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Heavy carriers, non-drude optical conductivity and transfer of spectral weight in MnSi

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Abstract

The optical properties of the weak magnetic metal MnSi were determined using reflectance at 80° (2–800 meV) and ellipsometry (0.8–4.5 eV). At low frequencies in the magnetic phase we observe a departure of the optical conductivity from Drude behavior: $m^*(\omega)/m$ is strongly frequency dependent and $1/\tau(\omega)$ is approximately linear in frequency. In fact, we show that $\sigma(\omega)/\sigma(0) = (1 + i\omega/\Gamma)^{-0.5}$. Moreover, in the magnetic phase, the plasma frequency shifts to the red indicating that spectral weight is transferred to high frequencies. This is opposite to the effect recently seen in other magnetic compounds.

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MnSi, a helimagnetic metal ($T_C = 29.5\,K$), has been the subject of intensive studies during the last 40 years. Several of its properties have been described as a result of spin fluctuations (SF) [1], in particular the T^2-dependence of the resistivity in the magnetic phase. However, the same theory predicts a $T^{5/3}$-dependence above T_C, which is not seen [2]. Recently, attention has been given to the quantum phase transition [3] at $p_c = 14.6\,kbar$, where T_C becomes zero. Such studies have suggested the non-Fermi liquid nature of MnSi in the normal state [3]. Despite these efforts, few attempts have been made to determine its optical properties. Here we present a detailed study of the optical response of MnSi. In the magnetic phase, at low frequencies, we show that the frequency dependent scattering rate and the effective mass deviate from the behavior expected for Fermi liquids. This can be understood from the fact that the optical conductivity is best described with an expression that departs from the usual Drude model. Another interesting aspect is the presence of a large transfer of spectral weight between low and high frequencies. This effect is opposite to that seen recently in other magnetic materials [4].

Single crystals of MnSi were grown using the floating zone method. The temperature dependence of the p-polarized grazing reflectivity at 80° (R_P) was measured from 20 to 6000 cm^{-1} with the intensities calibrated against gold evaporated in situ. From 20 to 100 cm^{-1} we measured R_P with 0.5 and 2 K intervals below and above 50 K, respectively. The dielectric function, $\varepsilon(\omega) = \varepsilon'(\omega) + i(4\pi/\omega)\sigma_1(\omega)$, in the range 6000–36,000 cm^{-1} was measured with a commercial ellipsometer from 4 to 300 K every 0.5 K. From the complete data set $\sigma(\omega)$ was obtained from 20 to 36,000 cm^{-1} at several temperatures using Kramers–Kronig relations. Remarkably, $\sigma(\omega)$ of MnSi is similar to the response of heavy fermion systems. Therefore, following a common procedure in the study of those systems, we have calculated the frequency dependent scattering rate, $1/\tau(\omega)$, and the effective mass, $m^*(\omega)/m$, from $\sigma(\omega)$

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using the extended Drude-model with $\omega_P = 18700\,\mathrm{cm}^{-1}$ (see below). Fig. 1a indicates at 10 K, at the lowest frequency, an enhancement of 4, and 17 when we extrapolate the data to $\omega = 0$. In comparison, De Haas–van Alphen experiments (at $T = 0.35\,\mathrm{K}$) [5] give an *average* enhancement of 4.5 times the cyclotron mass, although values as high as 14 were observed for some of the orbits. The behavior of $1/\tau(\omega)$ (Fig. 1b) is also remarkable. At high temperatures this quantity becomes frequency independent, as expected for a Drude peak but already at 100 K it is no longer constant. Approaching the phase transition, $1/\tau(\omega)$ becomes strongly frequency dependent between 30 and $300\,\mathrm{cm}^{-1}$ following approximately a linear frequency dependence. In contrast, theory predicts $1/\tau(\omega, T) = 1/\tau_o + a(\hbar\omega)^2 + b(k_B T)^2$ with $b/a = \pi^2$ for heavy fermions [6] and $b/a = (2\pi)^2$ for SF [7]. For 10 K these two possibilities are plotted in the inset of the lower panel of Fig. 1. A deviation from these predictions is evident.

Our results below $100\,\mathrm{cm}^{-1}$, show a gradual change in $1 - R_P(\omega)$ from the Hagen–Rubens law, $\omega^{1/2}$, to a linear frequency dependence when T is lowered. However, combining the Drude model with the Fresnel equations as the scattering rate decreases, a change from $\omega^{1/2}$ to a plateau is expected. From a fit to the measured quantities it turns out that the infrared properties together with the DC resistivity can be summarized in an economical way when we replace the Drude formula with $\sigma(\omega) = (\omega_P^2/4\pi)\mathrm{i}/([\omega + \mathrm{i}\Gamma]^{1-2\eta}[\omega + \mathrm{i}\Omega]^{2\eta})$ [8]. Such a fit gives $\omega_P \sim 18,700\,\mathrm{cm}^{-1}$, $\Omega \sim 2000\,\mathrm{cm}^{-1}$, $\eta \sim 0.25$ and a strong T-dependence of Γ. This equation has been also

