Heavy Carriers and Non-Drude Optical Conductivity in MnSi

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Optical properties of the weakly helimagnetic metal MnSi have been determined in the photon energy range from 2 meV to 4.5 eV using the combination of grazing incidence reflectance at 80° (2 meV to 0.8 eV ) and ellipsometry (0.8 to 4.5 eV). As the sample is cooled below 100 K the effective mass becomes strongly frequency dependent at low frequencies, while the scattering rate develops a linear frequency dependence. The complex optical conductivity can be described by the phenomenological relation $\sigma(\omega, T) \propto (\Gamma(T) + i\omega)^{-0.54}$ used for cuprates and ruthenates.

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The weakly helimagnetic metal MnSi ($T_C = 29.5$ K) has been the subject of intensive studies during the last 40 years. In the helimagnetic phase the resistivity has a $T^2$ dependence, which has been explained as resulting from a coupling of the charge carriers to spin fluctuations$[1]$. Recently interest has shifted to the quantum phase transition $[2, 3]$ at a critical pressure of 14.6 kbar where the Curie temperature becomes zero. The temperature dependence of the resistivity outside the magnetically ordered region, at high pressures, has been found to be proportional to $T^{3/2}$ in temperature range far larger than that predicted by the so-called nearly ferromagnetic Fermi-liquid theory (an extension of the Fermi-liquid picture)$[3]$. This fact has suggested the non-Fermi liquid nature of MnSi in the normal state $[2, 3]$. Despite these efforts in understanding the physics behind MnSi, few attempts have been made to determine and understand its optical properties. Measurements below $T_C$ of

![FIG. 1: DC resistivity as a function of temperature (solid curve). The open symbols represent $\rho_F(T) = (1/\rho(0) + 1/(AT))^{-1}$ with $\rho(0) = 286 \mu \Omega \text{cm}$ and $A = 1.62 \mu \Omega \text{cmK}^{-1}$. Top left inset: DC resistivity below 30 K (dots) and fit to $\rho_F(T) = \rho(0) + AT^\mu$. Lower right inset: Temperature dependence, $\mu(T)$, of the exponent in $\rho(T) = \rho(0) + AT^\mu$ (solid curve). The open symbols represent $d\ln \rho_F/d\ln T$, where $\rho_F(T)$ is the same function as in the main panel.

![FIG. 2: a) Grazing reflectivity at 80° angle of incidence measured at 10 and 300 K b) Expanded view of the reflectivity below 300 cm$^{-1}$. c) Real and imaginary part of the dielectric function in the visible part of the spectrum measured with spectroscopic ellipsometry.](image-url)
FIG. 3: a) Optical conductivity at four different temperatures (solid lines) and components of the non-Drude plus Lorentz oscillators fit at 300 K (dotted lines). b) Measured DC resistivity, and DC resistivity obtained by extrapolating the experimental $\sigma(\omega)$ using a Drude-Lorentz fit (stars) and using Eq. 3 (open circles). The fit-parameters are presented in Fig. 5.

FIG. 4: a) Effective mass (Inset: Behavior below 200 cm$^{-1}$ for 100, 75, 30, 20, 10 K starting from below) and (b) frequency-dependent-scattering rate (Inset: Behavior below 200 cm$^{-1}$ for 100, 75, 30, 20, 10 K starting from above) in MnSi as obtained from $\sigma(\omega)$ at different temperatures. Symbols represent the experimental data and thick lines the calculation from the non-Drude fit described in the text. The solid points at the left show $\rho_{DC}/(4\pi)$. The inset of the lower panel shows also the expected frequency dependence in the Fermi liquid theory calculated from Eq. 2 (dotted lines). This dependence is not compatible with our measured $1/\tau(\omega)$ and cannot be explained with errors coming from the measured reflectivity as shown by the error bars.

FIG. 5: Parameters for the non-Drude optical conductivity, Eq. 4 obtained by fitting the complete set of data (reflectivity, ellipsometry and DC resistivity): a) $\Gamma$, b) $\Omega$, c) $\omega_p$ and d) $\eta$. e) Temperature dependence of $\Gamma$ (see text). Inset: Temperature dependence of $\Gamma^{0.54}$ (circles) and of $\rho_{DC}/(4\pi\Omega^2\pi^2)$ (solid line).

dependent scattering rate, and the effective mass deviate from the behavior expected for Fermi liquids which can be described with an expression for $\sigma(\omega)$ that departs from the usual Drude model.

