LIGHT: AN IDEAL PROBE TO UNDERSTAND CONDENSED MATTER

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Ladies and Gentlemen,

It is a great honour for me to be awarded the Latsis Prize. During my short presentation, I will try to highlight the main goal of our experimental technique and the major achievements obtained so far, with a perspective to our future projects, as well.

1. Introduction

The interaction of light with the condensed matter is the basic idea and principal foundation of several experimental spectroscopic methods. With light, generally considered here as electromagnetic radiation, one can cover an extremely large range of frequency (or energy) and consequently one can investigate several aspects of the intrinsic solid state properties of matter. Optical methods are particularly suitable since, due to the fairly large penetration depth of light in the matter, the study of so-called bulk properties can be achieved. Consequently, optical techniques are very important for the investigation of the complete excitation spectrum of matter.

A short survey of the nowadays most investigated systems in solid state physics immediately shows us the variety of energy scales, characterizing their ground state. For instance, so-called broken symmetry ground state or many-body highly correlated materials display a large diversity of relevant characteristics scales. In the class of the broken symmetry ground state systems, we can find the superconductors or the charge and spin density (CDW or SDW) wave condensates, while the heavy-electron materials are the most exciting examples of highly correlated systems.

2. Heavy-electron systems

Heavy-electron systems have developed into a new branch of metal physics which is flourishing and presumably will attract research interest for many more years. One can speak of metals with strongly

correlated, yet delocalized electrons. In order to understand this, we shall first compare this new kind of metals with the more conventional simple metals. Usually, metals are characterized by typical properties like: increasing conductivity with decreasing temperature, small and temperature independent magnetic susceptibility and a linear temperature dependence for the specific heat at low temperatures.

In heavy-electron systems the resistivity does not change appreciably at high temperatures, but below a characteristic temperature there is a strong decrease of the resistivity by decreasing temperatures. The magnetic susceptibility follows at high temperature a Curie-Weiss law, and very often saturates at a constant value. The low temperature specific heat behaviour (yet not universal) of heavy-electron systems shows that there are far more low lying excitations present than in an ordinary metal. A most interesting finding is that there seems to be a one-to-one correspondence between the low-lying excitations in heavy-electron systems and those in a nearly free electron gas when the parameters of the latter (e.g., the mass or the magnetic moment of the electrons) are properly renormalized. The name 'heavy-fermion' stems from the observation that enormously large specific heat coefficient implies an effective mass m* of the quasiparticles which is several hundred times the free electron mass. The key aspect is the presence of localized f-electrons in all heavy-electron systems. These may be Ce ions with 4f electrons or U or Np ions with 5f electrons. The 'heavy-electron' can be considered as the 4f or 5f electron interacting with the bands of the delocalized electrons (d or s). Essentially, this interaction is a hybridization with the band states near the Fermi level (E_F) via the Kondo spin fluctuations through constant exchange spin-flip transitions of f-electron and band electrons in the vicinity of E_F. It is precisely this process which leads to strong mixing of the Fermi

electrons with the localized f-electrons. The final result is a renormalization of the Fermi surface and moreover a drastic enhancement of the effective mass of free band electrons at E_F.

The electrodynamics of several heavy-electron metals have been explored in detail [1]. In the coherent state the optical conductivity shows a narrow resonance centered at zero frequency. Moreover, the small spectral weight of this resonance is indicative of the large effective mass m* of the heavy quasiparticles. However, significantly less was known about the electrodynamics of the superconducting and magnetic states of these materials. Although magnetic-ordering and heavy-electron behaviour (eventually together with superconductivity) seem, at first sight, to be mutually exclusive, various experimental observations in recent years indicate that this is not necessary so. Both magnetic ordering out of a heavy-electron state and the formation of a heavy-electron state in a magnetically-ordered matrix seem possible. Examples of these two distinctly different situations are realized in the low-temperature properties of U_2Zn_{17}, UCu_5, UPd_2Al_3 and URu_2Si_2.

From our observations we have concluded that UPd_2Al_3 and U_2Zn_{17} undergo an antiferromagnetic phase transition involving mainly local magnetic moments. In contrast UCu_5 and URu_2Si_2 exhibit an excitation spectrum characterized by the sudden appearance of absorption in the far infrared (FIR) spectral range at a temperature coincident with the onset of the antiferromagnetic transition, which we tend to ascribe to a spin-density-wave (SDW) gap. This suggests that the magnetic state develops as the consequence of a Fermi surface anomaly, similar to the one at the SDW transition in Cr [2,3].

