Optical evidence for heavy charge carriers in FeGe

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The optical spectrum of the cubic helimagnetic metal FeGe has been investigated in the frequency range from 0.01 - 3.1 eV for different temperatures from 30 K to 296 K. The optical conductivity shows the evolution of a low energy (0.22 eV) interband transition and the development of a narrow free carrier response with a strong energy and temperature dependence. The frequency dependent effective mass and scattering rate derived from the optical data indicate the formation of dressed quasi-particles with a mass renormalization factor of 12. Similar to FeSi the spectral weight in FeGe is not recovered over a broad frequency range, an effect usually attributed to the influence of the on-site Coulomb interaction.

Cubic FeGe is a good metal at low temperature, which undergoes a transition to helimagnetic order\(^1\) at \(T_C = 280\) K with the magnetic moment at the iron sites of \(1\mu_B\). The helix changes its orientation in a temperature interval \(T_2 = 20\) K and shows pronounced temperature hysteresis\(^2\) between 211 K and 245 K. This material crystallizes in the \(B20\) structure and the cubic space group \(P2_13\) lacking a center of symmetry which is responsible for this long range order. The iso-electronic compound FeSi has the same crystal structure. It has a large magnetic susceptibility at room temperature, which vanishes as the temperature approaches zero due to a small (70 meV) semiconductor gap at \(E_F\). A continuous series FeSi\(_{1-x}\)Ge\(_x\) can be formed, where the metal insulator transition\(^3\) occurs for \(x \approx 0.25\). Theoretical models which have been proposed to explain this behavior, invoke disorder\(^4\), narrow bands and different ways of incorporating electron correlations\(^5,6,7,8\). The temperature dependent closing of the gap has been explained as a result of a correlation gap using a two-band Hubbard model\(^9,10\), and excellent agreement was obtained with optical data\(^10,11,12\) but it has been shown that vibrational disorder, if sufficiently strong, also closes the gap\(^13\).

Anisimov et al. have predicted a magnetic-field driven semiconductor to metal transition in FeSi\(_{1-x}\)Ge\(_x\), and argue that the difference in electronic structure between FeSi and FeGe in essence consists of a rigid relative shift of the majority and minority spin bands for the latter material. According to this model the optical spectra at low energies is expected to be the superposition of a Drude peak and an interband transition across an energy range corresponding to the forementioned relative shift of the majority and minority bands. Experimentally relatively little is known about the electronic structure of FeGe, for example no optical data have been published.

Here, we report optical measurements on a cubic FeGe single crystal at different temperatures. The real and imaginary parts of the dielectric function were derived from the reflectivity and ellipsometry measurements. Optical spectra of FeGe reveal the presence of an important interband transition at 0.22 eV and an unusual dynamics of the free carrier charge. In order to clarify the behavior of the optical conductivity the data were compared with local spin-density approximation (LSDA) calculations of the electronic structure.

Cubic FeGe single crystals were grown by chemical vapor transport method as described in detail in Ref.\(^[12]\). The single crystals were characterized by transport, magnetic and thermodynamic measurements. It was found that a first order phase transition to the helimagnetic state occurs at \(T_C = 280\) K. Optical properties of FeGe were obtained using spectroscopic ellipsometry at 0.75 - 3.1 eV and near normal incidence reflectivity spectra were measured in the energy range from 0.01 to 0.85 eV for different temperatures from 30 to 296 K. The complex dielectric function \(\epsilon(\omega) = \epsilon_1(\omega) + 4\pi i\sigma_1(\omega)/\omega\) on a broad frequency spectral range between 0.01 - 3.1 eV was calculated by combining the two sets of data and using a variational Kramers-Kronig constrained analysis\(^[10]\).

Figure\(^1\) shows the reflectivity \(R(\omega)\), the optical conductivity \(\sigma_1(\omega)\), and the dielectric function \(\epsilon_1(\omega)\) of single crystalline cubic FeGe at a series of temperatures from 30 to 296 K over a broad frequency range. Absolute values of the reflectivity were obtained by calibrating the instrument using a gold-layer deposited in-situ on the sample surface, without breaking the vacuum and without moving the sample. This calibration procedure is designed to fully compensate the frequency dependence of the instrument and the geometry of the sample. Repeated experiments showed that two weak features at 400 cm\(^{-1}\).
and 530 cm$^{-1}$ are not reproducible. They coincide with the frequencies of strong interference fringes of the thin polyethylene cryostat window. The fact that they are not fully removed by the calibration procedure is due to a gradual evolution of the window properties during the time (several hours) lapsed before and after depositing the gold layer on the crystal.

