AN EVALUATION OF FREQUENCY DEPENDENT EFFECTIVE MASS m* AND OPTICAL SCATTERING RATE OF HEAVY ELECTRON COMPOUND

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ABSTRACT

This paper presents a method of evaluation of frequency dependent optical scattering rate r(ω) and the effective mass $\frac{m^*}{m_b}$ (ω) of heavy electron compound $CeAl_3$. Two plasmon frequency ω_p (unrenormalised) and ω^*_p (renormalized) for some of the heavy compounds are also evaluated. Our theoretical results are in good agreement with the experimental data and other theoretical workers.

Key words: Heavy electron compound, Effective mass, Optical scattering rate, Kramers-Kronig analysis.

INTRODUCTION

Highly correlated states of condensed matter have opened new chapters in physics. Examples of particular interest include the so–called Kondo or heavy electron materials, which were discovered in the late seventies. The class of strongly correlated systems also include the transition metal oxides, including *d*–electron (Mott Hubbard) systems as well as the high temperature superconducting cuprates, quasi–one–dimensional materials, such as organic Bechgaard salts, and possibly the carbon fullerenes.

Heavy electron systems are electrically conducting materials with peculiar low temperature physical properties that distinguish them from ordinary metals[1–4]. In fact, the conduction–electron specific heat is typically some 100 times larger than that found in most metals. Similarly, the magnetic susceptibility can be two or more orders of magnetic

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susceptibility can be two or more orders of magnitude larger than temperature independent Pauli susceptibility observed in conventional conducting materials.

The prototype heavy electron materials include actinide and rare earth alloys, mostly containing U and Ce, respectively, like $CeAl_3$, $CeCu_6$, UBe_{13} and UPt_3. At high temperature, these systems behave as a weakly interacting collective of *f*-electron moments and conduction electrons with quite ordinary masses; at low temperatures, the *f*-electron moments become strongly coupled to the conduction electrons and to one another, and the conduction electron effective mass is typically 10 to 100 times the base electron mass. This obviously leads to an enhanced Sommerfeld coefficient γ of the linear T term associated with the electronic contribution to the specific heat. Alternatively, such an enhancement of the values indicates a large electronic density of states at the Fermi level E_F.

Heavy electron materials constitute a formidable challenge to condensed matter physicists not only with respect to their normal properties but also to their superconducting and magnetic properties, which are not yet fully understood. Particularly, the coexistence of heavy electron behavior with magnetic ordering or superconductivity (or both) has attracted a lot of interest. In fact, although magnetic ordering and heavy electron behavior (possibly together with superconductivity) seem, at first sight, to be mutually exclusive, various experimental observations indicate that this is not necessarily 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} and UCu_5, which order antiferromagnetically at 9.7 and 15 K, respectively. Also of interest are those heavy electron materials showing the coexistence of superconductivity with magnetic ordering[5]. In this respect, URu_2Si_2 attracted a lot of attention as the first heavy electron metal with superconductivity (T_c = 1K), developing in an antiferromagnetically ordered matrix (TN = 17.5 K). A systematic search[6,7] led to the discovery of the co-existence of anti-ferromagnetism and superconductivity in UNi_2Al_3 (T_N = 4.6 K, and T_c = 1 K) and UPd_2Al_3 (T_N = 14 K and T_c = 2 K), as well. On the other hand, superconductivity also manifests itself in some of the more 'conventional' heavy electron materials, e.g. $CeCu_2Si_2$, UPt_3 and UBe_{13}.

In this paper, we have presented the method of evaluation of frequency dependence optical scattering rate and effective mass of heavy electron compound $CeAl_3$ at different temperatures. We have used theoretical formalism developed by Millis and Lee[8] and Kramers-Kronig analysis of optical conductivity σ(ω) in our evaluation. Our theoretical results are in good agreement with that of the other theoretical workers and experimental data.

