

Dyson line and modified Dyson line in the EPR measurements

Volodymyr Popovych, Mariusz Bester, Ireneusz Stefaniuk, Marian Kuzma

Abstract. The difficulty in determining the electron paramagnetic resonance (EPR) line parameters of ferromagnetic semiconductors has been addressed. For these materials, the resonance line is very broad and lies at low resonance field, so that only a part of the line can be detected experimentally. Moreover, the line is of asymmetric (Dysonian) shape as described by the line shape parameter α . We have compared values of line parameters derived by computer fitting of the whole experimental EPR line to the Dyson function (or modified Dyson function) with the values obtained by applying this procedure to the left and the right half of the line.

Key words: EPR • FMR • Dyson line • CdCrTe

V. Popovych Drogobych State Pedagogical University,

24 Ivan Franko Str., 82100 Drogobych, Ukraine

M. Bester, I. Stefaniuk, M. Kuzma[™] Faculty of Mathematics and Natural Sciences, University of Rzeszow, 1 Pigonia Str., 35-959 Rzeszow, Poland, Tel.: +48 17 851 8605, +48 17 851 8705, Fax: +48 17 851 8524, E-mail: kuzma@ur.edu.pl

Received: 11 September 2014 Accepted: 27 March 2015

Introduction

In many experiments on EPR, the signal recorded is broad and asymmetric in relation to the Gaussian or Lorentzian distribution. It was a long time ago (in 1955) that the asymmetry of EPR signal was described theoretically by Dyson [1] for EPR experiments on free electrons in metals. Dyson studied the shape of the EPR line depending on the diffusion time of charge carriers, skin depth, and relaxation time. Several cases were investigated for various mutual relations of the values of these parameters. A clear and comprehensive view of these results is presented in this study [2].

Asymmetry of ferromagnetic resonance (FMR) or EPR resonance lines is attributed to the samples with dimensions larger than skin depth, which cause non-uniform distribution of the microwave field in the sample [2, 3]. Therefore, conducting materials, such as metals or low-resistivity semiconductors, exhibit asymmetrical line shape. The second reason of asymmetry is a great width of the line and small resonance field. The aim of this study is to show the modification of the wide resonance line shape in the region of very low resonance field, based on so-called modified Dyson line shape introduced by Joshi [4]. In particular, the case where the resonance line is not fully visible in the measured spectra has been studied and illustrated by experimental results of FMR in CdTe:Cr semiconductor (note that this type of semiconductor was predicted to be one of the most promising materials for spintronic applications [5]). We show that four parameters describing modified Dyson line can be delivered with good accuracy from experimental right half of the resonance line recorded.

Lorentz, Dyson, and modified Dyson line

Parameters describing Dyson line and its derivative are the same as for the Lorentz line: ΔB , B_0 , I, and additional A/B parameter, which is the ratio of amplitude of the left peak A to the right peak B in the derivative of resonance: $A/B = I_{\rm L}/I_{\rm R}$. The latter is equivalent to the asymmetry parameter α , which is the ratio of dispersion and absorption. Dyson line parameters can be determined either by the graphical method of direct measurement of the spectrum or by computer fitting of experimental data by Dyson function with four parameters: A, α , ΔH , and H_0 . One can also choose fitting the experimental spectrum to the polynomial:

(1)
$$F(x) = \frac{(1 - Cx - x)}{(1 + x^2)^2}$$

where $x = (B - Br)/\Delta B$. The procedure for the designation of parameters in such a way is described in the work of Peter [6] and Burgard [7].

For broad EPR line, the contribution of the component of microwave field, which rotates in the opposite direction to the Larmor precession, must be taken into account for description of the line shape. In fact, one can notice that for a circularly polarized microwave field, where the factor g is anisotropic and contains components of positive and negative sign, the EPR signal appears on both sides of the field B = 0. This phenomenon is described in the book of Bleaney and Abragam [8]. The impact of that effect on the shape of the broad EPR line was given attention in the work of Ivanshin [9] and was expressed by equations for Lorentz and Dyson broad lines [4]:

$$P_{\rm L} \propto \left[\frac{\Delta B}{4(B-B_0)^2 + \Delta B^2} + \frac{\Delta B}{4(B+B_0)^2 + \Delta B^2} \right]$$
(2)
$$P_{\rm D} \propto \left[\frac{\Delta B + \alpha (B-B_0)}{4(B-B_0)^2 + \Delta B^2} + \frac{\Delta B + \alpha (B+B_0)}{4(B+B_0)^2 + \Delta B^2} \right]$$

In Eq. (2) there is an asymmetry against the field B = 0 (Fig. 1a), although from a physical point of view, this asymmetry should not exist, because the reversal field B in the spectrometer should not change properties of the investigated system. The effect is visible only for broad lines (see Fig. 1a).