We observe that at low frequency the reflectivity increases with decreasing temperature. In contrast, the range between 500 cm$^{-1}$ and 1500 cm$^{-1}$ $R(\omega)$ is strongly suppressed and a dip develops at low temperature. The two distinct sharp excitations in the far infrared spectrum (230 cm$^{-1}$, 290 cm$^{-1}$) are due to optically active phonons. Consequently the far infrared region of the optical conductivity spectrum displayed in Fig. 1b is strongly temperature dependent. At low temperature $\sigma_1(\omega)$ shows a minimum around 0.1 eV, which vanishes at $T_C$. Below $T_C$ a peak in the optical conductivity appears at 0.22 eV which looks like an onset of interband transitions. We observe a narrowing of the free carrier response while the temperature is lowered to zero. However a large finite conductivity remains below the interband transition at 0.22 eV which appears to be part of free carrier contribution. The narrowing of the free carrier response signals a strong reduction of the scattering rate, whereas the simultaneous appearance of an interband transition is similar to observations in Kondo-lattices like URu$_2$Si$_2$, CeAl$_3$, CeCoIn$_5$ and CeIrIn$_5$.

The dielectric function (see Fig. 1c) has a zero-crossing at 0.4 eV for all measured temperatures, which we assign to the plasma resonance of the conduction electrons. A second zero crossing occurs below 150 K. This low frequency crossing is strongly temperature dependent and is shifting toward lower energy as the temperature is decreasing. Such a line shape of $\varepsilon_1(\omega)$ resembles the heavy fermion systems, where the low frequency plasmon is a characteristic feature of the heavy quasiparticles.

We calculated the electronic structure using the linear muffin-tin orbital (LMTO) code with the self-consistent LSDA method, resulting in a ferromagnetic groundstate with a magnetic moment of $1\mu_B$ per Fe-atom. The density of states, shown in Fig. 2 is consistent with the schematic density of states of Anisimov et al. (see Fig. 3 of Ref. 14). The theoretical optical conductivity has been calculated as a sum of all band transitions within 2.7 eV of $E_F$ at 9216 k-points of the irreducible Brillion zone, including the dipole matrix elements. The effect of thermal disorder and zero-point motion on the band structure is introduced as a band broadening, which is assumed to be equal for all bands. The result of disorder is also a smearing of the spectra at higher energies. The parameter for the band broadening is estimated from calculations of disordered supercell calculations for FeSi and FeGe.

The comparison between the calculated and measured optical conductivity (see Fig. 3) is not as good as for CoSi and FeSi. Theoretically we find the onset weak interband transition at an energy as low as 80 meV followed by a gradual increase of the optical conductivity to a maximum of 0.4 eV. The experimental data show a minimum at 0.1 eV followed by a peak at 0.22 eV. In addition two peaks are predicted at 1.6 eV and 2 eV. The experimental high frequency optical spectra is less structured compared to the calculated one. The reason could be that at high energy the scattering is very large. We associate the experimental maximum at 0.22 eV with the theoretical peak at 0.4 eV and the higher energy structures to the theoretical peak predicted at 1.6 eV.
have varied the lattice parameter in the LSDA calculation to see if this would improve the agreement with the experimental data, but the position of the peak at 0.4 eV turned out to be robust.

In order to further analyze the low frequency behavior we use the extended Drude formalism. This model is only meaningful in the energy region where the optical response is due to mobile carriers and not to the bound ones. The strong temperature dependence of the optical data (Fig. 4b) for frequencies below 0.1 eV, naturally assigned to the mobile carriers, suggest that this model can be applied at frequencies lower than 0.1 eV. According to this formalism the scattering rate $1/\tau(\omega)$ and the effective mass $m^*(\omega)/m$ are represented as follows

$$m^*(\omega)/m + i \omega\tau(\omega) = \frac{\omega_p^2}{\omega^2(\epsilon_{\infty} - \epsilon(\omega))},$$

where $h\omega_p = 4.4eV$ is the total Drude plasma frequency and $\epsilon_{\infty} = 60$ is the high-frequency dielectric constant, due to the bound charge polarizability. The value of $\omega_p$ was obtained by integrating $\sigma_1(\omega)$ up to the onset of interband absorption. Since there is not a complete separation between the intraband and interband responses (see Fig. 1b) the value of $\omega_p$, determined in this way is somewhat ambiguous. However, the choice of $\omega_p$ does not affect the frequency dependence of $\tau(\omega)$ and $m^*(\omega)$.

