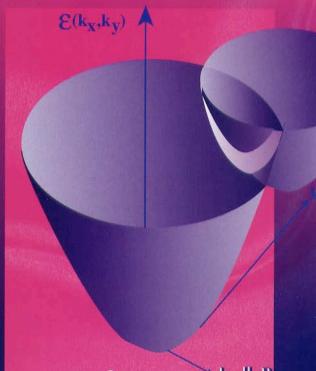
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35 Years of
Condensed Matter
and Related Physics

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# 35 Years of Condensed Matter and Related Physics

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## GAP ENHANCEMENT IN SUPERCONDUCTORS: WHERE DID IT GO?

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#### ABSTRACT

A review of the effects of microwave radiation on superconducting devices and the superconducting state is given with a focus on critical current and superconducting gap enhancement. Evidence indicates there exists at least one mechanism for critical current enhancement which does not involve an increase in the energy gap.

### 1. Introduction

The late 70's and early 80's saw the end of about twenty years of research on the effects of microwave radiation on the superconducting state. This time also signaled the end of a decade of rather intense research on nonequilibrium superconductivity in general. One unresolved issue from this era of physics research was the question: Can the superconducting energy gap be increased or enhanced by irradiation with high frequency fields, e.g., microwaves. Although most theorists and some experimentalist believed this occurred, it was not universally accepted and was, therefore, somewhat controversial.

The origin of the belief started with the observation by many experimentalists that an increase in the critical currents in superconducting microbriges and films occurred while simultaneously irradiating the superconductors with microwaves. Two models arose to explain this observation, one with the microwaves altering the superconductivity in an inhomogeneous superconductor and the other with the microwaves altering the superconducting state itself. Because there was not a complete resolution of the question at that time, the present work is an up to date review of the subject, fifteen years after the start of the controversy.

In what follows is a history of the general research on the effects of microwave radiation on the superconducting state and devices (more or less), including both experiment and theory, up to the early 80's at which time the area was considered to be mature. After this history, the results of the two camps or the opposing views of gap enhancement will be presented. Next will be discussion of the few experimental

papers after the early 80's, which focus on the nonequilibrium superconductivity area of gap enhancement. A critical discussion of the experimental papers will be followed by some final conclusions or at least discussion.

# 2. History of Microwave Irradiated Superconductors

The history of the effects of microwave radiation on the superconducting state begins with experiments specific to superconducting devices. In particular, we will look at the effects of microwave radiation on tunnel junctions current voltage characteristics and the critical currents of superconducting strips or weak links. We begin in 1962 when Dayem and Martin <sup>1</sup> observed the effects of microwave radiation from 25 to 63 GHz on Al/Pb, In and Sn tunnel junctions. They found an increase in the tunneling current at the voltages corresponding to the sum  $(\Delta_1 + \Delta_2)$  and difference  $(\Delta_1 - \Delta_2)$  of the two temperature dependent energy gaps  $(\Delta_1$  and  $\Delta_2)$  of the superconducting films, Al and either Pb, In or Sn, of the junction. These peaks are, of course, expected from the single particle picture for superconductor-superconductor tunneling. In addition to the sum and difference peaks, there was also increased current at voltages corresponding to  $(\Delta_1 + \Delta_2)/(n\hbar\omega/e)$  where  $\omega$  is the microwave radiation frequency, n is an integer, and e is the charge of an electron. These extra peaks found in the conductance correspond to either absorption or emission of an integer number of photons.

Tien and Gordon <sup>2</sup> explained the experimental observations of Dayem and Martin with a rather simple quantum mechanical model. Their model agreed with both the position of the current jumps in terms of the applied bias voltage and the microwave photon energies. In addition, their model correctly predicted the relative amplitude of the current jumps as a function of the incident microwave power.