Asymmetry can be explained based on the absorption and dispersion components of the susceptibility of the microwave electromagnetic field. Joshi [4] noticed that the absorption is symmetric with respect to the magnetic field, while the dispersion

B0=200mT, ΔB=250mT, α=0.5 b) dP/dB, arb. u. -600 -400 -200 Ò 200 $4\dot{0}0$ 600

Fig. 1. Asymmetry with respect to B = 0 in the shape of broad Dyson EPR lines (a), modified Dyson line (b).

B, mT

is antisymmetric. Composition of both components results in asymmetry observed in Fig. 1. In order to remove this asymmetry, Joshi [4] proposed to modify the Dyson function to the following form:

(3)
$$P_{\rm DM} \propto \left[\frac{\Delta B + \alpha (B - B_0)}{4 (B - B_0)^2 + \Delta B^2} + \frac{\Delta B - \alpha (B + B_0)}{4 (B + B_0)^2 + \Delta B^2} \right]$$

The graphical presentation of the modified Dysonian line is shown in Fig. 1b.

When the position of the resonance line in EPR or FMR measurements is in the vicinity of B = 0(B > 0), the observed derivative of the line is deformed by superimposing the line from B < 0 range. This effect is significant for the left part of the derivative.

Experimental

The sample for EPR measurements was cut from polycrystalline $Cd_{1-x}Cr_x$ Te ingot prepared by alloying of CdTe and Cr₂Te₃ powder components with nominal Cr concentration in the initial charge x =5%. The synthesis was performed at a temperature as high as 1320°C [10] in order to ensure complete melting of chromium telluride alloys, of which the highest melting point is 1283°C, according to the Cr-Te phase diagram [11]. Concentration of Cr in the ingot obtained was determined by XRF method and was found to corresponds to x = 0.02 composition. EPR studies were carried out on X-band spectrometer (9.4 GHz) at room and liquid nitrogen temperatures.



a)



Fig. 2. EPR spectrum for $Cd_{1-x}Cr_xTe$ at 300 K (a) and 77 K (b); spectra generated from Fig. 1a (300 K) using parameters obtained from the fitting of the left and the right half (c), (d), respectively.

Results and discussion

The spectrum of $Cd_{1-x}Cr_x$ Te sample at 300 K consists of one broad line (Fig. 2a) as well as at 77 K (Fig. 2b). The spectral line at T = 77 K is strongly shifted to the low field region. The g factor for this line is approximately equal to g = 3.47 at T = 300 K and g = 4.57at T = 77 K. Such great values of the g factor and its strong dependence on temperature may indicate that we are dealing with FMR instead of EPR.

The shape of the lines was adjusted to the Lorentz function, but good agreement with this function is observed only at room temperature (Fig. 2a).

We suppose two possible origins of ferromagnetic behavior of the sample: formation of $Cd_{1-x}Cr_xTe$ solid solution with exchange sp-d interaction in consequence of chromium dissolution in CdTe or formation of CrTe-related second phase precipitates in CdTe matrix. Both phases are thought to be ferromagnetic [12, 13]. It is known that for magnetic semiconductors, the magnetic resonance lines are usually broad [14].

The EPR line at 77 K is shifted towards low fields as a result of large increase of the internal magnetic field [10]. The shift is so considerable that only the right half of the signal derivative is fully visible, as is shown in Fig. 2b. Therefore, one has to determine its parameters solely from that part of the curve by computational matching of the analytical function to the experimental data. As a result, parameters derived from the part of the curve deviate from those derived from the whole curve, introducing an error. In order to determine the magnitude of the discrepancy, we performed the numerical analysis of the EPR spectra presented in Fig. 2a (T = 300 K) using standard procedure with the Origin software. The analysis consists of three stages.

At the first stage, the experimental EPR spectrum presented in Fig. 2a was fitted with Eq. (3). A very good agreement of the experimental curve with the modified Dyson function is observed in Fig. 2a. From this fitting, the parameters ΔB , B_0 and α were inferred (Table 1). During the second and the third stages, the parameters were calculated by the same method, but from the left or the right half of the derivative, respectively. These parameters differ from those obtained from the whole spectrum by less than 10% (see Table 1), except the parameter α , which weakly influences the shape of both the right and left part of the line. The discrepancy is considerably small in the determination of the resonance field B_0 . Modified Dyson lines were generated for parameters determined in the second and the third stage and were compared with the experimental EPR line (Figs. 2c and 2d).

Spectrum in Fig. 3 is the result of fitting the experimental data presented in Fig. 2b (T = 77 K) by modified Dyson formulae using parameters col-

Table 1. Line parameters derived from fitting of the modified Dyson function to the 'whole' EPR line and its parts (the left and the right half)

| Parameters | Dyson line | | | Modified Dyson line | | |
|-----------------|--------------|-----------|------------|---------------------|-----------|------------|
| | 'Whole' line | Left half | Right half | 'Whole' line | Left half | Right half |
| α | 0.63 | 0.50 | 0.68 | 1 | 1 | 1 |
| $B_0 [mT]$ | 195.15 | 196.29 | 188.29 | 197.94 | 195.96 | 188.53 |
| $\Delta B [mT]$ | 62.12 | 54.95 | 73.30 | 121.12 | 104.82 | 160.62 |



Fig. 3. Fitting experimental data from Fig. 2b (77 K) using modified Dyson function with parameters collected in Table 2.