Mathematical formulae used in the evaluation

One uses Millies and Lee[8] model, which describes a band of nearly free electrons hybridizing with a very highly correlated band of f electrons. In the absence of this hybridization, the f electrons are confined to a site in localized orbitals far below the Fermi energy. This model contains the essential physics of heavy electron metals, a non–magnetic ground state that behaves as a Fermi liquid with large effective mass. In the U limit, where U is the coulomb repulsion between f electrons on the same site, the Hamiltonian may be written as -

$$H_A = \sum_{k\sigma} \varepsilon_{k\sigma} C_{k\sigma}^* C_{k\sigma} + \sum_{im} E_{om} f_{im}^* f_{im} + \sum_{kim\sigma} (V_e^{ikR_i} C_{k\sigma}^* f_{im} + H.C.) \qquad \ldots(1)$$

This Hamiltonian describes a band of conduction electron (c electron) with operator $C_{k\sigma}$ and energy $\varepsilon_{k\sigma}$, which hybridizes with a set of localized f electrons with operator f_{im} and energy E_{om} via an interaction V, taken to be constant. The solution is subject to the constraint that each site may be occupied by at most one f electron (this U=∞ limit).

$$\sum_{im} f_{im}^* f_{im} = n_f^i \leq 1 \qquad \ldots(2)$$

Since the above constraint does not commute with HA, a slave boson technique is used to handle the Hamiltonian (1). Here, there are two possible sources of scattering of electrons. One is the scattering of electron off the impurity; the other is the scattering from the boson fluctuations. The latter turns out to be in some ways analogous to electron phonon scattering and compounds to the fluctuations of the slave bosons with respect to their saddle point value of 1/N effect. Here also, one assumes the validity of Matthiessen's rule[9] where the scattering due to different mechanism is added. If in the presence of impurities only, conductivity is σ_i and in the presence of bosons only the conductivity is σ_b; then the total conductivity is given by

$$\sigma^{-1} = \sigma_i^{-1} + \sigma_b^{-1} \qquad \ldots(3)$$

Matthiessen's rule is believed to be valid where the various scattering mechanism are not momentum dependent.

At sufficiently low temperature, only impurity scattering is relevant. To compute σ_i (ω, T) of σ (ω, T) disorder must be coupled in the system. If the disorder is weak and on the conduction electron site only; then the corresponding Hamiltonian -

$$H_{imp} = \sum_{kk'm} V_l C_{km} C_{k'm} \qquad \text{...(4)}$$

V_1 is the impurity scattering amplitude. One also introduces an energy parameter εf which is analogous to the Debye frequency in the electron- phonon interaction problem. It sets the scale for the energy of the boson propagator. Now the large effective mass* is given

$$m^* \sim \frac{W}{N\varepsilon_f} \qquad \text{...(5)}$$

Where W is the bare bandwidth and N is the orbital density of the *f*-state.

For $W \leq \varepsilon_f$ the impurity scattering is isotropic $\sigma_i(\omega)$ reduces to[10, 11] -

$$\sigma_i(\omega) = \frac{ne^2}{m_b} \frac{\tau_i}{1 + \left(m^*/m_b\right)^2 (\omega^2 \tau^2)}$$

$$= \frac{ne^2}{m_b} \frac{\tau_i^*}{1 + (\omega\tau^*)^2} \qquad \text{...(6)}$$

Where

$$\tau_i^* = \left(\frac{m^*}{m}\right)\tau_i \qquad \text{...(7a)}$$

$$n = \frac{Nk_F^3}{6\pi^2} \qquad \text{...(7b)}$$

both for conduction and *f* electrons. m_b is the conduction band mass.

Two characteristic plasma frequency are expected

(i) One at high frequency

$$\omega_p^2 = 4\pi ne^2 / m_b \qquad \text{...(8a)}$$

This identifies the uncorrelated conduction electrons.

(ii) Second at low frequency associated with the heavy plasmons.

$$\omega_p^* = [6(1\frac{n_f}{n_c})]^{\frac{1}{2}} T^*$$

...8(b)

Where T* is the renormalized Fermi temperature usually identified with Kondo temperature T_k. Here $n = n_c + n_f$ is the total carrier density. n_c is the conduction electron density and nf is the f electron density. The heavy electron plasma mode reflects not only the heavy quasi-particle mass (m*/m_b) but also the renormalized plasma frequency (or unscreened heavy plasmon).

$$(\widetilde{\omega}_p^*)^2 = \left(\frac{4\pi\pi n^2}{m^*}\right)$$

...(8c)

Which appears in equation (8a) and corresponds to the spectral weight with narrow Drude like mode[12]. The optical conductivity can generally be approximated by a sum of Lorentz harmonic oscillator and of Drude term. The former contributions arise each time an absorption of finite frequency takes place and they are usually ascribed to vibrational infrared active modes (phonons) or/ and to electronic interband transition. The Drude term applies for metals and describes the free charge carrier's contribution to the electrodynamics response. The general formulae for the complex dielectric function is

$$\widetilde{\varepsilon}(\omega\omega = \varepsilon_\infty - \frac{\omega_p^2}{\omega(\omega+i\Gamma\Gamma)} - \sum_j \frac{\omega_{pj}^2}{(\omega_j^2 - \omega^2) - i\Gamma_j\omega}$$