If we now consider the effects of the radiation from a classical direction, we will find a different result as was shown by Hamilton and Shapiro  $^3$ . To understand their treatment, consider the microwaves not as individual photons but as a high frequency voltage generator. In this case the current at any d.c. bias voltage will correspond to an average of the voltage dependent current over one cycle of the r.f. voltage centered about the d.c. applied voltage. Thus, any sharp structure in the tunneling characteristic should become smeared out into two sharp structures one at  $V = V_{dc} - V_{rfmax}$  and one at  $V_{dc} + V_{rfmax}$  where  $V_{dc}$  is the voltage of the original structure and  $V_{rfmax}$  is the maximum microwave voltage across the junction. An important point to realize is the cross over of the tunnel characteristic from the quantum mechanical picture to the classical picture will occur when the energy width of the structure in the tunneling characteristic goes from less than a single photon energy to more than a single photon energy.

Thus far the microwave source has been treated as a voltage generator which assumes the junction resistance to be large. The reverse of this (junction resistance less than impedance of microwave source) has been investigated experimentally by Thulin <sup>4</sup> for the classical case. His main result is that the smearing of a structure is observed only at  $V = V_{dc} + V_{rfmax}$ , not at  $V = V_{dc} - V_{rfmax}$ . In this work, computer fits to the data were accomplished by varying the ratio of the junction resistance to the microwave source.

We now turn to the effects of microwave radiation on the critical current of weak links and small superconducting strips. The first experimental work on the effects of microwave radiation on weak links was done by Anderson and Dayem  $^5$  and later in more detail by Dayem and Wiegand  $^6$ . Their results which are relevant to this work is the observation of an increase in the critical current of the weak links (consisting of a 2 x 0.1  $\mu$ m constriction in an evaporated Indium film) during exposure to microwave radiation.

The above experimental works were explained by three different models. The first theoretical explanation of this phenomena was by Hunt and Mercereau 7. Specific to the works by Anderson, Dayem, and Wiegand, Hunt and Mercereau argued if the two larger portions of a superconducting film, i.e., the portions separated by the constriction or the weak link remain separate in a superconducting sense, the quantum mechanical phases may be different thereby gaining an extra degree of freedom and thus possess a lower energy. If this energy is greater than the difference in energy of the link in the superconducting and normal states, then the constriction would remain normal below its expected  $T_c$  (same as the larger portions of the film). The energy lost by forcing the constriction to remain normal will be proportional to its volume but the length must be such that an electron traveling from one side to the other will lose its phase memory (greater than a coherence length). If we lower the temperature until the constriction becomes superconducting, the critical value for the current density would still be less than we would expect since it still "sees" the energy of having the bulk portions of the film uncoupled. Hunt and Mercereau proposed that the effect of the microwave radiation was to somehow couple the phases of the two superconducting regions and therefore lose some fraction of this energy for a given radiation frequency and power level. This would manifest itself by raising its  $T_c$  up to the proper temperature, thereby increasing its critical current to the anticipated value.

Although Hunt and Mercereau's model may be applicable to weak links, experimental work first by Shepard <sup>8</sup> and more compete studies later by Klapwijk and Mooij <sup>9</sup> showed the effect was not limited to weak links, but that microwaves could also increase the critical current in long superconducting films. In addition, Klapwijk and Mooij reported measurements of a critical current when the temperature was above the nonirratiated superconducting transition temperature of the films,

i.e., with the irradiation of the microwaves, they were able to enhance not only the critical current in the superconducting state but also the superconducting transition temperature.

A different model, applicable to both the weak links and the long films, was that of Lindelof <sup>10</sup>. If we consider the pair potential or energy gap in a given region of a superconducting film to be lower than that in the remainder of the film (for whatever reason) then the density of Cooper pairs in this region will be reduced from the surrounding regions. When one measures the critical current, it will be dominated by this weak region just as 'a chain is no stronger than its weakest link.' With the above assumed, Lindelof argues that the measured pair potential in this region, and therefore the critical current, can be increased in the following way.

