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Photoinduced spin polarization and microwave technology

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ABSTRACT

We report here on studies of optically pumped active microwave media based on various fullerene derivatives, with an emphasis on the use of these materials in microwave electronics. We have investigated a class of optically excited paramagnetic materials that demonstrate activity in the X-band as candidate materials. We found that a particular fullerene derivative, Phenyl-C_{61}-butyric acid methyl ester (PCBM), produced the largest electron paramagnetic resonance (EPR) emission signal compared to other organic compounds that have been suggested for use as microwave active materials. We also studied the effects of concentration, temperature, solvent etc. on the activity of the material. In these experiments, EPR studies using a commercial spectrometer were followed up by measurements of an RF signal reflected from a resonator loaded with the PCBM-based material. The activity was directly demonstrated through the change in the quality factor and RF coupling between the resonator and waveguide feed.

At the inception of these experiments the primary interest was the development of a microwave PASER. The PASER (particle acceleration by stimulated emission of radiation [1]) is a novel acceleration concept that is based on the direct energy transfer from an active medium to a charged particle beam. While the previous work on the PASER has emphasized operations at infrared or visible wavelengths, operating in the microwave regime has significant advantages in terms of the less stringent quality requirements placed on the electron beam provided an appropriate microwave active medium can be found. This paper is focused on our investigation of the possibility of a PASER operating in the microwave frequency regime [2] using active paramagnetic materials. While a high level of gain for PCBM was demonstrated compared to other candidate materials, dielectric losses and quenching effects were found to negatively impact its performance for PASER applications. We present results on development and bench testing for these new fullerene-based materials along with some conceptual designs for microwave PASERs. Other possible applications for active paramagnetic materials are suggested including low noise microwave amplifiers and tunable RF absorbers.

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1. Introduction

Recent theoretical work has shown that an electron beam can absorb energy from an active medium (one in which a population inversion exists) and be accelerated [3]. The active medium transfers the energy stored in it to the kinetic energy of an electron beam. This technique has the potential to be the basis of a new method of acceleration. The effect is similar to the action of a laser or maser with the stimulated emission of radiation being produced by the virtual photons in the electromagnetic field of the beam.

In the 1930s Latyschev and Leipunsky [4] performed an experiment that showed this effect using single electrons. Electrons were injected into mercury vapor that was illuminated by a mercury lamp. The electrons were found to gain energy on the order of few eV from the excited mercury atoms, through an inverse process to the Frank–Hertz [5] effect. When a bound electron drops from the excited state to the ground state it emits a photon which has the probability to be absorbed by a free electron. Free electrons will gain energy from an active medium. If an electron beam is modulated with the frequency that corresponds to the transition energy between the excited state and the ground state, a stimulated process occurs, as in lasing action [6]. A proof of principle experiment has been performed recently that produced results consistent with the PASER effect at infrared frequencies [1]. The active medium in the experiment was a CO_2 gas mixture.

Given the relative difficulty of studying the PASER effect in the infrared regime, we developed another approach [2] that employs a solid state active microwave medium to accelerate charged particles. In this new scheme the microwave active medium is pumped with a

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flash lamp, laser, or low energy electron beam. This method of acceleration effectively replaces the microwave power source used in conventional rf-based accelerators with a less expensive powerful light source. The success of the technique relies on the development of new types of microwave active media. A basic study of microwave active medium development and testing is presented in this paper.

From an electromagnetic standpoint, an active medium exhibits within some frequency range a negative imaginary part of its permittivity or permeability and hence an effective negative resistivity. Thus an electromagnetic wave with a frequency in this band or a charged particle beam with a Fourier frequency component of its current in this range will gain energy from the medium. For the case of a plane electromagnetic wave, the dispersion relation is $k=\sqrt{\varepsilon\mu}(\omega/c)$. Letting $\varepsilon=\varepsilon_r+i\varepsilon_i$ and $\mu=\mu_r+i\mu_i$, we obtain $k=(\omega/c)\left(\hat{k}_r+i\hat{k}_i\right)$, ($\varepsilon_r$, ε_i, μ_r, μ_i, $\hat{k}_r$, and $\hat{k}_i$ real) with

$$\hat{k}_i=\frac{(\mu_i\varepsilon_r+\mu_r\varepsilon_i)}{2\hat{k}_r}$$

and

$$\hat{k}_r=\frac{1}{\sqrt{2}}\sqrt{(\mu_r\varepsilon_r-\mu_i\varepsilon_i)+\sqrt{(\mu_r\varepsilon_r-\mu_i\varepsilon_i)^2+(\mu_i\varepsilon_r+\mu_r\varepsilon_i)^2}}$$

The amplitude of the wave is proportional to$\exp[-(\omega/c)\hat{k}_i z]$ (where $+z$ is the propagation direction); thus, depending on the sign of$\hat{k}_i$, the wave is either damped or amplified as it propagates. In particular we note that for an active paramagnetic medium, too large a positive value of ε_i can overwhelm the amplification from a negative μ_i. For an active medium inside a waveguide, the dispersion relation is modified and as will be discussed below, appropriate design of the waveguide can lead to amplification even in the case of large losses in the medium.