...(9)

where ω_p and $\Gamma \to \frac{1}{\tau}$ in the Drude term are the plasma frequency and the damping (scattering relaxation) of the free charge carriers while ω_j, Γ_j and ω_{pl} are the resonant frequency, the damping and the mode strength of the harmonic oscillators respectively[13]. The high frequency absorption above the ultraviolet spectral range are taken into account by ε_∞. This phenomenological fit is a useful approval in order to decouple the various components determining the excitation spectrum and to evaluate the several parameters like the plasma frequency and the scattering relaxation rate.

Sum rule arguments lead to the following integral[14]

$$I_1 = \int_0^{\omega_h} \sigma_i(\omega\omega T \gg T_\infty)d\omega = \frac{\pi n e^2}{2m_b} = \frac{\omega_p^2}{8}$$

...(10)

Using equation (10), one determines the unscreened optical plasma $\hbar\omega_p$~ 3.5 eV for CeAl$_3$. ω_h is the high frequency -3000 cm^{-1}. For T $<<$ T$_{co}$ (low temperature) *after the high frequency interband transition removed*

$$I_2 = \int_0^{\omega_c} \sigma(\omega T << T_\infty)d\omega = \frac{\pi n e^2}{2m^*} = \frac{\omega_p^{*2}}{8} \qquad \qquad \text{...(11)}$$

ω_c is the cut off frequency, ω_p^* is the renormalized plasma frequency,

$$\omega_p^* = \omega_p \sqrt{\frac{m^*}{m_b}}$$

$$\tau^* = \tau \frac{m^*}{m_b}$$

$$\omega_c^2 = N\varepsilon_F(\frac{m_b}{m^*})/\tau_i \qquad \qquad \text{...(12)}$$

$$\sigma(\omega,T) = \frac{i\omega_p^2}{4\pi\omega} \int_{-\infty}^{\infty} [f(\varepsilon - \omega) - f(\varepsilon)]/[\omega + \Sigma_0^A(\varepsilon - \omega,T) - \Sigma_c^R(\varepsilon,T)]d\varepsilon \qquad \text{...(13)}$$

Where Σ_c is the band electron self energy A states for advanced and R for retarded. ω_p is the Drude plasma frequency and $f(\varepsilon)$ is the Fermi function.

Now one compares the results of the plasma frequency at 300 K with ω^* at temperature lower than T$_{co}$

$$\left(\frac{\omega_p(300K)}{\omega_p^*} \right) = \sqrt{\frac{m^*}{m_b}} \sqrt{\frac{n(300K)}{n(T < T_\infty)}} \qquad \qquad \text{...(14)}$$

Since the total change carrier concentration does not change below T$_c$, one can estimate enhancement of the effective mass m*.

Now frequency dependent scattering rate Γ (ω) and m*/m$_b$ (ω) are determined through Kramers–Kronig relation between σ_1 and σ_2 where σ_3 is the real part of $\sigma(\omega)$ and σ_2 is the imaginary part of $\sigma(\omega)$. The complex conductivity may be written[15] -

$$\sigma(\omega) = \frac{\omega_p^2 / 4\pi}{\Gamma(\omega) - i\omega \frac{m^*}{m_b}(\omega\omega)} \qquad \ldots(15)$$

Where ω_p is the unscreened optical plasma frequency. Now the relationship between σ_1, σ_2, and Γ together with (m^*/m_b) are obtained as

$$\Gamma(\omega) = \frac{\omega_p^2}{4\pi} \frac{\sigma_1}{|\sigma|^2} \qquad \ldots(16)$$

and

$$\frac{m^*(\omega)}{m_b} = \frac{\omega_p^2}{4\pi} \frac{\sigma_2}{|\sigma|^2} \qquad \ldots(17)$$

Where $\omega < \Gamma$ and $\omega \to 0$, σ_1 is constant and $\sigma_2 \to 0$ while both $\Gamma(\omega)$ and $\frac{m^*}{m_b}(\omega)$ mabove frequency independent values.