A high frequency current in a superconductor will consist of both Cooper pairs and quasiparticles (except at T=0 K where there are no quasiparticles). This is because the inertia of self-inductance of the pairs will not allow an infinitely fast response to an external electric field and therefore before the electric field inside the superconductor is reduced to zero the quasiparticles will have been accelerated giving them a net velocity. The proportioning of the current between the pairs and quasiparticles is dependent on how many of each are present. Therefore in the region where the pair potential is small, one expects the current leaving this region to have a larger proportion of quasiparticles than the current coming into this region (charge neutrality must be maintained). This creates a momentary situation where the density of Cooper pairs in the weakened region is larger than it was at equilibrium. For this to work, however, the pairs must be injected into this region faster than it can restore equilibrium. If the microwave radiation is at a higher frequency than 1/relaxation time, then the average value of the Cooper pairs would be larger in this region. Since the measured critical current is that of the region with the smallest number of pairs then the critical current of the film would be increased.

The final model we will discuss to explain the observed effects was by <sup>11</sup> Eliashberg <sup>11</sup>. From the BCS theory of superconductivity the self consistent equation for the energy gap parameter ( $\Delta$ ) is given by

$$\Delta = g \int_{\Delta}^{\hbar\omega_D} \Delta/(\epsilon^2 - \Delta^2)^{1/2} [1 - 2f(\epsilon)] d\epsilon.$$
 (20),

where g is the BCS coupling parameter,  $\epsilon$  is the energy of a particle measured with respect to the fermi surface,  $\hbar$  is Planck's constant divided by  $2\pi$ ,  $\omega_D$  is the Debye frequency of the superconductor in question and  $f(\epsilon)$  is the distribution function of the quasiparticles. The energy denominator  $(\epsilon^2 - \Delta^2)^{1/2}$  is, of course, proportional to the density of states in the superconducting state. This equation shows the value of  $\Delta$  is related to the distribution of the quasiparticles through

 $f(\epsilon)$ . If we can somehow alter the distribution by raising the mean energy level occupied by the quasiparticles,  $\Delta$  will also increase. Eliashberg showed this can be accomplished with electromagnetic radiation if the energy of a single photon is less than that necessary to break apart a Cooper pair  $(h\nu/\epsilon < 2\Delta)$  but with a frequency greater than  $1/\tau$  the temperature dependent characteristic relaxation time for the quasiparticles to decay back to equilibrium. Thus, the radiation does not affect the pairs but alters the thermal distribution of the quasiparticles.

Not only would the Eliashberg model explain observed critical current enhancement, it was also believed it could explain the increase in the critical temperature. Although not simple to show with the above equation, the belief was the increase in the superconducting gap with the redistribution of the quasiparticles under the stimulation of microwave radiation could be considered as a lower energy state than the normal or nonirradiated state. Therefore with the radiation applied to the superconductor in the superconducting state and held constant, the sample could be heated to temperatures higher than  $T_c$  and the superconducting state would persist..

In concluding the historical review up to the late 70's we should note that there were other publications describing models to explain the experimental observations <sup>12</sup>, <sup>13</sup> and <sup>8</sup> but they are either variations or elaborations of the above. In general, we can classify the models to three categories, microbridge effects (Hunt and Mercereau [Hu67]), reduced or weakened energy gap effects (Lindelof [Li76]) and direct modification of the superconducting state (Eliashberg [El70]). One might argue the Hunt and Mercereau and the Lindelof models are, in principle, both inhomogeneous superconductor models but because the Lindelof length scale may be longer and the critical current enhancement is seen in films, we will not mention the Hunt and Mercereau mechanism in the following discussion.

# 3. Gap Enhancement Proved or Not?

Experimental evidence to support the Eliashberg model was published by Kommers and Clarke <sup>14</sup>. They measured the energy gap in superconductor-superconductor tunnel junctions prepared on single crystal sapphire substrates in the presence of 10 Ghz microwave radiation. Increases of the energy gap in aluminum films of almost a factor of two were claimed by the authors. Contradictory to this, investigations by Dahlberg, Schuller, and Orbach <sup>15</sup> found no evidence for gap enhancement in a long detailed investigation in which all previous effects of microwave radiation on the superconducting state and devices, including critical current enhancement were observed (there may be some experimental artifacts if care is not taken to shield the thermometry from the microwave radiation <sup>16</sup>). They stated that no gap

enhancement was observed even though it was specifically looked for in the experiments. This last result was consistent with the results of a number of researchers who also failed to observe gap enhancement but chose to not publish a null result (private communications to the authors from Pals, Wolter, and Lindelof).