A major issue and current topic of debate in the field of highly-correlated electron systems is the fundamental question whether these systems, in their normal state, may be described as simple Fermi liquids. For some heavy- electron compounds (e.g., UPt_3) this seems indeed to be the appropriate picture. Specific-heat and resistivity data, supported by results of de Haas-van Alphen experiments may be well explained in this way, if strongly renormalized Fermi-liquid parameters are introduced. More recent experimental work has indicated, however, that several heavy-electron compounds and related alloys display quite remarkable properties, which may much less well be related with 'conventional' Fermi liquid behaviour. Problems with Fermi-liquid type descriptions of relevant data were met on different occasions, including materials like $UCu_{5-x}Pd_x$ for $x=1.5$, $CeCu_{5.9}Au_{0.1}$, $U_{1-x}Th_xPd_2Al_3$ and also the alloy $U_xY_{1-x}Pd_3$ for $x=0.2$. Particularly in these latter compounds, a set of data of specific-heat, magnetic susceptibility and resistivity measurements was found to be at variance with the usual temperature dependence expected for these quantities in Fermi-liquid-type systems.

Optical investigations, extending over a broad frequency range and at various temperatures, are in principle suitable for the simultaneous study of the energy and temperature dependence of intrinsic parameters characterizing the system, like the energy and temperature dependence of the transport relaxation time τ. This is an important quantity in order to reveal possible non-Fermi liquid behaviour. Such investigations are in progress [4].

3. Quasi one-dimensional systems

It was first pointed out by Peierls in 1955 that a one-dimensional metal coupled to the underlying lattice is not stable at low temperatures. In fact, for a one-dimensional metal with a partly filled band

the regular chain structure will never be stable, since one can always find a suitable distortion for which a break (i.e., a gap) will occur at or near the edge of the Fermi distribution edge.

For a one-dimensional metal at T=0, in the absence of electron-electron or electron-phonon interaction, the electron states are filled up to the Fermi level; the lattice is a periodic array of atoms. In the presence of an electron-phonon interaction, it is energetically favourable to introduce a periodic lattice distortion. The distorted lattice introduces a periodic potential and the free electrons will try to screen it. This distortion also opens up a gap at the Fermi level. The development of a gap leads to a lowering of the electronic energy. In one dimension the single-particle gap is proportional to the amplitude of the periodic lattice distortion, and leads to a decrease of the energy of the electron gas. On the other hand, the distortion leads also to an increase of the elastic energy proportional to the square of the distortion. For a small distortion, the total energy of the coupled electron-phonon system is smaller than that of the undistorted metal, so that a new ground state can be achieved. The modification of the dispersion relation also leads to a position-dependent electron density much in the same way as in the nearly-free-electron theory of metals. This corresponds to a so-called charge-density-wave (CDW). In the case that the electron-electron interaction dominates, a SDW ground state will develop.

Our experiments led to the discovery of several features characterizing the electrodynamic response of these condensates: namely, the modes due to the internal excitations of the density waves, the collective pinned mode and the single particle gap. We have also applied optical methods in order to study how the so-called precursor or fluctuation effects affect the phase transition. Strong deviations from the prediction for a simple or conventional Fermi-liquid metal have been found [5,6].

There is also renewed interest in the behaviour of highly anisotropic conductors in their metallic state. The reason for this is twofold. Various theories based on the one-dimensional Hubbard model predict a non-Fermi liquid state with profound implications on the spectroscopic signatures in the so-called metallic state. The name Luttinger liquid is used in general to describe the new quantum liquid state which follows from the different solutions of the one-dimensional electron systems. Of course, real materials such as the linear chain compounds are not strictly one-dimensional but have finite interchain coupling or interchain single particle transfer integrals. This raises interesting questions about the extent to which strictly one-dimensional models are applicable to experiments performed in real materials. In addition, questions concerning the interplay between the periodicity of the electronic correlations and the lattice periodicity are among the central aspects of various such theories. This interplay may lead to unusual features such as discommensurations and nonlinear charge and spin excitations.

We are presently conducting thorough optical experiments (as a function of temperature and light polarization) on very large single crystals of the materials $(TMTSF)_2X$ ($X=PF_6$, AsF_6 and ClO_4), which are prototype materials undergoing a spin-density-wave (SDW) phase transition at temperatures between 6 and 15 K. By extending our optical investigations even lower than the FIR spectral range, we will be able to considerably improve the general understanding of the ground state properties of these quasi-one-dimensional materials [6-8].