Figure 4 displays the spectra of scattering rate and effective mass as a function of frequency for different temperatures. Note that at room temperature both the scattering rate and the effective mass are almost constant. As the temperature approaches the transition temperature, $1/\tau(\omega)$ shows strong frequency and temperature dependence. The value of the scattering rate at high temperature becomes quite large and it seems difficult to describe the metallic state of this material. On the other hand, at low temperature $1/\tau(\omega)$ is much smaller than at room temperature. This behavior of the scattering rate reflects the narrowing of the zero frequency peak. This suggests that the temperature dependence of the optical response shown in Fig. 1b cannot be explained in terms of the simple Drude model. In addition we notice a strong suppression of the scattering rate upon cooling, like in MuSi$^{22}$ and in the heavy fermion compounds URu$_2$Si$_2$,$^{17}$ CeAl$_5$,$^{18}$ CeCoIn$_5$ and CeIrIn$_5$,$^{19}$ suggesting the development of heavy quasiparticles at low temperature. As a result a renormalized Drude absorption due to these heavy quasiparticles is observed in the optical spectra. In relation to this, at low frequency the electrons are dressed by interactions giving them a large effective mass. For $\omega \to 0$ we observe $m^*(\omega)/m \sim 12$. At high frequency the electrons are no longer dressed by the interaction and the effective mass reduces to the band mass as can be seen in Fig. 4b. In contrast to the case of heavy fermions systems, FeGe has no 4f electrons but the large mass renormalization factor appears to be of the same order of magnitude as that observed in CeCoIn$_5$ and CeIrIn$_5$. In this context we speculate that FeGe can be considered as a 3d heavy fermion system.

The half metallic ferromagnet chromium dioxide has an optical conductivity,$^{22}$ very similar to that of FeGe. A suppression of $1/\tau(\omega)$ was observed below $T_C$, which was stronger than in FeGe (Fig 4b). This is a natural consequence of the halfmetallic ferromagnetism: Spin-flip scattering, which is the dominant scattering mechanism in ferromagnets, is completely suppressed for frequencies smaller than the gap separating the minority bands from the Fermi level. Note, that in disordered Co-doped FeSi, which is also a half-metallic ferromagnet, the opposite behaviour is observed, in that the scattering rate increases in the magnetically ordered state.$^{23,24,25}$

In FeSi the spectral weight removed at frequencies below 70 meV due to the opening of a gap at low temperatures, was observed not to be recovered for energies up
to at least 6 eV\textsuperscript{11,22}. Since this implies the coupling of the conduction electrons to high energy scale excitations, possibly on the scale of on-site Coulomb interaction, it may be an indication that the material has features in common with a Kondo lattice, and the insulating gap of FeSi is due strong local electron correlation effects presumably in the 3d-shell of the iron atoms. In view of the structural and chemical similarities between FeSi and FeGe one might suspect electron correlation effects to be equally important in the latter material. While FeGe has no insulating gap at low temperatures, the optical spectra depend strongly on temperature. In Fig. 5 the function \( N_{\text{eff}}(\omega) = \frac{2m_e}{\pi \hbar^2} \int_0^\omega \sigma_1(\omega) d\omega \) is displayed. For \( \omega_c \to \infty \) this represents the total number of electrons per unit of FeGe. In the region below 800 cm\(^{-1}\) we observe at low temperatures that \( N_{\text{eff}}(\omega) \) increases more sharply as a function of frequency than the high temperature data, which is a consequence of the fact that \( 1/\tau \) is smaller and the Drude peak narrower at low temperature. However, all curves cross at 800 cm\(^{-1}\), and above 1500 cm\(^{-1}\) \( N_{\text{eff}}(\omega, 296\, K) \) exceeds \( N_{\text{eff}}(\omega, 30\, K) \) by a constant amount of \( \simeq 0.005 \) per FeGe formula unit.

In other words: Just like in FeSi, cooling down the sample results in a loss of spectral weight in the frequency range 0.1 eV, which is not recovered over a broad frequency range. While due to the smallness of the sample the data-noise does not permit a quantitative analysis of the spectral weight transfer for energies larger than 0.5 eV, the temperature dependence of \( \epsilon_1(\omega, T) \) for \( h\omega \approx 1 \) eV observed directly with ellipsometry confirms quantitatively the trends seen in Fig. 5, suggesting that in FeGe, just as in FeSi, spectral weight is redistributed over an energy range of order 1 eV or higher when the temperature is varied. This behaviour may have its origin in the temperature dependence of the electron correlations resulting from the Hund’s rule interaction on the Fe-atoms\textsuperscript{11,14}, in a change of character of the bands near \( E_F \) due to thermal disorder, or in a combination of these two.

