Enhanced Sensitivity of Electron Spin Resonance using Absorption-Free Measurement

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Introduction

Absorption-free measurement (AFM) (called interaction-free measurement by Elitzur and Vaidman [1] the discoverers of the phenomenon) is a counterintuitive consequence of quantum mechanics which allows the detection of a classical or quantum object without the exchange of any energy or momentum with the object. Kwiat et al [2] have noted that AFM could be used to enhance spectroscopic measurements. This paper is concerned with the application of AFM to electron spin resonance (ESR) spectroscopy.

The sensitivity of ESR is proportional to the amplitude of the magnetic field of the microwave radiation in the sample which is positioned in a resonant microwave cavity [3]. The improvement of the sensitivity that can be achieved by increasing the microwave power is limited because the sensitivity is proportional also to the difference in populations of the paramagnetic levels producing the ESR spectrum and the difference in populations of the levels is reduced by the absorption of microwave power at resonance. The effect is negligible so long as the rate of absorption is much less than the rate at which the spins can release the absorbed microwave energy to the environment (which normally acts as a perfect sink for the absorbed microwave energy). At high enough microwave power the criterion is not satisfied, the difference in populations of the spin levels is reduced and the sample is said to saturate. The rate of release of the absorbed energy to the environment is inversely proportional to the spin-lattice relaxation time. The onset of saturation occurs at quite low microwave powers in samples with long spin-lattice relaxation times, which is quite common when measurements need to be carried out at low temperatures, and the consequent loss of sensitivity is a problem.

If the ESR can be detected by AFM then it appears that the sensitivity of ESR could be increased by using higher microwave powers than would be possible by conventional means because at least a proportion of the measurement takes place without the absorption of energy leading to less saturation for the same signal strength.

Absorption-free measurement (AFM)

A version of AFM [4] which is suitable for modification for ESR is shown in Fig. 1. The path lengths are arranged so that when the object is absent (Fig. 1(a)), the beams *b* and *c* reflected by the mirrors interfere destructively on path *d* and all the radiation leaves the interferometer by the entrance path *a*. If an absorbing object blocks path *c* (Fig. 1(b)), no interference takes place and the reflected beam on path *b* is partially reflected onto path *d*. Therefore the detection of radiation on path *d* reveals the presence of an object on path *c*. If a single photon enters on path *a* and is detected on path *d*, one can be certain that path *c* is blocked even though no energy or momentum is exchanged with the blocking object. In some repetitions of the experiment as described, an incoming photon will follow path *c* and will exchange energy or momentum with the object but refinements of the method [5] allow the detection of objects with probability one and with vanishing probability of energy or momentum exchange with the object no matter how many times the experiment is repeated.

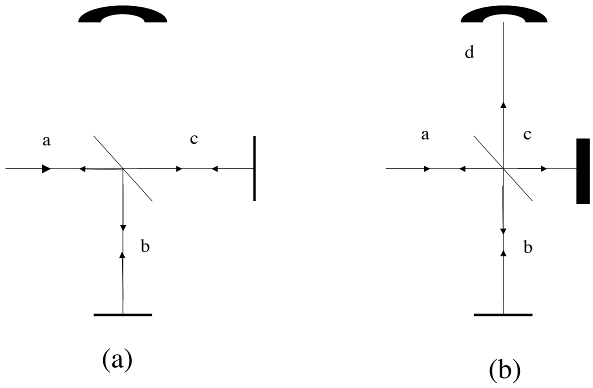

Figure 1: (a) Beam enters and exits on path *a* due to interference of reflected beams *b* and *c*. (b) Absorber on path *c* can lead to detection on path *d* of radiation that did not strike the absorber.

Electron spin resonance (ESR) and absorption-free measurement (AFM)

Understandably, the emphasis in the AFM literature has been on the paradoxical possibility of detecting an object without the exchange of energy or momentum with it. In applying the method to ESR, the emphasis is slightly different. To understand why, it is necessary to review the basic operation of an ESR spectrometer. It is worth noting that AFM is not exclusively a quantum phenomenon and a classical treatment which is what we will be using here is quite capable of dealing with AFM [6].

A typical set-up for ESR is shown in Fig. 2. The microwave power from the klystron is divided between arm *b* where the power is entirely absorbed in the matched load and arm *c* where it is almost entirely absorbed in the resonant cavity except for a slight mismatch so that some power is reflected. The reflected power divides at the beamsplitter (magic-T) and provides some power to the detector in arm *d* so that the detector operates in an optimum way. The ESR spectrum arises by scanning a magnetic field through the sample in the resonant cavity which changes the energy separation of the paramagnetic levels of interest. When the energy separation matches the microwave energy the sample absorbs microwave energy, changing slightly the *Q* of the cavity and resulting in a change in the microwave power reflected to the detector.

What is shown as a conventional beam-splitter in the centre of Fig. 2 is implemented in the case of the microwaves used in ESR by a magic-T hybrid junction which has the same transformation matrix as a symmetrical beam-splitter [7]. Note that the fourth arm of the magic-T, labelled *b* in Fig. 2, plays no active role in a conventional ESR spectrometer.