Single crystals were grown using the travelling floating zone technique. The temperature dependence of the resistivity is shown in Fig. 5. Fitting the resistivity to the equation $\rho(T) = \rho(0) + A T^\mu$ in the temperature interval 4K to 23K, we obtain $\rho(0) = 1.85\mu\Omega\text{cm}$, $A = 0.021\mu\Omega\text{cmK}^{-\mu}$, and $\mu = 2.1$. The resistivity increases more rapidly in the region between 23 K and the phase transition. For $T > 30$K the resistivity fits to the formula $\rho_p(T) = (1/\rho_\infty + 1/((AT)^\mu)^{-1}$ with $\rho_\infty = 286\mu\Omega\text{cm}$ and $A = 1.62\mu\Omega\text{cmK}^{-\mu}$. The remarkable accuracy of this description is further confirmed by the logarithmic derivative shown in the inset of Fig. 5. The tendency of the resistivity toward saturation at a value $\rho_\infty$ for $T \to \infty$ is in agreement with Calandra and Gunnarsson’s result that the resistivity saturates when the mean free path $l = 0.5n^{1/3}d$ (roughly the Ioffe-Regel limit), where $n$ is the density of the electrons and $d$ is lattice parameter. Also this indicates that, if the temperature saturation would be absent, the resistivity would be proportional to $T^\mu$, where the exponent $\mu = 1.0$ with
a very high accuracy. These observations stand in stark contrast to the $T^{5/3}$ temperature dependence predicted from the model of spin-fluctuations in itinerant electron magnetism.\[^{1}\] Yet the overall temperature dependence, and the strong reduction of $\rho(T)$ below $T_C$ indicate a dominant electronic (or spin) contribution to the scattering mechanism.

Grazing incidence reflectivity was measured in the range 20 to 6000 cm\(^{-1}\) using a Bruker 113v FT-IR spectrometer (see Fig. 2a and 2b). The temperature dependence was measured using a home-built cryostat, the special construction of which guarantees the stable and temperature independent optical alignment of the sample. The intensities were calibrated against a gold reference film which was evaporated in situ without repositioning or rotating the sample-holder. In the range 20 to 100 cm\(^{-1}\) we measured the temperature dependence of the grazing reflectivity with 0.5 K intervals below 50 K and 2 K intervals above 50 K. The complex dielectric function in the range 6000 to 36000 cm\(^{-1}\) was measured with a commercial (Woollam VASE32) ellipsometric spectrometer for the same set of temperatures as the grazing infrared reflectivity or rotating the sample-holder. In the range 20 to 6000 cm\(^{-1}\) was measured with a commercial (Woollam VASE32) ellipsometric spectrometer (see Fig. 2a and 2b). The temperature dependence was calculated from the complete data set (grazing infrared reflectance and visible ellipsometry) using Kramers-Kronig relations, following the procedure described in Ref. 7. Below 20 cm\(^{-1}\) the reflectivity data were extrapolated to fit the experimentally measured DC conductivities. The optical conductivity is shown for some temperatures in Fig. 3.

The first remarkable feature in the spectra is the similarity of the optical conductivity to the response of heavy fermion systems.\[^{5}\] In those materials, $\sigma(\omega)$ has almost no temperature dependence down to a frequency of $\sim$10 cm\(^{-1}\) and, below this frequency, a narrow mode centered at zero frequency is formed.\[^{5}\] Similar behavior has also been noticed for $\alpha$-cerium in the mid-infrared frequency range. Following a common procedure in the study of the dynamic response of heavy fermion systems, we have calculated $1/\tau(\omega)$ and $m^*(\omega)/m$ from the optical conductivity using the extended Drude-model 6:

$$\sigma(\omega) = \frac{\omega_p^2}{4\pi} \frac{1}{1/\tau(\omega) - i\omega m^*(\omega)/m}$$

(1)

adopting the value $\omega_p/2\pi c = 18700$ cm\(^{-1}\) for the plasma frequency, motivated by the least square fits which we will discuss below. Fig. 4a indicates a significant mass renormalization at low frequencies which, at the lowest measured temperatures, shows no indication of reaching a frequency independent value. Previous de Haas-van Alphen experiments (at $T=0.35$ K)\[^{10}\] provided an average enhancement of 4.5 times the cyclotron mass, although values as high as 14 were observed for some of the orbits. This average value was found to be compatible with the enhancement of the linear coefficient of the heat capacity $\gamma/\gamma_0 = 5.2$ calculated from specific heat data of Ref. 11. In comparison, our data show at 10 K and at the lowest measured frequency an enhancement of 4, and an enhancement of 17 when we extrapolate the data to $\omega = 0$.

