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Scaling between magnetization and Drude weight in EuB_6

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The low-temperature optical response of EuB_6 is strongly influenced by a ferromagnetic phase transition with a zero-field onset temperature T_C of $\sim$16 K. A marked blueshift of the reflectivity plasma edge with either decreasing temperature or increasing magnetic field is observed. The corresponding temperature and field-dependent plasma-frequency shift, combined with low-field Hall data, reveals a distinct reduction of the effective mass of the itinerant charge carriers, in parallel with the onset of ferromagnetic order. A universal scaling linking the experimental data on magnetization and the Drude weight is established.

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The spontaneous polarization of itinerant electronic moments in a metal may be viewed as a competition between the single-particle kinetic energy, favoring the paramagnetic state, and the Coulomb-interaction-based exchange energy, favoring the spin-polarized state.[1] This point of view is inherent in the conventional Stoner model,[2] where, paraphrasing the above concept, the gain in exchange energy upon spin polarization more than compensates for the cost in kinetic energy, which results from the Pauli principle. It is still debated which conditions favor ferromagnetism in metals. Key issues in this puzzle include ferromagnetic transition metals,[3] weak metallic ferromagnets,[4] and colossal magnetoresistance.[5]

New aspects of ferromagnetism in metals have possibly been found in experimental results probing hexaborides with divalent alkaline-earth and rare-earth metals, as cation elements.[6] Here, we focus on EuB_6, which orders ferromagnetically at low temperatures ($T_C \sim 16$ K) and exhibits large magnetoresistance.[7] Because of similarities of this latter feature with observations on transition-metal oxides,[8] the ordered state of EuB_6, due to the local $4f$ electron moments of the Eu^{2+} ions,[9] is of particular interest. In the paramagnetic state the temperature dependence of the electrical resistivity is of metallic character but the concentration of itinerant charge carriers is low.[10,11] It is, therefore, not *a priori* clear whether the spontaneous alignment of the localized moments is simply due to the usual Ruderman-Kittel-Kasuya-Yoshida (RKKY) interaction or whether other coupling mechanisms might be of significance.

Optical experiments offer insight into the microscopic electronic properties of materials. If EuB_6 is cooled through the ferromagnetic transition at T_C its optical reflectivity displays a giant blueshift of the plasma edge,[10] indicating a sizable increase of the charge-carrier concentration and/or a reduction of the effective mass of the itinerant charge carriers. Hall-effect data suggest that simultaneously with the onset of magnetic order the effective charge-carrier concentration increases significantly.[11] The concomitant loss of effective mass or undressing of the itinerant carriers,[11] occurring at T_C and necessary to explain the blueshift of the plasma edge,[10] has recently been claimed to offer new routes to understand metallic ferromagnetism.[12] Hirsch's new point of view suggests that metallic ferromagnetism is driven by band broadening or, equivalently, an effective mass reduction that occurs upon spin polarization.[12] In totality this leads to a decrease rather than a rise in kinetic energy. Alternative scenarios related to ferromagnetism and enhanced metallicity, based on polaron formation, have also been proposed.[13–15] Millis *et al.* have demonstrated that the double-exchange model must be complemented by a strong electron-phonon interaction, arising from Jahn-Teller effects and leading to polaron formation, in order to explain the colossal magnetoresistance in the series of manganese oxides.[13] An overlap of magnetic polarons, first suggested on the basis of Raman data,[14] was claimed to account for the enhancement of the electrical conductivity upon the onset of magnetic order in EuB_6.[15]

In this work we analyze data concerning the magnetic-field dependence of the optical response of EuB_6 at several temperatures above and below T_C, the zero-field Curie temperature. We find an equivalence between the temperature and the magnetic-field dependence of the plasma edge, and the extracted plasma frequencies scale universally with the magnetization. The data analysis reveals that the effective mass of the itinerant charge carriers decreases with increasing field and decreasing temperature, suggesting some kind of undressing of the itinerant quasiparticles upon spin polarization.

