{\Gamma_3} \quad (\text{ortho state}) , \tag{7a}$$

$$C_s(0)=\frac{1}{4}I\frac{\Gamma_0^{3\mathrm{ph}}}{\Gamma_0} \quad (\text{para state}) . \tag{7b}$$

On the other hand, in the presence of a magnetic field

$$C_{t1}(H)=\frac{3}{8}I\frac{\Gamma_3^{3\mathrm{ph}}}{\Gamma_3} \quad (m=\pm1 \ \text{ortho states}) , \tag{8a}$$

$$C_{t0}(H)=\frac{3}{8}I\frac{(\Gamma')_3^{3\mathrm{ph}}}{(\Gamma')_3} \quad (m=0 \ \text{ortho state}) , \tag{8b}$$

$$C_s(H)=\frac{1}{4}I\frac{(\Gamma')_0^{3\mathrm{ph}}}{(\Gamma')_0} \quad (m=0 \ \text{para state}) . \tag{8c}$$

We note that the sum of (8a) and (8b) reduce, for vanishing field, to (7a), by virtue of Eqs. (5a) and (6a). In both cases the remaining fraction $(1-I)$ of positrons not forming qPs will give a contribution—which is unaffected by the field—amounting to $(1-I)/372$.

The numerical coefficients in Eqs. (8a) and (8b) take into account that the observations are made not over the entire sphere surrounding the source, but with the detectors symmetrically arranged in a plane perpendicular to the magnetic field. In fact, according to the Drisko calculation,[32] it is found that the perturbed ortho state gives the same contribution to the three quantum events as the sum of the other two triplet sublevels—by assuming our geometrical arrangement. Furthermore, we note that a contribution to the three quantum yield also comes, in principle, from the para sublevel; this is due to the pickoff process, both in the presence as well as in the absence of the magnetic field; a further contribution results from the magnetic mixing.

RESULTS AND DISCUSSION

We are now able to introduce a "quenching parameter" Q. It is tempting to define Q as the three quantum rate from qPs in the presence of the field, normalized to the same quantity at zero field; however, this last

TABLE I. Lifetime τ (in ns), intensity I (%), and relative contact density η of the positron bound system in the investigated media.

	Byphenil[a]	Mylar	Naphthalene	PPO[b]
τ	1.15±0.02	1.82±0.04[c]	1.03±0.01[d]	1.07±0.03
I	22.5±0.3	16.0±0.3[c]	17.3±0.3[d]	20.5±1.4
η	0.76±0.05	0.78±0.05[e]	0.82±0.07[e]	0.78±0.04

[a]Reference 23.
[b]Reference 24.
[c]Reference 13.
[d]Reference 14.
[e]Present work.

definition would be not immediately comparable with the results of the experiment, since our signal (the three photons annihilation rate from qPs) is measured together with the spurious events (background) as well as with the three quantum yield from free positrons. Furthermore, the background cannot be obtained in the classical way by rotating one of the counters by 45° out of the plane of the two others,[33] because of the presence of the magnet. In order to overcome these difficulties, we measure the following parameter, according to Telegdi *et al.*:[10]

$$Q_{\text{expt}}=\frac{P_m(H)-P_b}{P_m(0)-P_b} , \tag{9}$$

where $P_m(H)$ [$P_m(0)$] is the three quantum annihilation rate measured in the material with (without) the field; P_b is the same quantity in beryllium, where it is known that no qPs formation occurs. Correspondingly, we define the quenching parameter Q as follows [from Eqs. (7) and (8)]:

$$Q=\frac{C_{t1}(H)+C_{t0}(H)+C_s(H)-I/372}{C_t(0)+C_s(0)-I/372} . \tag{10}$$

Expression (9) is the experimental counterpart of Eq. (10), if we assume that the spurious events, measured in the presence of beryllium as well as of the material, are equal. This assumption can be indirectly subjected to an experimental test; in fact, we can measure the background outside the magnet separately for the metal and for the other samples. A preliminary, accurate comparison of the results allowed us to conclude that there is no meaningful difference in the background among the various media. This is probably due to the use of beryllium as the reference metal, whose density (1.85 g/cm^3) is comparable to that of the other investigated materials. In connection to this, it is useful to note that we verified that, by repeating the experiment with aluminum of the same thickness (200 mg/cm^2), a small increase of the background was observed.

From definition (10) it follows that Q depends on the lifetime of ortho-qPs, as well as on its relative contact density η. Therefore, a preliminary knowledge of these parameters featuring the behavior of qPs in the investigated materials is necessary. This can be achieved from lifetime spectroscopy and, for instance, triple coincidence measurements. The data concerning our work are collected in Table I.