The second remarkable feature is the behavior of $1/\tau(\omega)$ (Fig. 4b). At high temperatures this quantity becomes frequency independent, as expected for a Drude peak. Already at 100 K $1/\tau(\omega)$ is no longer a constant. Approaching the phase transition $1/\tau(\omega)$ becomes strongly frequency dependent between 30 and 300 cm\(^{-1}\) and it follows approximately a linear frequency dependence in this frequency range. In contrast, other correlated systems, such as heavy fermions \[^{6,8}\] and perovskite titanates \[^{12}\], show a frequency-dependent scattering rate with an $\omega^2$ dependence at low frequencies. Indeed the theory of Fermi liquids \[^{13}\] predicts

$$1/\tau(\omega, T) = 1/\tau_0 + a(\hbar \omega)^2 + b(k_B T)^2$$

(2)

with $b/a = \pi^2$. The same expression was obtained by Millis and Lee considering the Anderson lattice model \[^{14}\], and qualitatively similar behavior has been calculated by Riseborough in the context of spin-fluctuations.\[^{15}\] MnSi has also a $T^2$ dependence of the DC resistivity below $T_C$ and the corresponding expected frequency dependence of $1/\tau(\omega)$ is plotted in the inset of the lower panel of Fig. 4 (dotted lines) for 10 and 20 K. There is a mismatch with the experimental $1/\tau(\omega)$, both in absolute value and the observed trend, which is outside the experimental error bars. However it can not be excluded, that at an even lower frequency the experimental $1/\tau(\omega)$ would cross over to a $\omega^2$ dependence.

Above, we have pointed out various striking results in the optical response of MnSi. In order to understand their nature, let us take a closer look at the low frequency data. From 300 to 75 K, $1-R_p(\omega)$ follows a $\omega^{1/2}$ behavior (see Fig. 2). This can be easily understood from the fact that at low frequencies, from the Fresnel formulae, $R_p$ can be written approximately as:

$$R_p = 1 - \frac{2\omega^{1/2}}{\cos \theta} \text{Re} \left[ \frac{1}{\sqrt{\pi} \sigma(\omega)} \right]$$

(3)

where $\theta$ is the angle of incidence. In the case that $\sigma_1$ is constant and $\sigma_2$ goes to zero, this expression reduces to the well known Hagen-Rubens law. In the Drude picture this corresponds to the frequency range where the scattering rate is larger than $\omega$. In contrast, below 75 K our measured $R_p$ does not follow a $\omega^{1/2}$ behavior. Combining the Drude model with the Fresnel equations for reflectivity, a plateau in the reflectivity is expected for intermediate frequencies (frequencies larger than the scattering rate but much lower than the plasma frequency). To check this more closely we measured $R_p$ below 100 cm\(^{-1}\)
in a finer temperature mesh. Our results show no sign of
a plateau, instead $1 - R_p(\omega)$ evolves gradually to a linear
frequency dependence when $T$ is lowered. We can then
conclude that either the peak centered at zero frequency
departs from the Drude picture or other modes appear
at low temperatures and at low frequencies.

To distinguish between these alternatives we have fit-
tted, simultaneously, the measured reflectivity, ellipsome-
try and resistivity with two models. First we modelled
the data with a Drude peak and a set of oscillators.
In this case, the fit fails to reproduce the measured DC re-
sistivities at low temperatures (stars in Fig. 3b). On the
other hand, if we give more fitting weight to $\rho_{DC}$, the re-
sult is a poor fit of $R_p$ at low frequencies. Although this
can in principle be remedied by introducing an arbitrary
number of oscillators at frequencies below 100 cm$^{-1}$,
the infrared properties together with the DC resistivity can
be summarized in an economical way (i.e. requiring a
minimal set of adjustable parameters) when we replace
the Drude formula with [16]

$$\sigma(\omega) = \frac{\omega_p^2}{4\pi(\omega + i\Gamma)^{1-2\eta}(\omega + i\Omega)^{2\eta}}$$  (4)

which for $\eta = 0.25$ corresponds to the model by Ioffe
and Millis[17], and for $\Gamma \ll \omega \ll \Omega$ to Anderson's [18]
power-law formula $\sigma(\omega) \propto (i\omega)^{2\eta-1}$, both in the
context of the optical conductivity of the cuprate high $T_c$
superconductors. Eq. 4 in the case $\Omega \gg \omega$, has been shown to
describe the optical conductivity of SrRuO$_3$, below 40 K,
in the range [6-2400]cm$^{-1}$ with $\eta = 0.3$ [19]. For SrRuO$_3$
this behavior has been justified as arising from the
coupling of electrons to orbital degrees of freedom [20], and
in the context of the discrete filamentary model of charge
transport [21].

Our new fit, non-Drude plus Lorentz oscillators (whose
individual components at 300 K are displayed in Fig. 3b),
gives the same overall result at high temperatures ($T > 75$
K) as the Drude fit. However, at low temperatures, the
non-Drude equation gives a better fit at low frequencies
and, what is more important, reproduces $\rho_{DC}$ at all
temperatures (open symbols in Fig. 3b). Therefore, we con-
clude that the low frequency optical response of MnSi is
best described by Eq. 4. From the fit we can extrapolate
the optical properties to lower frequencies (insets of Fig.
3). The extrapolation shows that at 10 K, for $\omega \rightarrow 0$,
$m^*(\omega)/m = 17$, with a gradual decay as a function of
increasing frequency. Similarly, $1/\tau(\omega)$ is approximately
proportional to $\omega$ in the frequency range below 300 cm$^{-1}$.
Above $T_C$, it has a weak $\omega^2$ frequency dependence.