To implement the idea of AFM in an ESR spectrometer it is only necessary to remove the matched load from arm *b* so that it reflects power back to the magic-T. We then have the implementation of AFM envisaged in Fig. 1(b) except that the "object" to be measured, which is the ESR sample in the cavity, is not going to block the path *c* but is going to change the reflectivity of the "mirror" or absorber at the end of path *c*. This is reminiscent of the implementation of the original Elitzur and Vaidman proposal suggested by Penrose [8] where the object becomes one of the mirrors in a Mach-Zhender interferometer. A further difference in the present case is that the absorption in the ESR sample may *increase* the reflectivity of the cavity. That is because in ESR spectroscopy the cavity is deliberately slightly mismatched to the waveguide to provide "leakage" to the detector and the change in sample absorption at resonance may drive the cavity closer to a perfect match with the waveguide, leading to an increase in the reflection of microwave power. In that case, the presence of the "object" increases the nett number of detection events in the detector(s), rather than reducing them in as in the case of previous AFM implementations.

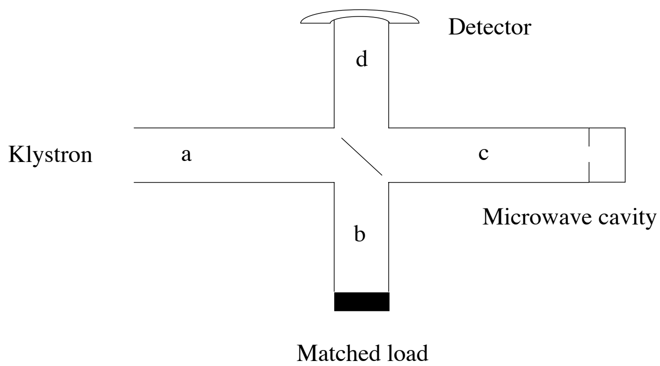

Figure 2: Conventional ESR spectrometer in schematic form.

Results

We will deal with a beam splitter with an amplitude reflection coefficient *r* and amplitude transmission coefficient *t* for the sake of generality. The results show that the optimum values are $|r| = |t| = 1\sqrt{2}$ which are the values for the magic-T. The amplitude of incoming waves (i.e. before reflection in arms *b* and *c*) in the arms *a*, *b*, *c* is represented by $A(\omega)$, $B(\omega)$, $C(\omega)$ respectively and the amplitude of outgoing waves (waves after reflection) is represented by $\overline{A}(\omega)$, $\overline{B}(\omega)$, $\overline{C}(\omega)$ and $\overline{D}(\omega)$. The power reflected by the cavity in arm *c* when the ESR sample is not absorbing microwave energy (applied magnetic field not producing paramagnetic energy levels satisfying the resonance condition with the microwaves) is $|\gamma|^2$ and the power reflected by arm *b* is $|\beta|^2$. The normal ESR case can be recovered by setting $|\beta| = 0$.

The evolution of the input amplitude a is

$$\begin{aligned} A(\omega) &\rightarrow rB(\omega)+tC(\omega) \\ &\rightarrow r(\beta\overline{B}(\omega)+\sqrt{1-|\beta|^2}B(\text{abs}))+t(\gamma\overline{C}(\omega)+\sqrt{1-|\gamma|^2}C(\text{abs})) \\ &\rightarrow r(\beta(r^*\overline{A}(\omega)-t\overline{D}(\omega))+\sqrt{1-|\beta|^2}B(\text{abs}))+t(\gamma(t^*\overline{A}+r\overline{D}(\omega))+\sqrt{1-|\gamma|^2}C(\text{abs})) \\ &= (\beta|r|^2+\gamma|t|^2)\overline{A}(\omega))+rt(\gamma-\beta)\overline{D}(\omega))+r\sqrt{1-|\beta|^2}B(\text{abs})+t\sqrt{1-|\gamma|^2}C(\text{abs}). \end{aligned}$$

The signal from the detector is proportional to the microwave power reaching the detector which in turn is proportional to $|r|^2|t|^2|\gamma-\beta|^2$. In the conventional ESR spectrometer the signal is due to the change $\Delta\gamma$ in the reflected amplitude due to the further mismatch between the resonant cavity and the waveguide due to the absorption of power by the electron spins when the magnetic field is scanned to the value allowing the paramagnetic transitions to take place. Thus the conventional ESR signal ($|\beta|=0$) is proportional to $|r|^2|t|^2|\gamma+\Delta\gamma|^2-|r|^2|t|^2|\gamma|^2 \sim 2|r|^2|t|^2|\gamma\Delta\gamma|$ since $|\Delta\gamma)| << 1$.

The ESR signal can be significantly enhanced by taking advantage of AFM. This can be done by allowing arm d to reflect power by not terminating arm b with a matched load so that $\beta > 0$. The detector signal due to the ESR resonance is then proportional to $|r|^2|t|^2(|\gamma+\Delta\gamma-\beta|^2-|\gamma-\beta|^2) \sim 2|r|^2|t|^2(|\beta|-|\gamma|)|\Delta\gamma|$. The factor $|\gamma|$ must be kept small because as $|\gamma|$ is increased, the microwave power incident on the sample in the microwave cavity is reduced. There is no restriction on the value of β and hence the detector signal can be enhanced considerably over the conventional case by choosing $|\beta| >> |\gamma|$. The larger value of $|\beta|$ also ensures that there is enough power reflected to the detector to bias it correctly thus avoiding a common problem of insufficient bias power in conventional ESR when low powers are used to avoid saturating a sample.

Conclusion

It is suggested here that it may be possible to enhance the ESR signal from a sample for which the sensitivity is otherwise limited because of a long spin-lattice relaxation time leading to the early onset of saturation. The ESR signal enhancement involves $|\beta|$ which is proportional to the amplitude of the microwaves in arm b none of which reaches with the cavity containing the paramagnetic sample in arm c. That is a manifestation of the AFM basis of the method. It would be interesting to confirm the present suggestion experimentally.

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