A number of possible configurations can be considered for a microwave PASER, provided a suitable active medium can be found. A resonator loaded with the medium and possessing a vacuum channel for the passage of the beam is the simplest possibility; such an accelerating technique is similar to a traditional RF-fed iris-loaded structure. One could also consider a two beam PASER accelerating scheme. In this case a high intensity (but low energy) drive beam produces a wakefield that pumps the active medium. A trailing witness beam gets accelerated to high energy by absorbing the energy stored by the drive beam in the active medium. Other possibilities for pumping include coherent or incoherent light, or direct absorption of a low energy electron beam in the active medium [7].

Realization of a microwave PASER will require a modulated beam or a bunch train with frequency content $\sim$10 GHz. Several methods of bunch train generation have been studied at the Argonne Wakefield Accelerator facility (AWA). One approach relies on temporal laser pulse shaping for a photocathode gun [8]. Another method makes use of a transverse-to-longitudinal emittance exchange beamline [9].

In this paper we present results of investigations into active microwave media based on paramagnetic media. This development process led to the identification of (6,6)-phenyl-C_{61}-butyric acid methyl ester (PCBM), a soluble derivative of fullerene (C_{60}), as a major candidate for future active medium applications. This part of the experimental work was done using EPR (electron paramagnetic resonance) spectroscopy measurements of various media as functions of temperature, concentration and other parameters. The EPR activity of the materials under test was produced by pumping with a 532 nm laser pulse.

This concept could find potential uses in microwave and accelerator applications in a frequency region from hundreds of MHz to up to 100 GHz. The frequency of the electron spin resonance is controlled by an applied magnetic field. Easily achieved kG-level magnetic fields correspond to X band or $\sim$10 GHz frequencies, while superconducting magnet Tesla-level fields yield frequencies in the 100–600 GHz range [10].

The measurement of activity using the EPR technique is very sensitive but indirect: it detects a small relative change in the RF coupling to a high-Q resonator. It does not directly provide the imaginary part of the permeability. We performed bench tests to directly observe RF emission and absorption in the form of changes in quality factor and coupling due to Zeeman splitting of energy levels. We also quantified the population inversion achieved in the PCBM sample in terms of the magnetic permeability by comparison with a calibration material of known spin density. These measurements are similar to studies of solid state MASERs (microwave amplification by stimulated emission of radiation). We plan to demonstrate the performance of an RF amplifier based on PCBM in a follow up experiment.

Traditional EPR-based solid-state masers require cryogenic temperatures [11]. This requirement is mostly due to the fast decrease of the triplet lifetime with rising temperature. A short lifetime yields low amplification and a short RF pulse. Levanon and Blank [12,13] demonstrated the possibility of extending the lifetime of the triplet state to the microsecond level at room temperature using fullerene and porphyrin solutions in organic solvents (liquid crystals). We used these results as a starting point and the design evolved to a fullerene derivative, PCBM, dissolved in polystyrene, which exhibited emission signals significantly larger than C_{60}.

2. Activity in three level paramagnetic systems

Three-level solid-state maser amplifiers based upon the paramagnetic properties of the unpaired electron exploit changes in the electron spin population of the magnetic Zeeman levels, thus allowing amplification of electromagnetic radiation in the bulk material. To achieve amplification in a paramagnetic based maser, an inverted spin population must be achieved, corresponding to a negative spin temperature.

The generic term for the process that produces the activity in the media is chemically induced dynamic electron spin polarization (CIDEP) [14]. CIDEP is formed in reactions of radicals resulting from optical pumping that in turn lead to the formation of free radicals or triplet state molecules. We note that the optical pulse serves in this case both to generate the EPR active excited state and to create the population inversion.

The details of the mechanism are complex, but for our purposes an adequate picture is given by the schematic energy level diagram shown in Fig. 1. Absorption of the laser pulse causes molecules to transition from the ground singlet state, S_0, to the first excited singlet state, S_1. Following a fast intersystem crossing the excitation transfers from S_1 to the lowest lying triplet T_0. In the triplet state the spin levels are split into three sublevels by the applied magnetic field. As a rule the rate constants of the transitions from the S_1 state to the T_X, T_Y, T_Z states are different resulting in electron polarization due to the triplet mechanism. In the case of a long triplet molecule lifetime the polarization of this triplet molecule can be observed by EPR.

3. Electron paramagnetic resonance measurements of active media

We tested various compounds known to form EPR-detectable triplet states under optical excitation: C_{60} (fullerene), TPP (Tetraphenylporphyrin), Zn-TPP, PCBM (Phenyl-C61-butyric acid methyl ester) and some others. A number of solvents were also used: toluene, polystyrene, MBBA (N-(4-Methoxybenzylidene)-4-butylaniline), EBBA (N-(4-ethoxybenzylidene)-4′-n-butylaniline),

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