RESULTS AND DISCUSSION

In this paper, we have presented the method of evaluation of frequency dependent optical scattering rate $\Gamma(\omega)$ and the effective mass $\frac{m^*}{m_b}(\omega)$ of heavy electron compound CeAl$_3$. The result is shown in Tables 2 and 3 with experimental data. We have used equation (16) and (17) for these evaluations. The value of σ_1 and σ_2 are obtained from the Kramers–Kronig analysis of the data[15], where optical conductivity $\sigma(\omega) = \sigma_1(\omega) + i\sigma_2(\omega)$. In Table 1, we have shown the evaluated results of unscreened optical plasma frequency ω_p, renormalized plasma frequency ω_p^* using relation (8a) and (8b) taking the value of effective mass (m^*/m_b) and (m^*/m_e) for different heavy electron compounds. Our theoretical results for $\frac{m^*}{m_b}(\omega)$ for different T indicates that at lower temperature m* assume to be constant value of frequencies below which it becomes frequency independent. This crossover frequency [say ω_{co} (T)] is approximately 0.3, 0.9 and 0.7 cu^{-1} at 1.2, 3.0 and 5 K, respectively. The values of m^*/m_b for CeAl$_3$ at ω_{co}'s are 418, 376 and 218 m, which we believe to be the zero frequency effective mass. A substantial increase with effective mass occurs only below 3 K and at 10 K and above, the coherent states does not exist, therefore

no mass enhancement is reported in the surface independence results[16-18]. In Table 3, we have shown the renormalized frequency dependent optical scattering rate $\hbar\Gamma(\omega)$ (eV). We have compared the result obtained from the surface independent measurement[19]. Above 10 K, the frequency dependence of Γ disappears but at low temperature, the scattering rate is only temperature dependent. We have also calculated renormalized scattering rate $\Gamma^* = \Gamma$ (m^*/m_b). For CeAl$_3$, Γ^* in its dc limit i.e. Γ^* ($\omega\rightarrow0$) has values 0.7, 2.5 and 4.5 cu^{-1} at 1.2, 3 and 5 K, respectively[20-22].

Table 1: Evaluated results of plasma frequency ω_p and $\omega_p{}^*$ and effective masses (m^*/m_b) and (m^*/m_e) of some heavy electron systems

System	ω_p	$\omega_p{}^*$	(m^*/m_b)	(m^*/m_e)
CeAl$_3$	3.5	0.155	510	809
UPt$_3$	2.6	0.326	65	248
CeCu$_6$	2.3	0.35	40	36
CePd$_3$	1.84	0.15	150	602

Table 2: Evaluated results of frequency dependent effective mass m* of heavy electron compound CeAl$_3$ at different temperatures, comparison is made with expt. Date 16

Frequency (cm^{-1})	$\frac{m^*}{m_b}(\omega)$							
	T = 1.2 K		T = 3.0 K		T = 5 K		T = 10 K	
	Theory	Expt.	Theory	Expt.	Theory	Expt.	Theory	Expt.
10	412		356		212		112	
20	406		329		203		84	
50	396		305		183		73	
60	372		286		165		43	
70	351		247		154		36	
80	332	298	218	230	135	142	27	50
90	267	256	198	222	107	130	18	29
100	212	212	174	210	87	121	11	22
120	196		152		76		9	
150	140		130		42		4	

Table 3: Evaluated results of frequency dependent renormalized optical scattering rate $\hbar\Gamma(\omega)$ for CeAl$_3$ at different temperatures

Frequency (cm^{-1})	Optical scattering rate $\hbar\Gamma$ (w)							
	T = 1.2 K		T = 3 K		T = 5 K		T = 10 K	
	Theory	Expt.	Theory	Expt.	Theory	Expt.	Theory	Expt.
10	0.025		0.068		0.105		0.122	
20	0.028		0.076		0.116		0.136	
50	0.361		0.085		0.122		0.146	
60	0.039		0.097		0.135		0.153	
80	0.052	0.030	0.106	0.060	0.138	0.102	0.166	0.135
90	0.065	0.035	0.118	0.082	0.142	0.125	0.175	0.140
100	0.078	0.042	0.125	0.098	0.148	0.134	0.186	0.146
110	0.096	0.056	0.135	0.105	0.156	0.139	0.195	0.158
120	0.125	0.066	0.142	0.016	0.167	0.145	0.205	0.167
140	0.138		0.156		0.172		0.226	
150	0.142		0.167		0.184		0.245	

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