We used the same well-characterized sample investigated in Refs. 10 and 11 with $T_C = 15.9$ K. Optical reflectivity $R(\omega)$ data at selected temperatures above and below T_C have been collected between 15 and 5000 cm^{-1} in magnetic fields ranging from 0 to 7 T. We used an Oxford magneto-optical cryostat with appropriate windows. In principle, the optical reflectivity with suitable extrapolations at low ($\omega < 15$ cm^{-1}) and high ($\omega > 5000$ cm^{-1}) frequencies can be used for the Kramers-Kronig transformations providing the optical functions.[10,16] For the purposes of this report, we concentrate on the reflectivity collected in the midinfrared spectral range. Figure 1 summarizes the general trend of $R(\omega)$ between 750 and 3000 cm^{-1}, at selected temperatures. We recognize a clear metalliclike plasma edge, which is blue-

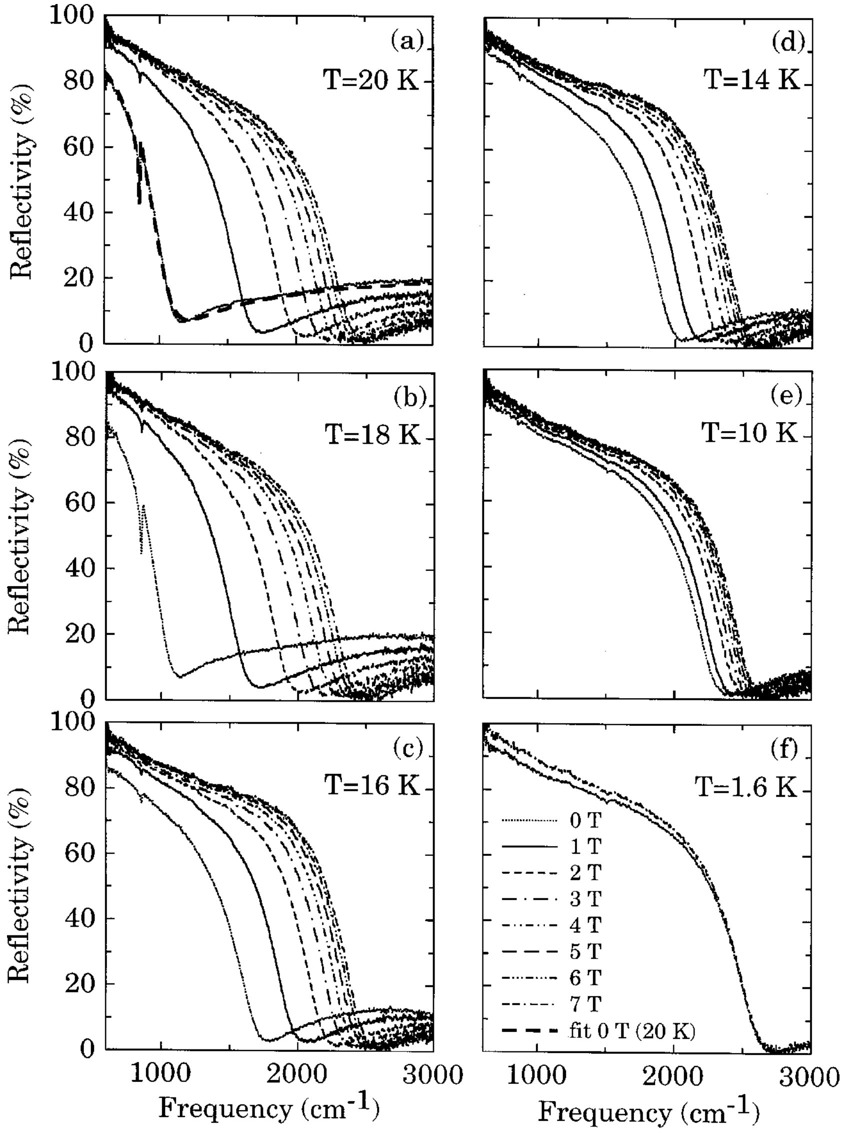

FIG. 1. Magneto-optical reflectivity of EuB_6 at (a) 20, (b) 18, (c) 16, (d) 14, (e) 10, and (f) 1.6 K in the midinfrared spectral range. At 20 K the fit for 0 T is presented as an example of the fit quality.