Table 2. Parameters of fitting of spectrum from Fig. 2b(77 K) to modified Dyson formulae

| $\Delta B [mT]$ | $B_0 [mT]$ | A/B | α |
|-------------------|-------------------|------|------------------|
| 335.19 ± 5.48 | 147.11 ± 1.74 | 1.03 | 0.56 ± 0.043 |

lected in Table 2. A very good accuracy of adjustment (better than 2%) was obtained. However, we must remember that the parameters were calculated only from part (right) of the line. In such case, the error is determined on 10% as in Table 1.

Conclusions

In this study, we have considered the accuracy of the determination of parameters of asymmetric EPR line in the case when only a part of the line can be recorded due to the great linewidth and low resonance field. EPR spectrum of $Cd_{1-x}Cr_xTe$ sample was analyzed for illustration. Although the shape of the line is not reproduced exactly by the parameters obtained by computer fitting of the experimental EPR line to the modified Dyson function, the linewidth and resonance field are determined with accuracy better than 10%.

Acknowledgments. The work was partially supported by the grant no. N N507 264740 of the Polish Committee for Scientific Research.

References

- 1. Dyson, F. J. (1955). Electron spin resonance absorption in metals. II. Theory of electron diffusion and the skin effect. *Phys. Rev.*, 98(2), 349–359.
- Feher, G., & Kip, A. F. (1955). Electron spin resonance absorption in metals. I. Experimental. *Phys. Rev.*, 98(2), 337–351. http://dx.doi.org/10.1103/PhysRev.98.337.
- Webb, R. H. (1964). Electron-spin-resonance line shape in spherical metal particles. *Phys. Rev.*, 158(2), 225–233. http://dx.doi.org/10.1103/Phys-Rev.158.225.
- Joshi, J. P., & Bhat, S. V. (2004). On the analysis of broad Dysonian electron paramagnetic resonance spectra. J. Magn. Reson., 168(2), 284–287. DOI: 10.1016/j.jmr.2004.03.018.
- Blinowski, J. P., Kacman, J. A., & Majewski, J. (1996). Ferromagnetism in Cr-based diluted magnetic semiconductions. J. Cryst. Growth, 159(1/4), 972–975.
- Peter, M., Shaltiel, D., Wernick, J. H., Williams, H. J., Mock, J. B., & Sherwood, R. C. (1962). Paramagnetic resonance of S-state ions in metals. *Phys. Rev.*, 126(4), 1395–1402. http://dx.doi.org/10.1103/ PhysRev.126.1395.
- Burgardt, P., & Seehra, M. S. (1977). Electron paramagnetic resonance in gadolinium near T_c. *Phys. Rev. B*, *16*(5), 1802–1807. http://dx.doi.org/10.1103/ PhysRevB.16.1802.
- 8. Bleaney, B., & Abragam, A. (1986). *Electron paramagnetic resonance of transition ions*. New York: Dover Publications.
- Ivanshin, V. A., Deisenhofer, J., Krug von Nidda, H. A., Loidl, A., Mukhin, A. A., Balbashov, A. M., & Eremin, M. V. (2000). ESR study in lightly doped La_{1-x}Sr_xMnO₃. *Phys. Rev. B*, 61(9), 6213–6219. DOI: 10.1103/PhysRevB.61.6213.
- Stefaniuk, I., Bester, M., & Kuzma, M. (2008). Ferromagnetic resonance in CdCrTe solid solution. *J. Phys.-Conf. Ser.*, *104*, 012010. DOI: 10.1088/1742-6596/104/1/012010.
- Sreenivasan, M. G., Bi, J. F., Teo, K. L., Liew, T. J. (2008). Systematic investigation of structural and magnetic properties in molecular beam epitaxial growth of metastable zinc-blende CrTe towards halfmetallicity. J. Appl. Phys., 103, 043908.
- Kuzma, M., Stefaniuk, I., & Bester, M. (2010). Theoretical models and EPR study of Cr based diluted magnetic semiconductors. J. Phys.-Conf. Ser., 213, 012035. DOI: 10.1088/1742-6596/213/1/012035.
- Akram, M., & Nazar, F. M. (1983). Magnetic properties of CrTe, Cr₂₃Te₂₄, Cr₇Te₈, Cr₅Te₆, and Cr₃Te₄ compounds. *J. Mater. Sci.*, 18, 423. DOI: 10.1007/BF00560631.
- Langreth, D. C., & Wilkins, J. W. (1972). Theory of spin resonance in dilute magnetic alloys. *Phys. Rev. B*, 6, 3189. http://dx.doi.org/10.1103/PhysRevB.6.3189.