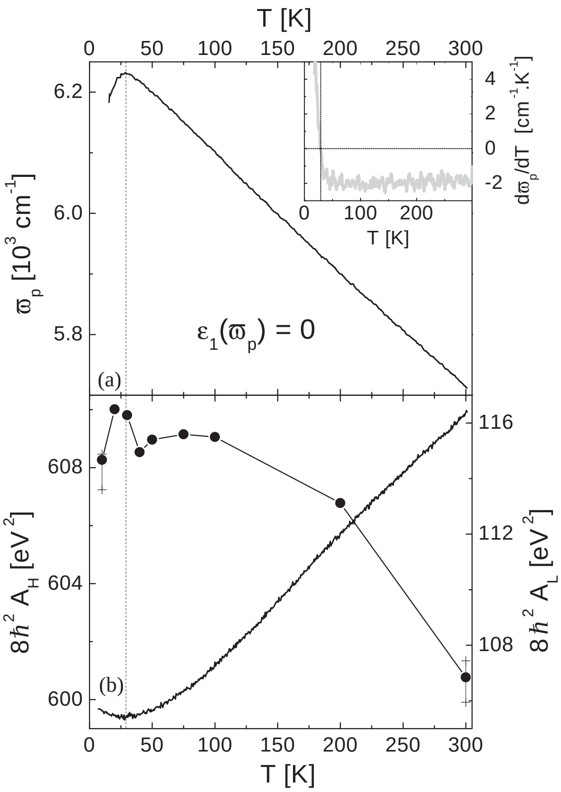

Fig. 2. (a) Plasma frequency and (b) spectral weight of $\sigma(\omega)$ above and below $6000\,\mathrm{cm}^{-1}$.

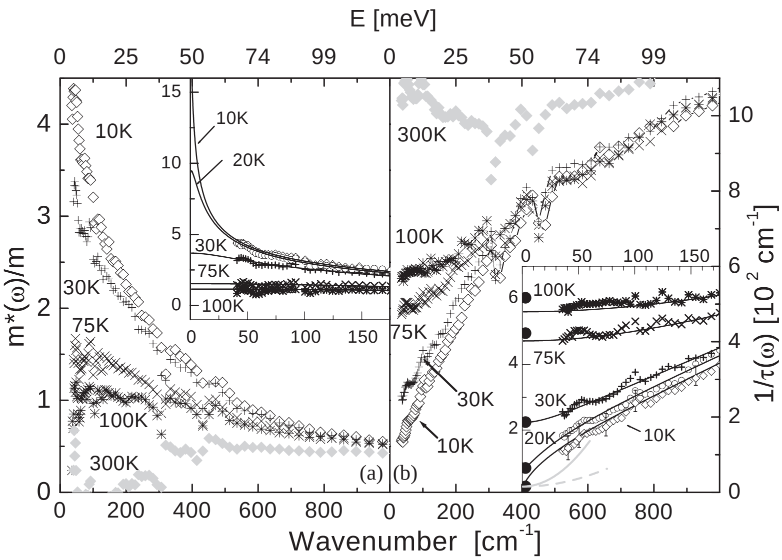

Fig. 1. (a) $m^*(\omega)/m$ and (b) $1/\tau(\omega)$ at different temperatures (insets: same quantities below $200\,\mathrm{cm}^{-1}$). Symbols: experimental data. Lines: calculation from the non-Drude fit. The inset of the right panel also shows $\rho_{DC}\omega_P^2/(4\pi)$ (solid points) and the expected frequency dependence for heavy fermions (gray solid line) and for spin fluctuations (gray dashed line) at 10 K.

shown to describe $\sigma(\omega)$ of $SrRuO_3$, below 40 K, in the range 6–2400 cm^{-1} with $\eta = 0.3$ [9].

Another important aspect in MnSi is the behavior of the high frequency response. From the ellipsometric data we have obtained the temperature dependence of ϖ_P defined as $\varepsilon_1(\varpi_P) = 0$ (Fig. 2a). From 300 K to T_C, ϖ_P shifts to the blue but below T_C shifts to the red (see also the first derivative of $\varpi_P(T)$ in the inset of Fig. 2a). This shift is the manifestation of a transfer of spectral weight from high to low frequencies. To confirm this, we have also calculated A_L and A_H, the spectral weight of the optical conductivity for frequencies below and above $W = 6000\,cm^{-1}$, respectively. W is the frequency where a minimum in $\sigma(\omega)$ occurs. The results (Fig. 2b) indicate a decrease of A_H as the temperature is lowered down to T_C. From that temperature, there is a small recovery of the spectral weight. Correspondingly, A_L shows the opposite trend. This is in contrast with the behavior seen in other magnetic materials such as $Ga_{1-x}Mn_xAs$, manganites and EuB_6 [4].

In conclusion, we have shown that for $\omega < 300\,cm^{-1}$ and $T < 100$ K, m^*/m increases as temperature and frequency are decreased. In the same range, $1/\tau(\omega)$ becomes frequency dependent and at the lowest temperatures is approximately linear. Phenomenologically this can be explained with the fact that the DC and optical conductivity follow $\sigma \propto (\Gamma(T) + i\omega)^{-0.5}$. Finally we presented a spectral weight transfer opposite to that seen in other magnetic materials.

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