shifted with increasing field. The temperature dependence of the plasma edge in zero field coincides with that of our previous results.[10] The plasma edge defines the screened plasma frequency $\tilde{\omega}_p$, the screening being caused by the electronic interband transitions at high energies. The phonon mode at 850 cm^{-1},[10] distinctly visible at 20 K and 0 T, is progressively hidden by the blueshifted plasma edge. In the far infrared spectral range, not shown here, a second optically active phonon mode is observed at about 150 cm^{-1},[10] which is also hidden by the progressively enhanced metallic reflectivity with increasing fields. The blueshift saturates at low temperatures. A magnetic-field-induced appearance of the plasma edge was previously observed for $Pr_{1-x}Ca_xMnO_3$.[17] In contrast to EuB_6, however, the appearance of Drude weight with the field in Mn oxides coincides with an insulator-to-metal transition at T_C, from a charge-ordered insulator to a ferromagnetic metal. No hysteresis effects in $R(\omega)$ were observed when sweeping the field up and down at all temperatures.[18]

From the reflectivity data we may extract the unscreened plasma frequency ω_p, i.e., $\omega_p^2 \sim \tilde{\omega}_p^2 \varepsilon_\infty$, ε_∞ being the optical dielectric constant representing the screening by the high-frequency transitions, using the standard phenomenological Lorentz-Drude approach.[10,19] We describe the complex dielectric function and ultimately the optical reflectivity with an appropriate ensemble including a Drude component for the itinerant charge carriers and Lorentz harmonic oscillators for non zero energy excitations, respectively.[19] The fits at any given combination of temperatures and fields are consistent with our previous analysis at 0 T.[10] As an example, a corresponding fit at 20 K and 0 T is shown in Fig. 1(a) the same fit quality is obtained for all data.[16] Figure 2(a) shows the temperature dependence of ω_p at various fields. We note the significant increase of ω_p below T_C in zero field.[10] With increasing field the enhancement of ω_p with decreasing temperature is less pronounced and ω_p tends to saturate around 6000 cm^{-1} at low temperatures. It is natural to correlate this

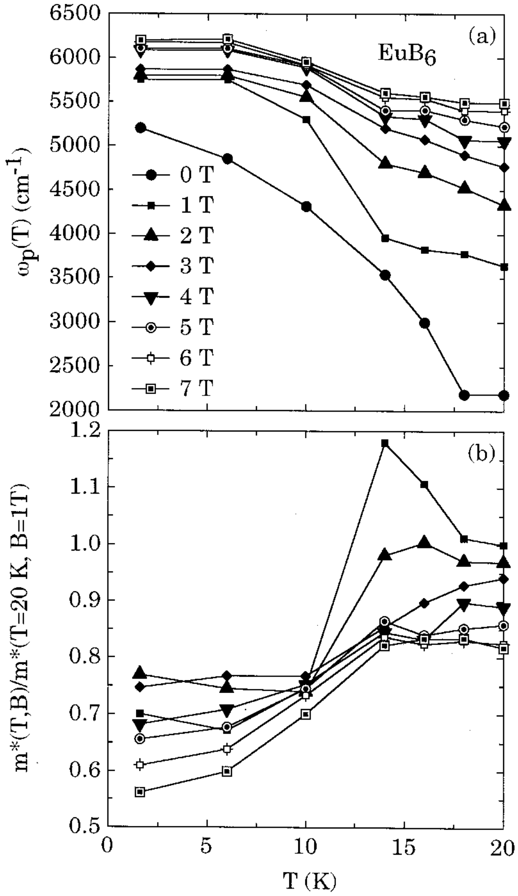

FIG. 2. Temperature dependence at various magnetic fields of (a) the plasma frequency ω_p and (b) the effective mass m^* of the itinerant charges carriers, calculated from Hall-effect data of Ref. 11 and normalized by the value at 20 K and 1 T.