The results of our measurements are displayed in Figs. 2–5 and in Table II, where the quenching parameter Q is shown as a function of the magnetic field. In each figure the continuous line refers to a quenching parameter calculated, according to Eq. (11), by assuming an "unperturbed" qPs, that is, with $\eta=1$. It is convenient to discuss the data for byphenil (Fig. 2) first of all; this material was chosen, since it is known from lifetime measurements that the magnetic quenching shows regular features.[23] If the data are fitted by the continuous line, we get the value 2.95 for the normalized χ^2, corresponding to $P(\chi^2>\chi^2_{\text{sp}})>99\%$. This means that the model of the unperturbed qPs is a bad approximation to our data. We

TABLE II. Magnetic fields in (teslas) and corresponding values of the quenching parameter Q in the investigated materials.

Material	Field	Q
Byphenil	0.21	1.000±0.023
	0.36	0.933±0.031
	0.43	0.958±0.028
	0.70	0.976±0.029
	0.95	0.838±0.032
	1.21	0.820±0.036
	1.33	0.741±0.035
Mylar	0.24	0.900±0.030
	0.32	0.888±0.034
	0.49	0.770±0.036
	0.72	0.773±0.040
	0.94	0.745±0.033
	1.08	0.750±0.031
	1.50	0.728±0.030
Naphthalene	0.21	0.922±0.046
	0.51	0.793±0.060
	0.60	0.838±0.058
	0.68	0.780±0.044
	0.85	0.583±0.060
	1.58	0.654±0.064
PPO	0.21	0.881±0.046
	0.50	0.824±0.051
	0.71	0.595±0.056
	0.93	0.560±0.045
	1.33	0.591±0.047
	1.60	0.471±0.056

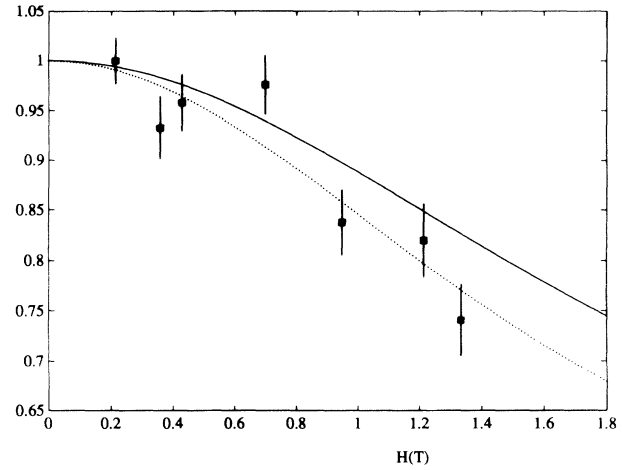

FIG. 2. Quenching parameter Q as a function of the applied magnetic field H (tesla) in byphenil. Continuous line: unperturbed qPs ($\eta=1$) and dotted line: qPs with $\eta=0.76$.

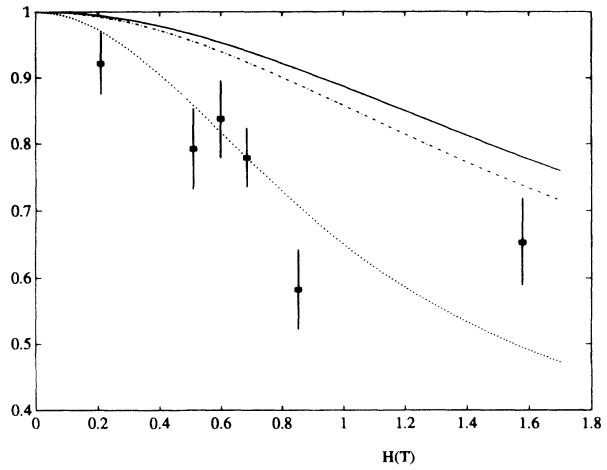

FIG. 4. Quenching parameter Q as a function of the applied magnetic field H (tesla) in naphthalene. Continuous line: unperturbed qPs ($\eta=1$); dash-dotted line: qPs with $\eta=0.82$; dotted line: qPs with $\eta=0.36$.

can improve the agreement by using for η the value 0.76, shown in Table I and obtained by other methods. In such a case $\chi^2=1.39$ and $P=80\%$. Of course, we can directly obtain the relative contact density from our measurements through a best fit procedure, by using Eq. (10) with η as a free parameter; we obtain $\eta=0.76\pm0.09$, which is in excellent agreement–within the errors–with the previous one. Therefore, this result confirms that the positron bound system in byphenil can be treated as a "relaxed" qPs. Furthermore, we can assert that the technique adopted here is reliable, since it leads to conclusions, which are consistent with those obtained by using the lifetime spectroscopy.

Let us now examine the other results. A glance at Figs. 3 to 5 allows us to realize that the model of the relaxed qPs cannot explain the data. Actually, in all cases the experimental values of Q systematically remain under the dash-dotted curve, which is drawn by assuming a relaxed qPs with the contact density given in Table I. The values of η were taken during a separate, triple coincidence experiment in the absence of magnetic fields, whose outline has been previously given.[23] One might think that the data could be fitted by using other values of the contact density. However, this is not the case: the "best" fits obtained by letting this parameter to be free supply $\chi^2=3.32$, 3.04, and 3.37 for Mylar (with $\eta=0.52$), PPO ($\eta=0.30$) and naphthalene ($\eta=0.36$), respectively. In any case, the corresponding P is always $>99\%$.