In summary, we have reported the optical properties of single crystalline cubic FeGe, which undergoes a helimagnetic transition at 280 K. At the temperature where magnetic order occurs, a distinct and narrow free carrier response develops, with a frequency dependent scattering rate and a moderate mass enhancement in the zero-frequency limit. Similar behaviour as in FeSi is observed in FeGe, where the low energy (Drude) spectral weight appears to be transferred to higher energies, on an energy scale of at least 1 eV, when the material is cooled down. This, together with the observed frequency dependent mass enhancement, indicates the important role of electron correlations in these materials.

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\begin{figure}
\centering
\includegraphics[width=0.5\textwidth]{figure5}
\caption{(Color online) The integrated spectral weight of FeGe for a number of representative temperatures.}
\end{figure}

\begin{thebibliography}{99}
\bibitem{1} R. Wäppling and L. Häggström, Phys. Lett. A 28, 173 (1968).
\bibitem{2} B. Lebech, J. Bernard and T. Frehoft, J. Phys. Condens. Matter 1, 6105 (1989).
\bibitem{3} S. Yeo, S. Nakatsuji, A. D. Bianchi, P. Schlootmann, Z. Fisk, L. Balicas, P. A. Stampe, and R. J. Kennedy, Phys. Rev. Lett. 91, 046401 (2003).
\bibitem{4} T. Jarlborg, J. Magn. Magn. Mater 283, 238 (2004).
\bibitem{5} V. Jaecarino, G. K. Wertheim, J. H. Wernick, L. R. Walker and Sigurds Arags, Phys. Rev. 160, 476 (1967).
\bibitem{6} Y. Takahashi, and T. Moriya, J. Phys. Soc. Jpn. 46, 1451 (1979).
\bibitem{7} G. Aeppli and Z. Fisk, Comments Condens. Matter Phys. 16, 155 (1992).
\bibitem{8} D. Mandrus, J. L. Sarrao, A. Migliori, J. D. Thompson, and Z. Fisk, Phys. Rev. B 51, 4763 (1995).
\bibitem{9} V. I. Anisimov, S. Y. Ezhov, I. S. Elfimov, I. V. Solovyev, and T. M. Rice, Phys. Rev. Letters 76, 1735 (1996).
\bibitem{10} K. Urasaki and T. Saso, Phys. B 282, 313 (2000).
\bibitem{11} Z. Schlessinger, Z. Fisk, Hai-Tao Zhang, M. B. Maple, J. F. DiTusa and G. Aeppli, Phys. Rev. Lett. 71, 1748 (1993).
\bibitem{12} A. Damascelli, K. Schulte, D. van der Marel and A. A. Menovsky, Phys. Rev. B 55, R4863 (1997).
\bibitem{13} T. Jarlborg, Phys. Rev. B 59, 15002 (1999).
\bibitem{14} V. I. Anisimov, R. Hubina, M. A. Korotin, V. V. Mazurenko, T. M. Rice, A. O. Shorkov, and M. Sigrist, Phys. Rev. Letters 89, 257203 (2002).
\bibitem{15} M. Richardson, Acta Chem. Scand. 21, 2305 (1967).
\bibitem{16} A. B. Kuzmenko, Rev. Sci. Instrum 76, 083108 (2005).
\bibitem{17} D. A. Bonn, J. D. Garrett, and T. Timusk, Phys.Rev.Lett. 61, 1305 (1988).
\bibitem{18} A. M. Awasthi, L. Degiorgi, G. Grüner, Y. Dalichaouch, and M. B. Maple, Phys. Rev. B 48, 10692, (1993).
\bibitem{19} F. P. Mena, D. van der Marel, J. L. Sarrao, Phys. Rev. B 72, 045119 (2005).
\bibitem{20} P. Pedrazzini, H. Wilhelm, D. Jaccard, T. Jarlborg, M.
\end{thebibliography}
Schmidt, M. Hanfland, L. Akselrud, H. Q. Yuan, U. Schwarz, Yu. Grin, and F. Steglich, Phys. Lett. 98, 47204 (2007).

21 F.P. Mena, (thesis), Univ. of Groningen, (2004).

22 E. J. Singley, C. P. Weber, and D. N. Basov, A. Barry, J. M. D. Coey, Phys. Rev. B 60, 4126 (1999).

23 N. Manyala, Y. Sidis, J. F. DiTusa, G. Aeppli, D. P. Young, and Z. Fisk, Nature (London) 404, 581 (2000).

24 N. Manyala, Y. Sidis, J. F. DiTusa, G. Aeppli, D. P. Young, and Z. Fisk, Nat. Mater. 3, 255 (2004).

25 F. P. Mena, J. F. DiTusa, D. van der Marel, G. Aeppli, D. P. Young, A. Damascelli, and J. A. Mydosh, Phys. Rev. B 73, 085205 (2006).