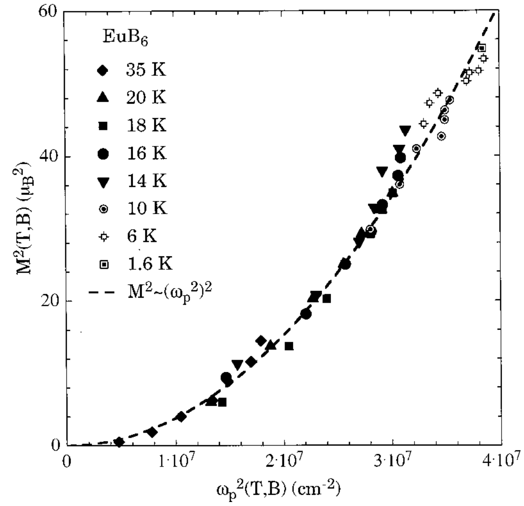

FIG. 3. Squared magnetization versus squared plasma frequency (i.e., Drude weight) of EuB_6.

temperature- and magnetic-field-induced variation of ω_p with the large negative magnetoresistance.[7,11]

Given the plasma frequency $\omega_p{}^2 \sim n_{\mathrm{eff}}/m^*$, we may estimate the effective mass of the itinerant charge carriers by using the Hall-effect data measured between 1 and 7 T.[11] The effective charge-carrier concentration n_{eff} extracted from the Hall number R_H is, in a perfect semimetal, related to the real electron (or hole) concentration n by $n_{\mathrm{eff}}=n(\mu_p+\mu_n)/(\mu_p-\mu_n)$, where μ_p and μ_n are the hole and electron mobilities, respectively. Thus, the real electron concentrations n in EuB_6 may even be smaller than n_{eff} used to calculate m^* from ω_p. Another difficulty arises from the nonlinearity of $R_H(H)$. However, since we are simply interested in the trend of the relative change of m^* versus field and temperature, we are confident that our approximation is not seriously affecting the main conclusions of our analysis. Figure 2(b) shows the calculated temperature dependence of m^* in fields between 1 and 7 T. We note a general reduction of the effective mass upon cooling to below T_C, which is more pronounced at lower fields.[11,20] In low fields and at temperatures close to and just above T_C, m^* increases [Fig. 2(b)] because of the small redshift of ω_p above T_C in zero field[10,20] and the rapid increase of n with respect to ω_p at low fields and at temperature close to T_C. This is consistent with some predictions of Ref. 12. In order to account for the blueshift of the optical plasma edge below T_C (Fig. 1), the enhancement of the free-charge-carrier concentration upon spin polarization, as evidenced from Hall data,[11] must be combined with a reduction of the effective mass [Fig. 2(b)]. These simultaneous changes in carrier concentration and effective mass that follow from our data contradict the findings reported in Ref. 21 where, upon the onset of ferromagnetism, no measurable modifications of the Fermi-surface dimensions or carrier masses have been seen. However, the high magnetic fields needed to observe quantum oscillations by far exceed the fields, which are needed for a full polarization of EuB_6 at any temperature below 30 K. Therefore, because of the saturated magnetization the Fermi surface keeps its shape.[15]