It is interesting to note the qualitative agreement between the present results and those obtained with lifetime spectroscopy: in fact, the data in Mylar (Fig. 3) turn out to settle along the dash-dotted curve for fields greater than 1.2 T; this behavior is similar to that shown in Ref. 12. On the other hand, the experimental Q values in na-

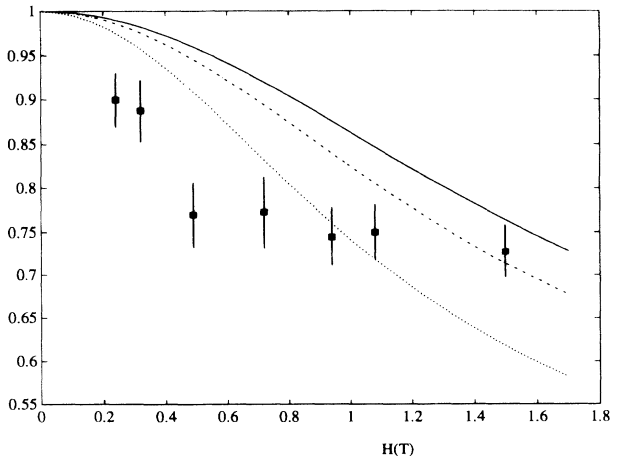

FIG. 3. Quenching parameter Q as a function of the applied magnetic field H (tesla) in Mylar. Continuous line: unperturbed qPs ($\eta=1$); dash-dotted line: qPs with $\eta=0.78$; and dotted line: qPs with $\eta=0.52$.

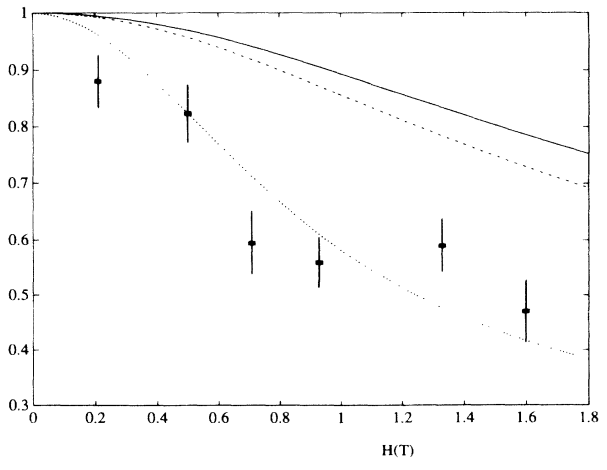

FIG. 5. Quenching parameter Q as a function of the applied magnetic field H (tesla) in 2,5-diphenyloxazole. Continuous line: unperturbed qPs ($\eta=1$); dash-dotted line: qPs with $\eta=0.78$; and dotted line: qPs with $\eta=0.30$.

phthalene (Fig. 4) and in PPO (Fig. 5) remain under the expected curve, even at the highest investigated fields; furthermore, the magnetic quenching in PPO is stronger than in naphthalene, and both these facts can be similarly noted by comparing the results of the previous investigations.[14,24]

Finally, we would like to point out a peculiarity of the technique presented here: since the perturbed orthostate has the same statistical weight as the sum of the two other, unperturbed, ortho sublevels [Eqs. (8a) and (8b)], the magnetic quenching effect is magnified with the triple coincidence technique in respect to lifetime spectroscopy. As a consequence, any physical effect that can be related to the magnetic quenching of qPs is expected to be "amplified" by measuring the three quantum yield.

CONCLUSIONS

In the present work we have carried out a magnetic quenching study in different molecular solids, some of which are known to show a stronger effect than could be expected on the basis of the Halpern theory. The technique used—the measurement of the three quantum annihilation rate in the presence of magnetic fields—gives results that confirm the data already obtained with lifetime spectroscopy, which pointed out the BR effect. We expressly remark that these results are considered "anomalous" only within the framework of the model of relaxed qPs, that is, the Halpern theory modified to take into account interactions of the positron-electron pair with the surroundings.

In those materials where the Halpern model is valid (byphenil, in our work), our technique is able to supply values of the relative contact density, which are perfectly comparable with those obtained from lifetime spectroscopy: in such a way, another independent determination of the contact density allows us a better knowledge of this important parameter.

On the other hand, we need to have more than an experimental method when we are faced with results requiring to be fitted with a multiparametrical model—as is the case with some explanations of the BR effect. In these cases, the triple coincidence technique, coupled with magnetic fields, is a reliable tool for investigating the features of positron bound systems in condensed matter.

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