4. The alkali metal doped fullerenes

The twin discoveries of molecular and bulk C_{60} have laid the foundation for a vast new field of studies in condensed matter physics. Much of the interest in this new form of carbon originates from the subsequent discovery of superconductivity when bulk C_{60} is

doped with alkali metals. After the initial discovery of enhanced conductivity and particularly of superconductivity in A_xC_{60}, it was shown that the fraction of superconducting material, probed by ac-susceptibility, was maximized using a starting composition near $x=3$.

Despite a variety of experiments, several issues were controversial either for the normal or for the superconducting state properties of A_3C_{60}. The central question, of course, was about the nature of the pairing mechanism. There has been debate about whether electron-electron interactions on the C_{60} ball or electron-phonon (e-ph) coupling mediate the pairing. Concerning the latter mechanism, there was quite a bit of controversy with respect to arguments, favouring electron-phonon interactions with low frequency *inter*molecular vibrations or with high frequency *intra*molecular modes. The energy scale of the various modes which mediate the e-ph coupling are different, and therefore, which of these are important, could in principle be decided by examining whether the weak or strong coupling limit applies. In the case of high frequency phonons for example, one expects, within the mean-field Bardeen-Cooper-Schrieffer (BCS) theory, the weak coupling limit to be appropriate with the ratio of the single particle gap Δ and the transition temperature T_c given by $2\Delta/k_BT_c= 3.52$. In the case of low frequency vibrations, on the other hand, $2\Delta/k_BT_c$ is expected to exceed the value which is appropriate for the weak coupling limit.

In this context, optical investigations, besides characterizing the normal state properties, are a powerful experimental tool in order to single out whether the weak or strong coupling limit of the BCS theory is more appropriate to describe the superconducting state. Our work mainly focused on our attempts to determine the relevant energy scale (i.e., the superconducting gap) of the alkali doped

superconducting fullerenes. From the analysis of the complete excitation spectrum of single crystals K_3C_{60} and Rb_3C_{60}, extending from the far infrared (FIR) up to the visible and ultraviolet (UV) frequency range, we evaluate several intrinsic parameters, characterizing the normal and superconducting state properties. Particularly, our reflectivity measurements show clear evidence of well defined superconducting gaps with $2\Delta/k_BT_c$ in full agreement with the prediction of the weak coupling BCS limit. The motivation of our optical work was somehow more ambitious. Indeed, we tried to fully exploit the optical investigation and to single out the relevant phonon excitation for the pairing mechanism and thus for superconductivity. In this respect, we discussed our experimental results within the standard Eliashberg electron-phonon theory of superconductivity. These calculations strongly support a pairing mechanism mediated by high-frequency *intra*molecular phonon modes, in accord with the implications of the weak coupling BCS limit [9,10].

Besides the superconducting alkali-metal doped fullerenes, there are other stable phases of A_xC_{60}. For quite a long time, the $x=3$ compound was believed to be the only electrically conducting fullerenes salt. However, a new phase with one alkali atom per fulleride ($x=1$) and metallic characteristics was recently discovered. This new member of the already rich family of C_{60} derivatives undergoes a first order structural phase transition at about 400 K. It has been suggested that the structural phase transition is the consequence of a polymerization of the C_{60} molecules. Therefore, the crystal structure of these polymer chains along the orthorhombic axis suggests a strongly anisotropic electronic structure with one electron per C_{60} forming an unfilled band. Furthermore, it has been suggested from magnetic measurements that the Rb_1C_{60} polymer is a quasi one-dimensional (1D) conductor with a spin-density-wave (SDW) ground state, developing at about 50 K.

We presented our optical data on the frequency dependent conductivity of A_1C_{60} polymers. All three A_1C_{60} materials appear to be conducting at ambient temperature but with an unusually strong frequency dependence in the far infrared. K_1C_{60} is a poor metal with a slowly decreasing low frequency conductivity from 4 K up to the depolymerization temperature of 400 K. Rb_1C_{60} and Cs_1C_{60} are also conductors at ambient temperatures but in contrast to K_1C_{60} display metal-insulator transitions at 50 and 40 K, respectively. At the lowest temperatures, the optical conductivity is depressed below 15 and 30 meV photon energy, indicating a vanishing density of states at the Fermi level. Our optical data reinforce the picture of a quasi one-dimensional instability towards a condensed SDW state for the phase transition at about 50 K in the Rb_1C_{60} and Cs_1C_{60} polymers [11].

This latter paragraph concludes this brief review. However, the physics of strongly correlated and broken symmetry ground states is still an alive and stimulating research area, which will attract for many more years the interest of several investigators. Therefore, it just remains to follow the provocative incitation of Leonardo da Vinci:

O miseri mortali, aprite gli occhi.

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