Now let us analyze the parameters of the non-Drude
conductivity as provided by the fit to Eq. 4 These values
are summarized in Fig. 5b-d where the error bars represent
the interval of confidence calculated for a vari-
ation of 1% of $\chi^2$. Within those error bars, the param-
eters $\omega_p$, $\Omega$ and $\eta$ are temperature independent, which
contrasts with the strong temperature dependence of $\Gamma$.

This has another interesting consequence in connection
with the DC resistivity. From Eq. 4 we can easily see that $\rho_{DC} = 4\pi\omega_p^2\Omega^{2\eta}\Gamma^{1-2\eta}$, but since $\omega_p$, $\Omega$ and
$\eta$ are temperature independent, $\rho_{DC}(T) \propto \Gamma(T)^{1-2\eta}$. For
our sample, using the values of $\omega_p = 18867$ cm$^{-1}$,
$\Omega = 2049$ cm$^{-1}$ and $\eta = 0.23$ (from Fig. 5), we obtain
$\rho_{DC} = 6.02\Gamma^{0.54}$ [\mu\Omega\cm]. Recently, Dodge et al. [19]
have emphasized a similar non-linear relationship be-
tween the DC resistivity and the parameter $\Gamma$ in the case
of the weak itinerant ferromagnet SrRuO$_3$. The con-
clusions for SrRuO$_3$ have been questioned recently by
Capogna et al. [22], who argued that the true tempera-
ture dependence of the optical properties may have been
masked by the large residual resistivity of the sample
used in Ref. 19. In the present work this problem is ab-
sent due to the low residual resistivity of single crystalline
MnSi. In fact, we can go a step further and try to give
a detailed picture of the temperature dependence of $\Gamma$.
For that purpose we fit the low frequency $R_p$ (at all the
measured temperatures) to Eq. 4 using the known values
of $\omega_p$, $\Omega$ and $\eta$. The values obtained for $\Gamma$ are displayed
in Fig. 5. The inset shows $\Gamma^{0.54}$ and $\rho_{DC}/6.02$. We
can see that the model represented by Eq. 4 describes
the measured data (reflectivity and resistivity) down to
approximately 20 K. Below this temperature a fit only
to reflectivity produces unphysical negative values for $\Gamma$.
However, introducing the measured $\rho_{DC}$ produces a $\chi^2$
which is not more than twice that obtained when fitting
only reflectivity. Apparently at low temperatures there
are still details which Eq. 4 is not able to describe.

At low frequencies, deviations from the Drude formula
of the optical conductivity have been seen accompanied
by deviations from $T^2$ in $\rho_{DC}$. Well known examples are
YBCO [25] and more recently CaRuO$_3$ [21]. Therefore,
a departure from Drude behavior has been usually con-
sidered as evidence against Fermi-liquid behavior. Here,
for MnSi, we are confronted with an atypical case. The
resistivity has a quadratic temperature dependence, but
the optical conductivity is better described by Eq. 4
with $\eta \approx 0.23$, a clear departure from the Drude for-
mulation. Moreover, instead of an $\omega^2$-type frequency depen-
dent scattering rate, which is usually observed in strongly
interacting Fermi-liquids [7, 8], here $1/\tau(\omega)$ has a linear
frequency dependence. Although Eq. 4 summarizes in a
compact way the low frequency optical response, differ-
ing in a fundamental way from conventional Drude behavior,
it's microscopic origin is as yet not fully understood.

For frequencies below 300 cm$^{-1}$ and for $T < 100$ K the
situation can be summarized as follows: (i) $m^*/m$
decreases from 17 to 1 as temperature and frequency are
increased. (ii) Phenomenologically the DC conductivity
and the optical conductivity follow $\sigma \propto (\Gamma(T) + i\omega)^{-0.5}$. In
this formulation $\Gamma(T) \propto T^4$ below $T_C$, whereas above $T_C$
the temperature temperature dependence is approxi-
ately linear. (iii) For $T > T_C$ the scattering rate
$1/\tau(\omega, T)$ is proportional to $T$ and $\omega^2$ in contradiction
with the theory of weak itinerant ferromagnetism. For $T < T_c$ the scattering rate is proportional to $T^2$ and $\omega$. Given the frequency range for this type of measurements, we can not exclude the possibility, that for frequencies below 30cm$^{-1}$ the scattering rate crosses over to the Fermi-liquid result $1/\tau \propto \pi^2 T^2 + \omega^2$.

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