In a direct comparison between the experiments of Kommers and Clarke and those of Dahlberg, Schuller, and Orbach, we can find no differences in the experimental techniques. The samples had similar  $T_c$ 's, comparable dimensions, the range of temperatures below  $T_c$  investigated were the same, the frequencies used were the same, and the range of junction resistances were the same. On occasion the junction resistances of Dahlberg et. al. were low enough to see Josephson coupling and microwave induced Josephson tunneling between the films, as Kommers and Clarke show in their data.

Finally, a most curious point is the following. As stated before, in the experiments of Dahlberg, Schuller, and Orbach, they observed enhancement of the critical currents by as much as a factor of ten. In their measurements of the gap, even when the microwave enhanced critical current was present, the gap remained unenhanced. It is interesting to recall that the original motivation to believe in gap enhancement was the increase in critical current. This phenomena, critical current enhancement, is very easy to observe in aluminum films, with at least a dozen experimental reports on the observation of critical current enhancement; observations of gap enhancement are considerably more scarce. This of course flies in the face of the original impetus for the theory of gap enhancement-the observation of microwave enhanced critical currents.

With the above setting the stage, there was a flurry of activity in the late 70's to resolve this issue. One experiment by Pals and Dobben <sup>17</sup> which supported gap enhancement was a measurement of the magnetic flux in a cylinder coated with a thin aluminum film. By relating the flux in the cylinder to the gap with a simple model, they claimed a 2% increase in the gap. This was, of course, a rather modest increase compared to that reported in the tunnel junction measurements. Also, a comment by the authors which casts some doubt on their reported gap enhancement was they were unable to correlate the 2% increase with the critical currents because of inhomogeneities in the film.

There was another group at the then NBS laboratory, Hall, Holdeman, and Soulen, who were able to observe enhancement in two aluminum tunnel junctions on BaF2 substrates in the range of 2 to 4 Ghz but not at 10 Ghz <sup>18</sup>. In their work, they saw enhancement of almost a factor of two in the energy gap in the junctions with the application of 3.72 Ghz radiation. Although not mentioned in this publication, a private communication from the authors indicated that the films of the tunnel junctions were Josephson coupled in both samples. This later fact may be important as the Josephson coupling could allow the Lindelof effect to be active between two physically uncoupled films.

# 4. Gap Enhancement after the Early 80's

Since 1980 there have only been four experimental papers dealing with gap enhancement. Two of the papers use microwaves as the agent to alter the quasiparticle distribution <sup>19</sup> and <sup>20</sup> and the other two use high frequency phonons <sup>21</sup> and <sup>22</sup>. Because of the paucity of experiments dealing with gap enhancement since 1980 we will include the works using phonons to disturb the quasiparticles. Starting with the work of Horstman and Wolter <sup>19</sup>, they claimed an increase of 40% in the energy gap with microwaves of 13.8 Ghz. If one were to be critical of this work, the focus would be on the I-V characteristics with the application of microwaves. There is not a signature of photon induced tunneling. This fact leads one to believe the effect of the microwaves on the junctions would be described by the classical model discussed above. In this case, what the authors report to be an observed increase in the gap, would be a voltage smearing of the gap with the peak in the current, not occurring at the gap, but at the gap plus the rf voltage generated across the junction (see for example <sup>14</sup> and <sup>23</sup>).

The work of Escudero and Smith  $^{19}$ , which also measured increases in the gap with application of microwaves, used tin-tin junctions. The anomalous result of this work is that although they reported increases of the gap of 5.5%, this was at 2.8K or at a  $T/T_c$  of only 0.72. At this low temperature there should be very few quasiparticles left for the microwaves to redistribute. At higher temperatures of 3.7K there was only a 3% effect. These results are opposite to what should occur if the Eliashberg mechanism is responsible for the observations and are also opposite to the observations of Kommers and Clarke.