The reduction of m^* is a manifestation of an undressing of the quasiparticles, in coincidence with the onset of the ferromagnetic state. As the magnetization increases, carriers undress and the spectral weight of the quasiparticles increases.[12] The calculated magnetoresistance, assuming the undressing of quasiparticles as the driving mechanism for ferromagnetism,[12] yields the same behavior for the magnetic-field dependence of the measured transport properties as is indeed observed.[7,11] These calculations do not necessarily invoke a substantial change in the quasiparticle scattering rate with magnetization[12] and they provide results that are consistent with our fits. In zero field, the scattering rate, extracted from the phenomenological Lorentz-Drude analysis, decreases at T_C (Ref. 10) but our results indicate that it does not change as a function of magnetic field at fixed temperature. This seems to imply that in view of the total scattering rate, spin-flip processes are not important. Although the idea of quasiparticle undressing in connection with ferromagnetism[12] is appealing and finds some experimental confirmation, the experimental data (magneto-optics and Hall effect) imply a more complicated situation, where variations of both n and m^* are important.

The enhancement of ω_p with decreasing temperatures and increasing fields is due to a shift of spectral weight collected between 2000 and 10^4 cm^{-1} into the spectral range below

2000 cm^{-1}.[16] Such a spectral weight shift in coincidence with the ferromagnetic transition is a key prediction of the double-exchange-based model of Millis *et al.*[13] The crucial point is that optical processes conserve spins. In the fully polarized ferromagnetic state it is impossible for an optical process to create a final state with the electron spin antialigned with the local moments, i.e., between the lower and upper spin band with an excitation energy of the order $2J$, J representing Hund's coupling. Therefore, only intrabandlike Drude transitions are possible.[13,22] A strong Hund's rule coupling to the local moments causes a perfect alignment of itinerant spins, thus reducing the possibility of spin-flip-scattering processes. This latter model prediction seems to be consistent with our findings on the temperature and field dependence of the Drude scattering rate. Nevertheless, a strong electron-phonon coupling, associated with the formation of Jahn-Teller polarons, is an essential ingredient of the model in order to explain the sharp drop in the resistivity at the transition.[13] In view of the ionic configuration of Eu^{2+}, no Jahn-Teller effects are expected for EuB_6.

Alternatively, the sharp drop in resistivity could be the consequence of magnetic-polaron overlap, claimed to develop around 30 K.[14,15] Such an overlap should lead to a metallization in EuB_6, manifest in a rapid increase of the conduction-electron concentration and a decrease of their mass.[15] While at present we are not able to explicitly identify polaronic features in our spectra, some consequences of both these polaron scenarios are in agreement with our optical results.

Finally, we report the unexpected identification of a universal relation between the magnetization **M** (Ref. 23) and the plasma frequency ω_p. As shown in Fig. 3, the scaling relates $\mathbf{M}^2$ to ω_p^2 and provides a strong argument in favor of the magnetic polarization involving simultaneous variations of n and m^*. The scaling is valid at temperatures both above and below the Curie temperature T_C in zero field, and points to an intimate relation between the optical weight of the itinerant charge carriers and the moment spin polarization. In the mean-field approach of the double-exchange model[24,25] the temperature dependence of $\mathbf{M}^2$ represents the variation of the spectral weight of the intraband excitations. A scaling between ω_p and **M** seems also to be implicit in Hirsch's model, although both quantities are claimed to be only affected by the effective-mass behavior as a function of both temperature and magnetic field.[12] In this sense we are not aware of a solid theoretical basis and it remains to be seen how the phenomenological trend, i.e., $\mathbf{M}^2 \sim (\omega_p{}^2)^2$, can rigorously be explained. The scaling is obviously not affected by the phase transition in different magnetic fields, a remarkable and theoretically challenging result.

In conclusion, we have shown that the optical response of EuB_6 is characterized by a huge blueshift of the plasma edge in reflectivity as a function of both temperature and magnetic field, in coincidence with the ferromagnetic transition, as well as by a surprising scaling between the Drude weight and the magnetization for temperatures above and below T_C. Identifying the reason for such a scaling is beyond the scope of this paper and new theoretical input is clearly needed. The undressing of quasiparticles and, more generally, the shift of spectral weight into the Drude term upon spin polarization-seem to be important secondary manifestations of the onset of ferromagnetism.

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