The first use of phonons for the quasiparticle redistribution to create gap enhancement was by Miller and Rutledge  $^{21}$ . As with Escudero and Smith, they also worked with tin-tin junctions and also saw enhancement at a low reduced temperature of  $T/T_c$  of only 0.41. The increase in the gap was equivalent to a cooling of the junction of 1.4 mK. Although rather a rather modest increase in the gap, one should remember that at the reduced temperature of 0.41 there would not be many quasiparticles to affect. Finally, Seligson and Clarke  $^{22}$  also used phonon irradiation but on aluminum- aluminum tunnel junctions. They found increases in the gap of up to approximately 400% at  $T/T_c$  of 0.99 but were not able to correlate the gap increase with critical current increases.

### 5. Discussion

As a starting point, it is fairly safe to assume that there is critical current enhancement which is distinct from gap enhancement (if it exists as formulated by Eliashberg). For evidence in addition to the above lack of simultaneous observation and/or correlation of critical current and gap enhancement, there is the question of how the Eliashberg effect can work at temperatures above  $T_c$ . Klapwijk, van den Bergh and Mooij 24 measured the increase in the critical currents of films with microwave radiation applied. They found that at temperatures above  $T_c$ , where no critical current was observed that they were able to generate a measurable critical current with application of microwave radiation. The Eliashberg model must then explain how the incident radiation on the normal state above  $T_c$  could alter the electron distribution in such a way that the superconducting state would be energetically favorable. This is very different from the original Eliashberg interpretation where the redistribution of the quasiparticles occurs in the superconducting state, i.e., the microwaves applied below  $T_c$ , stabilizes the superconducting state and allows it to be heated to a temperature higher than  $T_c$ . Certainly to claim the Eliashberg model can create a superconducting state from the normal state is not easy to understand conceptually.

The enhanced  $T_c$  effect was explained by Falco, Werner and Schuller <sup>25</sup>, not with the Eliashberg mechanism but with the Lindelof mechanism which requires an inhomogeneous superconductor. They found that fitting the temperature dependence of the critical current over a  $T/T_c$  range from 0.94 to 0.99 would determine a lower critical temperature for a superconducting aluminum film. They show that even if the bulk of the film had a single  $T_c$  there would be smaller regions with a higher  $T_c$  most likely due to the oxygen sensitivity of superconducting aluminum. Given this situation, even above  $T_c$  a supercurrent could be generated in the film with the Lindelof effect by the microwaves averaging the superconductivity in these small regions over the bulk in the film.

A second experimental result which is difficult for the Eliashberg model to explain is critical current enhancement in proximity effect junctions. Warlaumont, Brown, Foxe, and Buhrman <sup>26</sup> found increases in the critical currents in normal metal proximity bridges. For reasons similar to those given in the first paragraph in this section, it is difficult to understand how the Eliashberg mechanism can be responsible for this effect. Confronting the same logic, the authors of [26] attribute their observations to the Lindelof mechanism.

Turning now to the central question of this paper which is gap enhancement,

we must state that there are no papers published since 1980 which show conclusive proof of gap enhancement with the application of microwave radiation. One might ask as to why the simplicity of the Eliashberg model can be flawed. Possibly it is just that. It is to simple. It does not consider the heat transfer out of the film or junction, nor does it consider the effects of the nonequilibrium distribution of the quasiparticles on the various relaxation times, such as the quasiparticle relaxation time.

This makes one reconsider the original works which were argued to confirm its applicability. It may be possible that because of the paucity of experiments which have observed gap enhancement that the Lindelof effect has been working all along. Consider that of the, say, 100 tunnel junctions studied by all groups, only three or four showed the effect. Then maybe that was the three or four where the junction happened to probe a weak region where the weak region was in contact with a strong region. In this case, the microwaves will average the gap over the two regions, but the tunneling, measuring only the weak region, sees an increase in the gap.

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